Modeling and Analysis of Electrorheological Suspensions in Shear Flow

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ABSTRACT: A model capable of describing the flow behavior of electrorheological (ER) suspensions under different electric field strengths and over the full range of shear rates is proposed. Structural reformation in the low shear rate region is investigated where parts of a material are in an undeformed state, while aligned structures reform under the shear force. The model's predictions were compared with the experimental data of some ER fluids as well as the CCJ (Cho-Choi-Jhon) model. This simple model's predictions of suspension flow behavior with subsequent aligned structure reformation agreed well with the experimental data, both quantitatively and qualitatively. The proposed model plausibly predicted the static yield stress, whereas the CCJ model and the Bingham model predicted only the dynamic yield stress. The master curve describing the apparent viscosity was obtained by appropriate scaling both axes, which showed that a combination of dimensional analysis and flow curve analysis using the proposed model yielded a quantitatively and qualitatively precise description of ER fluid rheological behavior based on relatively few experimental measurements.

1. INTRODUCTION

Electrorheological (ER) fluids, which are suspensions of very fine particles in an electrically insulating fluid, are smart materials with structural and rheological properties that are significantly altered by 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 an applied electric field. The apparent viscosities can be reversibly changed over three or four orders of magnitude in response to an electric field at low shear rates.3–5 A typical ER fluid can quickly respond and behave as a liquid or a gel under an applied electric field, with response times on the order of milliseconds.6 The rapid reversible response of ER fluids is applicable to various electrically controlled mechanical devices that transform electrical energy into mechanical energy such as hydraulic valves, dampers, brakes, bullet-proof vests, shock absorbers, and actuators.7

In the absence of an electric field, particle-based ER fluids behave as viscous Newtonian or shear thinning fluids. Under application of a sufficiently large electric field, ER fluids show well-defined yield stresses beyond which shear thinning behavior is observed. The yield stress of an ER fluid generally increases as the applied electric field increases.2,8,9 At moderate shear rates, the typical steady-shear behavior of an ER fluids under these conditions resembles that of a Bingham-like fluid described by the expression:

\[
\tau = \tau_{dy}(E) + \eta_{pl} \dot{\gamma} \quad \tau \geq \tau_{dy}(E) \]

\[
\dot{\gamma} = 0 \quad \tau < \tau_{dy}(E) \quad (1)
\]

where \(E\) is the electric field strength, \(\tau_{dy}(E)\) is the dynamic yield stress at the field strength \(E\), \(\dot{\gamma}\) is the shear rate, and \(\eta_{pl}\) is the \(E\)-dependent plastic viscosity, which approaches the suspension viscosity at a sufficiently high shear rate.5,10 To describe the flow curves of ER fluids over a large range of shear rate and to account for the shear thinning behavior, other models, such as the De Kee-Turcotte model11 or the Hershel-Bulkley model,12 have been applied.

The dynamic yield stress, \(\tau_{dy}\), is commonly obtained by extrapolating the shear stress versus shear rate curve back to the shear stress intercept at a zero shear rate (Figure 1).8 The value obtained using this method is strongly influenced by the range of shear rates and by the rheological model selected for the extrapolation. The static yield stress, \(\tau_{sy}\), is the shear stress required to initiate shear flow in a fluid that is initially at rest (Figure 1). Thus, the dynamic yield stress (\(\tau_{dy}\)) is the yield stress for a completely broken down ER fluid by continuous shearing whereas the static yield stress (\(\tau_{sy}\)) is the yield stress for an undisrupted fluid.5 As Fossum et al. stated, \(\tau_{sy}\) measured for an ER fluid under continuous shear and under application of a large electric field can differ significantly from \(\tau_{dy}\) because
ER fluids are naturally thixotropic due to the destruction of the microstructure that resists flow-induced particle rearrangement. The static yield stress is thus an elusive property that depends on the shearing history of the sample under investigation. Previous models such as the CCJ model, the De Lee-Kee-Turcotte model, the Herschel-Bulkley model, and the Bingham model did not correctly predict the static yield stress, nor did they properly describe the fluid dynamics of the ER fluids including rupture and reformation of the fibrous or columnar structures. The static yield stress seems to be larger than the dynamic yield stress from Figure 1 based on this analysis, though reverse tendency has been reported in the case of some magnetorheological (MR) fluids. However, we contemplate that it is due to the model fitting of the stress curve. When the stress minimum due to structural change is not evident, curve fitting does not pass the stress minimum, but just bypasses it over the stress minimum. In this case, obtained dynamic yield stress value can be larger than the static yield stress because it is extrapolated from the larger value than the static yield stress, but its validity should be carefully checked. This can be another interesting subject in the future. In this report, we restrict our analysis to ER fluids case, though the model can be equally applied to MR fluids.

