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A general constitutive equation of an ER suspension based on the internal variable theory

B. Wang, Harbin, China, and **Z. Xiao,** Singapore

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Summary. A microstructural constitutive theory of ER suspensions was formulated in this investigation. The framework was based on the internal variable theory and the mechanism analysis. The ER suspension consists of fine particles with high dielectric constant and the supporting fluid. Under the action of the electric field, the polarized particles will aggregate together to form the chain-like structures along the direction of the electric field. As the size and orientation of the particle aggregates are volatile, and they adjust according to the applied electric field and strain rate, the energy conservation equation and the force equilibrium equation were thus established to determine the orientation and size of the aggregates. Following that, a three-dimensional, explicit form of the constitutive equation was derived based on the interaction energy and the dissipation function of the system. The response of the system under the action of a simple shearing load was considered and discussed in detail. It is found that the shear-thinning viscosity of an ER suspension is well approximated by the power-law $\propto (Mn)^{-0.82}$.

1 Introduction

An electrorheological fluid (ER) consists of a suspension of the dielectric particles in a liquid of low dielectric constant and low viscosity. Its apparent viscosity increases dramatically in the presence of an applied electric field. The phenomenon is reversible. Upon electric field cutoff, the system almost immediately resumes its original liquid state. These novel properties make ER fluids very attractive for many futuristic technologies. Recently they are also used as the components in some smart structures. It is now clear that the underlying mechanism for the transition is that the polarized particles in ER fluid will form the chain structures along the direction of the applied electric field. A lot of work has been done to understand the mechanism of the chain formation [22], interesting readers are referred to the comprehensive review paper by Parthasarathy and Klingenberg [17].

The macroscopic response of an ER suspension depends strongly on the applied electric field and its microstructural parameters, such as the volume fraction and dielectric constants of the particles. In order to reveal the relationship between the macroscopic response of an ER system and its microstructures, a constitutive equation should be established. This equation can also provide engineers the necessary tool to carry out the finite element analysis for a structure with ER systems as its components. Many researchers have done some fruitful investigations on the constitutive relation under the simple shear load. To name a few, Halsey et al. [9], [10] proposed a stimulating model to predict the shear-thinning viscosity of the fluid. Based on the continuum concept of unsymmetric stress state, Rosensweig [19] developed an efficient method to obtain

the general expressions for the yielding stress of magnetorheological fluid. Klingenberg and Zukoski [14] first considered a model ER suspension with an idealized structure. They calculated the elastic behavior under simple shearing deformation. Bonnecaze and Brady [1], [2] developed a molecular dynamics-like simulation method to efficiently compute the electrostatic interactions in suspensions with arbitray particle configuration under the action of an electri field and a flow field. Ginder and Ceccio [7], and Conrad et al. [5] carried out extensive theoretical and experimental investigation on the yield strength of ER systems. Based on the energy consideration, Bossis et al. [13] predicted the yield stress in magnetorheological and electrorheological fluids. Martin and Odinek [16] developed a non-linear rheological model of an ER system by considering the response of a fragmenting and aggregating particle chain to the prevailing hydrodynamic and electrostatic forces.

In fact, the response of an ER suspension is quite different in the different stages of the applied strain rate. For quasi-static loading, the chain structure will not break down when the strain is small. The particles will move a very limited distance in response to the applied strain. Under such kind of loading, the stress is related with the applied strain, not with the strain rate. The stress-strain relations obtained by many researches are the most suitable for this stage. For dynamic loading, or when the strain rate is high, the chain structure becomes volatile whose size adjusts in response to the flow, fragmenting and aggregating. As the shear strain rate increases or decreases, its orientation also adjusts in response to the flow. Such microstructural evolution will induce further energy dissipation, and constitute the main reason for the shear-thinning phenomena. In this stage of loading, the stress is related with the strain rate, instead of the strain. As was pointed out by Jordan et al. [12], [13], to develop the constitutive equation for ER fluid, two approaches can be followed. Namely, one is based on detailed microstructural electrohydrodynamics, while the other is based on continuum mechanics. The former has been successful in elucidating interrelations of system variables, but it has failed to generate closed form constitutive equations. Continumm mechanics based models, on the other hand, lack detailed microstructural information. In the current investigation, we attempt to establish a three-dimensional constitutive equation based on the internal variable theory [18], [23]. Starting with the microstructural consideration, the interaction energy and the dissipated energy under the action of an electric field and a flow field will be derived first. Then, based on the ellipsoidal aggregate assumption, the evolution equations of the internal variables, such as the orientation and size of the aggregates will be established. At last, the three-dimensional constitutive relation is obtained in the framework of the internal variable theory. The mechanism-based constituitve model obtained in the current research not only provides the engineer with the closed form equation to run FEM, but also establishes the relationship between the macroscopic response and their microstructures of ER suspensions.

2 The interaction energy of an ER suspension containing the aggregates of polarized particles

It is a well-known fact that the rheology of ER suspensions at low to moderate volume fractions is due to the aggregation of particles into volatile chainlike structures whose size and orientation adjust in response to the flow and applied electric field. The electrodes as the boundary of the system have a very strong image effect on the aggregation process of polarized particles when the size of the aggregates approaches to the distance between the electrodes. In establishing the microstructural constitutive relation of a material, a constitutive element is

usually taken as the subject of study. From a macroscopic view, the element should be small enough to represent the behavior of one point in the material, whereas from a microscopic view the element should be large enough to contain sufficient microstructural information. Therefore, an ER suspension confined by the electrodes can be considered to be piled up by a large number of such element, each of which may have different response due to the effect of non-uniform applied electric and flow field. Thus in establishing the constitutive relation of the material, the image effect of the electrodes should not be taken into consideration. The material element we consider is assumed as a suspension of the spheroidal aggregates of polarized particles in a fluid. This assumption will simplify our analysis since the polarization is constant inside an ellipsoid placed in a constant external field. Similar assumptions can be easily found in literature, such as Halsey et al. [9], Bossis et al. [3], etc. The element is subjected to an applied electric field $\vec{E}^0$ along the z-direction, and a general flow velocity field $\vec{V}^0$. If the size of a spheroidal aggregate is denoted by $a_1 = a, a_2 = a_3 = c$, it's volume is given by $v_a = \frac{4}{3}\pi a c^2$. In a unit volume of the ER suspension with the volume fraction of ϕ of the particles, the number of the aggregates in a unit volume is given by $N = \phi/v_a$. The aggregate consists of the dielectric particles with the isotropic, relative permittivity α^p, whereas the fluid has a lower relative dielectric permittivity α^f. When the suspension system is subjected to an applied electric field, the interaction depolarization energy due to the introduction of the dielectric aggregates can be obtained after determining the distribution of the electric field. In what follows, we will derive the electrostatic energy in a unit volume.

We first consider a single prolate spheroid aggregate in the fluid. To incorporate the effect of the other aggregates, the concept of the effective field $\vec{E}^m$ is introduced, which is defined as the volume average field in the surrounding fluid. In the local coordinate system where X, Y, and Z axes are connected with the semi-axes $a_1, a_2, a_3 (a_2 = a_3 = c)$ of the spheroid, the electric field inside the aggregate is related with the effective field similar to single dielectric inclusion problem [15], [16],

$$
\begin{aligned}
E_x^i &= \frac{E_x^m}{1 + (\alpha^p/\alpha^f - 1)n_x} = A_x E_x^m, \\
E_y^i &= \frac{E_y^m}{1 + (\alpha^p/\alpha^f - 1)n_y} = A_y E_y^m, \\
E_z^i &= \frac{E_z^m}{1 + (\alpha^p/\alpha^f - 1)n_z} = A_z E_z^m,
\end{aligned}
\tag{1}
$$

where $n_x = \frac{1-\beta^2}{2\beta^3}(\ln\frac{1+\beta}{1-\beta} - 2\beta), n_z = n_y = \frac{1}{2}(1 - n_x)$, and $\beta = \sqrt{1 - c^2/a^2}$.

