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Numerical Analysis on Magnetic-induced Shear Modulus of Magnetorheological Elastomers Based on Multi-chain Model

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Based on the magnetic interaction energy, using derivative of the magnetic energy density, a model is proposed to compute the magnetic-induced shear modulus of magnetorheological elastomers. Taking into account the influences of particles in the same chain and the particles in all adjacent chains, the traditional magnetic dipole model of the magnetorheological elastomers is modified. The influence of the ratio of the distance between adjacent chains to the distance between adjacent particles in a chain on the magnetic induced shear modulus is quantitatively studied. When the ratio is large, the multi-chain model is compatible with the single chain model, but when the ratio is small, the difference of the two models is significant and can not be neglected. Making certain the size of the columns and the distance between adjacent columns, after constructing the computational model of BCT structures, the mechanical property of the magnetorheological elastomers composed of columnar structures is analyzed. Results show that, conventional point dipole model has overrated the magnetic-induced shear modulus of the magnetorheological elastomers. From the point of increasing the magnetic-induced shear modulus, when the particle volume fraction is small, the chain-like structure exhibits better result than the columnar structure, but when the particle volume fraction is large, the columnar structure will be better.

Key words: Magnetorheological elastomers, Shear modulus, Magnetic dipole model

I. INTRODUCTION

Magnetorheological (MR) materials are a class of smart materials whose rheological properties can be controlled by the application of an external magnetic field. MR fluids can reversibly change their states between free-flowing, linear viscous liquids and semisolids having controllable yield strength within milliseconds after a magnetic field is turned on or off. MR elastomers, which are composed of rubber and micron-sized magnetizable particles, can be thought of a new generation of MR materials. When the mixture is cured in the presence of a magnetic field, the particles in the matrix are arranged in chains, or columns due to the MR effect, and these structures remain in the matrix after curing. This kind of composite exhibits significant field controlled performance, mainly, magnetic field induced increase in shear modulus [1-5]. The shear modulus of MR elastomers can be simplified as the summation of the shear modulus when no magnetic field is applied (which is not studied here) and the magnetic field induced change in shear modulus. The magnetic-induced shear modulus of MR elastomer, which represents the field controllable performance, is one of its main mechanical properties. Until now, only a few theoretical models of MR elastomers have been developed, among which, the magnetic dipole model is widely used. H. Dang *et al.* revised the model of MR elastomer on this basis of distributional chains [6]. A special function was used to describe the distribution of chains, then the MR effect of distributional chains and MR elastomers with distributional chains were studied. Current models of MR elastomers are based on the magnetic interactions between particles within a single chain, even the magnetic interactions between two adjacent particles. Then these magnetic interactions are averaged over the entire sample to yield the bulk magnetorheological effect. Computational model for complex aggregates of particles for MR elastomers has not been seen yet.

In this work, the influences of adjacent chains on the magnetic-induced shear modulus of MR elastomers are considered, and the computational model of body-centered tetragonal (BCT) structure is presented. The magnetic-induced shear modulus of MR elastomers containing columnar structures is computed, thus the traditional magnetic dipole model of the magnetorheological elastomers is modified. The model examines the field-induced increment in shear modulus of MR elastomers, and provides theoretical guide for fabricating MR elastomers.

II. MODELING OF MR ELASTOMERS

Upon application of a magnetic field, the shear modulus of MR elastomers will substantially increase. The mechanism responsible for this bulk effect is the induced

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magnetic interaction of particles within the matrix. The conventional model of MR elastomers are based on a single chain model, assuming that the distance between adjacent chains is large. In this work, it is first supposed that the particle chains align along the direction of the applied magnetic field, and the chains are evenly spaced. The magnetic interactions of particles in the same chain and the particles in all adjacent chains are considered.

Neglecting the effect of strain rate, a quasi-static model of the MR elastomers is developed. In order to model the magnetic dipole interactions, several assumptions must be made. The particles are assumed to be uniform and homogeneous spheres that can be treated as identical dipoles. The particles are regarded as evenly arranged in chains in the composite and the magnetic field is regarded as static. For two dipoles with magnetic dipole moment $\vec{m_1}$ and $\vec{m_2}$ respectively, if they are separated by the distance $\vec{r}$, the interaction energy of these two point dipoles is

$$E_{12}=\frac{1}{4\pi\mu_0\mu_f}\left[\frac{\vec{m_1}\cdot\vec{m_2}}{r^3}-\frac{3}{r^5}(\vec{m_1}\cdot\vec{r})(\vec{m_2}\cdot\vec{r})\right] \quad (1)$$

where μ_f is the relative permeability of matrix in the composite, μ_0 is permeability in vacuum.

