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Magneto-mechanical properties of elastic hybrid composites

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Abstract: The paper gives an overview of remotely controlled elastic magnetic composites based on silicon rubber matrix highly filled with a magnetic soft and hard filler. The magnetic soft phase, which is represented by iron microparticles, allows active control of the physical properties of the composites, while the magnetic hard phase (e.g. neodymium-iron-boron alloy microparticles) is mainly responsible for passive adjustment of the composite. Active control of these materials based on the magnetic hard phase is also possible, depending on the type of powder used. The control is performed by the application of an external magnetic field *in situ*, and passive adjustment is performed by means of pre-magnetization in order to change material remanent magnetization, i.e. the initial state. The potential and limits of active control and passive tuning of these composites in terms of their magneto-mechanical behavior are presented and discussed.

Keywords: soft magnetic composite, magnetorheology, anisotropy, smart materials

PACS: ...

1 Introduction

The combination of magnetic microparticles and an elastic soft polymeric matrix makes it possible to obtain an intelligent material with a wide range of remotely controlled properties. Similar to concentrated suspensions of magnetic microparticles, known as magnetorheological fluids, composites in which the liquid carrier medium is replaced by an elastic matrix are known as magnetorheological elastomers [1]. Other frequently used designations for them are magnetoactive or magnetosensitive elastomers [2]. The principal effect of a magnetic field on these composites is a change in their viscoelastic properties. To quantify this change, which is also referred to as the magnetorheological effect (MR effect), it is appropriate to use the relative change in the corresponding material parameter under the influence of an external magnetic field $\vec{H}$. In addition to the relative effect,

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the absolute effect is also to be taken into account, i.e. the difference between the material parameter in the magnetic field and that without it. Other important magnetically controlled effects are the magneto-deformational (strictional) effect [3, 4, 5, 6, 7, 8, 9, 10] and the rather poorly investigated shape memory effect [2, 11, 12]. In besides, the attention of researchers is attracted by the possibility of controlled modification of surface roughness of such composites [13, 14, 15], as well as their dielectric properties [16, 17, 18]. This set of unique features makes these composites attractive for various technical implementations, such as damping devices and suspensions, activators, sensors and grippers, including medical applications (see e.g. [19, 20, 21, 22, 23, 24]).

As a matrix of elastic magnetic composites it is proposed to use various materials: from biocompatible hydrogels to silicon compounds and vulcanized rubbers (see e.g. [25, 26, 27, 28]). The choice of matrix and magnetic filler depends on the intended application of the material and it must be ensured that the material properties are sufficiently controllable, e.g. that the MRE is significant. Thus, at high modulus of elasticity (from ∼1 MPa), even a high degree of filling of the composite with magnetic particles is not a guarantee of variability of controlled properties, i.e. the relative effect is very restricted [26]. Ultra-soft, gel-like matrix, having the modulus of elasticity of several kPa or lower, guarantees a high relative effect with a sufficient filling of the material with magnetic powder, but the absolute value of the increase of modules in the field may be insignificant [27]. The use of silicone compounds as a matrix highly filled with magnetic powder of microparticles has proven to be a promising option for magneto-controlled composites [2, 12, 29, 30]. Magnetic-soft iron powders are commonly used as a composite filler. Alternatively, approaches using magnetic hard powders were proposed [31, 32, 33, 34]. First of all, this allows one to change the initial state of the composite by changing its residual magnetization. Despite the residual magnetization, the application of an external magnetic field allows for reversible changes in the properties of these materials [35, 36, 37]. The use of complex powders, in particular mixtures of soft magnetic and hard magnetic particles, expands the possibilities of active control, while maintaining the possibility of passive tuning of the material [9, 38, 39].

In this contribution we present an overview of recent results obtained within experimental characterisation of magneto-controlled composites based on silicone rubber highly filled with different types of magnetic filler. The work is focused on magnetic properties and magnetorheological effect. Also, essential information about the components and the process of manufacturing composites is briefly presented. This article is correspondingly divided into three parts: section 2 deals with components and fabrication steps, section 3 presents magnetic properties and in section 4 magneto-mechanical response of the composites is considered.

2 Material composition and fabrication

The basic components of controllable magnetic elastic composites are a polydimethylsiloxane (PDMS) matrix and a magnetic filler. In Figure 1 chemical components of the matrix, polymerization reaction, schematic representation of the filled cross-linked matrix as well as optical microscopy images of various particle powders are provided. Below given a brief description of the manufacturing process, more details can be found in [40][1].

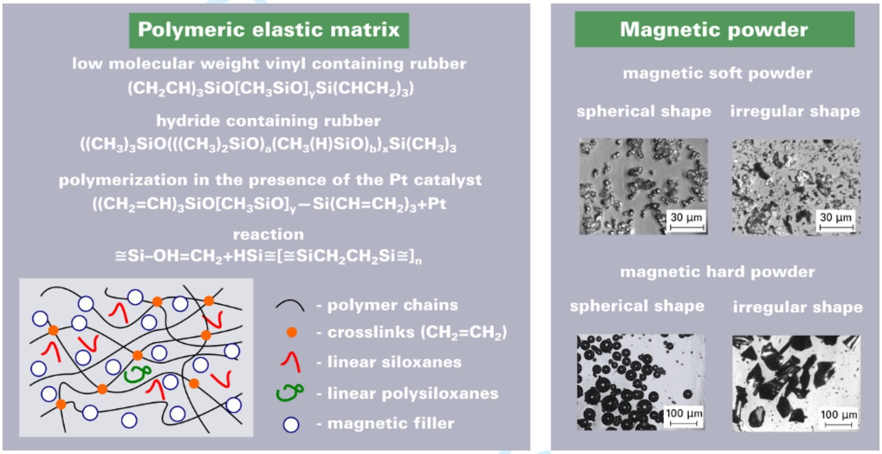

Fig. 1: Typical components used for hybrid magnetic elastic composites: chemicals, polymerization reaction and schematic representation of the filled PDMS matrix (left) and exemplary optical microscopy images of magnetic soft and hard microparticles of the powders of various shape (right).

