Properties of the magnetostrictive composite materials with the polyurethane matrix reinforced with Terfenol-D particles

L.A. Dobrzański a,b, A.E. Tomiczek a, A.W. Pacyna b
a Silesian University of Technology, ul. Konarskiego 18a, 44-100 Gliwice, Poland
b Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Sciences, ul. Radzikowskiego 152, 31-342 Kraków, Poland
* Corresponding e-mail address: leszek.dobrzanski@polsl.pl

Received 16.10.2012; published in revised form 01.12.2012

ABSTRACT

Purpose: The aim of this work is to obtain functional composite materials and to observe changes of magnetic properties of samples with different particle size distributions of magnetostrictive Terfenol-D (Tb0.3Dy0.7Fe1.9) powder. The influence of the concentration and particles size of the Tb0.3Dy0.7Fe1.9 on magnetic properties were investigated as function of applied magnetic field intensity, temperature and frequency.

Design/methodology/approach: The investigated samples were obtained by casting of the composite materials with the polyurethane matrix reinforced with Tb0.3Dy0.7Fe1.9 particles. Magnetizations versus applied field curves were registered using the Oxford Instruments Ltd. vibrating sample magnetometer (VSM). Volume magnetic susceptibility was determined as temperature function on the Cahn RG automatic electrobalance (Ventron Instruments, USA). Testing of the magnetic permeability in function of frequency was made using the Maxwell-Wien bridge system and the electrical properties were made by the resistivity measurements.

Findings: Analysis establishes a direct connection between physical properties and structural characteristics of the Tb0.3Dy0.7Fe1.9 powder size: the increases of particle size distribution of Tb0.3Dy0.7Fe1.9 powder in composite materials amplify the magnetic responses and - at the same time - causing growth of resistivity values also. Moreover, in the investigated frequency range, no effect was observed of frequency on the susceptibility value for the particular material, which suggests possibility of using these materials in the high-frequency magnetic fields.

Practical implications: The polyurethane matrix in investigated composite materials causes growth of resistivity, limiting these 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 Tb0.3Dy0.7Fe1.9 particles, with desired physical properties (including electrical one) in cost effective way in comparison to conventional giant magnetostrictive materials (GMM).

Keywords: Magnetostriction; Terfenol-D; Giant magnetostrictive materials; Composite materials

Reference to this paper should be given in the following way:
1. Introduction

The development of so-called Giant Magnetostrictive Materials began with tests using terbium - and later also samarium or gallium - with dysprosium compounds [1-3]. The materials with composition developed during the initial period presented high magnetostriiction, but only in magnetic fields of very high strength and/or in cryogenic temperature. The introduction of the RF₃₂ phase (where R is rare earth metal) enabled the shift of the Curie temperature of such materials above ambient temperature, while the magnetic field required for the magnetostrictive deformation was reduced by the appropriate selection of proportions between the concentration of terbium and dysprosium - elements with opposite magnetocrystalline anisotropy, which caused the compensation of the alloy effective anisotropy value close to zero in ambient temperature [4,5]. The discovery was the start of research on alloys defined by the general formula: TbₓDy₁₋ₓFeₙ (x = 0.27-0.3; y = 1.9-2), the wide group thereof was called „terfenols” (after the abbreviation of the elements composing them and the name of laboratory in which the discovery was made: TER-VE-Naval Ordnance Laboratory, USA).

The TbₓDy₁₋ₓFeₙ alloys are intermetallic phases with Laves C15 phase structure, very fragile in ambient temperature. Reaching a giant magnetostriiction in magnetic fields of low strength for low strength polycrystals is only possible when the grains are //₁₁₁ oriented (i.e. in the direction of easy magnetization), at the same time - this is the direction along which measurements are made, which is due to giant anisotropy of magnetostriiction (λ₁₁₁ // > λ₁₀₀). This is one of the factors causing that TbₓDy₁₋ₓFeₙ type alloys are usually produced by hard-to-perform methods of directional crystallization, which increases material costs. Furthermore, the production irregular form of materials is complicated and requires the application of precise machining at the final stage [6,7].

