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AN INTERFACIAL POLARIZATION MODEL FOR ACTIVATED ELECTRORHEOLOGICAL SUSPENSIONS

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Abstract – Surfactant influences the ER response in two different ways. At low surfactant concentrations, it enhances the ER response by enhancing the particle polarizability. While at large concentrations, the response degrades due to the non-linear conductivity in the continuous phase. The yield stress is proportional to the electric field strength squared at small surfactant concentrations, but increases more slowly with field strength at large concentrations. In this paper, an interfacial polarization model is introduced to predict the ER behavior of surfactant-activated ER suspensions. Maxwell-Wagner model was modified by incorporating the effects of surfactant adsorption and field-induced alteration of the surfactant structure. The modified interfacial polarization model predicts well the qualitative behavior of the surfactant activated ER suspensions over all surfactant concentration ranges.

Key words: *Electrorheology, Colloid, Surfactants*

INTRODUCTION

The electrorheological (ER) response is defined as the dramatic change in rheological properties of a suspension of small particles due to the application of a large electric field transverse to the direction of flow. ER suspensions are typically composed of nonconducting or semiconducting particles dispersed in a nonconducting continuous phase. The observation of a large ER effect was first reported by Winslow in 1949. Although many ER devices, such as dampers, clutches, and adaptive structures, have been brought successfully to the prototype stage, and despite much industrial activity in the U.S. and abroad, there are currently no commercially available devices. The main limitation of ER technology development is a lack of effective fluids [Weiss and Carlson, 1993]. Our inability to design effective fluids stems largely from a lack of a fundamental understanding of the mechanisms that control ER behavior.

Surfactants are added to ER suspensions for a variety of reasons [Deinega and Vingradov, 1984; Block and Kelly, 1988; Gast and Zukoski, 1989; Jordan and Shaw, 1989; Petrzhik et al., 1980; Trapeznikov et al., 1981; Chertkova et al., 1982; Kim, 1996], and can be used to tailor suspension properties. They are often used to promote colloidal stability, which is necessary to keep particles from irreversibly flocculating, and to control rheological properties in the absence of the electric field. Surfactants are also used to "activate" suspensions. Some suspensions display little or no ER activity unless a small amount of water or surfactant is added, while other suspensions exhibit a significantly enhanced response with activators present [Petrzhik et al., 1980; Trapeznikov et al., 1981; Kim, 1996]. Enhancing ER activity with surfactants offers advantages over other approaches, such as adding water which severely limits the allowable temperature range of operation, promotes corrosion, and also increases suspension conductivity and power consumption. Furthermore, additional independent variables (i.e., type and amount of surfactants) give flexibility to designing desired properties that is not possible by simply varying the materials of the disperse and continuous phases.

Surfactant influences the ER response in two different ways [Kim, 1996]. At low surfactant concentrations, it enhances the ER response by enhancing the particle polarizability. While at large concentrations, the response degrades due to the non-linear conductivity in the continuous phase. The dynamic yield stress initially increases with surfactant concentration, passes through a maximum, then decreases at large concentrations. The yield stress is proportional to the electric field strength squared at small surfactant concentrations, but increases more slowly with field strength at large concentrations. The surfactant concentration at the maximum yield stress is insensitive to the water content, particle type, temperature, and oil viscosity, but they depend on the surfactant type and particle surface area [Kim, 1996].

In this paper, a model based on Maxwell-Wagner polarization of the disperse phase is developed by introducing the effect of surfactant adsorption and non-linear conduction between particles. This modified Maxwell-Wagner model qualitatively reproduces both the rheological and dielectric data well.

