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Polymer 47 (2006) 227–233
polymer
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Magnetic field sensitive functional elastomers with tuneable elastic modulus

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Received 18 March 2005; received in revised form 1 August 2005; accepted 28 October 2005
Available online 21 November 2005

Abstract

The main purpose of the present work was to establish the effect of external magnetic field on the elastic modulus. We have prepared poly(dimethyl siloxane) networks loaded with randomly distributed carbonyl iron particles. It was found, that the elastic modulus of magnetoelasts could be increased by uniform magnetic field. In order to enhance the magnetic reinforcement effect, we have prepared anisotropic samples under uniform magnetic field. This procedure results in formation of chain-like structures from the carbonyl iron particles aligned parallel to the field direction. The effect of particle concentration, the intensity of uniform magnetic field as well as the spatial distribution of particles on the magnetic field induced excess modulus were studied. It was established that the uniaxial field structured composites exhibit larger excess modulus compared to the random particle dispersions. The most significant effect was found if the applied field is parallel to the particle alignment and to the mechanical stress. A phenomenological approach was proposed to describe the dependence of elastic modulus on the magnetic induction. The magnetic field sensitive soft materials with tuneable elastic properties may find usage in elastomer bearings and vibration absorber.

Keywords: Magnetic elastomers; Anisotropic elastomers; Elastic modulus

1. Introduction

Composite materials consisting of rather rigid polymeric matrices filled with magnetic particles have been known for a long time and are called magnetic elastomers. These materials are successfully used as permanent magnets, magnetic cores, connecting and fixing elements in many areas. These traditional magnetic elastomers have low flexibility and practically do not change their size, shape and elastic properties in the presence of external magnetic field.

The new generation of magnetic elastomers (abbreviated as magnetoelasts) represents a new type of composite, consisting of small (mainly nano- or micron-sized) magnetic particles dispersed in a highly elastic polymeric matrix [1–26]. The combination of polymers with magnetic materials displays novel and often enhanced properties. The magnetic particles couple the shape of the elastomer to the external magnetic field. Combination of the magnetic and the elastic properties leads to a number of striking phenomena that are exhibited in response to impressed magnetic field. Giant deformational effect, tuneable elastic modulus, non-homogeneous deformation and quick response to the magnetic field open new opportunities for using such materials for various applications. More information on the shape change induced non-uniform magnetic field can be found in our previous papers [6,8–11,14].

It has only recently been accepted that the elastic modulus of magnetic elastomers can be increased rapidly and continuously by application of external magnetic field [4,5,7].

If the magnetoelast contains magnetic particles dispersed randomly there are two basic experimental situations: The compressive force ($\mathbf{F}_x$) and the direction of magnetic field (characterized by the magnetic induction, $\mathbf{B}$) can be either parallel or perpendicular as shown on Fig. 1. In one of our previous papers we found a 20% increase in the modulus for a magnetoelast containing randomly distributed magnetic particles [25,26].

Elastic materials with tailor-made anisotropy can also be prepared under external field [5,7,25,26]. The anisotropy manifests itself both as a directionally dependent elastic modulus as well as a directionally dependent swelling. The anisotropic composite exhibited an increase in 21.5 kPa in the shear storage modulus upon application of a 740 Oe magnetic field [4,5]. Jolly et al. have found 0.6 MPa maximum increase in the shear modulus for iron loaded elastomer [7]. In these works the shear elastic behaviour perpendicular to the columnar structure, have been investigated. To the best of our knowledge, no information is available concerning other experimental situations. Depending on the direction of

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doi:10.1016/j.polymer.2005.10.139