It is first assumed that the chains in MR elastomers do not form aggregate structures, and the chains are equally spaced, parallel with the applied magnetic field. Denote the distance between adjacent chains by D_0, and denote the center-to-center distance between nearby particles in a chain by d_0, in addition, R denotes the radius of spherical particle. Figure 1 indicates the Cartesian coordinates, where coordinates of an arbitrary particle is (x, y, z).

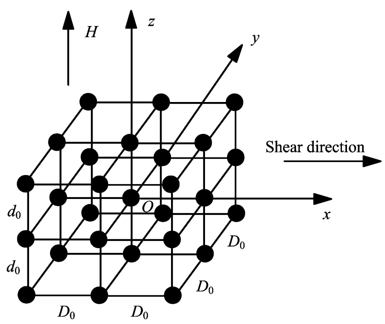

FIG. 1 Sketch of Cartesian coordinates

Denoting shear angle of the chains by θ, when the dipole moments are of identical strength m and are aligned in the same direction as applied field before deformation, Eq.(1) becomes

$$E_{12}=\frac{m^2}{4\pi\mu_0\mu_f}\left(\frac{1-3\cos^2\theta}{r^3}\right) \quad (2)$$

Assume that shear deformation is in the same direction as x axis, and u denotes the displacement of a particle in x direction, then the shear strain is $\gamma=\tan\theta=u/z$,

$$u=\gamma z \quad (3)$$

The chains are supposed to deform affinely with the shear strain, so the new coordinates of a particle after shear deformation is $(x+\gamma z, y, z)$, then

$$r=[(x+\gamma z)^2+y^2+z^2]^{1/2} \quad (4)$$

$$\cos^2\theta=\frac{z^2}{(x+\gamma z)^2+y^2+z^2} \quad (5)$$

Substitute Eqs.(4) and (5) into Eq.(2), and the following equation can be obtained

$$E_{12}=\frac{m^2}{4\pi\mu_0\mu_f}\frac{(x+\gamma z)^2+y^2-2z^2}{[(x+\gamma z)^2+y^2+z^2]^{5/2}} \quad (6)$$

Since the particle at origin of the Cartesian coordinates has magnetic interaction energy between all the particles in the composite, calculate the sum of Eq.(6), and then the interaction energy for a specific particle, for instance, at origin of the Cartesian coordinates, can be expressed as:

$$E=\sum\frac{m^2}{4\pi\mu_0\mu_f}\frac{(x+\gamma z)^2+y^2-2z^2}{[(x+\gamma z)^2+y^2+z^2]^{5/2}} \quad (7)$$

where $\sum$ indicates the sum over all the particles in the matrix.

For a MR elastomer in which particle volume percentage is ϕ, and total volume is V, the total magnetic energy density is:

$$\begin{aligned}E_d&=\frac{\phi V/2}{4\pi R^3/3}\frac{E}{V}=\frac{3\phi}{8\pi R^3}E\\&=\frac{3m^2\phi}{32\pi^2\mu_0\mu_f R^3}\sum\frac{(x+\gamma z)^2+y^2-2z^2}{[(x+\gamma z)^2+y^2+z^2]^{5/2}}\end{aligned} \quad (8)$$

The stress induced by the application of a magnetic field can be computed by taking the derivative of the magnetic energy density with respect to shear strain. The total number of particles in a MR elastomer is finite, so the sequence of sum and derivative can be reverse. The stress induced by the magnetic field is determined by

$$\begin{aligned}\tau&=\frac{\partial E_d}{\partial\gamma}=\frac{9m^2\phi}{32\pi^2\mu_0\mu_f R^3}\\&\times\sum\frac{z(x+\gamma z)[4z^2-(x+\gamma z)^2-y^2]}{[(x+\gamma z)^2+y^2+z^2]^{7/2}}\end{aligned} \quad (9)$$

Defining $x=kD_0$, $y=lD_0$, $z=nd_0$, since the particle chains are evenly spaced and parallel with each other, thus k, l, n are all integers. Defining $\lambda=D_0/d_0$, i.e., the ratio of the distance between adjacent chains to the distance between nearby particles in a chain, Eq.(9) can be written as:

$$\tau=\frac{9m^2\phi}{32\pi^2\mu_0\mu_f d_0^3R^3}\times\sum_{\substack{k=-k_{\max}\\l=-l_{\max}\\n=-n_{\max}}}^{\substack{k_{\max}\\l_{\max}\\n_{\max}}}\frac{n(k\lambda+\gamma n)[4n^2-(k\lambda+\gamma n)^2-(l\lambda)^2]}{[(k\lambda+\gamma n)^2+(l\lambda)^2+n^2]^{7/2}} \quad (10)$$

where $(k_{\max}D_0, l_{\max}D_0, n_{\max}d_0)$ is the coordinates of a particle whose distance to the origin is the largest.

