Modeling of Effect of Particle Size on Macroscopic Behavior of Magnetorheological Elastomers

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Abstract
In this work we report on an investigation of the effect of the magnetic particles size on the effective macroscopic behavior of magnetorheological elastomers (MREs). MREs are a class of smart materials known for their tunable deformation. They are composite materials which consist of magnetically permeable particles in a non-magnetic polymeric matrix. When subjected to an external magnetic field, MREs respond by changing their stiffness and damping properties accordingly. The property of MRE to change their mechanical properties is widely known as the magnetorheological effect. Several factors significantly influence the magnetorheological effect such as the polymer matrix, particles-volume fraction, properties, and size of the magnetic particles. In this study, using finite element simulation we determine the correlation between the latter and the macroscopic behavior of MREs. Based on continuum formulation theory, the constitutive and geometric properties on the microscale are considered to predict the composite's macroscopic behavior by means of a computational homogenization. Using COMSOL Multiphysics software, the magnetic and mechanical fields were defined and resolved. For a constant particle-volume fraction (φ=20%) and varying mean particle sizes (Φ=5, 10, 20 and 30 µm), a two-dimensional representative volume element (RVE) was developed, and applying periodic boundary conditions the simulations were performed for isotropic (unaligned) and anisotropic (aligned) microstructures. From the results, the particle size is found to have a significant effect in the mechanical response of the MRE materials. More specifically, the magneto-induced strain effect is observed to decrease with increase in particle sizes. Also, increasing the particle sizes, is observed to lead to a linear increase in the inter-particle distance for the aligned MRE when the sample is deliberately configured such that the vertical distance between the particles is kept constant for all the particle sizes.

Keywords: Magnetorheological elastomers, Finite element analysis, COMSOL Multiphysics, Macroscopic behavior, Magnetoelastic coupling.

Introduction

Magnetorheological elastomers (MREs) are composites consisting of a polymer matrix filled with micron sized magnetizable particles, typically iron particles. Due to their strong magnetoelastic coupling properties, these materials exhibit field-dependent material properties when under the effect of externally applied magnetic field. Specifically, their mechanical properties including stiffness and damping changes when subjected to an external magnetic field [1], [2]. The capability of MREs to respond rapidly, controllably and reversibly to the effect of external magnetic field has been found useful for a variety of applications including sensors, actuators, adaptive engine mounts, tunable vibration absorbers, and vibration isolators [3].

As a result, much attention has been focused on improving the efficiency of these materials, trying to achieve higher tunable modulus amplitude under magnetic field. Several factors have been found to significantly influence the magnetorheological (MR) effect, such as matrix modulus, plasticizers, working modes, magnetic field strength, type, concentration and distribution of magnetic particles [4]–[6]. Among those, the particle size is also found to be of great importance to the MR effect. Using experiment, Lokander et al [7], showed that unaligned MRE material with irregularly shaped particles with diameters sizes < 60 µm had greater MR effect than the same with 3-5µm. Qian et al [5] showed that for aligned samples the optimum particle size was ~74 µm beyond which the MR effect decreased with increase in particles sizes. All these studies showed there was an optimum particle size but neither of the studies compared the effect of the particles sizes for the unaligned and aligned MRE materials.

Inspired by the experimental work, we aim to investigate the effect of particle sizes on the effective behavior of unaligned and aligned MRE based on microscopically motivated continuum model. Continuum-based models as used by Dorfman and Ogden [8], Kalina et al [9], and Spieler et al [10] are considered as popular tools to understand magneto-elastic responses of these materials. Here, the local magnetic and mechanical fields are resolved explicitly. While for the calculation of the effective material behavior of the MRE an appropriate homogenization scheme is utilized [10]. Since in this modeling strategy each of the constituents is considered separately, influences of the microstructure can be taken into account. Thus, deformation mechanisms leading to material characteristic phenomena such as magnetostriction and the magnetorheological effect can be investigated systematically.

In this work, FEM methodology is utilized due to its capability to take into account different microstructures [9]. In particular, the results are evaluated for isotropic and anisotropic microstructures, respectively. Assuming the sample to have constant particles' volume fraction and shape, the effect of particle sizes on the effective composite behavior is determined for the different microstructures. To perform the simulation, commercial finite element analysis (FEA) software COMSOL Multiphyics is used as it has the capabilities to couple the magnetic and elastic behavior [11].

