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The microstructure of magnetorheological materials characterized by means of computed X-ray microtomography

Abstract: Magnetorheological materials are a class of “smart materials”, where mechanical material properties can be tuned by the application of externally applied fields. To accomplish the magneto-sensitive quality, magnetic particles are distributed in a host matrix. In the last year’s interest gained in materials based on solid matrices. In contrast to fluid systems, within a solid matrix, the particles are fixed within the material. This enables an evaluation of the structures formed by the particles by means of computed X-ray microtomography. As known from past investigations, the arrangement and movement of the magnetic particles within the matrix play a major role in determining the overall material properties. Computed X-ray microtomography proved to be a convenient tool, providing important new knowledge about those materials. This paper gives an overview of the application of the method of computed X-ray microtomography on several kinds of solid magnetorheological materials, the broad possibilities of data evaluation, and fundamental results obtained with this method and the described materials.

Keywords: magnetorheological effect; particle structure; smart materials; X-ray tomography.

1 Introduction

1.1 Magnetorheological materials

The core feature of magnetorheological materials is the tunability of mechanical properties, for instance, Young’s moduli, by the external application of magnetic fields. The embedment of magnetic microparticles within a surrounding matrix realizes the magnetic properties of the material.

The first smart materials reacting on magnetic fields were realized as suspensions of magnetic particles in appropriate carrier liquids. Rabinov [1, 2] accomplished the embedment of magnetic micron-sized particles, obtaining the first magnetorheological suspension. This fluid material featured strong yield stress in the presence of magnetic

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fields, thus exhibiting magnetorheological properties. Aiming for long-term stability without sedimentation led to the transition to magnetic nanoparticles. Papell [3] earned a patent on the realization of so-called ferrofluids: A stable suspension of magnetic nanoparticles in a carrier liquid featuring magnetically controllable flow and fluid properties.

Regardless of their numerous applications and usages [4, 5], fluid matrix materials exhibit a range of disadvantages and technical difficulties as leakage, sedimentation, and the overall challenge of realizing a long-term stable material. This issue was addressed by switching from liquid to solid matrix materials. The idea was to provide a more stable and easily feasible magnetorheological material offering comparable properties and features.

The most obvious solution is the usage of a soft elastomer as a host matrix. This approach leads to an elastic and mechanically stable material, where mechanical properties such as Young's modulus can be tuned by magnetic fields. These magnetorheological elastomers are an important subject of research for about 15 years [6–13]. This class of materials brings its own difficulties and most of its behavior characteristics and properties are not fully understood yet. Nevertheless, several technical and scientific applications of magnetorheological elastomers were successfully realized [14–22]. More information on technical applications of magnetorheological elastomers can be found in the articles “Field-controlled vibrational behavior of magnetoactive elastomers used as adaptable sensor elements” and “Actuator systems based on a controlled particle-matrix interaction in magnetic hybrid materials with the application for locomotion and manipulation”, both found within this issue.

Besides the choice of the matrix material, the properties of the embedded magnetic particles play a major role in the macroscopic material behavior. In addition to the most commonly used iron particles, magnetically hard particles are of special interest in the research of the past few years [23–27]. Most recent investigations even apply mixtures of magnetically soft and hard particles to combine the features and advantages of both particle types [25, 28]. Information on the impact of the composition of magnetorheological elastomers on their properties can be found in the article “Synthesis of MGE with a wide range of magnetically controlled properties”, found in this issue as well. Continuing the pass of searching for suitable matrix material led to the development of magnetorheological foams [29–33], and magnetorheological gels [6, 34–36], adding further aspects and functional properties to the obtained materials.

1.2 Evolution of evaluation methods

Already at the beginning of research regarding magnetorheological materials, it became apparent, that the magnetically induced movement and rearrangement of the magnetic particles within the matrix form the key influence determining the macroscopic material behavior. Thus, besides the evaluation of global material properties in

dependence of several material parameters, research focused on the evaluation of the internal particle structure.

Changes in the internal structure have been observed by microscopy [11, 37–39] or small-angle neutron scattering (SANS) [40, 41]. These methods provide either a two-dimensional projection of the internal structure or a statistical average over the particle ensemble.

