Electrorheological properties of pumice/silicone oil suspension

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Abstract Pumice particles-based electrorheological (ER) suspensions were prepared in silicone oil and its ER behavior was investigated as a function of shear rate, electric field strength, concentration and temperature. Sedimentation stabilities of suspensions were determined. ER activity of all the suspensions was observed to increase with increasing electric field strength, concentration and decreasing shear rate. Yield stress of pumice suspensions increased linearly with increasing applied electric field strength and with concentrations of the particles. The pumice suspensions show a typical shear thinning non-Newtonian viscoelastic behavior, in which viscosity of suspension decreased sharply with increasing shear rate. Effect of high temperature onto ER activity of pumice/silicone oil system was also investigated.

Introduction

An electrorheological fluid is typically a suspension of a solid (conductive or polarizable particle) or liquid material (liquid crystal) dispersed in insulating oil. The increase in the viscosity of suspensions upon application of an electric field is commonly known as electrorheological effect or Winslow effect [1]. The rheological properties (viscosity, yield stress, etc.) of an ER suspension can reversibly change in several orders

of magnitude under an electric field of several kilovolts per millimeter. Its mechanical properties can be easily controlled within a wide range almost from pure liquid to solid. The ER fluid can be used as an electrical and mechanical media in various industrial areas. Their wide potential stimulated a great deal of interests both in academic and industrial areas [2]. Most suspensions require additives such as surfactants or polar liquid (promoter), which are called wet (hydrous) ER fluids. Additives are added to improve sedimentation stability of dispersed particles also enhance ER activity [3].

A wide variety of particulate materials have been selected to prepare ER suspensions. They are: zeolite [4], silica [5], sepiolite [6], Chitosan [7, 8], phosphate cellulose [9], carboneus particle [10] and conducting polymer [11].

These materials have been tested as anhydrous ER fluid particles. On the other hand, the continuous phase including silicone or hydrocarbon oils posses' low conductivity and large dielectric breakdown strength.

The well-known electrostatic polarization model incorporates the field-induced polarization of the dispersed phase particles relative to the continuous phase [12], in which the driving force of the particle fibrillation originates mainly from the electrostatic interaction among the particles. In addition, it plays a crucial role on the dielectric mismatch between the dispersed and continuous phase, which causes this interaction [2–13].

Not many ER devices have been commercialized because of a number of limitations that must be solved, such as; their yield stress is not high enough, they have limited temperature range of use, they present problems of suspension's stability against sedimentation, and once contaminated may provoke abrasion of pipe or container, and others [14].

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In this study, pumice powder was dispersed in silicone oil with a particle concentration of 5–25 (%, m/m), and 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 (ρ = 0.965 g/cm^3, η = 200 mPa s, ε = 2.61 at 25 °C). Dispersed phase, pumice was kindly supplied by POMZAMER (Süleyman Demirel University pumice research and application center) and its chemical composition is given in Table 1.

Determination of particle size

The particle size of pumice sample was determined by Fraunhafer scattering using a Malvern Mastersizer E, version 1.2b particle size analyzer. The pumice sample was dispersed in distilled water and stirred at a constant temperature of 25 °C. The data collected was evaluated by the Malvern software computer according to Fraunhafer diffraction theory [15]. Average particle diameters of the pumice particles were determined as 53 μm.

Preparation of suspensions

Prior to suspension preparation, pumice was treated with 1 M HCl(aq), then washed with distilled water, and pumice particles and silicone oil were dried in a vacuum oven for 24 h at 150 °C and 4 h at 110 °C, respectively. Different volume fractions (5, 10, 15, 20, 25 %, m/m) were prepared with silicone oil.

Determination of sedimentation stability

The colloidal stability of suspensions against sedimentation was determined at constant temperature (25 °C). Glass tubes containing the suspensions were immersed into a constant temperature water bath and formation of first precipitates was taken to be the indication of colloidal instability.

Table 1 Chemical composition of pumice

Compound	SiO$_2$	Al$_2$O$_3$	Na$_2$O	K$_2$O	Other
Composition (%)	74.0	15.6	6.1	2.4	1.9

Rheological measurements

Suspensions of pumice in silicone oil were mechanically stirred before each measurement against sedimentation. Rheological properties of the suspensions were determined with a Termo-Haake RS600 parallel plate Electro-rheometer (Germany). The gap between the parallel plates was 1.0 mm and the diameters of the upper and lower plates were 35 mm. All the experiments were carried out at various temperatures (25–125 °C, with 25 °C increments). The voltage used in these experiments was also supplied by a 0–12.5 kV (with 0.5 kV increments) dc electric field generator (Fug Electronics, HCL 14, Germany), which enabled resistivity to be created during the experiments.

