Journal of Magnetism and Magnetic Materials 377 (2015) 373–377

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Effect of particle concentration on ferrogel magnetodeformation

A.Yu. Zubarev [a,*], D.Yu. Borin [b]

[a] Urals Federal University, Lenin Ave. 51, 620000 Ekaterinburg, Russia
[b] Technische Universität Dresden, Institute of Fluid Mechanics, 01062 Dresden, Germany

ARTICLE INFO

Article history:
Received 12 June 2014
Received in revised form
28 October 2014
Accepted 28 October 2014
Available online 31 October 2014

Keywords:
Ferrogels
Magnetodeformation

ABSTRACT

The aim of this work is a theoretical study of deformation of ferrogels under the action of a uniform magnetic field. Our analysis shows that an increase of the particle concentration in the composite can qualitatively change the type of this deformation. When the particle concentration is small and they are randomly distributed in the matrix, the sample, depending on its shape, can either contract or elongate in the field direction. Composites with a high concentration of the particles can only elongate due to effect of the short ranged order of the particles spatial disposition.

1. Introduction

Magnetopolymer composites (ferrogels and ferroelastomers) present a new type of soft smart materials for modern high technologies [1–5] due to a combination of valuable properties of polymer and magnetic systems. In particular, their ability to produce a mechanical stress and to be deformed by uniform magnetic field is very attractive for many mechanical applications [1].

One of the most interesting problems of the physics of ferrogels is the qualitative type of the composite deformation under the action of the external magnetic field – either it elongates or contract in the field direction. Both types of the magnetodeformation have been observed in various experiments (see, for example, Refs. [6–13].).

Phenomenological analysis [14] shows that the type of this deformation is determined by the competition between two following factors. The first one is the change of the sample demagnetizing shape-factor as a result of the deformation. This effect induces elongation of the composite along the field. The second factor is the change of the sample magnetic susceptibility. This factor can stimulate either contraction or elongation of the sample.

Microscopical analysis [15,16], based on a statistical physics approach demonstrates that for dilute composites with the gas-like distribution of the particles effect of change of the susceptibility stimulates contraction of the sample and dominates when the sample is either prolate or strongly oblate. Thus, the samples of these shapes must contract under the field action. The samples with the moderate shape must be stretched. This feature of the sample deformation has been called "the Procrustes effect" [15]. It should be noted that contraction of the system of magnetizable particles randomly distributed in an elastic matrix has been detected in computer simulations [17,18].

Theoretically the composites with the linear chain-like aggregates have been studied in Refs. [19,20]. The results show that compression of gaps between the nearest particles in the chains leads to contraction of the sample. This conclusion is supported by the experiments [6,7] and computer simulations [17]. On the other hand, if the particles in the chains are situated very closely and the chains are non-deformable, magnetic interaction between the chains stimulates the sample elongation [20,21].

Theoretically magnetodeformation of ferrogels with homogeneous distribution of the particles also have been studied in [11]. The change of the sample susceptibility as a consequence of the deformation has not been taken into account in this model. That is why the results of [11] predict elongation of the composite independently of its shape. Experiments [6,11] with the highly concentrated composites (about 30% of the particles volume fraction) demonstrate their elongation for all studied shapes of the samples.

Disagreement of these experimental results with the conclusions of the theoretical models [15,16], developed for composites with small concentration of the particles, allows us to suppose that increase of the particle concentration can change qualitatively the type of the magnetodeformation. The main microscopical cause of the concentration effects can be in appearance, with the concentration growth, of the short range order in the particles spatial disposition. Indeed, the computer simulations [18] demonstrate contraction of the samples with the absolutely random distribution of the particles. However, if appearance of some clusters of densely situated particles is allowed, the sample must rather elongate. Note, that these cluster in concentrated systems can take

* Corresponding author. Fax: +7 343 3507401.
E-mail address: andrey.zubarev@usu.ru (A.Yu. Zubarev).

http://dx.doi.org/10.1016/j.jmmm.2014.10.141
0304-8853/

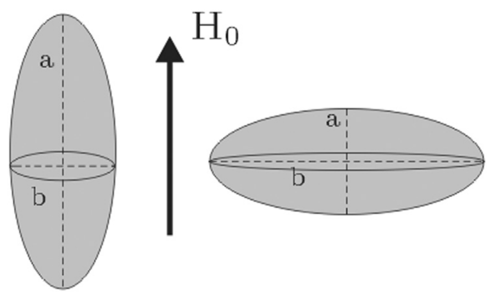

Fig. 1. Sketch of the initially prolate (left) and oblate (right) sample.

place due to usual entropic effects which lead to appearance of maximums of the pair correlation function of the particles spatial distribution [21].

The aim of this work is a theoretical analysis of the concentration effects on the macroscopical magnetodeformation of the ferrogels. We suppose that the composite was cured without magnetic field, therefore, distribution of the particles in the matrix is isotropic and chaotic, without chains and other heterogeneous aggregates. The particles are considered as identical paramagnetic linearly magnetizable spheres.

