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Analysis of giant electrorheological fluids

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1. Introduction

Electrorheological (ER) fluids (suspensions of very fine particles in an electrically insulating fluid) are smart materials with structural and rheological properties that are significantly altered by the application of an external electric field [1,2]. The suspended particles attract one another and instantly form solid-like networks of fibers along the direction of the applied electric field. At low shear rates, the apparent viscosity can be reversibly changed over three or four orders of magnitude, in response to an electric field [3–7]. A typical ER fluid responds guickly to the applied electric field with the response times on the order of milliseconds and shows the behavior of a liquid or a gel depending on the applied electric field [1,2,6]. The maximum yield stress of a usual ER fluid is on the order of several kilopascals whereas the apparent maximum yield stress of similar magnetorheological (MR) fluids (whose properties vary under the application of a magnetic field) can reach up to 100 kPa [8]. A lot of efforts have been dedicated for the development of smart ER fluids showing the yield stress over 100 kPa. The appearance of GER fluids changed the paradigm: these fluids exhibit yield stresses higher than 100 kPa [9-11]. Huang et al. ascribed the mechanism of GER fluids to the saturation of polarization under a higher energy [11]. The yield stress behavior of GER fluids was also considered to be different from that of normal ER fluids [9,12].

For an idealized ER system where uniform dielectric hard spheres are dispersed in a Newtonian fluid medium, the yield stress (τ_y) depends on the dielectric constant mismatch between the particles and the continuous media ("polarization model"),

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ABSTRACT

The yield stress dependence on electric field strength for giant electrorheological (GER) fluids over the full range of electric fields was examined using Seo's scaling function which incorporated both the polarization and the conductivity models. If a proper scaling was applied to the yield stress data to collapse them onto a single curve, the Seo's scaling function could correctly fit the yield stress behavior of GER suspensions, even at very high electric field strengths. The model predictions were also compared with recently proposed Choi et al.'s model to allow a consideration of the universal framework of ER fluids.

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and can be expressed as $\tau_y \propto \phi k_f f(\beta) E_0^2$, where ϕ is the volume fraction of the particle, $\beta = (k_p - k_f)/(k_p + 2k_f)$ is a dimensionless dielectric mismatch parameter, E_0 is the electric field strength, and k_p and k_f are the dielectric permittivities of the particle and the fluid, respectively [2,4,13]. However, as noted by Choi et al. [13], experimental yield stress data deviate significantly from this equation and are better represented by the power law; $\tau_y \propto E_0^m$ (m < 2) at high electric field strengths. This is ascribed to the electrical breakdown of ER fluids under high-strength electric fields [1,3]. Davis and Ginder showed that $\tau_y \propto E_0^{3/2}$ for $E_0 > E_c$, where E_c is the critical electric field strength (the "conduction model") [14]. Based on their experimental observations, Sheng et al. recently reported the linear dependence of the yield stress on the applied electric field ($\tau_y \propto E_0$) for GER fluids [9,11,12]. They proposed a "saturation polarization model".

For the purpose of designing ER fluids, a more precise knowledge of the relationship between the rheological properties of the suspension and variables such as the deformation rate, the applied electric field strength, and the composition, is required. Although predictive models have provided insights into the mechanisms governing the behavior of ER fluids, they have not been sufficiently quantitative to be applied in a design process. In addition, these models do not yet incorporate the yield stress behavior of GER fluids [15,16]. Since the yield stress is a critical design parameter in ER applications, a universal yield stress of GER fluids and normal ER fluids.

Choi et al. [15,16] recently reported that they could model the static yield stress of Cheng et al.'s GER fluids [17] using their scaling function up to 1.96 kV/mm. To fit the linear dependence up to 5 kV/mm, Choi et al. [16] proposed a new universal yield stress scaling equation shown later; this equation could predict the yield



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stress behavior of GER fluids for both low electric field strength region where polarization effects dominated and high electric field strength region where polar molecules dominated. However, this model did not adequately fit the yield stress behavior of normal ER fluids at high electric field strength region. Recently, we proposed a new scaling function to simulate the yield stress behavior of ER fluids over the full range of electric fields [18]. In spite of its simple form, comparison of the model predictions with the experimental data for both ac and dc fields and with the polarization model and conduction model demonstrated that the proposed model worked correctly and universally to fit the yield stress behavior.

In the present work, we applied the same scaling function to GER fluids and compared the results with Choi et al.'s scaling function. Although reported experimental data for the yield stress of GER fluids were one order of magnitude larger than the theoretical limit of Sheng et al. [10], our modeling demonstrated that accurate modeling by the same scaling function as general ER fluids was possible for the GER fluid as long as appropriate scaling is applied.

2. New modeling results and discussion

The variation of the yield stress as a function of *E* is divided into two regions by the critical electric field strength, E_c , which depends on several factors, including the particle conductivity, the conduction mismatch between the particle and the liquid medium, and the particle volume fraction [1,2]. As mentioned above, the static yield stress corresponding to the stress at low shear rates is quadratically related to the electric field strengths at low electric field strength ($\tau_y \propto E_0^2$) while the dependence becomes weaker at high electric field strengths ($\tau_y \propto E_0^{3/2}$). However, GER fluids was shown to follow a linear dependence at high electric field strengths [9– 11,18]. A decade ago, Choi et al. showed that proper scaling allows the yield stress data for ER fluids to be collapsed onto a single curve [13]. If this scaling allows the GER fluids data to be collapsed into that single curve, our scaling function can fit the experimental data to analyze the GER fluids as well as normal ER fluids [18].