2.1 Composite manufacturing

The main technological stages of composite manufacturing include (I) preparation of magnetic filler (powder modification), (II) mixing of filler and liquid components of matrix, (III) vacuumization of composition and its casting into moulds, and, finally, (IV) polymerization (cross-linking) process.

1 The study [40] is going to be published in the current issue.

The aim of stage *I* is to ensure the compatibility of the powder microparticles with the silicone matrix and to prevent its possible impact on the cross-linking process. This is accomplished by coating the particle surface with a layer of siloxane liquid. Detailed information on particle treatment is given, e.g. in the studies [34, 38]. The determining criterion when choosing a soft magnetic filler is its high saturation magnetisation and minimum coercivity. According to this criterion, iron and iron oxide (e.g. Fe_2O_3, Fe_3O_4) microparticle powder is commonly used as a magnetic soft filler. Iron powders with microparticles of various sizes and shapes are commercially widely available, e.g spherical carbonyl iron powders from BASF. The choice of the hard magnetic phase is based on the high remanence magnetisation of the powder material as well as its high coercivity. Powder metallurgical technologies are usually used in the manufacture of permanent magnets, due to this, hard magnetic materials are also commercially available in powder form. Among materials with high coercivity are known so called high-energy ones, which are samarium cobalt (SmCo) and neodymium-iron-boron (NdFeB) alloys. Moreover, there is a technology for obtaining spherical microparticles for NdFeB alloys [41]. The use of spherical powders is preferable in terms of comparing experimental results with theoretical researches. On the other hand, the use of particles with a shape other than spherical is interesting from a practical point of view. When microscopic mechanisms of magnetic interactions for spherical and non-spherical particles differ, this will affect the macroscopic properties of composites. Thus, preference may be given to magnetic hard powders based on NdFeB, which are available with particles of different shapes (e.g. Magnequench^{tm} powders).

In stage *II*, the treated filler is mechanically mixed with the initially liquid components of the polymer matrix. For this purpose, one can use a stirrer for the coarse dispersion and, if necessary 3-roll dispersing machine for fine dispersion. The matrix can be diluted with silicon oil in order to tune the elasticity of the composite. In addition, in order to prevent the composition from separation, the suspension can be enriched with structural additives that increase its viscosity.

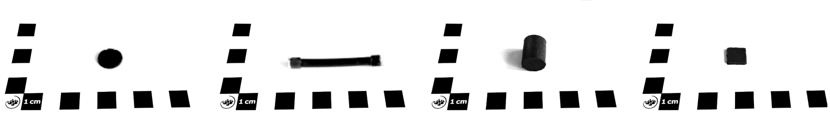

Fig. 2: Composite samples of various shapes: disk, rod, cylinder and plate.

Degassing of the mixture in stage *III* is necessary to remove air bubbles in order to obtain a defect free sample. The resulting degassed mixture is poured into a mould with a particular geometry, which is determined by the further purpose of the sample. The mold walls can be coated with a surfactant layer that blocks the polymerization of the silicone polymer. Figure 2 shows examples of composite samples of various shapes.

The curing process (stage IV) can take place both at room and at increased temperature. Heating increases the reaction rate and therefore can has a positive effect on the homogeneity of particle distribution in the matrix volume. As an alternative to heating in a convection furnace, microwave heating can be used, which increases the speed of the set temperature inside the sample [34]. Also effective at preventing particle sedimentation is the rotation of the sample at low speeds (e.g. ∼10 rpm) during the cross-linking process.

Optionally, during the stage IV, the sample can be placed in a homogeneous external magnetic field (without rotation). In this case, an anisotropic, i.e. structured in the direction of the applied field, composite will be obtained. The use of various filler concentrations and conditions of magnetic field application at this stage is used to obtain different types of internal structure morphologies [42, 43, 44, 45]. Strategies of the targeted structuring of magnetic microparticles in a polymer composite are recently discussed in [46]. Composites pre-structured during the process of curing are not the focus of this work.

2.2 Composite pre-magnetization

The pre-magnetization of the specimens containing magnetically hard phase is realized using a uniform field provided by the electromagnet B-E25 (Bruker, Germany). Typically external magnetizing fields are used up to B_M=1500 mT are used. Cylindrical and rod-shaped samples are magnetized along the vertical axis and disk- as well as plate-like samples are magnetized in the direction perpendicular to the plain. The pre-magnetization results in the induced microstructural anisotropy of the composites due to the particles rearrangements when the matrix is soft enough as shown in [37]. It's worth noting that effective magnetic fields affecting inside the samples with different geometries and various pre-magnetizations are not equal for identically applied external fields, due to the samples own demagnetizing fields. However, it is not always possible to take this issue into account, since demagnetization factors for the composites with an induced microstructural anisotropy remain unknown. On this reason, we handle with external fields in this work.

2.3 Additional processing

In addition to the above specified manufacturing steps, it is necessary to mention the additional processing of samples, namely their adaptation for subsequent fixation in experimental setup. Depending on what kind of experiment the sample of certain geometry is intended for, it is additionally supplied with structural elements made of solid polyethylene. For example, for strong fixation of rod specimens in the clamps of testing machines, annular plugs are glued using cyanoacrylate adhesive to their ends (see e.g. rod specimen in Figure 2). Solid polyethylene discs with diameters equal to or greater than the diameter of the specimen diameter shall be glued to the ends of the cylindrical specimens. Thin discs for magnetic measurements are completely fixed in a hollow thin-walled cylindrical holder with a height equal to the thickness of the sample (see Figure 3). Shear test specimens (e.g. discs and plates) are usually glued directly to the measurement geometry of the testing equipment.