Result and discussion

Sedimentation stability

When the density of particles is not the same as that of medium, the particles with micron size settle down according to Stoke's law [16]. In order to solve the traditional problem of particle sedimentation, several works have developed different solutions [17]. Density mismatch between dispersed phase and continuous phase plays an important role in sedimentation ratio of the ER fluid [18]. Figure 1 shows the sedimentation ratio in time of the ER fluids. The highest ratio is about 78% in 30 days at 5 (%, m/m) concentration, which is satisfactory and meet, likely the commercial requirements. It was observed that as the particle concentration of suspensions decreases, their sedimentation
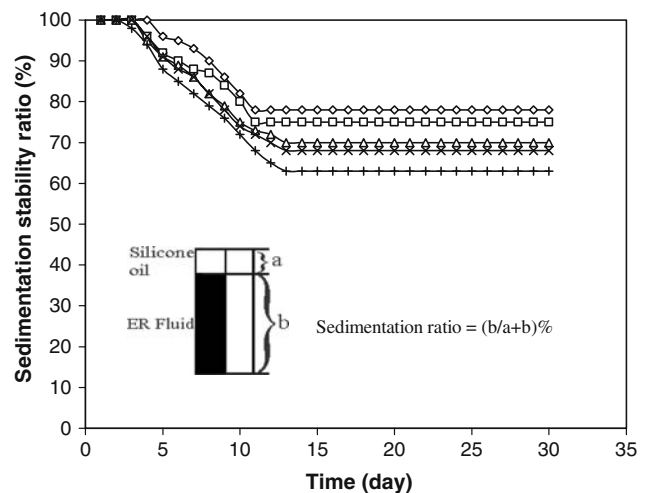

Fig. 1 Sedimentation ratio of the pumice suspension vs. time. c = (◇) 5%, (□) 10% , (△) 15%, (×) 20%, (+) 25%

stability increases. Similar results were reported by Zhao [19] Dong [20] and Wu [21] for organic/inorganic colloidal hybrid, FCCS and BaTiO$_3$ in silicone oil, respectively.

Electrorheological properties

Optimum particle concentration

Figure 2 shows the change in electric field viscosity ($\eta_{E \neq 0}/\eta_{E = 0}$) with suspension concentration at various shear rates (1–20 s^{-1}). Suspension concentration exerts principle effect on the ER effect.

At a concentration below 15%, the viscosity ratio increases with increasing particle concentration. This trend may be understood by considering the effects on polarization forces between particles. In dilute suspensions (large distance between particles), the magnitude of this polarization force in the direction of the applied electric field (E) is [22].

$$F = 6\varepsilon_2 r^6 E^2 / \rho^4 \tag{1}$$

where ε_2 is the dielectric constant of the particle, ρ is the distance between particles, and r is the radius of the particle.

As shown by this equation, an increased suspension concentration will decrease the distance between the particles, which will result in an increased polarization force. When the particle concentration is above 15%, viscosity ratio decreases with increasing particle concentration. Also, t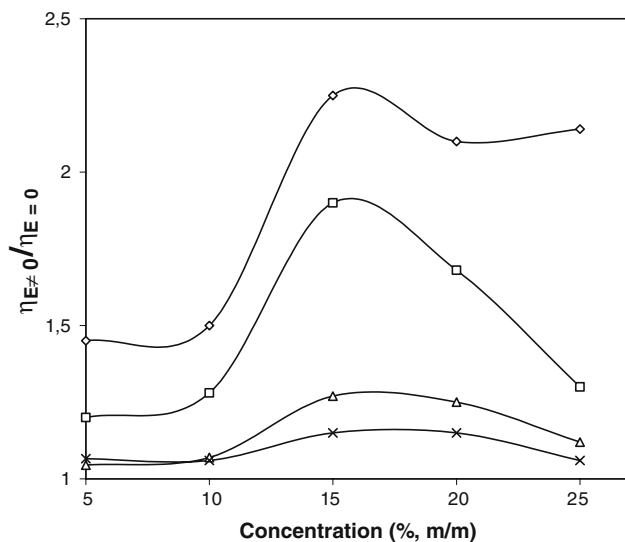he ratio of apparent viscosity to zero-field viscosity reaches its maximum at about 15% concentration. This is because, at higher suspension concentrations, particles are close to each other and the electric double layers around particles overlap. Therefore, the mutual action between particles increases and the electric double layers may drop out of particles. As a result, electric field viscosity of suspension, in other words ER effect, decreases. The ER effect was also observed to increase with decreasing shear rate. Wu and Shen [7] reported an 11% optimum concentration in the ER studies of chitin and chitosan suspensions in silicone oil, Kordonsky et al. [23] reported a 20% optimum concentration in the ER studies of carboxy methylcellulose suspensions in transformer oil, and Langelova et al. [24] reported 10–15% optimum concentration in the ER studies of polyaniline suspensions in silicone oil.

