Polish Society of Composite Materials
Polskie Towarzystwo Materiałów Kompozytowych

12: 3 (2012) 210-215

COMPOSITES THEORY AND PRACTICE

Jerzy Kaleta, Michał Królewicz, Daniel Lewandowski*, Michał Przybylski, Piotr Zając

Wroclaw University of Technology, Institute of Materials Science and Applied Mechanics, ul. Wybrzeze Wyspianskiego 27, 50-370 Wroclaw, Poland
**Corresponding author. E-mail: daniel.lewandowski@pwr.wroc.pl*

Received (Otrzymano) 16.02.2012

SELECTED MAGNETOMECHANICAL PROPERTIES OF MAGNETORHEOLOGICAL ELASTOMERS WITH THERMOPLASTIC MATRICES

The goal of the paper is to present the process of preparing magnetorheological composite materials with thermoplastic matrices called magnetorheological elastomers (MRE) and the possibilities of modelling them using Kevin-Voight relations. At the beginning, the MRE components are presented: a specific thermoplastic elastomer matrix material and ferromagnetic fillers. Both polymer and iron particles are mixed together in specific proportions on the basis of of calculations of the Critical Particle Volume Concentration. Moreover, the method of mixing and the process parameters are described in this paper as they are crucial to achieve the desired structure of the composite. The next important step in testing the MR elastomer is preparation of the sample. At the beginning, the material samples are cut and mounted in such away as to be able to test the influence of magnetic and mechanical fields. Such material is cut into pieces and glued between claddings that support it during testing. The material is put simultaneously under the influence of a high temperature and pressure in a magnetic field in order to achieve an anisotropic structure. Tests performed on a hydraulic pulsator MTS allowed us to expose the MRE to various conditions, which gives better understanding of the behaviour of the material. On the basis of the results, it is possible to observe the viscous and elastic properties of the material and thanks to that, model it and simulate its behaviour, which is presented at the end of this paper.

Keywords: **smart magnetic materials, magnetomechanical cross-effects, viscoelastic model, magnetorheological composites**

WYBRANE WŁAŚCIWOŚCI MAGNETOMECHANICZNE KOMPOZYTÓW MAGNETOREOLOGICZNYCH O OSNOWIE Z POLIMERÓW TERMOPLASTYCZNYCH

Celem pracy jest przedstawienie procesu przygotowania, badania i modelowania kompozytowych materiałów magnetoreologicznych z termoplastyczną osnową nazywanych magnetoreologicznymi elastomerami (MRE). Zaprezentowano proces wykonywania z wykorzystaniem konkretnych typów materiału osnowy elastomeru termoplastycznego i wypełniacza ferromagnetycznego. Zarówno polimer, jak i cząstki żelaza są mieszane ze sobą w określonych proporcjach ustalonych na podstawie obliczeń. Procesy mieszania i polaryzowania zostały pokazane, ponieważ mają zasadnicze znaczenie dla osiągnięcia pożądanej struktury kompozytu. Kolejnym ważnym krokiem było przygotowanie próbek i ich badanie. Próbki materiału były wycinane oraz mocowane w taki sposób, by uzyskać możliwość badania wpływu pola magnetycznego i mechanicznego. Tworzono strukturę próbki z dwóch elementów materiału i trzech elementów mocujących. Wykorzystywano technikę klejenia. Taki sposób mocowania umożliwił badanie w stanie zbliżonym do czystego ścinania. Próbki po przygotowaniu badano na przystosowanym do tego stanowisku, na którym możliwe było realizowanie cyklicznych obciążeń. Wyniki badań realizowano w postaci rejestracji sygnałów naprężenia i odkształcenia dla zadanej wartości zewnętrznego pola magnetycznego. Na podstawie wyników możliwe było zaobserwowanie lepkich i sprężystych właściwości materiału. Z wykorzystaniem prostego modelu materiału Kevina-Voight wykonano próbę zamodelowania badanego materiału. Wyznaczono wartości parametrów modelu dla poszczególnych pętli histerezy. Zaprezentowano porównanie pętli rzeczywistych i obliczonych na podstawie modelu.