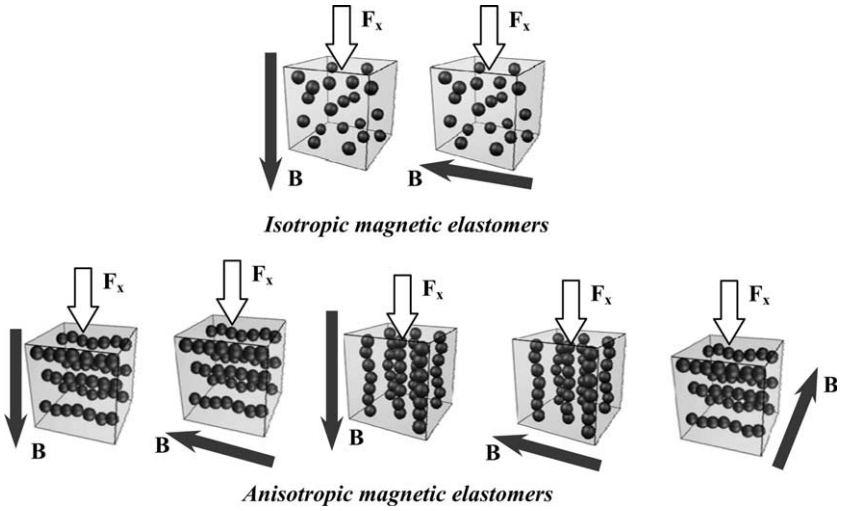

Fig. 1. Experimental possibilities in the study of the influence of external magnetic field on the elastic modulus of the isotropic and anisotropic elastomers. White arrow indicates the direction of the force; black arrow shows the direction of magnetic field.

the magnetic field, the columnar structure and the mechanical stress, the elastic modulus of the anisotropic magnetoelasts can be determined by using five different experimental set-ups as shown on Fig. 1.

The main purpose of the present work was to establish the effect of external magnetic field on the elastic modulus. For the experiments poly(dimethyl siloxane) elastomers loaded with carbonyl iron (Fe) particles were used. The improvements in our understanding of the magneto–mechanical properties of magnetoelasts should assist the development of new type of dampers where the magnetoelastic response can be actively controlled in real time.

1.1. Magnetic elastomers in uniform field, a theoretical approach

Let us consider first a piece of cube-shaped ideal rubbery material without external magnetic field. Let define the initial (reference) and deformed length of the cube in the x-direction of the force denoted by h_0 and h_x, respectively. The principal deformation ratio along the axis x is $\lambda_x = h_x/h_0$. Those for perpendicular directions are λ_y and λ_z. The strain energy density function, W_{el} for an ideal network can be expressed as follows [27,28]:

$$W_{el} = \frac{1}{2} G_0(\lambda_x^2 + \lambda_y^2 + \lambda_z^2 - 3) \tag{1}$$

where G_0 represents the elastic modulus. If the deformation is performed at constant volume, then one may write for the deformation ratios: $\lambda_x{*}\lambda_y{*}\lambda_z = 1$. In case of unidirectional compression λ_y equals to λ_z, and consequently both deformation ratios reduce to $\lambda_x^{-1/2}$. The strain energy density function can be expressed in the terms of λ_x as well as the elastic modulus, G_0:

$$W_{el}(\lambda_x) = \frac{1}{2} G_0 \left(\lambda_x^2 + \frac{2}{\lambda_x} - 3 \right) \tag{2}$$

The nominal stress, σ_{el} can be derived from Eq. (2) by a standard method [27,28]:

$$\sigma_{el} = \left(\frac{\partial W_{el}}{\partial \lambda_x} \right) \tag{3}$$

and the stress–strain dependence of unidirectional deformation of an ideal polymer network can be described by neo-Hookean stress–strain relation [29]:

$$\sigma_{el} = G(\lambda_x - \lambda_x^{-2}) = G_0 D \tag{4}$$

where $D = \lambda_x - \lambda_x^{-2}$. It follows from the above equation that the elastic modulus can be defined as follows:

$$G_0 = \frac{1}{3} \lim_{\lambda_x \to 1} \left(\frac{\partial \sigma_{el}}{\partial \lambda_x} \right) \tag{5}$$

It is worth mentioning, that in the small strain regime one can approximate the experimental data by Hooke's law with Young modulus of $E = 3G$.