If the applied electric field is denoted by $\vec{E}^0$, it should be equal to the volume average of the electric field as follows:

$$
(1 - \phi)\vec{E}^m + \phi\vec{E}^i = \vec{E}^0. \tag{2}
$$

Substitution of Eq. (1) into Eq. (2) gives the effective electric field as follows:

$$
\begin{aligned}
E_x^m &= (1 - \phi + \phi A_x)^{-1} E_x^0, \\
E_y^m &= (1 - \phi + \phi A_y)^{-1} E_y^0, \\
E_z^m &= (1 - \phi + \phi A_z)^{-1} E_z^0.
\end{aligned}
\tag{3}
$$

The total induced dipole moment in a spheroid aggregate is thus given by

$$P_x = v_a A_x(\alpha^p - \alpha^f)\alpha_0(1 - \phi + \phi A_x)^{-1}E_x^0,$$

$$P_y = v_a A_y(\alpha^p - \alpha^f)\alpha_0(1 - \phi + \phi A_y)^{-1}E_y^0, \tag{4}$$

$$P_z = v_a A_z(\alpha^p - \alpha^f)\alpha_0(1 - \phi + \phi A_z)^{-1}E_z^0,$$

where α_0 is the dielectric permittivity of the vacuum, and v_a is the volume of the ellipsoidal aggregate.

The interaction energy of a single aggregate is defined as the electrostatic energy change due to the introduction of one aggregate into the fluid, and is given by [15]

$$\begin{aligned} u &= -\frac{1}{2}\vec{E}^m \cdot \vec{P} \\ &= -\frac{1}{2}v_a\alpha_0(\alpha^p - \alpha^f)[A_x(1 - \phi + \phi A_x)^{-2})(E_x^0)^2 \\ &\quad + A_y(1 - \phi + \phi A_y)^{-2}(E_y^0)^2 + A_z(1 + \phi + \phi A_z)^{-2}(E_z^0)^2]. \end{aligned} \tag{5}$$

The energy of the ER suspension can be considered to consist to two parts: one is the bulk depolarization energy of the dielectric aggregates, the other part is the surface energy of the aggregates. As pointed out by Halsey et al. [9], the surface energy arises also from the dipole interaction, but it should depend on the lattice constant of the aggregates. Bossis et al. [4] also pointed out that the origin of the surface energy was the difference between the local field on a particle situated on the surface of an aggregate relatively to the local field on a particle situated inside the aggregate. As a result, the surface energy is much smaller than the bulk depolarization energy. To simplify the analysis, we neglect the surface energy in this paper as the majority of previous investigations did.

In a unit volume there are N aggregates, therefore the total interaction energy can be written as

$$\begin{aligned} \Psi &= Nu \\ &= -\frac{\phi}{2}(\alpha^p - \alpha^f)\alpha_0[A_x(1 - \phi + \phi A_x)^{-2}(E_x^0)^2 \\ &\quad + A_y(1 - \phi + \phi A_y)^{-2}(E_y^0)^2 + A_z(1 - \phi + \phi A_z)^{-2}(E_z^0)^2]. \end{aligned} \tag{6}$$

For our problem, the applied electric field E^0 is along the z-axis of the global coordinate system. In the local coordinate system, (X, Y, Z)axes are connected with the semi-axes a_1, a_2, a_3, which can be specified as follows: X is the symmetric axis, and Z lies in the (x, y) plane of the global coordinate system. Thus, the components of the applied electric field in the local coordinate system can be expressed in the form as

$$\begin{Bmatrix} E_x^0 \\ E_y^0 \\ E_z^0 \end{Bmatrix} = \begin{bmatrix} \sin\theta\cos\varphi & \sin\theta\sin\varphi & \cos\theta \\ -\cos\theta\cos\varphi & -\cos\theta\sin\varphi & \sin\theta \\ \sin\theta & -\cos\varphi & 0 \end{bmatrix} \begin{Bmatrix} 0 \\ 0 \\ E^0 \end{Bmatrix}, \tag{7}$$

where θ is the angle between the symmetric axes X and z, and φ is the angle between the projection of X onto the plane (x, y) and axis x.

By substituting Eq. (7) into Eq. (6), and noting $A_y = A_z$ for spheroidal aggregates, one obtains the interaction energy

$$\Psi = -\frac{\phi}{2}(\alpha^p - \alpha^f)\alpha_0(E^0)^2[A_x(1-\phi+\phi A_x)^{-2}\cos^2\theta + A_y(1-\phi+\phi A_y)^{-2}\sin^2\theta]. \tag{8}$$

It is very clear that the specific interaction energy of the suspension system depends on the applied electric field, the orientation, size, and volume fraction of the aggregates. Further, the interaction energy is not the linear function of the volume fraction of the particles since we introduced the concept of the effective field to consider the interaction effects among the aggregates.

3 The dissipation potential of ER suspensions subjected to microstructure evolution

To establish the constitutive relation of the suspensions, one needs first to derive the dissipation potential corresponding to the microstructural evolution. The dissipation potential is defined as the dissipated energy in unit volume and unit time. In the current model, the suspension element is subjected to an electric field $\vec{E}^0$ along the z-direction, and a linear flow velocity field $\vec{V}^0$, (i.e., a constant strain rate field). The orientation of the aggregates changes with the velocity $\dot{\theta}, \dot{\varphi}$. The size of the aggregates also adjusts in response to the flow and electric field. If the translation velocity of the **i**th aggregate center is denoted as $\vec{U}_i$, which is assumed to be the same as the flow velocity of the fluid at that point, the velocity of the fluid adhering to the particle surfaces can be written as follows:

$$\vec{V}_i = \vec{U}_i + \vec{w}_i \times \vec{r}_i, \quad \text{on each} \quad S_i, \tag{9}$$

where $\vec{w}_i$ refers to the angular velocity of the ith aggregate, which can be expressed in terms of $\dot{\theta}, \dot{\varphi}$; $\vec{r}_i$ denotes the position vector drawn from the center of the aggregate, and S_i is the surface of the ith aggregate. Without the aggregates, the flow field of the fluid is given by

$$\vec{V}_i^{\,0} = \vec{U}_i + \tilde{\gamma}^0 \cdot \vec{r}_i, \tag{10}$$

where $\tilde{\gamma}^0$ is the applied strain rate tensor.

For quasi-static creeping flows in the absence of external body forces, the kinetic energy of the fluid-particle system is negligible, and the potential energy of the fluid remains constant. Accordingly, the rate Φ at which energy is being dissipated within the confines of the apparatus is equal to the rate of work done by the stresses over all the surfaces bounding the fluid. In general, this overall surface includes both the apparatus boundaries and the particle surfaces. Hence,

$$\Phi = \int\int_{S_0+\sum S_p} \prod_{ij} V_i n_j \, ds, \tag{11}$$

where S_0, S_p are the surface of the material element and the spheroid aggregates, V_i is the velocity vector of the fluid on the surface, n_j is the normal vector of the surface, which is directed outward the fluid, and $\prod_{ij}$ is the stress tensor. For Newtonian supporting fluids, it can be expressed in the form:

$$\prod_{ij} = -p\delta_{ij} + \mu(\partial_i V_j + \partial_j V_i), \tag{12}$$

in which p is the hydrostatic pressure, and μ is the shear viscosity of the fluid.

By using the condition $V_i = V_i^0$ on the element surface S_0, Eq. (11) can be expressed in the form:

$$\Phi = \int\limits_{S_0+\Sigma S_p}\int \prod_{ij} V_i n_j \, ds$$

$$= \int\limits_{S_0+\Sigma S_p}\int \prod_{ij} V_i^0 n_j \, ds + \int\limits_{\Sigma s_p}\int \prod_{ij} (V_i - V_i^0) n_j \, ds. \tag{13}$$

By introducing the stress tensor $\prod_{ij}^0$ corresponding to the applied flow field V_i^0 in the homogeneous fluid without particles, the reciprocal theorem [11] gives

$$\int\limits_{S_0+\Sigma S_p}\int \prod_{ij} V_i^0 n_j \, ds = \int\limits_{S_0+\Sigma S_p}\int \overset{0}{\prod_{ij}} V_i n_j \, ds. \tag{14}$$

The above relationship is used to replace the first integral in Eq. (13), and by using the boundary condition again, Eq. (13) becomes

$$\Phi = \int\limits_{S_0+\Sigma S_p}\int \prod_{ij} V_i n_j \, ds$$

$$= \int\limits_{S_0}\int \overset{0}{\prod_{ij}} V_i^0 n_j \, ds + \int\limits_{\Sigma S_p}\int \overset{0}{\prod_{ij}} V_i n_j \, ds + \int\limits_{\Sigma S_p}\int \prod_{ij} (V_i - V_i^0) n_j \, ds. \tag{15}$$

The second integral vanishes when the inertia effects and body force are absent since $\partial_j \prod_{ij}^0 = 0$. Therefore, the energy dissipation rate can be expressed in the form as

$$\Phi = \int\limits_{S_0}\int \overset{0}{\prod_{ij}} V_i^0 n_j \, ds + \int\limits_{\Sigma S_p}\int \prod_{ij} (V_i - V_i^0) n_j \, ds. \tag{16}$$

