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A THEORETICAL MODELLING OF THE CHAIN STRUCTURE FORMATION IN ELECTRORHEOLOGICAL FLUIDS*

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Abstract: *A model was developed to understand the aggregation process of the particles in electrorheological (ER) fluids under the action of an applied electric field. By establishing a generalized virtual work principle based on the consideration that the released electromagnetic energy accompanying the growth of the chain should equal to the dissipated energy related with friction resistance of the viscous fluid in the chain formation, the governing differential equation of the chain growth was established. Based on this energy model, the velocity of the chain forming, and the response time of ER fluid can be predicted. The present model can also predict the effect of the temperature and some microstructural parameters, such as the dielectric constants and concentration of the particles, etc., on the response of an ER system.*

Key words: electrortheological fluid; response time; dynamic model

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Introduction

An electrorheological (ER) fluid consists of a suspension of fine particles in a liquid of low dielectric constant and low viscosity. Its apparent viscosity increases dramatically in the presence of an applied electric field. If the electric field exceeds a critical value, the ER fluid turns into a solid whose yield stress increases as the field is further strengthened. This phenomenon is completely reversible. Upon electric field cutoff, the system almost immediately resumes its original liquid state. The time scale for the transition is of the order of millisecond. The phenomenon of electrorheology was first discovered by Winslow (1949)[1], and is sometimes termed as the "Winslow effect." Because of their fast response and low power requirements, ER fluids provide the possibility of rapid-response coupling between mechanical devices and electronic control. These properties also make ER fluid attractive for many futuristic technologies. To predict their response, a crucial problem is to gain a clear understanding of the physical mechanism of the ER phenomenon. Consider a system of small particles of dielectric constant ε_p

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suspended in a fluid of dielectric constant ε_f, $\varepsilon_p > \varepsilon_f$. The whole system is placed between two parallel plates, upon which a voltage is applied to produce an electric field. When the dielectric particles have a density close to the liquid density, the buoyant force neutralizes the gravity and creates a low-gravity environment. Before applying the electric field, the thermal motion makes particles randomly distributed in the space and forms a uniform liquid suspension. As the electric field is applied, the particles obtain an induced dipole moment. When the applied electric field increases, the depolarized particles will begin to aggregate one another, and form a chain structure along the electric field. Due to the formation of these chains, the shear strength and viscosity of ER fluid increase by orders. Therefore, the formation of chains underlies all phenomena of ER fluid.

Many researches have been carried out to understand the mechanism of the structure formation. Tao *et al*. (1991)[2] published a series of works about the structure formation in ER fluid under the action of the external field based on phase transition theory from a disordered state to an ordered state. Klingenberg *et al*. (1989)[3] carried out two- and three-dimensional molecular-dynamics-like simulation on the structure formation in electrorheological suspensions. Recently much effort is being devoted to the synthesis of new ER fluids possessing desirable rheological, electrical, chemical, and tribological properties. Receiving particular attention are those materials that exhibit large yield stress and short response time of the field-induced structure change. Detailed descriptions of the research works on the mechanism can be found in the review paper by Parthasarathy and Klingenberg (1996)[4].

To use ER fluid successfully in engineering and develop some high performance materials, it is important to understand the mechanism by which the chain structure is formed. From our knowledge of ER fluid, at present stage, the molecular-dynamics-like simulation is widely used to simulate the motion and aggregation of the particles. But the analysis is restricted by the limitation of the numerical calculation. In this paper, a generalized virtual work principle was established for chain formation process based on thermodynamics analysis. By assuming the forming chain as a prolate spheroid, its aspect ratio was selected as the generalized degree of freedom of the system. Then the governing differential equation of the chain growth was established. Based on this energy model, the velocity of the chain forming, and the response time of ER fluid can be predicted.

