



Characterization of composite materials with giant magnetostriction

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ABSTRACT

Purpose: This paper concerns manufacturing and researching a new group of the magnetostrictive composite materials, consisting $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles in different polymer matrix.

Design/methodology/approach: The following investigation studies have been carried out: the structures of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders, the topography of composite materials fractures with the use of electron scanning microscopy; chemical composition of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders using the EDS method; phase composition of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders and composite materials using X-ray diffraction; as well as properties of the composite materials with polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles including: density, thermal conductivity (in magnetic field also), electrical and magnetic properties, as well as magnetostriction in magnetic field intensity up to 800 kA/m using a three terminal capacitance technique.

Findings: Based on the examination carried out it was found that the highest magnetostriction equals to $805 \cdot 10^{-6}$ (with magnetic field intensity of 766 kA/m) characterizes a composite material with polyurethane matrix, reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder of 20% volume fraction and particles size in the range of 38-106 μm . Simultaneously, the materials is characterized with coercion intensity $H_c = 5.39$ kA/m, remanence $B_r = 0.013$ T and magnetic permeability $\mu_r = 1.13$. Moreover, it was found that the maximum magnetostriction value for this material assumes values approximate to the magnetostriction of monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ alloy. Additionally, it was confirmed that the correlation exists between the diversification of the volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder in the matrix, its particle distribution and the maximum magnetostriction and magnetic properties of the developed composite materials. The key factor determining the energy transfer efficiency between $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ and matrix is – in addition to the size and volume fraction of the magnetostrictive particles in the matrix – the method of combining those two phases.

Originality/value: The paper presents the research involving the composite material with giant magnetostriction.

Keywords: GMM; $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ (Terfenol-D); Composite materials; Functional (smart) materials

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MATERIALS

1. Introduction

The rapid civilization progress caused that numerous fields of technology offer products whose desired applications can be achieved by combination of mechanical, IT and electrical or electronic elements only. The examples include anti-blockier, anti-skid systems and airbags in the automotive industry, manipulation systems and robots in automation and micro-mechanical devices of medical technology. For the correct operation of such devices – called mechatronic systems – the appropriate system of sensors and actuators, as well as the application of mathematical models to obtain information from measured signals are required [1-3].

Like the automatic regulation systems, intelligent materials (defined also as smart/adaptive/active or multifunctional materials) – whose dynamic growth has been observed for the last few years – perform function as the sensor (through recording external stimulation), processor (analyzing the changing environmental conditions) and actuator (adapting its properties to the changed environmental conditions) – at the same time proving the reversibility of such changes and the feedback effect [1,4-7].

One of the important targets in the field of active intelligent materials is to construct a device in which maximum deformation will be achieved with electricity of magnetic energy consumption as low as possible. Due to the beneficial ratio between the deformations obtained and the volume of a given executive element, the magnetostrictive materials are significantly distinguishable against the stress sources used so far. Their key importance is the result of the potential to transport mechanical into magnetic energy (sensor) and magnetic into mechanical energy (actuator) of very high efficiency (Fig. 1) [8,9].

Summarizing, these materials should be characterized with significant magnetostriction and minimum impact of external factors on its changes. The major inconvenience related to using magnetostrictive materials is the necessity to use magnetic stimulation coil and circuit, which increases the equipment size and deteriorates their competitiveness in relation to piezoelectric and electrostrictive materials. Therefore, they are mainly used wherever large deformations are required, while sizes play a minor role.

The development of magnetostrictive materials began in 1842 with experimental observations of J.P. Joule who found that magnetization of a ferromagnetic sample (iron) causes change of its dimensions. Such phenomenon was later shown also in cobalt, nickel and other metal alloys (Table 1) – however, the change of the sample length along

the magnetic field direction occurring in strong magnetic fields is scarce for those materials [10-12].

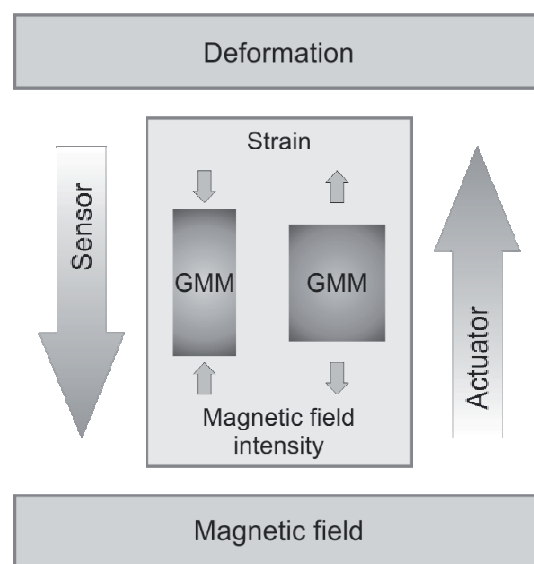


Fig. 1. Schematic view of energy transformations in materials with giant magnetostriction [8]

Table 1.

Comparison of achieved strain in conventional materials [12]

Material	Strain, 10^{-6}
Nickel	-46
Nickel alloys	120
Piezoelectric ceramic	< 1000
Tb _{0.3} Dy _{0.7} Fe _{1.9}	800-1200

The source of magnetostriction – like crystalline anisotropy – is the magnetic coupling of the spin and orbital moment of electrons (Fig. 2). Under the influence of an external magnetic field, spin moment follows the direction of the orbital moment, which is coupled to the crystal lattice, which encounters high resistance of material. At sufficiently high magnetic fields it leads to a change in the length of bonds – depending on the direction – in the crystal lattice, i.e. magnetostriction, which is numerically expressed by the relation [13-16].

$$\lambda = \frac{\Delta l}{l} \quad (1)$$

where:

Δl – changing the length of the sample under the influence of magnetic field;

l – initial length of material.

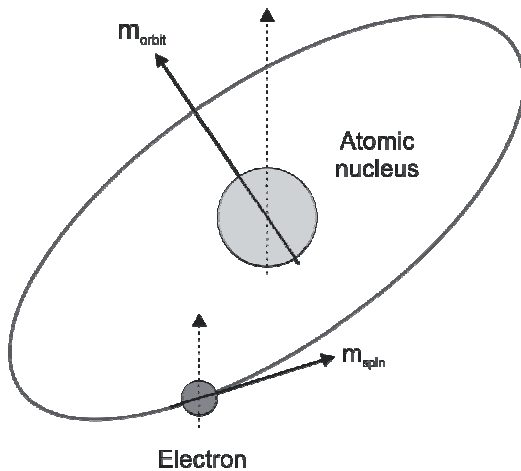


Fig. 2. Spin magnetic moment orienting (m_{spin}) under the influence of a magnetic field intensity H [15]

Assuming that the deformation of crystal depends on the direction of the spontaneous magnetization with respect to the axis of the crystal (i.e. from the directional cosines) α_1 , α_2 and α_3 , magnetostriction can be described by following equation [14,17]:

$$\lambda = \frac{3}{2} \cdot \lambda_{[100]} (\alpha_1^2 \beta_1^2 + \alpha_2^2 \beta_2^2 + \alpha_3^2 \beta_3^2 - \frac{1}{3}) + 3 \cdot \lambda_{[111]} (\alpha_1 \alpha_2 \beta_1 \beta_2 + \alpha_2 \alpha_3 \beta_2 \beta_3 + \alpha_3 \alpha_1 \beta_3 \beta_1) \quad (2)$$

where:

- $\alpha_1, \alpha_2, \alpha_3$ – direction cosines of the spontaneous magnetization direction of the crystal axis;
- $\beta_1, \beta_2, \beta_3$ – direction cosines of the vector in the non-deformed crystal;
- $\lambda_{[100]}, \lambda_{[111]}$ – saturation magnetostriction measured along the respective direction.

As a result of magnetostriction, such ferromagnetics as iron or nickel have distorted crystal structure above Curie temperature only. In lower temperatures – as a result of spontaneous magnetization – the balance conditions in the crystal change, causing spontaneous deformation, which leads to reduction of the anisotropic energy; moreover, each magnetization change causes forced magnetostriction, however this dependency is reversible – the external stresses affect the magnetization course [13,15-18].

Magnetostriction of various intensity and direction takes place in the direction of both easy and difficult crystal magnetization and its absolute magnitude and its plus or minus sign depend on the ferromagnetic type (Fig. 3), its chemical composition, thermal and mechanical processing,

as well as the direction and value of the magnetic field and ambient temperature [14,19,20].