Comparing Eqs. (3) and (5), one can express the elastic modulus by the following alternative expression:

$$G_0 = \frac{1}{3} \lim_{\lambda_x \to 1} \left(\frac{\partial^2 W_{el}}{\partial \lambda_x^2} \right) \tag{6}$$

In case of a magnetoelast the overall energy density is the sum of $W_M(H_{eff})$ magnetic and $W_{el}(\lambda_x)$ elastic energy contributions:

$$W(\lambda_x, H_{eff}) = W_{el}(\lambda_x) + W_M(H_{eff}) \tag{7}$$

where H_{eff} stands for the effective magnetic field strength. The magnetic energy density, W_M of a piece of magnetoelast is related to the effective field:

$$W_M = \mu_0 M(H_{eff}) H_{eff} \tag{8}$$

where $M(H_{eff})$ function describes the magnetization curve and μ_0 denotes the magnetic permeability of the vacuum.

Deformation of a magnetoelast involves a change in the particle arrangement, which affects on the magnetic energy density, $W_M(H_{eff})$. As a consequence, the deformation of

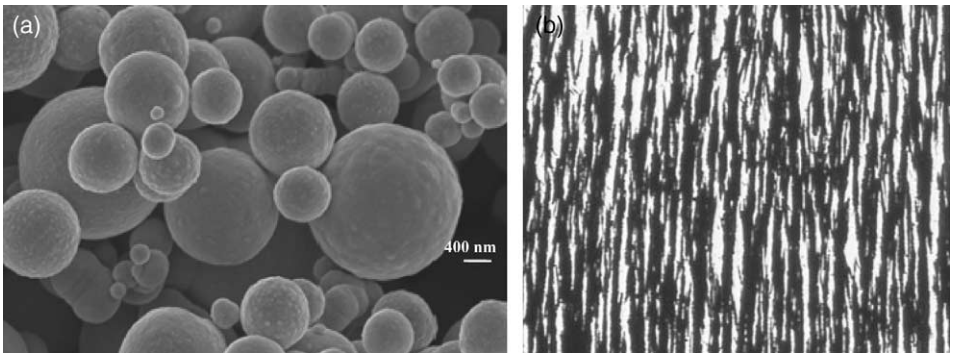

Fig. 2. SEM picture of the carbonyl iron particles in the lack of external magnetic field (a), chain formation of carbonyl iron particles under uniform magnetic field (b) seen by light microscope.

magnetoelast requires more energy in a uniform magnetic field since one must overcome both the change in the elastic, and the magnetic energy as well. In practice, this magnetic effect manifests itself as an increase in the elastic modulus. We call this effect as temporary reinforcement due to uniform magnetic field.

On the basis of Eqs. (6) and (7) the elastic modulus measured under uniform external field can be expressed as the sum of two contributions:

$$G = G_0 + G_{\mathrm{M}}^E \tag{9}$$

where G_{M}^E denotes the magnetically induced excess modulus. We can characterize the magnetic field intensity by the magnetic induction, B which is the product of the magnetic permeability of the vacuum and the field intensity: $B = \mu_0 H_{\mathrm{eff}}$. The magnetic contribution of the modulus can be calculated from the change of magnetic energy due to the distortion of the composite. Similar to Eqs. (6) the excess modulus can be defined as follows:

$$G_{\mathrm{M}}^E = \frac{1}{3} \lim_{\lambda_x \to 1} \left(\frac{\partial^2 W_{\mathrm{M}}}{\partial \lambda_x^2} \right) \tag{10}$$

Since the magnetic polarization is a fast process, consequently G_{M}^E develops quickly when the external field is turned on and disappears immediately when the external field is turned off. The analytic calculation of the magnetic excess modulus, G_{M}^E is a rather complicated task due to the fact that the magnetic energy density $W_{\mathrm{M}}(H_{\mathrm{eff}})$, can not be given in an analytical form.

2. Experimental part

2.1. Preparation of magnetic elastomers

In order to prepare magnetic field responsive PDMS composites, carbonyl iron particles were used as magnetic filler particles. Commercial product of BASF (Germany) was used for the preparation without further purification. The average size of the spherical iron particles is 2.5 μm. The specific surface area determined by BET method is 0.7 m^2/g. The concentration of the solid magnetic materials was varied between 10 and 30 wt% in the polymer matrix. Fig. 2 shows the scanning electron microscope (SEM) picture of the carbonyl iron particles. It can be seen on the figures that the polydisperse carbonyl iron particles have spherical shape with rather smooth surface.