1 Generalized Principle of Virtual Work for the Chain Growth Process

As discussed above, under a given temperature, when a strong electric field is applied on the electrodes, the polarized particles will aggregate to form chains along the direction of the applied electric field. With the increases of the electric field, more particles will aggregate on the chain to form thick column. Due to the image effect of the electrodes, the column is usually wide in the two ends, whereas comparatively thin in the middle. But in this paper, we only consider the initial stage of the chain formation without considering the later aggregation and thickening process. Therefore, only one chain is considered here and it will grow only along the direction of applied electric field. To simplify the analysis, the single chain is assumed to take the shape of a prolate spheroid with the semi-axes as a_1 and a_3, where a_1 is the radius of the particles. The kinetics of the chain formation is determined by the change of a_3, or the aspect ratio $\beta = a_3/a_1$,

with respect to time t. We consider the system which consists of all the polarized particles to form a chain. In the chain formation process, the temperature of the huge environment remains constant. Therefore the temperature of the system can be also considered to remain constant. In fact, the dissipated energy due to the friction force in the chain formation process will be converted into heat and transferred back to the environment.

The virtual work done by the system corresponding to the infinitesimal change $\delta\beta$ should include those done by the electric field, magnetic field, viscous friction force and the inertia force if the applied mechanical load is zero. Therefore, the generalized principle of the virtual work can be expressed in the following form:

$$\delta W_e + \delta W_m + \delta W_f + \delta W_i - T\delta S = 0, \tag{1}$$

where δW_e, δW_m, δW_f and δW_i are the virtual words done by the system through the electric, magnetic, viscous friction and inertia forces, respectively. δS is the change of the system entropy due to the change $\delta\beta$. In what follows, we will derive the terms in Eq.(1) one by one, and establish the evaluation equation of β.

2 The Virtual Work Done by the Electric Field and Magnetic Field

Consider that a suspension of fine dielectric particles in a liquid of low dielectric constants is put between two parallel plates, when the applied electric field is zero or very small, the thermal motion makes the particles randomly distributed in the space and form a uniform suspension (Fig. 1). As the electric field increases, the particles obtain an induced dipole moment. Thus the system consisting of those particles to form a chain will have some amount of electrostatic energy before aggregation. When the particles aggregate to form a chain, the electrostatic energy of the system will change. Since it is a dynamic process of the chain formation, it will also induce a magnetic field around it. Therefore the system will have some amount of magnetic energy in the chain formation process. The work done by the electric and magnetic field of the system should equal to the decrease of the electromagnetic energy of the system. Therefore if we can derive the change of electromagnetic energy of the system, we obtain the work done by the electric and magnetic field.

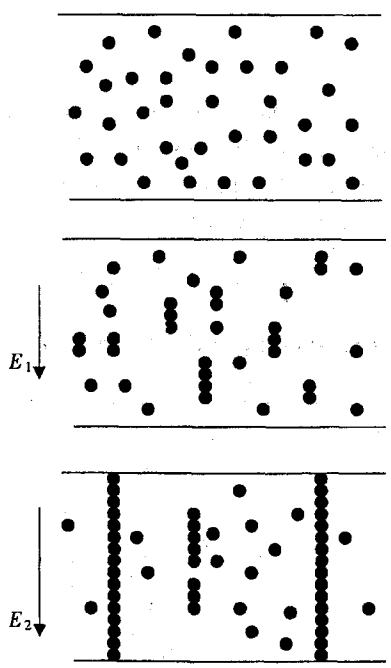

Fig.1 Schema of the chain formation process of the particles in ER fluid under the action of an applied electric field

2.1 The electromagnetic energy of spherical particles

Before aggregation, the system contains N spherical particles in ER fluid, the expression of

electromagnetic energy for the system can be derived. First consider a single spherical polarized particle with polarization $\boldsymbol{P}_s$ in a homogeneous dielectric material under the action of the external electric field. The internal electromagnetic energy can be expressed as

$$f = \frac{1}{2}\iiint_V \boldsymbol{E}\cdot\boldsymbol{D}\mathrm{d}v = \frac{1}{2}\iiint_V (\boldsymbol{E}^e + \Delta\boldsymbol{E})\cdot(\boldsymbol{D}^e + \Delta\boldsymbol{D})\mathrm{d}v = \frac{1}{2}\iiint_V E_i^e D_i^e \mathrm{d}v - \iiint_\Omega E_i^e p_i^s \mathrm{d}v - \frac{1}{2}\iiint_\Omega \Delta E_i p_i^s \mathrm{d}v, \tag{2}$$

where $\boldsymbol{E}^e$, $\boldsymbol{D}^e$ are the applied electric field and corresponding electric displacement, $\Delta\boldsymbol{E}$, $\Delta\boldsymbol{D}$ are the induced electric field and displacement due to the polarization prescribed in the particle $\boldsymbol{P}_s$. The interaction energy caused by introducing a single particle is

$$\Delta f = -\iiint_\Omega E_i^e p_i^s \mathrm{d}v - \frac{1}{2}\iiint_\Omega \Delta E_i p_i^s \mathrm{d}v. \tag{3}$$