For the purpose of designing ER fluids, more precise knowledge of the relationships between the suspension rheological properties and such variables as the deformation rate, the applied electric field strength, and the composition are required. Even though predictive models have provided insights into the mechanisms governing ER fluid behavior, they have not been sufficiently quantitative for the purpose of design. In addition, they are not suitable for the predicting static yield stresses. To fit the ER fluid flow curves over the full range of shear rates under a high electric field strengths, Choi et al. proposed an empirical six parameter model (the CCJ model). Although this model described the overall fluid behavior, the values of the fitting parameters were not consistent with the physical phenomena, nor could the model predict the static yield stress. Another method for correlating or reducing experimental data is, therefore, desirable. In this study, we propose a simple model that describes the flow behavior of ER fluids, particularly the static yield stress \( \tau_\text{sy} \), the appearance of the minimum of shear stress versus shear rate, and the dynamics of the structural reformation. The model comprehensively describes the flow behaviors over the full range of shear rates and the parameters are associated with physical phenomena. A combination of flow curve analysis using the proposed model and dimensional analysis permitted a quantitatively and qualitatively precise description of the rheological behaviors of ER fluids using relatively few experimental measurements.

2. NEW MODELING

To describe ER fluids over a wide range of shear rates, Choi et al. proposed an empirical six parameter model (CCJ model):

\[
\tau = \frac{\tau_\gamma}{\left(1 + (t_1\dot{\gamma})^{t_2}\right)^{t_3}} + \eta_\text{fl} \left(1 + \frac{1}{(t_2\dot{\gamma})\eta_\text{fl}}\right)\dot{\gamma}
\]

(2)

In eq 2, \( t_1 \) and \( t_2 \) are time constants, \( \dot{\gamma} \) is the shear rate, \( \tau_\gamma \) is the yield stress, and \( \eta_\text{fl} \) is the viscosity at higher shear rates. The first term on the right-hand side represents the shear stress behavior at low shear rates, in which the shear stress decreases with shear rate. The second term describes the shear stress behavior at high shear rates. To simulate the stress variation as a function of the shear rate, the CCJ model incorporates the shear rate dependence into the Bingham model (the numerator of the first term on the right-hand side of eq 2) and a shear thinning viscosity term (the second term on the right-hand side of eq 2). Although this six-parameter model fits the experimental data well, its optimized parameters are sometimes inconsistent with experimental results as described further below. More importantly, the CCJ model can be misleading in analyzing the dynamics of ER fluids during structural reformation processes. Because the CCJ model is based on the Bingham fluid model, it does not include the static yield stress; rather, it predicts only the dynamic yield stress. Breaking and reforming aligned particle structures under shear deformation and an applied electric field induce some ER fluids to exhibit shear stresses that initially decrease with increasing shear rate before they begin to increase once again. This behavior depends on the applied electric field strength. The yield stress calculated by using the CCJ model is sometimes lower than the measured stress. If flow occurs, the first term on the right-hand side of eq 2 is inapplicable because it describes \( \tau_\text{sy} \) in the absence of flow; however, if the shear stress exceeds the yield stress, the fluid should be in the yielding region and flow as a Newtonian or shear thinning fluid (Figure 1). This paradox is predicted upon the application of the Bingham model, which implicitly assumes a homogeneous dispersion of particles, even though ER suspensions do not have a homogeneous structure in the low shear rate region.

