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Towards a theory of mechanical properties of ferrogels. Effect of chain-like aggregates

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HIGHLIGHTS

- The strain–stress dependence in ferrogels, in ferrogels with chains is nonlinear.
- In the presence of magnetic field the chains induce finite stress at zero strain.
- The magnetically induced stress non monotonic, with maximum, depends on the strain.

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ABSTRACT

The paper deals with theoretical study of deformation of a ferrogel filled by magnetizable particles united in the chain-like aggregates. The sample is placed in a magnetic field parallel to the chains. Uniaxial elongation of the sample along the field is studied. Analysis shows that interaction of the particles in the chains and rupture of the chains by the elastic forces lead to non linear dependence of the macroscopical stress on the elongation strain. The theoretical results are in principle agreement with known experimental study of the ferrogels deformation.

1. Introduction

Magnetic polymers, ferrogels and ferroelastomers, consist of the nano- or micron sized magnetic particles imbedded in a polymer matrix. Combination of rich set of physical properties of the polymer materials with the high reaction on magnetic field makes these smart systems very attractive for many industrial and bio-medical applications [1].

One of the interesting, from the scientific point of view, and valuable, from the viewpoint of practical applications, features of ferrogels is their ability to change mechanical properties and behavior under the action of external magnetic field. Magnetic field induces either elongation or contraction of the composites in the field direction (see discussion of this problem in Ref. [1]) and significantly changes mechanical properties of these systems. These magnetomechanic effects are especially strong in the systems with internal heterogeneous structures (chains, columns, etc.) formed by the particles [2–4]. Usually these aggregates are created by using magnetic field, at the stage of the polymer matrix curing, when it is liquid. Polymerization of the matrix fixes the internal structures in the composites (see, for example, Refs. [2,4–8]).

For example, experiments [2] demonstrate significant increase, under the field action, of the elastic stress σ at a given uniaxial strain ε in magnetic elastomers with the chain-like aggregates. The dependence of σ on ε, measured in Ref. [2], was nonlinear, with especially fast increase of σ at small ε. Effect of the field H on the stress in the same systems with

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homogeneous and isotropic spatial distribution of the particles was rather negligible; in the isotropic composites [2] the stress linearly depended on the strain. It is interesting to note that non monotonic, N-like shape of dependence of the difference $\delta\sigma = \sigma(H) - \sigma(0)$ on the tensile strain ε in the composites with the chains was detected in Ref. [2].

The nonlinear relation between σ and ε in soft ferrogels, placed in magnetic field, has been observed in experiments [3]. This non-linearity was induced by formation, under the field action, of heterogeneous chain-like structures in the gel. Without magnetic field the composites demonstrated the linear dependence of σ on ε. Under the action of quite moderate magnetic field (1–3 kOe) the elastic stress increased, at least, several times as compared with the case of zero field.

The non-linear relation between the shear stress and shear strain in magnetic elastomers with the anisotropic structures has been observed in experiments [4].

Therefore one can conclude that appearance of the anisotropic heterogeneous aggregates in magnetic polymers significantly enhances mechanical properties of these composites and produces the nonlinear dependence of the mechanical stress σ on the deformation ε.

The aim of this work is theoretical study of the stress–strain dependence in ferrogels with the chain-like aggregates. To be specific, we focus on the modeling of the uniaxial deformation of the sample in the chains direction. We suppose that the chains have been created, as in Refs. [2,4–8], by the application of external magnetic field at the stage of the gel curing. That is why the chains have the same direction, parallel to the field of polymerization. As in experiments [2,3], the actual field $\mathbf{H}$ is imposed in the direction of the chains.

2. Mathematical model and results

The rheological properties of ferrogels depend on the properties of carrier polymer matrix; on concentration of the imbedded particles; on the law of their magnetization and magnetic interaction; on the length of the chains and details of the particles dispositions in these aggregates. For the maximal simplification of the physical analysis and transparency of the mathematical part of the problem, we will use the following approximations.

First, we will estimate magnetic force of the particle interaction in the dipole–dipole approximation. It is supposed that magnetic moment of the particle is not changed by the chain deformation.

Note that the similar approximation of the dipole–dipole interaction between the particles in the chains has been used successfully in the theory of rheological properties of suspensions [9,10] and polymer composites [11–13] of magnetizable particles. Possible ways of generalization of this approximation are discussed in the Conclusion.

Secondly, we will use the linear stress–strain relation for the polymer matrix. Physically this means that only small deformations of the matrix are considered. It should be noted the mechanical non-linear effects in the composites with the chains have been detected in the region of ε corresponding to the linear law of deformation of the pure matrix [2,3]. We will neglect fluctuations of the chains shape and suppose that centers of all particles in the chain are situated on a straight line parallel to the applied field $\mathbf{H}$; all particles in the chain are in physical contacts with their nearest neighbors. This means that the gel was cured under the action of very strong magnetic field. We will suppose also that all chains consist of an identical number of particles; this number is determined by the features of the matrix curing. The length of the chain, like in experiments [2–4], is less than the size of the composite, i.e. the chains do not percolate the sample. The infinite chains, percolating the sample, are considered in Ref. [14].

Diameter of the particle is supposed much more than the characteristic size of the gel cell, therefore, with respect to the particles, the gel can be considered as a continuous media. Taking into account that the typical size of the cells in synthetic gels is about several nanometers, for the micron-sized magnetizable particles this condition is well justified. Next, we will neglect any interactions between the chains. This approximation is based on the results of Ref. [15] which show that, in formation of macroscopic properties of the composites, effects inside the chains play dominate role as compared with effects of the interchain interaction. The considered model of the chain is illustrated in Fig. 1.

We will study deformation of a single chain under the assumption that the composite experiences uniaxial deformation in the chain direction. For definiteness we will suppose that the chain consists of an odd number of the particle. Obviously, this assumption is not of principle for the physical analysis.

We will denote the mean vector of a material point displacement in the composite as $\boldsymbol{u}$.

In the coordinate system, shown in Fig. 1, the vector $\boldsymbol{u}$ of the mean composite displacement (i.e. displacement at the infinitive distance from the chain), can be presented as: $u_z = \varepsilon z$. The other components of $\boldsymbol{u}$ do not play a role in the framework of this model that is why we omit them here.

Let the total number of the particles in the chain be $2N + 1$; n be number of a particle in the chain, starting from the central one, whose number is 0. In the framework of the used approximations, equations of stationary displacement of the particles in the chain can be presented in the following form:

$$\beta(\varepsilon z_n - u_n) + \kappa\left[\frac{1}{(d + u_{n+1} - u_n)^4} - \frac{1}{(d + u_n - u_{n-1})^4}\right] + f_{n+1,n} - f_{n,n-1} = 0, \quad -N < n < N \tag{1}$$

$$\beta(\varepsilon z_N - u_N) - \kappa\left[\frac{1}{(d + u_N - u_{N-1})^4}\right] - f_{N,N-1} = 0,$$