For constant polarization and spherical inclusion, $\Delta\boldsymbol{E}$ is given by $\Delta\boldsymbol{E} = (-p_s/6\varepsilon)\boldsymbol{k}$. For the system containing N polarized particles, the polarization can be expressed in the form as $P_j^s = \sum_{i=1}^{N} p_j^s \mathrm{H}(\Omega_i)$, where $\mathrm{H}(\Omega_i)$ is the Heaviside function defined on the i-th particle. Without considering the interaction between the particles, the total interaction energy is obtained as

$$\Delta F = -\iiint_\Omega E_i^e P_i^s \mathrm{d}v - \frac{1}{2}\iiint_\Omega \Delta E_i(P_s) P_i^s \mathrm{d}v. \tag{4}$$

As assumed in this paper that the chain is formed by attracting particles to aggregate one by one along the direction of the applied field, the number of the separated particles in the system will decrease with time. Therefore, the change rate of the electromagnetic energy with the reduction of separated particles is given by

$$\Delta\dot{U} = \frac{\mathrm{d}(\Delta F)}{\mathrm{d}t} = \left[-\iiint_\Omega E_i^e p_i^s \mathrm{d}v - \iiint_\Omega \Delta E_i p_i^s \mathrm{d}v\right]\frac{\mathrm{d}N}{\mathrm{d}t}. \tag{5}$$

2.2 The electromagnetic energy of the spheroidal chain

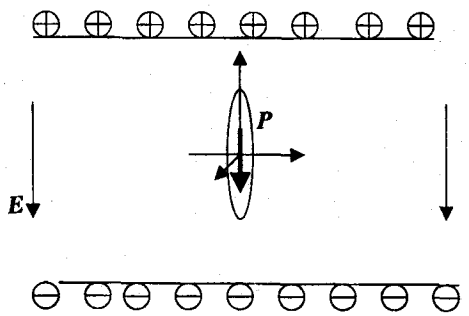

Fig.2 Schema of the spheroidal chain formed by the polarized particles under the action of an applied electric field

In the chain forming process, part of the electromagnetic energy will disappear with the reduction of separated particles. At the same time, part of the electromagnetic energy will appear with the aggregation of particles. Next we consider the change rate of the electromagnetic energy accompanying the growth of the chain. In the finite element V of the ER fluid there is a chain of polarized particles (Fig.2). The electromagnetic energy of the chain under the action of the applied field is given by

$$U = \frac{1}{2}\iiint_V (\boldsymbol{E}\cdot\boldsymbol{D} + \boldsymbol{H}\cdot\boldsymbol{B})\mathrm{d}v, \tag{6}$$

where V is the total volume of the ER material element. The change rate of the energy is given by

$$\dot{U} = \frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{1}{2}\iiint_V (\boldsymbol{E}\cdot\boldsymbol{D} + \boldsymbol{H}\cdot\boldsymbol{B})\mathrm{d}v\right] = \iiint_V [-\boldsymbol{E}\cdot\boldsymbol{J} - \nabla\cdot(\boldsymbol{E}\times\boldsymbol{H})]\mathrm{d}v. \tag{7}$$

In deriving Eq.(7), we have used the Maxwell equation to replace the time-derivative factors in the integrand. Eq.(7) is known as Poynting's theorem. $\boldsymbol{S} = \boldsymbol{E}\times\boldsymbol{H}$, where $\boldsymbol{S}$ is called the Poynting vector, which is identified as the energy flux, or energy flow rate per unit area.