Effect of electric field and shear rate on viscosity

Figure 3 shows the change in viscosity of the suspension with electric field strength at constant concentration (c = 15%, m/m) and various shear rates. There are several important features seen in this figure, which are almost common in all particle concentrations studied. First, the effect of the field strength is very significant at low shear rates and the increment of the ER effect is smaller at higher shear rates. Second, suspension's ER effect also increases with the increasing field strength. Third, the suspension's ER effect decreases with the increased shear rate. Under an applied electric field strength, the magnitude of the interparticle interaction (polarization forces) between particles increases, and this in turn, increases the chain length (formed by particles), thus resulting in the enhancement of the viscosity. Under an applied shear field, the particles are
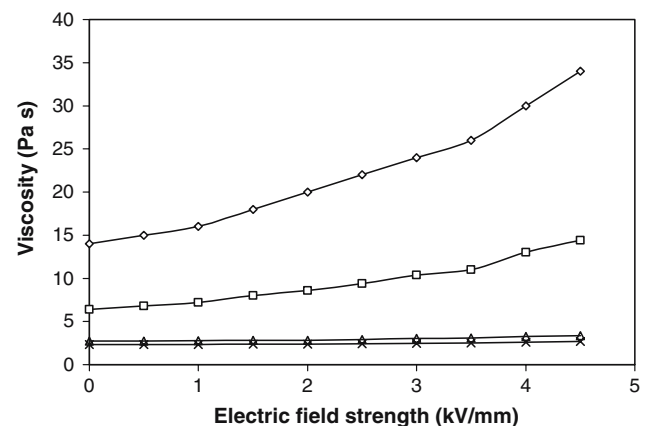
Fig. 2 The change in viscosity with concentration. T = 20°C, E = 0 V/mm and 0.5 kV/mm, γ (s^{-1}). = ($\diamond$)1 , ($\square$) 2, ($\triangle$) 10%, ($\times$) 20

Fig. 3 The change of viscosity with electric field strength. T = 20°C, c = 15 (%, m/m), γ (s^{-1}): ($\diamond$) 0.4, ($\square$) 1, ($\triangle$) 4, ($\times$) 20

also acted on by the viscous forces, which are modulated by hydrodynamic interactions with other particles in the suspensions. The viscous forces scale as:

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

where η_s is the viscosity of suspension, r the radius of the particle, and γ the shear rate.

At higher shear rates the suspension viscosity almost becomes independent of the applied electric field strength. It may be concluded that, at higher shear rates, the viscous forces are dominant, and the fibrillar structure of the suspension does not vary appreciably with field strength.

A similar trend was observed by Dürrschmith and Hoffmann [25] in ER studies of saponite suspensions prepared in silicone oil and by Pavlinek et al. [26] in ER studies of poly(glycidyl methacrylate) suspensions prepared in silicone oil.