With technical advances in tomography and digital image evaluation, computed X-ray microtomography turned out to be one of the most convenient methods for this task [12, 23, 42–48]. It enables a nondestructive evaluation of three-dimensional information and thus allows the evaluation of geometrical information of several thousands of particles at once. Furthermore, it is possible to combine the measurement with additional triggers such as mechanical strain, and most importantly: magnetic fields.

The high resolution of X-ray microtomography combined with sophisticated algorithms in digital image processing provides the possibility to obtain single-particle resolution. These recent advances allow the tracking of particles after the application of magnetic fields or mechanical loads. The obtained detailed geometrical information of the particle structures can serve as input data for theoretical descriptions of the material. Furthermore, the experimental data can provide a benchmark for proving the results of numerical simulations of the material behavior [49–57].

The scope of this contribution will be to outline the technical possibilities of computed X-ray microtomography for evaluating particle structures inside magnetorheological materials and to describe the used methods of digital image processing. This description of the evaluation of microscopic particle distributions will be accompanied by investigations of the mechanical properties of those materials to provide the link between microscopic structural changes and macroscopic material behavior.

2 Computed X-ray microtomography

The method of X-ray absorption tomography is based on a mathematical theorem by Johann Radon [58] stating, that the two-dimensional distribution of a quantity A can be evaluated if the line integrals of this quantity are known for all angles of observation.

Lambert-Beer's attenuation law determines the attenuation of X-rays when passing through the material.

$$I(y) = I_0 \cdot e^{-\mu y} \quad (1)$$

I denote the transmitted intensity, I_0 the initial X-ray intensity, μ the linear absorption coefficient for X-rays, and y the thickness of the passed material. The term in the exponential function changes into a line integral over the absorption coefficients along the pass of the X-rays, if the sample is composed of different materials with different absorption coefficients.

$$I(Y) = I_0 \cdot e^{-\int_0^Y \mu(y) dy} \quad (2)$$

The negative logarithm of the ratio between the transmitted and the incident X-ray intensity is called the projection:

$$p(Y) = -\ln\left(\frac{I(Y)}{I_0}\right) = \int_0^Y \mu(y) dy \quad (3)$$

It provides the line integral of the absorption coefficient along the pass, the X-ray's pass took through the sample. As a result, if X-ray absorption images are recorded under different observation angles, Radon's theorem can be discretely realized.

To get a three-dimensional distribution of the absorption coefficients, a reconstruction of the projection images is conducted. Commonly, algorithms based on the Fourier slice theorem are used. With this approach, the Fourier transform of a parallel projection under a certain observation angle is equal to a line within the two-dimensional Fourier transform. Thus, if a Fourier transformation of the projection data is taken for many observation angles, a discrete representation of the two-dimensional Fourier transform of the distribution of the linear absorption coefficients in the sample is obtained.

If conventional cone-beam X-ray tubes are used to capture the absorption images, the Fourier Slice Theorem can be implemented with the Feldkamp–Davis–Kress (FDK) algorithm. Within the resulting 3D images, the gray value corresponds to the spatial density distribution of the material, as the linear absorption coefficient depends on the density. Those images can be processed to extract the distribution of particles inside a magnetorheological elastomer. A rendered 3D model of reconstructed tomography data is shown in Figure 1, depicting particle structures in a magnetorheological elastomer.

Figure 1: The images show magnetic particles obtained from reconstructed tomography data. The pictures are excerpts of a dataset counting approx. 12.500 particles. The left side shows the initial isotropic particle structure, the right side shows the chain-like structure, as a result of an application of a 2T magnetic field before tomography. The images show the necessity of a particle separation process after binarization to evaluate the individual particles [12, 23].

Many complex aspects like the energy dependence of the absorption coefficient and the general treatment of artefacts in tomographic data sets are not discussed within this introduction of computed X-ray microtomography. More detailed information regarding these points are given in [59, 60].

The tomographic data sets presented in this contribution were recorded using the home-built tomography setup TomoTU [42]. This setup uses a conventional nano-focus cone-beam X-ray tube with a maximum acceleration voltage of 160 kV and a focus spot size of $1.5\text{ }\mu\text{m}$. A sample manipulation stage, consisting of various positioning stages, a rotational stage to rotate the sample enables the capture of absorption images from different observation angles. Here, a directly converting X-ray detector (Shad-O-Box 6K HS) with 2304×2940 pixels and a pixel size of $49.5\text{ }\mu\text{m}$ is used.