The increase in shear modulus due to the magnetic field is simply stress divided by shear strain, i.e.

$$\Delta G = \frac{\tau}{\gamma} = \frac{9m^2\phi}{32\pi^2\mu_0\mu_f d_0^3 R^3\gamma} \times \sum_{\substack{k=-k_{\max}\\ l=-l_{\max}\\ n=-n_{\max}}}^{\substack{k_{\max}\\ l_{\max}\\ n_{\max}}} \frac{n(k\lambda+\gamma n)[4n^2-(k\lambda+\gamma n)^2-(l\lambda)^2]}{[(k\lambda+\gamma n)^2+(l\lambda)^2+n^2]^{7/2}} \tag{11}$$

This is the modified computational model for magnetic-induced shear modulus of MR elastomers in which the chains are evenly arranged and parallel with the applied field before shear deformation.

According to the traditional single chain model, the magnetic-induced shear modulus of MR elastomers is Ref.[1-3]:

$$\Delta G' = \frac{9m^2\phi\zeta}{4\pi^2\mu_0\mu_f {d_0}^3 R^3} \tag{12}$$

where $\zeta=\sum\limits_{n=1}^{\infty}(1/n^3)\approx1.202$. The ratio of Eqs.(11) to (12) is

$$\frac{\Delta G}{\Delta G'} = \frac{1}{8\zeta\gamma} \times \sum_{\substack{k=-k_{\max}\\ l=-l_{\max}\\ n=-n_{\max}}}^{\substack{k_{\max}\\ l_{\max}\\ n_{\max}}} \frac{n(k\lambda+\gamma n)[4n^2-(k\lambda+\gamma n)^2-(l\lambda)^2]}{[(k\lambda+\gamma n)^2+(l\lambda)^2+n^2]^{7/2}} \tag{13}$$

Thus, the relative error of traditional single chain model, which is independent of the distance between the adjacent particles in a chain, can be obtained.

A. Analysis of magnetic-induced shear modulus of chain-like structure

When subjected to a magnetic field, the magnetizable particles form a number of chains in the direction of the external magnetic field. It is assumed that the chains are evenly spaced and parallel with the direction of applied field, and particles are evenly distributed along the chains, i.e., chain-like structure.

Pick out a unit cell volume, as depicted in Fig.2. The unit cell is a cuboid, and D_0 is the length and width, in addition d_0 is the height. Extending the unit cell, we can get the chain-like structure. Defining $d_0=aR$, $\lambda=D_0/d_0$, the relationship between particle volume fraction and the dimension of the unit cell is:

$$\phi = \frac{4\pi R^3}{3{D_0}^2 d_0} = \frac{4\pi}{3\lambda^2 a^3} \tag{14}$$

$$\lambda = \sqrt{\frac{4\pi}{3\phi a^3}} \tag{15}$$

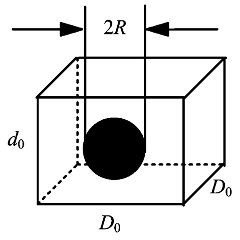

FIG. 2 Schematic cell

Limited by the capability of calculation, and without loss in generality, as a kind of approximating, while computing, the values of $k_{\max}$, $l_{\max}$, $n_{\max}$ are 300, 300, 1000, respectively. For small shear deformation, γ=0.0001 has been used for simplicity. Ratio of the increases in shear modulus due to the field calculated from two models for various λ is shown in Fig.3.

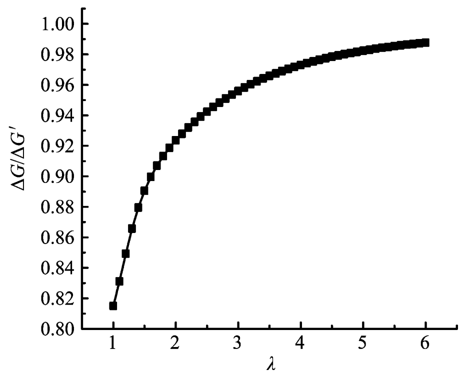

FIG. 3 Ratio of the increases in shear modulus due to the field calculated from two models

Figure 3 shows that the magnetic-induced shear modulus of the modified model is smaller than the one of traditional single-chain model, i.e., conventional point dipole model has overestimated the magnetic-induced shear modulus of the magnetorheological elastomers. Assuming the distance between adjacent particles in a chain is constant, when λ is small, namely, the distance between adjacent chains is small, compared to the modified model, the error of the conventional point dipole model will be significant, so the influences of particles in other chains can not be neglected. When λ is large, thus the distance between nearby chains is large, the proposed model is compatible with the traditional magnetic dipole model.