Bingham fluids include an inherent discontinuity in the stress at the critical shear rate. An important feature of plastic behavior in this case is that parts of the fluid may flow whereas other part may act as solids. Papanastasiou explained that the difficulties of the Bingham model arise from the necessity of tracking randomly distributed yield material surfaces. Other models that describe ER fluid yield behavior, such as the DeeKee-Turcotte model or the Herschel-Bulkley model, suffer the same problem. To avoid the discontinuity in the flow curve due to incorporation of the yield criterion, Papanastasiou proposed a simple equation that can describe the entire yield flow curve, before and after yield:

\[
\tau(\dot{\gamma}) = \tau_\gamma [1 - \exp(-a\dot{\gamma})] + \eta \dot{\gamma}
\]

(3)

where \( \eta \) is the viscosity of the yielded material, \( \tau_\gamma \) is the yield stress, and \( a \) is the time constant. This model is suitable for both yielding and unyielding regions; thus, it can handle nonuniform stresses in a material in which part of the material may flow and the rest may act as a solid. Depending on the
exponent $a$, quick stress growth can be achieved with very small changes in the strain rate, consistent with the behavior of yielding materials.

The actual yield stress of an ER fluid under a strong electric field should be $\tau_{sy}$ because this is the stress required to initiate the shear flow in an ER fluid initially at rest and it varies with the fluid motion due to the structural realignment (Figure 1). Hence, the stress should include $\tau_{sy}$ and stress variations ($\Delta \tau_{sy}$) due to structural realignments at low shear rates, i.e., $\tau \approx \tau_{sy} - \Delta \tau_{sy}$. Considering these and Papanastaiou’s approximation, we propose a four-parameter model,

$$\tau = \tau_{sy} \left(1 - \frac{1 - \exp(-a\dot{\gamma})}{1 + (a\dot{\gamma})^a}\right) + \eta_\infty \dot{\gamma}$$

(4)

where $\tau_{sy}$ is the static yield stress, $\eta_\infty$ is the viscosity at a high shear rate, and $a$ is the time constant, the reciprocal of the critical shear rate where the rheological behavior changes. The power-law form of the shear rate in the denominator of the first term was used to describe the breaking and reformation of the aligned particle structure, similar to the von-Mises criterion for power-law form of the shear rate in the denominator of the first critical shear rate where the rheological behavior changes. Depending on the parameter values, eq 4 can describe Newtonian fluids ($\tau_{sy} = 0$), Bingham fluids ($a = 0$) and can simulate the De Kee-Turcotte ($a = 0$, $\alpha \neq 0$) and Herschel-Bulkley ($a = 0$, $\eta_\infty = m\dot{\gamma}^{\alpha-1}$) models. Because the predicted yield stress is naturally $\tau_{sy}$ at low shear rates including $\tau_{dy}$ corresponding to the absence of motion, inconsistencies with the CCJ model do not occur.

Each parameter of the model controls a particular aspect of the flow curve. Figure 2 shows the stress curves for various parameter values. The effect of time constant $a$ is shown in Figure 2a in which the other parameters are kept constant values at the optimized ones after fitting the experimental results of an ER fluid (dodecyl benzene-sulfonic acid (DBSA)-doped polyaniline dispersed in a silicone oil) under a 3.5 kV/mm electric field in Table 1. Increasing $a$ brings about a more rapid decrease in the shear stress as the structure deforms. Figure 2b shows that the recovery of the broken structure occurs more rapidly with a smaller power-law index value. Hence, the overall balance between breaking and reforming of the aligned structures is decided by these two parameters. It is obvious from these figures that the yield stress is $\tau_{sy}$ not $\tau_{dy}$. The effect of the viscosity, $\eta_\infty$, is also shown in Figure 2c. The stress reaches a minimum, then increases rapidly with the viscosity.