By varying relative positions of the X-ray tube, sample, and detector, a magnification of up to 20 can be realized. This enables a spatial resolution of about $2\text{ }\mu\text{m}$ in the final tomogram. The setup of TomoTU is pictured in Figure 2. The insert shows a sample stage, capable to apply different stimuli on magnetorheological elastomer samples during tomography.

Figure 2: This CAD model visualizes the main components of the used tomography setup TomoTU [42]: the detector unit (left), the rotating sample stage (middle), and the X-ray source (right). The insert shows the sample setup which provides the stimuli to the sample. Two permanent magnets provide magnetic fields of up to 270 mT during tomography and stress can be applied to the sample by a plunger [12, 47].

2.1 Application of magnetic fields and mechanical stress

The sample stage shown in Figure 2 contains two permanent magnets positioned below and above the sample holder, their distance can be varied. This setup can provide homogeneous magnetic fields of up to 270 mT with homogeneity of 94% within the sample dimensions [23]. Furthermore, this setup is equipped to apply mechanical loads to the sample. With this feature experiments with coupled external magnetic and mechanical stimuli can be conducted [47]. To ensure comparable magnetic fields within accompanying mechanical measurements, the used stress-strain-testing device is equipped with two permanent magnets in the same manner. This setup was successfully used in several previous works, providing valid information about the behavior of the investigated materials under influence of magnetic fields [23, 45, 47, 61].

2.2 Digital image processing and particle separation

As visible in Figure 1, a direct evaluation of the particle geometry and location from reconstructed tomography datasets is not possible. The raw data as a result of the reconstruction consists of gray value images. A simple thresholding does not lead to a satisfying extraction of the particles, as those particles appear very often connected to each other. To accomplish a precise separation of the particles without altering their actual shape, sophisticated image processing is necessary. Furthermore, the usage of highly absorbent neodymium-iron-boron (NdFeB) particles lead to pronounced reconstruction artefacts and decrease the overall image quality of the raw data. The applied particle separation procedure, as described in this chapter, was conducted with Matlab and DipImage [62]. The process with its intermediate steps is visualized in Figure 3.

Figure 3: Separation process of the particles, converting raw gray value images, as given by the reconstructed tomography data, to precisely separated and labeled particles. An explanation regarding the individual steps is given in the text.

- Image 1: Gray value image as a result of tomography data reconstructions. The particles are visible, along with artifacts, noise, and blur.
- Image 2: After an application of a moderate median filter to reduce noise, local maxima indicating the particles are detected. Several particles will exhibit two or more maxima, which would result in over separation. As those maxima are very close to each other, this issue can be solved by a binary dilatation, which joins those maxima. After these steps exactly one maximum for every particle is present.
- Image 3: The negation of the initial image forms the base for the following watershed algorithm.
- Image 4: With images 2 and 3 a seeded watershed algorithm is applied, providing the borders which will separate the particles [63].
- Image 5: A threshold is applied to accomplish the transition from a gray value image to a binary image. With the tomography results presented here, a local threshold was calculated. As seen in image 1, the space between particles very close to each other is often brighter than the rest of the background. This issue leads to unsatisfactory results when applying a fixed threshold to the whole image. For the calculation of the local threshold, an additional strongly smoothed version of the original image is involved. The local threshold is calculated with

$$D_5 = D_1 > (A \cdot D_{1\text{smoothed}} + B) \quad (4)$$

where A is a factor, B is a global offset and D denotes the images according to Figure 3. A and B have to be adapted to the type and quality of the provided raw data to accomplish optimal results. The offset B is chosen to match the resulting proportion of detected particle area to the actual volume concentration of particles, which is determined during synthesis. The resulting binary image shows the particles, still touching, with rough boundaries and artifacts.

- Image 6: Image 4, showing the borders, is subtracted from the binary image 5. This finally provides clearly separated particles.
- Image 7: A combination of binary erosion and dilatation eliminates most remaining reconstruction artifacts, smoothes the particle surfaces, and restores the particle shape to a quality which corresponds to the visual appearance of the particles — found in the initial gray value image — As the last step, labeling is conducted to distinguish the individual particles.