Substituting Eqs. (9), (10) and (12) into Eq. (16) and assuming that the pressure at the boundary of the element is zero, one can derive

$$\Phi = \int\limits_{S_0}\int \overset{0}{\prod_{ij}} V_i^0 n_j \, ds + \int\limits_{\Sigma S_p}\int \prod_{ij} (V_i - V_i^0) n_j \, ds$$

$$= 2\mu \gamma_{ij}^0 \gamma_{ij}^0 + N D_{ik} \gamma_{ik}^0 - N D_{ik} \varepsilon_{ijk} \omega_j, \tag{17}$$

where ε_{ijk} is the permutation symbol, having the following properties: it is zero if any two of the three indices are equal; it has the value $+1$ if (i, j, k) is an even cyclic permutation of the integers (1, 2, 3); it has the value -1 if (i, j, k) is an odd cyclic permutation of the intergers (1, 2, 3). And

$$D_{ik} = -\int\limits_{S_p}\int \prod_{ij} n_j x_k \, ds. \tag{18}$$

In deriving Eq. (17), we used $\prod_{ij}^0 = 2\mu\gamma_{ij}^0, V_i^0 = \gamma_{ij}^0 x_j$ on the surface of the suspension element. The angular velocity of the aggregate can be expressed in the following form:

$$\vec{\omega} = -\sin\varphi\dot{\theta}\,\vec{i} + \cos\varphi\dot{\theta}\,\vec{j} + \dot{\varphi}\,\vec{k}\,. \tag{19}$$

The tensor D_{ij} created by the linear ambient flow field γ_{ij}^0 and the rotational movement of the aggregate ω_i is derived in Appendix I, taking the form:

$$D_{ij} = M_{ijkl}\gamma_{kl}^0 + H_{ijk}\omega_k, \tag{20}$$

where the tensors M_{ijkl}, H_{ijk}, depending only on the orientation and size of the spheroid aggregates, are shown in Appendix I. Substitution of Eq. (20) into Eq. (17) gives

$$\begin{aligned}\Phi &= 2\mu\gamma^0_{ij}\gamma^0_{ij} + ND_{ik}\gamma^0_{ik} - ND_{ik}\varepsilon_{ijk}\omega_j \\ &= 2\mu\gamma^0_{ij}\gamma^0_{ij} + NM_{ij\alpha\beta}\gamma^0_{ij}\gamma^0_{\alpha\beta} + N(H_{\alpha\beta k} - M_{ij\alpha\beta}\varepsilon_{ikj})\gamma^0_{\alpha\beta}\omega_k - NH_{ij\alpha}\varepsilon_{ikj}\omega_k\omega_\alpha. \end{aligned} \tag{21}$$

From Eq. (21), one knows that the dissipation potential is a quadratic form of the rates γ^0_{ij}, ω_k. With the aid of the interaction energy given in Eq. (8) and the dissipation potential given in Eq. (21), we can not only establish the general constitutive relation of the system, but also derive the governing equations related to the microstructural evolution $\dot{\theta}, \dot{\phi}$.

4 Constitutive relation of an ER suspension

4.1 General formulation of the internal variable theory

It is well-known fact that the thermodynamic state of an ER suspension at a given time is not only a function of the instantaneous value of the strain rate γ^0_{ij}, but also depends on the previous history of γ^0_{ij}. The investigation of the thermodynamic state may be dealt with in various manners. One effective method is the ''internal variable theory'' [18], [23]. To completely define a thermodynamic state of a suspension, one needs to introduce some internal variables that describe the microstructural change of the material during loading, besides identifying the instantaneous strain rate. In such way, the dependence of the material response on loading history can be replaced by a dependence on what it has produced. Namely, the current pattern of structural arrangement on the microscale of the material element is represented by the current value of internal variables. When the internal variables are fixed, the response of the material only depends on the instantaneous value of the strain rate γ^0_{ij}. But generally speaking, the values of the internal variables depend on the loading history. The internal variable theory is based on the fundamental principle of thermodynamics [23]. In its framework, one can not only establish the relation between the stress and strain, but also derive the evolution equation of the microstructures. Consider a unit volume element of an ER suspension; its state variables are denoted as the strain tensor η_{ij}, absolute temperature T and a group of internal variables ϑ_k. In other words, the variables η_{ij}, ϑ_k and T can give a complete description of the state of the system. The first fundamental law of thermodynamics can be expressed in the following form:

$$dW = dU - dQ, \tag{22}$$

where U is the internal energy of the system, dW is the elementary work done on the system and dQ is the heat supply to the system.

The second fundamental law of thermodynamics states that there exists a state function $S(\eta_{ij}, \vartheta_k, T)$, called entropy, such that

$$TdS \geq dQ. \tag{23}$$

If (23) holds with the equality sign, the process is referred to as reversible, otherwise as irreversible. The entropy can be written in the following form:

$$dS = d^{(r)}S + d^{(i)}S, \tag{24}$$

where

$$d^{(r)}S = \frac{dQ}{T} \tag{25}$$

is the reversible increment of S, called the entropy supply from outside, whereas

$$d^{(i)}S \geq 0 \tag{26}$$

is the irreversible increment, referred to the entropy production inside the system. The combination of (22), (23) and (24) leads to

$$dW = dU - dQ = dU - Td^{(r)}S = dU - TdS + Td^{(i)}S. \tag{27}$$

If the applied stress field on the material element is denoted as τ_{ij}, the elementary work done on the system can be written as

$$dW = \tau_{ij}d\eta_{ij}. \tag{28}$$

On account of the fact the U and S are state functions, Eq. (28) can be replaced by the relation

$$\tau_{ij}d\eta_{ij} = \left(\frac{\partial U}{\partial \eta_{ij}} - T\frac{\partial S}{\partial \eta_{ij}}\right)d\eta_{ij} + \left(\frac{\partial U}{\partial \vartheta_k} - T\frac{\partial S}{\partial \vartheta_k}\right)d\vartheta_k + \left(\frac{\partial U}{\partial T} - T\frac{\partial S}{\partial T}\right)dT + Td^{(i)}S. \tag{29}$$

For the pure heating case, (29) is reduced to

$$\left(\frac{\partial U}{\partial T} - T\frac{\partial S}{\partial T}\right)dT + Td^{(i)}S = 0. \tag{30}$$

The second term is nonnegative, whereas the quantity inside the parentheses is a state function and hence is independent of dT. Since (30) must hold for both positive and negative values of dT, we have

$$\frac{\partial U}{\partial T} - T\frac{\partial S}{\partial T} = 0. \tag{31}$$

It is noted that the above result is generally valid and independent of the type of process even though we have obtained it by considering a special process. Equation (29) can be simplified if we introduced another state function, the so-called free energy of the system, defined by

$$\Xi = U - TS. \tag{32}$$

Then

$$\tau_{ij}d\eta_{ij} = \frac{\partial \Xi}{\partial \eta_{ij}}d\eta_{ij} + \frac{\partial \Xi}{\partial \vartheta_k}d\vartheta_k + Td^{(i)}S. \tag{33}$$

As mentioned by Ziegler [23], the term $Td^{(i)}S$ has the form of an elementary work, and can be expressed in the following form:

$$Td^{(i)}S = A_{ij}d\eta_{ij} + B_k d\vartheta_k. \tag{34}$$

Substitution of Eq. (34) into Eq. (33) gives

$$\tau_{ij}d\eta_{ij} = \left(\frac{\partial \Xi}{\partial \eta_{ij}} + A_{ij}\right)d\eta_{ij} + \left(\frac{\partial \Xi}{\partial \vartheta_k} + B_k\right)d\vartheta_k. \tag{35}$$

Since η_{ij}, ϑ_k are independent state variables, the above equation implies

$$\tau_{ij} = \frac{\partial \Xi}{\partial \eta_{ij}} + A_{ij}, \tag{36}$$

$$\frac{\partial \Xi}{\partial \vartheta_k} + B_k = 0. \tag{37}$$

In fact, Eq. (36) is the constitutive relation of the material, and Eq. (37) can be used to determine the values of the internal variables.