Magnetic PDMS networks were prepared from a commercial product of a two component reagent (Elastosil 604 A and Elastosil 604 B) provided by Wacker Co. These chemicals were used without further purification. Component A contains polymers and the Pt containing catalyst while component B provides the cross-linking agent. Component B was varied from 2.5 to 3.5 wt%. The carbonyl iron particles were dispersed in the Elastosil 604 A. After mixing it with the Elastosil 604 B component, the solution was transferred into a cube-shaped mould. The cross-linking reaction was carried out at ambient temperature for 4.5 h to obtain magnetoelasts. After gelation, the cube shaped samples were removed from the moulds.

Anisotropic magnetic elastomers were prepared under uniform magnetic field. The mixture of the silicone prepolymers and the carbonyl iron particles, confined in the cube-shaped mould was placed between the poles of a large electromagnet (JM-PE-I (JEOL, Japan). The mixture was subjected to $B = 400$ mT uniform magnetic field for 5 h. In uniform field magnetic dipoles are induced. As a result, mutual particle interactions occur if the particles are so closely spaced that the local field can influences their neighbours. This mutual interaction can be very strong leading to significant change in the structure of particle ensembles. The particles attract each other when aligned end to end, and repel each other in side-by-side situation. This results in formation of chain-like structures aligned parallel to the field direction as shown in Fig. 2(b). The chain formation takes a few minutes. If the chemical reaction proceeds slowly, then there is enough time to lock the pearl chain structure of the carbonyl iron particles into the network by the cross-linking reaction. After the reaction was completed, the silicon cubes were removed from the moulds.

We have varied the concentration of the carbonyl iron particles at constant cross-linking density. The concentration of component B was 3.5 w/w% in every case.

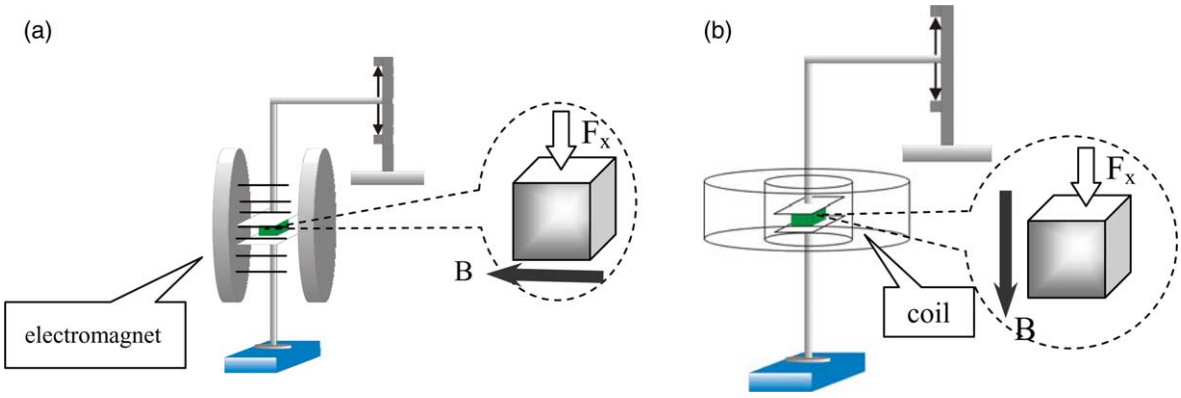

Fig. 3. Schematic representation of the experimental set-up measuring elasticity under uniform magnetic field. F_x represents the force and B denotes the magnetic induction.

2.2. Mechanical measurements under static magnetic field

The elastic modulus of magnetoelasts was measured under uniform magnetic field at 293 K using two different home made apparatus. The magnetic induction, **B** was measured by a Phywe Teslameter. Comparing the direction of the force and the magnetic field there are two different situations: the force and the magnetic field are either perpendicular or parallel as shown on Fig. 3(a) and (b).

