Mechanical Properties of an Ideal Electrorheological Fluid: Metal-Particle/Insulating Oil System *

ZHAO He-ping(赵鹤平)[1,3], LIU Zheng-you(刘正猷)[1], SHEN Jia-rui(沈家瑞)[2], LIU You-yan(刘有延)[1,4]
[1]*Department of Physics,* [2]*Department of Polymer Materials, South China University of Technology, Guangzhou 510641*
[3]*Department of Physics, Jishou University, Jishou, Hu'nan 416000*
[4]*International Centre for Materials Physics, Chinese Academy of Sciences, Shenyang 110015*

(Received 12 October 1998)

The electrorheological properties of a model system consisting of metal particles with insulating coating in a dielectric oil are analyzed by extending Rayleigh identity based on a multipole expansion theory. Evaluation of the effective dielectric constant is emphasized. The dependence of the shear stress τ on the shear strain θ and the shear modulus G for the electrorheological solid is presented. It is found that the model system shows much higher electrorheological performance than the traditional suspensions.

PACS: 83.80.Gv, 41.20.−q

It has been known for a long time that metal particles embedded in a nonconducting matrix form an ideal dielectric. One would naturally suppose then that a system consists of metal particles in an insulating oil would be an excellent electrorheological (ER) fluid.[1] Of course, the particles must be coated with an oxide or other insulating layer to prevent electrical shorts through chains of contacting particles. The purpose of this paper is to deal with the mechanical properties of this kind of ER fluid by extending Rayleigh identity based on a multipole theory. Evaluation of effective dielectric constant is emphasized, and from its dependence on the shear strain, the dependence of the shear stress τ on the shear strain θ and the shear modulus G for the ER solid structure is presented specially.

The new ER system consists of the spherical particles of uniform size with radius R in a dielectric oil of dielectric constant $\epsilon_{\rm f}$. The particle has a metal core of radius $R_{\rm M}$ and an insulating coating of thickness $\delta = R - R_{\rm M}$, which we suppose to be much smaller compared to the radius R. The dielectric constant of the dielectric outer layer is $\epsilon_{\rm p}$ and both the insulating coating and the ambient medium have zero conductivity. The ER system is subjected to an applied electric field E_0. It has been found that the favored structure of ER solid is body-centered tetragonal (BCT),[2−4] for simplicity, we assume that the particles have been aligned along the applied field into chains and the solid structure has formed. The electric potential ψ can be expanded about a sphere centre O in the following ways:

$$\begin{aligned}
&\psi(r,\theta,\phi) = 0, \quad r < R_{\rm M},\\
&\psi(r,\theta,\phi) = \sum_{l=0}^{\infty}\sum_{m=-l}^{l}(A'_{lm}r^{l} + B'_{lm}r^{-l-1})Y_{lm}(\theta,\phi),\\
&\qquad\qquad R_{\rm M} < r < R,\\
&\psi(r,\theta,\phi) = \sum_{l=0}^{\infty}\sum_{m=-l}^{l}(A_{lm}r^{l} + B_{lm}r^{-l-1})Y_{lm}(\theta,\phi),\\
&\qquad\qquad r > R, \qquad (1)
\end{aligned}$$

where $Y_{lm}(\theta,\phi)$ is the spherical harmonics of order (l,m). The boundary conditions at the surface ($r = R_{\rm M}, R$) of the sphere result in the following relationships between the coefficients in the expansion of the potential outside the particle,

$$A_{lm} = \frac{B_{lm}}{R^{2l+1}} \cdot \frac{[l\alpha + (l+1)] + (l+1)(\alpha-1)(R_{\rm M}/R)^{2l+1}}{l(1-\alpha) - [l + (l+1)\alpha](R_{\rm M}/R)^{2l+1}}, \qquad (2)$$

where $\alpha = \epsilon_{\rm p}/\epsilon_{\rm f}$ is the ratio of the dielectric constant of insulating coating of the sphere relative to that of ambient medium.