We now rewrite Eq. (34) by replacing the differentials by time derivatives as follows:

$$\Phi = A_{ij}\gamma^0_{ij} + B_k\dot{\vartheta}_k, \tag{38}$$

where Φ is the dissipation function which is the rate of work done by the dissipative forces. Equation (38) cannot determine the dissipative force, the tensor A_{ij} and vector B_k even if we know the dissipation function. To determine A_{ij} and B_k, we introduce the following orthogonality condition: the dissipative force corresponding to the velocity γ^0_{ij} or $\dot{\vartheta}_k$ is orthogonal to the dissipation surface $\Phi = \Phi_0$ in the end point. Therefore, one can obtain

$$\begin{aligned} A_{ij} &= \lambda_1 \frac{\partial \Phi}{\partial \gamma^0_{ij}}, \\ B_k &= \lambda_2 \frac{\partial \Phi}{\partial \vartheta_k}, \end{aligned} \tag{39}$$

where λ_1, λ_2 are proportional factors determined on account of (38) by

$$\begin{aligned} \lambda_1 &= \left(\frac{\partial \Phi}{\partial \gamma^0_{ij}}\gamma^0_{ij}\right)^{-1}\Phi, \\ \lambda_2 &= \left(\frac{\partial \Phi}{\partial \vartheta_k}\vartheta_k\right)^{-1}\Phi. \end{aligned} \tag{40}$$

Since we have derived that Φ is a quadratic function of the velocities, Eq. (40) yields

$$\lambda_1 = \lambda_2 = \frac{1}{2}. \tag{41}$$

As the strain and strain rate are symmetric tensors, substitution of Eqs. (39) and (41) into Eqs. (36) and (37) yields

$$\tau_{ij} = \frac{1}{2}\left(\frac{\partial \Xi}{\partial \eta_{ij}} + \frac{\partial \Xi}{\partial \eta_{ji}}\right) + \frac{1}{4}\left(\frac{\partial \Phi}{\partial \gamma^0_{ij}} + \frac{\partial \Phi}{\partial \gamma^0_{ji}}\right), \tag{42}$$

$$\frac{\partial \Xi}{\partial \vartheta_k} + \frac{1}{2}\frac{\partial \Phi}{\partial \dot{\vartheta}_k} = 0. \tag{43}$$

It is worth to note that as discussed by Ziegler [23] the orthogonality condition is equivalent to the principle of maximal dissipation rate.

4.2 Constitutive equation

4.2.1 The constitutive equation of an ER suspension before yielding

Experimental data reveal that when the applied shear stress is smaller than the yielding strength of an ER suspension, the ER suspension behaves like an ordinary solid material, its strain increases almost linearly with the applied stress. An identifying characteristic of ER suspensions under such a static condition is that upon application of an electric field the particles align into a chain-like structure along the direction of the field. Under the action of an applied shear loading, the fibril aggregates of the particles will keep intact, but they will rotate slightly as Shown in Fig. 1. In this stage, we further assume that there is no slipping between the electrodes

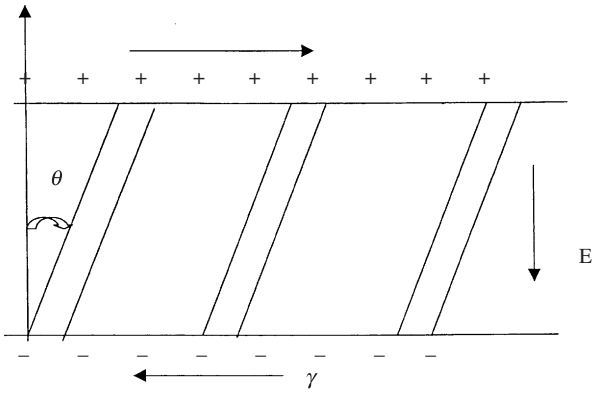

Fig. 1. Schematic of ER structure at the initial stage

and the induced ER structures. Under such conditions, the tilt angle of the droplets is directly related with the applied strain. If the applied shear strain is $\eta_{13} = \eta_{31}$, one can find

$$2\eta_{13} = \eta_{13} + \eta_{31} = tg\theta \approx \theta. \tag{44}$$

By substituting it into Eq. (8), the interaction energy of the system is given as follows:

$$\Psi = -\frac{\phi}{2}(\alpha^p - \alpha^f)\alpha_0(E^0)^2\Big\{A_x(1-\phi+\phi A_x)^{-2} - [A_x(1-\phi+\phi A_x)^{-2} - A_y(1-\phi+\phi A_y)^{-2}](\eta_{13}+\eta_{31})^2\Big\}. \tag{45}$$

For an isothermal process, one can write

$$\frac{\partial \Xi}{\partial \eta_{ij}} = \frac{\partial \Psi}{\partial \eta_{ij}}. \tag{46}$$

The shear stress can be derived by using Eq. (42) and neglecting the dissipation terms as follows:

$$\tau_{13} = 2G^*\eta_{13} = \frac{1}{2}\left(\frac{\partial \Psi}{\partial \eta_{13}} + \frac{\partial \Psi}{\partial \eta_{31}}\right) = 2\phi\alpha_0(\alpha^p - \alpha^f)(E^0)^2[A_x(1-\phi+\phi A_x)^{-2} - A_y(1-\phi+\phi A_y)^{-2}]\eta_{13}, \tag{47}$$

where the effective shear modulus is given by

$$G^* = \phi\alpha_0(\alpha^p - \alpha^f)(E^0)^2[A_x(1-\phi+\phi A_x)^{-2} - A_y(1-\phi+\phi A_y)^{-2}]. \tag{48.1}$$

If the dielectric permittivity of the particles does not approach infinity, i.e., they are not conductors, the fibril aggregates can be assumed to be cylindrical dielectrics, the depolarizing factors are $n_x = 0, n_y = n_z = \frac{1}{2}$. Therefore, the effective shear modulus is given by

$$G^* = \phi\alpha_0(\alpha^p - \alpha^f)(E^0)^2\left[1 - \frac{2\alpha^f}{\alpha^p + \alpha^f}(1 - \phi\frac{\alpha^p - \alpha^f}{\alpha^p + \alpha^f})^{-2}\right]. \tag{48.2}$$

If the dielectric permittivity of the particles does approach infinity, the aggregates cannot be assumed to be infinitely long cylinders as assumed in deriving Eq. (48.2), in such case, Eq. (48.1) should also give reasonable results.

From Eq. (47), it is very clear that at the initial stage the ER suspensions behave as an ordinary elastic material with the shear modulus given by Eq. (48). However, it should be mentioned that this result is based on the assumption that the tilt angle is very small, as given by Eq. (44), and the droplets will not slip on the electrodes. When the applied shear strain

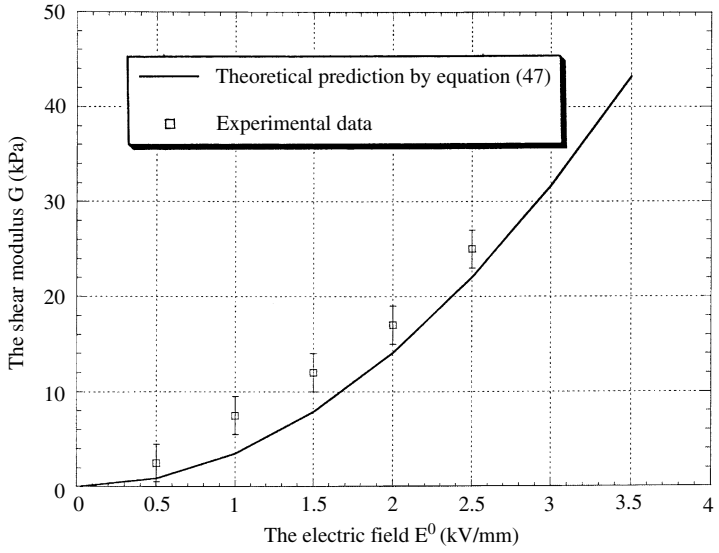

Fig. 2. The shear modulus versus the applied electric field

reaches its critical value η_{13}^c, the droplets cannot keep intact. The corresponding shear stress given by Eq. (47) is the static yielding stress of the ER system.

The experimental data obtained by Ginder and Davis [18] are used to verify our theoretical prediction. The model fluids utilized in their study were composed of barium titanate particles having the relative permittivity $\alpha^p = 2000$, suspended at volume fractions $\phi = 0.2$ in dodecane having the relative permittivity $\alpha^f = 2$. The permittivity of free space is $\alpha_0 = 8.85417 \times 10^{-12} F/m$. The shear modulus versus the applied electric field is shown in Fig. 2. The predicted shear modulus is slightly lower than the experimental data in the barium titanate system. This departure may be due to the strong image effect of the electrodes.

