

# Particle Shape Dependence of Polar Molecule Dominated Electrorheological Effect

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**Abstract.** We investigated the relations between the yield stress of polar-molecule dominated electrorheological (PM-ER) fluids and the shape of ellipsoidal particles of the fluids. It is found that, when the short axis of an ellipsoidal particle is along an external electric field while the volume of the particle keeps constant, the shorter of the short axis is, the stronger the attraction between the two adjacent particles would be. The equipment used for yield stress determination, the experiment and calculation of the condition under which the sample are short-axis chained, the detailed process of sample preparation and sample characterization will be described and discussed in an extended form of this paper.

## 1. Introduction

In the PM-ER model, the interaction between charges and polar molecules adsorbed on particle surface is a key to the PM-ER effect.[1] In this work, we will discuss the relations between the shape of ellipsoidal dielectric particles and their attractive force according to the PM-ER theory. The result indicates that the yield stress increases by changing the shape of particle from spherical to ellipsoidal.

## 2. Theory

We will first discuss the interaction between charges and polar molecules adsorbed on particle surfaces. According to PM-ER theory [1], the polar molecules are relatively free for rotation and potentially to be oriented in the direction of a strong electric field.[2,3] The attractive force density  $f_{m-e}$  of molecule-charge can be expressed as  $f_{m-e} = e\mu(2\pi\epsilon_0\epsilon_r d_{m-e}^3)^{-1}$ . When two or more polar molecules, which are adsorbed on particle surface, exist in the gap between two adjacent particles, the attractive force  $F_{m-e} = Nf_{m-e}$ , where  $N$  is the number of active polar-molecule attractions in the gap.  $N = \rho_m n(E) S$ , where  $\rho_m$  is the density of the polar molecules adsorbed on the particles, the probability  $n(E)$  of the aligning polar molecules being nearly proportional to  $\mu E_{loc}$ [4]. When the dielectric particle is spherical, the effective area  $S$  is nearly  $\pi r d_{m-e}$ , [1] where  $d_{m-e}$  is the distance from molecule centre to charge, and the molecule size  $d \approx 2 d_{m-e}$ .

When the particle is ellipsoidal, the effective area  $S$  can be calculated by following process.

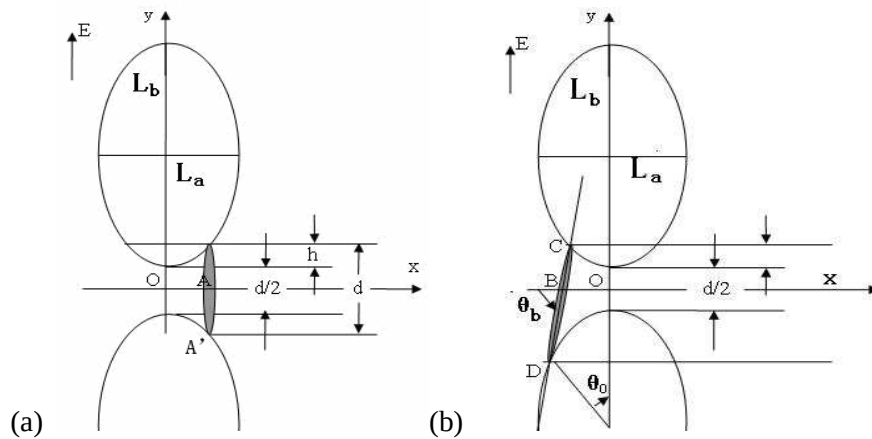


**Figure 1** Ellipsoid particles and a polar molecule parallel to  $E$  (a) and not parallel to  $E$  (b).

Polar molecules rotate under external electric field  $E$  and turn to the external field direction, as shown in figure 1(a). The molecule size  $d=0.4\text{nm}$ , which is approximately a double of the distance  $d_{m-e}$  from molecule center to charge. Suppose the gap between two particles is  $d/2$ , so  $h=d/4$ . The length of semi-axis along  $x$  is  $L_a$ , and that along  $y$  is  $L_b$ . The Langevin function [4] can be expressed as  $L(a) = \overline{\cos \theta} = a/3 - a^3/45 + 2a^5/945 - 2a^7/9450 + \dots$ , where  $a = \mu E/kT$ . When  $a$  is not very large,  $L(a) \approx a/3$ , or  $L(a) = \mu E_{loc}/3kT$ .

If polar molecules orientate along external field direction,  $\theta = 0$ , one has  $L(a)=1$ ,  $E_{loc}=3kT/\mu \approx 186 \text{ kV/mm}$  when the dipole moment  $\mu=2D$ .