The final data obtained by the separation process enables an evaluation of all desired geometrical information for every individual particle. This procedure is applicable to samples containing several 10 thousands of particles, evaluating all at once. Furthermore, the obtained geometrical information is accurate enough to distinguish the particle from its neighbors. This enables the recovery of the same particle in another data set and thus enables particle tracking. Figure 4 visualizes the results of particle separation process by comparing a 3D particle structure before and after separation.

Figure 4: These images visualize the impact of the particle separation process on the dataset already shown in Figure 1. The sample contains 40 wt% of NdFeB-particles after a magnetization at 2 T. The left side shows the results of a simple thresholding, the right side shows the result of the particle separation process. The pictures are excerpts of a dataset counting approx. 12.500 particles [23].

3 Data evaluation

3.1 Global structure evaluation

As a first step tomographic data sets enable an evaluation of the overall spatial distribution of magnetic particles inside a magnetorheological elastomer. This is of special interest for anisotropic magnetorheological elastomers, as those which were synthesized in presence of a magnetic field. The magnetic particle interaction induced by the applied magnetic field leads to a structure formation of the particles, as the particles are able to move freely within the liquid matrix as long as the crosslinking process is not finished. This leads to anisotropic magnetorheological elastomers with fixed chain-like or rod-like particle structures

With such samples, the dependence of the obtained particle structure on characteristic parameters is of central interest. In previous investigations, the impact of the particle concentration and the magnetic field applied during crosslinking on the resulting anisotropic particle chain structures were conducted [42, 43]. It was shown that varying particle concentration can lead to various columnar, rod-like, and tubular structures [42]. The variation of the magnetic field applied during crosslinking leads to increasing chain diameters and chain-to-chain distances with stronger fields [43].

With recent investigations, a visual insight into the obtained reconstructed tomography data sets remains an important benchmark to enable a critical check regarding the found results.

3.2 Particle positions, homogeneity, and pair correlation function

Concerning image processing, the analysis of structures inside anisotropic magnetorheological elastomers, as outlined in the previous chapter, has been quite simple. An analysis on a single particle basis requires the separation of the particles as described in Section 2.2. After the successful separation of the particles, the position of every individual particle can be evaluated. With this information, the spatial homogeneity can be examined. As sedimentation of the particles is a known problem within sample synthesis, the validation of spatial homogeneity is a crucial necessity for almost every study presented here [23, 47, 48, 61, 64].

The pair correlation function (PCF) can be used to evaluate particle structures based on the particle positions. For theoretical descriptions of the material behavior, the spatial distribution of the particles is of central interest [65, 66]. The PCF is a statistical method which describes the relation between macroscopically measurable density ρ and the arrangement of microscopic objects as the probability to find an object at the radius r around a reference object [67, 68]. The method evaluates near- and long-range order or crystal-like structures of object ensembles. Thus, the PCF is widely used to characterize atomic structures in magnetic materials [69–71] and magnetically altered particle structures in ferrofluids [72, 73]. Up to this point, no direction depending on information can be obtained. Thus, the benefit of this method directly applied to anisotropic structures is limited. Two-dimensional approaches (radial distribution function, RDF) can be applied to evaluate macroscopic deformation of particle structures [47, 66] as well, but is limited to the evaluation of distances between particle chains, whereas the PCF enables an evaluation of three-dimensional data. To obtain information regarding different spatial directions, the method was modified to satisfy this requirement [61].

To realize a direction-dependent quality, a new control volume was introduced. Only particles with a limited angle between their directional vector and the considered spatial direction were taken into account for evaluation. The new control volume was defined as an intersection of the original spherical shell with a cone, opening in the considered direction. A more detailed explanation of the direction depending approach is given in [61]. Figure 5 visualizes the control volumes for the three spatial directions used here. The PCF is calculated separately for every one of the three spatial directions, giving information on the distance distribution of particles regarding the three directions. A similar approach has been chosen in ref. [74].