3 Magnetic properties

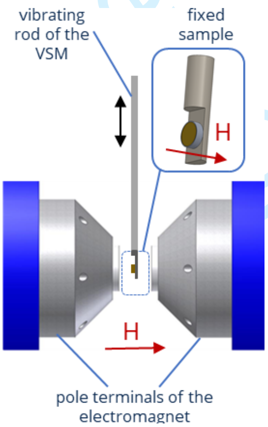

Fig. 3: Sample orientation in the measurements of its magnetic properties on a vibrating sample magnetometer.

A common method of determining the magnetic properties of magnetic hybrid composites is to obtain their magnetization curves by means of vibrating sample magnetometry. This method consists in measuring the magnetic moment of a sample oscillating with a given frequency and amplitude between a set of pickup coils in an external uniform magnetic field. We utilize the Lake Shore 7407 magnetometer and a nickel sphere with a diameter of 3 mm having a magnetic moment of m=6.92 mA$\cdot$m^2 at external field H=398 kA/m and T=298 K as a calibration sample. The method provide an accuracy of the experimental sample magnetization measurement of at least 1 A/m. For composites magnetic characterization, disc-shaped specimens were used (diameter 4.7 mm, height 1-1.5 mm). The sample plane in the experiment is perpendicular to the external magnetic field as shown in Figure 3.

The magnetic properties of the elastic composites are primarily determined by the material and concentration of the filler's powder, but the matrix in which the powder particles are embedded can also influence the magnetization processes. Depending on the rigidity of the matrix, the filler particles will have different mobility levels. If the matrix is soft enough, the microparticles will move, rotate and form structures by the effect of the applied magnetic field. The microstructural rearrangement of the particles results in the correspondingly forced changes of the macroscopic behaviour of the composite material. Theses statements are confirmed both from an experimental and theoretical point of view [2, 8, 35, 36, 37, 47, 48, 49, 50]. In Figure 4 measured magnetization curves of two samples with different matrices filled with 20 vol.% of spherical iron microparticles are shown. Despite the fact that the filler is a soft magnetic material, the magnetization curve of the composite based on a soft matrix is a hysteresis. In particular, the magnetization of the soft sample in a decreasing field is higher, than the magnetization in the increasing field. As mentioned above such behavior is related to the hysteresis movement of particles inside the matrix. Curves run is reproduced by repeated measurements if negative magnetic fields are not used. In the case of a negative field, irreversible particle movements relative to the matrix (in particular, particle rotation) are possible. In this case, the so-called training of the sample (several repeated runs) is required to achieve reproducibility of the magnetization curves. Such behavior is associated not only with the mobility of particles (particle-matrix interaction), but also with possible matrix defects. As an analogy of such behavior, we may mention the Mullins effect known from the mechanical response of filled rubbers [51].

Another important characteristic of composites based on a soft magnetic filler is the initial magnetic susceptibility χ_{ini}. The susceptibility χ_{ini} is defined as the slope of the initial linear part of the magnetization curve. For magnetic composites χ_{ini} is a function of magnetic powder concentration. Previously, it was proposed to use the so-termed Maxwell-Garnett approximation [52] to estimate the magnetic susceptibility of the composite depending on the volume concentration of the

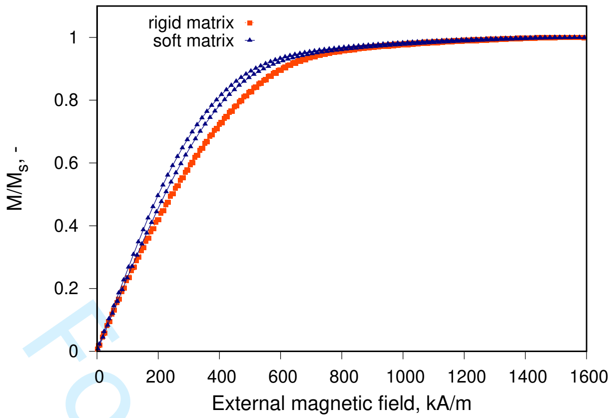

Fig. 4: Magnetization curves of the elastic composites with ~20 vol.% of iron microparticles (BASF CC powder) embedded into a rigid (E ~1 MPa) and soft (E ~0.3 MPa) PDMS matrix. Magnetization of the samples M is normalized to their saturation magnetization M_s.

magnetic phase [4, 53, 54]:

$$\chi_{\text{ini}} = \frac{3\phi}{1-\phi}. \tag{1}$$

The equation 1 provides χ/ϕ close to 3 for diluted composites ($\phi << 1$). Experimental measurements (Figure 5) show that equation 1 satisfactorily predicts values of χ_{ini} only for solid matrix composites, in which structuring and movement of particles is restricted, at filler concentrations not exceeding ~15 %. It is obvious that structuring increases initial magnetic susceptibility. Comparing the results obtained for elastic composites with the results obtained for samples based on the liquid matrix, we assume that the factor determining the susceptibility is the size of structures formed in the magnetic field, i.e. mobility of the particles. In the case of elastomers the effect of structuring is restricted due to the elastic forces immobilizing the particle movements.

