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MAGNETOMECHANICAL PROPERTIES OF COMPOSITE MATERIALS WITH GIANT MAGNETOSTRICTION

WŁASNOŚCI MAGNETOMECHANICZNE MATERIAŁÓW KOMPOZYTOWYCH O GIGANTYCZNEJ MAGNETOSTRYKCJI

The aim of this work was to observe the changes in the magnetomechanical properties of composite materials with different $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ (Terfenol-D) powder particle-size distributions and varying volume fractions in the polyurethane matrix. The results show a direct relationship between the properties and the particle size of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder: the increases in the particle-size distribution of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder in the matrix amplify the magnetostrictive responses and the compressive modulus values. Moreover, it was found that the key role in efficiency of the transformation of magnetic energy into mechanical plays the initial compressing pre-stress.

Keywords: Giant Magnetostrictive Materials, Terfenol-D, Magnetomechanical properties, Composite Materials

Celem pracy jest określenie zmian własności magnetomechanicznych materiałów kompozytowych o zróżnicowanej wielkości cząstek $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ oraz zmiennym udziale wzmocnienia w osnowie poliuretanowej. Otrzymane wyniki wskazują na bezpośrednią zależność pomiędzy własnościami a rozmiarem cząstek $Tb_{0.3}Dy_{0.7}Fe_{1.9}$: zwiększenie ich wielkości powoduje wzrost wartości magnetostrykcji oraz modułu ściskania. Stwierdzono ponadto, że kluczową rolę w efektywnej transformacji energii magnetycznej w mechaniczną odgrywa naprężenie wstępne.

1. Introduction

All over the world intensive research is being conducted into the increasing efficiency of magnetostrictive transducers, which are an attractive alternative to piezoelectric materials, mainly because of the size of the gained deformations and forces. Constant aspiration to obtain the most effective energy transformation possible has become a basic factor causing the development of a materials group, among which $Tb_xDy_{1-x}Fe_y$ ($x = 0.27 \div 0.3$; $y = 1.9 \div 2$) alloys, which are distinguished for their giant magnetostriction, are remarkable. The type of material most commonly used today is $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ (Terfenol-D), which saturation magnetostriction for particular directions reaches $\lambda_{[111]} = 1640$ ppm, $\lambda_{[100]} = 9$ ppm and $\lambda_{[112]} = 1200$ ppm [1-3]. Moreover, in this material it is possible to obtain deformations equal to up to 2000 ppm, which is feasible thanks to creating compressive pre-stress (≥ 20 MPa) [4,5]. After application of a magnetic field, such working conditions enable complete rotation of the magnetic domains; without compressive pre-stress the material magnetization process takes place through a 90° domain wall motion [6-8].

The advantages of magnetostrictive particle dispersion into the polymer matrix include – among others – a wider transducer frequency band, lower brittleness, the possibility of adjusting the properties for specific application and the competitive price of the composite materials in comparison to monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ [9-11]. Proper selection of the matrix material affects the characteristics of the composite materials: it has been established that the properties of these materials are highly dependent on the matrix properties, the volume fraction of the reinforcing particles and their distribution [12-14]. In [15] an opinion has been presented that the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder parameters (particle size and shape) have inconsiderable influence on the static and dynamic properties of the composite material. Further research has proven this state and since then study has focused mostly on obtaining the maximum magnetostriction in the function of the composite material components: the size [10, 15-17] and shape [17-20] of the magnetostrictive particles, as well as the matrix properties [9, 10, 17, 18, 21, 22].

The goal of this work is the preparation of magnetostrictive composites and the investigation of the particle size and concentration in the polymer binder response to the magnetic and magnetomechanical properties.

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2. Experimental details

Composite materials of the polyurethane matrix (Smooth-on Inc., USA), reinforced with 10%, 15% and 20% $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ (Terfenol-D) powder by volume (Etrema Co., USA) were fabricated by casting. Samples differing in particle size distribution, denoted by their manufacturer as 38-106 μm (powder A) and 106-212 μm (powder B), were made.

The particle-size distribution of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder was analysed using a Analysette 22 Microtec Plus (Fritsch) analyser and observation of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle's structure were undertaken using a Zeiss SUPRA 35 scanning electron microscope (SEM). Examination of the magnetic properties was carried out on an Oxford Instruments Ltd. vibrating sample magnetometer (VSM) and the remanence B_r, as well as the coercivity H_c quantities of the powder and composite samples were determined, based on the measured magnetization and the applied field intensity values.