2. Macroscopical consideration

For maximal simplification of mathematical part of the work, let us suppose that the sample presents an ellipsoid of revolution with the axes equal to $a \neq b = c$. The ellipsoidal shape provides the homogeneity of the internal magnetic field H inside the sample and allows us to use the well-known explicit expressions [15] for the sample demagnetizing factor. That is very important for analytical study of the magnetomechanical effects.

We suppose also that the external magnetic field H_0 is aligned along the main axis a of the sample. This model situation is illustrated in Fig. 1. The polymer matrix is assumed to be incompressible. In many real situations this condition is not ensured. However any small deformations can be presented as combinations of the sample's shape change at the constant volume and change of its volume at the constant shape. A linear approximation allows us to consider these deformations separately. In our model the deformations at the constant volume of the sample are evaluated.

The lengths of the axes a and b for the nondeformed sample are denoted as a_0 and b_0, respectively. We will consider only small deformations of the sample and suppose that the axes of the deformed ellipsoid are

$$a = a_0(1+\varepsilon), \quad b = b_0\left(1 - \frac{\varepsilon}{2}\right), \quad \varepsilon \ll 1. \tag{1}$$

When the size of the specimen is not fixed the positive sign of ε corresponds to the elongation of the sample in the field direction and the negative sign corresponds to the contraction of the sample.

The free energy F of the deformed sample is expressed as a sum of the elastic component F_{el} and the magnetic component F_m. The elastic component F_{el} corresponds to the macroscopic deformation of the matrix whereas the magnetic component F_m corresponds to the magnetic interaction of the particles with an applied magnetic field and with each other. The elastic component F_{el} is positive and proportional to ε^2 for small deformations. The change of the magnetic free energy F_m is proportional to ε as shown below. Thus, when $\varepsilon \ll 1$ the magnetic component F_m is dominating and the sign of F_m determines the type of the sample deformation.

Keeping in mind a determination of the qualitative type of the deformation (not its amplitude), we will consider below only the change δF_m of the magnetic free energy F_m.

For maximal simplification of calculations we suppose that the linear law of the particle magnetization is fulfilled.

The free energy of a magnetizable ellipsoid in an external field H_0 is [14]:

$$F_m = -\frac{\mu_0}{2}\frac{\chi_e}{1+\chi_e N}H_0^2 \tag{2}$$

Here μ_0 is the vacuum magnetic permeability, χ_e is the component of the tensor of the sample effective magnetic susceptibility and N is the demagnetizing factor in the direction of the field H_0 (i.e. in the direction of the axis a). For simplicity we suppose that the sample volume equals to unity.

A deformation of the sample changes both N and χ_e. [14] In the linear approximation with respect to ε the change of the magnetic free energy F_m is calculated as

$$\delta F_m = -\frac{\mu_0}{2}\frac{H_0^2}{\left(1+\chi_e N\right)^2}\left[\delta\chi_e - \chi_e^2\,\delta N\right] \tag{3}$$

Here $\delta\chi_e$ and δN are changes of χ_e and N in the field direction due to the sample deformation. The condition of the thermodynamic equilibrium requires that the free energy should be a minimum, that corresponds to decrease of F_m, i.e. $\delta F_m < 0$.

Let us denote $R = a/b$. The case $R > 1$ corresponds to the prolate ellipsoid, the case $R < 1$ to the oblate one. Taking into account Eq. (1), in the linear approximation in ε the change of the demagnetizing factor is

$$\delta N = \frac{\partial N}{\partial R}\frac{\partial r}{\partial \varepsilon}\varepsilon = \frac{3}{2}\frac{\partial N}{\partial R}R_0\varepsilon \tag{4}$$

where $R_0 = a_0/b_0$.

By using the well known formulas for the demagnetizing factor N (see, for example [14]), one can present:

$$N = \begin{cases} \dfrac{R^{-2}}{2(1-R^{-2})^{3/2}}\left[\ln\dfrac{1+\sqrt{1-R^{-2}}}{1-\sqrt{1-R^{-2}}} - 2\sqrt{1-R^{-2}}\right], & R<1 \\[2ex] \dfrac{1+\sqrt{R^{-2}-1}}{(R^{-2}-1)^{3/2}}\left[\sqrt{R^{-2}-1} - \mathrm{a\,tan}\sqrt{R^{-2}-1}\right], & R>1 \end{cases} \tag{5}$$

3. Change of the magnetic susceptibility

The effective magnetic susceptibility χ_e of a system with the linearly magnetizable spheres can be presented in the following form (see, for example, [14,22]):

$$\chi_e = 1 + \chi_p\varphi\frac{\langle h_p\rangle}{H} \tag{7}$$

Here H is the macroscopical field inside the sample, h_p is the field inside an arbitrary "trial" particle, $\langle h_p\rangle$ is the field, averaged over positions of all other particles, χ_p is the magnetic susceptibility of the particle material and φ is the volume concentration of the magnetic particles. The magnetic permeability of the polymer matrix equals to unity.

A strict solution of the problem of the determination of h_p is possible neglecting magnetic interactions between particles