Investigation of Electrorheological Properties of Kaolin Suspensions under DC Fields

Hasim Yilmaz,¹ Ummihan Yilmaz²

¹Chemistry Department, Harran University, Science Faculty, 63190 Sanliurfa, Turkey ²Chemistry Department, Gazi University, Science Faculty, 06500 Ankara, Turkey

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ABSTRACT: In this study, the electrorheological (ER) behavior of suspensions prepared from $d_{50} = 7 \ \mu m$ kaolin particulate, dispersed in insulating silicone oil (SO) medium, was investigated. ER activity of all the suspensions was observed to increase with increasing electric field strength (*E*), concentration (*c*), and decreasing shear rate ($\dot{\gamma}$). Shear stress ($\dot{\gamma}$) of kaolin suspensions increased linearly with increasing concentrations of the particles and with the applied electric field strength. The viscosity (η) of all suspen-

sions was decreased sharply with increasing shear rate and showing a typical shear thinning non-Newtonian viscoelastic behavior. It was observed that kaolin/silicone oil system studied in the present work was sensitive to high temperature. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 3798– 3802, 2007

Key words: electrorheological fluids; kaolin/silicone oil suspensions; shear stress; temperature

INTRODUCTION

Electrorheological (ER) fluids consist of dispersions of solid particulates within an insulating liquid. Application of an external electric field causes changes in their flow behavior and can completely suppress the flow until a critical shear stress is reached.¹ Important factors that influence the ER effect are electric field strength, electric field frequency, shear rate, fluid composition, temperature, and the addition of a polar promoter.² There is very wide range of potential applications for ER fluids in such areas as damping, robotics, hydraulics, couplings, and automotive. The patent literature on the subject suggests a growing interest in such devices after a period of research and assessment.³ A major limiting factor is still the need for fluids with better overall performance.

There is also a need for fluids with enhanced colloidal stability against sedimentation and sludge deposits formation.⁴ Most studies reported in the literature are on the ER activity of acrylate salts and zeolitic materials;⁵ none of these researchers has investigated the influence of colloidal stability of suspensions on ER activity. Another target is for ER fluids with long service stabilities, particularly at high temperatures and rigid environmental conditions.⁶ Among various polarizable particles for dry-base ER materials, semi-

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conducting polymers, including poly(acene quinone) radicals,⁷ polyaniline,⁸ copolyaniline,⁹ polyphenylenediamine,¹⁰ poly(2-acrylamido-2-methyl-1-propane sulfonic acid),¹¹ polyaniline nanocomposite,¹² and polyacrylonitrile/diatomite composites,¹³ poly(Li-HEMA)co-poly(4-vinyl pyridine) copolymeric salt suspensions in the SO,^{14,15} PMMA-b-PSt,¹⁶ and poly(Li-2-hydroxyethyl methacrylate)/SO system,¹⁷ have been adopted as dry-base ER fluids as a result of handling and superior physical properties.

In the present study, kaolin powder was dispersed in silicone oil with a particle concentration of 5– 30 wt %. These suspensions were evaluated as ER fluids as a part of our continuing research interest in various potential ER materials.

EXPERIMENTAL

Materials

The continuous phase was silicone oil (SO) ($\triangle = 0.97$, $\eta = 200 \text{ mPa} \cdot \text{s}$, $d = 0.965 \text{ g/cm}^3$, $\epsilon = 2.61 \text{ at } 25^{\circ}\text{C}$). Kaolin powders (dispersed phase) were kindly supplied by Omya Mining Co. (Istanbul, Turkey) (see Table I for composition). The average particle size of kaolin was 7 μ m before suspension preparation; kaolin particles and silicone oil were dried in a vacuum oven for 24 h at 150°C and 4 h at 110°C.

Preparation of suspensions

Suspensions of kaolin suspensions were prepared in silicone oil at a series of particle concentrations (5–30 wt %).

Correspondence to: H. Yilmaz (hasim@harran.edu).

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Chemical Composition of Kaolin			
SiO ₂ (%)	Al ₂ O ₃ (%)	Fe ₂ O ₃ (%)	TiO ₂ (
47.00	35.07	0.76	0.77

TADIT

Determination of sedimentation stability of kaolin suspensions

The sedimentation stability of kaolin suspensions prepared in silicone oil was determined at 25°C. Glass tubes containing the suspensions, prepared at a series of kaolin concentrations, were immersed in a constant temperature water bath; the formation of the first particle precipitates was recorded as the indication of colloidal instability (Fig. 1).

Electrorheological tests

Flow rate measurements of the suspensions were carried out between two brass electrodes. During the measurements, these electrodes were connected to an external high-voltage dc electric source (E = 0 -10 kV with 0.5-kV increments) and a voltmeter, and the electrodes were dipped into a vessel containing the kaolin suspension. The stepwise electric fields were then applied to the suspension in the direction perpendicular to the parallel plates. After a few seconds, the vessel was removed, and the flow time for complete drainage was measured, using a digital stopwatch under E = 0-kV and E $\neq 0$ -kV conditions.