Composite samples containing a magnetic hard phase are characterized by a loop of magnetic hysteresis with significantly different from zero residual magnetization M_r and coercive force H_c. Figure 6 demonstrates the initial magnetization curves for two types of hard magnetic powder embedded into epoxy resin comparing to magnetization of the carbonyl iron powder. In Figure 7 dependencies of hard magnetic powders residual magnetization on the applied magnetization field are given. In the composites with an elastic matrix structuring and rotation of particles can be forced by an external magnetic field. This can in turn significantly influence the magnetization process as shown in theoretical and experimental studies

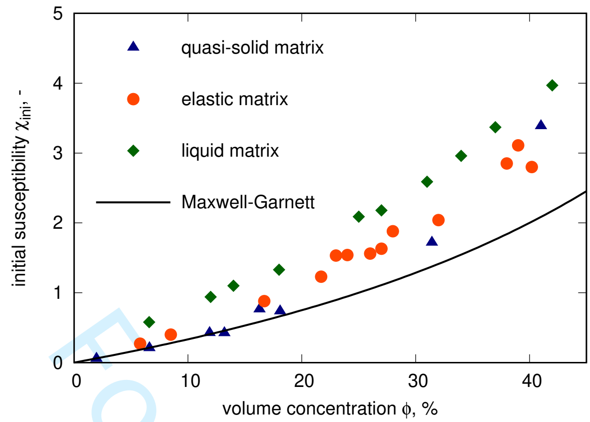

Fig. 5: Measured initial magnetic susceptibility of the composites based on different matrices as a function of the BASF CC carbonyl iron powder volume concentration. Results are given for specimens based on an epoxy resin (quasi-solid, E ~2 GPa), PDMS (elastic, E ~0.3 MPa), and silicon oil (liquid, $\eta = 0.35$ mPa·s). For comparison, the Maxwell-Garnett approximation (equation 1) is as well provided.

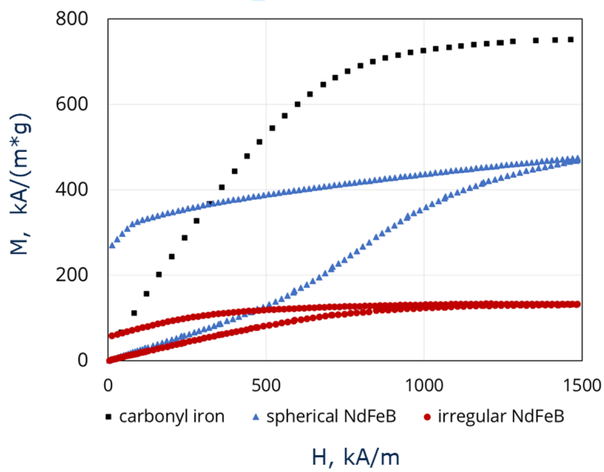

Fig. 6: Initial magnetization curves of the carbonyl iron powder and two types of NdFeB-alloy powder embedded into epoxy resin.

[35, 36, 49, 50, 55, 56, 57, 58]. Figure 8 presents full magnetic hysteresis curves for composites based on spherical magnetically hard powder embedded into the matrices of various elasticity. The processes of both magnetization and rotation

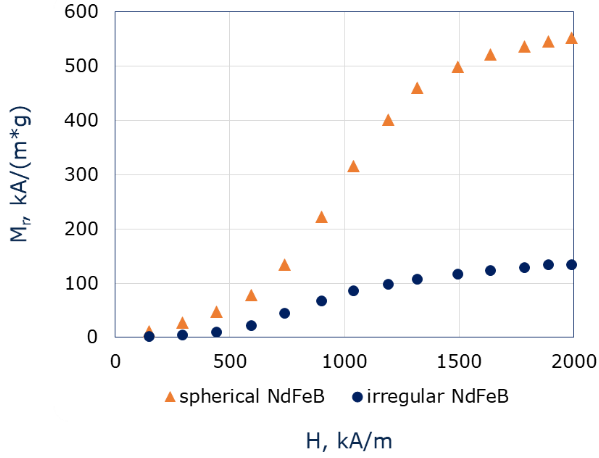

Fig. 7: Dependency of remanence magnetization M_r (by weight) on the external field B_M which was applied to magnetize two different types of magnetic hard powder.

of particles are reflected in the material behaviour. The hysteresis loop of soft

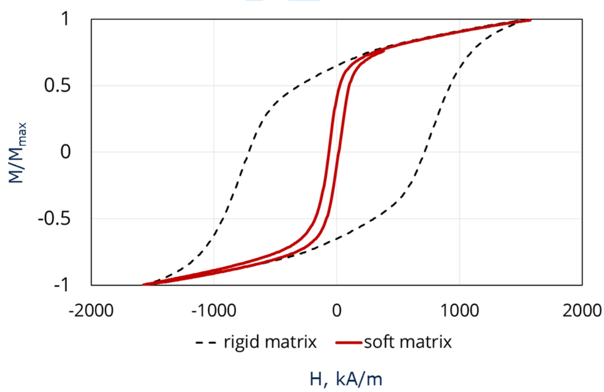

Fig. 8: Magnetization loops of the composite filled with $\sim$40 vol.%. of spherical NdFeB-alloy embedded into a rigid matrix ($E > 1000$ kPa) and soft matrix ($E \sim 60$ kPa). Initial curves are not shown.

composite is much narrow and asymmetrical. This is determined by the direction of primary magnetization of the magnetic elastomer. Moreover, at a certain relationship of the residual magnetization of the magnetic filler, the magnetic field strength applied at re-magnetization and the elasticity of the polymeric matrix, the nominal coercive force may have a negative value, i.e. the magnetization of the material is positive in the negative magnetic field [49]. The effect is caused by rotation of the magnetized particle to its initial state by elastic forces of the polymeric matrix. Another effect that can be seen in the process of magnetizing soft samples with hard magnetic phase is the appearance of significant difference between first consecutive magnetization loops (training effect) [55]. This effect is demonstrated in Figure 9. Furthermore, recently, an influence of the under-magnetization of hard magnetic powder on the training effect in elastic composites was analyzed experimentally and theoretically [58]. In the case of composites with a complex filler, i.e. powder based on a mixture of hard and soft magnetic microparticles, the first-order reversal curve (FORC) measuring method is an effective tool for investigating magnetic interactions within the material. In the context of magnetic elastomers, the FORC method was firstly used in [59]. It has been demonstrated that FORC distributions are also subject to significant influence of structuring and rotation processes of hard magnetic particles dominating over rather energetically unfavorable domain processes within the particles. In addition, the FORC diagrams reflect both irreversible and reversible contributions of the magnetic soft phase to the magnetization of the major loop.