To allow for the coupling of the magnetic and mechanical fields in the composite the AC/DC and the structural mechanics module in COMSOL Multiphyiscs are used [12]. A two- dimensional representative volume element (RVE) is developed as depicted in Figure 1. Periodic conditions are assigned and the Maxwell stress tensor is introduced to the boundaries of the particles in order to couple the mechanical and magnetic fields and the macroscopic response of the MRE is studied as a function of particles sizes.

The paper is organized as follows: In Section 2, the theory of numerical simulation in finite deformation [8], [13] is briefly discussed. In Section 3, using FEM the RVE models developed for numerical simulations and the microstructure of the sample are discussed. In Section 4, the results of the numerical analysis are presented and discussed. Finally, in Section 5 concluding remarks are given.

Theory

The continuum theory of magnetoelasticity in magnetic elastomer composites has been well developed Danas et al [1], Dorfmann and Ogden [8] and Kankanala and Triantafyllidis [14]. Therefore, in this section no aim is made to redevelop the theory, but to use it to study the magnetic particle sizes deformation dependent behavior of the composite material. Here, we consider the polymer matrix and the magnetic fillers as continuum with distinct materials properties in contrast to the phenomenological approaches [8], [14] where the material model is fitted to the experimental data obtained from a homogenization.

Following the basics in continuum mechanics, the deformation gradient F and its determinant J are defined as

$$\boldsymbol{F} = \partial \boldsymbol{x} / \partial \boldsymbol{X}, \qquad J = \det \boldsymbol{F} > 0 \tag{1}$$

where $\boldsymbol{X}$ and $\boldsymbol{x}$ represent the positions of a typical material point in the reference (undeformed) and current (deformed) configuration, respectively and J is the Jacobian. The Jacobian is a measure of the materials volume change due to deformation. When a magnetic field is applied to MRE, the material deforms, the local deformation can then be characterized based on the deformation gradient and the Jacobian [15], [16]. Based on the reference configuration (Lagrangian description), the strain measure in terms of materials coordinates is defined using the symmetric right Cauchy Green tensor $\boldsymbol{C} = \boldsymbol{F}^T\boldsymbol{F}$. This should be differentiated from that in spatial coordinates (Eulerian description) where the strain measure is defined using the symmetric left Cauchy Green tensor $\boldsymbol{b} = \boldsymbol{F}^T\boldsymbol{F}$. In this work, a FEM approach that uses an Arbitrary Lagrangian-Eulerian (ALE) formulation with finite deformation kinematics to account for large deformation is used. Thus, for the remainder of the work, the discussion of the model will be based on the Lagrangian description.

Magnetostatics

Considering static magnetic field, the Maxwell's equations for stationary magnetic field are described by the relations,

$$\Delta . \boldsymbol{B} = 0, \tag{2}$$

$$\nabla \times \boldsymbol{H} = \boldsymbol{J}. \tag{3}$$

Where $\boldsymbol{B}$ is magnetic flux density, $\boldsymbol{H}$ is magnetic field strength and $\boldsymbol{J}$ is current density. Across the boundary surface the boundary conditions for $\boldsymbol{B}$ and $\boldsymbol{H}$ are given as,

$$\boldsymbol{n}.[\boldsymbol{B}^+ - \boldsymbol{B}^-] = 0, \ \boldsymbol{n} \times [\boldsymbol{H}^+ - \boldsymbol{H}^-] = 0 \tag{4}$$

Where the square brackets indicate a jump of a quantity across the surface and $\boldsymbol{n}$ is the outwards normal to the surface. Magnetization **M**, which is magnetic moments, induced in the magnetic material per unit volume can be related to **B** and **H** by the constitutive relation,

$$\boldsymbol{B} = \mu_0(\boldsymbol{H} + \boldsymbol{M}) \tag{5}$$

Where μ_0 is permeability of free space. In numerical simulation, which is the method used in this work, the modeling of the magnetic behavior involves choosing the magnetic vector potential **A** as the independent variable in the constitutive laws defined above. The independent variable **A** is then related to **B** by the relation,

$$\boldsymbol{B} = \nabla \times \boldsymbol{A} \tag{6}$$

and using the Coulomb gauge transformation which is defined by $\Delta.\boldsymbol{A} = 0$, the Gauss's law for magnetism is automatically satisfied whereas the Ampere's circuital law stated in (3) remains to be solved. In addition, equations 2 and 6, and the coulomb gauge imply the continuity of the magnetic vector potential across a boundary surface of discontinuity, hence [**A**]=0.