2.3 The input electromagnetic energy through the boundary

The rate of the energy input by the outside environment through the boundary Γ is given by

$$\dot{W} = -\iint_\Gamma \boldsymbol{S}\cdot\mathrm{d}\boldsymbol{s}. \tag{8}$$

2.4 The work done by the electric and magnetic field

The change rate of the electromagnetic energy of the system is given by

$$\dot{G} = \dot{W} - \dot{U} + \Delta\dot{U} = p_s\iint_S E_3^1\dot{R}(t)\mathrm{d}s - \iint_S [\boldsymbol{S}]\cdot\mathrm{d}\boldsymbol{s} + \Delta\dot{U} = \iint_S \{p_s E_3^1\dot{R}(t) - [S_i]n_i\}\mathrm{d}s + \Delta\dot{U}, \tag{9}$$

where the integration is carried out on the surface of the chain. As side before, the work done by the electric and magnetic field of the system should equal to the decrease of the electromagnetic energy of the system. Therefore

$$\dot{W}_e + \dot{W}_m = -\dot{G}. \tag{10}$$

In fact, $\dot{G}$ represents the driving force for the growth of the chain. If $\dot{G} > 0$, that means the positive work was done on the system through the electric and magnetic field accompanying the growth of the chain. With the aid of the expressions of the electric and magnetic field both inside and outside the ellipsoidal inclusion, the change rate of the energy is given by

$$\dot{G} = \frac{p_s^2}{\varepsilon_f}\frac{\mathrm{d}N}{\mathrm{d}t}v_a\left[\frac{1}{6} - \frac{I_3(\beta)}{8\pi}\right] - \frac{p_s^2}{4\pi\varepsilon_f}\frac{1}{c_f^2}\iint_\Gamma \frac{\partial^2\Phi}{\partial t^2}\dot{R}(t)\mathrm{d}s, \tag{11}$$

where $\Phi(\lambda) = \frac{1}{2}[I(\lambda) - x_n x_n I_N(\lambda)]$, $x_n x_n I_N(\lambda) = x_1^2 I_1(\lambda) + x_2^2 I_2(\lambda) + x_3^2 I_3(\lambda)$,

and

$$\begin{cases} I(\lambda) = \dfrac{4\pi a_1 a_3}{\sqrt{\beta^2 - 1}}\operatorname{arccosh}\bar{b}, \\ I_3(\lambda) = 4\pi\beta(\operatorname{arccosh}\bar{b} - \bar{d}/\bar{b})/(\beta^2 - 1)^{3/2}, \\ I_1(\lambda) = I_2(\lambda) = 2\pi\beta(\bar{b}\bar{d} - \operatorname{arccosh}\bar{b})/(\beta^2 - 1)^{3/2}, \end{cases} \tag{12}$$

where $\bar{b} = \sqrt{(a_3^2 + \lambda)/(a_1^2 + \lambda)}$, $\bar{d} = \sqrt{(a_3^2 - a_1^2)/(a_1^2 + \lambda)}$, and λ is the largest positive root of the equation

$$\frac{x_1^2}{(a_1^2 + \lambda)} + \frac{x_2^2}{(a_2^2 + \lambda)} + \frac{x_3^2}{(a_3^2 + \lambda)} = 1. \tag{13}$$

When $\boldsymbol{x}$ is inside the inclusion, $\lambda = 0$.

The energy release rate can also be expressed in the form of

$$\dot{G} = g\frac{\partial \beta}{\partial t}, \tag{14}$$

where $g = \partial G/\partial \beta$ is the energy change per unit increase of the aspect ratio of the chain, and

$$\frac{\partial \beta}{\partial t} = \frac{\mathrm{d}N}{\mathrm{d}t}, \tag{15}$$

where $\mathrm{d}N/\mathrm{d}t$ is the change rate of the particles numbers aggregating on the chain. From Eq.(10), one can find

$$\delta W_e + \delta W_m = -g\delta\beta, \tag{16}$$

where

$$g = \frac{p_s^2}{\varepsilon_f} v_a \left[\frac{1}{6} - \frac{I_3(\beta)}{8\pi}\right] - \frac{p_s^2}{4\pi\varepsilon_f}\frac{1}{c_f^2}\iint_\Gamma \frac{\partial^2 \Phi}{\partial t^2}\frac{\partial R}{\partial \beta}\mathrm{d}s. \tag{17}$$