3. RESULTS AND DISCUSSION

This model simulates the stress variation in various ER fluids over a wide range of shear rates. The model is applied first to our previous data based on flow curves for an ER fluid (DBSA-doped PA (dodecyl benzene-sulfonic acid (DBSA)-doped polyaniline (PA) dispersed in silicone oil)). The particles were prepared by emulsion polymerization. The particles had an irregular plate-like structure with a particle size distribution in the range of 1–15 μm. Particle doping was performed to render the PA particles semiconducting. Particles were then dried under vacuum for 24 h, then 15 wt % amount was added to the silicone oil with a kinematic viscosity of 30 cS and a density of $\rho = 0.95$ g/mL. The rheological properties were measured using a Physica rheometer (MC120) with a Couette geometry and a high voltage generator. Figure 3 shows the flow curves of this ER fluid for different electric field strengths.
structures that hinder flow are formed by cohesion of the particles due to the dipolar interactions among particles under the electric field, the ER fluids exhibits a static yield stress at low shear rates that increases with the electric field. On the other hand, hydrodynamic forces tend to destroy the ER structures and promote flow at high shear rates. As the shear rate increases, the effects of the hydrodynamic forces are expected to dominate the effects of the electrostatic forces. The ER fluid is then completely broken down with no chainlike structure, and it behaves as a shear thinning fluid.9,10

The proposed model fits the flow curve very well and describes the structural changes over the full shear rate range under different applied electric field strengths. The parameters for the CCJ model and the proposed model are summarized in Table 1. As mentioned, all fitting parameters for the CCJ model show random variations because the six parameters were optimized simultaneously to minimize the total error in the optimization procedure, which identified local minima in the relative error difference. The time constants (t1 and t2) vary inconsistently with the electric field. The parameter β in the CCJ model should be between 0 and 1, because dτ/dγ ≥ 0; however, some of the simulated fitting values exceeded 1. Most importantly, the yield stress, τy, fluctuates with the electric field strength showing values smaller than the fluid shear stresses at low shear rates because it is τy, the dynamic yield stress in the absence of fluid motion. On the other hand, the proposed model provided yield stresses higher than the shear stress at low shear rates because it is the static yield stress, τy. The static yield stress increased consistently with the electric field strength, as did the plastic viscosity. The critical shear rate for achieving deformation can be defined as the inverse of the time constant a, i.e., γ = 1/a. This is the shear rate at which the stress function slope changes. This critical shear rate also increased with increasing electric field strength, indicating that the deformation behavior occurred at a higher shear rate under a stronger electric field. The viscosity, η∞, first decreased due to breaking among the aligned structures, then later increased due to the rapid reformation of the broken chains under the applied electric field. In the absence of an electric field, the viscosity was 0.18 Pa·s. The power-law index was negative when structural deformations occurred, and it decreased as the electric field strength increased indicating a more rapid decrease in the stress. The proposed model provides a consistent prediction of the flow curves over the full range of shear rates and under different electric field strengths, using fewer parameters.

For comparison, another example of an ER fluid with a different structure was simulated. Choi et al. recently reported the synthesis of anisotropic (snowman-like) particles by coating seed particles (poly(methyl methacrylate)) with a semi-conducting polyaniline layer. They then took advantage of the enhanced dielectric properties and demonstrated the particles’ utility in forming ER fluids.9 Particles with a large aspect ratio generally provide better ER effects through preferential alignment;15 however, this ER fluid exhibited weak ER effects because fewer particles are placed between two electrodes than for spherical microparticles due to the anisotropic snowman-like shape, and the polarized interactions among particles were reduced due to the asymmetric polarization density.9 Nonetheless, the fluid showed ER effects under strong electric fields. The ER fluid yields a very short relaxation time which indicates a more rapid response to an electric field. The ER fluid also exhibits shear stress decreasing at low shear rates and increasing at higher shear rates than the critical value due to the breakage and reformation of the fibrillation of ER particles under shear flow. The model fits were compared with the flow curves obtained from this data (Figure 4). The optimal parameter values for the CCJ model and the proposed model are summarized in Table 2. Both models seemed to fit the experimental data well. However, subtle differences in the behavior were observed at both low and high shear rates. The yield stress values from both models steadily increased with the electric field strengths, but stress prediction by our model converged to the yield stress at low shear rates whereas the predictions of the CCJ model continuously increased with decreasing shear rates. The stress convergence to the yield stress at low shear rates was not evident in the CCJ model. The CCJ model predictions at high shear rates deviated from the experimental data, whereas the current model prediction agreed well (Figure 4 b). Differences between the CCJ model and the current model were apparent at high shear rates due to viscosity differences. The viscosity parameters obtained using the CCJ model were 1.5 times larger than those obtained using the current model. When an electric field was not applied, the viscosity was 0.054 Pa·s. The viscosity