Słowa kluczowe: **materiały magnetyczne „smart", efekty krzyżowe magnetomechniczne, model lepkosprężysty, kompozyty magnetoreologiczne**

INTRODUCTION - RESEARCH GOAL

Magnetorheological composites with solid elastomer matrices (nonporous), also called magnetorheological elastomers (MRE), is a new group of smart materials. They are characterized by a reversible change of specific mechanical and rheological quantities under an external magnetic field. Such a phenomenon is known as the magnetorheological effect. MR elastomers have a working range in the pre-yield region, unlike magnetorheological fluids (MRF) which normally work in the post-yield regime. Typical magnetorheological elastomers consist of two components: a non-magnetic polymer matrix and a ferromagnetic active filling (soft magnetism). Thanks to the cross-linked polymer structure, the filler particles are not able to move freely.

There are many materials which might be used as the matrix of an MRE. The most common are silicone or natural rubbers. The filler particles are usually made of ferromagnetic materials such as carbonyl iron. Both the shape and size of the magnetically active elements have a major influence on the properties of the whole MR composite. Most often, spherical carbonyl iron particles with a diameter of several micrometres are used.

Based on previous papers and previously conducted research, the need to test the possibilities of modelling magnetorheological elastomers was stated. The key tasks were defined as the following:

- to design and prepare the manufacturing procedure of magnetorheological elastomers
- to select components: matrix material, magnetically active particles and additives
- to prepare MRE samples and use the constructed test stand for experimental examinations
- to verify the possibilities of modelling MRE with a simple Kevin-Voight model.

Magnetorheological elastomer material - matrix

A typical magnetorheological elastomer is composed of a ferromagnetic powder, an elastomer matrix bonding the powder, and additives to improve the mechanical properties of the material. The elastomer matrix is one of the two most important parts of the material as it determines the mechanical properties of the elastomer. So far, several types of materials have been tested as matrices for MRE: rubber (silicone, natural, acrylonitrile, isobutylene-isoprene, acrylonitrile-butadiene), silicone gels and modifications of the aforementioned rubbers.

Almost all of those rubbers are cross-linked which means covalent bonds between the polymer chains have been created and they cannot be easily recycled and formed again. This is a huge disadvantage of those materials that can be overcome by using a thermoplastic elastomer, which can be plasticized by heating the material to form it again in any needed shape. In theory, there should be no change in the properties after each recycling of the material, however, it has not been tested yet.

As there was a need for a soft thermoplastic elastomer with good mechanical properties, Téfabloc TO.222 30A produced by CTS Cousin-Tessier cts was chosen as the best match. This is a TPE-S polymer that, according to the manufacturer, combines the parameters of a block copolymer SBS (styrene-butadiene-styrene) and hydrogenated block copolymer SEBS (styrene-ethylene-butadiene-styrene). Its features are: resistance to acids, bases, detergents, oxidation and ozone; good resistance to chemicals, UV and weather; good thermal stability. This material is also food contact acceptable. Téfabloc's hardness varies from 30 to 80 a (on the Shore scale), the softest variation of this material was chosen as it should give a good magnetorheological effect in the material matrix.

Ferromagnetic powder

Magneto-active particles are the second most important component of magnetorheological elastomers. They determine the magnetorheological properties of the composite. On the basis of tests performed by J. David Carlson and Mark R. Jolly [1], the two most important characteristics of the ferromagnetic powder used for MRE can be defined as magnetic permeability and the density of magnetic induction. Those parameters are responsible for the forces acting between the particles, and by increasing those parameters, the attraction between the particles inside the magneto-active material can be increased. Soft ferromagnetic materials (iron) best meet those parameters and that is why it is most commonly used as an active material in magnetorheological elastomers. The shape and size of the particles are also very important. The experiments performed by Martin Lokander [2] showed that an irregular shape and the size of tens of micrometers give the best magnetorheological effect in the material.

a) b)

