

전기유변유체의 Universal Yield Stress Scaling Function

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Universal Yield Stress Scaling Function for Electrorheological Fluids

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Introduction

An electrorheological (ER) fluid commonly consists of a suspension of micron-sized particles, which have a higher dielectric constant and/or conductivity than that of suspending fluids, in a non-conducting fluid. The rheological properties of ER fluids are drastically changed by an applied electric field (E_0), because the suspended particles attract each other to form a solid-like network of fibers aligned in the field direction. This phenomenon is also reversible, *i.e.* the fibrillation structures disappear when E_0 is removed. The main characteristics of ER fluids are a high yield stress and enhanced viscosity under an applied electric field, a response time on the order of milliseconds, and full reversibility, *i.e.* the fluid recovers its original properties when the electric field is removed. The yield stress is one of the critical design parameters in an ER device and has attracted considerable attention both experimentally [1-4] and theoretically [5]. Many published experimental data for the yield stress dependence on the electric field strength are inconsistent due to the various material systems and measuring conditions, including the actual measuring devices [6]. In this work, we present a generalized scaling function for the normalized yield stress via scaling of the applied electric field strengths, which is convenient to ER/magnetorheological (MR) fluid experimentalists for their data analyses. Several models have been proposed to describe the ER phenomena via shear stress equations [5,7-9]. The original model, the so-called "polarization model", attributed the attractive force between particles to Maxwell-Wagner's interfacial polarization and employed the point-dipole approximation. Within this framework, the yield stress (τ_y) is represented as

$$\tau_y \propto \phi K_f E_0^2 f(\beta), \quad (1)$$

where ϕ is the volume fraction of the particles and $\beta = (K_p - K_f)/(K_p + 2K_f)$ is the dimensionless dielectric mismatch parameter. Here, K_p and K_f are the dielectric permittivities of the particle and the fluid, respectively. This polarization model shows excellent agreement [1] with data for small ϕ and E_0 . However, yield stress data significantly deviate from Eq. (1) at high electric field strengths [7] and are better represented by the power law; $\tau_y \propto E_0^m$ ($m < 2$). As the gap between the conducting particles in the fluid decreases (*e.g.*, increasing concentration), the electric response of the fluid becomes nonlinear through electrical breakdown (or partial discharge) under high electric field strengths. Davis and Ginder [7,10] incorporated this nonlinear conductivity effect with the bulk conducting particle model and constructed an alternative yield stress model, showing that power law index "m" approaches 3/2 for high electric field strengths. Davis showed that over a range of unscaled E_0 greater than critical electric field (E_c), the slope of E_0 vs. τ_y (on log paper) is 3/2, while the slope approaches 2 for small E_0 . To represent the yield stress data for a broad electric field strength range, we introduce the following simple hybrid equation:

$$\tau_y(E_0) = \alpha E_0^2 \left(\frac{\tanh \sqrt{E_0/E_c}}{\sqrt{E_0/E_c}} \right), \quad (2)$$

where α depends on the dielectric constant of the fluid, the particle volume fraction, and β . E_c represents the critical electric field originated from nonlinear conductivity model, and is descriptive for crossover behavior and defines (or links) the two regimes in the E_0 vs. τ_y plot (Figs. 1 and 2).

Experimental

Various particles used for the ER fluids were synthesized. Polyaniline (PANI) was synthesized through chemical oxidation process. After the reaction, PANI particles were ground using a ball mill and passed through a sieve. To obtain semiconducting polyaniline, we dedoped the PANI particles by reducing the pH of the aqueous medium which contained the particles to pH 9.0 using an aqueous NaOH solution. The pH-controlled particles were filtered and washed using distilled water, ethanol, and cyclohexane in order to remove oligomer and excess monomer and to make the particle surface hydrophobic. Finally, products were dried in a vacuum oven at room temperature. The synthesized PANI was identified from characteristic IR peaks obtained by a Fourier Transform Infrared (FT-IR) spectrometer.

In addition, synthesized PANI particles were also used as the core material for the encapsulation, and prepolymer MF as the shell materials. A mixture of 25 g of a 20 % citric acid (Sigma Chemical Co.) or poly(styrene sulfonic acid) (Alco Chemical Co.) and 100 g of distilled water was prepared. The pH of the solution was then controlled to 4 using 1M NaOH. Dried PANI particles were put into this solution. After stirring this mixed solution for 2 hr at 40~45°C in a water bath, the solution of prepolymer melamine (DAE YANG Chemical Co.) and 37 % formaldehyde (Sigma Chemical Co.) was added while stirring. As the temperature rose to about 65°C, MF resins were cured, and encapsulation of the PANI particles was proceeded. After completion of the encapsulation reaction, the particles were filtered and washed with distilled water several times to remove the unreacted residuals. Finally, the products were dried in a vacuum oven at room temperature.

Furthermore, semiconductive poly(p-phenylene) (PPP) particle was synthesized following the method of Kovacic and Oziomek. Benzene was converted to poly(p-phenylene) using anhydrous aluminum chloride and cupric chloride under nitrogen atmosphere and polymerization temperature was kept at 45°C. At early stage in the reaction, water was added as initiator. After polymerization, the mixture was washed by using water and ethyl alcohol to remove CuCl and aqueous acid.

