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On anisotropic mechanical properties of heterogeneous magnetic polymeric composites

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Summary

The study is devoted to the magneto-mechanical characterization of heterogeneous magnetoactive elastomers based on an elastic polydimethylsiloxane matrix with embedded spherical magnetic soft microparticles and magnetic hard microparticles of irregular shape. An issue of the anisotropic mechanical properties of these smart composites is considered. Non-magnetized and pre-magnetized specimens are characterized using a planar shear and axial loading in an externally applied homogeneous magnetic field. The field direction differs relative to the direction of the field used for the specimens pre-magnetization. Results of the different methods allow to compare the tensile shear moduli for the samples with an initially identical composition. Obtained results demonstrate a strong correlation between the composite behavior and orientation of the magnetic field used for the pre-magnetization of the sample relative to the external field applied to a sample during the test. Composites pre-magnetized in the direction parallel to an applied mechanical force and external magnetic field show higher magnetorheological response than composites pre-magnetized transversally to the force and the field. Application of the external field directed opposite to the direction of the pre-magnetization reduces the observed stiffening. Moreover, in this situation a softening of the material can be observed, depending on the magnitude of the external field and the field used for pre-magnetization.

Introduction

Magnetoactive elastomers (MAEs) belong to a class of smart materials with remote controllable physical properties. These elastomers are heterogeneous composites of a polymeric matrix with embedded magnetic particles. There are a couple of designations used in the scientific literature for these composites, e.g. such as magnetorheological (MR) elastomers, ferroelastomers, magnetic polymers, magnetic gels, ferrogels etc [1,2,3,4,5]. Conspicuous is the fact that there is no unity in the usage of these definitions despite the quite different material compositions and consequently different physical performances. Therefore, it is reasonable to distinguish at least between composites filled with nanoparticles and those filled with microparticles, as well as between composites with soft gel-like matrices and composites with significantly stiffer resin-like matrices. In this context, ferrogel is a suitable designation for a material based on the nanosized particles embedded into a gel-like matrix, while by the term MAE a composite based on an elastomer with magnetic microparticles is to be understood, as is the case with ferrofluids and magnetorheological suspensions [6,7]. It must be additionally noted that materials which are filled with magnetic powder and which are having a very stiff matrix, such as a rubber, vulcanized under pressure, whose rheological properties are not controllable with an externally applied magnetic field, shouldn't be accounted to the MAEs.

As a filler for MAEs, magnetically soft iron microparticles are commonly used. Nevertheless it is as well possible to use a magnetic hard powder, e.g. NdFeB-alloy particles. The magnetic hard component allows the change of the initial state of the MAE tuning the composite remanence magnetization [8,9]. Recently, an approach to use a mixture of magnetic soft and hard particles has been proposed [10,11,12,13,14]. This approach enhances the range of an active control and simultaneously provides a possibility to passively tune the material properties.

A MAE prior to a polymerization process represents a kind of magnetorheological fluid, which contains movable particles. When a MAE is cross-linked under conventional conditions, one deals with a composite with an isotropic particles