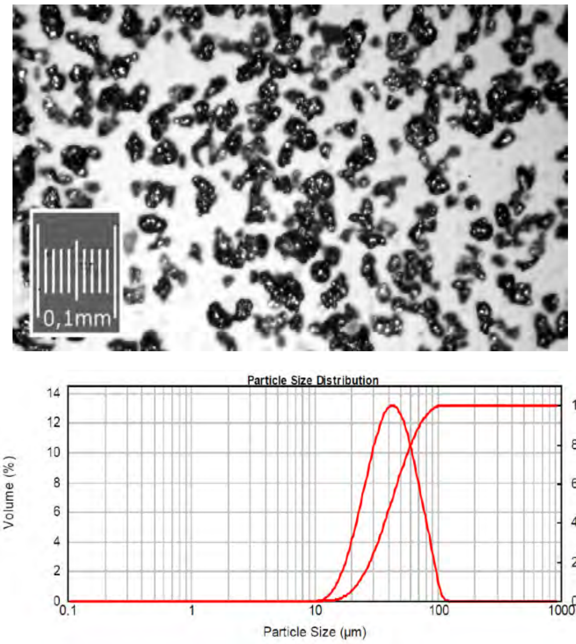

Fig. 1. Picture of Höganäs AB ASC 300 iron particles taken using light microscope (a) and size distribution of particles (b) (Mastersizer 2000, thanks to help of Faculty of Chemistry at Wroclaw University of Technology)

Rys. 1. Zdjęcie żelaza ASC 300 firmy Höganäs AB wykonane przy użyciu mikroskopu świetlnego (a) i rozkład wielkości cząstek (b) (Mastersizer 2000, dzięki pomocy Wydziału Chemii Politechniki Wrocławskiej)

Ferromagnetic particles used to produce MRE were chosen on the basis of previous experiments performed in the Institute of Material Science and Applied Mechanics I-19 where this paper was prepared. ASC 300 iron made by the Swedish company Höganäs AB was chosen as the one that best fulfils the requirements described above. According to its manufacturer, the size of the particles should be about 60 μm and they have an irregular shape.

Setting the composition

The key to producing a good magnetorheological elastomer lies in the proportion of the matrix and filler material. To find the proportion called the Critical Particle Volume Concentration (CPVC) [3], equation 1 has to be applied. The bulk density of the ASC 300 iron powder given by the manufacturer is equal to 2.88 g/cm^3 and the density of iron is equal to 7.87 g/cm^3, which gives a CPVC = 36.6%. Considering the result, to maximize the magnetorheological effect in the composite, the volume of magnetoactive particles should be close to the CPVC value. In previous tests conducted by the author, MREs were made with 28 to 35% ASC 300 iron powder with various additives. The best result was achieved for the magnetorheological elastomer with 35% iron powder with paraffin oil [4], which is close to the CPVC value. CPVC formula for iron:

$$CPVC_{Fe} = \frac{\rho_n \cdot 100\%}{\rho_{Fe}}$$

After taking into consideration all of that information, the composition of the magnetorheological elastomer was set as presented in Table 1.

TABLE 1. **Chosen composition of MRE**
TABELA 1. **Wybrany skład MRE**

Element	Téfabloc TO..222 30A	Iron powder ASC 300	Paraffin oil
Weight	20.8 g	125 g	5.2 g

PREPARATION OF MAGNETORHEOLOGICAL ELASTOMERS

Each test sample of the magnetorheological elastomer was prepared in three stages. The first one was to create the material, then it had to be formed into samples and polarized. In the end, the samples were glued in between laminate plates that held them together during the tests.

Preparation of the material

To prepare the magnetorheological elastomer, the elastomeric matrix has to be mixed with the ferromagnetic powder and other additives at a proper temperature and using the right amount of force. To obtain the right conditions for mixing, the Plasti-Corder Lab-Station made by the Brabender company was used. The progress of the mixing was presented on the screen of the computer as a graph of torque in time (Fig. 2), thanks to which the time of feeding of the additives could be determined.

In Figure 2, each point presents the moment of feeding additives to the mixer. At the beginning of the process, the matrix material was poured into the mixing chamber, which caused an increase in torque up to point *A*. Then it stabilized up to point *B* when the iron powder was added. At this point, the torque on the rotors rapidly increased up to point *C* where it reached its maximal value and again it stabilized up to point *D* where the paraffin oil was added to the mixing chamber. The end of the process is represented by point *E* and from the graph it can be read that the whole process took about 8 minutes and that the temperature inside the chamber during the process was about 160÷170°C, which was enough to plasticize and mix the polymer matrix with the additives. After the mixing was done, the prepared material was removed from the chamber and left aside to cool down.