| Table 1. Optimal Parameters in the CCJ Model (eq 3) and the Proposed Model (eq 4) Obtained by Fitting the Models to the Flow Curves of ER Fluids (Dodecyl Benzene-Sulfonic Acid (DBSA)-Doped Polyaniline (PA)) at Applied Electric Field Strengths |
|---|---|---|---|---|
| model | electric field strength [kV/mm] | parameters | τy | η∞ | α |
| CCJ (eq 3) | 0.5 | 20 | 250 | 190 | 0.09 | 0.09 | 0.2 | 0.1 | 0.5 | 1.2 | 0.4 | 0.8 | 1.04 |
| Present model (eq 4) | 3.5 | 24.8 | 153.5 | 394 | 0.17 | 0.153 | 0.26 | 15.7 | 2.2 | 0.32 | 0.020 | −0.045 | −0.085 |

Optimal parameter values from ref 6.

Figure 3. Flow curves of the ER fluid (dodecyl benzene-sulfonic acid (DBSA)-doped polyaniline (PA)) at various electric field strengths of (●) 0 kV/mm, (△) 0.5 kV/mm, (□) 1.5 kV/mm, (○) 3.5 kV/mm (taken from ref 6). The lines are calculations according to the proposed model, eq 4. Optimal parameter values for the model are in Table 1.

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of the ER fluid under the applied electric field should be less than this value due to the shear thinning behavior of the ER fluids and the paucity of particles between electrodes. The current model showed a viscosity slightly lower than the limiting value, whereas the CCJ model overestimated the viscosity. The calculated viscosity based on the present model steadily increased with the electric field strength, whereas the CCJ model viscosity presented fluctuations. The critical shear rate for deformation is defined as the inverse of the time constant \( a_i \), i.e., \( \gamma_\text{c} = 1/a_i \), where the rheological behavior changes. This critical shear rate did not show a clear trend as the electric field strength was increased due to the weak ER effects. The proposed model afforded better prediction with fewer parameters than the CCJ model for all flow curves over the full range of shear rates and applied electric field strengths.

Another intriguing simulation was conducted for an ER fluid of the aminated chitosan which shows trembling shear behavior, as shown in Figure 5. Ko et al. ascribed the sinusoidal flow behavior to the formation of fully developed chain and lamellar structures, the destruction of the formed structure, and the flow after destruction.\(^{16}\) Figure 5 replicates this behavior into four regions. In the region I, the shear stress decreases with the shear rate due to the breakage of chains and the boundary friction force which decreases with the shear rate. In the region II, shear stress increases with the shear rate due to little influence of the boundary friction force. In region III, shear stress decreases because of the strong shear field which prevents the ordered structure formation that gives a strong shear stress. In region IV, the viscosity term is dominant over the electric field induced shear stress to show the increase of the shear stress with the shear rate.\(^{16}\) Figure 5 case resembles the previous cases, except that the stress decreasing region I is not so clear. Ko et al. analyzed this behavior using their seven parameter model, which included the oscillating structural patterns characteristic of the particles. The current model fits the shear stress variations of this trembling ER fluid very well in

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**Table 2. Optimal Parameters for Models Obtained from the Flow Curve of 13 wt % Snowman-Like Anisotropic Microparticle Based ER Fluids at Various Electric Field Strengths**