For small gaps between particles in a chain, i.e., the particles are close to each other within the chain, a=2 can be assumed. When particle concentration is ϕ=0.1, λ can be determined from Eq.(15), i.e., λ=2.288. Correspondingly, from Fig.3, $\Delta G/\Delta G'\approx0.935$ is obtained, namely, the relative error of conventional point dipole model is 6.5%. When the particle volume fraction increases, the value of λ will decrease, and then the relative error of the traditional dipole model will be more considerable.

B. Analysis of magnetic-induced shear modulus of columnar structure

For MR fluids, it is well known that the chains will aggregate into columnar structures, and when the particles are spherical, the steady state of the aggregate structure inside the column is BCT structure [6,7]. Since MR elastomers are analogue of MR fluids, the physical phenomena in MR elastomers are very similar to those in MR fluids. It can be assumed that during the curing procedure of fabricating MR elastomers, the chains will aggregate to form columns, and inside the columnar structures, the BCT structures exist. In the aspect of modeling of MR elastomers, there has not been any model concerning the BCT structures. In this section, computational model of BCT structures is constructed to calculate the magnetic-induced shear modulus of MR elastomers. The method will also be suitable for MR fluids.

For unit cell of BCT structure, the relative value of its dimension in three directions is $\sqrt{6}$:$\sqrt{6}$:2. Assume that the shortest edge of the BCT structure is in the same direction as the chains, and is parallel with the applied magnetic field. Expanding the BCT structure infinitely yields two classes of chains, which are all evenly spaced and parallel with the applied field. The discrepancy of the two classes is only the half unit space in each of three directions. Then the columnar structure is composed of two classes of chains and each class consists of chains evenly spaced and parallel with each other.

Analysis of magnetic-induced shear modulus of columnar structure is similar to that of chain-like structure, i.e., calculating the Eq.(11). The difference is that, for chain-like structure, there is only one class of chains; but for columnar structure, the computation is done for two classes of chains. Since the dimension of the column perpendicular to the applied field is finite, the magnetic interaction energy of particles in different chains inside the same column is somewhat discrepant. Average value for magnetic interaction energy of particles in different chains inside the same column is used when calculating the magnetic energy density in the composite.

Similar to the chain-like structure, it is assumed that the columns in MR elastomers are evenly spaced and parallel with each other. Then there are two questions to be resolved before the calculating of columnar structure: the dimension of the columns and the spacing between two adjacent columns. Assume that the columns are very long in the direction of the applied field, but in the direction perpendicular to the applied field, the dimension of the column is finite. For approximating, it is supposed that in a single column there are N^2 unit cells of BCT structure in the plane perpendicular to the applied field, namely, in each direction perpendicular to the magnetic field, the number of unit cells is N. When N varies, the dimension of the single column will also change. The total number of chains in one column is determined by

$$\mathrm{num} = (N+1)^2 + N^2 \tag{16}$$

Assume that ϕ denotes the particle volume fraction in the elastomer, and distances between adjacent particles in a chain is d_0=aR, then the number of particle chains in a unit cross section perpendicular to the applied field is

$$\mathrm{total} = \frac{\phi a R}{4/3\pi R^3} = \frac{3\phi a}{4\pi R^2} \tag{17}$$

The number of columns in a unit cross section perpendicular to the applied magnetic field is

$$N_t = \frac{\mathrm{total}}{\mathrm{sum}} = \frac{3\phi a}{4\pi R^2[(N+1)^2+N^2]} \tag{18}$$

So the distance between adjacent columns is equal to

$$d_{\mathrm{column}} = \frac{1}{\sqrt{N_t}} = \sqrt{\frac{4\pi R^2[(N+1)^2+N^2]}{3\phi a}} \tag{19}$$

Then the positions of the columns and the chains in the columns are all determined. Applying Eq.(13), sum over all the chains in columns. Limited by computational capacity, only 20 layers of columns surrounding one column are considered, i.e., the number of columns, which are evenly spaced and parallel with each other, is 40×40=1600 when calculating the magnetic energy density in the composite. For small gaps between particles in a chain, namely, the particles are close to each other within the chain, a=2 can be assumed. Given that particle volume fraction is 0.3, ratio of the increases in shear modulus due to the field calculated from two models as a function of N is shown in Fig.4 (a). For the case when there are hundreds of chains in a column [8], the value of N can be taken as 7, and ratio of the increases in shear modulus due to the field calculated from two models as a function of particle volume fraction ϕ is depicted in Fig.4 (b).

