A comparison between micro- and macro-structure of magnetoactive composites

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Abstract. A possibility to determine the microstructure of magneto active composites using micro-computed tomography (µ-CT) [1] was investigated in this work. The main interest of the current study has been an observation of the magnetic field dependent shift of individual particles from their initial positions inside an elastomeric matrix. For this purpose a µ-CT system has been combined with a sample holder coupled with two permanent magnets, enabling the investigation of the micro-structure under influence of an external homogeneous field. In the experimental investigations samples based on carbonyl iron particles have been used. The particles have been dispersed in a polymeric matrix and the polymer has been created in the presence of a magnetic field driving structure formation of the particles. After the characterization of the sample in its initial state, i.e. without external stimuli, it has been subjected to the magnetic field and its internal structure has been once more studied by µ-CT. As a result a comparison of the macroscopic change of the sample structure and the particle displacement could be undertaken.

1. Introduction
Magneto active composites, also known as magneto-rheological elastomers (MRE) are a kind of smart materials which consists of soft magnetic particles dispersed in an elastomeric matrix. The mechanical properties of this composite material can be changed actively and reversibly by applying a magnetic field. An essential reason for the property changes is an internal magnetodipolarstriction, i.e. a change of the local positions of the particles, which has been widely studied theoretically, e.g. in [2, 3]. The proposed theoretical models show a connection between the local positions of the particles and the behaviour of the matrix. These effects have to be tested using experimental methods.

To investigate different effects of MREs, it is important to distinguish between two scales: A microscopic scale that describes the behaviour of the soft magnetic particles and the interaction between these particles and the elastomeric matrix and a macroscopic scale that describes the behaviour of the composite in a range that is visible to the eye. A typical microscopic effect is the shift of the particles inside the matrix due to the influence of an applied magnetic field. Macroscopic effects are e.g. dynamic-mechanical deformations of the composite due to an applied mechanical stress and/or a magnetic field, e.g. the shape memory effect [4]. Several papers regarding these macroscopic effects were published in the last years [4, 5].

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Most of these studies were performed on composites, consisting of a silicon rubber as matrix material and carbonyl-iron-powder as particles. Advantage of this composite-system is the liquid state of the silicon components before starting the polymerisation synthesis. This makes it possible to suspend the particles in the silicone and produce samples with an isotropic particle concentration and samples with a chain like structure of the particles parallel to the applied magnetic field, also called anisotropic samples [6].

2. Experimental

2.1. Sample preparation

The particle material that have been used for the preparation were carbonyl-iron powder produced by Höganäs AB with a mean diameter of 35-45µm. Figure 1 shows the volume weighted sum function (circles) and the volume weighted density function (triangles) of the particles, which are used for preparation.

![Graph showing volume weighted sum function and density function of carbonyl-iron particles](image)

As silicon matrix a low-molecular vinyl-containing rubber and a hydride-containing cross-linking agent (compound SIEL™) was used [7]. This elastomeric matrix allows Young’s modulus smaller than 50 kPa.

The work of D. Günther [1] shows, that the best proportion of soft magnetic particles inside the silicon matrix is around 10 wt.%. A higher proportion of carbonyl iron particles causes that the particles cannot be fully resolved with the µ-CT due to their absorption and a lower content of particles causes a lower macrostructural deformation.

The production of the samples was as follows: The powder was mixed to the silicone and added to the crosslinker. After mixing and degassing under vacuum the sample polymerized in a homogeneous magnetic field. For the polymerisation process, we used a water heating system and placed this in an electromagnetic setup from Bruker Corp. The polymerisation time was 2 hours by 95 °C heating temperature and within a magnetic flux density of 250 mT. The results of the preparation were anisotropic samples with a height of 10 mm and a diameter of 8 mm.

2.2. Experimental setup

The analysis of the samples was performed by the same µ-CT system, which was used in [1]. It is based on a nano-focus tube, a movable sample stage and a detector. Especially for studies under the influence of a magnetic field, a new sample holder was produced. A principle scheme of this new sample holder is shown in Fig. 2. The sample holder consists of a plastic tube with a low absorption
coefficient, the sample with a height of 10 mm and a diameter of 8 mm on a table and two adjustable permanent magnets. The magnets are neodymium magnets with a remanent flux density of 1.46 T. This configuration enables to vary the magnetic flux density between the magnets from 0 to 250 mT.

To verify this magnetic field, simulations for the magnetic flux density were undertaken. For this purpose, the simulation software COMSOL FEMLAB Multiphysics was used. The simulation parameters were given by the geometrical properties of the permanent magnets and the remanent flux density of $B_r = 1.46$ T. Figure 3 shows the dependence of the magnetic flux density of the radius in the vertical centre of the sample. The minimum distance of 15 mm between the magnets is defined by the combined height of the sample and the sample stage. The magnetic flux density at minimum magnet distance in the centre of the sample is 250 mT and shows a homogeneity across the radius of more than 90%. At the maximum magnet distance of 130 mm, the magnetic flux density inside the sample equals zero.

The tomographic measurements were undertaken at two conditions, the first tomogram without field and the second tomogram within a magnetic flux density of 250 mT. After tomography the images have been evaluated with digital image processing software.

