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Short communication

Rheological analysis of graphene oxide coated anisotropic PMMA microsphere based electrorheological fluid from Couette flow geometry

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ABSTRACT

Based on flow curve of graphene oxide coated polymeric microsphere based electrorheological (ER) fluid, model independent yield stress and shear rate, $\dot{\gamma}(\tau)$, were determined using a Tikhonov regularization technique developed to solve the inverse problem of the shear rate in Couette flow viscometry. The model independent yield stress data were compared with that extracted from yield stress fluid models of Bingham plastic and Cho-Choi-Jhon (CCJ) used to fit the $\dot{\gamma}(\tau)$ data. The agreement between model independent and Bingham yield stress was excellent, while the CCJ model yield stresses are generally slightly higher.

Introduction

Electrorheological (ER) fluids are suspensions of polarizable particles or composite particles in a non-aqueous solvent such as silicone oil that can undergo a liquid–solid transition in response to an applied electric field [1–6]. The liquid–solid phase transition is reversible and the time-scale of this transition process can be regarded as almost instantaneous; a few microseconds. They acquire solid-like properties when exposed to an applied electric field and become liquid-like once this field is removed. The solid-like properties can be accompanied by orders of magnitude increase in the yield stress/stiffness depending upon the strength of the applied field. The ability to control the fluid yield stress/stiffness easily and at will by electric field strength is a highly desirable property in certain applications. These smart field-responsive fluids therefore have many potential applications such as clutch, brake, vibration control fluid, microfluidic pump, haptic devices, shock absorber, dampers, biomedical engineering and others [7–11].

Currently, much of the research focus is on new materials as ER fluids. Inorganic particles including high dielectric TiO_2 and $BaTiO_3$, zeolite, mesoporous silica, and even carbon materials (such as carbon nanotubes, graphene oxide (GO)) in pure or modified state have been reported as ER candidates [12–15]. In addition, semi-conducting polymers like polyaniline (PANI), polypyrrole, poly(p-phenylene) and their derivatives with undoped or dedoping state have been suggested for ER materials due to their relatively facile synthesis, low density, and high potential ER effects [16,17]. Notably, the hybridized materials composed of inorganics or semi-conducting polymers are appealing as novel ER materials due to their enhanced chemical and physical properties.

The ER characteristic is also influenced by the particle geometry, such as particle size and morphology, structure and aspect ratio. Hong et al. [18] have reported the enhanced ER activity with increasing aspect ratio (ER activity: nanotube > nanorod > nanosphere). Recently, PANI nanofibers have been described with improved ER effects and even improved dispersion and thermal stability [19]. Compared to the traditional wrapped or intercalated composites, the core–shell structured particles possess more outstanding behaviors with well controlled core size and shell thickness [20,21].

For the ER fluid-based devices to work as designed, the relationship between the yield stress and applied electric field

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strength needs to be known precisely. This means that the yield stress needs to be determined accurately. Currently, power law models are generally used to describe the relationship between the yield stress and applied electric field strength [22,23]. The exponent value appeared to vary. A value of 2.0 was given by the polarization theory for particle–particle interactions in conventional ER fluids [1,2]. When conduction becomes important, a value of 1.5 was obtained. For another class fluid known as giant ER fluid which is described by the surface polarization layer model the value is 1.0. A value of 1.0 was also reported for an ER fluid based on starch-mesophase carbon-gas oil generally not regarded as giant ER fluid [24]. The theories describing the relationships between yield stress and electric field strength and microstructure [1,2] formed are beyond the scope of this paper. Other aspects include the many factors that affect the yield stress such as particle size, shape, thickness of coating, particulate or its composite properties, particulate concentration and the conductivity mismatch between the particles and the solvent [25–28] are also outside the scope of this paper.

In this work, we proposed a new method of determining the yield stress of ER fluids without the need to assume of a fluid model from measured torque-rotational speed data of a commercial concentric cylinder viscometer designed for ER characterization, especially applying for our previously studied graphene oxide coated polymeric microsphere based ER fluid. This technique is based on Tikhonov regularization developed primarily to compute the model independent shear rate. It was developed by Yeow et al. [29] to solve the inverse problem of the shear rate in concentric cylinder viscometry. It has been applied successfully to a range of yield stress materials such as blood [30] and liquid foods [31]. This is the first time that this technique was applied to a field-responsive smart material subjected to an applied electric field. The yield stress obtained will then be compared to that obtained from other techniques such as those extracted from the yield stress fluid models used to fit shear stress-shear rate data. Bingham plastic model is commonly used to describe the flow behavior of ER fluids [1,2].