In perpendicular case the magnetic elastomer was placed on the sample stage between two magnetic poles (Fig. 3(a)) and the stress, which is perpendicular to the magnetic field, was measured by moving the silver rod connected to the strain gauge. The gap between the magnetic poles is 40 mm and the cross section of the pole is 19.6 cm^2. The magnetic field around the sample stage can be considered to be uniform. We were able to measure the stress–strain dependence under magnetic field between 0 and 400 mT.

When the stress and the strain are parallel (Fig. 3(b)), a coil was used to create the magnetic field. In this case we were able to measure the stress–strain dependence between 0 and 100 mT. In the experiments the magnetic field intensity was varied and the elastic modulus was measured as a function of magnetic induction, B.

3. Results and discussion

3.1. Effect of uniform magnetic field on the elastic modulus

Fig. 4 shows the effect of uniform magnetic field on the modulus of the magnetic PDMS samples containing randomly distributed Fe particles. Depending on the direction of the field to the mechanical stress, two kind of experimental arrangements were used: parallel and perpendicular. One can see on both figures that there is a slight increase in the modulus due to the external field. This finding is in accordance with our previous result where similar temporary reinforcement effect was reported for magnetite (Fe_3O_4) loaded poly(vinyl alcohol) hydrogels [20]. It is also seen on the figures that the concentration of the filler particles increases the field free modulus, G_0. Larger amount of Fe particles results in larger elastic modulus.

We have performed systematic studies on the magnetic reinforcement effects. All of the experimental arrangements seen in Fig. 1 have been investigated. The magnetic field intensity was varied and the elastic modulus was measured as a function of magnetic induction, B. The effect of the cross-linking density as well as the concentration of carbonyl iron particles has also been investigated.

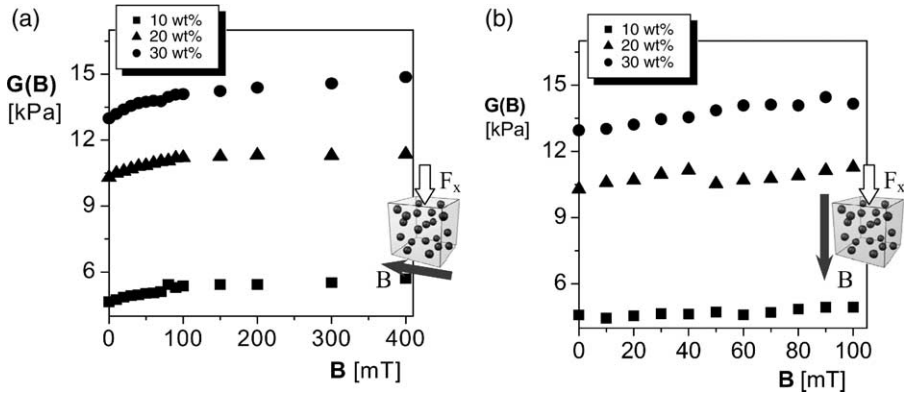

Fig. 4. Dependence of the magnetic field intensity on the elastic modulus for magnetoelasts containing different amount of carbonyl Fe particles. The concentration of the filler particles is indicated in the figure.