The advantages of the Tb₀.₃Dy₀.₇Fe₁.₉ include most of all the high value of the linear magnetostriiction, being 0.2%, the ability of non-contact carrying of loads from 500 to 600 MPa with very short signal response time (~1 μs in ambient temperature and <150 kA/m in low fields) - compared to the conventional materials. Moreover, these alloys are characterized with high reliability, large deformation energy density (~20 kJ/m³), low sound speed and high magnetomechanical coupling coefficient [8-10].

In spite of indubitable advantages of giant magnetostrictive materials (GMM), making them an important active module of executive devices, using them in industrial scale is limited by some factors [11-13]. The optimization of magnetostrictive alloys’ properties was made through synthesizing the materials with the strength of the specific elements. The tests proved that, in spite of the ability to combine high magnetostriiction with low anisotropy in these materials, the alloys hold defects characteristic for metal materials, including the induction of eddy currents, caused by the low resistivity of the material. Such factors restrict and sometimes even impede the application of the materials in transducers operating in power frequency magnetic fields [14-16].

It seems that an effective solution of the above issues is the production of a composite material with end properties requiring transmission of the specific effect from strain hardening to the matrix, which can be obtained through mechanical, electrical, optical, magnetic, thermal and/or chemical coupling between the components [17]. Polymer materials - thanks to their insulation properties and good adherence to the fiber or particle surface - provide the correct transmission of load onto the reinforcing phase in the composite materials and enable the formation of practically any shapes of the material. Furthermore, the polymer matrix produces non-conductive layers between the magnetostrictive particles, thus eliminating, thanks to increased resistivity, the losses for eddy currents in high frequencies of the transducer’s operation. The matrix also makes the resultant material more elastic and in consequence better adapted to operation in complex load conditions [18,19].

The first literature reports about the magnetostrictive composite materials with the polymer matrix appeared in 1994 [20] and in 1996 [21]. Since then there have been conducted intensive studies concerning to magnetostrictive composite materials with various polymer matrix [8,10,22].

The aim of this work is to investigate of magnetic and electrical properties of the newly-developed magnetostrictive composite material with polyurethane matrix.

2. Materials and methodology

2.1. Materials

Examinations were made on Terfenol-D (i.e. TbₓDy₀.₇Fe₁.₉ powder (Etrema Co, USA), demonstrating the giant magnetostriiction, as well as on samples of the composite material reinforced with them. The size of TbₓDy₀.₇Fe₁.₉ particles were denoted by their manufacturer as: 38-106 μm, 106-212 μm and 212-300 μm. The Smooth-cast 325 (Smooth-on Inc., USA) two-component polyurethane resin with low viscosity was used as the matrix in order to ensure sufficient particle wetting.

Composite materials with cylindrical shape were made by casting and for the particles sedimentation reasons, moving the mould continuously was required until the matrix gelation was completed. After cross-linking process finished, specimens were placed in an oven at 338 K for 5 h to ensure full cure of the resin. In this work, specimens containing particles volume fraction given in the Table 1 were made.

Table 1.

<table>
<thead>
<tr>
<th>Volume fraction of TbₓDy₀.₇Fe₁.₉, %</th>
<th>Mass fraction of TbₓDy₀.₇Fe₁.₉, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>48.1±0.5</td>
</tr>
<tr>
<td>15</td>
<td>60.1±0.5</td>
</tr>
<tr>
<td>20</td>
<td>68.1±0.5</td>
</tr>
</tbody>
</table>

2.2. Methodology

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 and afterwards - the remanence BR, permeability μmax, as well as coercive field Hc, quantities of powder and composites samples were determined.
Volume magnetic susceptibility $\chi$, as value specifying ability of given substance to change its magnetization, was determined as temperature function for composite materials and Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powders on the Cahn RG automatic electrobalance (Ventron Instruments, USA) with sensitivity of $10^{-7}$ cm$^3$/g and with Faraday’s method in the constant magnetic field $H_0 \sim 17$ kA/m, as well as in temperature range 50-350 K. For measurements taken in temperature range above ambient temperature the oven or for temperature range below 273 K - liquid helium flow cryostat were used. Moreover, magnetic susceptibility versus frequency dependence for Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powders and composite samples were registered using the Lake Shore 7225 AC susceptometer/DC magnetometer in the range up to 10 kHz.