To accommodate the dependence of the GER fluid yield stress on the electric field strength, Choi et al. [16] proposed a scaling function,

$$\tau_{\mathbf{y}}(E_0) = \frac{\alpha E_0^2 I_1\left(\frac{E_0}{E_c}\right)}{\left(\frac{E_0}{E_c}\right) I_0\left(\frac{E_0}{E_c}\right)} \tag{1}$$

where I_1 and I_0 are modified Bessel functions of order 1 and 0, respectively. α depends on the dielectric constant of the fluid as well as the particle volume fraction and β , and E_c represents the critical electric field strength originating from the nonlinear conductivity model. This expression also defines the crossover between different power-law regimes in the τ_y vs. E_0 plot (Fig. 1). This equation can describe the limiting behaviors at low and high electric field strengths; For $E_0 \ll E_c$, I_1 (E_0/E_c)/ E_0 and I_0 (E_0/E_c) = constant, which gives

$$\tau_y = \alpha E_0^2 \tag{2a}$$

and for $E_0 \gg E_c$, $I_1(x)$ and $I_0(x) \propto e^x / \sqrt{2\pi x}$, hence,

$$\tau_{y}(E_{0}) = E_{0} \tag{2b}$$

Therefore, the scaling function of Eq. (1) looks promising to fit the yield stress in GER fluids (which is known to show a quadratic dependence at low electric field strengths, but a linear dependence at high electric field strengths) [9,12]. To fit the experimental data, Choi et al. normalized Eq. (1) with $\tau_{yc}(E_c) = \alpha E_c^2 I_1(1)/I_0(1) = 0.446 \alpha E_c^2$, giving rise to a dimensionless form:

$$\hat{t} = \frac{2.242 \, \hat{E} I_1(\hat{E})}{I_0(\hat{E})} \tag{3}$$

where $\hat{E} = E_0/E_c$, and $\hat{\tau} = \tau_y(E_0)/\tau_y(E_c)$. By normalizing the data with $\tau_y(E_c)$ and E_c , the experimental data could be collapsed into a single curve (Fig.1). Although normalizing the data with the scaling constant value of $I_1(1)/I_0(1) = 0.446$ afforded a good fit to the experimental data, the predictions of Eq. (1) gave the largest difference from the true value at the critical value of $\hat{E} = 1$. This was similar to the behavior observed for their previous model for normal ER fluids [13]. Nonetheless, this scaling function is unsuitable for normal ER fluids which show $E_0^{3/2}$ dependence at high electric field strengths rather than the linear dependence of GER fluids.

Based on physical reasoning and the rheological behavior of Bingham fluids, Seo [18] recently introduced a simple equation to fulfill the necessary conditions for normal ER fluids,

$$\tau_{y}(E_{0}) = \alpha E_{0}^{3/2} \left(1 - \exp\left(-m'\sqrt{E_{0}}\right) \right)$$
(4)

where α is a parameter depending on the dielectric constant of the fluid, the particle volume fraction and m' is a fitting parameter. This equation has the following two limiting behaviors at low and high electric field strengths, respectively

$$\tau_y = \alpha \, m' E_0^2 \propto E_0^2 \quad \text{for} \quad E_0 \ll E_c \tag{5a}$$

$$\tau_y = \alpha E_0^{3/2} \propto E_0^{3/2} \quad \text{for} \quad E_0 \gg E_c \tag{5b}$$

Normalizing Eq. (4) with E_c and $\tau_y = \alpha E_0^{3/2}$ gives the following equation:

$$\hat{\tau} = \hat{E}^{3/2} \left(1 - \exp(-m\sqrt{\hat{E}}) \right) \tag{6}$$

where $\hat{\tau} = \tau_y / \alpha E_0^{3/2}$, $\hat{E} = E/E_c$, $m = m' \sqrt{E_c}$. This model demonstrated a very good fit with the experimental data [18,19].

To test the ER behavior of GER fluids, we used the same experimental data as that used by Choi et al. i.e., data of urea-coated BaTiO(C_2O_4)₂ (UBT, core-shell particles) suspended in silicone oil [9] and calcium and titanium-precipitated SiO₂ (CTP-SiO₂, core-shell particles) suspended in silicone oil [17]. The UBT particles were fabricated by first dissolving barium chloride in distilled water at 50–70 °C. Separately, oxalic acid was dissolved in water at 65 °C in an ultrasonic tank, with titanium tetrachloride added slowly to the solution. The two solutions were mixed in an ultrasonic bath, at 65 °C. Nanometer-sized barium titanyl oxalate particles were fabricated by incorporating CTP layers on spherical SiO₂ surfaces via a coprecipitation process. The CTP-SiO₂ nanoparticles



Fig. 1. The yield stress vs. electric field strengths for the GER fluids experimental data in log–log scale: ((\Box) UBT from Ref. [9] and (\bigcirc) SiO₂ (2.3 wt.%) – CTP ER fluid from Ref. [18]). *E*_{c1} and *E*_{c2} are critical electric field strength of each suspension [16].

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