Constitutive equations

From the basic balance principles of continuum mechanics such as the linear momentum and the angular momentum balance principle, the equation of mechanical equilibrium is given as,

$$\nabla.\boldsymbol{\tau} + \rho \boldsymbol{f} = \rho \dot{v} \tag{7}$$

Where $\boldsymbol{\tau}$ is stress tensor, ρ is density and $\boldsymbol{v}$ is velocity. In the stationary case, there is no acceleration and hence the force balance equation becomes,

$$\nabla.\boldsymbol{\tau} + \rho \boldsymbol{f} = 0 \tag{8}$$

For the case of coupling magnetic and elastic behavior there are different methods of defining the body forces, stresses, and traction. The deformation on the material due to magnetic field can be incorporated into the force balance equation in terms of magnetic force per unit volume $\mathbf{f}_m$,

$$\nabla.\boldsymbol{\tau} + \rho \boldsymbol{f} + \boldsymbol{f}_m = 0 \tag{9}$$

Or in terms of total stress tensor $\boldsymbol{T}$ which is considered to be the sum of the mechanical and magnetic ($\mathbf{T}_m$) contributions as follows,

$$\mathbf{T} = \boldsymbol{T}_m + \boldsymbol{\tau} \tag{10}$$

Hence, the equilibrium mechanical equation can be written as;

$$\nabla . \mathbf{T} + \rho \boldsymbol{f} = 0 \tag{11}$$

In general, the expressions for the total stress tensor depend on the type of the constitutive model. They stem from a combination of concepts including thermodynamics, continuum mechanics, material and electromagnetic field theory. A summary of the various forms of constitutive equations, traction conditions and body forces for finitely strained magnetic elastic composites can be found in Kankanala and Triantafyllidis [14] .
The boundary conditions for total stress tensor at the external boundary of the body is given as;

$$\mathbf{n} . \mathbf{T} = \mathbf{t} \tag{12}$$

Where **t** is traction vector. For a magnetic (nonzero **M**) elastic solid material, the stress tensor is determined based on Maxwell stress tensor which is written as

$$\boldsymbol{T}m = \frac{1}{u_0}\boldsymbol{B} \cdot \boldsymbol{B}^{\top} - \boldsymbol{M}\boldsymbol{B}^{T} - {}^{1}\!/_{2\mu_0}\,(\boldsymbol{B}.\boldsymbol{B} - \boldsymbol{M}.\boldsymbol{B})\mathrm{I} \tag{13}$$

In this work, the Maxwell stress tensor is applied over the boundary surface of the magnetic particles in order to approximate the magnetomechanical coupling in MREs [12], [17].

Modeling of MRE’s

To simulate the effect of particle sizes on the macroscopic behavior of MREs a two-dimensional RVE is developed in COMSOL Multiphysics as shown in Figure. 1. In composite materials, a RVE is the smallest volume over which the measurements can be made that will yield a value that accurately represents the overall macroscopic constitutive response. Since the RVE is usually smaller than the size of the sample, the boundary conditions are different from those on a macroscopic specimen. Hence, periodic boundary conditions (PBC), are often used to simulate the large system by modeling a small part that is far from its edge. Since in the FEM simulations, PBC are applied to the RVE models it is thus required that the RVE have periodic microstructures. To satisfy the continuity conditions of stress, the interface between every two adjacent RVEs must have the same displacement and stress field. For a given average deformation gradient **F** applied to the RVE model, the PBC can be represented by the following general format [16]

$$\boldsymbol{x}\,(\mathrm{Q1}) - \boldsymbol{x}\,(\mathrm{Q2}) = (\boldsymbol{F} - \mathbf{I})\,[\boldsymbol{X}\,(\mathrm{Q1}) - \boldsymbol{X}\,(\mathrm{Q2})], \tag{14}$$

$$\mathrm{V}\,(\mathrm{Q1}) = -\mathrm{V}\,(\mathrm{Q2}), \tag{15}$$

where Q1 and Q2 represents the general nodes of the opposite adjacent faces of the RVE, **I** is the identity matrix, V is the force applied on the node. Equation (13) represents the periodic displacements, while (14) represents the antiperiodic traction conditions.