4.2.2 The constitutive relation after yielding

In the dynamic regime after yielding, the behavior of an ER suspension is often approximated by a Bingham solid, i.e.,

$$\tau = \tau^y + \mu^* \gamma^0, \tag{49}$$

where τ^y is the yielding stress of the system, and μ^* is the viscosity of the suspension. Experimental data revealed that most ER suspensions showed a shear-thinning viscocity, i.e. the viscosity of the suspension decreases with increasing shear rate. Klingenberg and Zukoski [14] suggested that this shear-thinning behavior was due to the formation of condensed boundary layers near the electrodes, so that the velocity gradients appeared only on a portion of the sample. Halsey et al. [9], [10] assumed that the size and orientation of the aggregates would adjust with the flow field, and predicted that the shear-thinning effect was due to the bulk properties of the fluid. Shulman et al. [20] considered the similar problem for magnetorheological suspensions.

In this paper, the size parameters, a and c, and the orientation of the aggregates, θ and φ, are denoted as the internal variables of the system. When they are fixed, the material becomes an ordinary suspension. That means, if the internal variables are given at an instant time, the response of the system depends only on the instantaneous value of the strain rate, not on its history. In fact, the effect of the loading history has been considered by the values of the

internal variables. Furthermore we do not consider the existence of currents or of interfacial polarization due to a nonzero conductivity of the solid or of the liquid phase. These conditions apply principally to the category of ER fluids based on a large electronic polarizability of the constituent particles and acted on by electric fields whose frequency is high enough to neglect ionic polarization and charge accumulation on the electrodes. Substituting Eq. (21) into Eq. (42), we have

$$\begin{aligned}\tau_{mn} &= \tau^{y}_{mn} + \frac{1}{2}\left(\frac{\partial \Psi}{\partial \eta_{mn}} + \frac{\partial \Psi}{\partial \eta_{nm}}\right) + \frac{1}{4}\left(\frac{\partial \Phi}{\partial \gamma^{0}_{mn}} + \frac{\partial \Phi}{\partial \gamma^{0}_{nm}}\right) \\ &= \tau^{y}_{mn} + 2\mu\gamma^{0}_{mn} + \frac{1}{4}N(M_{mn\alpha\beta} + M_{nm\alpha\beta} + M_{\alpha\beta mn} + M_{\alpha\beta nm})\gamma^{0}_{\alpha\beta} \\ &\quad + \frac{1}{4}N[H_{mnk} + H_{nmk} - (M_{ijmn} + M_{ijnm})\varepsilon_{ikj}]\omega_{k}, \end{aligned} \tag{50}$$

where τ^{y}_{mn} is the static yielding stress. In deriving Eq. (50), one should notice that the interaction energy is independent of the applied strain in the dynamic regime. Once the suspension reaches the steady state where the size and orientation of the aggregates do not vary with time any more, the last term in Eq. (50) becomes zero, therefore,

$$\begin{aligned}\tau_{mn} &= \tau^{y}_{mn} + 2\mu\gamma^{0}_{mn} + \frac{1}{4}N(M_{mn\alpha\beta} + M_{nm\alpha\beta} + M_{\alpha\beta mn} + M_{\alpha\beta nm})\gamma^{0}_{\alpha\beta} \\ &= \tau^{y}_{mn} + 2\mu\gamma^{0}_{mn} + \phi\bar{M}_{mn\alpha\beta}\gamma^{0}_{\alpha\beta}, \end{aligned} \tag{51}$$

where $\frac{16}{3}\pi a c^{2}\bar{M}_{mn\alpha\beta} = M_{mn\alpha\beta} + M_{nm\alpha\beta} + M_{\alpha\beta mn} + M_{\alpha\beta nm}$, ϕ is the volume fraction of the particles.

4.2.3 Determination of the internal variables θ, φ, a, c

Since the tensors $M_{ijkl}, H_{ij\alpha}$ are functions of the size and orientation of aggregates, one needs first to determine how these internal variables change with the external condition. By using Eq. (43), the evolution equations for $\dot{\theta}, \dot{\phi}$ are derived as

$$\frac{\partial \Psi}{\partial \theta} = -\frac{1}{2}\frac{\partial \Phi}{\partial \dot{\theta}}, \tag{52}$$

$$\frac{\partial \Psi}{\partial \varphi} = -\frac{1}{2}\frac{\partial \Phi}{\partial \dot{\varphi}}. \tag{53}$$

Equations (52) and (53) jointly give the first-order differential equations system which can be used to determine $\theta(t), \varphi(t)$ under the given initial condition, the applied strain rate γ^{0}_{ij} and the electric field. By using Eq. (8), we can have

$$\begin{aligned}&\frac{\partial \Psi}{\partial \theta} = -\frac{\phi}{2}(\alpha^{p} - \alpha^{f})\alpha_{0}(E^{0})^{2}\sin 2\theta[A_{y}(1 - \phi + \phi A_{y})^{-2} - A_{x}(1 - \phi + \phi A_{x})^{-2}], \\ &\frac{\partial \Psi}{\partial \varphi} = 0. \end{aligned} \tag{54}$$

And using Eq. (21) and Eq. (19), one can obtain

$$\begin{aligned}\frac{\partial \Phi}{\partial \dot{\theta}} &= \Lambda^{(\theta)}_{\alpha\beta}\gamma^{0}_{\alpha\beta} + \Delta^{(\theta)}_{k}\omega_{k}, \\ \frac{\partial \Phi}{\partial \dot{\varphi}} &= \Lambda^{(\varphi)}_{\alpha\beta}\gamma^{0}_{\alpha\beta} + \Delta^{(\varphi)}_{k}\omega_{k}, \end{aligned} \tag{55}$$

where

$$\begin{aligned}
\Lambda_{\alpha\beta}^{(\theta)} &= N\cos\varphi(H_{\alpha\beta 2} - M_{ij\alpha\beta}\varepsilon_{i2j}) - N\sin\varphi(H_{\alpha\beta 1} - M_{ij\alpha\beta}\varepsilon_{i1j}),\\
\Lambda_{\alpha\beta}^{(\varphi)} &= N(H_{\alpha\beta 3} - M_{ij\alpha\beta}\varepsilon_{i3j}),\\
\Delta_{k}^{(\theta)} &= N\sin\varphi(H_{ij1}\varepsilon_{ikj} + H_{ijk}\varepsilon_{i1j}) - N\cos\varphi(H_{ij2}\varepsilon_{ikj} + H_{ijk}\varepsilon_{i2j}),\\
\Delta_{k}^{(\varphi)} &= -N(H_{ij3}\varepsilon_{ijk} + H_{ijk}\varepsilon_{i3j}).
\end{aligned} \tag{56}$$

Substituting Eqs. (54) and (55) into Eqs. (52) and (53), the first-order differential equations for θ, φ are thus established as

$$\begin{aligned}
(\Delta_1^{(\varphi)}\sin\varphi - \Delta_2^{(\varphi)}\cos\varphi)\dot{\theta} - \Delta_3^{(\varphi)}\dot{\boldsymbol{\varphi}} - \Lambda_{\alpha\beta}^{(\varphi)}\gamma_{\alpha\beta}^{0} &= 2\frac{\partial\Psi}{\partial\varphi},\\
(\Delta_1^{(\theta)}\sin\varphi - \Delta_2^{(\theta)}\cos\varphi)\dot{\theta} - \Delta_3^{(\theta)}\dot{\boldsymbol{\varphi}} - \Lambda_{\alpha\beta}^{(\theta)}\gamma_{\alpha\beta}^{0} &= 2\frac{\partial\Psi}{\partial\theta}.
\end{aligned} \tag{57}$$

As we discussed before, one needs to solve the differential equation (57) for a given loading history to determine the value of the internal variables.

To derive the equilibrium size of the spheroidal aggregate, Halsey [9] divided the polarization energy into two parts, the first is the depolarization energy as given by Eq. (8), and the second part is the surface energy of the droplet. Balancing these two effects, he obtained the dependence of the size on the Mason number. As mentioned by Halsey [10], the surface tension is a somewhat subtle effect, it also arises from the dipolar interactions as the bulk depolarization energy. The surface energy is specially a dipolar lattice effect. In our formulation, by considering that every particle attached to the aggregates should keep in equilibrium under the action of the hydrodynamic force and electrostatic force, it seems more appropriate to establish the force equilibrium equation to determine the length of the aggregate. Consider a spheroidal aggregate in the ER suspension, on the tip of it, a spherical particle exists as shown in Fig. 3. When the system reaches its steady state, all the forces on particles in the aggregates should keep in equilibrium, i.e., the electrostatic force and the hydrodynamic force acting on the particle should be balanced with each other. For a small spherical particle, we can assume the local electric field $\vec{E}^{\,e}$ acting on it is uniform. Therefore, the electric dipole moment of the dielectric particle can be determined by using Eq. (4) with $n_x = n_y = n_z = 1/3$ for a spherical particle as follows:

$$\vec{P} = P_s\vec{E}^{\,e}, \tag{58}$$

where

$$P_s = \frac{4\pi\alpha^f(\alpha^p - \alpha^f)}{\alpha^p + 2\alpha^f}\alpha_0 r_0^3 \tag{59}$$

and r_0 is the radius of the particle. The electrostatic force on the particle can be obtained by calculating the external force on the electric dipole sitting at the center of the particle as follows:

$$\vec{F}^{\,e}(\alpha + r_0, 0, 0) = \vec{\nabla}(\vec{P}\cdot\vec{E}^{\,e}) = P_s\vec{\nabla}\left[(E_x^e)^2 + (E_y^e)^2 + (E_z^e)^2\right]\big|_{X=a+r_0, Y=Z=0}, \tag{60}$$

in which the local electric field outside the spheroidal dielectrics is given in Appendix II.