Testing of the magnetic permeability in function of frequency was made using the measurement system whose main element was the alternating current Maxwell-Wien bridge (Agilent E498A Precision LCR Meter). During investigation the inductance, as well as resistance values of the induction coil with 31 turns were measured. Inside this coil the samples with cylindrical shape with diameter of 8.7 mm and the length of 40 mm has been placed. The measurement was taken for two cases: the first one when current intensity has variable values in the range 0.5-100 mA (for frequency equals to 1 kHz) and the second one - when current intensity has constant value equals to 50 mA and frequency was changed from 20 Hz up to 2 MHz. The measurements were carried out on the samples locating inside the coil with shackle, and the effective magnetic permeability were estimated based on the following relation:

$$\mu_{ef} = \frac{LI}{\mu_0 Z^2 A}$$

(1)

where:

- $L$ - the inductance of the coil, H;
- $I$ - the length, m;
- $\mu_0$ - the magnetic permeability of vacuum;
- $Z$ - the number of turns in the induction coil;
- $A$ - the cross-section area of transverse section of the rod, m$^2$.

Testing of electrical properties was made on stand consisting of the stabilized DC power unit, slide rheostat, ammeter, voltmeter, and sample holder. To ensure precise contact of the sample surface with electrodes, to ensure the homogeneous distribution of electrical charge, and 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. The resistivity values were estimated by using Ohm principle based on the following relation:

$$\delta = \frac{U}{I}$$

(2)

where:

- $\delta$ - the resistivity, $\Omega$ m;
- $I$ - the length of the sample, m;
- $A$ - the cross-section area of transverse section of the rod, m$^2$;
- $U$ - the voltage, V;
- $I$ - the current intensity, A.

3. Results and discussion

Basing on taken measurements of magnetic susceptibility $\chi$ as function of temperature, volume magnetic susceptibility $\chi$, as function of frequency in the range up to 10 kHz, magnetic permeability $\mu_\chi$ in function of frequency in the range of 20 Hz-2 MHz as well as magnetization versus applied field curves, the magnetic properties of the Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powders and the newly - elaborated composite materials has been assigned.

Based on the magnetization curves in the function of magnetic field intensities for Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powders and composite materials reinforced with them (Table 2), it has been found that - in case of composites - along with reduced ration of the Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ fraction in the matrix, the magnetisation changes and the saturation magnetisation $M_s$ assumes values approximate for the particular ratios of the reinforcement materials, changing in direct proportion to them: for composite materials with 10% Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ fraction concentration in polymer matrix and reaches highest $1213.73$ kA/m - for composite materials reinforced with powders of particle size of the range 38-106 µm and 10% volumetric ratio. The values of this level for composite materials reinforced with powders of particle size from 212 to 300 µm are distinguished by coercive field equal to 3.67 kA/m (with the 20% powder volumetric ratio) to 5.39 kA/m (with 20% Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder of particle size from the range 38-106 µm.