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distribution, neglecting or avoiding an effects of the gravitational sedimentation of the particles. Experimental microstructural investigations [15,16], as well as theoretical studies [17,18], demonstrate an appearance of the particles structures in an initially isotropic MAEs and ferrogels when an external magnetic field is applied to an already cross-linked sample. These structures disappear after the field is switched off for MAEs based on a magnetic soft powders. The formation of particles structure as well as particles movements inside an elastic matrix is known to be an essential reason for the magnetorheological effect, i.e. an increase in an elastic modulus and loss modulus of a composite. On the other hand, when an external magnetic field is applied during the sample cross-linking, particles form elongated structures in the field direction are appeared, which are preserved in the cross-linked elastomer network. As a result, a material with the structural anisotropy is obtained [19,20]. It has been shown in the past [21] that composites with a magnetic soft filler that were pre-structured in an uniaxial field exhibit much higher MR effects in an externally applied uniform magnetic field than samples with an isotropic distribution of particles. Additionally, an internal structural anisotropy obviously causes macroscopic anisotropic properties of MAEs. Influence of the direction of the applied field, particles alignment and the mechanical stress on the mechanical properties of the composites with magnetic soft filler was previously studied in [21]. The most significant MR effect was observed for the case when orientations of field, load and particle structures were parallel to each other. In the MAEs containing a magnetic hard component, the particle structures can persist after switching off the external field. For that, the strength of an applied to the specimen magnetic field must be high enough to provide a sufficiently high remanence magnetization. Consequently, an initially not structured MAE with a magnetic hard component will obtain anisotropic microstructures as it has been proved in [16]. The MR effect in pre-magnetized MAEs solely based on a magnetic hard filler as well as in MAEs with a hybrid composition, i.e. based on a mixture of magnetic hard and soft fillers, was recently studied in [14]. The direction of the magnetic field used for the specimen's magnetization, the external field and mechanical force applied during the characterization in [14] were parallel to each other. In [9] a MAE based on a magnetically hard filler was studied using oscillatory shear. Samples were pre-magnetized perpendicular to the shear strain direction applied in the experiments, while an application of an external field during the shear loading was not considered. Moreover, as shown in [22] results for oscillatory shear can be misleading, when performed with a standard rheometric configuration. Thus, to the best of our knowledge, the anisotropy of the MR effect in MAEs with a hybrid compositions has still not received any specific attention. However, it is an important issue which should be addressed towards a thorough understanding of the complex MAEs' behavior and their practical applications. The current experimental study is devoted to the anisotropic mechanical testing of a MAE with a complex hybrid composition. Non-magnetized and pre-magnetized samples based on a polydimethylsiloxane (PDMS) matrix, with a mixture of magnetically soft and hard particles embedded, are studied under the shear and axial loading. These different loading conditions have allowed to compare the elastic modulus E and shear modulus G for samples with initially identical compositions. Due to a different configuration of a measuring setup in the shear and axial loading tests, the direction of the externally applied magnetic field could be changed relative to the direction of the field used for the specimens pre-magnetization.

The paper is organized as follows. First, basic information on the samples composition and fabrication as well as on the used setup and methods are given. Subsequently results of experiments and their discussion are presented subsequently. Finally, conclusions and outlook are given.

Samples and methods

Samples composition and fabrication

Experimental samples of MAE have been fabricated using the two-component silicone elastomer compound (SIEL-254, product of the Russian State Scientific Institute for Chemical Technologies of Organoelement Compounds). The SIEL®-254 compound is initially liquid and can be cross-linked using a hydride-containing agent. Additionally, Baysilone® oil M100 was mixed with the SIEL-254 in a ratio 1:1 to obtain a sufficiently soft matrix. The magnetic filler is a mixture of carbonyl iron powder (BASF CIP grade CC) and magnetic hard NdFeB-alloy powder. Particles of BASF carbonyl iron powder (CIP) type CC are spherical (average size ~5 μm), while particles of NdFeB-alloy powder have irregular geometry with a size in a range between 40-100 μm (Figure 1). The surface of the powders particles is modified using the Baysilone® oil M100 to avoid particles aggregation and, making the particles' surface hydrophobic, to provide a better compatibility with the matrix as reported in [9].The overall weight concentration of powder in samples is ~82%, and the weight ratio between magnetic soft and hard fractions was 1:4. The mixed filler is mechanically stirred into the liquid matrix. After thoroughly stirring and degassing of the suspension, it is cast into moulds and cured at ~100^{o}C. In this way, cylindrical samples with a diameter of 14 mm and a height of 15-18 mm as well as plate-like samples with a size of 12x12x2 mm are prepared (Figure 2). More details on a multi-step process of the MAE synthesis can be found elsewhere, e.g. [14].