ER PROPERTIES OF SURFACTANTS-ACTIVATED SUSPENSIONS

A typical dependence of the yield stress on the electric field strength is depicted in Fig. 1. Here, the yield stress divided by the field strength squared is plotted against Brij®30 concentration (f_E=500 Hz). At small Brij®30 concentrations, the curves

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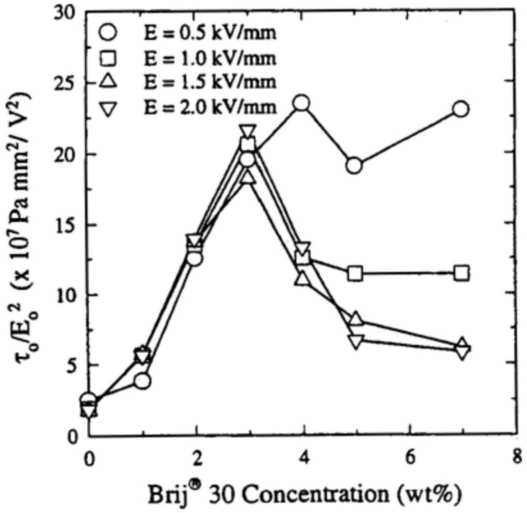

Fig. 1. Yield stress divided by the electric field strength squared as a function of Brij®30 concentration for 20 wt% dried neutral alumina suspensions in silicone oil.

at different electric field strengths superpose and the yield stress scales with the electric field strength squared (linear region). However, at larger Brij®30 concentrations (>3 wt%), the yield stress deviates from the field squared dependence, increasing approximately linearly with field strength (non-linear region).

In the linear region, the ER enhancement arises from the interfacial polarization. The yield stress increases with surfactant concentration and is proportional to the field strength squared. The electrostatic polarization model [Klingenberg and Zukoski, 1990; Klingenberg et al., 1989, 1991; Bonnecaze and Brady, 1992] describes the yield stress in terms of the electrostatic force acting between particles, induced by the applied electric field-this force increases quadratically with the particle polarizability for weakly polarizable particles. Surfactants readily adsorb to the particles and, together with adsorbed water, plays a role to increase the surface conductivity on the particles [Kim, 1996]. As a result, interfacial polarization would be enhanced, enhancing the ER response.

The non-linear ER behavior, at large surfactant concentrations, arises from the nonlinear, nonhomogeneous conduction due to the formation of surfactant-rich bridges between particles [Kim, 1996]. The dc-conductivity of Brij®30 solutions in SF96 as a function of electric field strength is presented in Fig. 2 for the surfactant solutions of 0 wt%, 0.5 wt%, 1 wt%, and 7 wt% Brij®30, respectively. The dc-conductivities of 0 wt% and 0.5 wt% Brij®30 solutions are independent of the applied electric field up to 2.0 kV/mm. For 1 wt% Brij®30 solution, the conductivity is constant up to 1.5 kV/mm and then starts to increase with the electric field above 1.5 kV/mm. The conductivity of 3 wt% Brij®30 solution starts to increase from low electric field and then reaches a new steady state-in this region, the conductivity is constant regardless of the applied electric field. This result shows that the conductivity of Brij®30 solutions is a function of the applied electric field, showing two critical electric fields where the conductivity behavior changes. The conductivity is constant up to the first critical electric field and then starts to increase with the electric field strength. The conductivity reaches a new steady state at the second critical electric field and then remains constant with further increase in the electric field strength.

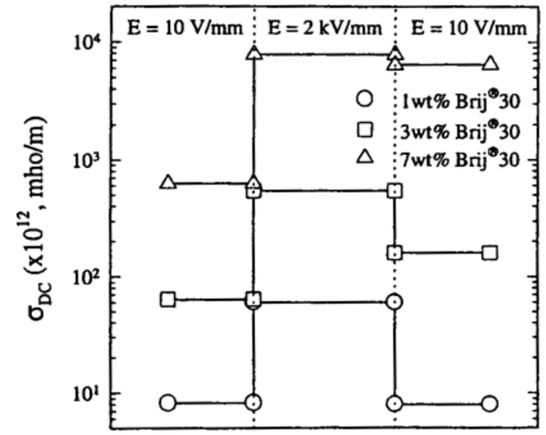

Fig. 2. DC-conductivity as a function of dc-electric field for 0 wt%, 0.5 wt%, 1 wt%, and 7 wt% Brij®30 solutions in SF96.