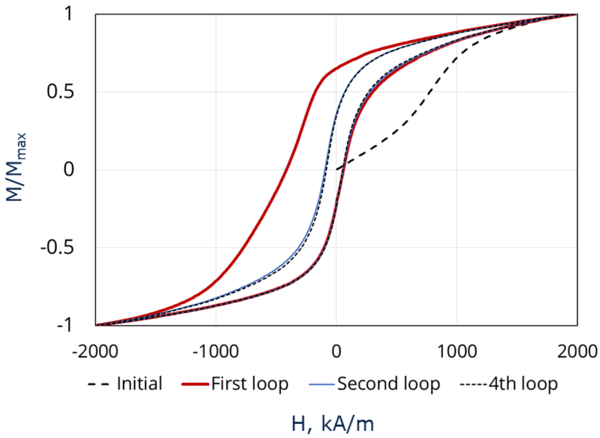

Fig. 9: Magnetization loops of the composite filled with spherical NdFeB-alloy powder at concentration of ∼11 vol.%. The tensile modulus of the filled composite is $E \sim 130kPa$. The presence of training effect is clearly demonstrated: gradual diminution of discrepancy between consecutive magnetization loops. The training effect manifests itself most strongly between the first and second loops, while it quickly fades out after that.

From an applied point of view, the residual magnetization of the composite is an important aspect of irreversible magnetization processes. This parameter determines the initial state of the material, including its mechanical behavior, and it plays a key role in the passive tuning of the magneto-mechanical properties of a composite [33]. In order not to change the initial state of a material with the complex filler, the magnetic field used for active control should not change the residual magnetization. The initial magnetization curves of the hard magnetic phase provide a possibility to estimate the range of the magnetic field, which should be used both for passive adjustment of the composite and for its active control. Beside, taking into account the fact that such undesirable process as the "training effect" which is the peculiarity of complete magnetization cycles, it is expedient not to use the change of magnetic field polarity when controlling under-magnetized composites with hard magnetic phase. Thus, for a practical estimation of possibility of adjustment of a material containing a various ratio of hard magnetic and soft magnetic phases it is at least necessary to estimate residual magnetization of a magneto-hard phase and initial curves of magnetization of the pre-magnetized composite, i.e. minor half-loops of the hysteresis. The issue of the mutual influence of the soft and hard magnetic components on the macroscopic magnetization of the material is considered in details experimentally and theoretically in very recent work [60].

4 Magneto-mechanical response

There are several approaches to measure the magneto-mechanical properties of hybrid elastic composites in conventional setups with the addition of an external homogeneous magnetic field (see e.g. Figures 10 and 11). In the past rheometric devices were adapted to the measurement of magnetorheological fluids with commercial magnetic cells [61, 62, 63]. Therefore, such devices are very often proposed to use for magnetic elastomers. It uses a plate-plate configuration with a disc-shaped specimen (Figure 10a). Critical issues of the method are discussed in [64]. This method should be avoided without special modifications made in a commercial cell that is not originally designed for elastic materials. As shown in [64], when using a rheometer, measuring cell configuration for rod-shaped samples can be used effectively for quasi-static elongation and torsional tests as well as for oscillating torsional tests (Figure 10b). From the field of solid mechanics, a universal test machine can be adopted to conduct a quasi-static and dynamic axial and shear loading (Figure 11). Results of the axial and shear loading provide an information on the tensile modulus E and shear modulus G correspondingly. To the best of

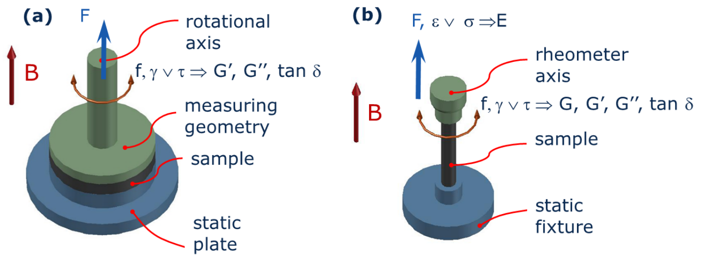

Fig. 10: Configurations of the experimental setup using rotational rheometer: (a) plate-plate configuration for the disc-shaped sample; (b) testing of the rod-shaped specimen; B - external magnetic field applied during the test, F - axial mechanical force, f - oscillating frequency; γ - shear deformation, τ - shear stress, G' - storage shear modulus, G'' - loss shear modulus, tanδ - loss factor, ϵ - tensile deformation, σ - tensile stress, E - tensile modulus.