Change in yield stress with electric field strength

The yield stress (τ_y) is one of the critical design parameters in the electrorheological (ER) phenomenon and has attracted considerable attention both experimentally and theoretically [27]. τ_y is plotted as a function of the field strength E at optimum particle concentration (15 %, m/m) and shear rate ($\gamma = 20$ s^{-1}) and temperature (20 °C) in Fig. 4. The yield stress values, which represent the limiting values of the shear stress as the shear rate approaches to zero shear rate, were obtained by extrapolating the shear stress-shear rate data to the zero shear rate. The phenomenon of the yield stress in the ER suspension has been commonly represented by the Bingham constitutive equation [28]:

$$\tau = \tau_y + \eta_0 \gamma \tag{3}$$

where τ_y is the yield stress, and η_0 the zero-electric field viscosity. As shown in Fig. 4 the yield stress increases with the applied electric field. This suggests that the system structure in ER suspensions is more stable in a strong electric field. At high field strength, the increase of the yield stress becomes less significant. This is due to the saturation of the particle structure with the electric field. It is observed, in Fig. 4 that the yield stress greatly depends on the electric field. For the 15% suspension in silicone oil, the yield stress reaches up to 78 Pa, under 4.5 kV/mm, respectively, suggesting that the ER fluid is solidified under external electric field and the degree of solidification increases with electric field. It is clearly illustrated in Fig. 4 that the yield stress linearly increases
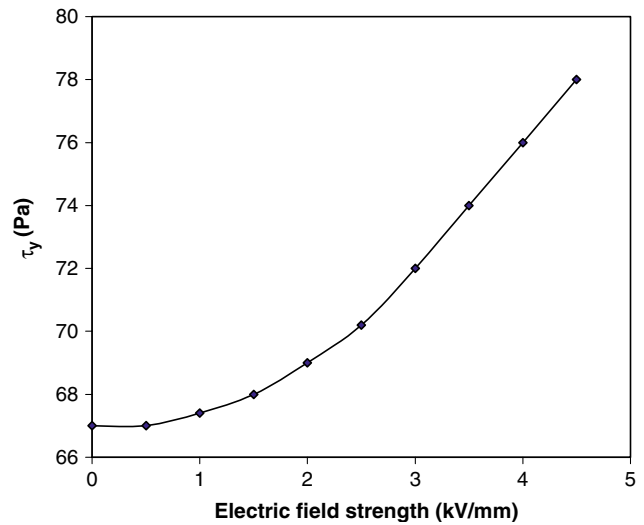

Fig. 4 The change of yield stress with electric field strength. c = 15 %, $\dot{\gamma} = 20$ s^{-1}, T = 20 °C

with the squared electric field, in good agreement with the studies carried out before [29].

Change in yield stress with concentration

Figure 5 shows a parabolic dependence between yield stress and concentration, and describes adequately the variation of the yield stress with the suspension concentration. Such dependence is a result also found by other authors [14–31], but disagrees with the commonly admitted linear relationship between τ_y and concentration (c). In fact, according to the
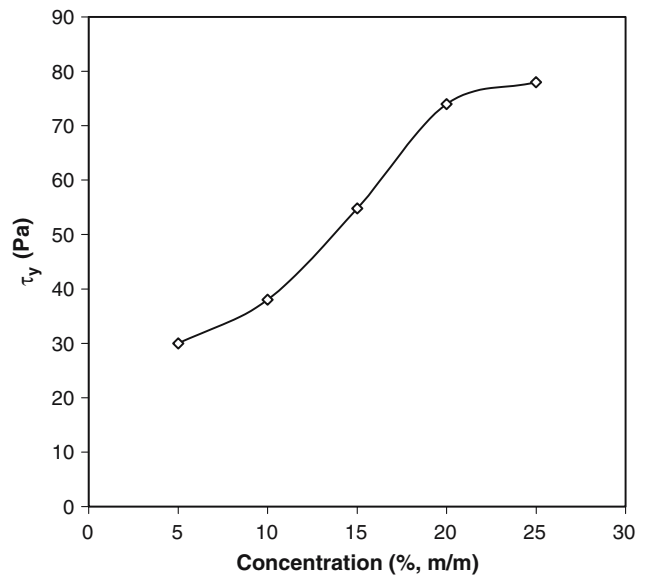

Fig. 5 The change of yield stress with concentration. $\dot{\gamma} = 20$ s^{-1}, T = 20 °C, E = 2.5 kV/mm

polarization and conduction theories of the ER effect, the yield stress should be proportional to the particle concentration. Such dependence has been found, particularly over a wide concentration range [32], but other exponents are possible in a τ_y and c^n relationship: n is believed to be less than one at low and high concentrations, and > 1 at intermediate concentration values [14–30]. This spread is probably related to changes in structure with the concentration of particles [33].