In figure 1(b), the angle between the polar molecule and the external field direction is  $\theta$ . When  $L_a$  and  $L_b$  are constants, the elliptic equations of two particles are expressed as

$$x_1^2/L_a^2 + (y_1 - L_b - d/4)^2/L_b^2 = 1, \quad (1)$$

$$x_2^2/L_a^2 + (y_2 + L_b + d/4)^2/L_b^2 = 1. \quad (2)$$

The slope  $s$  of line  $CD$  can be calculated by coordinates of the point  $C(x_1, y_1)$  and  $D(x_2, y_2)$ ,  $(y_1 - y_2) = s(x_1 - x_2)$ . The distance  $d$  between  $C$  and  $D$  is polar molecule size,  $(x_1 - x_2)^2 + (y_1 - y_2)^2 = d^2$ . Solving above equations, we can get the value of  $x_1, x_2, y_1$  and  $y_2$ . When polar molecules turn to the external field direction and the molecules touch both two adjacent particles as shown in figure 1(a), coordinate of contact point  $A'(x_{A'}, y_{A'})$  on the lower particle can be expressed as  $x_{A'} = L_a \left[ 1 - (y_{A'} + L_b + d/4)^2/L_b^2 \right]^{1/2}$ , and  $y_{A'} = -d/2$ .

The distribution of local electric field calculated with finite element analysis shows a sharp peak with peak value of about  $200 \text{ kV/mm}$  when the external electric field  $E=2\text{kV/mm}$  and  $L_a=15\text{nm}$ .  $E_{locA}$ , the local electric field at centre point  $A$  of the polar molecule parallel to the external field can be obtained through local electric field distribution.  $E_{locA}$  is close to  $E_{locA'}$ . The Langevin function  $L(a) = \overline{\cos \theta} \approx \mu E_{loc}/3kT$ , where  $k$  is Boltzmann constant,  $T=300\text{K}$ ,  $\mu=2D$ . It is supposed that the polar molecule does not align along the external field direction as shown in figure 1(b) and  $x_B$  is slightly smaller than  $x_{A'}$ . We can also get the value of  $E_{locB}$  through the local electric field distribution. The value of slope  $s$  is  $\tan(\theta_B)$ , so  $\theta_B = \arccos(\mu E_{locB}/3kT)$ .

Define  $d_B$  as the distance between two intersections of two ellipses with a line which is passing through point  $B$ . In figure 1(b),

$$d_B = CD = (x_1 - x_2)(\cos \theta_B)^{-1} = 3kT(x_1 - x_2)(\mu E_{locB})^{-1} \quad (3)$$

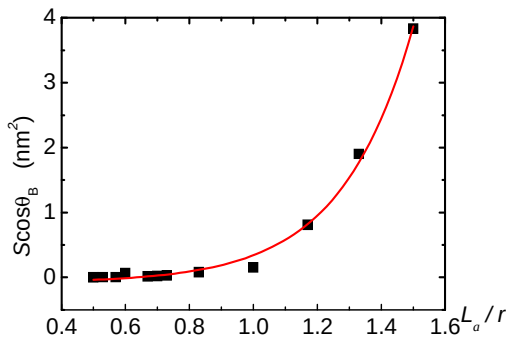


**Figure 2** Effective area which is proportional to attractive force vs. axis length perpendicular to E.

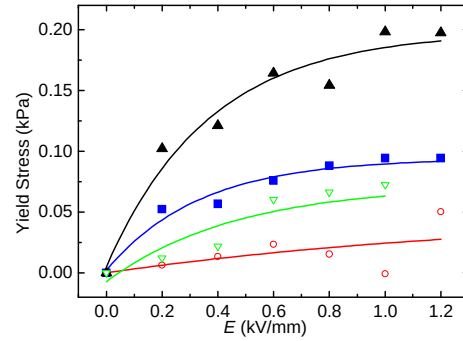


**Figure 3** The yield stress of ellipsoidal particles versus electric field (see text for notations).

The value of  $x_B$  can be changed slightly and  $CD=d_B$  in figure 1b can be closer to the molecule size  $d$ . When  $|d_B - d|/d \leq 10^{-3}$ , the polar molecule is thought to be adsorbed on two adjacent particles. The effective contact area  $S$  is an area of a particle on which all polar molecules are adsorbed on two adjacent particles respectively under an electric field. The effective contact areas  $S_<$  (for  $L_a < L_b$ ),  $S_ =$  (for  $L_a = L_b$ ) and  $S_>$  (for  $L_a > L_b$ ) can be calculated as follows.