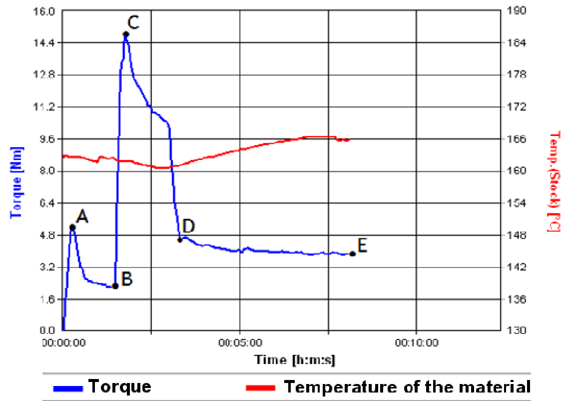

Fig. 2. Plastogram of MRE mixture (points presented in graph are described in text)

Rys. 2. Plastogram z mieszania MRE (punkty przedstawione na wykresie są opisane w tekście)

Preparation of test samples

The next step was to form the material into 40x40x4 mm cubic pieces of MRE to glue them between the claddings. To do so, the material had to be cut into small pieces and placed into a form. The material was heated under light pressure for 2 minutes at a temperature of 190°C to plasticize the elastomer and after that it was formed under pressure at a high temperature for another 2 minutes. The still hot material was placed together with the form between two permanent magnets to cool down and solidify. The magnets that were used had a magnetic field vector perpendicular to the surface of the magnets and generated a magnetic field *H* of about 400 kA/m. The samples were left in such conditions until they cooled down and then they were taken out of the form. The pieces of MRE that were taken out of the form were not ready to be glued into the claddings as they were too large and had rough edges. To prepare the samples to be glued into the claddings, they had to be cut into cuboids (Fig. 3a). Each pair of samples cut out of one piece of MRE were glued to glass-epoxy laminate TSE-5 claddings. The gluing was performed using Loctite 406 glue. Each side of the sample was glued to the cladding and left under slight pressure for 24 hours to bind properly. Figure 3b presents a completed test sample.

a)

b)

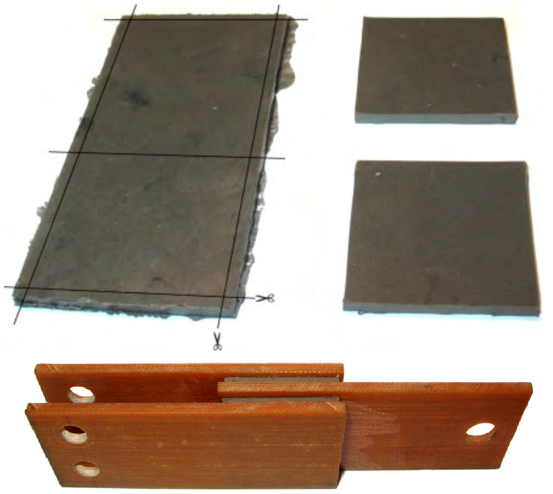

Fig. 3. Piece of MRE taken from form and samples cut out (a), test sample, which is composed of two pieces of magnetorheological elastomer glued between three glass-epoxy laminate claddings (b)

Rys. 3. MRE wyjęte z formy i wycięte próbki (a), próbka testowa, wykonana z 2 próbek elastomeru magnetoreologicznego wklejonego pomiędzy okładziny z laminatu szklano-epoksydowego Internal structure of anisotropic MRE (b)

Magnetorheological materials are composite materials. To present the internal structure of an MRE, very detailed pictures of its structure were taken using a scanning electron microscope (SEM) (Fig. 4a and 4b).

a)

b)