To measure the ER strength of ionomeric suspensions, a Thermo-Haake RS600 Rheometer was used with a plate–plate system. The applied shear rate was varied between $\dot{\gamma}$ = 0.1–1000 s⁻¹, gap between the plates was 1.000 mm. To measure the electric field viscosity, an electric field was created in the fluid perpendicular to the plates, and the rotor was forced to rotate. The voltage used in electrorheological experiments was supplied by a 0–10 kV (with 0.5-kV increments) FUG dc external electric field generator, which enabled resistivity to be created during the experiments. All measurements were carried out at 25–125°C.

RESULTS AND DISCUSSION

Since the ER phenomena are widely attributed to the chaining of micron-sized polarizable particles, when subjected to an external electric field, flow, and ER studies are conducted to observe the viscosity change and ER response of kaolin suspensions.

Sedimentation stability

When the density of particles is not as same as that of the medium, the particles of micron order size will settle down, according to Stoke's law.¹⁸ To solve the traditional problem of particle sedimentation, several researchers have developed different solutions. Density mismatch between dispersed and continuous phase plays an important role in the sedimentation stability of an ER fluid. The sedimentation stability results obtained from the kaolin/SO system at 25°C are given in Figure 1. According to Figure 1, the kaolin/SO suspensions possess an excellent anti-sedimentation stability. The sedimentation ratio is about 67-87% within 10 days. Less than 67% oil of total volume fluid could be observed after 10 days, and even for longer periods. It is obvious that the kaolin/SO suspensions do not deposit even when they are static for more than 10 days. It was observed that as the particle concentration of the suspensions decrease, their colloidal stabilities increase. These expected results are in accordance with the earlier studies reported in the literature.^{19,20}

Flow measurements

To observe the effect of dc electric field on the ER activity; flow rate measurements are carried out on the kaolin/silicone oil suspensions at several concentrations (c = 5-30%, m/m). Flow times were measured under E = 0.0 kV and $E \neq 0.0$ -kV conditions. The results obtained are depicted in Figure 2. As reflected in the graph, the flow times of the suspensions showed little increase with increasing electric field strength up to 1.5 kV/mm and sharp increases for several concentrations studied, after the threshold energies were supplied. Maximum flow times were



Figure 1 Change of sedimentation stability ratio of kaolin/ silicone oil suspensions. $T = 25^{\circ}C$; \blacklozenge , 5%; \diamondsuit , 10%; \blacktriangle , 15%; \bigtriangleup , 20%; \blacksquare , 25%; \Box , 30%.

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Figure 2 Effect of electric field strength on flow time. ♦,

5%; ◊, 10%; ▲, 15%; △, 20%; ■, 25%; □, 30%.

observed to be 184 s for kaolin particles c = 30%, respectively. Flow times given for the suspensions are the maximum flow times, which could be observed under the applied field. When *E* was further increased, stronger bridge formation was occurred for all the suspensions and no flow was observed. Similar behavior was reported by Unal and Yilmaz²¹ for sepiolite/silicone oil suspensions.

Rheometry

Change in shear stress and electric field viscosity with concentration

The change in electric field viscosity and shear stress with suspension concentration of kaolin/SO system is shown in Figure 3. As reflected in the graph, electric field viscosity increases with increasing suspension concentration. The maximum electric field viscosity was observed as 264 Pas for 30% particle concentrations (m/m %) at 1.0 s⁻¹ constant shear rate. As a result of polarization forces acting between the kaolin particles, electric field viscosity (or ER activity) was observed to increase with increasing kaolin concentration in the suspensions. Similar behavior was reported by Unal and Yilmaz¹¹ for PAMPS/silicone oil suspensions.

The effect of suspension concentrations on the shear stress was evaluated by changing the concentrations of suspensions in c = 5-30% range (Fig. 3). It is clear that the shear stress increase is directly related to the suspension concentrations. The change of maximum shear stress was obtained in kaolin/silicone oil system as 2.46 kPa for 30% (m/m %) particle concentrations.

Increase in the shear stress with increasing concentration may be attributed to the increased interparticle interactions with raising particle concentration in the suspension, which results with an enhanced ER activity. When electric field was applied to the suspension, polarization forces caused aggregation of the particles and a chain formation between the upper and lower plates occurred. The relation of the magnitude of viscous forces (F), with viscosity of suspension (η_s), the average shear rate ($\dot{\gamma}$) and radius of particle (r) can be written as follows:²²

$$F = 6\pi \eta_s r^2 \dot{\gamma} \tag{1}$$

As reflected in eq. (1), increased suspension concentration will decrease the distance between kaolin particles, resulting in increased polarization force. As the suspension concentration increases above 30%, a substantial reduction in relative viscosity was observed. That is, 30% is the optimal concentration for achieving maximum ER activity in the kaolin/SO system. Similar behavior was reported by Yilmaz¹⁶ for PMMA-*b*-PSt copolymer/silicone oil suspensions.