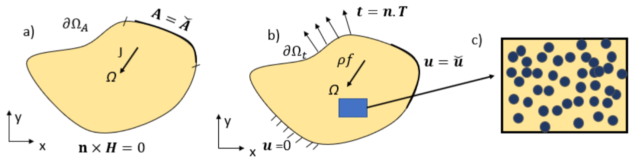

Figure 1. Different boundary conditions defined to the material body a) Electromagnetic boundary value problem (BVP) -prescribed magnetic vector potential **A** and current J in material body domain Ω b) Coupled magneto—mechanical BVP - prescribed displacements **u**= $\breve{\boldsymbol{u}}$ on $\partial\Omega$**,** surface tractions **t** and mechanical body force density Pf in Ω c) Schematic diagram of MRE RVE

To compute the macroscopic magnetic and the coupled magneto-mechanical response of the MREs from results obtained in an FEM, a computational homogenization method is used [10], [18]. Assuming the filler particles to be circular in shape and with volume fraction ~20%, the simulation is performed for varying mean particles sizes (Φ=5, 10, 20 and 30 µm). The particles are also assumed to be tightly bonded to the matrix, so that both displacement and traction are continuous across the interface.

Material properties

Different material properties established from experimental data and literature values are assigned to the matrix and the filler[17]. The matrix which is modeled as a silicon rubber is assumed to follow the Mooney-Rivlin free energy density function *W*, given as.

$$W = C_{10}(\bar{\boldsymbol{I}}_1 + 3) + C_{01}(\bar{\boldsymbol{I}}_2 + 3) + \frac{K}{2}(\boldsymbol{J} - 1)^2 \tag{16}$$

Where C_{10}, C_{01} are material constants, *K* is the bulk modulus and J again is the Jacobian, $\bar{I}_1$ and $\bar{I}_2$ are the modified invariants. Based on literature values [17] the material constants C_{10} and C_{01} are assumed to be 0.4 and 0.1 MPa respectively. The initial shear modulus of the matrix can be calculated from $G = 2(C_{10} + C_{01})$ as 1MPa. The Poisson’s ratio υ is assumed to be 0.47 for small compressibility. Using shear modulus and the Poisson’s ratio the initial Young modulus E and the bulk modulus K can be calculated to be 2.94 MPa and 16.33MPa, respectively. It is also assumed the relative permeability of the matrix is 1. The density of the matrix is assumed to be 1250kg/m^3. The particles which are assumed to be iron (Fe) particles are modeled as linear elastic material with young modulus E, υ, and density assumed to be 210GPa, 0.33 and 7860 kg/m^3 respectively. The magnetic properties of the Fe particles are defined by the B-H curve obtained from the materials library in COMSOL Multiphysics.

The model is considered as a two-dimensional RVE restricted on the left face as shown in Figure 2. The model consists of two sections, the composite RVE and of the surrounding representing the free space/air region. The basic idea behind our modeling is to allow the sample to freely deform as expected of MRE materials in the presence of an external magnetic field. Thus, a fixed constraint (u=0) where u is displacement, is imposed on the left face of the sample (Figure. 2) while other boundaries can freely deform.

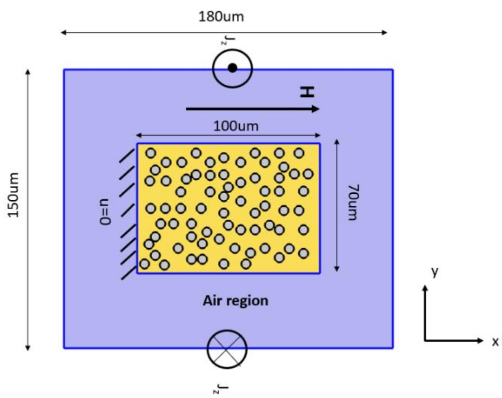

Figure 2. 2-D model description for the numerical simulation of the MRE sample where J_z represents the applied surface current in the z-coordinate (circle with a dot is out and with a x is into the board)

