IOP Conference Series: Materials Science and Engineering

PAPER • **OPEN ACCESS**

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To cite this article: P Melenev and A Ryzhkov 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **581** 012039

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IOP Conf. Series: Materials Science and Engineering **581** (2019) 012039 doi:10.1088/1757-899X/581/1/012039

Simulation of Uniaxial Deformation of a Ferrogel Sample Exposed by the External Magnetic Field

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Abstract. Ferrogels are magnetoactive composite materials with very soft hydrogel matrix, filled by ferromagnetic particles. This work is devoted to a numerical investigation of magneto-mechanical behaviour of a small sample of ferrogel. Our model is based on a coarse grained molecular dynamics approach: polymeric matrix is emulated by lattice of bead-strings chains and magnetic single-domain particles are considered as rigid spheres and placed in a nodes of the lattice. This model was previously successfully used for a research of equilibrium magnetization of the material. Here we examine of the reaction of ferrogel sample on uniaxial tension in presence of an external magnetic field. Modelling proves that key role in a changes of mechanical response during the magnetization plays an internal structure of material, specifically, the existence of particles aggregates, induced by forces of dipolar magnetic interaction. Also the influence of magnetic anisotropy on a process is considered.

1. Introduction

Soft magnetic composites (SMC) are interested for researches due to pronounced interrelation between mechanical and magnetic characteristics of SMC, caused by the ability of sufficient magnetically induced rearrangements of magnetic filler inside the soft polymeric matrix. Ferrogels may be considered as an “extreme case” of SMC with respect to matrix softness. These materials consist of hydrogels with very low elastic modules (characteristic value is less than 10 kPa) filled by ferromagnetic particles which usually have sizes within tens of nanometers and so have single-domain magnetic structure. This combination allows ferrogels to demonstrate noticeable response to magnetic field even of relatively low strengths (less then 0.1 T) which is substantial from the perspective of utilization of ferrogels in a bio-medical applications. Beyond the mechanical properties magnetic action could change the shape of the ferrogel samples [1-3].

The complex composition of SMC accounts for their uncial magneto-mechanical behavior, but simultaneously it is “headache” for the researches who try to investigate theoretically (and numerically) of these materials. Indeed, a mechanical response of composites is determined to a great extent by the local structure of their filler. Which in case of SMC may be altered by magnetic forces, induced by the external field and/or particle dipolar interaction. In turn the distribution of the magnetic field in the sample may also depends on filler allocation. This strong linkage between macrocharacteristics of the material and its internal structure complicates application of conventional continuous methods for the SMC theoretical study.

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One of the possible way of direct examination of the micro-structure of magnetic composite is application of particle-based methods, where both polymeric matrix and dispersed filler are modelled by discrete objects, namely, particles and bonds. This simplifies sufficiently the description of composite components and because of corresponding decrease of computation cost allows to consider in simulations systems which contain tens or even hundreds of thousands of magnetic particles [4, 5]. This approach is especially prospective for examination of ferrogel since real objects made of these materials developed for bio-medical applications may have submicron size. So it is possible to model the whole material sample [6]. On the other hand, there are examples of multiscales models [7-10] where data, obtained on the base, structural level of magnetic composite, are used for an evaluation of macroscopic material characteristics.

A nice review paper [11] contains description of various approaches of theoretical treatment of ferrogels, including several particle-based models. They could be divided into two types regarding way of account of polymer matrix. In the first one, matrix is represented exclusively by bonds between the filler particles, thus polymeric excluded volume is not considered (e.g. in [12]). This approach allows to perform a calculations of systems with larger amount of particles, but simultaneously complicates the procedure of identification of bonds parameters based on experimental and theoretical data regarding microstate of real polymers. Also it appears that in this case examination of composite with low or moderate filler concentration would not efficiently due to oversimplified model of the matrix. The second type of models presents polymer network as a set of chains of weakly deformable or rigid particles, which could be associated with polymer segments or blobs. This approach allows to model various types of matrix structure and variants of inclusion of filler particles into the polymer. The model, employed in our work here, belongs to this method with bead-strings description of polymer network.