All ER fluids were prepared by dispersing the particles within a silicone oil. The density and kinematic viscosity of the silicone oil are 0.95g/ml and 30cS at 25°C, respectively. Rheological properties were determined by a rotational Physica rheometer (MC120) with Couette type geometry, a high-voltage generator, and an oil bath for temperature control. An electric field was applied for five minutes in order to obtain an equilibrium chain-like, or columnar, structure before applying the shear.

Results and discussion

Figure 1 shows our data for an ER fluid consisting of microencapsulated PANI (MCPA) with a melamine-formaldehyde (MF) resin dispersed in silicon oil, which are in excellent agreement with Eq. (2). MCPA composites are denoted as MCPA_i with $i = 1, 2, 3$. Here 1, 2, and 3 are related with thickness of MF resin. Because microencapsulation entraps

core materials with polymers, we used this method to modify the PANI particles. E_c for the three different MCPA composites are 1.09 (MCPA1), 1.07 (MCPA2), and 1.05 (MCPA3) kV/mm, respectively. In our preliminary investigation with various ER fluid data [11-13], we also found that E_c is influenced by the conductivity mismatch between the particle and medium liquid and is weakly dependent on the volume fraction.

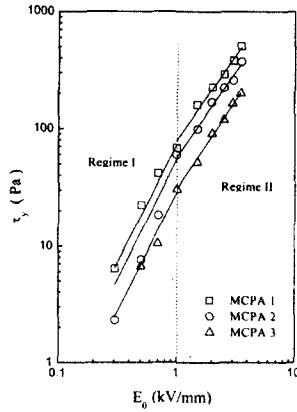


Figure 1. τ_y vs. E_0 for 20 wt% suspensions of MCPA particles encapsulated with MF resin. The ratios by weight of polyaniline to MF resin are 10/114 (MCPA1), 10/152 (MCPA2), and 10/190 (MCPA3), respectively.

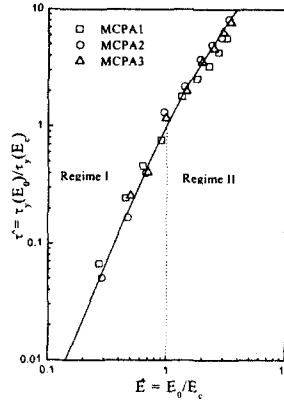


Figure 2. $\hat{\tau}$ vs. \hat{E} for 20 wt% suspensions of MCPA particles encapsulated with MF resin in silicone oil. The line is obtained from Eq. (4).

To collapse the data into a single curve, we normalized Eq. (2) with E_c and $\tau_y(E_c) = \alpha E_c^2 \tanh(l) = 0.762 \alpha E_c^2$:

$$\hat{\tau} = 1.313 \hat{E}^{3/2} \tanh \sqrt{\hat{E}}, \quad (3)$$

where $\hat{E} = E_0/E_c$ and $\hat{\tau} = \tau_y(E_0)/\tau_y(E_c)$. The data shown in Fig. 1 collapsed onto a single curve by using Eq. (3). Figure 2 represents these collapsed data. To better fit experimental data, we introduced one additional scaled parameter b:

$$\tau_y(E_0) = \alpha E_0^2 \left(\frac{\tanh(E_0/E_c)^{0.5+b}}{(E_0/E_c)^{0.5+b}} \right) \quad (4)$$

To obtain a universal correlation, we rescaled $\hat{\tau}$ and \hat{E} with $\hat{\tau} = \hat{\tau} \hat{E}^{4b}$ and $\hat{E} = \hat{E}^{1+2b}$ as

$$\hat{\tau} = 1.313 \hat{E}^{1.5} \tanh \hat{E}^{0.5} \quad (5)$$

Data for an ER system with PPP particles doped with $FeCl_3$ in aqueous solution collapsed onto a single scaling curve using Eq. (5). In addition, for the 10 wt% polyaniline particles in the mineral oil, yield stress as a function of the field strength agrees with Eq. (5) by setting $b = 0.38$ with our scaling curve as shown in Fig. 3.

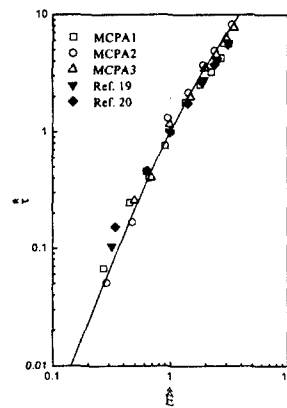


Figure 3. $\hat{\tau}$ vs. \hat{E} for 20 wt% suspensions of MCPA particles encapsulated with MF resin in silicone oil, 10 wt% polyaniline particle suspension in mineral oil (Data are taken from Ref. 19) and 20 vol% 1.5 μm doubly coated particles in silicone oil (Data are taken from Ref. 20). The line is obtained from Eq. (6).

It is noted that Gonon *et al.* [8] predict the theoretical development of $b = 0.38$. To examine the effect of the frequency, we plotted the yield stress data measured under a 50 Hz ac field applied between the parallel plates by Tam *et al.* [14] using 1.5 μm doubly coated particles. As shown in Fig. 3 (set $b = 0.28$), the yield stress obtained under the ac field also fit well to the scaling yield stress function [Eq. (5)].

We believe that Eq. (5) is very useful in constructing the master yield stress curve for ER/MR fluids. Even though the proposed correlations (Eqs. (4) and (5)) are not derived from first principles, they are important for representing data by experimentalists. The beauty of these correlations is the fact that most of the ER/MR experimental data can be collapsed with two parameters E_c and b .

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