The influence of the pre-stresses on the values of the magnetostriction λ for the composite materials was verified on a materials testing machine MTS 810, which was equipped with a coil. The sample, with a glued optical Fiber Bragg Grating (FBG) sensor registering the sample elongation, was placed inside this coil. The measurements were taken in the directions parallel to the sample axis, at room temperature and according to the procedure described in [9]. The frequency of the measured signals was below 100 Hz under magnetic fields up to 175 kA/m. Measurements for the non-stressed state, as well as for the compressive pre-stresses, equal to 1, 2 or 3 MPa were registered.

Compression tests were undertaken on a MTS 810 machine at room temperature and for different values of the magnetic field intensity (equals to 0, 52.5, 105, and 165 kA/m) in order to estimate the compression modulus and evaluate the influence of the magnetic field intensity on these values.

3. Results and discussion

Fig. 1 shows the morphology of the particles observed using a scanning electron microscope. The significant difference in the shape and size of the particles, which are irregular in the entire range, was observed. Moreover, a tendency for powder agglomeration was noticed (Fig. 1(A)). The results of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle-size distribution are shown in Fig. 2; powder with granulation determined as A is clearly characterized by particles with the smallest dimensions, which is confirmed by the median and quantile 0.9 values, equal to 39.86 μm and 105.73 μm respectively. The particle size range for A powder varies from 1.44 to 240 μm. The most coarse particle-size distribution with the median value of 176.66 μm is for powder denoted as B, which was shown of Fig. 1 (B). Moreover, the size distribution of the B powder is characterized by two peaks which could be explained by the fact that the powder is a mixture of particles with a largely differentiated size (from 1.91 to 550 μm). It should be noted also that 90% of particles have a diameter smaller than 313.01 μm ($q_{0.9}$). The results presented correlate well with accumulated surface value (S_{KUM}), a higher value (0.26 m^2/g) which is obtained for finer (i.e. A) powder. The properties of the magnetostrictive composite materials obtained should be considered in a context of full particle-size distribution – one may suppose that composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with the same granulation but different static distribution parameters might not show analogous physical and mechanical properties.

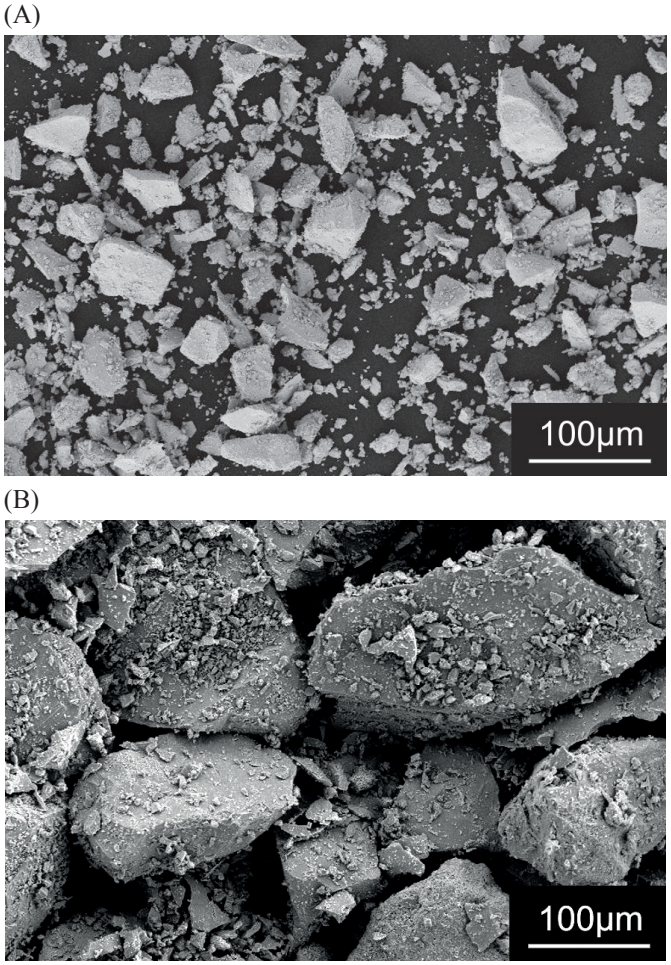

Fig. 1. Morphology of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders with granulation: 38-106 μm (A) and 106-212 μm (B); SEM