$$S_< = \pi \int_0^{\theta_0} L_a \sin \theta \left[ (-L_b \sin \theta)^2 + (L_a \cos \theta)^2 \right]^{1/2} d\theta$$

$$= \pi L_a L_b \left[ (1 - \varepsilon^2)^{1/2} + \varepsilon^{-1} \arcsin \varepsilon - \cos \theta_0 (1 - \varepsilon^2 \cos^2 \theta_0)^{1/2} - \varepsilon^{-1} \arcsin (\varepsilon \cos \theta_0) \right]; \quad (4)$$

$$S_ = \pi L_a L_b \int_0^{\theta_0} \sin \theta d\theta = \pi L_a L_b (1 - \cos \theta_0); \quad (5)$$

$$S_> = \pi \int_0^{\theta_0} L_a \sin \theta \left[ (-L_b \sin \theta)^2 + (L_a \cos \theta)^2 \right]^{1/2} d\theta$$

$$= \pi L_a L_b \left\{ (\varepsilon^2 + 1)^{1/2} + \varepsilon^{-1} \ln \left[ \varepsilon + (\varepsilon^2 + 1)^{1/2} \right] - \cos \theta_0 (\varepsilon^2 \cos^2 \theta_0 + 1)^{1/2} \right. \\ \left. - \ln \left[ \cos \theta_0 + \varepsilon^{-1} (1 + \varepsilon^2 \cos^2 \theta_0)^{1/2} \right] \right\}, \quad (6)$$

where  $\varepsilon = [(L_b^2 - L_a^2)/L_b^2]^{1/2}$ , and  $\theta_0 = \arctan \left[ -x_2 (L_b + d/4 + x_2 \tan \theta_B)^{-1} \right]$ .

Effective area  $S$  can be calculated through an integral and expressed analytically.  $E_{\text{locB}}$  is the local field around the edge of effective area on the plane across the center point of the particle gap. This plane is perpendicular to the external field direction.  $N$  is the number of active polar-molecule interactions in gap, and  $N = S \rho_m n(E)$ . If the density of polar molecule adsorbed on particle surface keeps constant,  $N$  is proportional to  $Sn(E)$ .

### 3. Results and discussions

The particle-particle attractive force  $F$  can be expressed as  $F = N f_{m-e}$ ,  $F \propto Sn(E) \propto Scos\theta_B$ , so we get figure 2, where  $r$  is the diameter of spherical particle when the volume of a particle keeps constant.

As shown in figure 2, the effective contact area, which is proportional to the attractive interaction of two ellipsoid particles, is simulated when  $L_a/r = 1.5$ , and it is about 25 times larger than that of two spherical ones.

If the ellipsoid particles are soft and the contact surface of particles can be flattened, the effective area increases, and so is the yield stress. If we can get the image of particles and the density of polar molecule adsorbed on particle surface through experiment, an exact model is found through a finite element analysis and a definite attractive force will be presented.

This modeling approach has many advantages. Finite element analysis can deal with non-sphere particles such as ellipsoid. The model can be easily integrated into complex shape particle simulation. Multi-field including magnetic field and optical field is allowed in finite element analysis. Finally, the model can be extended to hundreds or more particles simulation.

Ellipsoidal dielectric particles will orient by its longest dimension along the external field direction (z-direction) because z component of dielectric constant is larger than x or y component. If x or y component of dielectric constant is larger than z component, ellipsoidal dielectric particles will align along short axis under external electric field so as to larger yield stress.

#### 4. Experimental Section

We prepared submicrometer-sized mesoporous silica SBA-15 with cuboidlike morphology. The channels of the material run parallel to the short axis of the cuboidlike SBA-15. A ferroelectric material such as potassium dihydrogen phosphate ( $\text{KH}_2\text{PO}_4$ ) with large dielectric constant was interpolated in the channels. The channels of the material run parallel to short axis.

The yield stress of several cuboid-like SBA-15ER fluids (solid volume fraction is about 25%) versus  $E$  is shown in figure 3. Solid upper triangles (or solid squares) denote the yield stress of the elliptic plate sample doped with  $\text{KH}_2\text{PO}_4$  and adsorbed with (or without) polar molecules; open lower triangles (or open circles) denote that for samples without  $\text{KH}_2\text{PO}_4$  but adsorbed with (or without) polar molecules. Solid curves are the fitting for the corresponding data points. The yield stress of ER fluid consisting of the  $\text{KH}_2\text{PO}_4$ -doped material with coating of polar molecules in silicon oil has highest yield stress. This is because that adding ferroelectric materials in the channels parallel to the short axis would make the particles contact to each other with their short axis parallel to the external electric field, and this would increase the effective area, as well as the yield stress. The calculation of the condition under which the sample are short-axis chained, as well as the detailed process of sample preparation and sample characterization will be described and discussed elsewhere.

#### 5. Conclusion

The shape of particles suspended in a liquid has a direct effect on ER effects. Effective area on particle surface plays a crucial role in PM-ER fluids. The ellipsoid particles with short axis parallel to the external field direction have much larger effective area and attractive force than spherical ones when the volume of one particle keeps constant. The yield stress will increase by 25 times when the length of the axis perpendicular to field direction is 1.5 times longer than the sphere radius according to our simulation. Adding ferroelectric materials in the channels parallel to the short axis would make the short axis parallel to the field direction, and the PM-ER fluid consisting of such ellipsoidal particles should have higher yield stress in comparison with spherical ones.

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