In the surrounding, the magnetic field depends on the vector potential **A** which is determined from prescribing appropriate boundary conditions at the external boundaries (Figure 2). The direction of the resulting magnetic field in the surrounding space is then parallel along the longer axis (x-axis). Due to the large displacements we adopt a moving mesh mode for the calculation of the magnetic field. The mesh movement inside the internal subdomain and at its boundaries is determined by the displacements of the composite due to the applied magnetic fields. The mesh is fixed at the air region external boundaries allowing smoothing of the deformation of the mesh in the surrounding domain toward these boundaries.

Surface current (J_z) is applied as depicted in Figure. 2, giving rise to a magnetic field aligned with +x-direction. A parametric sweep of the surface current is carried out to determine the influence of the magnetic flux density on the effective deformational behavior of the MRE. According to reported experimental data on MREs large B values are needed to effect any significant changes on the MREs, thus large surface current is provided in the range of 0 to 8e5A/m in step sizes of 5e3A/m for the initial range 0-1e5A/m and step sizes of 10e3A/m for the 2e5-8e5A.m leading to large B values ~0.8T.

Results and discussions

Due to the material and geometric nonlinearities, associated with these types of materials, convergence is usually very challenging in the numerical simulations and a simulation with very refined mesh (extra fine, extremely fine) could take a while to get completed. Hence, an investigation of the accuracy of the physics defined coarse mesh (total number of elements being 16856, 1298 in the surrounding and 15558 elements in the MRE) to the refined mesh (normal and finer) with total number of elements being 30912 and 34266 respectively to predict the response of the RVE models is first performed. Triangular elements are used for the meshing of both the surrounding and the MRE. The simulation is performed for unaligned MRE with mean particle size of 5 um and using computational homogenization the effective magneto-induced strain ($\bar{\varepsilon}_{11}$) in the direction of the applied magnetic field is plotted for the different mesh sizes as shown in Figure. 3. From the analysis, the normal mesh size is observed to give larger ($\bar{\varepsilon}_{11}$) values as the effective magnetic flux density ($\bar{B}$) increases especially to larger values compared to the coarse and finer mesh sizes which are observed to overlap. This shows that using very refined (finer) mesh sizes reduces the accuracy of the response of these materials. Since only a small change is observed in the compared mesh sizes, the coarse mesh size is selected for the rest of the modeling to minimize the simulation time especially for the sample with smaller sized particles.

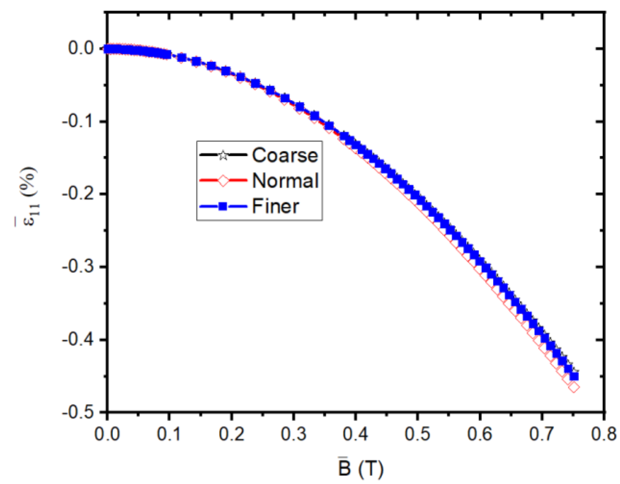

Figure 3. Effective magnetostrictive in the direction of the applied magnetics for varying physics defined mesh sizes.