To determine the hydrodynamic force acting on the particle, we need to derive the local fluid velocity $\vec{V}^{\,L}$ around the particle, which is given in Appendix III. The hydrodynamic force on the particle is given by the Stokes resistance

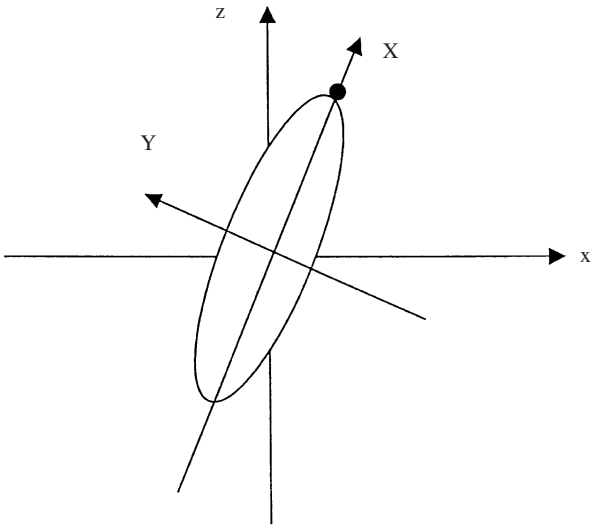

Fig. 3. Schematic of the interactions between the spheroidal droplet and the particle

$$\vec{F}^{h}(a+r_0,0,0) = 6\pi\mu r_0 \vec{V}^{L}(a+r_0,0,0). \tag{61}$$

Generally speaking, the length of the aggregates is very sensitive to the applied electric field and strain rate. Whereas their perpendicular size is comparatively stable. Therefore we can fix the size parameter c by balancing the force along the symmetric axis X to determine the length:

$$F_X^e(a+r_0,0,0) + F_X^h(a+r_0,0,0) = 0. \tag{62}$$

In fact, to obtain the constitutive relation given in Eq. (51), we need to know the aspect ratio a/c only. Through the calculation, it is found that the aspect ratio of the aggregates is independent of the selected size c. If one considers that the hydrodynamic forces are transmitted from one particle to the other through lubrication zones between the particles, the maximum hydrodynamic force between the particles occurs at the center of the aggregate.

By solving the simultaneous equations (57) and (62), the internal variables a, c, θ, φ can be obtained as functions of the applied strain rate and the electric field. When the system reaches the steady state, $\dot{\theta}, \dot{\varphi}$ become zero. After solving a, c, θ, φ, and substituting these values into the expression of M_{ijkl}, then with Eq. (51) the nonlinear constitutive relation is thus established.

5 The constitutive equation of an ER suspension under simple shear loading

As an example in a special case, in this section we focus our attention on the simple shear loading condition. The applied strain rate is $\gamma_{13}^0 = \gamma_{31}^0$. Since the symmetry axis is in the x-z-plane, one can write $\varphi = 0$, while the unit directional vector $\vec{d}$ along the symmetry axis is expressed in the form of

$$\vec{d} = \{\sin\theta, 0, \cos\theta\}. \tag{63}$$

By substituting Eq. (63) into the expressions of M_{ijkl}, H_{ijk} in Eqs. (A2) and (A6), then further into Eq. (57), it is found that

$$\Delta_3^{(\varphi)}\dot{\boldsymbol{\varphi}} = 0, \tag{64}$$

$$\Delta_2^{(\theta)}\dot{\theta} = 12\pi\mu\phi Y^H\left(\frac{a}{c}\right)^2\gamma_{13}^0\cos 2\theta + \phi\alpha_0(\alpha^p-\alpha^f)(E^0)^2[A_y(1-\phi+\phi A_y)^{-2}$$
$$-A_x(1-\phi+\phi A_x)^{-2}]\sin 2\theta = G\cos 2\theta + F\sin 2\theta, \tag{65}$$

where

$$F = \phi(\alpha^p-\alpha^f)\alpha_0(E^0)^2[A_y(1-\phi+\phi A_y)^{-2} - A_x(1-\phi+\phi A_x)^{-2}],$$
$$G = 12\pi\mu\phi Y^H\gamma_{13}^0\left(\frac{a}{c}\right)^2, \tag{66}$$
$$\Delta_2^{(\theta)} = 8\pi\phi\mu\left(\frac{a}{c}\right)^2 Y^C,$$

and Y^H, Y^C are determined by Eqs. (A4) and (A7).

Equation (64) means that for such a shear loading the symmetry axis of the aggregate rotates only in the plane xoz if initially $\varphi = 0$. Equation (65) gives the solution of the rotational angle as a function of time t for a given electric field E^0 and shear strain rate field γ_{13}^0 as follows:

$$tg\theta = \frac{1}{G}[F + \sqrt{G^2+F^2}\tanh(\chi)],$$
$$\chi = \frac{\sqrt{G^2+F^2}}{\Delta_2^{(\theta)}}t + \operatorname{arctanh}\frac{-F}{\sqrt{G^2+F^2}}. \tag{67}$$

When time t approaches infinity, $\chi \to \infty, \tanh(\chi) \to 1$. Therefore the tilt angle reaches the steady value for the given condition, which can be determined by setting $\dot{\theta} = 0$, or

$$tg2\theta_{eq} = -\frac{G}{F}. \tag{68}$$

The rotational angle as a function of non-dimensional time $t^* = t\gamma_{13}$ for different Mason number M_n which gives the ratio of hydrodynamic force to polarization force $M_n = \mu\gamma_{13}/[\alpha_0(\alpha^p-\alpha^f)(E^0)^2]$, is shown in Fig. 4.

By substituting Eq. (68) into Eq. (62), and solving Eq. (62) numerically, one can obtain the equilibrium length a of the aggregate for a given value of c. As said before, the obtained aspect ration a/c is independent of the value of c. The result of a/c versus the Mason number is depicted in Fig. 5. From the log-log plot, we found that $a/c \propto (Mn)^{-0.48}$. Using the molecular-like dynamics simulation, Takimoto [21] also revealed such a power-law relation, and the apparent exponent was approximately equal to -0.5. Through balancing the depolarization

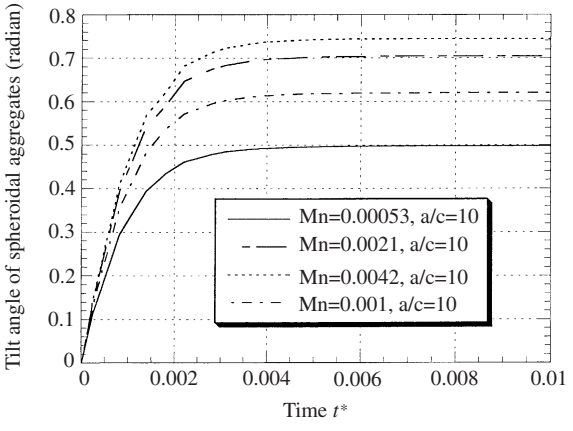

Fig. 4. The rotational angle as a function of nondimensional time $t^* = t\gamma_{13}$ for different Mason numbers Mn