3.3 Particle size distribution, shape, and orientation

In addition to the particle positions, geometrical information of the individual particles can be evaluated from the tomography data. This procedure is applicable for larger particles, as those are most often anisotropically shaped. Furthermore, larger particles

Figure 5: To realize a direction-dependent approach of the pair correlation function (PCF), a new control volume (full colored lines) was introduced as an intersection of the spherical shell (dashed gray lines) and a cone (dashed colored lines). Only objects located within this new control volume are taken into account for evaluation. Only one cone for each direction is shown here to enhance visibility. For calculation, a pair of cones for positive and negative direction is used. [61]

are necessary to obtain a sufficient resolution to precisely evaluate the geometrical information of the particles. Up to this point of technical advances with the used setup, this accounts for particles larger than approx. $40\ \mu\text{m}$. The geometrical properties which can be evaluated include i.e., particle volume or surface area. The shape of most particles can be approximated with an ellipsoid, enabling the evaluation of dimensions and directions of the three ellipsoid diameters of every individual particle.

Zingg's classification [75, 76] provides a convenient approach in evaluating particle shapes. The values of the three ellipsoid diameters as obtained by tomography were used to calculate the aspect ratios a/b and c/b . The longest ellipsoid diameter refers to a , the intermediate to b , and the short ellipsoid diameter to c . The graph depicted in Figure 6 shows the aspect ratios of approx. 11.000 particles, giving an impression of the distribution of particle shapes found in a magnetorheological elastomer containing NdFeB-particles with particle sizes ranging from $100\text{--}200\ \mu\text{m}$.

Furthermore, the gathered geometrical information of the particles can be used to make global statements about changes in the internal arrangement of the particles induced by magnetic fields. With anisotropically shaped particles their rotation to align to magnetic fields can be observed. This alignment of particles is visible in Figure 4. To evaluate the orientation of the particles, the angle of the ellipsoid axes can be measured. To calculate a characteristic angle between the particle and the magnetic field, the angle of the longest ellipsoid axis, called β , is evaluated. The here used NdFeB-particles feature a strong magnetic anisotropy leading to their orientation perpendicular to the applied magnetic fields. A phenomenon which is discussed in detail in ref. [23].

Figure 6: Zingg's classification of particle shapes depending on their volume applied to approx. 11.000 NdFeB particles, present in a magnetorheological elastomer.

3.4 Particle tracking

Aside global statements about the particle positions and geometrical information of the particles, an identification of particles allow tracking of particle motion between two situations. This procedure demands a very precise separation of the particles, as changes in measured geometrical properties of one particle from one to the other data set prohibits a correct assignment of the same particle in two data sets. Furthermore, it is necessary to equip the sample with a fixed spatial reference. For the specific experiments on magnetorheological elastomers described here, two small copper wires attached to the samples were used. The fixed position of this reference enables registration of several tomographic data sets, captured at different external stimuli.

For the first step, all particles in a near neighborhood of the original particle position as present in the first data set are taken into account in the second data set. This neighborhood has to be larger than the maximal possible translation of the particle from one situation to the other. In the next step, all those found particles are taken into account for an assignment to the original particle. To check the quality of the assignment of the particles between the two different data sets, a quality function which compares certain characteristics like the particle volume V and surface A , it's asphericity $P2A$ [29] and position x, y, z , was introduced [12]. This enables a validation of whether a correct assignment has been achieved or not. The following two Eqs. (5) and (6) were used to quantify the quality of a particle assignment. The particles considered for an assignment are named i and j . The smaller the values of G_{position} and G_{shape} , the higher the possibility of a correct assignment becomes. Thus the best fitting candidate for an assignment is chosen for every particle.

$$G_{\text{postion}} = \sqrt{(x_j - x_i)^2 + (y_j - y_i)^2 + (z_j - z_i)^2} \quad (5)$$

$$G_{\text{shape}} = \sqrt{\left(\frac{V_i - V_j}{V_i}\right)^2 + \left(\frac{A_i - A_j}{A_i}\right)^2 + \left(\frac{P2A_i - P2A_j}{P2A_i}\right)^2} \quad (6)$$

Using this procedure of particle assignment, it is possible to allocate and identify several thousand particles in one sample and to quantify changes in their spatial location and orientation induced by the external stimuli. Now it is possible not just to evaluate the present distribution of particle angles, but to calculate the exact rotational angle for every individual particle from one data set to the other. Furthermore, translation of the particles can be evaluated and particle trajectories can be observed [23]. Figure 7 shows correctly assigned particles from two datasets, one captured with and one without an application of a 240 mT field.