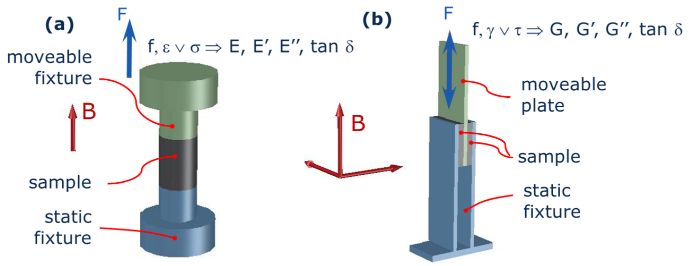

Fig. 11: Configurations of the experimental setup using universal test machine: (a) axial loading of the cylindrical specimen; (b) shear test of the plate-shaped composite using a layout with two coupled shear gaps; E' and E'' - storage tensile and loss tensile modulus correspondingly, other designations are as given in the caption of Figure 10.

our knowledge, a direct systematic comparison of the tensile and shear moduli of magnetic elastic composites obtained using different experimental methods is missing from scientific publications. Certain aspects on this topic were addressed e.g. in [65] as well as recently by us in [39]. On the other hand, essential is the fact, that there are no commercially available magnetic cells for tensile testing machines as well as for rod-shaped specimen rheometry. The easiest solution for creating a homogeneous magnetic field is to use a cylindrical magnetic coil of a given size. The coil provides a magnetic field oriented parallel to the direction of the axial mechanical force F. However, in order to realize the possibility of changing the direction of the applied field relative to mechanical stress, it is necessary to use a magnetic yoke with appropriate geometry. In the case of a tensile testing machine, this feature is most easily implemented for the shear test using a layout with two coupled shear gaps (Figure 11b). However, this configuration requires that the two samples be fully identical, including the conditions of their fixation in the measuring cell. This is not always feasible, especially in the context of soft composites. When using a combination of the rotational rheometer and conventional electromagnetic coil, the direction of the magnetic field with respect to mechanical stress is also different in the case of tensile and torsional loading (Figure 10b). The critical factor determining the correctness of the results obtained using this arrangement is the lack of magnetic elements in the construction of the rheometer, e.g. couplings, rotational axis etc. However, standard commercial device configurations (e.g. various modifications of Anton Paar and Thermo Scientific rheometers) contain magnetizing structural elements that distort the results of torque and axial force measurements. Another important methodological factor is the need to critically assess the raw data received from the instrument. Unfortunately, this is not always possible with commercial devices with proprietary software.

Taking the above into account, the most suitable experimental method to evaluate the magneto-mechanical response of elastic hybrid elastomers is the tensile compression test using table testing machine. Within the method either quasi-static or dynamic loading is possible.The maximum displacement s used for basic characterization of the composites should be limited to the area of linear deformation and to avoid significant influence of radial deformation of the specimen on the measurement results. In quasi-static measurement, a dependence of axial force F on displacement s is experimentally obtained. The ratio F/s gives the stiffness of the material. To determine the tensile modulus E, the measured forces F and displacements s are transformed into stress σ and strain ϵ values, respectively, taking into account the geometry of the specimen. The modulus E is when defined as the slope of the stress-strain curve. The modulus E is when defined as the slope of the stress-strain curve. As a result of the dynamic loading of a viscoelastic material one obtains hysteretic F(s) loops which allow to access such material parameters as the storage tensile modulus E' and the loss factor η. The loss factor in turn represents a ratio between the loss and storage tensile modulus $\eta = \tan\delta = \mathrm{E}''/\mathrm{E}'$. The slope k of the major axis of the force-displacement loop provides the dynamic stiffness of the specimen, while the area A within the loop provides the dissipated energy per cycle of loading $A = \int F ds = \pi \eta k \Delta s^2$ (see as well Figure 12a). The storage modulus E' is when obtained from the slope of the main axis of the stress-strain loop by obtaining values of σ and ϵ from the force F and displacement s, respectively. Figure 12 shows an example of a carbonyl iron-based composite response (particle concentration ~40 vol. %) resulting from dynamic loading in an external homogeneous magnetic field of various induction. For quantitative estimation of influence of a magnetic field on viscoelastic properties of a composite

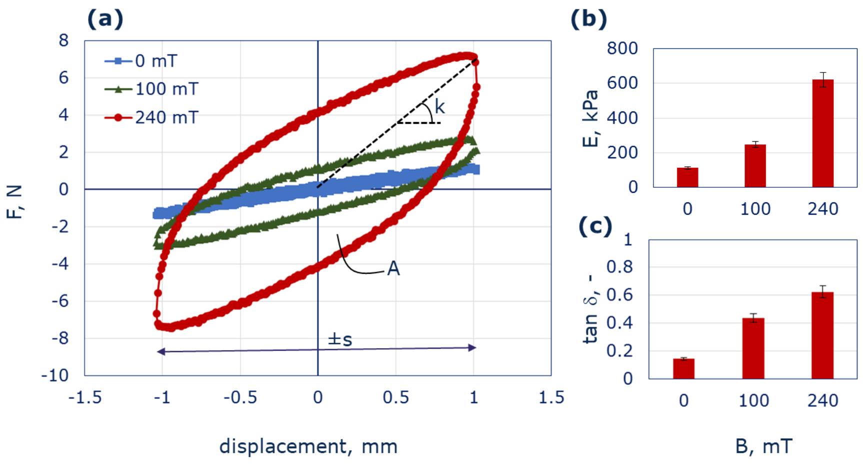

Fig. 12: Force-displacement loops (a), storage modulus E' (b) and loss factor η (c) measured at f=0.1 Hz, ϵ ~5% for a magnetic hybrid elastic composite based on carbonyl iron microparticles with concentration ~40 vol. % at various magnetic fields.

material it is convenient to use the MR effect calculated from the experimentally obtained values of a certain physical parameter of a material. For example, for the elastic storage modulus E' and the loss factor η, the active magnetorheological effect R_a under the externally applied magnetic field B is calculated correspondingly as follows:

$$R_{a\_E'} = \frac{E'(B) - E'_0}{E'_0}, \tag{2}$$

$$R_{a\_\eta} = \frac{\eta(B) - \eta_0}{\eta_0}, \tag{3}$$

where E'_0 and η_0 are storage and loss factor at zero field ($B = 0$). For samples containing a magnetically hard component, the concept of passive MR effect (R_p) is introduced, which takes into account the effect of pre-magnetization on the viscoelastic properties of the composite. Similar to equations (2) and (3), this effect is quantified as

$$R_{p\_E'} = \frac{E'(B_M) - E'_0}{E'_0}, \tag{4}$$

$$R_{p\_\eta} = \frac{\eta(B_M) - \eta_0}{\eta_0}, \tag{5}$$

where B_M denotes a flux density of the external field applied to magnetize a composite sample, prior to it's characterization.