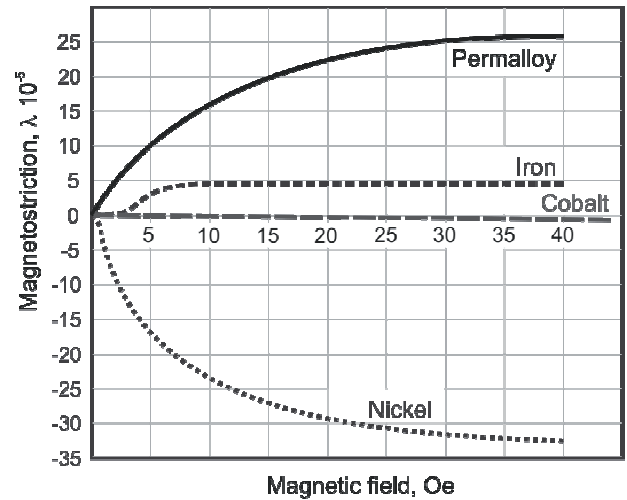


Fig. 3. The longitudinal magnetostriction dependence of the magnetic field [14]

The slight deformation changes obtained in iron and nickel (Table 2) caused that the materials were not widely applied. The turning point in this scope took place in the 1960's, when it was discovered in Naval Ordnance Laboratory (USA) that rare earth elements under temperature ca. 0 K show magnetostriction several hundred times higher than that of the said materials. However, the problem in practical applications of the Tb-Fe compound is that, because of a large magnitude of intrinsic magnetocrystalline anisotropy, a large magnetic field is required to achieve the giant magnetostriction. This problem can be solved with a great success by the partial substitution of Dy for Tb. Attempts have been made to reduce the magnetocrystalline anisotropy by controlling the microstructure – the theoretical background for this is that the magnitude of magnetocrystalline anisotropy is “effectively” reduced when the size of grains is smaller than the ferromagnetic exchange length. The reduction of magnetocrystalline anisotropy of the Tb-Dy-Fe alloy (Fig. 4) is known to be caused by a cancellation of the anisotropy constants of Dy and Tb, which have opposite signs, in the crystal symmetry of the cubic Laves (Tb,Dy)Fe₂ phase. In the case of an alloy with a Tb/Dy ratio where a complete cancellation of the anisotropy occurs, the cancellation may not be achieved completely if the cubic structure is distorted [21-23].

Table 2.
Comparison of magnetostrictive materials selected properties [24]

Material	Density, g/cm ³	Young's modulus, GPa	λ_s , %	Curie temperature, K
Fe	7.86	190	-0.0014	633
Ni	8.9	205	-0.005	1043
Permalloy (65% Fe, 45% Ni)	-	-	0.0027	713
SmFe ₂	8.53	-	-0.2340	688
DyFe ₂	9.28	-	0.0650	635
Tb _{0.3} Dy _{0.7} Fe _{1.9}	9.21	29	0.2000	653
TbFe ₂	9.06	-	0.2630	703

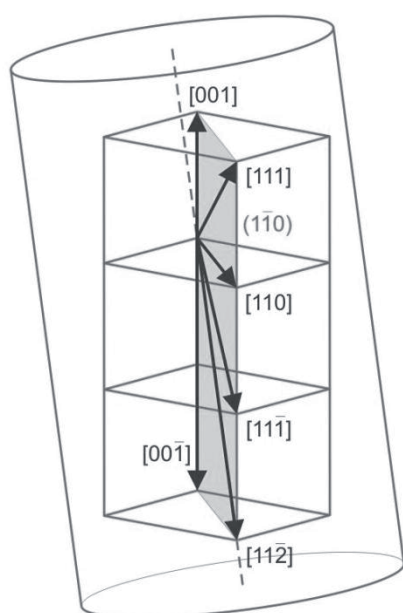


Fig. 4. Crystallographic orientation of Tb_{0.3}Dy_{0.7}Fe_{1.9} [25]

The research with the use of terbium [26] – and later also samarium [27-29] or gallium [21,23] – with dysprosium – began the development of Giant Magnetostrictive Materials (GMM). In 1986 ETREMA Products Incorporation (USA) launched Tb_{0.3}Dy_{0.7}Fe_{1.9} (Terfenol-D) on the market – nowadays the most commonly used material with giant magnetostriction; its saturation magnetostriction for the particular directions being: $\lambda_{[111]}=1640$, $\lambda_{[100]}=9$ and $\lambda_{[112]}=1200$. Furthermore, it is also possible with giant magnetostrictive materials to produce deformations equal to 2000 ppm even, which is possible thanks to evoking compressive pre-stress (≥ 20 MPa) (Fig. 5) [2,30-32].

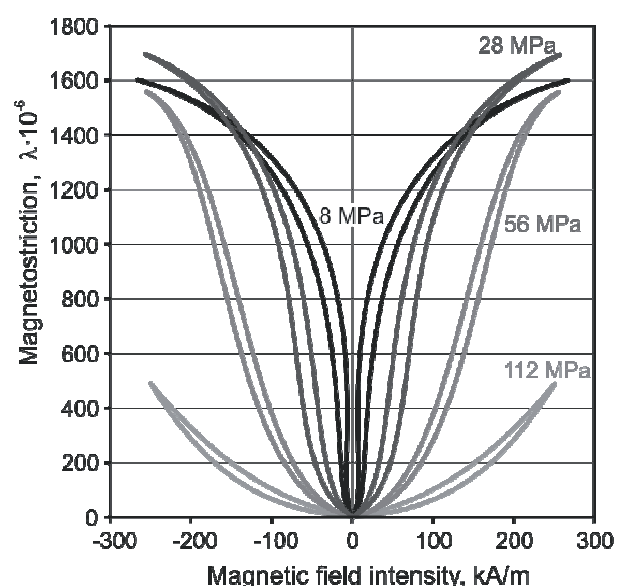


Fig. 5. Tb_{0.3}Dy_{0.7}Fe_{1.9} magnetostriction dependence of the magnetic field intensity at various initial stresses [33]

The application of the pre-stress beneficially affects the magnetostriction value, particularly in low magnetic fields. From the practical point of view, starting a particular transducer may be more efficient thanks to the minimization of the losses occurring in the coil. Due to the fact that the $\lambda=f(H)$ curve is non-linear and show a hysteresis, both the field value and uniaxial pre-stress should be appropriately matched, in order to obtain the optimum transduction value [12,33,34].

The factors restricting the application of Giant Magnetostriction Materials 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 [23,35]:

- 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);
- corrosion strength;
- simple production technologies;
- low price.

Attempts have been made to restrict the defects specified above, including without limitation doping $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ with copper, both in the amorphous and crystalline form, and through the explosive compaction technology [36]. In spite of their increased strength – in comparison to the basic material – the alloys have not been applied in practice, because their magnetostriction is just 18% of that of solid $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$. On the other hand, it will be beneficial to applications if low-cost metal can be totally or partially substituted for Tb or Dy while competitive properties can still be maintained.

The optimization of magnetostrictive alloys' properties was also made through synthesizing the materials with the strength of the specific elements corresponding to: $\text{Tb}_x\text{Dy}_{1-x}(\text{Fe}_{0.8}\text{Co}_{0.2})_2$ ($0.20 \leq x \leq 0.40$) [37], $\text{Pr}_{0.15}\text{Tb}_{0.3}\text{Dy}_{0.55}\text{Fe}_2$ [38], $\text{Tb}_x\text{DyHo}_{1-x-y}\text{Fe}_{1.95}$ [39], $\text{Tb}_{0.27}\text{Dy}_{0.73}(\text{Mn}_{1-x}\text{Fe}_x)_2$ ($x=0; 0.2; 0.4; 0.6; 0.8$) [40], $(\text{Tb}_{0.3}\text{Dy}_{0.7})_{40}(\text{Fe}_{0.95}\text{X}_{0.05})_{60}$ ($\text{X}=\text{Nb}, \text{Zr}$) [41], $\text{Tb}_{0.27}\text{Dy}_{0.73}(\text{Fe}_{1-x}\text{Co}_x)_2$ ($x=0-1$) [13], $\text{Tb}_{0.3}\text{Dy}_{0.7-x}\text{Pr}_x(\text{Fe}_{0.9}\text{Al}_{0.1})_{1.95}$ [42].

Studies have shown that, in spite of the possibility of combining high magnetostriction with low anisotropy in these materials, the alloys have disadvantages characteristic for metallic materials, including induction of eddy currents, caused by low resistivity of the material. These factors limit and sometimes prevent their use in transducers operating in the magnetic fields with rapidly changing frequencies.

The magnetostrictive composite materials composed of alloy powder with magnetostrictive properties dispersed in a non-magnetic matrix of aluminum [43], copper [21,43] or glass [44,45] have been developed in order to minimize such defects as fragility and losses for eddy currents in monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ alloys. However, a significant reduction of magnetostriction was recorded in these materials, which was a prerequisite for searching for some more efficient solutions.