Initially, this procedure was limited to very small particle concentrations [45]. Recent advances enabled particle tracking at a particle concentration of $\Phi = 40$ wt%. Now, this method can be applied to materials which exhibit magnetorheological

Figure 7: Here, the assigned particles without (green) and with a 240 mT field (red) are shown. The black lines represent the trajectory of the assigned particles. A rotation of the particles is visible as well. [23]

effects [23], which allows a direct linking of results concerning mechanical behavior and particle movement.

3.5 Tomography data as input for calculation and simulation

As stated before, the experimental data can be used as data input for theoretical investigations [46, 49–51] and can act as a benchmark for theoretical predictions and simulations [52–55].

The combination of experimentally obtained results providing a comprehensive description of macroscopic and microscopic material properties and internal particle structures with theoretical modeling and simulation gives the scale bridging approach required for a deeper understanding of the magnetic field-driven complex behavior of magnetorheological materials. Only by combining theoretical and experimental investigations of identical materials gives access to key parameters, for instance, the internal magnetic fields, which will provide fundamental new results regarding magnetorheological materials in the future. More information on using tomographically captured particle data as an input for theoretical approaches can be found in the articles “Multiscale modeling and finite element simulation of magnetorheological elastomers based on experimental data” and “Modeling and theoretical description of magnetic hybrid materials - bridging from meso-to macro-scales” within this issue.

4 Magnetorheological foams

Magnetoelastic foams are a relatively new kind of magnetic field-responsive smart materials developed in the last few years [30–33]. Owing to their high elasticity, soft foams loaded with magnetic particles show a great potential for use as magnetorheological material. Because of their porous structure, the arrangement of particles in the polymer leads to highly complicated structures and complicated magnetic and mechanic properties. The mechanical properties of foams are strongly influenced by their structure. The pore size distribution can be interpreted as a universal description of the foam structure in general. Detailed knowledge of the distribution and its dependence on given process parameters are the basis for a better understanding of the properties of the foam and the foaming process. The foam structure can be evaluated by Computed X-ray microtomography. Figure 8 shows an excerpt of evaluated foam pores, captured by X-ray microtomography.

It was found, that the Weibull cumulative distribution allows a well-fitting description of the pore volume distribution of the polyurethane foam discussed in refs. [29] and [77]. Furthermore, it was shown, that the pore volume distribution function is changed in a specific way if carbonyl iron particles are incorporated. Figure 9 shows the

Figure 8: Foam pores of a polyurethane foam sample as obtained by tomography after the separation process [29].

Figure 9: A Weibull plot showing pore volume distribution functions of polyurethane foams with different amounts of carbonyl iron particles. A strong increase in the number of small pores with the addition of particles is visible [29].

pore volume distribution functions of polyurethane foams with varying concentrations of carbonyl iron particles.

The observed effects can be explained with known foam-stabilization mechanisms. The drainage of liquid foam is retarded by the particles. As a result, the process of merging smaller pores to larger ones is decelerated [29]. Furthermore, the impact of a magnetic field applied during the foaming process of polyurethane foams loaded with

carbonyl iron particles was investigated [77]. An impact of the field on the pore volume distribution function is apparent and elongation of the pores as a result of field application was verified [77].

5 Magnetorheological elastomers containing carbonyl iron particles

Carbonyl iron powder is the most widely used choice for magnetic particles when synthesizing magnetorheological elastomers. Those materials provide strong and fully reversible magnetorheological effects, meaning an increase of Young's modulus induced by an applied magnetic field [47, 78, 79]. The magnetorheological effects of MRE is calculated from Young's moduli measured with an applied magnetic field E_B and without E_0 , using

$$\text{MRE} = \frac{E_B - E_0}{E_B}. \quad (7)$$

Computed X-ray microtomography offers an insight into the changes in microscopic particle structure which is highly responsible for the macroscopic mechanical behavior. A reversible formation of particle chains can be observed if the magnetic field is applied during tomography. Figure 10 shows the particle structures present in a magnetorheological elastomer with 40 wt% of carbonyl iron particles. The application of a 250 mT field to this material leads to a strong rearrangement of the particles to particle chains. This behavior is accompanied by a measured magnetorheological

Figure 10: The images show a 3D excerpt of the reconstructed tomography data after the separation process. Here 40 wt% of carbonyl iron particles were used. On the left, the initial particle microstructure without a magnetic field is shown, on the right the same sample at a field of 250 mT, applied vertically. Here, particle chains are present [47].

effect of $\text{MRE} = 633\% \pm 55\%$. The samples in these investigations feature Young's modulus of $E_0 \approx 10 \text{ kPa}$.