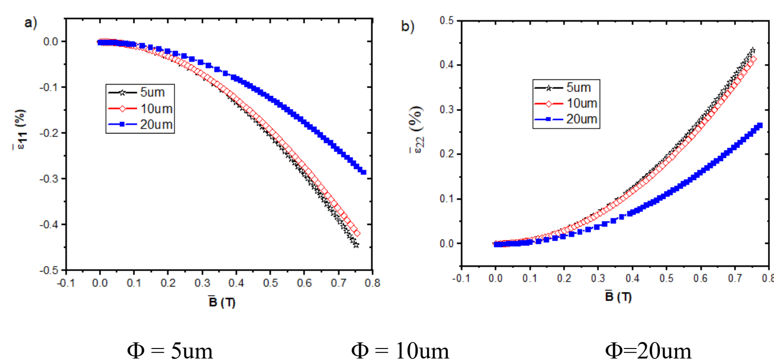

Figure 4. Effective magneto – induced strain in unaligned (a) parallel to the direction of the applied field (b) transverse to the direction of the applied magnetic field. The deformation of the RVE for different particle sizes is as shown in the legend. (The scale factor is 10).

The effect of particle sizes in the parallel ($\bar{\varepsilon}_{11}$) and transverse ($\bar{\varepsilon}_{22}$) directions of the applied magnetic fields for the unaligned microstructures was determined as shown in Figures 4 (a) and (b). The dominant mechanism of performance for MREs, depends on interaction between neighboring particles [19] hence as seen from other scholarly works [20], the interparticle distance is a significant parameter to consider when evaluating the macroscopic behavior of these materials. Noteworthy, the isotropic MREs are not dependent on interparticle distance, hence no attention is paid to the distance between the particles in these structures. However, for the anisotropic microstructures we deliberately configured these structures so that the interparticle distance parameter is taken into consideration. To compare the results, the vertical/horizontal distance parameters *a* and *r* (Figure. 5), respectively are kept constant for all the particle sizes. Figures 5 (a) and (b) shows the negative/positive effective magneto-induced strain for aligned microstructure parallel / transverse to the direction of the magnetic field with varying particles sizes and the distances *a* and *r* set to 10 and 4 um respectively.

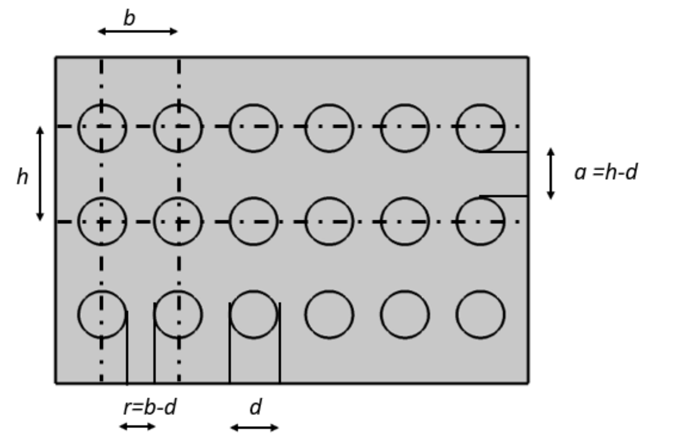

Figure 5. Schematic diagram of the aligned MRE. The geometric parameters include, particle diameter *d*, the vertical distance *h* and the horizontal distance *b* between two neighboring particles.

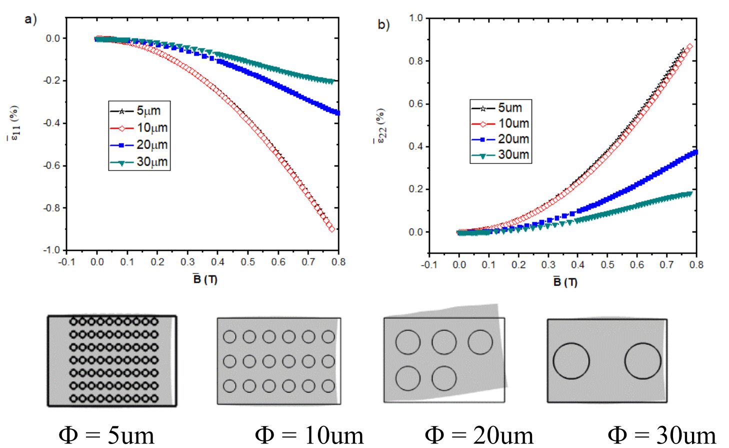

Figure 6. Effective magneto – induced strain in aligned MRE (a) parallel to the direction of the applied field (b) transverse to the direction of the applied magnetic field. The deformation of the RVE for different particle sizes is as shown in the legend. (The scale factor is 10).