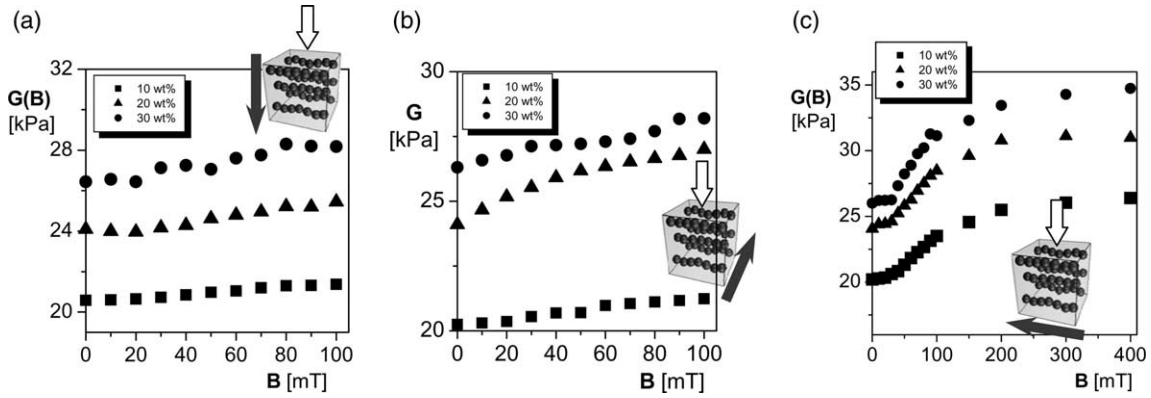

Fig. 5. Effect of the magnetic field intensity on the elastic modulus. The iron content of the elastomers is indicated on the figure. The white and black arrows show the direction of the force and the uniform magnetic field, respectively.

3.1.1. Compressive force is perpendicular to the direction of particle chains

The effect of the uniform magnetic field on the elastic modulus was studied when the applied mechanical stress was perpendicular to the particle alignment. By varying the direction of the applied magnetic field and the columnar structure of the particles, we have three possible experimental arrangements: the direction of the field is perpendicular to the particle chains (Fig. 5(a) and (b)) and parallel to the particle-chains (Fig. 5(c)).

On the basis of the experimental results shown on Fig. 5 it may be concluded that the spatial orientation of the force, the field and the particle arrangement play a decisive role in the temporary reinforcement effect. A weak effect has been found when the field is perpendicular to the particles alignment (Fig. 5(a) and (b)).

If the columnar arrangements of the particles are parallel to the direction of the magnetic field, the elastic modulus increases significantly (Fig. 5(c)). At small field intensities up to 30 mT a slight increase has been observed. Above 30 mT the modulus increases significantly. At higher field intensities (from 200 mT) the elastic modulus tends to level off. It is also seen that by increasing the concentration of the iron particles in the polymer matrix the elastic modulus, G_0 also increases. The strongest magnetic reinforcement effect was found if the magnetic field is parallel to the particle alignment.

3.1.2. Compressive force is parallel to the direction of particle chains

Fig. 6 shows the reinforcement effect when the direction of the applied mechanical stress is parallel to the particle alignment. In this case there are two different experimental situations. The direction of the magnetic field is parallel or perpendicular to the direction of the columnar structure as seen on Fig. 6. When the applied uniform magnetic field is perpendicular to the direction of the particle chains (Fig. 6(b)), the magnetic field does not make its significant influence felt on the elastic modulus. The strongest magnetic reinforcement effect was found when the applied uniform magnetic field is parallel to the particle alignment and to the mechanical stress (Fig. 6(a)). The relative increment of the modulus has the highest value.

When the iron content is increased, the mutual particle interaction increases which induces higher excess modulus. The elastic modulus increases significantly with the magnetic field intensity if the particles arrangements are parallel to

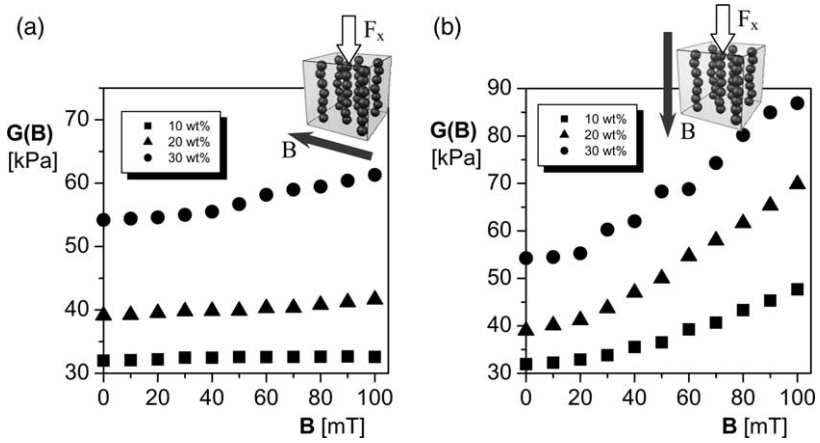

Fig. 6. Dependence of the magnetic field intensity on the elastic modulus. The arrangements of the particles in the polymer networks are parallel to the applied mechanical stress while the applied uniform magnetic field is parallel (a) or perpendicular (b) to the columnar structure. The concentration of the carbonyl iron particles in the polymer matrix is indicated on the figure.