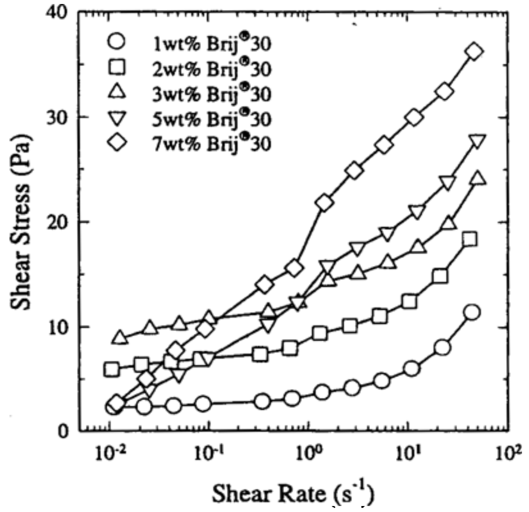

Fig. 3. Shear stress as a function of shear rate for 20 wt% dried neutral alumina suspensions with varying Brij®30 concentration (E=2.0 kV/mm and f_E=500 Hz).

This non-linear conductivity behavior arises from the dependence of the surfactant strand formation on the applied electric field [Kim, 1996]. The critical electric fields shift to higher electric field as surfactant concentration decreases. Therefore, the ER response of low surfactant concentration shows linear behavior because the critical electric field is very large, while the ER response of high surfactant concentration shows non-linear behavior because the critical electric fields is low-allowing the non-linear conductivity behavior, consistent with the rheological data [Kim, 1996].

The dependence of shear stress on shear rate for 20 wt% dried neutral alumina suspensions with various Brij®30 concentrations is presented in Fig. 3. The shear stress of suspensions with higher surfactant concentrations is larger than that with lower surfactant concentrations at high shear rate. Here, the shear stress is proportional to E_o^2, indicating that the ER response is linear and that enhanced interfacial polarization is active at higher shear rates. However, the shear stress for large surfactant concentrations decreases rapidly with decreasing shear rate, below that for suspensions with smaller surfactant concentrations at very small shear rates. Here, the response is non-linear at large surfactant concentrations. Thus, the mechanism producing non-linear behavior is only active at small shear rates.

To explain the ER behavior of nonionic surfactant-activated suspensions, we selected an interfacial polarization model and modified it by incorporating the observations: (1) the effect of surfactant adsorption to the particle polarizability and (2) the non-linear conductivity in the continuous phase. The Maxwell-Wagner model is selected as an interfacial polarization model because of its simplicity and modified as follows.

MODIFIED MAXWELL-WAGNER MODEL

1. Maxwell-Wagner Model

The simplest treatment of frequency-dependent particle polarization controlled by interfacial polarization is the Maxwell-Wagner model [Dukhin, 1970]. In this model, the complex dielectric constant of each phase, k, is written in terms of its real dielectric constant, ε_k, and bulk conductivity, σ_k,

$$\varepsilon_k^*(\omega)=\varepsilon_k-\mathrm{j}\,\sigma_k/\omega\,\varepsilon_0, \tag{1}$$

where ω is the angular frequency, ε_0 is the permittivity of free space, and the ε_k's and the σ_k's are assumed to be independent of both the electric field strength and frequency. Charge neutrality is assumed to be maintained everywhere within the suspension except within an infinitesimally thin layer near the fluid-particle interface (hence the label "interfacial polarization"). Since the average electric field, averaged over a volume containing many particles, is uniform, the effective medium dielectric constant can be obtained by dividing the volume-averaged dielectric displacement by the volume-averaged electric field strength [Dukhin, 1970]. Maxwell's approximate expression, which can be obtained from a heuristic mean field argument, is given by [Dukhin, 1970]

$$\varepsilon^*=\varepsilon_c^*[1+2\beta^*(\omega)\Phi]/[1-\beta^*(\omega)\Phi], \tag{2}$$

where ε_c^* is the complex dielectric constant of the continuous phase, Φ is the particle volume fraction, ω is the electric field frequency, and β^* is the complex relative polarizability defined as

$$\beta^*(\omega)=(\varepsilon_p^*-\varepsilon_c^*)/(\varepsilon_p^*+2\varepsilon_c^*), \tag{3}$$

where ε_p^* is the complex dielectric constant of the particle. Thus, even though the permittivities of the phases are frequency independent, the suspension permittivity is not, described in terms of the frequency dependent complex relative polarizability, $\beta^*(\omega)$.