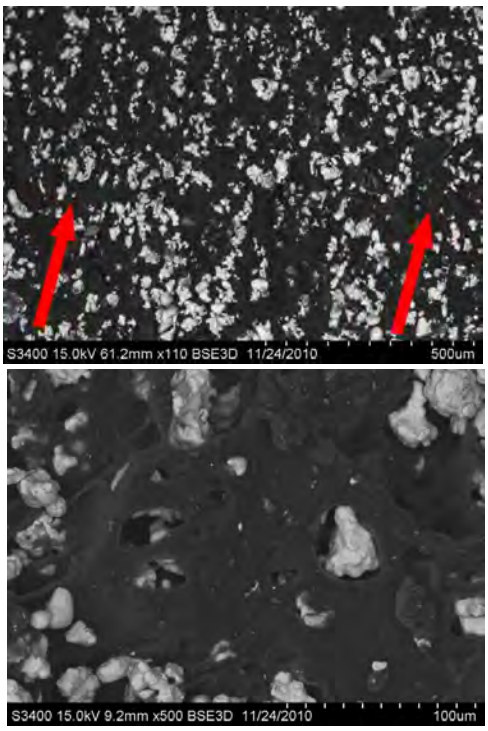

Fig. 4. Internal structure of anisotropic magnetorheological elastomer: a) aligned particles inside material with direction of polarization presented by red arrows and, b) presents close up of MRE structure (thanks to help of Labolatory of Sol-Gel Materials and Nanotechnology)

Rys. 4. Wewnętrzna struktura anizotropowego elastomeru magnetoreologicznego: a) uszeregowane cząstki wewnątrz materiału, z kierunkiem polaryzacji zaznaczonym przez czerwone strzałki, b) zbliżenie na strukturę MRE (dziki pomocy Laboratorium Materiałów Zol-Żelowych i Nanotechnologii)

Test stand

According to previous tests performed in previous papers [4], the most appropriate method of testing MRE samples is the oscillatory shear test performed on a hydraulic pulsator. Figure 5 presents the scheme of a test sample with forces acting on it during a shear test. As can be seen, the outer claddings are fixed and only the middle one moves and transfers force to the MRE samples. In addition, the magnetic field generated by the magnetic coil acts perpendicularly to the surface of the samples.

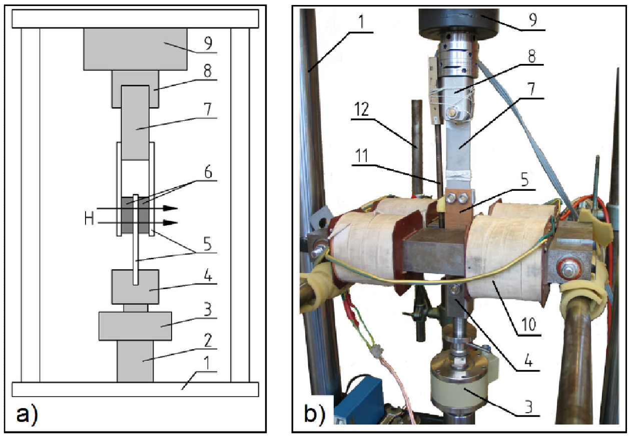

Fig. 5. Scheme of test stand (a), picture of test stand (b), where: 1) hydraulic pulsator MTS, 2) pulsators piston, 3) force sensor PCB, 4, 8) clamping jaws, 5) claddings, 6) magnetorheological elastomer, 7) distancing element, 9) force sensor MTS, 10) magnetic coil, 11) Hall probe, 12) frame

Rys. 5. Schemat ukłądu pomiarowego (a), zdjęcie stanowiska badawczego (b), gdzie: 1) pulsator hydrauliczny MTS, 2) tłok pulsatora, 3) czujnik siły PCB, 4, 8) szczęki, 5) okładziny, 6) elastomer magnetoreologiczny, 7) elementy dystansujące, 9) czujnik siły MTS, 10) cewka magnetyczna, 11) sonda Halla, 12) rama

TESTING OF MRE

The tests of the magnetorehological elastomer presented in this paper done on an MTS were conducted according to a program with changes in the magnetic field and strain amplitude with a constant frequency of 1 Hz. An example of the executed test program is presented in Figure 6.