<table>
<thead>
<tr>
<th>model</th>
<th>parameter</th>
<th>0.1 kV/mm</th>
<th>0.5 kV/mm</th>
<th>1.0 kV/mm</th>
<th>1.5 kV/mm</th>
<th>2.0 kV/mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bingham</td>
<td>( \tau _y )</td>
<td>8.67</td>
<td>11.98</td>
<td>17.56</td>
<td>22.56</td>
<td>30.45</td>
</tr>
<tr>
<td></td>
<td>( \eta _{pl} )</td>
<td>0.067</td>
<td>0.065</td>
<td>0.065</td>
<td>0.060</td>
<td>0.060</td>
</tr>
<tr>
<td>CCJ</td>
<td>( \tau _y )</td>
<td>8.67</td>
<td>12.23</td>
<td>19.14</td>
<td>25.30</td>
<td>35.20</td>
</tr>
<tr>
<td></td>
<td>( \eta _{pl} )</td>
<td>0.078</td>
<td>0.081</td>
<td>0.10</td>
<td>0.080</td>
<td>0.065</td>
</tr>
<tr>
<td></td>
<td>( \alpha )</td>
<td>0.57</td>
<td>0.44</td>
<td>0.43</td>
<td>0.41</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td>( \beta )</td>
<td>0.85</td>
<td>0.87</td>
<td>0.91</td>
<td>0.68</td>
<td>0.70</td>
</tr>
<tr>
<td></td>
<td>( \gamma _1 )</td>
<td>1.21</td>
<td>1.89</td>
<td>2.50</td>
<td>1.10</td>
<td>1.30</td>
</tr>
<tr>
<td>Present model(eq 4)</td>
<td>( \gamma _1 )</td>
<td>0.016</td>
<td>0.018</td>
<td>0.019</td>
<td>0.011</td>
<td>0.012</td>
</tr>
<tr>
<td></td>
<td>( \gamma _2 )</td>
<td>8.0</td>
<td>11.9</td>
<td>15.3</td>
<td>19.3</td>
<td>23.0</td>
</tr>
<tr>
<td></td>
<td>( \alpha )</td>
<td>0.152(^b)</td>
<td>1.26</td>
<td>1.03</td>
<td>1.04</td>
<td>1.2</td>
</tr>
<tr>
<td></td>
<td>( \beta )</td>
<td>0.046</td>
<td>0.044</td>
<td>0.046</td>
<td>0.051</td>
<td>0.05</td>
</tr>
<tr>
<td>Newtonian viscosity(^c)</td>
<td>( \eta _N )</td>
<td>0.053</td>
<td>0.05</td>
<td>0.053</td>
<td>0.056</td>
<td>0.056</td>
</tr>
</tbody>
</table>

\(^a\)The Bingham and CCJ model parameter values are from ref 9. \(^b\)At 0.1 kV/mm, the shear stress did not vary noticeably at low shear rates; hence, the critical shear rate was not the value of the minimum stress but the shear rate for yielding. \(^c\)The viscosity obtained from the slope of the shear stress at high shear rates. \(^d\)The viscosity obtained from the slope of the shear stress at high shear rates.

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**Figure 4.** (a) Flow curves for ER fluids containing 13 wt % snowman-like particles (core–shell structures of PMMA/polyaniline) dispersed in silicone oil (Taken from ref 9) at electric field strengths of (○) 0.1 kV/mm, (Δ) 0.5 kV/mm, (◊) 1 kV/mm, (▽) 1.5 kV/mm, (□) 2 kV/mm. The dotted and solid lines indicate the predictions according to the CCJ model and proposed model, eq 3. Optimal parameter values for different models are in Table 2. (b) Enlarged view of (a).
Figure 5. Shear stress vs shear rate for the aminated chitosan ER fluid at an electric field strength of 3 kV mm\(^{-1}\).\(^{15}\) The solid line shows a fit using the present model. Parameter values are \(\tau_c = 443\) Pa, \(a = 0.002\), \(\eta_m = 0.3893\), \(\alpha = -2.453\). Although the flow curve was trembling, the present model fits the experimental data very well, demonstrating its flexibility and adaptability.