The evaluation of particle angles from these tomographic data sets shows a significant rotation of the particles and an impact on the radial distribution function if a magnetic field is applied [47]. The application of mechanical stress during tomography was incorporated to give a more realistic reflection of the material behavior at the measurement of Young's moduli, where mechanical stress is present as well. An evaluation of the radial distribution function of the particle structure in presence of mechanical stress of $\varepsilon = 12\%$ and the magnetic field shows a significant shift of the first maximum indicating the distance of the particles within the chains. This shift represents a reduction of particle distance and thus compression of the particle chains [47]. Figure 11 shows these results.

The evaluation of the volume of every individual particle enables an analysis of the particle size distribution, as conducted in [51, 64]. Thus it was shown, that the particle size has a severe impact on the magnetorheological properties of magnetorheological elastomers composed from particles with different particle size distributions [64]. Figure 12 shows 3D models of reconstructed tomography data sets from samples composed of four different sieve fractions of carbonyl iron particles. As seen in Figure 13 their size distribution can be evaluated by computed X-ray microtomography. With these samples it was shown, that increasing particle size leads to increasing magnetorheological effects [64], from $\text{MRE} = 30 \pm 5.5\%$ for the smallest particles to $\text{MRE} = 56 \pm 7\%$ for the largest particles. The particle concentration of $\Phi = 40 \text{ wt\%}$ was kept constant. With the evaluation of the particle shape distribution by means of

Figure 11: The graph shows the radial distribution function of the particle ensemble in presence of a 250 mT field applied in an axial direction, with and without $\varepsilon = 12\%$ mechanical strain. The distribution function $g(r)$ is calculated perpendicular to the field direction. The shift of the maximum to smaller distances indicates compression of the particle chains. [47]

Figure 12: From left to right, excerpts of the 3D models of reconstructed tomography data sets visualizing particles with of size fractions of 80–100 μm , 63–80 μm , 40–63 μm , and 20–40 μm [64].

Figure 13: Volume weighted size distributions of the particle size classes obtained by tomography. The results correspond to the visualized particles in Figure 12. The particle sizes are given as the largest ellipsoid diameter. Plotted are the measurement results and a Gauss fit. [64]

Zingg's classification, it was shown, that the particle shape remained nearly constant through the particle size classes, indicating that the found effects can be clearly addressed to the particle size and not their shape.

6 Magnetorheological elastomers containing neodymium-iron-boron particles

The usage of magnetically hard NdFeB-particles provides additional functionality to magnetorheological elastomers compared to those containing magnetically soft iron particles. The possibility of remanent magnetization enables nonreversible structure

formation and change in mechanical properties by the application of strong magnetic fields. As shown in ref. [23] the known features like particle rotation and magnetorheological effect remain mostly reversible when using small magnetic fields. The tomography data corresponding to the results presented in the following paragraphs was already shown in Figures 1 and 4. The used samples feature a Young' modulus of $E_0 \approx 5$ kPa and exhibiting a magnetorheological effect of $MRE = 60\%$ after magnetization at 2 T. The NdFeB-particles sizes range from 100–200 μm . As stated before, the particles feature a strong magnetic anisotropy leading to their orientation perpendicular to the applied magnetic fields. This behavior was analyzed by means of X-ray diffraction measurements [23].