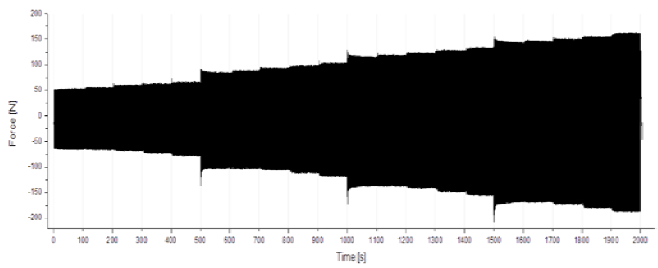

Fig. 6. Example of executed test program on test stand

Rys. 6. Przykład wykonanego programu badań na układzie pomiarowym

It presents the force signal in relation to time. Every 500 seconds, a significant change in force can be observed as the amplitude of deformation is increased by 6.25 x 10^{-3} with every leap. Within each amplitude, smaller changes of force can be observed as every 100 seconds the magnetic force is increased by 25 kA/m.

MODEL OF MAGNETORHEOLOGICAL ELASTOMER

To describe the behaviour of the magnetorheological elastomers, a basic model and its parameters have to be determined or calculated. To find the parameters of the model, a program written in HP Vee Software (Agilent) was used, which is based on the equations of an viscoelastic body. As the input data for the program, a single load cycle from the data obtained during tests was used and calculations were performed for each sample of material.

To present the approximated values of the parameters model and how they correspond to real data, stress-strain hysteresis loops were created presenting real and calculated data according to an equation describing the Kelvin-Voigt model.

In order to show the difference between the values of real and approximated values of elasticity and viscosity, graphs were prepared to present the stress values obtained during the tests and the stress calculated on the basis of approximated parameters.

Figures 7 and 8 present the real and approximated hysteresis graphs of stress in relation to strain for the maximal and minimal amplitude of displacement as well as the maximal and minimal values of the magnetic field used during the tests.

Figure 7a presents two hysteresis loops of real and approximated stress for the maximal amplitude of deformation used during tests without the magnetic field. The second graph shown in Figure 7b presents the same sample but under the influence of a magnetic field equal to 100 kA/m.

Figure 8 also presents the hysteresis loops of stress in relation to strain where Figure 8a presents the situation without a magnetic field and Figure 8b presents the situation with a magnetic field equal to 100 kA/m, but those two graphs represent the situation where the amplitude of deformation is the smallest used during the tests.

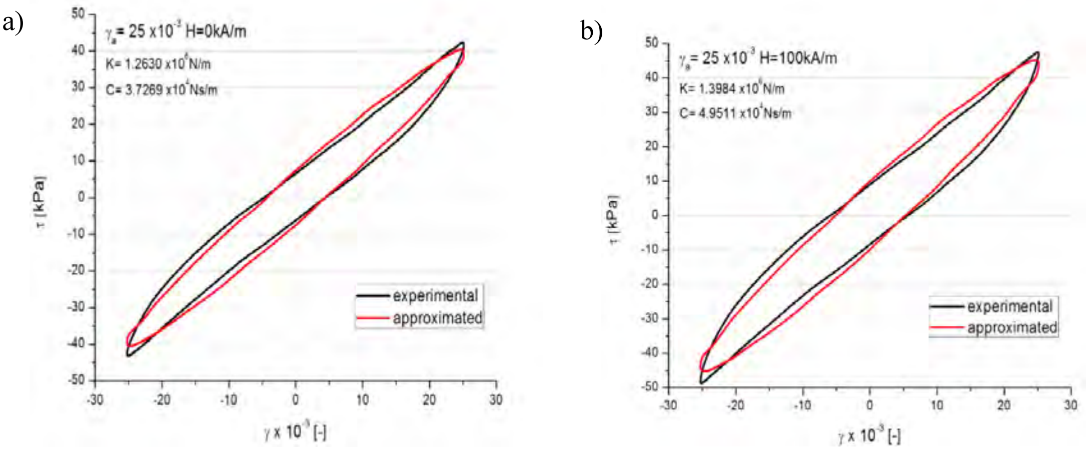

Fig. 7. Hysteresis loops of experimental and approximated values of stress in relation to strain

Rys. 7. Pętle histerezy dla wyników eksperymentalnych i przybliżonych wartości naprężeń w stosunku do odkształceń