As low frequency dispersion is commonly attributed to the relatively large-distance migration of charged species, and since we find that the addition of surfactant increases this dispersion [Kim, 1996], we assume that the surfactant adsorption only significantly influences the dc-conductivity of the particulate phase (i.e., not its dielectric constant). A linear relationship is assumed to describe this dependence:

$$\sigma_p=\sigma_{p0}+k\theta, \tag{4}$$

where σ_{p0} is the conductivity of the particulate phase in the absence of surfactant, and k is the proportional constant describing the surfactant adsorption dependence of the conductivity in the particulate phase. σ_{p0} and k are parameters to be determined from the dielectric data. σ_{p0} (=2.3×10^{-10} mho/cm) was obtained by fitting the dielectric data of 20 wt% dried neutral alumina suspensions (i.e., when θ is 0) to the Maxwell-Wagner model. ε_p (=8.6 [Hippel, 1954]) is assumed to be independent of surfactant concentration. Measured values of ε_c and σ_c were employed in the model.

Brij®30 adsorption isotherm on dried neutral alumina particles in SF96 is presented in Fig. 4. The coverage, θ, is modeled by the two-step adsorption model [Zhu, 1991]

$$\theta=\mathrm{k}_1\mathrm{C}_{equil}(1/\mathrm{n}+\mathrm{k}_2\mathrm{C}_{equil}^{n-1})/[1+\mathrm{k}_1\mathrm{C}_{equil}(1+\mathrm{k}_2\mathrm{C}_{equil}^{n-1})], \tag{5}$$

where the constants (n, k_1, and k_2) are determined from a least-squares fit of the experimental adsorption data. The solid line in Fig. 4 is the least-square fit. The constants obtained from the fitting are: n=2.257, k_1=1 kg/gmole, and k_2=650 (kg/gmole)^n.

Dielectric data for a 20 wt% dried neutral alumina suspension with 3 wt% Brij®30 are presented in Fig. 5. Included in this figure is the least-squares fit of Maxwell-Wagner model. The constant k (=8.25×10^{-8} mho/cm) was determined by a least-squares fit of both the ε and tanδ data to the model. The agreement is not perfect, but certain features are captured. The predicted dielectric constants and loss tangents are within 20% of the data (above ~50 Hz) and the low frequency dispersion is reproduced by the model, as expected. We note that this fit requires a much larger increase in the particle conductivity than that measured for the continuous phase conductivity [$k\theta/\triangle\sigma_c$= 3483, $\triangle\sigma_c=\sigma_c$ (3 wt% Brij®30) $-\ \sigma_{c0}$], consistent with the strong surfactant adsorption and that the adsorbed surfactants act to in-

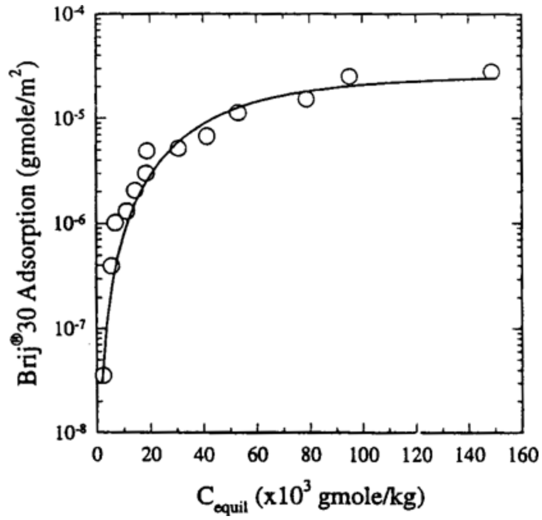

Fig. 4. Brij®30 adsorption isotherm on dried neutral alumina particles in SF96 (C_{equil} is the equilibrium surfactant concentration in the continuous phase).