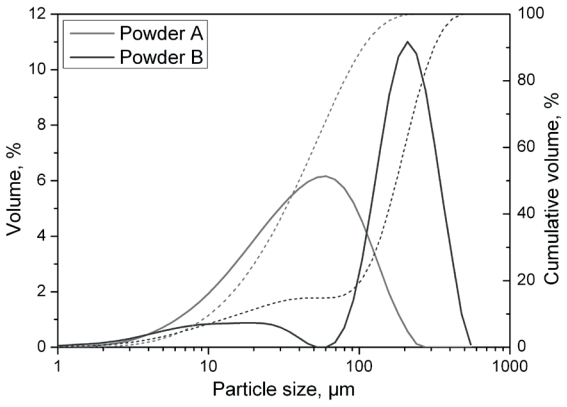

Fig. 2. Particle-size distribution of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powders used in the experiments

The magnetic property investigations (Fig. 3) indicate that A powder is characteristic of coercivity H_c equal to 8.25 kA/m and remanence B_r = 0.077 T. Composite materials with a polyurethane matrix reinforced with this powder (10%A) have coercivity in the range from 3.67 kA/m to 5.39 kA/m (20%A) and the remanence equals 0.007 T and 0.013 T, respectively. The larger particle size causes the higher coercivity – the lowest registered H_c value is 1.79 kA/m (for 10%B) and the decrease in remanence to the minimal registered value for analysed materials equals 0.003 T. The measurements taken show that composite materials reinforced with the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with finer granulation (10%A, 15%A and 20%A) have inferior magnetic properties: the remanence of 20%A material equal 0.013 T with coercivity of 5.39 kA/m, while composite materials with identical concentrations of B particles (20%B) show coercivity equal to 2.91 kA/m and remanence 0.008 T.

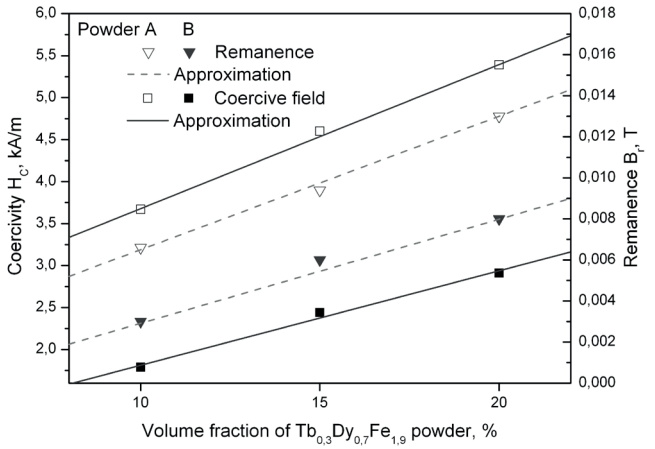

Fig. 3 Dependence of coercivity and remanence of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder concentration and its granulation in the composite materials

The characteristics describing the influence of the magnetic field's intensity changes on the magnetostriction of the analysed composite materials with a volume fraction of 20% are presented in Figs 4 and 5 and comparison of the initial stress (σ_0) influence on the magnetostriction of those materials are presented in Fig. 6.

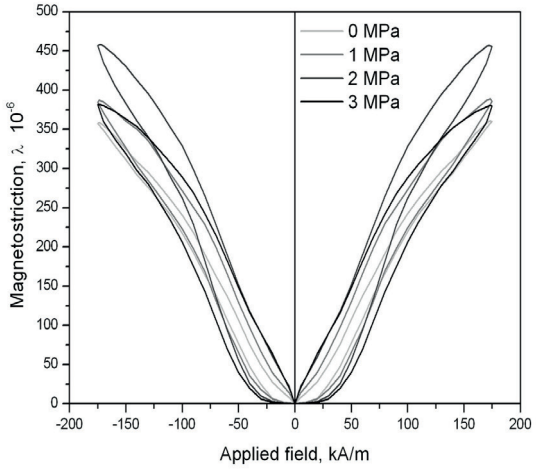

Fig. 4 Magnetostrictive dependence of the magnetic field intensity of the composite materials with the polyurethane matrix reinforced with 20% volume fraction of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with granulation A