We consider the behavior of small sample of ferrogel contains up to 1000 single-domain magnetic particles, arranged in a nodes of polymer network. Our simulation is based on coarse grained molecular dynamics approach. Earlier this model was used in simulations of equilibrium state of the samples with various values of filler concentration and particle characteristics in presence of permanent magnetic field. Particularly we considered dependence on the field and material properties of the sample volume and filler structure [13-14]. Development and studies of real ferrogels which may be utilized, among other, as micromachines (or their details) require ability to model a material response to mechanical loading. Also, the following calculations may be necessary for the identification of model parameters based on experimental data. This work is devoted to numerical examination of mechanical response of small ferrogel sample to uniaxial tension and examination of dependence of material reaction on the external magnetic field.

2. Simulation

2.1. Ferrogel sample model

Here we briefly describe the model of ferrogel, used in calculations. More details may be found in [13]. Filler particles are presented as rigid spheres of diameter d_p, each of which has magnetic moment of constant magnitude m_p. Dipolar magnetic interaction operates between the particles. Also each particles possesses uniaxial magnetic anisotropy with finite value of energy barrier E_A.

Polymer macromolecules is modelled by the chains of identical rigid spheres – beads (with diameter $d_\mathrm{m} < d_\mathrm{p}$), linked by practically non-deformable bonds. Chains may bend freely – flexural stiffness is absent. Chains is formed to quasi-cubic lattice some of nodes of which are occupied by magnetic particles. The portion of occupied nodes depends on filler concentration.

The system may be exposed by the external permanent and uniform magnetic field with strength H_0. The presence of constant temperature T, essential for the simulation of processes on submicron scales, is accounted by introduction to the model of Langevin thermostat. Thus the mechanical reaction of matrix is determined by the mechanism of entropic elasticity. Then, characteristic energy of interaction of dipoles with each other and with the external field as well as the energy of particle magnetic anisotropy are estimated relatively to thermal energy and specified in terms of corresponding dimensionless parameters:

$$\lambda = \frac{\mu_0}{4\pi}\frac{m_\mathrm{p}^{\,2}}{d_\mathrm{p}^{\,3}k_\mathrm{B}T}; \quad \xi = \mu_0\frac{m_\mathrm{p}H_0}{k_\mathrm{B}T}; \quad \sigma = \frac{E_\mathrm{A}}{k_\mathrm{B}T}, \tag{1}$$

here μ_0 denotes the vacuum permittivity.

The calculations are performed using ESPResSo software package [15] with our modifications, charged with magnetic anisotropy simulations.

2.2. Simulation of deformation process

At the start of each numerical experiment the thermostating of the sample is performed. Earlier it was found that in samples with high filler concentration and strength of dipolar interactions in absence of the external magnetic field elongated and chaotically oriented chain-like aggregates of particles are formed (see figure 2 in [13]). External field attempts to turn these chains along its direction, but degree of such an orientation depends on the field magnitude and material properties, particularly magnetic anisotropy energy. In the samples where dipolar forces are not able to form particles aggregates without external field, magnetization practically does not lead to filler clustering – usually, only small number of new particle pairs appears.

When system has reached equilibrium an uniaxial tension of the sample starts. Because of irregular relief of the sample's sides for performing of hard loading we use the following trick. On two opposite sides the layers are chosen, each with thickness of several d_p . Model elements – magnetic particles and polymer beads – inside these layers are fixed, which means their position do not change according to equation of motion integration. During the calculation one of the layer – "base" – remains immobile. And all of elements of the second one were moved in direction of tension to new positions which ensured the deformation ε of the sample in limit of 5%. Then the system has time to reach the new equilibrium state. And this operation repeats until achievement $\varepsilon = 50\%$ or fracture of the sample. The reaction of the sample is described by the net force f_b acts on the magnetic particles in "base" level. The presence of thermofluctuations of model elements causes to fluctuate the instantaneous value of this force. Therefore the time averaging of f_b is performed during given number (in practice – several tens) of "relaxation" numerical steps. Obtained value of force $\langle f_\mathrm{b} \rangle_t$ is used in calculation of mechanical module of material:

$$G = S_0^{-1} \frac{\Delta \langle f_\mathrm{b} \rangle_t}{\Delta \varepsilon}, \tag{2}$$

where S_0 is initial area of sample cross-section.