Among the advantages of the magnetostrictive composite materials with polymer matrix are: simple

technology, possibility of predicting their properties, production cost reduction thanks to the lack of costly finish processing, as well as reduction of material losses, resulting from the possibility to form practically any shape of the final materials. Furthermore, the stresses occurring in the polymer material during its cross-link process evoke a stress exerted on $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles, restricting the need to apply external pre-stress, necessary for the favorable orientation of domains in the monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ [46-50].

The factors were the inspiration for the development and improvement production methods of magnetostrictive composite materials with polymer matrix (Fig. 6), including:

- binding magnetostrictive material in form of powder [34,45,46,51-61] or fibers [62] with the polymer material (so-called 0-3 type composite materials);
- binding $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with polymer material and aligned magnetostrictive particles in the magnetic field (1-3 fiber composite materials, „pseudo-fiber”) [16,52,53,55,57,60,63-65], including materials, in which magnetostrictive particles are $\langle 112 \rangle$ -oriented (Crystallographically Aligned $\langle 112 \rangle$ Magnetostrictive Particle Composites, CAMP) [30,37,46,62,66];
- laminate materials [67,68], including thin layers for Micro-Electro-Mechanical System (MEMS) application [69-72];
- hybrid smart structures in which the magnetostrictive materials is combined with e.g. piezoelectric [67,73-77];
- Polymer Infiltrated Grain Aligned Composite (PIGAC) – the composite consists of bulk RFe_2 dendrites with a preferred orientation of $\langle 112 \rangle$ and binder between the dendrites [78-79].

Hitherto, the following materials has been used as the matrix for the magnetostrictive composites: epoxy resin [44,45,53,54,60,63,66], vinyl-ester resin [30,46,66], phenol-type resin [38,44,55,58], thermoplastic polyurethane [51,56,65] or unsaturated polyester resin [80]. Proper selection of matrix material have a significant influence on magneto-mechanical characteristic will achieved.

The purpose of the scientists throughout the last 30 years (mainly in the USA and in East Asian countries) is the production of composite materials with possibly the highest magnetostriction with simultaneous limitation of the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ fraction share in the matrix. Even the best magnetoelastic, quasi-static and dynamic properties of these materials, achieve significantly lower values, however, than their monolithic equivalents, which

dramatically restricts the possibility of their commercial use [30,45,63].

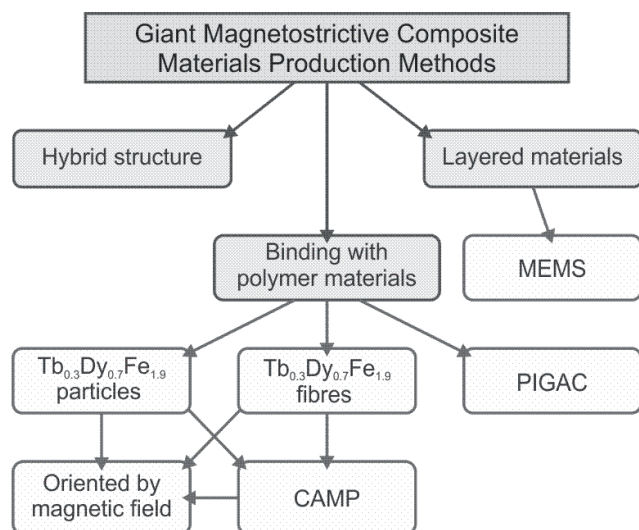


Fig. 6. Scheme of manufacturing magnetostrictive composites methods

Contradictory reference information on the factors affecting the magneto-mechanical behavior of magnetostrictive composite materials [2,43,44,53] and the need for less expensive alternative for the conventional GMM, make us search for optional solutions, in order to maintain the advantages resulting from dispersion of the magnetostrictive particles in polymer matrix with simultaneous provision of the most efficient energy transformation.

The integration of components with opposite properties in magnetostrictive composite materials and the variety of factors affecting their magneto-mechanical properties cause that the issue of appropriate selection of the matrix material and $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ share is complex. Considering the factors mentioned above, the idea of research concerning new group of magnetostrictive composite materials has been purposeful, in order to obtain high magnetostriction with simultaneous reduction the material costs, increased resistivity (reducing the occurrence of losses evoked by eddy currents), reduction of the necessity to set the initial stress, and – in the same time – restriction of the magnetically neutral phase effect on the magnetic properties, including the values of magnetic permeability, coercive force and saturation induction.

2. Material for investigation

The material for the tests has been prepared using as reinforcement the commercially available $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder (ETREMA Products Inc., USA) produced according to the USA patents no: 4308474, 4609402, 4770704, 4849034 or 4818304, by grinding in the ball mill in neutral gas atmosphere. The basic properties of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ are listed in the Table 3. As the matrix materials, the following polymers have been used (Fig. 7): polyester resin, polyurethane resin, epoxy resin, high density polyethylene (PE-HD) or acrylonitrile butadiene styrene (ABS). The matrix material selection determined the technology of composite material production, which included: casting (for polyester, polyurethane and epoxy resins), compacting (for PE-HD) and injection moulding (for ABS). The application of such methods minimized the increased temperature unfavorable impact on the composite materials physical properties because the conditions of forming them were limited by the maximum temperature of polymer processing.

Table 3.
Properties of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ [33]

Property	Value
Density	9250 kg/m ³
Curie temperature	653 K
Mechanical properties	
Young's modulus	25-35 GPa
Tensile strength	28 MPa
Compressive strength	700 MPa
Thermal properties	
Coefficient of thermal expansion	12 ppm/°C
Specific heat	0.35 J/kg·K
Thermal conductivity	13.5 W/m·K
Electrical properties	
Resistivity	58·10 ⁻⁸ Ω·m
Magnetomechanical properties	
Strain (estimated linear)	800-1200 ppm
Energy density	14-25 kJ/m ³
Relative permeability	3-10 H/m
Coupling factor	0.75

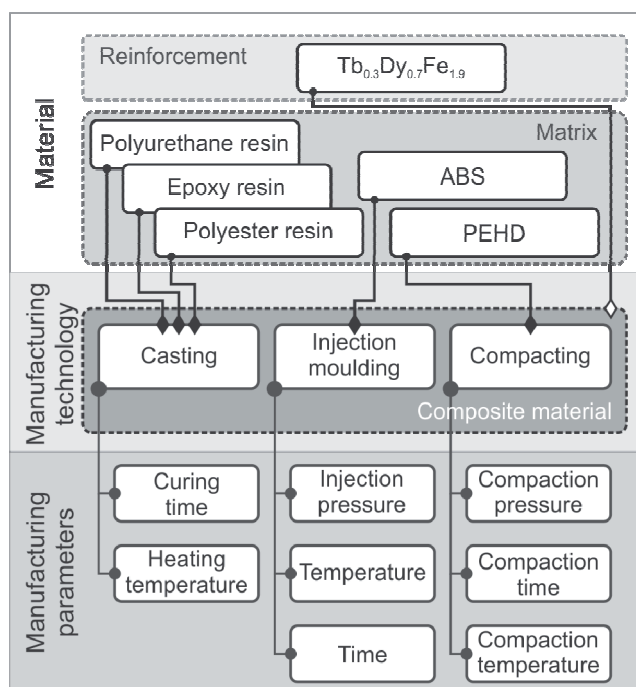


Fig. 7. Magnetostrictive composite material scheme of preparation (first stage of the study)

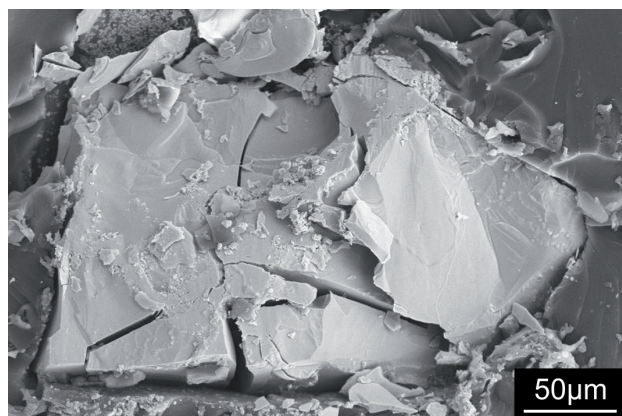


Fig. 8. Fracture of the composite material with polyethylene (PE-HD) matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles

Basing on the materialographic observations and magnetostriction measurements [51,56,81,82], it was found that the most beneficial combination of magnetomechanical properties with simplicity of production (significant due to the potential possibility to use the materials developed as the final control elements

of sensors and actuators) are presented by composite materials produced by casting, i.e. with polyurethane matrix reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles.