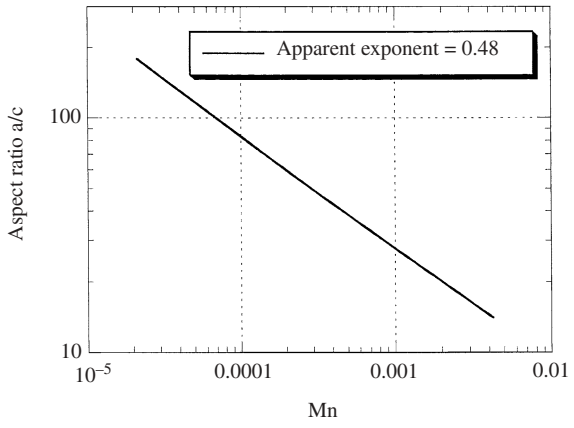

Fig. 5. The aspect ratio a/c versus Mason number in the log-log plot

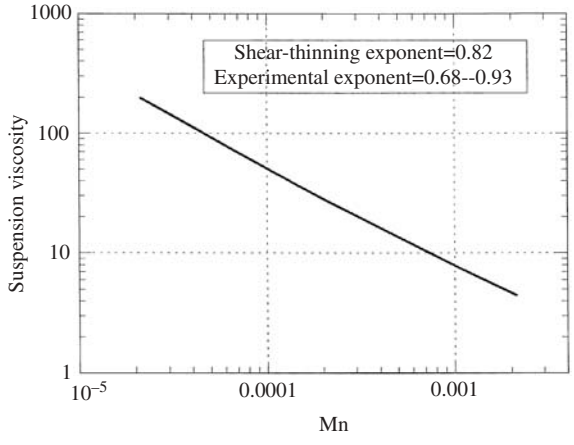

Fig. 6. The nondimensional suspension viscosity $(\mu^* - \mu)/\mu$ versus Mason number in the log-log plot

energy of a spheroidal droplet with its surface energy, Halsey et al. [9], [10] obtained that $a/c \propto Mn^{-\upsilon}$, and the exponent $\upsilon = 1/3$.

By substituting the equilibrium values of the rotational angle and the aspect ratio into the expression of M_{ijkl} in Eq. (A2). then into Eq. (51), we obtain the stress and strain rate relation under simple shear loading condition as

$$\begin{aligned}\tau_{13}^0 &= \tau_{13}^y + 2\mu\gamma_{13}^0 + \frac{5}{3}\pi\mu N\alpha^3\gamma_{13}^0[(3X^M + Z^M)\sin^2 2\theta_{eq} + 4Y^M\cos^2 2\theta_{eq}] \\ &= \tau_{13}^y + 2\mu\gamma_{13}^0 + \frac{5}{4}\frac{\phi\mu\gamma_{13}^0}{G^2 + F^2}\left(\frac{a^2}{c^2}\right)[(3X^M + Z^M)G^2 + 4Y^M F^2] \\ &= \tau_{13}^y + 2\mu^*\gamma_{13}^0, \end{aligned} \tag{69}$$

where the viscosity μ^* of the ER suspension is given by

$$\mu^* = \mu + \frac{5}{8}\frac{\phi\mu}{G^2 + F^2}\left(\frac{a^2}{c^2}\right)[(3X^M + Z^M)G^2 + 4Y^M F^2], \tag{70}$$

where X^M, Y^M, Z^M are dependent only on the aspect ratio, and are given by Eq. (A4). As shown in Eq. (66), the functions G and F and the aspect ration a/c depend on the strain rate γ_{13}^0 and the applied electric field $(E^0)^2$. Therefore Eq. (70) predicts a shear-thinning viscosity of the ER

suspension. By substituting the obtained aspect ratio into Eq. (70), one can find the variation of the reduced suspension viscosity $(\mu^* - \mu)/\mu$ with the Mason number Mn. The relationship is plotted in log-log scale in Fig. 6. From the Figure, it can be found that the suspension viscosity can be well approximated by a power function $\propto (Mn)^{-\Delta}$ with the shear-thinning exponent of $\Delta \approx 0.82$. Rheological measurement by Halsey et al. [9] on a model fluid consisting of monodisperse silica spheres immersed in a dielectric liquid showed a power-law dependence $\mu^* \propto (Mn)^{-\Delta}$ of the apparent viscosity on the Mason number with $\Delta = 0.68 - 0.93$.

6 Concluding remarks

In this paper, a microstructural constitutive theory of ER suspensions was formulated. The framework was based on the internal variable theory and the mechanism analysis. The ER suspension consists of fine particles with high dielectric constant and the supporting fluid. Under the action of the electric field, the polarized particles will aggregate together to form the chain-like structures along the direction of the electric field. The size and orientation of the particle aggregates are volatile. They will adjust according to the applied electric field and strain rate. Therefore, a model was established to determine the size and orientation of the aggregates. Then a three-dimensional, explicit form of the constitutive equation was derived based on the interaction energy and the dissipation function of the system. The response of the system under the action of a simple shearing load was considered and discussed in detail. It is found that the shear-thinning viscosity of an ER suspension can be well approximated by the power-law $\propto (Mn)^{-0.82}$. Since the evolution equation of the aggregate orientation is a loading history dependent differential equation, after solving it for a given loading history, one can predict the constitutive behavior of the ER suspension for the loading history.

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Appendix I

The derivation of the tensor D

The stress field on the surface of the aggregate is created by the linear ambient flow field γ_{ij}^0, and rotational movement of the aggregate ω_i. For a steady creeping flow considered in this paper, the stress tensor can be obtained by summing up the two microhydrodynamic solutions. We can express the tensor D_{ij} as follows:

(i) The force dipole D_{ij}^1 for linear ambient flow

The force dipole D_{ij}^1 is defined as follows:

$$D_{ij}^{1} = -\int_{S_p}\int \prod_{ik}^{1} n_k x_j \, ds, \tag{A.1}$$

where $\prod_{ik}^{1}$ is the stress field on the surface of the aggregation created by linear ambient flow. For ellipsoidal inclusion, the three fundamental problems – translation, rotation, and linear ambient field were solved in the paper of Oberbeck, Edwardes, and Jeffery. For a prolate spheroid aggregate ($a > b = c$), denoting the unit directional vector along the symmetry axis by $\underline{d}$, D_{ij}^{1} can be expressed in the form [13]:

$$\begin{aligned} D_{ij}^{1} &= M_{ijkl}\gamma_{kl}^{0} \\ &= \left\{4\pi\mu a^3 Y^H \varepsilon_{ijm}\varepsilon_{mk\beta}d_\beta d_l + \frac{20}{3}\pi\mu a^3[X^M d_{ijkl}^{(0)} + Y^M d_{ijkl}^{(1)} + Z^M d_{ijkl}^{(2)}]\right\}\gamma_{kl}^{0}, \end{aligned} \tag{A.2}$$

where

$$\begin{aligned} d_{ijkl}^{(0)} &= \frac{3}{2}\left(d_i d_j - \frac{1}{3}\delta_{ij}\right)\left(d_k d_l - \frac{1}{3}\delta_{kl}\right), \\ d_{ijkl}^{(1)} &= \frac{1}{2}(d_i\delta_{jl}d_k + d_j\delta_{il}d_k + d_i\delta_{jk}d_l + d_j\delta_{ik}d_l - 4d_i d_j d_k d_l), \\ d_{ijkl}^{(2)} &= \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il} - \delta_{ij}\delta_{kl} + d_i d_j\delta_{kl} + d_k d_l\delta_{ij} - d_i d_k\delta_{jl} - d_j d_k\delta_{il} \\ &\quad - d_i d_l\delta_{jk} - d_j d_l\delta_{ik} + d_i d_j d_k d_l, \end{aligned} \tag{A.3}$$

$$\begin{aligned} Y^H &= \frac{4}{3}e^5[(1+e^2)L - 2e]^{-1}, \\ X^M &= \frac{8}{15}e^5[(3-e^2)L - 6e]^{-1}, \\ Y^M &= \frac{4}{5}e^5[2e(1-2e^2) - (1-e^2)L]\{[2e(2e^2-3) + 3(1-e^2)L][(1+e^2)L - 2e]\}^{-1}, \\ Z^M &= \frac{16}{5}e^5(1-e^2)[3(1-e^2)^2 L - 2e(3-5e^2)]^{-1}, \\ L &= \ln\left(\frac{1+e}{1-e}\right), \end{aligned} \tag{A.4}$$

in which $e = (a^2 - c^2)^{1/2}/a$ is the eccentricity of the generating ellipse.