It is significant to note that used in present calculations values of model parameters do not correspond to some concrete examples of real ferrogels. And in this work we are focused on obtaining of qualitative results only. Particularly, we were not interested to find absolute values of mechanical module but try to examine it changes, caused by different factors: deformation, external magnetic field, magnetic anisotropy and strength of dipolar interaction of the particles.

3. Results

Simulation proves the key role which plays the internal filler structure in mechanical behaviour of the ferrogel. Thus, in absence of the external field the samples where dipolar forces able to form particle aggregates have higher value of G in comparison with analogous (in respect of particle concentration and anisotropy) samples with weaker dipolar interaction. Samples of the latter type, characterised by $\lambda < 3$, in absence of the field demonstrate practically linear elastic response for whole range of tension. For the materials with stronger magnetic forces the growth of mechanical module during the deformation also is not dramatic and does not exceed 10%, see dashed lines on figure 1. Magnetisation leads to noticeable increase of G in case of deformation along the external field. Also mechanical response of the samples with high values of λ becomes sufficiently nonlinear. Meanwhile in materials with weaker magnetic interactions much lower increase of mechanical module is observed, see solid lines on figure 1. The found difference of field dependence of G may be described by the origin of anisotropy of filler structure in the samples, where the external filed orients already existed particle aggregates. This assumption is supported by the simulation of deformation perpendicular to the direction of magnetisation, where only a small growth of value of G was observed – for all considered samples it was several times lower, than in case of deformation along the field, see figures 2-3. It is interesting that we obtained results similar to the experimental data for deformations of samples of soft magnetic elastomers with polydimethylsiloxane matrix, filled by microparticles of carbonyl iron [16], which

emphasizes the similar sense of dipolar interaction in forming of mechanical response of different types of SMC.

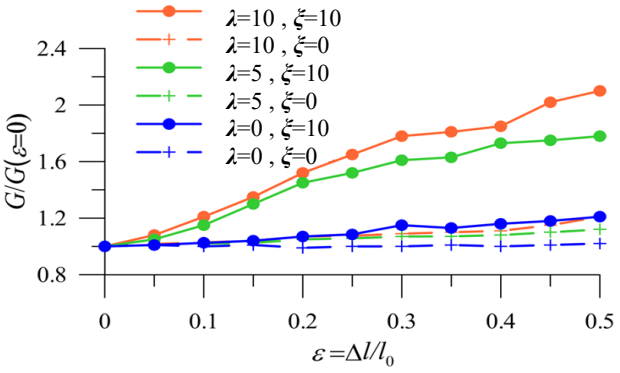

Figure 1. The dependence of relative value of mechanical module on strain. Presented results obtained for particles with uniaxial magnetic anisotropy, $\sigma = 2$.

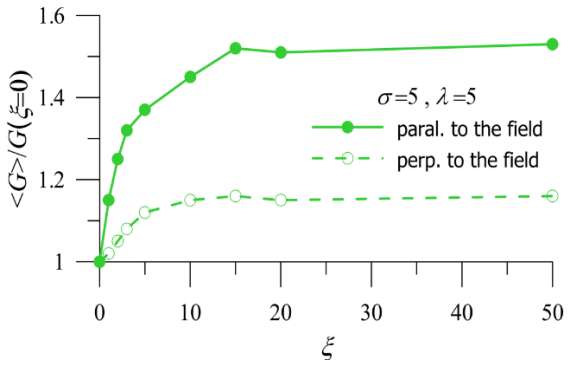

Figure 2. The dependence of the relative value of mechanical module on the external field. Presented results obtained for the system with $\sigma = 5, \lambda = 5$. Dashed line corresponds deformations perpendicular to field direction and solid line – parallel case.