It has been further found that the pressure applied for the production of composite materials through injection molding or compacting – due to generating internal stresses in $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles (Fig. 8) – has a negative effect on the magnetic properties of the materials developed [81]. Taking into account above factors, a self-deaerating, two component liquid unsaturated polyurethane resin Smooth Cast 325 (SMOOTH-ON Inc., USA) was used as optimal matrix (Table 4).

Table 4.
Properties of Smooth Cast 325 resin [83]

Property	Value
Density	1070 kg/m ³
Viscosity	0.1 Pa·s
Shrink	1 %
Pot time	2 min
Demold time (at 296 K)	10 min
Young's modulus	965.27 MPa
Tensile strength	21.86 MPa
Compressive strength	24.13 MPa
Ultimate Shore hardness	72

Matrix material upon combination with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder was casted to a die of 8.7 mm diameter and 40 mm height, maintained – upon sealing – in permanent motion throughout the matrix material gelation time, in order to prevent sedimentation of the reinforcing powder. After the time necessary for the polyurethane resin cross-linking, the samples were taken out of the die and reheated for 5 hours in the temperature of 65°C. Then the magnetostrictive composite materials with polyurethane matrix and reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder were optimized through the selection of reinforcing particle sizes (38-106 µm, 106-212 µm or 212-300 µm according to denoting of manufacturer) and their volume fraction in the matrix (10, 15 and 20%). The selection of the reinforcement material shares for magnetostrictive composite materials was related to the application aspects, while the restriction of the powder share to no more than 20% by volume resulted from the insufficient

binding of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with the matrix due to excessive increase of the mixture viscosity, preventing its casting in ambient temperature.

3. Investigation methodology

The morphology of the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders and transverse fracture structure were examined using a SUPRA 35 (ZEISS) scanning electron microscopy (SEM) at the accelerating voltage of 20 kV. The composite materials fractures were prepared for observation as a result of breaking in boiling temperature N_2 and prior to the imaging, each sample was sputtered with a 30 nm layer of gold. The qualitative and quantitative analysis of the chemical composition of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder was carried out using the X-ray energy dispersive spectroscopy (EDS) with the application of the EDS LINK ISIS spectrometer of the Oxford Company being a component of the scanning electron microscope.

The analysis of the phase composition of the composite materials and $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders representing their reinforcement was carried out using PANalytical X'Pert PRO X-ray diffractometer in a goniometric system, using the filtered K radiation of the cobalt lamp at 40 kV voltage and 30 mA heater current. The reflected radiation intensity measurements were made in the 2θ angle range from 35 to 95° every 0.05° and counting time of 10 seconds.

The composite materials density was determined according to the standard [84], while theoretical density was calculated using the rule of mixtures.

The thermal conductivity of the composite materials in the magnetic field of 1 tesla induction was examined on a Physical Property Measurement System (PPMS) device (QUANTUM DESIGN). The tests were made on samples with dimensions: 8x2x2 mm using the double-contact method in the temperature range of 293-333 K.

Electrical properties testing were made on workstation presented in the [56]. In order to provide the precise contact between the sample surface and the electrodes, enabling a homogeneous distribution of the electric charge on the measured current intensity value was, the samples prepared for the tests had copper pulleys included on the ends; their diameter was 8.7 mm and length 3 mm.

The test of the dependency of magnetization on the magnetic field was recorded by a vibrating sample magnetometer (VSM) (Oxford Instruments) with magnetic field intensity up to 1250 kA/m. Based on the magnetization dependency on magnetic field intensity

curves recorded hysteresis loops were plotted, enabling the determination of the coercive force intensity H_c , saturation induction B_s , remanence B_r and magnetic permeability μ_r of the newly developed composite materials and $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders. The measurements were made in ambient temperature on samples with dimensions: 2x3x6 mm.

The magnetostriction was examined with the magnetic field intensity up to 800 kA/m using a three terminal capacitance technique. The measurements were made lengthwise and crosswise in relation to the sample axis (Fig. 9) on the sites constructed at the Institute of Physics Polish Academy of Sciences in Warsaw (Poland). In this method the elongation of the sample is observed as a change of the capacity of capacitor with moving electrode in the external magnetic field created by the superconducting magnet. As a result of the magnetostrictive deformation the sample presses the moving electrode, thus changing the condenser's capacity. The magnetostriction measurements parallel to the sample axis are enabled by a capacitive camera [82].

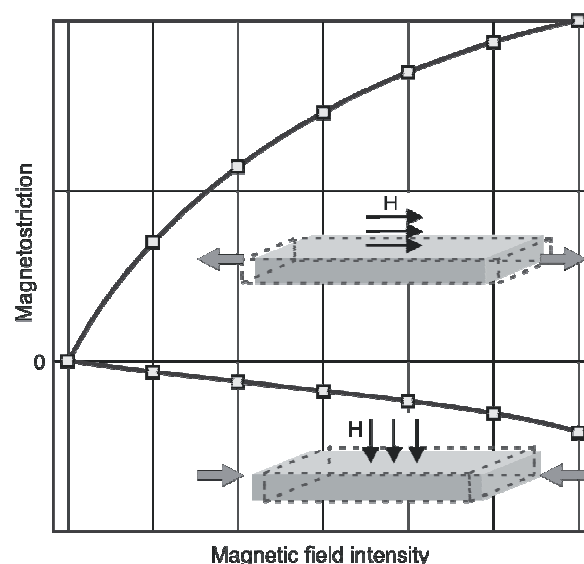


Fig. 9. A method of sample orientation in regard to the direction of the magnetic field during the measurement of magnetostriction

The tests were carried out in ambient temperature on samples of 2x3x4 mm dimensions and the results presented are values obtained from the second measurement cycle – the results from the first cycle were rejected due to the unknown initial orientation of the magnetic domains.

4. Description of achieved results

The $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders' morphology is presented in the Fig 10 – it can be seen that the powders are differ both with size and shape of grains, which are irregular in the entire range.

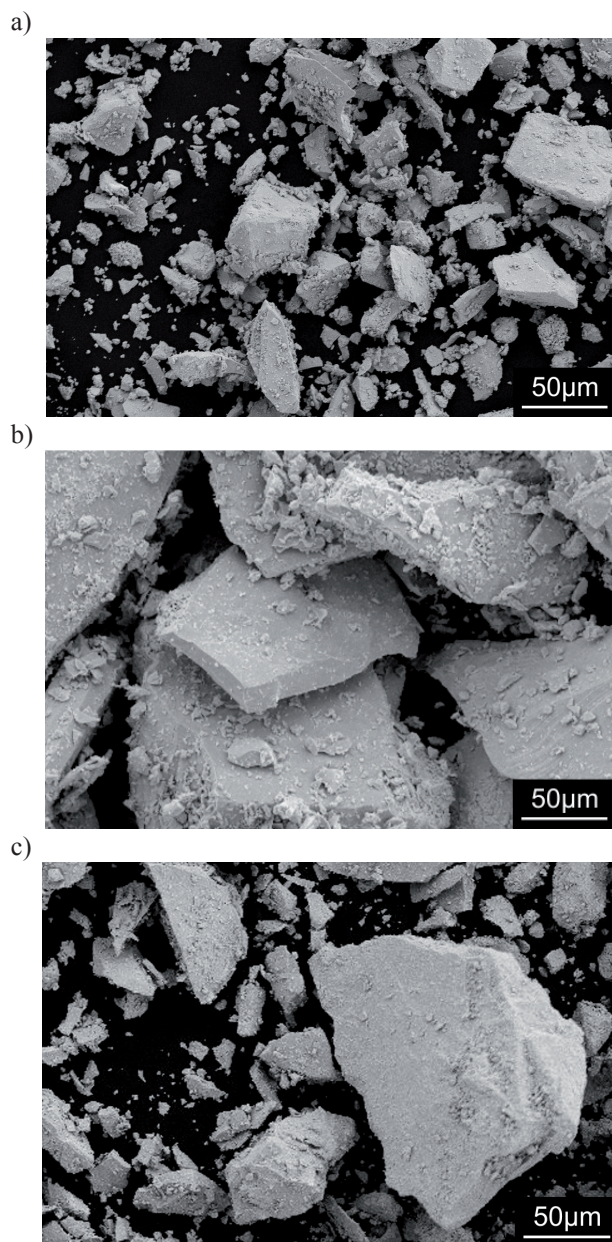


Fig. 10. Scanning electron micrographs of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size of: a) 38-106 μm ; b) 106-212 μm ; c) 212-300 μm

The qualitative X-ray analysis of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder provides the information about appropriate elements (Fig. 11), and in effect of the quantitative analysis the information about mass concentration of particular elements was obtained (Table 5).