<table>
<thead>
<tr>
<th>Tb$<em>{0.3}$Dy$</em>{0.7}$Fe$_{1.9}$ powders grain size range, µm</th>
<th>Volume fraction of powder, %</th>
<th>$M_s$, T</th>
<th>$H_{max}$, kA/m</th>
<th>$H_c$, kA/m</th>
<th>$B_t$, T</th>
<th>$B_s$, T</th>
</tr>
</thead>
<tbody>
<tr>
<td>38-106</td>
<td>10%</td>
<td>0.094</td>
<td>1230.03</td>
<td>3.67</td>
<td>0.007</td>
<td>1.626</td>
</tr>
<tr>
<td></td>
<td>15%</td>
<td>0.136</td>
<td>1228.48</td>
<td>4.60</td>
<td>0.009</td>
<td>1.664</td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>0.199</td>
<td>1224.59</td>
<td>5.39</td>
<td>0.013</td>
<td>1.723</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>0.981</td>
<td>1213.73</td>
<td>8.25</td>
<td>0.077</td>
<td>2.492</td>
</tr>
<tr>
<td>106-212</td>
<td>10%</td>
<td>0.079</td>
<td>1225.37</td>
<td>1.79</td>
<td>0.003</td>
<td>1.603</td>
</tr>
<tr>
<td></td>
<td>15%</td>
<td>0.134</td>
<td>1224.59</td>
<td>2.44</td>
<td>0.006</td>
<td>1.659</td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>0.196</td>
<td>1226.53</td>
<td>2.91</td>
<td>0.008</td>
<td>1.721</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>0.980</td>
<td>1224.59</td>
<td>4.61</td>
<td>0.029</td>
<td>2.509</td>
</tr>
<tr>
<td>212-300</td>
<td>10%</td>
<td>0.077</td>
<td>1226.92</td>
<td>2.55</td>
<td>0.005</td>
<td>1.602</td>
</tr>
<tr>
<td></td>
<td>15%</td>
<td>0.131</td>
<td>1226.92</td>
<td>3.22</td>
<td>0.008</td>
<td>1.659</td>
</tr>
<tr>
<td></td>
<td>20%</td>
<td>0.209</td>
<td>1226.92</td>
<td>3.71</td>
<td>0.011</td>
<td>1.738</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>0.910</td>
<td>1213.73</td>
<td>5.11</td>
<td>0.028</td>
<td>2.422</td>
</tr>
</tbody>
</table>
The powder is also characterised with coercive intensity $H_c$ equal to 8.25 kA/m, saturation induction $B_s = 2.492$ T and remanence $B_r = 0.077$ T. The composite materials with polyurethane matrix, reinforced with this type of powder are distinguished by coercive field equal to 3.67 kA/m (with the powder volumetric ratio 10%) to 5.39 kA/m (with 20% volumetric ratio) and saturation induction equal to 1.626 T and 1.723 T, respectively. The increase of the size of Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder particles causes the decrease of the coercive intensity - the lowest value recorded is 1.79 kA/m (for the composite material reinforced with Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder with 106-212 µm granulation and 10% of its volumetric share in the matrix) and decrease of saturation induction $B_s$ to the minimum value recorded for the materials tested - equal to 1.603 T (for the composites reinforced with Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder with 106-212 µm, as well as 212-300 µm granulation and 10% of its volumetric share in the matrix). The measurements made enabled finding that the composite materials reinforced with Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ particles with finer granulation, i.e. 38-106 µm powder show lower magnetic properties. The saturation induction of materials in which the particles of such granulation represent 20% of the volumetric ratio is 1.723 T, with 0.013 T magnetic residue and 5.39 kA/m coercive. However, the composite materials of identical ratio of Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ particles with 106-212 µm granulation are characterised with coercive intensity equal to 2.91 kA/m, saturation induction equals 1.721 T and remanence of 0.008 T. In case of composite materials reinforced with powder within the particle size range from 212 to 300 µm, coercive intensity is in the ranges from 2.55 kA/m (composite materials of 10% Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder volumetric ratio) to 3.71 kA/m (with 20% reinforcement ratio volumetrically), while saturation induction is on the level 1.602 T and 1.738 T, respectively. The maximum intensities of magnetic field $H_{\text{max}}$ reach a slight diversification of values in the range from 1213.73 kA/m for powders of particle size of the range 38-106 µm and 212-300 µm to 1230.03 kA/m for composite material reinforced with Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ particles with 38-106 µm granulation and 10% volumetric ratio. The values of this level for composite materials reinforces with powders of particle size 212-300 µm are constant and are 1226.92 kA/m, while for the composite materials reinforced with powders of particle size from 38 to 106 µm, $H_{\text{max}}$ decreases along with the increase of the ratio of reinforcement material in the matrix, reaching the lowest - i.e. 1213.73 kA/m - value for the powder of this granulation.

Based on susceptibility versus temperature relationship curves the effect was analyzed of temperature on magnetic properties of Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powders and of the magnetostrictive composite materials. These characteristics (Fig. 1) have the non-linear character with the maximum susceptibility values obtained at the temperature of about 200 K. With the further temperature rise, magnetic susceptibility turn in lower values and at the ambient temperature (i.e. about 300 K), change of the susceptibility versus temperature relationship occurs; one can see that in the plot as the slightly ascending line segments. Introducing various concentrations of Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ magnetic particles into polymer matrix changes the flow of curve and character of the temperature relationship only slightly. Regardless of Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder particles size, along with lowering of the volume concentration of powders in the composite material, shifting of the curve is observed to the lower susceptibility values in the entire examination temperature range. This behaviour is mainly affected by the magnetic fraction concentration in the particular material.