By considering that Φ is only the function of β, Eq.(17) can be rewritten in the form as

$$\begin{aligned} g = {} & \frac{p_s^2}{\varepsilon_f} v_a \left[\frac{1}{6} - \frac{I_3(\beta)}{8\pi}\right] - \\ & \frac{p_s^2}{8\pi\varepsilon_f}\frac{1}{c_f^2}\left[\left(\frac{\mathrm{d}\beta}{\mathrm{d}t}\right)^2 \iint_\Gamma \frac{\partial^2 \Phi}{\partial \beta^2}\frac{\partial R}{\partial \beta}\mathrm{d}s + \frac{\mathrm{d}^2\beta}{\mathrm{d}t^2}\iint_\Gamma \frac{\partial \Phi}{\partial \beta}\frac{\partial R}{\partial \beta}\mathrm{d}s\right] = \\ & A(\beta)\frac{\mathrm{d}^2\beta}{\mathrm{d}t^2} + B(\beta)\left(\frac{\mathrm{d}\beta}{\mathrm{d}t}\right)^2 + C(\beta), \end{aligned} \tag{18}$$

where

$$A(\beta) = -\frac{p_s^2}{8\pi\varepsilon_f}\frac{1}{c_f^2}\iint_\Gamma \frac{\partial \Phi}{\partial \beta}\frac{\partial R}{\partial \beta}\mathrm{d}s, \tag{19}$$

$$B(\beta) = -\frac{p_s^2}{8\pi\varepsilon_f}\frac{1}{c_f^2}\iint_\Gamma \frac{\partial^2 \Phi}{\partial \beta^2}\frac{\partial R}{\partial \beta}\mathrm{d}s, \tag{20}$$

$$C(\beta) = \frac{p_s^2}{\varepsilon_f} v_a \left[\frac{1}{6} - \frac{I_3(\beta)}{8\pi}\right]. \tag{21}$$

3 The Virtual Work Done by the Inertia Force

Usually in computer simulations, the inertia force was neglected. But if the mass density of particles has great effect on kinetics of the chain formation, the inertia force should be included in the analysis. In the following, we propose two simple models for the motion of particles, and derive the virtual work done by the inertia force.

3.1) The particle moves in a constant acceleration toward the chain

When an initially static particle is attracted to attach on the chain, the particle should move with acceleration. This acceleration can be considered to produce D'Alembert inertia force, which should do some work when the particle is moving. In this part, we will derive the expression for the work done by the inertia force. To simplify our analysis, we assume that the particle will move in a constant acceleration a toward the chain. The average distance between the particle and the chain is Δx, which can be determined if one knows the concentration of particles. If the velocity of the chain growth is denoted as $\dot{\beta}$, the time for one particle to aggregate on the chain is given by

$$t = 1/\dot{\beta}. \tag{22}$$

Since the attachment of one particle on the chain corresponds to that the aspect ratio β increases by one unit. Therefore, the constant acceleration can be determined through the following equation:

$$\Delta x = \int_0^t v\mathrm{d}t = \int_0^t at\mathrm{d}t = \frac{1}{2}\frac{a}{\dot{\beta}^2}. \tag{23}$$

In derivation of Eq.(23), we used Eq.(22). Thus

$$a = 2\Delta x\,\dot{\beta}^2. \tag{24}$$

Since the mass density per unit aspect ratio is the mass of one particle, the inertia force acting on an infinitesimal element $\delta\beta$ is

$$f_i = m\delta\beta a = 2\Delta x\,\dot{\beta}^2 m\delta\beta, \tag{25}$$

where m is the mass of one particle. The inertia force is in the direction opposite to the acceleration, therefore, the work done by the system through the inertia force is given by

$$\delta W_i^{\mathrm{I}} = f_i\Delta x = 2\Delta x^2\dot{\beta}^2 m\delta\beta. \tag{26}$$

That means that the inertia force does the negative work on the system.

3.2) The velocity of the particle will reduce to zero when it aggregates on the chain

Initially the particle keeps static. Thus if the initial and final velocity of the particle are all zero as assumed, the kinetic energy will keep unchanged, therefore the work of the inertia force will keep zero as follows:

$$\delta W_i^{\mathrm{II}} = \int_0^{\Delta x} f_i\mathrm{d}x = m\delta\beta\int_0^t \dot{v}v\mathrm{d}t = m\delta\beta\int_0^t v\mathrm{d}v =$$