For both unaligned and aligned microstructures, the effective magneto-induced strain in the parallel ($\bar{\varepsilon}_{11}$) and transverse ($\bar{\varepsilon}_{11}$) direction is observed to increase with increase in $\bar{B}$. Furthermore, the particle sizes are seen to have a significant effect in the macroscopic response of the composite. More specifically, the $\bar{\varepsilon}$ is observed to decrease with increase in particles sizes, as the particle surface area decreases, and the composite becomes less permeable with larger particle distance hence the particles are less magnetized. This agrees with experimental observations as seen in the scanning electron microscopy (SEM) micrographs of MRE samples with different particle sizes [5], [21]. The resulting larger area of the elastomer reduces the overall reinforcing effect of the mechanically stiffer particles and thus decreased deformational effect in the MRE. The effect of linearly increasing the particle sizes on the distance of separation between the neighboring particles is determined as plotted in Figure 7. The results show a linear relationship between the particles sizes and the interparticle distance of

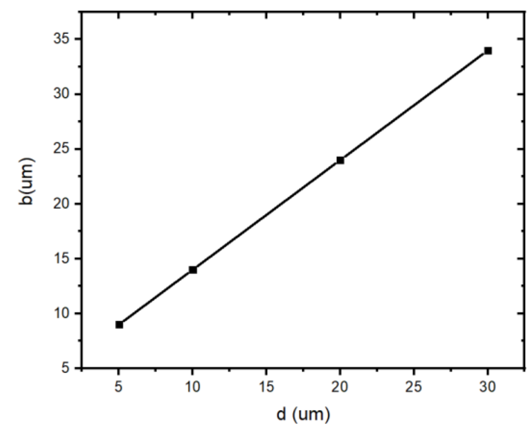

Figure 7: Variation of interparticle distance with the mean particle size.

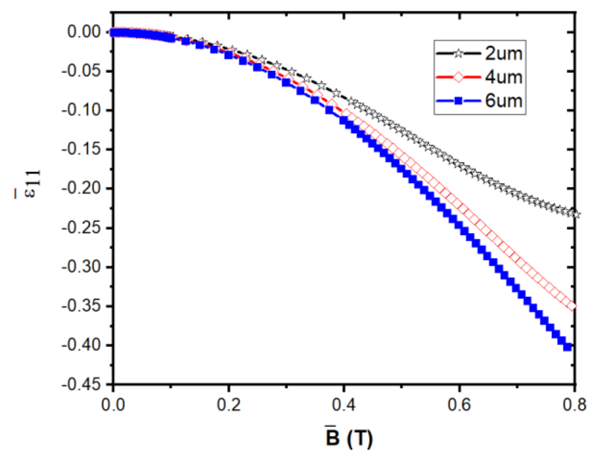

Figure 8. Effective magneto-induced strain for varying distances between neighboring particles for 20um sized particles.

between the particles. That is, a linear increase in the average particle size leads to a linear increase in the distance between neighboring particles. This agrees with the theoretical prediction of the effect of particles sizes on interparticle distance of such composites[5], [21], [22].
Further, keeping the parameter *a* constant and changing *r* (2, 4 and 6um), a study is carried out for the 20um sized particle to determine the effect of the distance between particles on the $\bar{\varepsilon}_{11}$ as shown in Figure 8. In general, macroscopic magneto-induced strain $\bar{\varepsilon}_{11}$ is observed to increase with increase in the distance *r* between particles. From the analysis the distance between neighboring particles is shown to have significant influence on the macroscopic response of MREs.

Conclusion

From the simulation study, we have shown that the size of the particles strongly influences the macroscopic magneto-induced strain in the MRE for both the unaligned and aligned particles. The effective magneto-induced strain effect is observed to decrease with increase in the average particle sizes. Also, increasing the particle sizes is observed to lead to a proportional increase in the distance of separation between the particles. That is, the larger the particle is, the longer the distance of separation between the neighboring particles is in the direction of the magnetic field. This agrees with the scanning electron microscopy images of MREs as showed by Qian et al[5] and theoretical calculation. Furthermore, the distance between the particles is observed to have a significant effect in the macroscopic mechanical behavior of MRE.

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