The regions of II, III, and IV as a function of the structural changes, though the lack of the experimental data in region I does not allow the fitting in that region. Coincidentally, the yield stress (443 Pa) approaches that of Ko et al. (476 Pa).\(^{16}\)

An accurate yield stress prediction is important because it can imply different electrical interactions. The yield stress \((\tau_y)\) of an ER fluid is generally expressed as a non-analytic power-law form in terms of the electric field strength \((E_o)\) according to \(\tau_y \propto E_o^{n}\).\(^{11,15}\) Davis and Ginder showed that \(\tau_y \propto E_o^{0.2}\) for \(E_o > E_c\) and \(\tau_y \propto E_o^{2}\) for \(E_o < E_c\), where \(E_c\) is the critical electric field strength.\(^{18}\) Thus, the variation of the yield stress with \(E\) is divided by the critical electric field strength, \(E_c\), into two regions. Table 2 lists the yield stresses obtained from the fitting of different models for the flow curve of a 13 wt % snowman-like anisotropic microparticle based ER fluid at various electric field strengths. These yield stresses are plotted versus the electric field strength in Figure 6. The Bingham fluid model predicted the second-order relationship with the electric field strength. The yield stresses obtained from the CCJ model also displayed second-order dependence, whereas the value predicted by the current model varied linearly. The values of \(\tau_y\) predicted by the CCJ model or the Bingham model were higher than \(\tau_y\) predicted by the current model (Table 2) because the CCJ model or the Bingham model simulated the stress section at low shear rates as if the stress did not vary with the low shear rate without passing through the minimum (this is evident in Figure S(a) of ref 9). The linear dependence of the static yield stress by the current model may indicate that the electrical forces between particles do not follow the relationship defined by the square of the field strength \((\tau_y \propto E^2)\).\(^{13}\) These deviations from the theoretical polarization model predictions have been observed by other researchers using different materials.\(^{15,19}\) Delgado et al. ascribed these deviations to the liquid medium’s response to the large fields to increase its conductivity.\(^{19}\) This process may be inevitable in the small gap between adjacent particles due to the large magnitude of local electric field, and it is expected mainly at the electrodes due to the charge injection.\(^{19–21}\) However, this behavior is applicable only to strong ER fluids and may not be applicable to weak ER fluids, such as this snowman-like anisotropic microparticle-based ER fluids.

If the primary forces governing the behavior of ER fluids are electrostatic polarization forces induced by an applied electric field and hydrodynamic forces caused by the particle motion relative to the continuous phase, the nondimensionalized ER fluid properties should depend only on the ratio of the electrostatic polarization force to the magnitude of the hydrodynamic force.\(^{2}\) Using dimensional analysis, Marshall et al. revealed that suitably nondimensionalized ER fluid’s properties should depend only on the magnitude of the hydrodynamic force to the ratio of the electrostatic polarization force.\(^{2,23}\) More precisely, the dipole–dipole interactions are proportional to the square of the electric field intensity, \(E^2\), whereas the shear strength acting on a particle within an ER chain is proportional to the local shear constraint, i.e., the shear rate \(\dot{\gamma}\). Hence, the normalized shear that allows for the comparison of the shearing processes to the cohesive ER structures present in different runs is \(\dot{\gamma}/E^2\), which is proportional to the Mason number \((M_n = \mu_0\dot{\gamma}/2\epsilon_0\beta E^2)\), where \(\mu_0\) is the medium viscosity, \(\beta\) is the dielectric contrast factor \(=(\epsilon_p - \epsilon_s)/(\epsilon_p + 2\epsilon_s))\), \(\epsilon_p\) is the particle dielectric constant, \(\epsilon_s\) is the dielectric constant of the liquid medium phase, and \(\epsilon_0 = 8.854 \times 10^{-14}\) F/cm is the vacuum permittivity.\(^{2}\) Thus, the apparent viscosity \(\eta (=\tau/\dot{\gamma})\) is proportional to \(M_n^{-1}\) if the volume fraction of the particle remains constant.\(^{22}\) The utility of this dimensional analysis is that the shear rate and field strength dependence of an ER fluid’s rheological properties can be described in terms of a single independent variable proportional to the Mason number (or \(\dot{\gamma}/E^2\)).\(^{10,15}\) However, aforementioned nonlinear particle polarization due to a nonspherical morphology, as well as weak ER properties, does not enable the ER flow curves shown in Figure 3 to be represented as a function of only the Mason number. Fossum et al. claimed that application of a scaling factor \(s(E)\), to the vertical axis, permits all flow curves to overlap.\(^{3,10}\) This implies that, by rescaling horizontal axis by \(E^2\) and the vertical axis by a suitable factor \(s(E)\), a master curve may be achieved. For a suitable form of \(s(E)\), Fossum et al. suggested a power-law form of the electric field strength