$$\frac{1}{2}m\delta\beta[v^2(t) - v^2(0)] = 0. \tag{27}$$

4 The Virtual Work Done by the Friction Force of Viscous Fluid

When the particles move to aggregate on the chain, the fluid will induce hydrodynamic drag resistance on the particle. If the velocity of the particle is denoted as v, which is not too high, i.e., the Reynolds number $Re \leqslant 0.2$, the hydrodynamic resistance on the particle is approximately given by the Stokes resistance

$$f_v = 6\pi\eta_0 a_1 v, \tag{28}$$

where η_0 is the viscosity of the dispersing fluid, and a_1 is the radius of the particle. For infinitesimal element $\delta\beta$, the drag force is $f_v\delta\beta$. Therefore the work done by the friction force to cross the distance Δx is given as follows:

$$\delta W_f = \int_0^{\Delta x} f_v\delta\beta\mathrm{d}x = \int_0^t f_v\delta\beta v\mathrm{d}t = 6\pi\eta_0 a_1 a^2\delta\beta\int_0^t t^2\mathrm{d}t. \tag{29}$$

Substituting Eqs.(22) and (24) into Eq.(29) yields

$$\delta W_f = 8\pi\eta_0 a_1\Delta x^2\dot{\beta}\delta\beta. \tag{30}$$

5 The Basic Differential Equation of the Chain Formation

Substitution of Eqs.(16), (26) and (30) into the expression of virtual work, Eq.(1) gives the basic differential equation of chain formation

$$A(\beta)\frac{d^2\beta}{dt^2}+[B(\beta)-2m\Delta x^2]\left(\frac{d\beta}{dt}\right)^2-8\pi\eta_0 a_1\Delta x^2\frac{d\beta}{dt}+T\frac{dS}{d\beta}+C(\beta)=0. \quad (31)$$

Equation (31) is the governing equation of the chain growth including the effect of inertia force based on our energy model. The form of Eq.(31) is similar to that used in computer simulation for one particle, where the term containing the temperature and entropy is similar to the Brownian force.

The parameter c_f given by Eq.(18) in the expressions of $A(\beta)$ and $B(\beta)$ as shown in Eqs.(19) and (20) represents the propagation speed of electromagnetic wave in the fluid. It should be much larger than the speed of the chain growth $d\beta/dt$, and when the chain formation approaches a stationary state, the first term in Eq.(31) can also be neglected. Therefore Eq.(31) becomes

$$2m\Delta x^2\left(\frac{d\beta}{dt}\right)^2+8\pi\eta_0 a_1\Delta x^2\frac{d\beta}{dt}-T\frac{dS}{d\beta}-C(\beta)=0. \quad (32)$$

The solution of Eq.(32) for the velocity of the chain formation is given by

$$\frac{d\beta}{dt}=\frac{1}{4m\Delta x^2}\left\{\sqrt{(8\pi\eta_0 a_1\Delta x^2)^2+8m\Delta x^2[C(\beta)+TdS/d\beta]}-8\pi\eta_0 a_1\Delta x^2\right\}, \quad (33)$$

in which we omit one solution which gives the negative velocity.

From Eq.(33), it is very clear that the velocity of the chain formation depends on the applied electric field, the dielectric constants of particles and fluid, which are contained in the expression of $C(\beta)$, the particle concentration, size, and mass density.

If we adopt the second model of particle's motion by substituting Eqs.(16), (27) and (30) into the expression of virtual work, Eq.(1) gives the basic differential equation of chain formation

$$A(\beta)\frac{d^2\beta}{dt^2}+B(\beta)\left(\frac{d\beta}{dt}\right)^2-8\pi\eta_0 a_1\Delta x^2\frac{d\beta}{dt}+T\frac{dS}{d\beta}+C(\beta)=0. \quad (34)$$

Using the same reasoning as obtaining Eq.(32), one can derive

$$8\pi\eta_0 a_1\Delta x^2\frac{d\beta}{dt}-T\frac{dS}{d\beta}-C(\beta)=0. \quad (35)$$

The velocity is given by

$$\frac{d\beta}{dt}=\frac{1}{8\pi\eta_0 a_1\Delta x^2}\left[C(\beta)+T\frac{dS}{d\beta}\right]. \quad (36)$$