It was found based on the volume magnetic susceptibility versus frequency measurements made (Fig. 2), that susceptibility of the composite materials is proportional to the Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ fraction concentration in polymer matrix and reaches highest values for materials reinforced with powder with particle size range 38-106 µm. The obtained results are being complementary to magnetic permeability dependence of frequency in the range 20 kHz to 2 MHz (Fig. 3).

**Fig. 1.** Susceptibility versus temperature relationship curve for composite materials reinforced with Tb$_{0.3}$Dy$_{0.7}$Fe$_{1.9}$ powder with granulation from 38 to 106 µm
The results of examination of the magnetic permeability dependence of frequency confirmed that permeability of the composite materials increase if the particle size decreases. The permeability value of composite materials with 20% volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder with particle size range 38-106 µm at frequency 50 Hz is equal to 1.24, whereas for composite materials with the same volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder, but with particle size range 212-300 µm, the effective permeability is equal to 1.16. It was also noticed that the susceptibility value would either rapidly decrease due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of several orders of magnitude or this transition will be broadened by several orders of magnitude or this transition will be broadened.

The composite materials tested are characterised with high resistivity depending on the Tb₃Dy₀.₂Fe₁.₉ powder 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 Tb₃Dy₀.₂Fe₁.₉ powder with particle size 212-300 µm and volumetric ratio of 20% to 30.5 kΩ·m (for 38-106 µm size Tb₃Dy₀.₂Fe₁.₉ and volumetric ratio 10%) (Fig. 4). Based on the recorded current-voltage characteristics it has been found that their course is typical for dielectric materials. With the increasing ratio of matrix in the composite materials, the weak contact between the Tb₃Dy₀.₂Fe₁.₉ particles causes high resistivity, while for composite materials with 20% ratio, the resistivity decreases due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of the composite materials produced. Compared to its value for monolithic Tb₃Dy₀.₂Fe₁.₉ (i.e. 58·10⁻⁸ Ω·m) causes that the materials developed represent an attractive alternative for them, which is of particular importance in the context of their application in magnetic fields of high frequency of magnetisation.

**Fig. 2.** Susceptibility - frequency relationship for the Tb₃Dy₀.₂Fe₁.₉ powders (a) and the magnetostrictive composite materials reinforced with Tb₃Dy₀.₂Fe₁.₉ powders with particle size: 38-106 µm (b), 106-212 µm (c), 212-300 µm (d)

The permeability value of composite materials increase if the particle size decreases. The permeability of composite materials with 20% volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder with particle size range 38-106 µm at frequency 50 Hz is equal to 1.24, whereas for composite materials with the same volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder, but with particle size range 212-300 µm, the effective permeability is equal to 1.16. It was also noticed that the susceptibility value would either rapidly decrease due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of several orders of magnitude or this transition will be broadened by several orders of magnitude or this transition will be broadened.

The composite materials tested are characterised with high resistivity depending on the Tb₃Dy₀.₂Fe₁.₉ powder 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 Tb₃Dy₀.₂Fe₁.₉ powder with particle size 212-300 µm and volumetric ratio of 20% to 30.5 kΩ·m (for 38-106 µm size Tb₃Dy₀.₂Fe₁.₉ and volumetric ratio 10%) (Fig. 4). Based on the recorded current-voltage characteristics it has been found that their course is typical for dielectric materials. With the increasing ratio of matrix in the composite materials, the weak contact between the Tb₃Dy₀.₂Fe₁.₉ particles causes high resistivity, while for composite materials with 20% ratio, the resistivity decreases due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of the composite materials produced. Compared to its value for monolithic Tb₃Dy₀.₂Fe₁.₉ (i.e. 58·10⁻⁸ Ω·m) causes that the materials developed represent an attractive alternative for them, which is of particular importance in the context of their application in magnetic fields of high frequency of magnetisation.