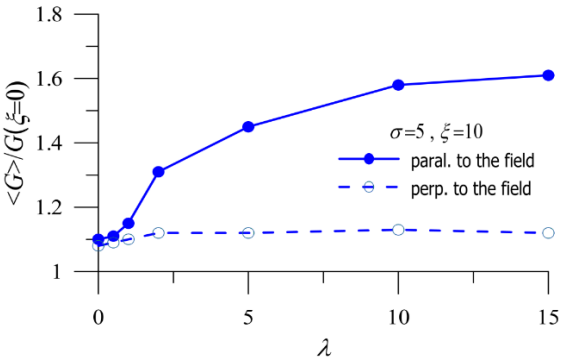

Figure 3. The dependence of the relative value of mechanical module on dipolar interaction strength. Presented results obtained for the system with $\sigma = 5, \xi = 10$. Dashed line corresponds deformations perpendicular to field direction and solid line – parallel case.

The distinguish feature of our ferrogel model is capability to consider particles with finite energy of magnetic anisotropy. The simulation of the samples with various E_A values shows that in materials with intense dipolar interactions and noticeable magnetic anisotropy (i.e. $\sigma \geq 1$) the increase of E_A entails some growth of mechanical module, which is evidently connected with the presence of local stresses occurred in matrix due to rotations of particles induced by reorientations of their magnetic moments. The magnetization, as it was already described, leads to increase of mechanical module (see figure 2). But for samples with intense dipolar interactions relative value of field-induced variation of G decreases within growth of σ, see figure 4. We suppose that this happens because above-mentioned initial hardening of the material hampers the rearrangements of the filler in the external field. So lesser changes of internal structure leads to lower growth of the mechanical module during the magnetization.

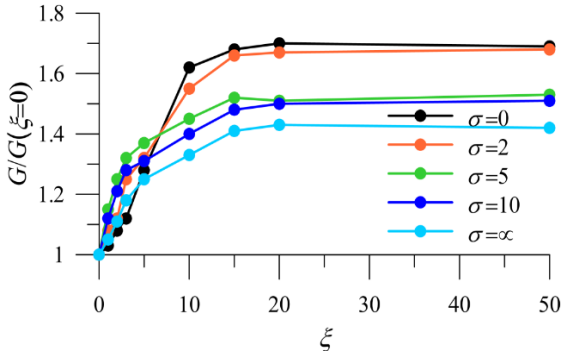

Figure 4. The dependence of relative value of mechanical module on the external for the systems with different values of anisotropy energy. Presented results obtained for samples with $\lambda = 10$ at strain $\varepsilon = 25\%$.

4. Conclusions

The results of numerical investigations on the process of uniaxial tension of small ferrogel sample in presence of permanent magnetic field, performed with employ of coarse grained molecular dynamics approach, confirm the key role of internal structure in forming of material response. In cases, when dipolar magnetic forces form chain-like aggregates of particles even in absence of the external field, the magnetisation leads to orientation of these clusters along the field. Occurred structure anisotropy provides the much larger growth of mechanical module than one, observed for the samples with uniform distribution of particles. Also it was found that the samples with higher energy of magnetic anisotropy are stiffer and, at the same time, demonstrate lower field-induced variation of mechanical module.

The performed calculations prove the applicability of chosen model of ferrogel for simulation of deformation processes. Now we can point out the following primary prospects of prolongation of this work. Firstly, it is necessary to clarify the influence of temperature on the mechanical response, because in our model thermofluctions alongside the polymer network structure determine the elastic properties of the matrix. Secondly, it would be interested to consider material compression, during which in the sample the regions of high particle concentration could occur. It may favour the formation of new particle aggregates and thus sufficient changes of mechanical properties of the sample.

Acknowledgments

The work was supported by Russian Foundation for Basic Research, project 17-41-590123. The calculations were performed using computer clusters of the Institute for Computational Physics of University of Stuttgart (Germany) and the Institute of Continuous Media Mechanics of Perm Federal Research Centre (Russia).

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