$C(\beta)$ given by Eq.(21) increases from zero to $p_s^2 v_a/6\varepsilon_f$ when β changes from 1 to infinity. The velocity given by Eq.(36) versus the aspect ratio β is shown in Fig.3 where, for simplicity, we neglect the heat transfer between the system and its environment. From the figure, it is very clear that the velocity of the chain formation approaches its maximum steady value very soon. From Eqs.(33) and (36), the maximum velocity of the chain formation is given by the following equations, respectively

$$\left(\frac{d\beta}{dt}\right)_{max\,1}=\frac{1}{4m\Delta x^2}\left\{\sqrt{(8\pi\eta_0 a_1\Delta x^2)^2+8m\Delta x^2[p_s^2 v_a/6\varepsilon_f+TdS/d\beta]}-8\pi\eta_0 a_1\Delta x^2\right\}, \quad (37)$$

$$\left(\frac{d\beta}{dt}\right)_{max\,2} = \frac{1}{8\pi\eta_0 a_1 \Delta x^2}\left[\frac{p_s^2 v_a}{6\varepsilon_f} + T\frac{dS}{d\beta}\right]. \tag{38}$$

The time needed for the chain to cross the distance between the electrodes is also given by Eq.(36), which is shown in Fig.4, where $t^* = \bar{\mu}\varepsilon_f \Delta x^2/(p_s^2 v_a)$. From Figs.3 and 4, one can find that the chain grows in a uniform speed soon after it starts to propagate.

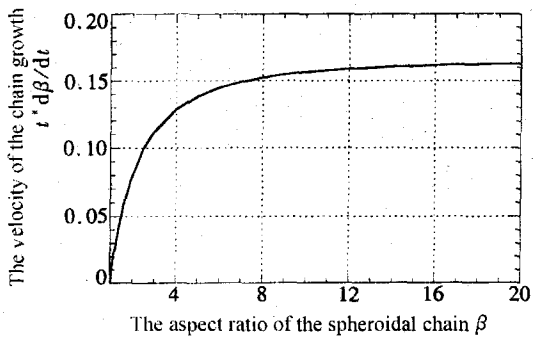

Fig.3 The velocity of the chain growth $t^* d\beta/dt$, where $t^* = \bar{\mu}\varepsilon_f \Delta x^2/(p_s^2 v_a)$, versus the aspect ratio of the chain

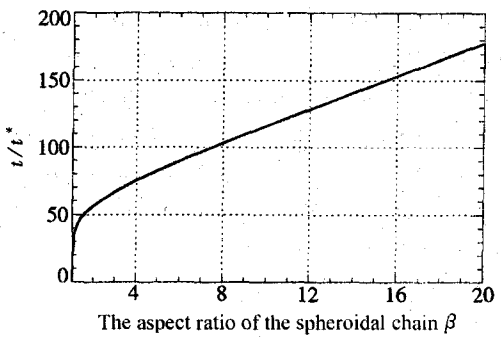

Fig.4 The formation time t/t^* of a whole chain versus the distance between the electrodes, where $t^* = \bar{\mu}\varepsilon_f \Delta x^2/(p_s^2 v_a)$

If we define the critical field for the phase transition as the minimum value of the applied electric field to induce the chain formation, this electric field can be determined by the following condition:

$$C(\beta) + T\frac{dS}{d\beta} = 0. \tag{39}$$

When $\beta \to \infty$, one can determine the lower bound of the electric field that induces the transition happening:

$$E_c = \sqrt{\frac{32\pi^2 RT}{3a^2 \varepsilon_f v_a}}, \tag{40}$$

where $R = -dS/d\beta$. In deriving Eq.(40), we used Eq.(31). The form of Eq.(40) is very similar to that obtained by Tao *et al.*[2]. Based on this model, one need first to know the entropy change accompanying the chain formation process to incorporate the effect of temperature

6 Comparison with Experimental Results and the Molecular-Dynamics-Linke Simulation Results

Based on our model, the response time, which is defined as the time to form the first bridging chain over the electrodes, can be derived through the integration of Eq.(36) as follows:

$$\tau = 8\pi\eta_0 a_1 \Delta x^2 \int_0^{L/a_1} \left[C(\beta) + T\frac{dS}{d\beta}\right]^{-1} d\beta, \tag{41}$$

where L is the gap between the electrodes. By neglecting the heat transfer during the chain formation process, and substituting Eq.(21) into Eq.(41), one obtains

$$\tau = 8\pi\eta_0 a_1 \Delta x^2 \frac{\varepsilon_f}{p_s^2 v_a}\int_0^{L/a_1}\left[\frac{1}{6} - \frac{I_3(\beta)}{8\pi}\right]^{-1} \mathrm{d}\beta. \tag{42}$$