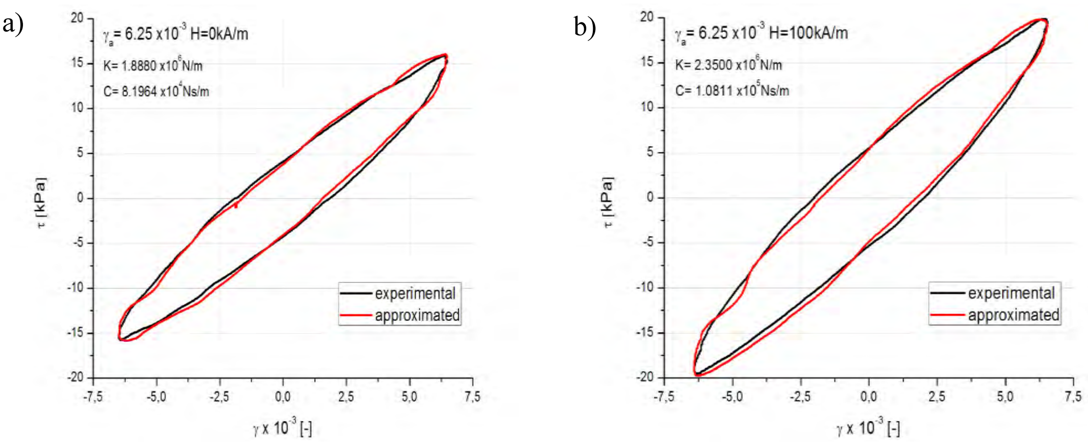

Fig. 8. Hysteresis loops of experimental and approximated values of stress in relation to strain

Rys. 8. Pętle histerezy dla wyników eksperymentalnych i przybliżonych wartości naprężeń w stosunku do odkształceń

As can be easily observed, the hysteresis loops presenting the stress signal under a magnetic field are much bigger than those presenting the stress signal of the sample without the influence of a magnetic field. This presents the magnetorheological effect that can be observed in MR elastomers. However, what is even more important, the hysteresis loops presenting the approximated values of stress in all the four examples are very similar to the real ones, which means that the approximation was quite accurate. The shape of the approximated loops is not identical to the real ones, especially the ends of the loops differ a lot. The approximated loops were calculated on the basis of the displacement signal after the noise reduction described earlier in this section. If they had been calculated on the basis of the displacement signal perfectly sinusoidal, those loops would have had a perfectly elliptical shape and would have corresponded much better to the real loops.

SUMMARY

The main objective of this work was to present the process of preparing magnetorheological composite materials and show their basic magnetomechanical properties. The manufacturing procedure for these materials also has been presented. The material for the matrix (Téfabloc thermoplastic elastomer) and the magnetically active particles (ASC 300 iron with 60 μm particles) were used. Samples with polarized (anisotropic) particle alignments were fabricated. The specimens were investigated using a specially designed test stand. The magnetorheological elastomers were subjected to cyclic shearing with a constant frequency of 1 Hz. The tests were conducted at various strain amplitudes with the application of a magnetic field. The results were presented in the form of hysteresis loops. The influence of the magnetic field and strain amplitude on the shape and size of the loops has been noticed. The possibilities to model the tested material with a simple viscoelastic model has been presented. The parameters of the model such as elasticity and viscosity have been calculated and the results as well as the hysteresis loop were compared to the experimental data.

Acknowledgement

The research was supported by the Wrocław Research Centre EIT+ within the project "The Application of Nanotechnology in Advanced Materials" - NanoMat (POIG.01.01.02-02-002/08) financed by the European Regional Development Fund (Innovative Economy Operational Programme, 1.1.2).

REFERENCES

[1] Carlson J.D., Jolly M.R., MR fluid, foam and elastomer devices, Mechatronics 2000, 10, 555-569.

[2] Lokander M., Performance of Magnetorheological Rubber Materials. PhD thesis, KTH, Stockholm 2004.

[3] Kallio M., The elastic and damping properties of magnetorheological elastomers. PhD thesis, VTT, Finland 2005.

[4] Zajac P., Kaleta J., Lewandowski D., Gasparowicz A., Isotropic magnetorheological elastomers with thermoplastic matrices: structure, damping properties and testing, Smart Materials and Structures 2010, 19, 045014.