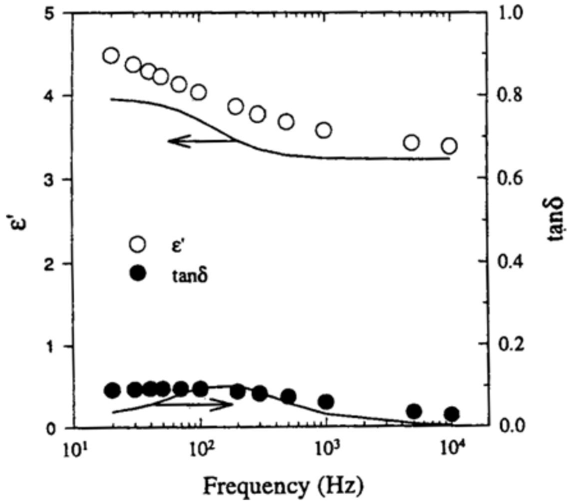

Fig. 5. Dielectric constants as a function of electric field frequency for 20 wt% dried neutral alumina suspension with 3 wt% Brij®30 (the symbols represent experimental data and the solid curve represent the model predictions).

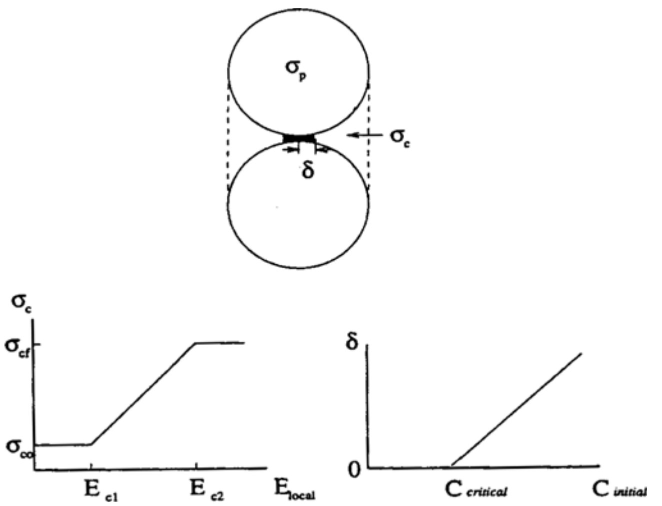

Fig. 6. Schematic diagram showing the dependence of conductivity on the electric field strength and the dependence of the surfactant structure size on the surfactant concentration.

crease the conductivity of the particles. As adsorbed species are known to increase the surface conductivity of hydrophilic particles dispersed in hydrophobic liquids [Deinega and Vinogradov, 1984; Block and Kelly, 1988], and since these alumina particles are very porous, an increase in the apparent bulk particle conductivity is not surprising.

2. Effect of the Non-linear Conduction

The enhanced interfacial polarization at low surfactant concentrations, where the ER response is linear, can be described by the Maxwell-Wagner model as discussed in the previous section. For small electric field strengths, the suspension dielectric properties are also described by this model at all surfactant concentrations. However, the model does not describe the nonlinear ER behavior at high surfactant concentrations and large electric field strengths. Here, the classical Maxwell-Wagner model fails because it does not account for the formation of highly conductive surfactant-rich bridges between particles. The Maxwell-Wagner model is modified below to incorporate this non-linear effect, as well as the enhanced interfacial polarization, on the ER response.

The dependence of the conductivity of the surfactant-rich phase between two particles on the electric field and surfactant concentration is illustrated in Fig. 6. The conductivity increases with increasing volume of the surfactant-rich bridge, which depends on the surfactant concentration and electric field strength. The conductivity in the continuous phase between particles can be modeled as

$$\sigma_c = \sigma_{co} + k_{cs}(E - E_{c1})\,H(E - E_{c1}) - k_{cs}(E - E_{c2})\,H(E - E_{c2}), \quad (6)$$

where the coefficient k_{cs} depends on surfactant concentration, E_{c1} is the first critical electric field strength where the conductivity starts to increase due to the formation of the surfactant-rich phase, E_{c2} is the second critical electric field strength from where the conductivity reaches the large field strength plateau, and H(x) is the unit step function. E_{c1} and E_{c2} are decreasing functions of the surfactant concentration. Their dependencies on the equilibrium surfactant concentration are modeled as

$$E_{c1} = k_{E1}/(C_{equil} + 0.0001), \quad (7)$$

$$E_{c2} = k_{E2}/(C_{equil} + 0.0001), \quad (8)$$

where k_{E1} and k_{E2} are constants and the factor 0.0001 is incorporated to prevent floating point errors.