**Fig. 2.** Susceptibility - frequency relationship for the Tb₃Dy₀.₂Fe₁.₉ powders (a) and the magnetostrictive composite materials reinforced with Tb₃Dy₀.₂Fe₁.₉ powders with particle size: 38-106 µm (b), 106-212 µm (c), 212-300 µm (d)

The permeability value of composite materials increase if the particle size decreases. The permeability of composite materials with 20% volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder with particle size range 38-106 µm at frequency 50 Hz is equal to 1.24, whereas for composite materials with the same volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder, but with particle size range 212-300 µm, the effective permeability is equal to 1.16. It was also noticed that the susceptibility value would either rapidly decrease due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of several orders of magnitude or this transition will be broadened by several orders of magnitude or this transition will be broadened.

The composite materials tested are characterised with high resistivity depending on the Tb₃Dy₀.₂Fe₁.₉ powder 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 Tb₃Dy₀.₂Fe₁.₉ powder with particle size 212-300 µm and volumetric ratio of 20% to 30.5 kΩ·m (for 38-106 µm size Tb₃Dy₀.₂Fe₁.₉ and volumetric ratio 10%) (Fig. 4). Based on the recorded current-voltage characteristics it has been found that their course is typical for dielectric materials. With the increasing ratio of matrix in the composite materials, the weak contact between the Tb₃Dy₀.₂Fe₁.₉ particles causes high resistivity, while for composite materials with 20% ratio, the resistivity decreases due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of the composite materials produced. Compared to its value for monolithic Tb₃Dy₀.₂Fe₁.₉ (i.e. 58·10⁻⁸ Ω·m) causes that the materials developed represent an attractive alternative for them, which is of particular importance in the context of their application in magnetic fields of high frequency of magnetisation.

**Fig. 2.** Susceptibility - frequency relationship for the Tb₃Dy₀.₂Fe₁.₉ powders (a) and the magnetostrictive composite materials reinforced with Tb₃Dy₀.₂Fe₁.₉ powders with particle size: 38-106 µm (b), 106-212 µm (c), 212-300 µm (d)

The permeability value of composite materials increase if the particle size decreases. The permeability of composite materials with 20% volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder with particle size range 38-106 µm at frequency 50 Hz is equal to 1.24, whereas for composite materials with the same volume fraction of the Tb₃Dy₀.₂Fe₁.₉ powder, but with particle size range 212-300 µm, the effective permeability is equal to 1.16. It was also noticed that the susceptibility value would either rapidly decrease due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of several orders of magnitude or this transition will be broadened by several orders of magnitude or this transition will be broadened.

The composite materials tested are characterised with high resistivity depending on the Tb₃Dy₀.₂Fe₁.₉ powder 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 Tb₃Dy₀.₂Fe₁.₉ powder with particle size 212-300 µm and volumetric ratio of 20% to 30.5 kΩ·m (for 38-106 µm size Tb₃Dy₀.₂Fe₁.₉ and volumetric ratio 10%) (Fig. 4). Based on the recorded current-voltage characteristics it has been found that their course is typical for dielectric materials. With the increasing ratio of matrix in the composite materials, the weak contact between the Tb₃Dy₀.₂Fe₁.₉ particles causes high resistivity, while for composite materials with 20% ratio, the resistivity decreases due to the conducting properties of the reinforcement material. The resistivity values correspond to the losses on eddy currents of the composite materials produced. Compared to its value for monolithic Tb₃Dy₀.₂Fe₁.₉ (i.e. 58·10⁻⁸ Ω·m) causes that the materials developed represent an attractive alternative for them, which is of particular importance in the context of their application in magnetic fields of high frequency of magnetisation.
Properties of the magnetostrictive composite materials with the polyurethane matrix reinforced with Terfenol-D particles

4. Conclusions

An important factor - especially from a point of view of newly elaborated composite materials as an final control elements for example in active intelligent systems - is necessity of taking into consideration an influence of frequency and temperature on their properties in operating conditions. Higher temperature indispensably accompanies changeable magnetic field, and high frequencies in particular - as a result eddy currents causing energy losses are being induced in conductive materials also. In order to verify chosen selection of materials with regards for application, complementary research has been made regarding magnetization versus applied field curves, susceptibility in a function of temperature as well as susceptibility and permeability in function of frequency.