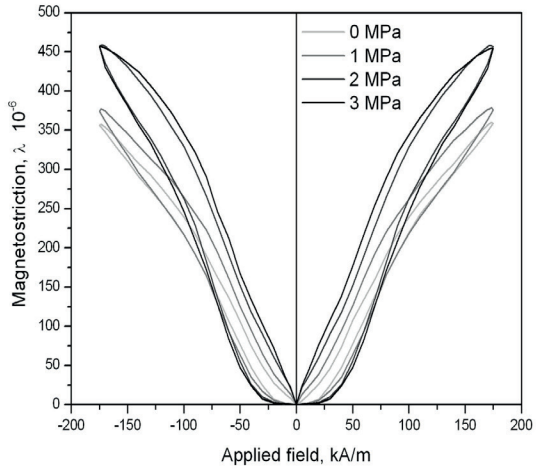

Fig. 5. Magnetostrictive dependence of the magnetic field intensity of the composite materials with the polyurethane matrix reinforced with 20% volume fraction of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with granulation B

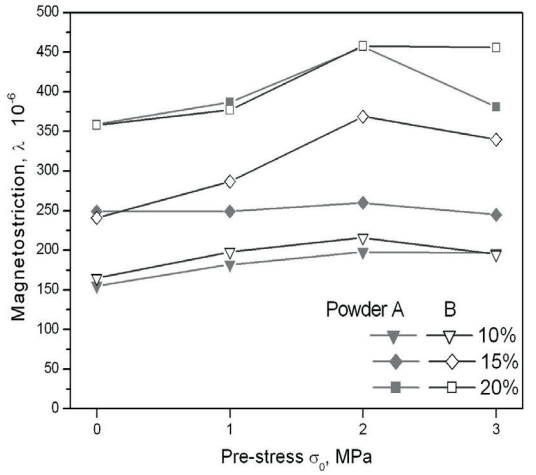

Fig.6. Comparison of the compressive pre-stress influence on magnetostriction for the composite materials

For σ_0 = 0, the highest magnetostriction in the magnetic field intensity up to 175 kA/m reaches values close to a particular concentration of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ in the matrix, attaining the highest rate for materials with a 20% volume fraction in the reinforcing phase. At the same time, decreasing the magnetostriction in the composite materials with the same contribution of the B powder has been stated to correspond to the share of the materials reinforced with A particles. In the considered range of magnetic field intensity and for σ_0 = 0, the lowest magnetostriction value equalling approximately $155 \cdot 10^{-6}$ has been reported for composite materials reinforced with the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with volume fraction of 10%.

Initial stress has a favourable influence on the magnetostriction of newly-elaborated materials in comparison with its values for measurements taken without its application. Increasing the pre-stress values causes increasing magnetostriction as well – this phenomena is caused by magneto-elastic interaction; however, for σ_0 > 2 MPa the influence of pre-stress on the magnetostriction value becomes

less important. In the case of the composite materials reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles marked B, a more definite increase in magnetostriction has been reported for σ_0 = 2 MPa (max 64 % compare with magnetostriction values without application of the pre-stress), than for composite materials reinforced with the A powders (for which the difference equals 28% accordingly). Application of 3 MPa pre-stress causes that magnetostriction to increase less significantly than for σ_0 = 2 MPa.

Applying pre-stress has a beneficial influence on the magnetostriction values, particularly in low-intensity magnetic fields. From the practical point of view, activation of a given transducer can, in that case, be more effective thanks to minimizing the losses occurring in the coil. Taking into account that the $\lambda = f(H)$ curve is non-linear and shows hysteresis – in order to obtain optimal transduction – both field values and uniaxial initial stresses ought to be properly chosen [1,3, 21].

Comparison of the compression modulus of newly-elaborated magnetostrictive composite materials with the polyurethane matrix (Fig. 7) revealed that the compression modulus values significantly depend on the volume fraction of the reinforcing material and – to a lesser degree – on the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles size, which is particularly noticeable for the B powders. In the case of these materials, the compression modulus has a value of 0.77±0.02 GPa for 10%B samples and approx. 0.8 ±0.04 GPa for 15%B and 20%B. For the composite materials reinforced with A powders, the lowest values of the compression modulus were obtained, reaching a level of 0.75±0.02 GPa (10%A). It has been established that increasing the volume fraction of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder in the matrix, causes an increase in the compression modulus.