The coefficient k_{cs} is a function of the surfactant concentration. From the rheological and adsorption data, the surfactant-rich phase appears to form above a critical surfactant concentration. It is assumed that k_{cs} is linearly proportional to the surfactant concentration above this critical surfactant concentration,

$$k_{cs} = k_o(C_{total} - C_{critical})\,H(C_{total} - C_{critical}), \quad (9)$$

where k_o is a constant, C_{total} is the total surfactant concentration in the ER suspension (including both adsorbed and dissolved surfactants), and $C_{critical}$ is the critical surfactant concentration where the surfactant-rich phase starts to form. The critical surfactant concentration is taken to 3 wt% Brij®30 to be consistent with the ER response, which shows a maximum yield stress at 3 wt% Brij®30.

3. Normalized Rheological Response

Rheological properties can be related to the dielectric properties within the framework of the electrostatic polarization model [Klingenberg and Zukoski, 1990; Klingenberg et al., 1989, 1991; Bonnecaze and Brady, 1992]. Once the many particle electrostatic problem is solved, the force on an individual particle can be determined from [Jackson, 1975]

$$\mathbf{F}_i^{el} = \int_{Si} \sigma^M \cdot \mathbf{n}\, dS, \quad (10)$$

where $\sigma^M = \varepsilon_0 \varepsilon_c(\mathbf{E}\,\mathbf{E} - \delta\,|\mathbf{E}|^2/2)$ is Maxwell's electrostatic stress tensor. The time-average force, <F>, between two point-dipoles under a time-varying electric field, $E_o e^{j\omega t}$, is given by [Kim, 1996]

$$\langle \mathbf{F} \rangle = 6\pi\varepsilon_0\varepsilon_c a^2 E_o^2(\beta^*\beta^{c*})\,\mathbf{f}(\beta^*). \tag{11}$$

The electrostatic contribution to the (mechanical) stress tensor is given by

$$\sigma = -(1/V)\sum_i \mathbf{r}_i\,\mathbf{F}_i^{el}. \tag{12}$$

Therefore, regardless of the level of approximation used in calculating the electrostatic forces, the yield stress must scale as

$$\tau_o \propto (\beta^*\beta^{c*})\,E_o^2\,f(\beta^*) \tag{13}$$

where E_o is the magnitude of the applied electric field, and $f(\beta^*)$ is an increasing function of β^*. For weakly polarizable particles, the function $f(\beta^*)$ is only weakly dependent on β^*, and the yield stress is proportional to $\beta^*\beta^{c*}$. Note that within the framework of this model, the frequency and surfactant concentration dependence of the yield stress lies entirely within the "normalized" yield stress, $\beta^*\beta^{c*}$. The $f(\beta^*)$ term becomes more important as the polarizability increases, producing a more rapid increase in yield stress with particle polarizability [Klingenberg and Zukoski, 1990].

The normalized time-average yield stress, τ_{on}, is defined as

$$\tau_{on} = \beta^*\beta^{c*} \equiv |\beta^*(\omega)|^2, \tag{14}$$

where

$$\beta^*\beta^{c*} = \beta_d^2([(\omega\tau)^2 + \beta_c/\beta_d]^2 + (\omega\tau)^2[1-\beta_c/\beta_d]^2)/([1+(\omega\tau)^2]^2), \tag{15}$$

and

$$\beta_d = (\varepsilon_p - \varepsilon_c)/(\varepsilon_p + 2\varepsilon_c) \tag{16}$$

$$\beta_c = (\sigma_p - \sigma_c)/(\sigma_p + 2\sigma_c) \tag{17}$$

$$\tau = \varepsilon_0(\varepsilon_p + 2\varepsilon_c)/(\sigma_p + 2\sigma_c). \tag{18}$$

Note that non-linear conduction appears in σ_c, as described in Eq. (6); the enhancement in the particle polarization appears in σ_p.

DISCUSSION

After selecting suitable proportional constants for this modified model, the normalized yield stress is calculated for various conditions and compared with experimental results to scrutinize the modified model. The constants for this modified model were selected to capture the behavior of the experimental normalized yield stress as a function of surfactant concentration, and the values are: k_{E1}=5 V/mm, k_{E2}=1000 V/mm, and k_o=3.0.