![Figure 2. New setup for µ-CT investigations of MRE’s under influence of a magnetic field. It consists of two adjustable permanent magnets, a sample stage and a plastic tube with a low absorption coefficient.](image)

![Figure 3. Simulation of the magnetic flux density in the vertical centre of the sample. Shown are the minimal distance (15 mm) and the maximal distance (130 mm) between the magnets.](image)
3. Results and discussion
The undertaken measurements were taken at an X-ray voltage of 90 kV. The magnification of the measurement was nine. This corresponds to a resolution limit of 8 µm which is equal to the measurement error in all measurements.

3.1. Macrostructural behavior
Figure 4 shows the tomogram of the sample without the influence of a magnetic field (left) and within a magnetic flux density of 250 mT. Using a digital image software, the dimensions of the sample were determined. The results are also shown in Figure 4. A deformation of the sample in the vertical direction (z-direction), respectively parallel to the applied magnetic field could be detected. In horizontal direction (x-y-plane), it was not possible to detect a deformation due to the spatial resolution of the tomogram. The result shows an extension of the sample of 60 µm. The sample is elongated in the presence of a magnetic field. This effect, also called magnetodeformational effect in magnetoelastics is described in many publications, e.g. in [8] and can be explained by the dipole-dipole interaction of the magnetic particles in the chains [8].

3.2. Microstructural behavior
Using µ-CT, it is now possible to zoom into the structure and to determine the microscopic behaviour. Figure 5 shows the microstructure of the anisotropic sample for the situation without magnetic field influence (left) and the arrangement with magnetic field influence (right). It can be seen that the particles are arranged in chains, because of the preparation method.

Using the same image processing program as for the evaluation of the macroscopic behaviour it was possible to separate the individual particles from each other. For this step, various filters and the watershed algorithm have been used. The particles were identified by their characteristics such as size, major and minor axes respectively. Using MATLAB with DIPimage it was possible to assign the particles in the case without magnetic influence to the particles within magnetic field influence.

For the associated particles the displacement of the centre points was determined. This shift can be represented by vectors. Figure 6 shows the displacement of the particle centre as vectors. For a better view, the shift in the z-x-plane is shown. Due to the resolution limit, it was found, that there is no difference between the x and the y-direction. This can also explained with the fact that the sample and the magnetic field have been arranged symmetrical with respect to the long axis of the sample cylinder.
This type of analysis requires a large amount of memory. Due to the long computation time, only some particles could be evaluated in the whole sample. Thus only a qualitative description of the particle displacement can be given there. But this result shows a particle shift in vertical direction ($z$), that is one order of magnitude larger than the particle shift in horizontal direction ($x$). This could be explained with the parallelism of the vertical direction and the magnetic field. It is also clear that the shift of the particles in vertical direction is higher than the macroscopic deformation of the sample in the same direction.

**Figure 5.** Microstructural view of the sample, specially the chain like structure of the particles without the influence of a magnetic field (left) and within the influence of a magnetic flux density of 250 mT (right).

**Figure 6.** Microscopic change of the structure of a MRE, shown is the displacement of the centre of some particles at different positions in the Matrix. The axes are not scaled.
4. Conclusions
In this work, a new experimental method has been developed to examine the movement of particles in an elastomeric matrix under the influence of a magnetic field and to evaluate the interaction between the matrix and the suspended particles. It could be shown that the particle movement parallel to the applied magnetic field is higher than the particle movement perpendicular to the applied magnetic field. In addition, it has been found that the particle shift is larger than the macroscopical deformation of the sample. This finding leads to the conclusion that the particles in this kind of MR-elastomer can move relative to the matrix with small influence on the matrix deformation only.

Future work will show the overall behaviour of the particles in the matrix. Due to the gradient in the magnetic field, it could be possible that the particle displacement is different between the centre and the edges of the sample. For this purpose, all of the particles have to be segmented and compared. Also the macroscopic deformation at higher content of iron particles will be investigated.

Acknowledgments
This project is funded by the European Union and the Free State of Saxony.

References
[1] Günther D, Borin D Yu, Günther S, and Odenbach S 2012 X-ray micro-tomographic characterization of field-structured magnetorheological elastomers Smart Mater. Struct. 21 015005
[2] Stolbov O V, Raikher Yu L and Balasoiu M 2011 Modelling of magnetodipolar striction in soft magnetic elastomers Soft Matter 7 8484
[3] Davis L C 1999 J. Appl. Phys. 853348
[4] Stepanov G V, Borin D Yu, Raikher Yu L, Melenev P V and Perov N S 2008 J. Phys.: Condens. Matter 20204121
[5] Böse H and Röder R 2009 J. Phys.: Conf. Ser. 149 012090
[6] Borbarth T, Günther S, Borin D Yu, Gundermann Th and Odenbach S 2012 XµCT analysis of magnetic field-induced phase transitions in magnetorheological elastomers, Smart Mater. Struct. 21 105018
[7] Stepanov G V, Borin D Yu, Odenbach S 2009 J. Phys.: Conf. Ser. 149 012098
[8] Nikitin L V, Stepanov G V, Mironova L S and Gorbunov A I 2004 JMMM 272-276 2072-2073