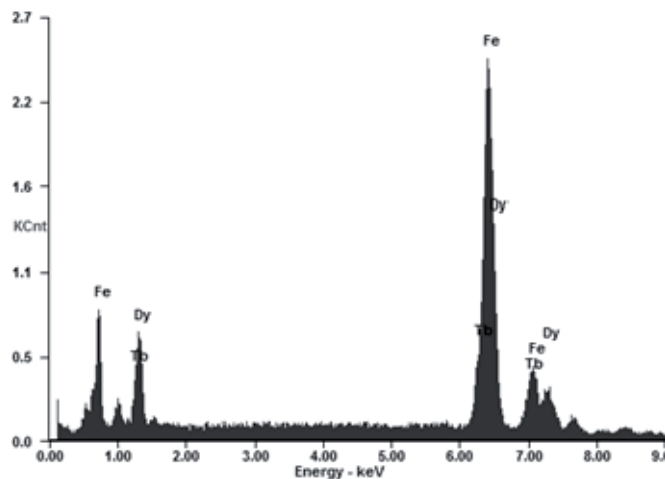


Fig. 11. X-ray energy dispersive spectrum for the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder

Table 5.

The result of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder quantitative chemical analysis

Element	Mass concentration of element, wt. %
Tb	17.2
Fe	41.8
Dy	41.0

Based on the X-ray diffraction analysis, it was found out that the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders (Fig. 12a), as well as composite materials reinforced by them (Fig. 12b) are composed of TbFe_2 and DyFe_2 magnetostrictive phases. Due to their isomorphism and small differences (at 0.01° level) between the values of the diffraction angles of reflection for the different phases, distinguishing them clearly by this research technique was difficult.

As a result of reinforcing the composite materials with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size distribution analyzed herein, slight changes of composite materials density have been found (in the range from 1.756 to 1.812 g/cm^3 for composite materials with 10% volume fraction of reinforcing phase), more and more noticeable along with the increase of the reinforcing phase share by volume in the matrix (Fig. 13).

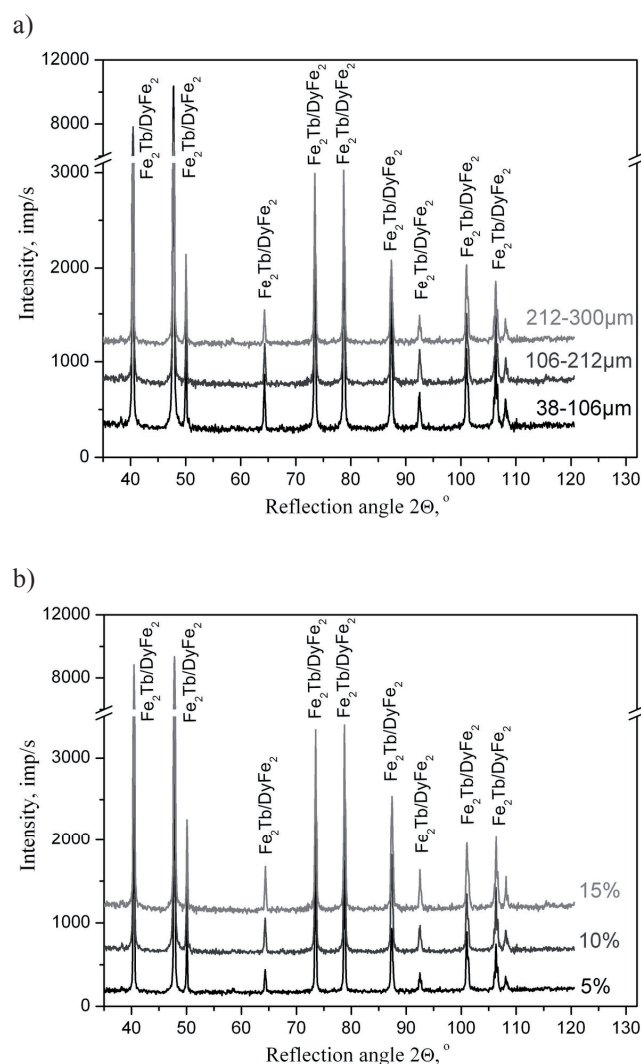


Fig. 12. Results of the X-ray phase analysis for the: a) $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder; b) composite materials with the polyurethane matrix; diffraction patterns were shifted along the vertical axis to show the results more clearly

Dispersion of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles in the polymer matrix obviously causes the reduction of density and increase of resistivity of the composites (Fig. 14), as compared to the monolithic alloy. The course of the current-voltage characteristics [56] is typical for dielectric materials. The high values of resistivity causes that the developed composite materials could be alternative for the monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$, in which eddy currents are induced during operation in magnetic fields with variable frequencies. The lowest resistivity value (for $U=200$ V) is

equal to $1 \text{ k}\Omega\cdot\text{m}$ and was recorded for the composite materials reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles in the range of $212\text{--}300 \mu\text{m}$ and with 20% share by volume; it is several times higher than for the monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ – which should be considered, however, in the context of dielectric losses related thereto, under operating conditions of these materials, causing heating of the polymer material [85].

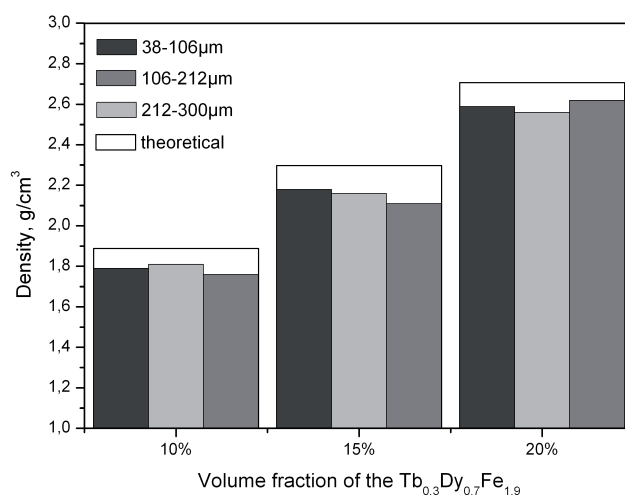


Fig. 13. Relationship between density and the volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder in composite materials

The thermal conductivity values of the composite materials oscillate from 0.221 W (for the composite material reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size in the range of $38\text{--}106 \mu\text{m}$ and 10% volume fraction) to 0.556 W (with 20% fraction of that powder) and – like the values of density – increase monotonically, along with the growth of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ share in the matrix. The changes results from higher density (9250 kg/m^3) and thermal conductivity ($13.5 \text{ W/m}\cdot\text{K}$) of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$, in comparison to the values for the polyurethane resin (1040 kg/m^3 and $0.035 \text{ W/m}\cdot\text{K}$, respectively). Composite materials thermal conductivity independency of temperature and magnetic field set during measurements with induction of 1 tesla (Fig. 15), proves the parameters are meaningless in relation to the operating properties of the materials developed. Therefore, the heat generated during their operation in fast-changing frequencies shall not significantly affect their thermal properties.