Rayleigh[5] recognized that the terms in Eq. (1) for $r > R$ with negative powers of r are contributions to the potential ψ around the sphere centered at the origin coming from the induced multipole moments on the sphere itself. But the terms with positive powers of r come from the applied field and the multipole moments on all other spheres. These arguments enabled him to write the following identity:[5]

$$\sum_{lm} A_{lm}r^{l}Y_{lm}(\theta,\phi) = E_0 z + \sum_{i\neq 0}\sum_{lm}\frac{B_{lm}}{\rho_i^{l+1}}Y_{lm}(\theta_i,\phi_i), \qquad (3)$$

where the sum over i is understood as running over all spheres other than the central one. Coordinates (ρ_i,θ_i,ϕ_i) are measured relative to the centre of the i-th sphere and the applied field is assumed to be in the direction of the negative Z axis.

Equation (3) is valid for all points in the immediate environment of the sphere at the origin. Using the general form of the addition theorem of spherical harmonics and its completely orthonormal nature,[6] we get the extended Rayleigh identity:

$$\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left[\frac{\delta_{l,l'}\delta_{m,m'}(l+m)!}{T_l R^{2l+1}(l-m)!} + \frac{(-1)^{l'+m'+1}}{(l-m)!(l'-m')!} \cdot U_{l+l'}^{m-m'}\right]B_{l,m} = E_0\delta_{l',1}\delta_{m',0}, \qquad (4)$$

*Supported by the National Natural Science Foundation of China under Grant No. 19874021, and in part by the Foundation of Educational Committee of Hu'nan Province of China under Grant No. 98B087.
©by the Chinese Physical Society

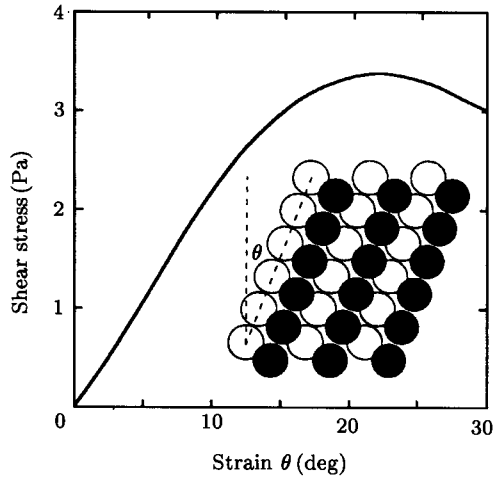

Fig. 1. Calculated stress in units of Pascal plotted as a function of strain, i.e., the tilted angle θ as defined in the inset. The parameters used in the calculation are $\epsilon_p/\epsilon_f = 5.0$, $E = 2.0\,\mathrm{kV/mm}$, $f = 0.2$, and $R_M/R = 0.985$.

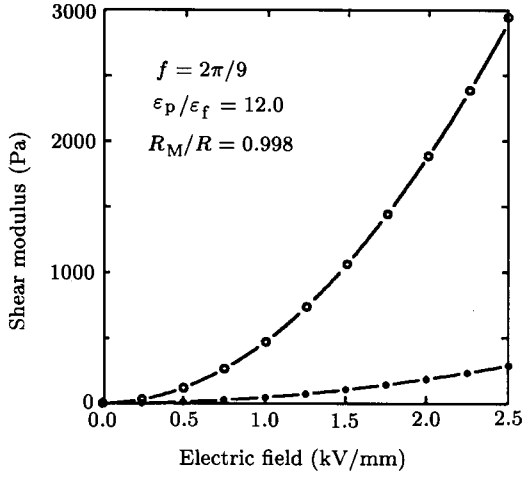

Fig. 2. Shear modulus G vs applied electric field E_0. Open circle indicates the shear modulus of the new ideal ER system and the closed circle shows that of the traditional suspension ($R_M = 0$).