Below is an overview of the experimental results obtained for samples of hybrid elastic composite with different magnetic fillers in the context of magnetorheological effect. All given results are obtained in the axial cyclic loading of cylindrical samples at a frequency of 1 Hz and maximal deformation of 5%. Concentration of the powder in all considered below samples is ∼40 vol.%. The matrix composition is selected so that the initial module of the all samples is more or less same and is E_0 ∼120-140 kPa. Figures 13 and 14 show an influence of the externally applied field B and the used for the pre-magnetization field B_M on the elastic modulus E and loss factor η respectively for hybrid composites with a various filler (Table 1).

specimen	carbonyl iron	spherical NdFeB	irregular NdFeB
s1	100%	0%	0%
s2	0%	100%	0%
s3	0%	0%	100%
s4	25%	75%	0%
s5	25%	0%	75%

Tab. 1: Composition of specimens filler. Overall powder concentration in each specimen is ∼40 vol.%.

the carbonyl iron based composite ($s1$), composites filled with spherical ($s2$) and irregularly shaped ($s3$) NdFeB-alloy particles, as well as composites $s4$ and $s5$ with a mixed powder. In samples $s4$ and $s5$ 25% of NdFeB-alloy particles replaced with carbonyl iron particles. The influence of different ratios of magnetically hard and soft phases on the magneto-mechanical response of hybrid composites was considered in particular in [38]. The current overview provides representative results obtained for samples of mixed composition with 25% carbonyl iron. Sample $s1$ serves more as a reference one, as its passive magnetic tuning is not possible.

First consider MR effect of the composites without pre-magnetization. The field B is not high enough to change initial state of the samples $s2$ and $s2$ containing magnetically hard phase. In this case, the sample $s3$ containing magnetically hard particles of irregular shape reacts to an external field, while the sample $s2$ with spherical magnetically hard particles remains uncontrollable. It is noteworthy that the magnetic susceptibility of particles in sample s3 is lower than that of particles in the sample $s2$. This mismatch of the reaction of the composite to the external magnetic field should be explained in terms of the variable microstructure. As demonstrated in performed microCT studies, see e.g. [37], particles of irregular shape are rotating reversibly in already moderate fields, while their structuring is not occurred as well as a structuring of the spherical magnetically hard particles. The rotation of spherical particles can not be tracked using microCT method, due to obvious reasons, however, it can be assumed according to the results of the magnetic

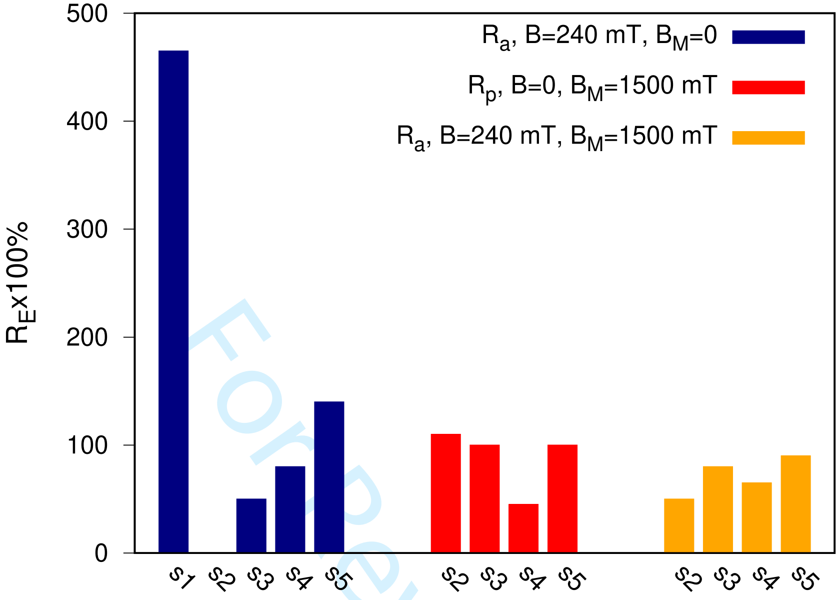

Fig. 13: Active and passive MR effects (R_a and R_p) respective elastic storage modulus E' for composites with various filler.

measurements [49]. Thus, the rotation of irregularly shaped particles in the applied field is sufficient to make the originally non-magnetized composite magnetically controlled. In the sample $s1$, the filler particle material has the highest magnetic susceptibility (see Figure 6). Due to the high dipole-dipole interaction induced by the magnetic field, the particles are aggregated in the matrix. It is obvious that as a result the MR effect of this composite significantly exceeds the MR effect of the sample $s3$. When to use complex filling, that is to add to composites with magnetically hard particles carbonyl iron, it is possible to increase their controllability considerably. Thus, sample $s4$, in which part of spherical NdFeB-alloy particles is replaced by carbonyl iron (25%) becomes actively controlled. In sample $s5$ the same fraction of irregular NdFeB-alloy particles is replaced by carbonyl iron and its MR effect also becomes higher than in the sample $s3$. It can be assumed that the MR effect in the sample $s4$ is caused by formation of structures of particles, while in the sample $s5$, in addition to the structuring, the rotation of irregularly shaped particles additionally affects the magneto-mechanics of the composite.