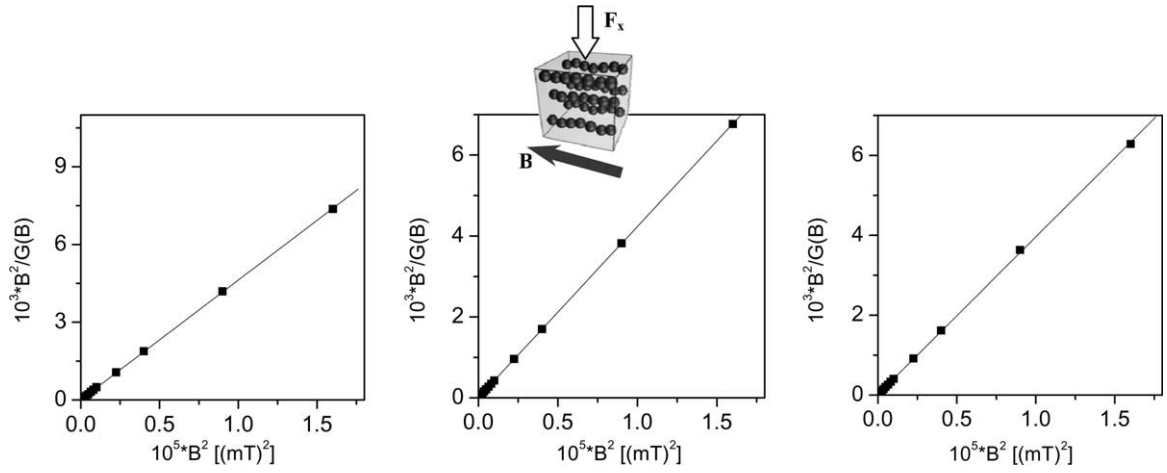

Fig. 7. Dependence of $B^2/G^E_M(B)$ on B^2 for magnetic PDMS samples. The concentration of carbonyl iron varies from left to right: 10, 20 and 30 wt%. The experimental arrangement is shown on the figure.

the force and to the applied uniform magnetic field. However, the magnetic field does not influence the elastic modulus significantly when the applied field is perpendicular to the mechanical force and to the particle-chains.

3.2. Interpretation of the experimental results

We have demonstrated that in the presence of built in magnetic particles it is possible to modify the elastic modulus. The measure of magnetically induced excess modulus, G^E_M depends on the concentration and spatial distribution of the magnetic particles as well as on the strength of applied field. For randomly dispersed magnetic particles G^E_M slightly depends on the particle concentration and on the magnetic induction. It was found to vary between 0 and 2.8 kPa. More significant magnetic reinforcement effect was found for anisotropic samples containing oriented particle chains, instead of randomly distributed particles. If the mechanical stress and the direction of columnar structure as well as the magnetic induction are all parallel G^E_M approaches to 32.7 kPa when the magnetic PDMS sample contains 30 wt% carbonyl iron (Fig. 6(a)).

The analytic calculation of the magnetic modulus, G^E_M is a rather complicated task due to the unknown $W_M(H_{eff})$ function.

It was found that at low field intensities the magnetic excess modulus is proportional to the square of the magnetic induction: $G^E_M \propto B^2$. At high field intensities the magnetization saturates and as a result G^E_M approaches a maximum value of $G_{M,\infty}$.

These two limiting cases can be phenomenologically described by the following equation:

$$G^E_M(B) = G_{M,\infty}\frac{B^2}{a_B + B^2} \tag{11}$$

where a_B represents a material parameter.