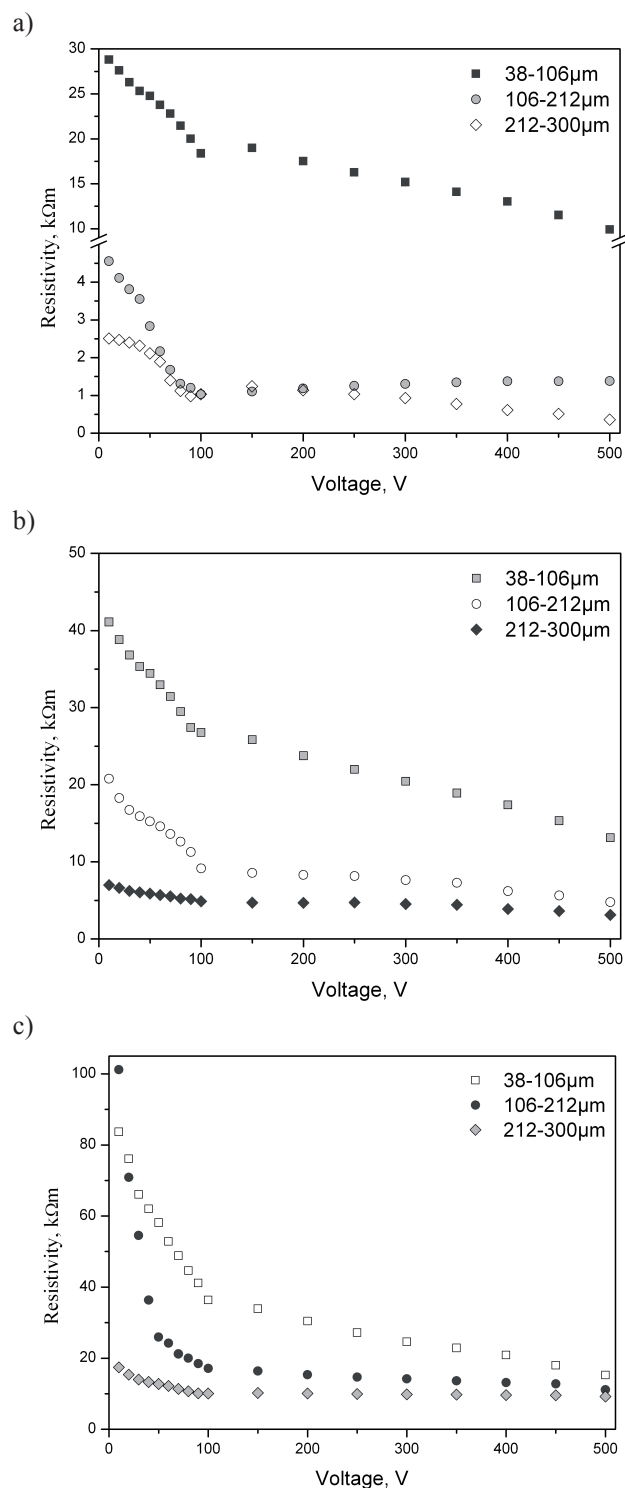


Fig. 14. The voltage dependence of resistivity for composite materials reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles of: a) 10 %; b) 15 %; c) 20 % volume fraction in polyurethane matrix

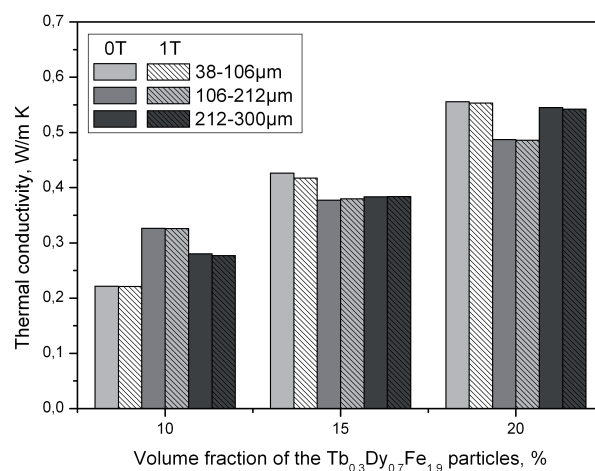


Fig. 15. Relationship between thermal conductivity and the volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder in composite materials

Low viscosity polyurethane resin – used as a matrix for the newly developed composite materials – makes $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles sufficient wetting. Moreover, the discontinuous connection between these two phases no exist, what was confirmed on the basis of images from the scanning electron microscope (Fig. 16). Thanks to proper coherence favorable energy transfer between boundary of the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles and the matrix is ensured. Moreover, with 20% fraction by volume of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ in the matrix, the magnetostrictive particles contact each other, which affects the increase of thermal and electric conductivity of the composite materials. No percolation paths for the materials with 10% fraction by volume of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ in the matrix causes that these properties – from the point of view the materials operation in fast-changing magnetic fields frequency – are worse. Such situation, however, does not affect the magnetic properties of the composite materials that result from the share of non-magnetic fraction causing dispersion of the external magnetic field.

In case of magnetic materials, the principal effect on deterioration of the properties, compared to their monolithic equivalents, is made by demagnetization and superparamagnetism resulting from the fact that – contrary to the monolithic materials that are generally characterized by a multi-domain structure – the nano-particles are single-domain. Simultaneously the particles with significant anisotropy of shape may maintain a single-domain structure with significantly larger diameters. The transfer from multi-domain to a single-domain structure is accompanied by a significant growth of coercive force H_c associated with a change of the magnetization reversal,

taking place for multi-domain particles, mainly through the domain wall movement. The increase of H_c value results from a greater energy input, necessary for generation of magnetic streams in small particles, therefore the dimension of magnetic domains is identical with that of the particles [11,15,86,87].

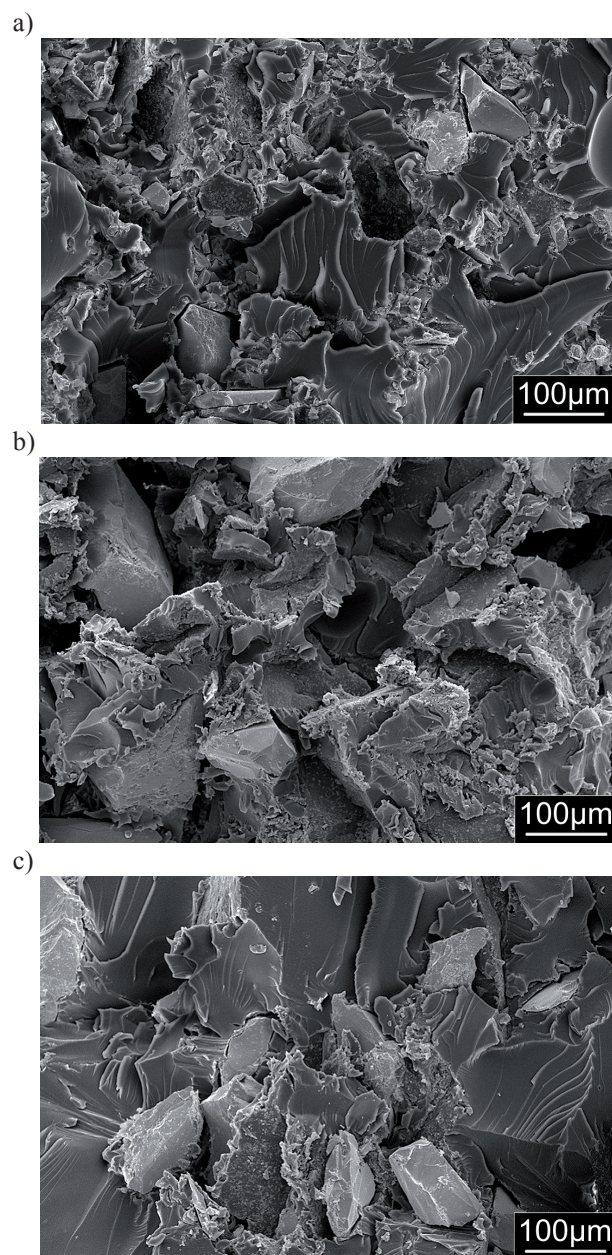


Fig. 16. Fractures of the composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder of 10% volume fraction in polyurethane matrix and particle size in the range of: a) 38-106 μm ; b) 106-212 μm ; c) 212-300 μm)