Application of much higher external fields to a composite containing a magnetically hard phase caused appearance of the remanence magnetization M_r (see

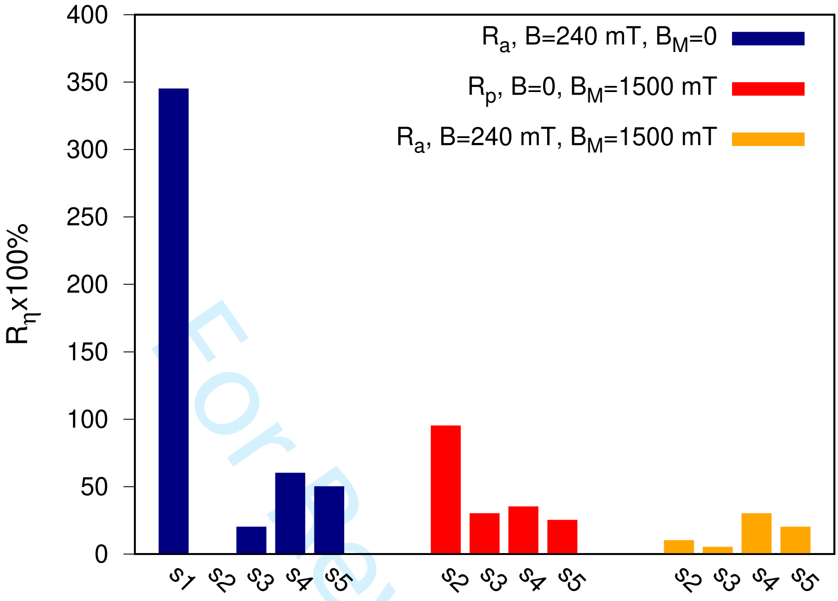

Fig. 14: Active and passive MR effects (R_a and R_p) respective loss factor η for composites with various filler.

Figure 7). Besides the appearance of residual magnetization, the microstructure of such a composite is irreversibly altered [37]. Figures 13 and 14 as well demonstrate how the magnetizing field B_M changes an initial state of the composite specimens filled with various powders. Changes of up to approximately 100% in material elasticity are possible. Replacing the magnetically hard fraction with a soft one reduces the possibility of the passive tuning. This is most noticeable for a spherical NdFeB-alloy particle based composite (specimen $s4$). On the other hand, pre-magnetization provides active control of sample $s2$, but reduces the range of active control of composites with mixed filler $s4$ and $s5$. Comparing an influence of the pre-magnetization on the modulus E' with the influence on the loss factor η one observes that the results for the samples $s2$ and $s3$ are inconsistent. While pre-magnetization increases the control range of the elasticity, their loss factor becomes almost uncontrollable.

Obviously, the explanations for the observed behavior of composites with different fillers are related to microstuctural changes induced by the applied magnetic field. It should be distinguished between the processes of the structuring and rotation of magnetic microparticles inside the matrix. Both processes are confirmed within microstructural observations, e.g. in [37, 66]. Use of the complex mixed powder provides broad passive tuning and remote active control of the composite which can be chosen for the demand of certain application. The NdFeB-alloy powder with an irregular morphology provides enhanced tuning of the hybrid elastic composite viscoelastic response despite weaker magnetization and lower magnetic susceptibility of the powder particles than compared to spherical NdFeB-alloy particles.

5 Summary and outlook

An overview of the basic magneto-mechanical properties of the hybrid elastic composite highly filled with various types of magnetic microparticles is given. Composite based on carbonyl iron particles is compared with specimens containing magnetically hard NdFeB-alloy powders of two different morphology and with specimens containing mixed powder of carbonyl iron and NdFeB-alloy particles. Material based on spherical NdFeB powder only is not controllable in a non-magnetized state, while when using NdFeB particles of irregular shape the composite responds to an external field without having a pre-magnetization. It is related to a rotation of particles within the matrix as follows from performed elsewhere microstrucural observations, see e.g. [37] and [66][2]. The addition of magnetically soft carbonyl iron microparticles into the composite with NdFeB powder enhances the active MR effect of the non-magnetized samples significantly, while the presence of magnetically hard particles still provoke the composite to have a remanence magnetization and, consequently, adjustable initial state. Pre-magnetization of the composite based on a mixed powder slightly restricts the possibilities of its active control.

The presented results on the evaluation of active control and passive tuning of magnetic hybrid elastic composites show wide prospects of their potential applications. However, despite great efforts in the field of experimental characterization of these composites, theoretical approaches are still based on simplified assumptions, e.g. [7, 8, 47, 67, 68][3]. On the other hand, the studies on modeling of magnetic hybrid composites are currently limited to elastic polymers with one type of magnetic particles and does not consider the complex mixed filling of matrices with magnetically soft and hard powders [69, 70][4]. This is due not only to the complexity of understanding the physical processes of interaction between magnetic particles

2 The study [66] is going to be published in the current issue.
3 The study [68] is going to be published in the current issue.
4 The studies [69, 70] are going to be published in the current issue.

of different types and the elastic polymer matrix. It is also problematic that the use of different experimental techniques leads to controversial results, the correct interpretation of which is difficult. Thus, in addition to proposition of further more deeper theoretical approaches, first and foremost ways must be found to develop and implement standardized mechanical testing methods to experimental characterization of magnetic hybrid elastic materials. Furthermore, since a high cyclic stability of the material will obviously required for a technological application of the magnetic hybrid materials, procedures will also have to be developed to investigate and quantify the change of the materials by repeated magnetic stimulation. The corresponding data obtained have to be in turn reflected back into the synthesis process in order to enable higher cyclic stability with constant magnetorheological effects. In this context, the morphology of magnetic microparticles and their chemical-physical coupling with the polymer matrix will play an important role. The first steps in this respect are e.g. studies of the magneto-mechanical coupling of single domain particles into viscoelastic polymeric matrices [71][5].

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5 The study [71] is going to be published in the current issue.

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