\[
\tau(E) = (E/E_c)^a (\dot{\gamma}/E^2)
\]  

(5)
where the functional relation $f$ represents the master curve. Once the apparent suspension viscosity is defined as $\eta_{app} = \tau / \dot{\gamma}$, it has the same functional relationship as $\tau$. After non-dimensionalization according to the viscosity at an electric field strength of zero ($\eta = 0.18$ Pa s) and plotting it as a function of $\dot{\gamma}/E^2$, the dimensionless apparent viscosities of the ER fluid of (DBSA-doped PA (dodecyl benzene-sulfonic acid (DBSA)-doped polyaniline (PA)) in Figure 3 collapse onto a single curve, as shown in Figure 7 for $\alpha = 1$. At small $\dot{\gamma}/E^2$, the log($\eta_{app}/\eta_0$)/($E/E_c$) curve has a slope of $-1.1$, whereas at large $\dot{\gamma}/E^2$, the curve smoothly approaches unity. Using the Bingham constitutive equation, Marshall et al. correlated the apparent viscosity with the Mason number having the slope of $-1$. This small difference is ascribable to the non-Bingham fluid behavior due to structural reformations as well as the nonspherical particle shapes. The data collapse onto a master curve indicating that the combination of flow curve analysis according to the proposed model and dimensional analysis enables not only qualitative but also quantitative prediction of ER fluid behavior with relatively few experimental measurements.

We applied the functional relationship between $(\eta_{app}/\eta_0)(E/E_c)$ and $(\dot{\gamma}/E^2)$ for the ER fluids of the anisotropic (snowman-like) particles. After non-dimensionalization according to the viscosity at zero electric field strength ($\eta_0 = 0.054$ Pa s) and being plotted as a function of $\dot{\gamma}/E^2$, the data collapse onto a single curve, as shown in Figure 8 for $\alpha = 1.7$. A somewhat large $\alpha$ value implicates the weak ER effect of the ER fluid. The converted data span 6 orders of magnitude along either axis and all data nicely collapse onto the master curve. Again, the collapse of data onto the master curve indicates that the combination of flow curve analysis according to the proposed model and dimensional analysis correctly predicts the ER fluid behavior with relatively few experimental measurements.

4. CONCLUSIONS

The flow behavior of ER fluids that display both static yield stress, $\tau_{sy}$, and aligned structure deformations (breaking and reformition of the formed structures) at high shear rates was modeled using a new rheological model. This model combined the nonuniform stress distribution proposed by Papanastasiou in the context of Bingham fluids with the stress variation due to aligned structure reformation. The four-parameter model, eq 4, correctly describes the phenomena underlying the flow curve, and provides a phenomenological explanation for the structural deformations associated with yielding at different stress levels. The obtained parameters correlated well with the experimental data for three different ER fluids both quantitatively and qualitatively. Most of all, the model plausibly predicted the static yield stress, $\tau_{sy}$ rather than the dynamic yield stress, $\tau_0$. The model could correctly predict variations in the parameters under an applied electric field strength. The apparent viscosity of the ER fluid could be represented by the function of $\dot{\gamma}/E^2$, which resulted in collapse of the experimental data at various electric field strengths and shear rates onto a master curve when properly rescaled with a shift factor $s(E)$. This implies that the current model affords precise determination of the rheological behavior of ER fluids over a range of electric field strengths and shear rates with the requirement for relatively few experimental measurements.

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**REFERENCES**


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