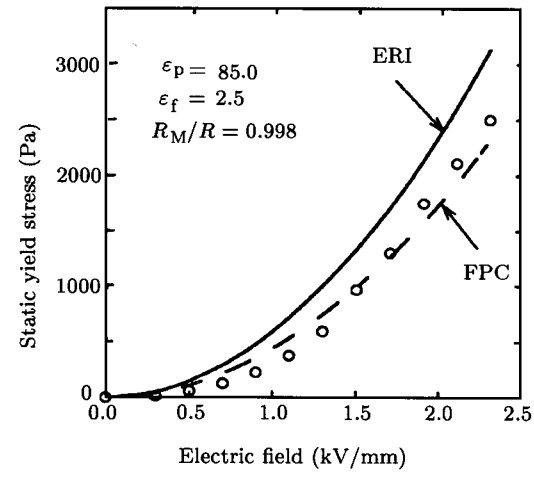

Fig. 3. Experimental and theoretical results of static yield stress vs the electric field. The solid line is the results of our extended Rayleigh identity (ERI) and the dashed line is from Tam's first principle calculation (FPC). Open circles indicate the experimental results by Tam *et al.*

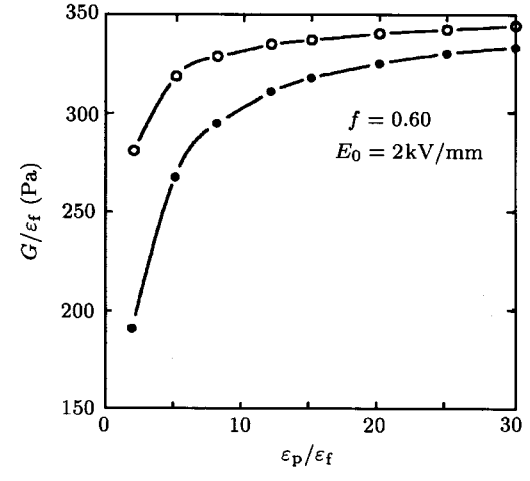

Fig. 4. Shear modulus G divided by ϵ_f vs ϵ_p/ϵ_f for various thickness of dielectric coating, $R_M/R = 0.985$ (open circles) and $R_M/R = 0.955$ (closed circles).

where

$$U_l^m = (l-m)! \sum_{i=1}^{\infty} \rho_i^{-l-1} P_l^m(\cos\theta_i)\cos(m\phi_i), \quad (5)$$

and m, m' range from $-l$ to l, and $-l'$ to l', respectively.

It turns out that ϵ_{eff} depends only on the coefficient $B_{1,0}$ in Eq. (1). The term in Eq. (1) containing $B_{1,0}$ is the potential of the induced dipole moment on a sphere. Comparing this term with the standard form of a dipole potential, we find the induced dipole per sphere is

$$p = -4\pi\epsilon_f B_{1,0}, \quad (6)$$

pointing to the Z direction. Then the effective dielectric constants is given by

$$\epsilon_{\mathrm{eff}} = \frac{D}{\epsilon_f E_0} = 1 - \frac{4n\pi B_{1,0}}{VE_0}, \quad (7)$$

where n is the number of particles containing in volume V.

In order to calculate the shear stress τ, it is necessary to perturb the system away from its original structure. We assume the BCT structure to be sheared by angle θ (inset in Fig. 1) and the shear rate to be infinitesimal. In the absence of nonelectrostatic forces, the free energy of the system changes $\Delta(-\boldsymbol{E}\cdot\boldsymbol{D}/2)$, thus shear stress is

$$\tau = -\frac{1}{2}\epsilon_0 \frac{\partial\epsilon_{\mathrm{eff}}(\theta)}{\partial\theta} E_0^2. \quad (8)$$

When a small shear applied to the system, the interparticle spacing d along z direction increases, which results in the principal variation of ϵ_{eff}. In this case, $\partial d/\partial\theta = \theta d$ and $\tau = G\theta$, so

$$G = \frac{1}{2}\epsilon_0(-d\frac{\partial\epsilon_{\mathrm{eff}}}{\partial d})E_0^2 \quad (9)$$

subject to the constraint of constant particle volume fraction.

By solving identity (4) for periodic structure of particles, we can obtain $B_{1,0}$ which may be used in Eq. (7) to calculate the effective dielectric constant. In order to investigate the influence of the increase of α on effective dielectric constants, we have calculated ϵ_{eff} of several kinds of structures for various α. It has been found that it is not as great as we had expected in such ER system. This may be due to the use of new particles.