Change in viscosity with shear rate

In Fig. 6, the viscosity is plotted versus shear rate for a sample containing 15% of pumice in silicone oil. With and without applied E, the viscosity of the suspension decreases with an increasing shear rate. The sample demonsrates shear-thinning non-Newtonian viscolelastic behavior. Similar results were reported for the studies of polyisoprene-co-poly(tert-butyl methacrylate) [34], sepiolite [35], molecular sieve [36], poly(naphtalene quinone) radical [37], and polyaniline [38], in which silicone oil was used as continuous phase for the suspensions.

Effect of temperature

Figure 7 shows the changes in the shear stress of pumice/silicone oil suspensions under different temperatures at optimum concentration (15% m/m). There are two reasons that temperature might change the ER effect substantially. The first is that temperature can change the polarizability of the ER suspension. The
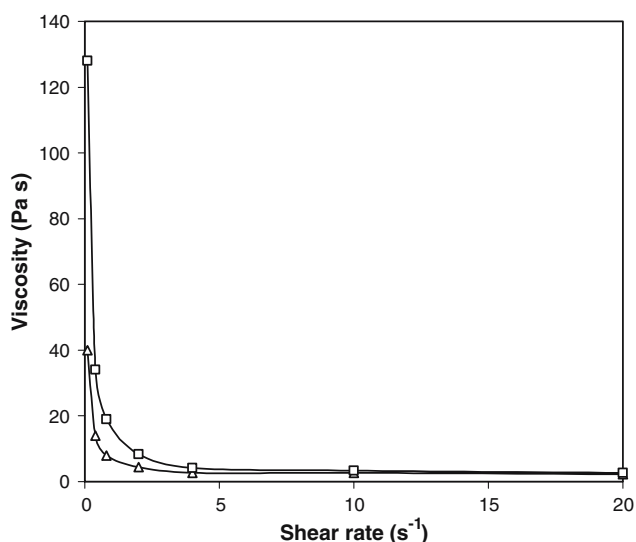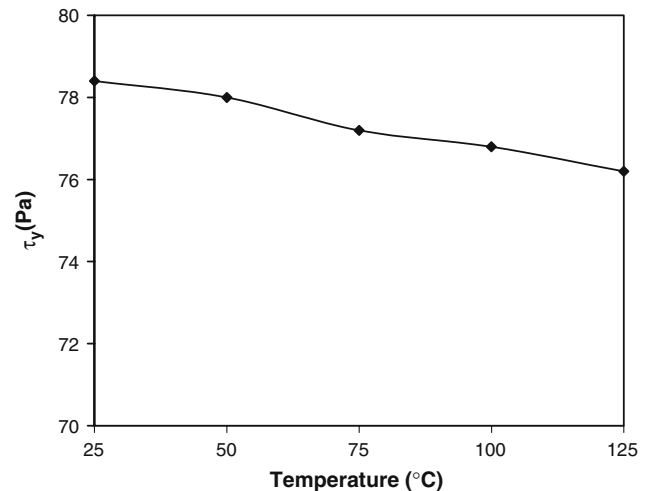
Fig. 7 The change in yield stress with temperature. c = 15 (%, m/m), E = 2.5 kV/mm

second is that temperature can directly impact particle thermal motion. If the Brownian motion could be intensified at high temperature and could become strong enough to compete with the particle fibrillation, the ER effect would become weak [39]. Similar results were reported by other researchers [34–40].

Conclusion

This study was conducted to investigate the ER behavior of pumice/silicone oil suspensions, and the following conclusions were drawn. Sedimentation stability of pumice/silicone oil system was determined as 75 percentage 30 days for c = 5% suspension concentration. ER activity of all the suspensions was observed to increase with increasing electric field strength, concentration and decreasing shear rate. Yield stress of pumice/silicone oil suspensions was increased linearly with increasing electric field strength and concentration, and τ_y = 78 Pa yield shear stress was obtained. The electric field viscosity of all the suspensions was decreased sharply with increasing shear rate, thus showing a typical non-Newtonian viscoelastic behavior. Pumice/silicone oil system was observed to be slightly sensitive to high temperature.

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Fig. 6 The change in viscosity with shear rate (△) E = 0, (□) E = 2.0 kV/mm, c = 15 (%, m/m), T = 20 °C

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