Concentric cylinder viscometers are often used to characterize the flow behavior of ER fluids [32–36]. The instrument manufacturer often applies the small gap assumption to approximate the shear rate at each rotational speed, i.e. the rotational speed of the bob divided by the gap width, or uses the Newtonian shear rate expression. The Tikhonov regularization technique will also be applied here to determine the model independent $\dot{\gamma}(\tau)$ for an ER fluid subjected to a range of applied electric field strengths. ER fluids displayed complex yield stress behavior in the presence of an applied electric field and this technique of determining the model independent shear rate would be highly desirable for such material. The parallel plate is another geometry used to characterize the rheological properties of ER fluids [1,37,38]. Instead of the shear rate, the shear stress at the rim of the plate is a function of the fluid model. Newtonian expression for the shear stress is normally assumed by instrument. The Tikhonov regularization technique can also be used to compute the model independent shear stress [39,40]. The shear rate span across the plate can be very wide especially at high rotational speed, zero at the center of the plate to a maximum at the rim [39,40]. The concentric cylinder geometry is therefore more suitable for ER fluid flow behavior characterization due to a near constant shear rate across the gap.

In this study, Tikhonov regularization technique was employed to analyze rheological data of the graphene oxide (GO)-adsorbed snowman-like anisotropic poly(methyl methacrylate) (SPMMA) microsphere based ER fluid [41], especially focusing on flow curve and dynamic yield stress as a function of applied electric fields. In addition, we described a numerical procedure for extracting the yield stress and other rheological parameters from raw torque-rotational speed data from the rheometer used without referring to any fluid model. Real fluids may or may not conform to any of the commonly encountered rheological constitutive equations. Thus our model-free results are closer to true properties of the fluids than model-based results. Our model-free results can also be used to check the various established constitutive equations or to develop new ones. In that sense our model-free results can be regarded as the empirical data for model verification and model development. At a more fundamental level of investigation, the model-free results can used to develop links between observed flow behaviors to the physical structures and chemical properties of the constituents of the fluid. These include, for example, the size and shape distributions of the particles, surface forces between particles and their electric and magnetic properties.

Governing equations and numerical method

In the characterization of a fluid flow behavior using a concentric cylinder viscometer, a set of torque (Γ) versus rotational speed (ω) readings is obtained. The Γ is usually converted first to wall shear stress, τ. The governing equation for the concentric cylinder Couette flow used in the determination of the shear rate, $\dot{\gamma}(\tau)$, is given by:

$$\omega = \frac{1}{2}\int_{\substack{\tau_{\mathrm{Out}}\\ \text{or}\\ \tau_{\mathrm{Y}}}}^{\tau_{\mathrm{In}}} \frac{\dot{\gamma}(\tau')}{\tau'}\,d\tau' \qquad (1)$$

Solving for the unknown $\dot{\gamma}(\tau)$ using measured torque-rotational speed data without the need to assume a fluid model, is an inverse problem. Yeow and coworkers [29–31] have solved this inverse problem for $\dot{\gamma}(\tau)$ for a range of fluids including yield stress materials using the Tikhonov regularization technique.

The relationship between $\dot{\gamma}(\tau)$ and the inner and outer wall shear stresses and rotational speed data ($\tau_{\mathrm{In}1}$, $\tau_{\mathrm{Out}1}$, ω_1), ($\tau_{\mathrm{In}2}$, $\tau_{\mathrm{Out}2}$, ω_2), . . ., (τ_{InND}, τ_{OutND}, ω_{ND}) of a Couette viscometer is shown in Eq. (1) [42]. For ER fluids without yield stress the lower limit of integration is τ_{Out} and for those with yield stress, it is τ_{Out} or τ_{Y} whichever is larger. An ER fluid with a yield stress will not deform if $\tau \leq \tau_{\mathrm{Y}}$, hence

$$\dot{\gamma}(\tau_{\mathrm{Y}}) = 0 \qquad (2)$$

This equation can be used to determine τ_{Y} only if the yield stress is within the range of the wall shear stress covered by the data set, i.e. when there are one or more data points in which the ER fluid in the annular gap is partially sheared.

Following Yeow et al. [29], the following dimensionless variables

$$X = \frac{\tau - \tau_{\min}}{\tau_{\max} - \tau_{\min}}, \quad \Omega = \frac{\omega}{\omega_{\max}}, \quad f = \frac{\dot{\gamma}}{\omega_{\max}} \qquad (3)$$

are defined. Subscripts max and min denote the maximum and minimum values in the data set. In dimensionless form, the viscometry data become ($X_{\mathrm{In}1}$, $X_{\mathrm{Out}1}$, Ω_1^{M}), ($X_{\mathrm{In}2}$, $X_{\mathrm{Out}2}$, Ω_2^{M}), . . ., (X_{InND}, X_{OutND}, $\Omega_{\mathrm{ND}}^{\mathrm{M}}$). Superscript M denotes measured rotational speeds. The dimensionless equivalent of Eqs. (1) and (2) are

$$\Omega_{\mathrm{C}}^{i} = \frac{1}{2}\int_{\substack{X_{\mathrm{Out}i}\\ \text{or}\\ X_{\mathrm{Y}}}}^{X_{\mathrm{In}i}} \frac{f}{(X + X_{\mathrm{L}})}\,dX \quad i = 1, 2, 3, \ldots N_D \qquad (4)$$

$$f(X_{\mathrm{Y}}) = 0 \qquad (5)$$