Since the electric dipole moment density p_s is in proportion to the applied electric field E^e, the response time is scale as $1/(E^e)^2$. Hence, such prediction seems in coincidence with the simulation result and experimental results. The dependence of the response time on the volume concentration of particles is reflected through the parameter Δx, which is the average distance for a particle to move to aggregate on the chain. How the distance Δx depending on the volume concentration of particles is a complicated geometry problem, since the particles are randomly distributed in space. Consider that the chain will be formed along the line oo', which connects the upper and lower surfaces of electrodes. If the number of particles in volume v is assumed to follow Poisson distribution, i.e., the probability that the volume v contains k particles is given by

$$Pr(i = k) = \frac{(nv)^k}{k!}e^{-nv}, \tag{43}$$

where n is the average number of particles in unit volume. Next, we try to derive the average nearest distance between the particles and the chain position oo'. One can consider a cylindrical region with radius r surrounding oo'. The probability that there is no particle in the cylindrical region is given by

$$Pr(i = 0) = e^{-nv} = e^{-\pi nLr^2}. \tag{44}$$

The probability density function of particle's position is $2\pi nLr$. Hence, the probability density function of the nearest distance between the particles and oo' is the conditional probability density function given that there is no particle in the cylindrical region with radius r, therefore

$$f(r) = 2\pi nLre^{-\pi nLr^2}. \tag{45}$$

The average distance Δx is given by the integration

$$\Delta x = \int_0^\infty rf(r) = \int_0^\infty 2\pi nL\, r^2 e^{-\pi nLr^2}\mathrm{d}r =$$

$$2\pi nL\int_0^\infty r^2 e^{-\pi nLr^2}\mathrm{d}r = (\pi nL)^{-1/2}\Gamma\left(\frac{3}{2}\right). \tag{46}$$

The average number of the particles in unit volume is related with the volume concentration of particles as follows:

$$n = \Psi/v_a, \tag{47}$$

where Ψ is the volume concentration of particles, and v_a is the average volume for one particle.

Substitution of Eq.(47) into (46) gives

$$\Delta x = (\pi L)^{-1/2}\Gamma\left(\frac{3}{2}\right)\left(\frac{v_a}{\Psi}\right)^{1/2} \tag{48}$$

By substituting Eq.(48) into the expression for response time, Eq.(42), one can find that the response time scales as $\tau \propto \Psi^{-1}$. Klingenberg *et al.* (1993)[4] carried out the experimental measurement on the response time. They found that the data had a considerable scatter, and the

response time varied as $\tau \propto \Psi^{-2.8}$, while their computer simulation gave $\tau \propto \Psi^{-3.2}$. Exact measurement on the response time is a challenge at present stage.

7 Concluding Remarks

At last, we would like to summarize the work presented in this paper.

1) In this paper, a generalized principle of virtual work for the chain formation process in ER fluid is developed based on thermodynamics analysis. Based on the energy approach, the governing differential equation for the chain formation in ER fluid is derived. The advantage of such kind of energy approach over classic molecular-dynamics-like simulation method is quite similar to the advantage of Langrange analytical mechanics over Newton mechanics. Using the energy approach, one can consider a few numbers of entities as generalized coordinates to represent the degrees of freedom of the system. In this paper, we only use the aspect ratio β of the chain as the generalized coordinate. The number of generalized coordinates is of course likely to be much less than the number of particles. In fact, as a specific case, if one denotes three degrees of freedom for each particle, the dynamic governing differential equations for computer simulation can also be obtained through such an energy approach.

2) The velocity of the chain formation is obtained explicitly. It is found that the chain grows in a uniform speed soon after it starts to propagate.

3) The response time of an ER system is also obtained and compared with experimental and numerical results.

4) Even though the energy approach developed in this paper is quite general for more complicated ER structures, the assumptions adopted in this investigation, such as neglecting the image effect of the electrodes and taking the prolate spheroid shape of the chain will limit the application of the results. To deal with more complicated ER structures, on can adopt more generalized coordinates to represent the degrees of freedom of the system instead of only using the aspect ratio β of the chain.

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