By considering the magnetic permeability \( \mu_r \) in dependence of frequency, it can be assumed that in lower frequency permeability of fine \( \text{Tb}_0.3\text{Dy}_{0.7}\text{Fe}_{1.9} \) powders (i.e. from the range 38-106 \( \mu m \)) attains higher values then for the coarse ones (i.e. for particle size in the range 106-212 \( \mu m \) and 212-300 \( \mu m \)). Although this tendency is confirmed in literature reports, there is no unequivocal explanation this phenomena. In that cases - according to data from literature [23,24] - the natural resonance frequency of composite materials reinforced with fine particles is lower than for the coarse ones. One can suppose that this phenomena is connected with dispersion of permeability, which may originated from [23,25]:

- the domain walls resonance (vibrating Bloch’s walls due to the force acting on walls in the presence of high frequency external ac magnetic field),
- the natural ferromagnetic resonance (the forced precession of magnetization vectors in domains due to the presence of effective magnetic anisotropy),
- the relaxation of magnetization.

Moreover, it was also noticed that the values of permeability for particular composite materials is nearly constant in investigated frequency range. Increasing of extorting signal frequency up to 2 MHz causing that magnetic permeability for composite materials decrease of about 0.01 \% which suggest possibility of using newly-developed composite materials as intelligent actuators and sensors working in the high-frequency magnetic fields. In such cases, one of the indispensable factor accompanying to fast-changing magnetic field is raised temperature - its influence onto magnetic properties of \( \text{Tb}_0.3\text{Dy}_{0.7}\text{Fe}_{1.9} \) powders, as well as magnetostrictive composite materials have been analyzed on the base of magnetic susceptibility versus temperature curves. These curves have the non-linear character with the maximum susceptibility values obtained at the temperature of about 200 K. With the further temperature rise up to 273 K, these curves reach local minimum responding to anisotropy compensation. In the temperature range near 300 K, the change of the susceptibility versus temperature relationship occurs; one can see that in the plot as the slightly ascending line segments. Taking into account that thermal vibration accompanying to rising of temperature cause destroying of ordered spin magnetic moments, which leads to decay of magnetic domains, one may suppose that at the Curie point (i.e., 653K) which is the ferromagnetic/paramagnet transition temperature, the susceptibility value would either rapidly decrease by several orders of magnitude or this transition will be broadened within the temperature range. It was also noticed that decreasing of volume fraction of reinforced powders in matrix - irrespective of their size - causing lowering of magnetic susceptibility-temperature curve.

Results show that - thanks to the fact that the magnetically indifferent material has been used as the matrix - the magnetic properties of those composites depend on volume fraction of reinforcement and - to a lesser extend - on \( \text{Tb}_0.3\text{Dy}_{0.7}\text{Fe}_{1.9} \) particles size. The best results (i.e. \( H_c = 2.91 \) kA/m, \( B_r = 0.008 \) T and \( B_t = 1.721 \) T) were obtained for the composite materials reinforced with 20 % volume fraction of \( \text{Tb}_0.3\text{Dy}_{0.7}\text{Fe}_{1.9} \) powder with particle size range from 106 to 212 \( \mu m \).
Acknowledgements

Research was financed partially within the framework of the Scholarship No 51201116 of the International Visegrad Fund realized by Anna Tomiczek, Ph. D., Eng.

References


[10] A. Tomiczek, Magnetostrictive composite materials with the polymer matrix reinforced with Tb0.3DY0.7Fe1.95 particles, Ph.D. Thesis - unpublished, Main Library of the Silesian University of Technology, Gliwice, 2012 (in Polish).


[15] J. Liu, W. Ren, D. Li, N. Sun, X. Zhao, J. Li, Z. Zhang, Magnetic transitions and magnetostrictive properties of TbDy1-x(Fe0.8Co0.2)2 (0.20≤x≤0.40), Physical Review B 75 (2007) 064429.

[16] X. Zheng, P. Zhang, F. Li, Z. Cheng, B. Shen, A magnetic, magnetostrictive and Mössbauer study of Tb0.3Dy0.7-xPrx(Fe0.9A10.1)1.95 alloys, Journal of Magnetism and Magnetic Materials 321/23 (2009) 3842-3846.