The parameters (a_B and $G_{M,\infty}$) can be determined by linearization of Eq. (11)

$$\frac{B^2}{G^E_M(B)} = \frac{a_B}{G_{M,\infty}} + \frac{1}{G_{M,\infty}}B^2 \tag{12}$$

By plotting the quantity of $B^2/G^E_M(B)$ against B^2, the slope provides the $1/G_\infty$ value, and the intercept gives the ratio $a_B/G_{M,\infty}$. We have found that the proposed phenomenological equation is in rather good agreement with the experimental results. Fig. 7 shows the experimental data plotted according to Eq. (12).

We determined the parameters $G_{M,\infty}$ and a_B (Table 1) by linear least square method for the samples shown in Fig. 7.

On Fig. 8 the experimental data and the phenomenological approach are presented. It is seen that the agreement between experimental data and phenomenological description is quite satisfactory. It is a further important task to investigate how the parameters of Eq. (11) depend on the particle concentration as well as on the spatial distribution. This will be the subject of our forthcoming paper.

4. Summary of the main results

The main purpose of the present work was to perform systematic investigations of how the uniform magnetic field influences the elastic modulus of PDMS samples loaded with carbonyl iron particles. We have prepared

Table 1
Parameters of Eq. (12) for the sample shown in Fig. 7 determined linear least square method

Iron content [wt%]	a_B [(mT^2)]	$G_{M,\infty}$ [kPa]
10	12059.55	6.68
20	6945.07	7.33
30	10915.83	9.35

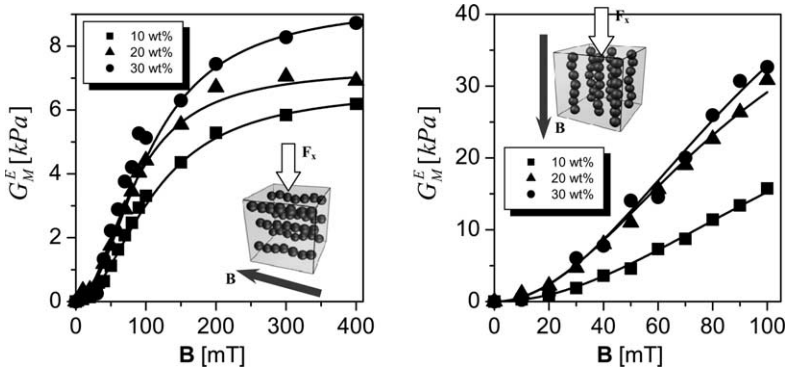

Fig. 8. Dependence of G_M^E on the magnetic induction for two kind magnetoelasts having different experimental arrangement as shown on the figures. Solid lines were calculated on the bases on Eq. (11).

magnetoelasts with randomly distributed carbonyl iron particles. It was found, that the elastic modulus of magnetoelasts could be increased by external magnetic field. This effect is called as temporary reinforcement. A slight increase in elastic modulus on the magnetic field was observed. In order to enhance the magnetic reinforcement effect, we have prepared anisotropic samples by varying the spatial distribution of the magnetic particles in the elastic matrix. We have shown that uniaxial field structured composites exhibit much larger increase in modulus than random particle dispersions. Mechanical properties like elastic modulus, stress–strain behaviour of samples characterized by parallel- and perpendicular chain-like structure are significantly different. It was found that the temporary reinforcement effect was most significant if the applied field, the particle alignment and the mechanical stress are all parallel with each other. A phenomenological approach was proposed to describe the dependence of elastic modulus on the magnetic induction. Within the experimental accuracy the prediction of phenomenological equation is supported by the experimental data.

These magnetic field sensitive materials with tuneable elastic properties may find usage in elastomer bearings and vibration absorber. Anisotropic magnetic elastomers are worth further experimental and theoretical study. An understanding of magneto–elastic coupling in anisotropic magnetoelasts will hasten the engineers to develop more powerful smart vibration and shock absorber.

Acknowledgements

This research was supported by the Széchenyi NRDP No. 3/043/2001 and the Hungarian National Research Fund (OTKA, Grant No. T038228 and F046461). This research is sponsored by NATO's Scientific Division in the framework of the Science for Peace Programme (NATO SFP 977998).

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