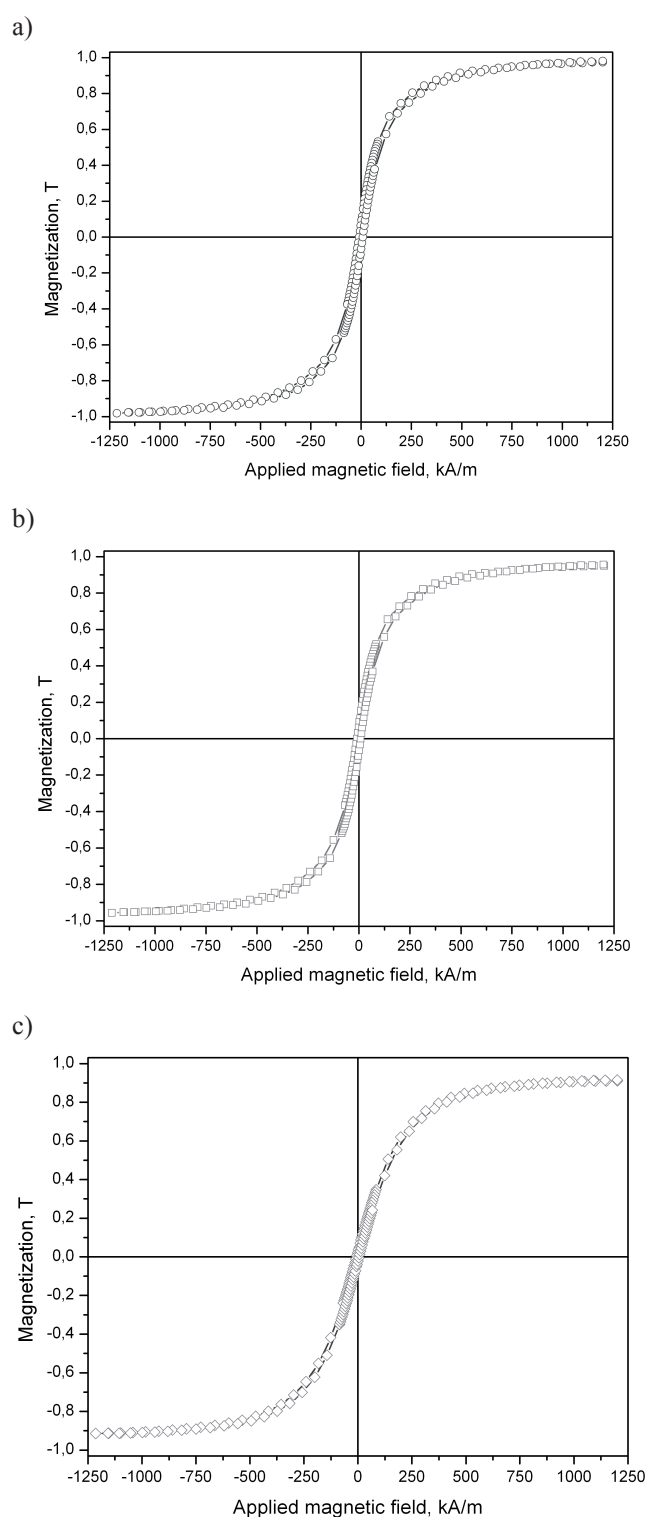


Fig. 17. Applied magnetic field dependency of magnetization for $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with particle size in the range of: a) 38-106 μm ; b) 106-212 μm ; c) 212-300 μm)

The research on the magnetic properties of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders with diverse particles size and composite materials with the polyurethane matrix (Fig. 17) indicate that the grain of the powders does not significantly affect the composite properties that – for particular volume fraction – are characterized with comparable magnetizing values and those of saturation induction and remanence. Based on the magnetic hysteresis loops plotted for the composite materials and $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders, it has been found that the saturation induction value scarcely depends on the size of the reinforcing particles. However, it should be noted that the value is lower for composites, in comparison to the values obtained for the specific fractions of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders and its reduction is strictly connected with the reduction of magnetic fraction in composite materials, in relation to the monolithic material.

The dependence of coercive force and remanence of composite materials on the size of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles has been presented in the Table 6. The magnetic properties of metal alloys in the form of powders mainly depend on their chemical composition, structure, internal stresses occurring during production and demagnetization coefficient, resulting from its geometry. Its value is directly affected by the material magnetic permeability: the higher it is, the larger is the value of demagnetization coefficient. Due to the fact that the magnetic permeability of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ used as reinforcement for the composite materials developed with polyurethane matrix is low, the coefficient was not considered in the analysis.

The value of coercive force for the composite materials mainly results from the structural defects arising during the production of powder. Small particles, due to their large

surface area, in comparison to their mass, have more surface defects than large particles, which affects the immobilization of domain walls during magnetization. Moreover, the fine granulation powder has high surface energy, which negatively affects the homogeneity of the reinforcement material distributions within the polymer matrix. The lowest coercive force H_c and remanence B_r values among the composites tested in case of composite materials reinforced by the powders with particle size range of 106-212 μm reflect their low anisotropy in room temperature.

The magnetization characteristics in the function of magnetic field strength corresponds with the magnetic permeability values of the composite materials developed. While analyzing the changes of this value (Fig. 18), attention needs to be paid to their characteristic course: starting with some initial value, the permeability grows fast to the maximum value and decreasing its value tends to the parallel course in relation to the axis of abscissae. Magnetic permeability μ_r reaches its maximum value for magnetic field strength equal to the coercive force, thus indicating a relatively easy 180° movement of domains in this range. Increasing the H value causes the reduction of permeability to the value corresponding to the condition in which the material becomes totally magnetized [14]. The use of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders as reinforcement for the composites causes that the permeability of the materials is significantly reduced in relation to the monolithic materials and is fixed on the level from 1.05 (for materials reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size in the range of 106-212 μm or 212-300 μm and 10 % volume fraction up to ca. 1.14 (for materials reinforced with particles with 20% volume fraction).

Table 6.

Comparison of characteristic magnetic properties of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders and composite materials

$\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders particle size range, μm	Volume fraction of powder, %	M_s , T	H_{\max} , kA/m	H_c , A/m	B_r , T	B_s , T	μ_r
38-106	10%	0.094	1230.03	3.67	0.007	1.626	1.07
	15%	0.136	1228.48	4.60	0.009	1.664	1.90
	20%	0.199	1224.59	5.39	0.013	1.723	1.13
	100%	0.981	1213.73	8.25	0.077	2.492	1.65
106-212	10%	0.079	1225.37	1.79	0.003	1.603	1.05
	15%	0.134	1224.59	2.44	0.006	1.659	1.09
	20%	0.196	1226.53	2.91	0.008	1.721	1.13
	100%	0.980	1224.59	4.61	0.029	2.509	1.64
212-300	10%	0.077	1226.92	2.55	0.005	1.602	1.05
	15%	0.131	1226.92	3.22	0.008	1.659	1.08
	20%	0.209	1226.92	3.71	0.011	1.738	1.14
	100%	0.910	1213.73	5.11	0.028	2.422	1.60

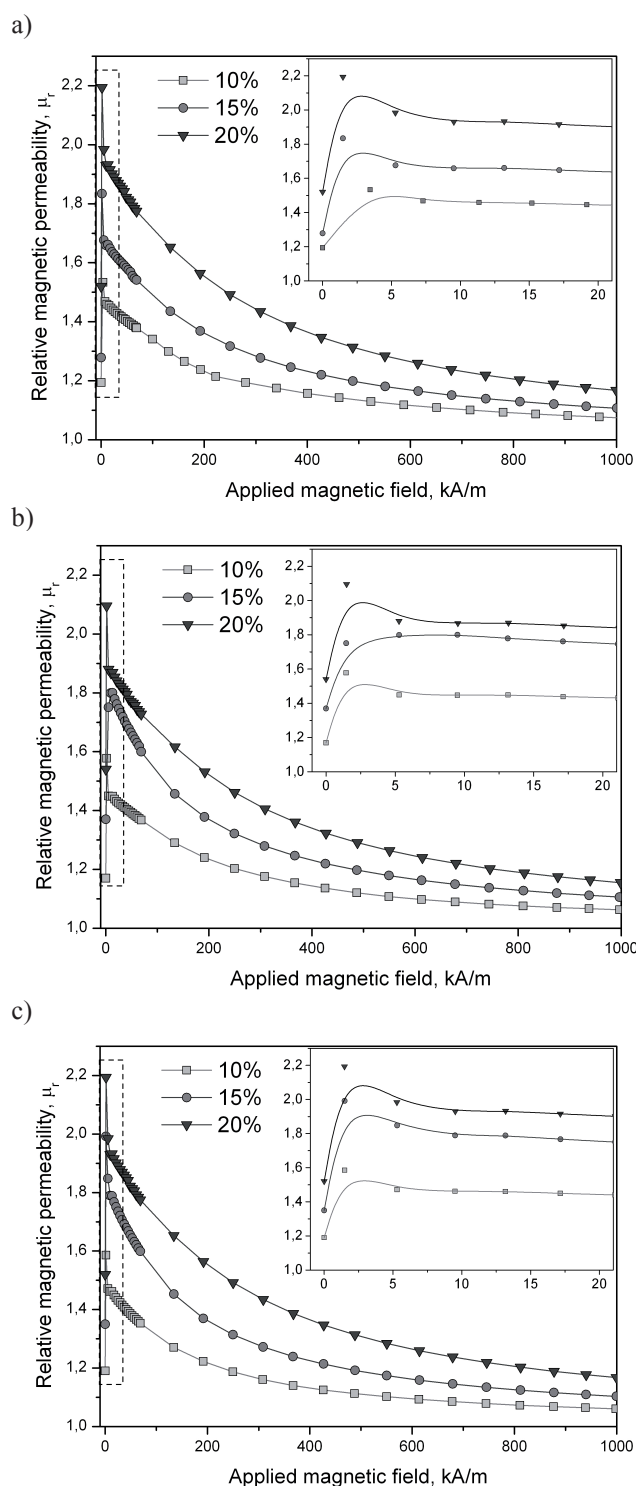


Fig. 18. The dependence of relative magnetic permeability μ_r of applied magnetic field for $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders and composite materials reinforced by particles with size in the range of: a) 38-106 μm ; b) 106-212 μm ; c) 212-300 μm)

Due to the fact that a magnetically neutral material was used as matrix for the magnetostrictive composite materials developed, the magnetic properties of such materials depend on the fraction of the reinforcing phase in the matrix and – to a lesser extent – on the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles dimension. It was found that the most favorable magnetic properties are obtained for composite materials with 20% volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size in the range of 106-212 μm , which equals: $H_c=2.91$ kA/m, $B_r=0.008$ T, $B_s=1.721$ T and $\mu_r=1.13$.