Provided that the electrostatic ground state has a unique local spatial structure, the target configuration for the maximum ϵ_{eff} can be limited to periodic structures. We have performed numerical calculation of ϵ_{eff} for six periodic structures, the body-centered tetragonal, the face-centered cubic, the hexagonal, the body-centered cubic, the simple cubic and the diamond. It is found that at any given concentration of particles, body-centered tetragonal has the largest ϵ_{eff}. The fact that BCT is the favored structure of ER solid is in agreement with previous calculations based on dipole approximation[2] as well as more rigorous approach.[3,4]

Shear stress τ has been calculated for a typical volume fraction, $f = 0.2$, a typical electric field 2 kV/mm, and $R_{\text{M}}/R = 0.985$. From Fig. 1, we can see that τ almost increases linearly for small shear, the slope being the shear modulus G. As θ increases, τ reaches a maximum which is defined as the static yield stress τ_s, which is found to be approximately one half of G. Then the stress tends to decrease. It means that the lattice structure becomes unstable. That agrees with the result of Ma *et al.*[7]

Shear modulus G of the new ideal ER fluid has been compared with that of ER fluids consisting of dielectric particles with no metal core ($R_{\text{M}} = 0$) in the same dielectric medium in Fig. 2. The efficacy of the metal particles/insulating oil system is clear. The enhancement with orders of magnitude in the shear modulus is obtained by using the new ER fluids, when compared to that of traditional system. We have also compared our computated results τ_s to experimental and theoretical results by Tam *et al.*[8] in Fig. 3. We can see satisfying quantitative agreement (to the extent we can expect from such complex system). The discrepancy may come from the differentiation of ER systems, though they thought that the marterial for the core of their doubly-coated particles is immaterial to ER effect.

The dependence of shear modulus G divided by ϵ_{f} on the ratios of ϵ_{p} to ϵ_{f} for various thickness is shown in Fig. 4. We can see that the smaller δ leads to larger shear modulus for the same $\epsilon_{\text{p}}/\epsilon_{\text{f}}$. This agrees with the result of Davis[1] and Tam *et al.*[8] However, a substantial increase in G cannot be obtained by decreasing δ, because this would increase the field in the gap between the particles to near the break-down field.

A simple physical picture emerges from our results above. The mechanical properties of ER fluids are closely related to the electrostatic energy of the system. For a given applied electric field, the electrostatic energy can be maximized by field distribution and the dielectric constant of the materials. The metallic core of the particles limits the field distribution to the dielectric areas occupied by insulating coating and ambient dielectric medium. The thin dielectric outer layer produces high fields by keeping small separations among particles. That results in the dramatic ER enhancement.

In conclusion, we have presented a theoretical study of the mechanical properties of a new ER system which serves as a useful model to determine the ultimate performance of an ER fluid based on a polarization mechanism. Considering the function of the metal core, the particle density and the resultant sedimentation, it is suggested that the metal core can be replaced by the spherical dielectric particle with small density coated with thin metal film in practical experiment. Our results also show that there can be a path of continuous improvement through which optimal ER fluid performance may be achieved. Equation (4) has a general form. It can be used to investigate other properties of ER fluids.

REFERENCES

[1] L. C. Davis, J. Appl. Phys. 73 (1993) 680; M. M. Kharadly and W. Jackon, Proc. Electron. Eng. 100 (1952) 199.
[2] R. Tao and J. M. Sun, Phys. Rev. Lett. 67 (1991) 398.
[3] H. P. Zhao, Z. Y. Liu, Y. Y. Liu and J. R. Shen, Phys. Lett. A 239 (1998) 393.
[4] H. Clercx and Bossis, Phys. Rev. E 48 (1993) 2721.
[5] L. Rayleigh, Philos. Mag. 34 (1892) 481.
[6] R. C. McPhedran and D. R. McKenzie, R. Soc. London. Ser. A 359 (1977) 45.
[7] H. Ma, W. Wen, W. Y. Tam and P. Sheng, Phys. Rev. Lett. 77 (1996) 2499.
[8] W. Y. Tam, G. Yi, W. Wen, H. Ma, M. M. T. Loy and P. Sheng, Phys. Rev. Lett. 78 (1998) 2987.