Figure 14 shows the distribution of the angle between the longest ellipsoid axis of the particles and the direction of the magnetic field applied to the elastomer called β , for six different magnetic field situations. First, the angle has been evaluated for all particles, for a situation without any field applied. Afterward, a magnetic field of 240 mT has been applied and again, the particle orientation has been evaluated. Finally, the magnetic field has been removed to check, whether hysteresis in particle orientation appears. As one can see, the application of the magnetic field forces an alignment of the long major axis of the particles with the magnetic field direction, increasing the percentile of the total number of particles with small angles. After switching of the magnetic field, most of the particles rotate back to their initial position and only a small portion remains a bit more aligned with the magnetic field direction, a matter of fact which shows that the particles are well connected with the polymeric network, which exhibits a strong restoring force, nearly disabling any kind of hysteretic behavior. This mostly fully reversible rotation was further evaluated by scanning-

Figure 14: The frequency distribution of the particle angle β of all evaluated particles indicating a rotation of the particles induced by the magnetic fields [23].

confocal microscopy [23, 80, 81], proving a fully elastic deformation of the surrounding matrix, leading to a reversible rotation. This knowledge can be used to assign the observed nonreversible effects to the nonreversible magnetic behavior of the magnetically hard particles.

After the sample is magnetized at a 2 T field, the particles feature a pronounced remanence, leading to a nonreversible particle chain formation. Furthermore, the magnetization leads to a strong increase of the amount of particles with a large angle, indicating a severe orientation of the particles perpendicular to the applied field. Even at that state, an application of the 240 mT field leads to further rotation, which is partly reversible if the field is removed.

With the conduct of particle tracking, the rotation of the particles as seen in Figures 1 and 4 can be evaluated in detail. To do so, the difference of the particle angles between before and during application of the 240 mT field is calculated as $\Delta\beta = \beta_B - \beta_O$. The results are visualized in Figure 15. With these results a mean rotational angle of the particles can be evaluated, a result which is not within reach without particle tracking. Furthermore, a clear dependence of the initial particle angle β_O on the resulting rotational angle $\Delta\beta$ was found, and a significant impact of the particle shape [23].

As stated before, because of the remanent magnetization of the neodymium-iron-boron particles, the chain formation induced by a 2 T field is nonreversible. The direction-dependent PCF enables a convenient evaluation of the chain formation process. Figure 16 shows results for the PCF in chain direction of a NdFeB-particle loaded elastomer after application of magnetic field up to 2 T.

Figure 15: Particle tracking enables the calculation of the change of the angle for every particle individually, providing the rotational angle $\Delta\beta$. Regarding the Gaussian fit, the results show a significant rotation of every particle of approx. 10° for the unmagnetized material, and approx. 5° after magnetization at 2 T [23].

Figure 16: PCF regarding the z-direction for the particle structure obtained by tomography after application of magnetic fields up to 2 T oriented along the z-axis [61].

The results shown in Figure 16 correspond to the particle structures shown in Figure 17. As visible, the particle chains are not as prominent as those found with carbonyl iron particles as seen in Figure 10. Nevertheless, the evaluation of the direction-dependent PCF offers a high sensitivity detecting anisotropy, even for less pronounced particle chains.

Figure 17: Excerpts of three reconstructed 3D models obtained by tomography showing the particle structure. (a) Shows the initial structure before application of any fields, (b) was captured after application of $B = 1250$ mT, (c) after $B = 2000$ mT. At this point particle chains in the field direction, in this picture: vertically, are clearly visible [61].

7 Summary and outlook

The microscopic arrangement and movement of the embedded magnetic particles is the key parameter governing the macroscopic mechanical behavior of magnetorheological materials. Computed X-ray microtomography provides a sophisticated method to evaluate the particle structure, giving information on particle distribution, individual particle positions, and geometrical properties of the particles. With advanced image processing methods, the obtained 3D information can be processed to enable evaluation methods ranging from overall structure analysis to evaluations based on single-particle information and even particle tracking. With this toolbox, the results obtained by accompanying measurements regarding the macroscopic mechanical behavior can be linked to the observed changes in particle structure. This combination provides a comprehensive and coherent description of the material properties and enables a deeper understanding of the underlying physical mechanisms found with magnetorheological materials.

Besides outlining the method of X-ray microtomography applied to magnetorheological materials, the presented work gave an overview of findings, which were not within reach without the accomplished recent advances with computed X-ray microtomography and the accompanying digital image processing methods.

The presented results show the potential of this method and enable future investigations to systematically analyze the important characteristics of magnetorheological materials as choice and embedment of particles, varying matrices, and synthesis methods. A comprehensive comparison of these results will provide important knowledge to approach one of the main goals of research regarding those materials: A targeted tailoring of magnetorheological materials to fit their desired properties.

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