The predicted normalized yield stress is presented as a function of Brij®30 concentration in Fig. 7, for 20 wt% dried neutral alumina suspensions. The modified model captures the increase and decrease of the yield stress as the surfactant concentration increases, as observed experimentally (Fig. 1). It also captures the linear behavior at low surfactant concentrations and the non-linear behavior at high surfactant concentrations.

To further scrutinize this model, the predicted frequency dependence of the yield stress is compared to the experimental results of 20 wt% dried neutral alumina suspension with 3 wt% Brij®30 at 1.5 kV/mm in Fig. 8. The modified Maxwell-Wagner model correctly predicts the frequency dependence of the yield stress, decreasing with electric field frequency, consistent with interfacial polarization. The discrepancy at low electric frequencies (below 50 Hz) between the experimental data and predictions apparently arises from electrode polarization [Zukoski, 1993].

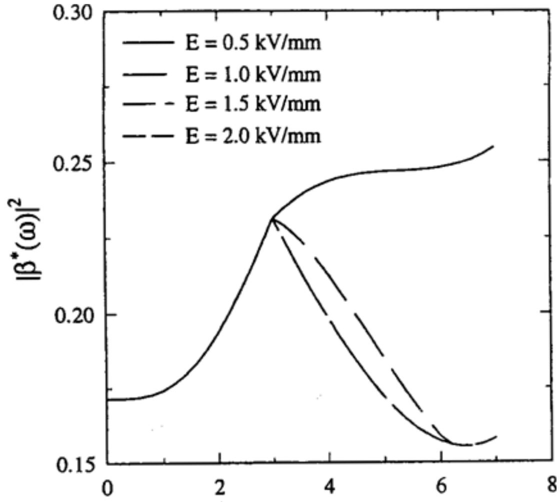

Fig. 7. Normalized yield stress as a function of Brij®30 concentration under various electric field strengths.

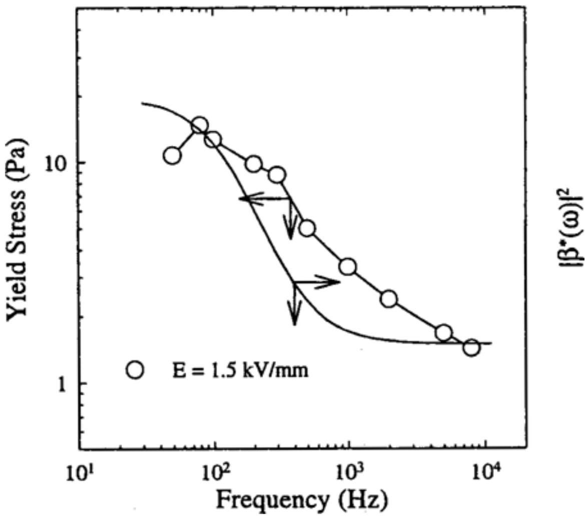

Fig. 8. Yield stress as a function of electric field frequency under an electric field strength of 1.5 kV/mm (the symbols represent experimental data for 20 wt% dried neutral alumina suspensions with 3 wt% Brij®30 and the solid curve is from the modified Maxwell-Wagner model).

The predicted frequency dependence of the yield stress is compared with experimental data for 20 wt% neutral alumina suspensions with 7 wt% Brij®30 at 2.0 kV/mm in Fig. 9. In this case, the yield stress data increases with electric field frequency, which is again captured by the model.

In the low frequency (dc) limit ($\omega \rightarrow 0$), the normalized yield stress approaches

$$\beta^*\beta^{c*} = [(\sigma_p - \sigma_c)/(\sigma_p + 2\sigma_c)]^2. \tag{19}$$