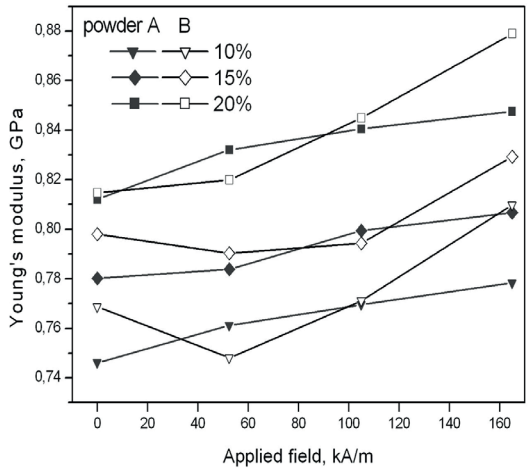

Fig. 7. Comparison of compression modulus as a function of magnetic field intensity for composite materials reinforced by different granulations of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder

Comparison of the compression modulus' dependence on the magnetic field intensity of values equal to 52, 105 and 165 kA/m made it possible to conclude that its influence on the compression modulus of composite materials is insignificant (average level of 5%), while the lowest values for particular samples were reported for measurements taken without a magnetic field, the highest with the given field of intensity equal to 165 kA/m (20%B). The maximum change in the compression modulus – in the range from 0.81±0.08 GPa (for H = 0) to 0.88±0.02 GPa (for H = 165 kA/m) has been given for the composite material reinforced with $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles with B granulation and a volume fraction of 20%.

4. Conclusion

In this paper, the magnetic and magnetomechanical properties of the composite materials with the polyurethane matrix reinforced by $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder with varying particle sizes, as well as its volume concentration, have been investigated. The 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 the volume fraction of the reinforcement and – to a lesser extent – on the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle size. The best results (i.e. H_c = 2.91 kA/m, B_r = 0.008 T) were obtained for the 20%B materials.

In the case of the elaborated composite materials, applications of pre-stress result in increasing the magnetostriction from 387 $\cdot 10^{-6}$ at σ_0 = 1 MPa (materials reinforced with 20% volume fraction of reinforcement) to approximately 458 $\cdot 10^{-6}$ at σ_0 = 2 MPa. The main reason for this phenomenon is the crosslinking of polyurethane material used as a matrix which – thanks to that – contracts and inducts mechanical compressive stress onto the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles causing the privileged irreversible 180° domain wall motion as the external initial compressive stress in the monolithic material [7, 16-17]. By decreasing the volume fraction of $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ in the composite material, i.e. by increasing axial stress, more domains would orientate perpendicularly to the magnetic field. It has been stated that the minimal value of pre-stress should average 2 MPa in order to orientate domains appropriately – magnetic moments for this value (under the influence of a magnetic field) change their orientation between crystallographic directions, i.e. from perpendicular to a direction close to axial [4].

It can be seen that increasing the magnetic field intensity up to 175 kA/m has an influence on the magnification of the pre-stress causing greater distortion and decreasing the slope of the magnetostriction in the function of the applied magnetic field intensity. When pre-stress used to rotate the magnetic domains exceeds the critical value of 2 MPa, the dependence of the magnetostrictive distortion versus magnetic field intensity lessens (Fig. 5). For pre-stresses below that value, in some cases, the 180° domain wall motion has occurred and – by this – the magnetization process without distortion takes place (as in the application of compressive pre-stress at a level of 28 MPa for monolithic material) [7, 9]. Moreover, increasing the compression stress values for particular composites causes a growth in the area of the magnetostrictive hysteresis loops, showing more and more magnetomechanical damping. At the same time, higher non-stability of the measurement taken has been revealed, affecting the high standard deviation values.

As a result of using the matrix for newly developed composite materials such as the polyurethane resin, whose compression modulus is significantly lower than its value for monolithic $Tb_{0.3}Dy_{0.7}Fe_{1.9}$, the main significance in transferring the tension is attributed to the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particles. The compression modulus of the composites increasing

is a function of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ particle volume fraction and this dependence is more distinctive for the composite materials reinforced by the B powder. Although the high value of the compression modulus obtained for composites with a 20% volume fraction of reinforcement is desirable in some applications, it is also important to limit the concentration of the non-magnetic matrix in order to assert the high efficiency to volume ratio of the magnetostrictive material.

Newly developed magnetostrictive composite materials are characterized by favourable magnetomechanical properties. High values of magnetostriction and the possibility of adjusting them in a wide range – depending on the volume fraction of the $Tb_{0.3}Dy_{0.7}Fe_{1.9}$ powder in the matrix, as well as its particle-size distribution – indicate the possibility of utilizing these materials as the final control elements of intelligent actuators and sensors.

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