(ii) The force dipole $D_{ij}^{(2)}$ for rotational motion of the spheroid aggregate

$D_{ij}^{(2)}$ is defined as

$$D_{ij}^{(2)} = -\int_{s_p}\int \prod_{ik}^{(2)} n_k x_j \, ds, \tag{A.5}$$

where $\prod_{ik}^{(2)}$ is the stress field created by the rotational motion of the aggregate. Using the solution for spheroidal inclusions, one can express the tensor $D_{ij}^{(2)}$ in the following form [13]:

$$\begin{aligned} D_{ij}^{(2)} &= H_{ijk}\omega_k \\ &= \{4\pi\mu a^3\varepsilon_{ij\alpha}[X^C d_\alpha d_k + Y^C(\delta_{k\alpha} - d_k d_\alpha)] - 4\pi\mu a^3 Y^H(\varepsilon_{ikl}d_j + \varepsilon_{jkl}d_i)d_l\}\omega_k, \end{aligned} \tag{A.6}$$

where

$$X^C = \frac{4}{3}e^3(1-e^2)[2e-(1-e^2)L]^{-1},$$
$$Y^C = \frac{4}{3}e^3(1-e^2)[(1+e^2)L-2e]^{-1}. \tag{A.7}$$

Appendix II

The electric field outside a dielectric spheroid under a uniform external field

In the book of Laudau et al. [15], the field potential outside an uncharged conducting spheroid was expressed in explicit form. Following the similar procedure, we can derive the electric field potential outside a dielectric spheroid under a uniform external field in explicit form as follows. In the local coordinate system (XYX) with the X-axis along the symmetry axis of the spheroid, the external electric field is denoted as $\{E_X^0, E_Y^0, E_Z^0\}$. The electric field potential outside the spheroid can be expressed in the following form:

$$\phi^e = \phi_1 + \phi_2 + \phi_3, \tag{A.8}$$

where

$$\phi_1 = -E_X^0 X\left\{1+\left(\frac{E_X^i}{E_X^0}-1\right)\cdot\frac{\tanh^{-1}\sqrt{(a^2-c^2)/(a^2+\xi)}-\sqrt{(a^2-c^2)/(a^2+\xi)}}{\tanh^{-1}\sqrt{1-c^2/a^2}-\sqrt{1-c^2/a^2}}\right\},$$
$$\phi_2 = -E_Y^0 Y\left\{1+\left(\frac{E_Y^i}{E_Y^0}-1\right)\cdot\frac{\sqrt{(a^2+\xi)}/(c^2+\xi)-(a^2-c^2)^{-1/2}\tanh^{-1}\sqrt{(a^2-c^2)/(a^2+\xi)}}{a/c^2-(a^2-c^2)^{-1/2}\tanh^{-1}\sqrt{1-c^2/a^2}}\right\},$$
$$\phi_3 = -E_Z^0 Z\left\{1+\left(\frac{E_Z^i}{E_Z^0}-1\right)\cdot\frac{\sqrt{(a^2+\xi)}/(c^2+\xi)-(a^2-c^2)^{-1/2}\tanh^{-1}\sqrt{(a^2-c^2)/(a^2+\xi)}}{a/c^2-(a^2-c^2)^{-1/2}\tanh^{-1}\sqrt{1-c^2/a^2}}\right\}, \tag{A.9}$$

where the internal electric field inside the spheroid is given by Eqs. (1) and (3), and the coordinate ξ is related to X, Y and Z by

$$\frac{Y^2+Z^2}{c^2+\xi}+\frac{X^2}{a^2+\xi}=1, \tag{A.10}$$

with $0 \le \xi \le \infty$ in the space outside the spheroid. The electric field outside the dielectric spheroid can be obtained by

$$\vec{E}^{\,e} = -\vec{\nabla}\,\phi^e. \tag{A.11}$$

Appendix III

The velocity field solution for a spheroid in the linear ambient field $v_i^0 = \gamma_{ij}^0 X_j$

When the spheroidal aggregate reaches its equilibrium state, it will be fixed in the fluid. Under the action of the linear ambient flow field, the velocity field will change greatly due to the existence of the fixed spheroid. Jeffery obtained the solution for an ellipsoid long time ago,

which was shown in the book by Kim and Karrila [13]. For a spheroid, we can reproduce his solution to the following explicit-form expression:

$$V_i^L = \gamma_{ij}^0 X_j - \frac{3}{32\pi\mu}\left(S_{jk} + \frac{1}{2}\varepsilon_{jlk}T_l\right)\frac{\partial}{\partial X_k}\left[G_1\delta_{ij} - X_j\frac{\partial G_1}{\partial X_i} + \frac{a_J^2}{4}\frac{\partial^2 G_2}{\partial X_i \partial X_j}\right], \tag{A.12}$$

where

$$G_n = \int_\xi^\infty \left(\frac{X^2}{a^2+\lambda} + \frac{Y^2}{c^2+\lambda} + \frac{Z^2}{c^2+\lambda} - 1\right)^n \frac{d\lambda}{\Delta(\lambda)}, \tag{A.13}$$

with $\Delta(\lambda) = \sqrt{(a^2+\lambda)}(c^2+\lambda)$ and the ellipsoidal coordinate ξ defined as the positive root of

$$\frac{X^2}{a^2+\xi} + \frac{Y^2+Z^2}{c^2+\xi} = 1. \tag{A.14}$$

T_i, S_{ij} are the torque and stresslet on the spheroid, which are given by

$$\begin{aligned} T_i &= -8\pi\mu a^3 Y^H \varepsilon_{ijl} d_k d_l \gamma_{jk}^0, \\ S_{ij} &= \frac{20}{3}\pi\mu a^3 [X^m d_{ijkl}^0 + Y^m d_{ijkl}^1 + Z^m d_{ijkl}^2]\gamma_{kl}^0. \end{aligned} \tag{A.15}$$

The definition of the symbols in Eq. (A.15) is the same as that in Appendix I. Since the symmetry axis is connected with the X-axis, $d_1 = 1, d_2 = d_3 = 0$.

For a spheroidal aggregate, Eq. (A.12) can be reduced to the explicit form:

$$\begin{aligned} G_1 &= X^2 I_1 + (Y^2+Z^2)I_2 - I, \\ \frac{\partial G_1}{\partial X_i} &= 2X_i I_I, \\ \frac{\partial G^2}{\partial X_i \partial X_j} &= 8X_i X_j I_{IJ} + 4\delta_{ij}[X^2 I_{1I} + (Y^2+Z^2)I_{2I} - I_I], \end{aligned} \tag{A.16}$$

where the following summation convention has been used: repeated lower case indices are summed up from 1 to 3; upper case indices take on the same number as the corresponding lower case ones but not summed. And

$$I = \int_\xi^\infty \frac{d\lambda}{\Delta(\lambda)} = \frac{2}{\sqrt{a^2-c^2}}\text{arccosh}\bar{b},$$

$$I_1 = \int_\xi^\infty \frac{1}{a^2+\lambda}\frac{d\lambda}{\Delta(\lambda)} = 2(\text{arccosh}\bar{b} - \bar{d}/\bar{b})/(a^2-c^2)^{3/2},$$

$$I_2 = I_3 = \int_\xi^\infty \frac{1}{c^2+\lambda}\frac{d\lambda}{\Delta(\lambda)} = (\bar{b}\bar{d} - \text{arccosh}\bar{b})/(a^2-c^2)^{3/2},$$

$$I_{11} = \int_\xi^\infty \frac{d\lambda}{(a^2+\lambda)^2\Delta(\lambda)} = \frac{2}{3}(a^2-c^2)^{-1}\left[\frac{3}{2}I_1 - \frac{1}{(a^2+\xi)^{3/2}}\right],$$

$$I_{12} = I_{13} = I_{21} = I_{31} = \int_{\xi}^{\infty} \frac{d\lambda}{(a^2+\lambda)(c^2+\lambda)\Delta(\lambda)} = \frac{2}{a^2-c^2}\left[\frac{3}{2}I_2 - \frac{1}{(a^2+\xi)^{1/2}(c^2+\xi)}\right],$$
$$I_{22} = I_{33} = I_{23} = I_{32} = \int_{\xi}^{\infty} \frac{d\lambda}{(c^2+\lambda)^2\Delta(\lambda)} = -\frac{1}{2}\frac{1}{a^2-c^2}\left[\frac{3}{2}I_2 - \frac{1}{(a^2+\xi)^{-1/2}(c^2+\xi)^2}\right], \tag{A.17}$$

where $\bar{b} = \sqrt{(a^2+\xi)/(c^2+\xi)}$ and $\bar{d} = \sqrt{(a^2-c^2)/(c^2+\xi)}$. Thus for any linear ambient field, one can express the velocity field around the spheroid in explicit form.

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Author's address: B. Wang, Electro-Optics Technology Center and Research Center for Composite Materials, Harbin Institute of Technology, Harbin, P. R. China (E-mail: mmwangb@polyu.edu.hk); Z. Xiao, School of Mechanical and Production Engineering, Nanyang Technological University, Nanyang Avenue, Singapore