In Fig.4, it is shown that conventional single-chain point dipole model has overrated the magnetic-induced shear modulus of the magnetorheological elastomers again. The discrepancy will be bigger when the number of chains in a column increases, i.e., when the column is thicker. When particle concentration ϕ is constant, magnetic-induced shear modulus has a trend to decrease when the number of chains in a column increases. So when fabricating MR elastomers, the situation that the chains aggregate into very thick columns should be avoided. When the particle volume fraction is large, thus distance between adjacent columns is small, the discrepancy between the proposed model and the traditional model will be significant.

It is shown in Fig.4 (b) that, when ϕ=0.1 and N=7, for columnar structure, ratio of magnetic-induced shear modulus for modified model to conventional model is 0.915, which is slightly smaller than 0.935 of chain-like

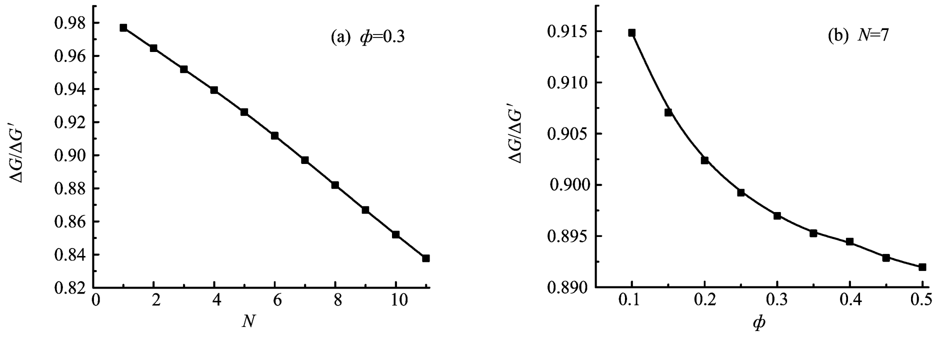

FIG. 4 Ratio of the increases in shear modulus due to the field calculated from two models

structure. When ϕ=0.4, λ equals to 1.144 for chain-like structure, and the ratio of magnetic-induced shear modulus for modified model to conventional model is 0.839, while the ratio for columnar structure is 0.894, which indicates that increment in shear modulus due to the field of columnar structure is larger than that of chain-like structure.

So it is obvious that when the particle volume percentage is small, in order to increase the magnetic-induced shear modulus of MR elastomers, the chain-like structure exhibits better result than the columnar structure, but when the particle volume fraction is large, the columnar structure will be better. Eq.(12) indicates that the magnetic-induced shear modulus of MR elastomers increases linearly with the particle volume fraction. Though Eq.(12), i.e., the traditional model is not accurate, but the error is not large enough, so the change in shear modulus of the elastomer in an applied magnetic field will still increase with the particle concentration. So when fabricating MR elastomers, in order to get bigger magnetic-induced shear modulus, people always incline towards choosing relative large particle volume fraction, and the columnar structure exhibits well, which is in accordance with people's general cognition [8].

III. CONCLUSION

In this work, on the basis of magnetic interaction energy, taking into account the influences of adjacent chains, a model is proposed to compute the magnetic-induced shear modulus of magnetorheological elastomers. This approach can improve the accuracy of previous models in the literature where only dipole interactions in a chain are considered. The calculation has been done for two kinds of structures: chain-like structure with chains evenly spaced and aligned in the same direction as external magnetic field, and columnar structure containing body-centered tetragonal (BCT) cells.

Results show that, conventional single-chain point dipole model has overrated the magnetic-induced shear modulus of the MR elastomers. If only consider the magnetic interaction energy, when the amount of chains in a single column is about one hundred, from the point of increasing the magnetic-induced shear modulus, when the particle volume fraction is small, the performance of chain-like structure is better than that of columnar structure, but when the particle volume fraction is large, the columnar structure will be better. For columnar structure, when the volume concentration of magnetizable particles is given, magnetic-induced shear modulus will decrease when the number of chains in a column increases.

It should be pointed out that, when the particles are close to each other, the theory of dipole model will not be accurate, and the effect of local field cannot be neglected and should be added into the proposed model. Especially for columnar structure, the particles pack tightly, and the effect of local field is significant. Furthermore, the influences of friction when shearing and the host matrix which is viscoelastic should be considered, so the multi-chain model needed to be improved further.

IV. ACKNOWLEDGMENT

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