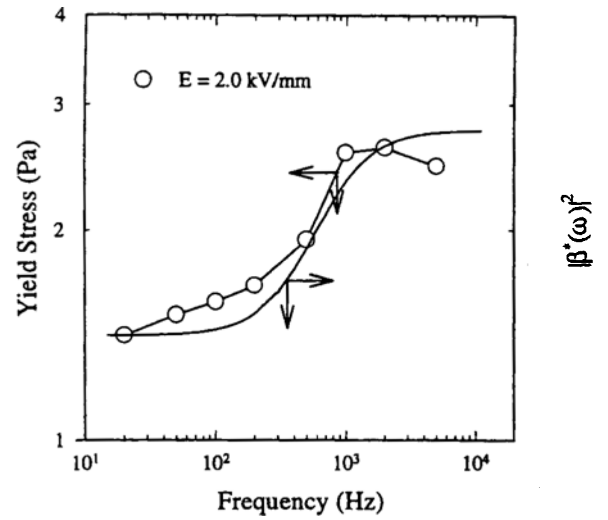

Fig. 9. Yield stress as a function of electric field frequency under an electric field strength of 2 kV/mm (the symbols represent experimental data of 20 wt% dried neutral alumina suspensions with 7 wt% Brij®30 and the solid curve is from the modified Maxwell-Wagner model).

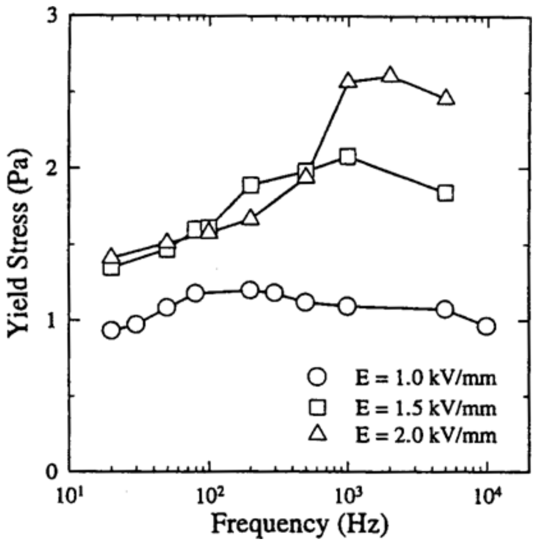

Fig. 10. Yield stress as a function of electric field frequency for 20 wt% dried neutral alumina suspensions with 7 wt% Brij®30.

Conductivities dominate polarization and the yield stress in this region. For low surfactant concentrations, the conductivity in the continuous phase is smaller than the enhanced particle conductivity and is independent of the electric field strength. The yield stress in this low electric field frequency region is large and the resulting ER response is linear. As the electric field frequency increases, mobile charges can no longer keep up with the varying electric field, and the polarization decreases, becoming dominated by the permittivity difference between the phases. As this difference is smaller than the dc conductivity difference, the polarization and yield stress decrease with electric field frequency (Fig. 8).

At large surfactant concentrations, the conductivity in the continuous phase between particles is comparable to the enhanced particle conductivity, due to the formation of the surfactant-rich phase between particles under the large electric fields. The yield stress in the dc limit is therefore smaller than that for small surfactant concentrations. The ER response is non-linear due to the non-linear behavior of the conductivity in the continuous phase between particles. As the electric field frequency increases, permittivity differences began dominate the yield stress, and thus the yield stress increases (Fig. 9). Here, the response is now linear since the permittivities are independent of field strength. This linear ER behavior at high electric field frequencies, even at large surfactant concentrations, is observed experimentally for 20 wt% dried neutral alumina suspension with 7 wt% Brij®30, as depicted in Fig. 10.

SUMMARY

Maxwell-Wagner model was modified to predict the ER behavior of the nonionic surfactant activated ER suspensions. The model was modified by incorporating: (1) the dependence of the particle conductivity on the surfactant adsorption and (2) the dependence of the suspension conductivity on the formation of the surfactant-rich phase between particles, which depends on the surfactant concentration and electric field strength. Although this model is crude, it qualitatively captures the salient features of the influence of surfactants on the ER response. Quantitative prediction will be considerably more challenging, as this requires solving for the electrostatic potential in a three phase system.

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NOMENCLATURE

E : electric field
k : constant
r : position vector
V : volume

Greek Letters

ε^* : complex dielectric constant
β^* : complex relative polarizibility
σ : conductivity
ω : angular frequency
Φ : particle volume fraction
θ : surfactant adsorption coverage
τ : stress

Superscripts

* : complex number
c : conjugate

Subscripts

c : continuous phase
p : particulate phase
c1 : first critical point
c2 : second critical point
i : particle

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