The courses of magnetization dependency on applied magnetic field (Fig. 17) converge with the diagrams of magnetostriction dependency on magnetic field strength (Fig. 19) for the particular composites, at the same time indicating value of the magnetic field for which the saturation occurs. It has been found that the composite materials do not reach saturation magnetostriction within the magnetic field range tested. Based on the magnetization characteristics in the magnetic field strength function one may draw a conclusion that the saturation would have been reached in a field of ca. 1000 kA/m. Reinforcing the magnetostrictive composite materials with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles with growing granulation causes the growth of magnetostriction, which mainly results from minimizing the demagnetization effect, that prevents deformation of a single particle by the resultant magnetic field.

The composite material with 20% volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder with particles size in the range of 38-106 μm reaches the highest magnetostriction value throughout the whole range of the magnetic field strength up to 766 kA/m, achieving its maximum value equal to $805 \cdot 10^{-6}$. The significant aspect is that the value is comparable to the minimum magnetostriction value declared by the manufacturer for the monolithic material.

It has been found that the maximum magnetostriction for the given composites depends on the grain size of the $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particle and their fraction in the matrix and its value for the magnetic field strength ~ 780 kA/m in case of the materials discussed oscillates in the range from $54 \cdot 10^{-6}$ (for the composites reinforces with powder with particle size in the range of 212-300 μm and 10% fraction by volume and with perpendicular orientation of the sample to the magnetic field line) up to $805 \cdot 10^{-6}$ (for materials reinforced with particles size in the range of 38-106 μm and 20% fraction by volume).

The magnetostriction measurements made with the perpendicular or parallel orientation of the samples tested in relation to the magnetic field direction, confirmed its

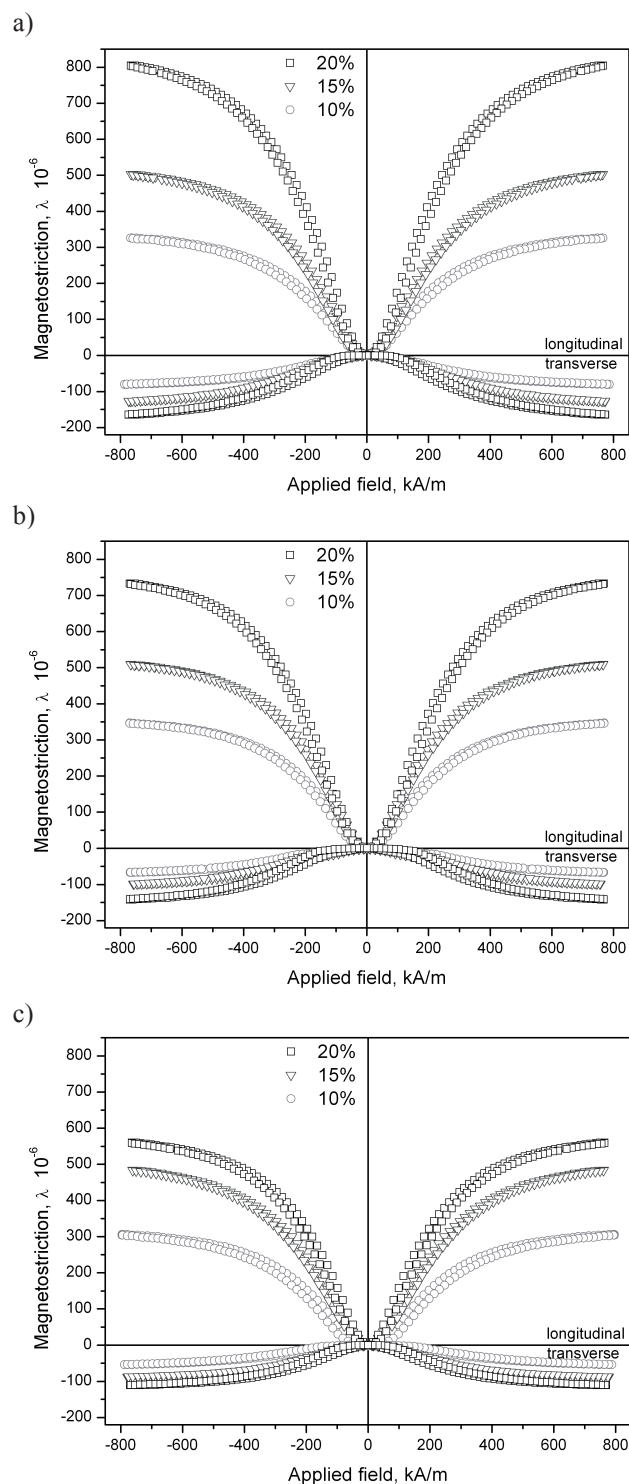


Fig. 19. Applied magnetic field dependency of magnetostriction for the composite materials reinforced by $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles with size in the range of: a) 38-106 μm ; b) 106-212 μm ; c) 212-300 μm)

effect on the magnetostriction loops recorded. In case of magnetostriction measurement applying the magnetic field with perpendicular orientation to the sample axis, the magnetostriction in each case reaches lower values than the measurements performed parallel to the sample axis (Fig. 20). The highest value for that orientation – equal to $165 \cdot 10^{-6}$ – was obtained for the composite reinforced with particles size in the range of 38-106 μm and their 20% fraction by volume. Within the analyzed magnetic field strength range (up to 800 kA/m), the lowest magnetostriction values have been reached for material with 10% fraction by volume of powder with particle size in the range of 212-300 μm , which is due to, without limitation, the large specific surface area of the reinforcing particles and their percentage fraction by volume in the matrix. In all composite materials tested, the growth of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders fraction causes the increase of magnetostriction, which is not only the result of enhanced share of the magnetostrictive phase, but also of the increased direct mutual contact of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles (Fig. 16), thus leading to a better energy transfer between the composite material components. This dependency also confirms that the main factor responsible for the magnetostriction value is the response of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles to the magnetic field action. The results of tests carried out as part of this study, in turn, confirmed that the interaction between the said particles and the matrix also represent a significant factor.

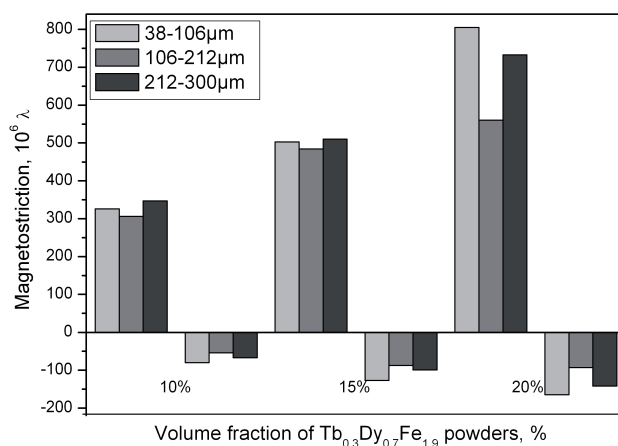


Fig. 20. Relationship between maximum magnetostriction and the volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powders in composite materials

5. Conclusions

Based on analysis of the investigation results for the newly developed magnetostrictive composite materials the following conclusions were made:

- The highest magnetostriction equals to $805 \cdot 10^{-6}$ (with magnetic field intensity of 766 kA/m) characterizes a composite material with polyurethane matrix, reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder of 20 % volume fraction and particles size in the range of 38-106 μm . Simultaneously, these materials are characterized with coercive force intensity $H_c=5.39$ kA/m, remanence $B_r=0.013$ T and magnetic permeability $\mu_r=1.13$. It was found that the maximum magnetostriction value for this material assumes values approximate to the magnetostriction of monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ alloy.
- It was confirmed that the correlation exists between the diversification of the volume fraction of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ powder in the matrix, its particle distribution and the maximum magnetostriction and magnetic properties of the developed composite materials, however, the impact in case of magnetization and saturation induction and remanence is limited. The values of coercive force intensity, permeability and susceptibility, in turn, are proportional to the fraction of the reinforcing material in the matrix and assume values approximate to those for the composite materials with polyurethane matrix reinforced with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ particles, with diversified granularity and corresponding volume fractions.
- It was also found that the key factor determining the energy transfer efficiency between $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ and matrix is – in addition to the size and volume fraction of the magnetostrictive particles in the matrix – the method of combining those two phases.

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