Effect of electric field strength on viscosity and shear stress

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Figure 4 shows the change in viscosity and shear stress with electric field strength for kaolin/silicone oil system. As reflected in the graph, the increased ER activity of the suspensions is directly proportional to the increase in the applied electric field strength. The higher kaolin concentration results in a denser particle structure organized in the electric field with a higher



Figure 3 Effect of concentration on shear stress and viscosity. $T = 25^{\circ}C$, $\dot{\gamma} = 1.0 \text{ s}^{-1}$; \blacksquare , shear stress; \blacktriangle , viscosity.





Figure 4 Change of shear stress and viscosity with electric field strength. $\dot{\gamma}$ = 1.0 s⁻¹; T = 25°C; *c* = 30%, **I**, shear stress; **A**, viscosity.

resistance against flow. Also, as a result of polarization forces acting between the kaolin particles, electric field viscosity (or ER activity) increases with increasing kaolin concentration in the suspensions.

When kaolin/silicone oil suspensions were subjected to an external applied electric field, a fibrillar structure was formed across the direction of shearing force, which leads to increased viscosity of suspensions. Similar behavior was reported by Yilmaz²³ for diatomite/silicone oil suspensions.

Change in shear stress and viscosity with shear rate

The effect of shear rate on shear stress and viscosity was studied at constant electric field strengths (E = 2.0 kV/mm); the results obtained are shown in Figure 5. The electric field-induced viscosity of suspensions was found to decrease sharply with increasing shear rate, giving a typical curve of shear thinning non-Newtonian viscoelastic behavior. As reflected in the graph, shear stress was increased with increasing shear rate and shown also typically non-Newtonian behavior. These characteristic behaviors of the ER suspension are related to the internal particle structure induced by an applied external electric field. Before shearing the ER fluid, the dispersed particles are aligned through the electric field direction, making columnar structures, and these structures become stronger at higher electric fields. Similar results were reported for the studies of poly(lithium-2-acrylamido-2-methyl propane sulfonic acid),¹¹ sepiolite,²¹ poly (naphthalene quinone radical,²⁴ cellulose,²⁵ PANI,²⁶ calcium carbonate,²² and mesoporous molecular sieve,²⁷ in which silicone oil was used as continuous phase for the suspensions.



Figure 5 Effect of shear rate on ER activity; $T = 25^{\circ}C$; E = 2.0 kV/mm; \blacksquare , shear stress, \blacktriangle , viscosity.

Effect of temperature on viscosity

Change of viscosity and shear stress of suspensions with temperature (Fig. 6) was investigated; it was observed that the viscosity and shear stress of suspensions sharply decrease with increasing temperature, yielding a typical curve of shear thinning non-Newtonian viscoelastic behavior. For this type of ER fluid, the ER effect is due to the polarization of mobile solid particles subjected to external electric field. Since the polarizability of the material is temperature dependent, the shear stress of ER fluid is influenced by the environment's temperature. As reflected from the



Figure 6 Change of shear stress with temperature. $\dot{\gamma} = 1.0 \text{ s}^{-1}$; E = 2.0 kV/mm; c = 20%; \blacksquare , shear stress; \blacktriangle , viscosity.

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graph, we obtained a high shear stress ($\tau = 1.12$ kPa) for c = 30% kaolin/SO system at T = 25°C, and this τ value decreased sharply with raising temperature, reaching $\tau = 384$ Pa at T = 125°C. The loss at the τ of kaolin/SO system for $\Delta T = 100$ °C temperature change is approximately $\Delta \tau = 0.74$ kPa, which is extremely high τ loss in terms of potential high temperature industrial applications.

The temperature effect of ER fluid is one of the important parameters to evaluate ER effect.²⁸ Generally, the temperature has two effects on the ER fluid: one is the effect on the polarization intensity of particle and another is Brownian motion. The increase of the temperature results in a decrease in the activation energy polarized, but in increased polarization ability of the particle. On the other hand, Brownian motion does not contribute to chain formation by kaolin particles. As a consequence, the ER effect decreases while the leaking current density increases (< 35 μ A/cm²). Yanju et al.²⁹ reported similar shear stress loss behavior for the inorganic/polymer blend in silicone oil. Similar behavior was reported by Unal et al.¹⁷ for poly (HEMA)/silicone oil suspensions.

CONCLUSIONS

This study was conducted to investigate the ER behavior of kaolin/silicone oil suspensions, and the following conclusions were drawn: Colloidal stability of kaolin/silicone oil system was determined to be 10 days for $d = 7.0 \,\mu\text{m}$ at 5 wt % suspension concentration. ER activity of all the suspensions observed to increase with increasing electric field strength and concentration, decreasing shear rates. Shear stress of kaolin/SO suspensions were increased linearly with increasing electric field strength and concentration; $\tau = 2.64$ -kPa high excess shear stress was obtained. The electric field viscosity of all the suspensions decreased sharply with increasing shear rate and showing a typical non-Newtonian viscoelastic behavior. The kaolin/

SO system was found to be slightly sensitive to high temperature.

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