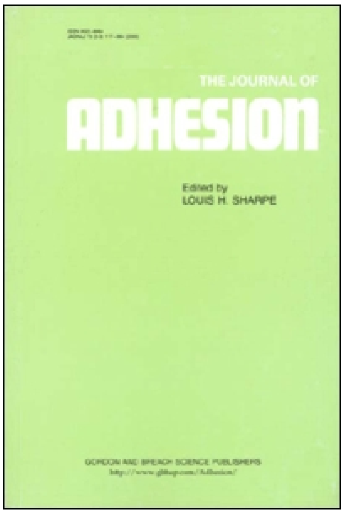

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Effect of Surface Moisture on the Polarization of Individual Glass Spheres*

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When glass particles adsorb water from humid environments their low frequency dielectric properties change drastically. To investigate the polarization mechanism, variable frequency measurements upon individual glass particles ($\sim 50\,\mu m$ diameter) suspended in silicone oil were performed with a computer-controlled dielectrophoretic levitator. The electrical properties of individual particles have been measured in the frequency range between 10 Hz and 100 kHz. A relaxation in the induced dipole moment of moistened glass particles suspended in silicone oil is clearly evident in measured spectra. Surface moisture causes the characteristic relaxation times of soda-lime glass particles to change by up to 5 orders of magnitude. The relaxation spectra are satisfactorily modeled by the Debye equation and a model based on ohmic surface conduction appears to explain the phenomenon. This research has potential implications with respect to the controversy surrounding the influence of moisture on the performance of electrorheological fluids.

Keywords: Particles; spectra; relaxation; moisture; water; Debye; glass; polarization; dipole moment; surface; conduction; electrorheological fluid

1. INTRODUCTION

The induced polarization of a dielectric particle can be drastically altered by surface phenomena. A thin ohmic surface layer screens

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electric fields from the bulk of a particle, thereby greatly increasing the static polarization. Furthermore, the finite response time of charge in the surface layer introduces a time dependence that manifests itself as a relaxation in the polarization. Current theoretical models describing particle polarization originated from Maxwell's [1] work on heterogeneous dielectric materials. Generalized by Wagner [2] to include conductivity, Maxwell's theory describes the polarization of disperse systems of particles but does not account for surface polarization. The importance of surface effects, however, is apparent in a variety of interesting particulate systems.

For many small particle systems, where the surface-to-volume ratio is large, surface phenomena dominate and must be accounted for in polarization models. Technologies in which electrical surface phenomena are critical include electrorheological fluids, electrophotography, electrofusion of biological cells, and electrophoretic separation. Electrorheological (ER) fluids are particularly reliant on mechanisms of surface polarization. In this article, the surface polarization of small particles is investigated by studying a model ER fluid: moistened glass particles in an insulating oil. Characterizing the polarization mechanism is a crucial step in understanding and developing ER fluids.

Theoretical models describing the surface polarization of particles evolved from two separate ideas: electric double layer theory and conductive film theory. These two theories are distinguished by the process controlling the movement of charge. In an electric double layer the competition between diffusion and electrical potential gradients governs charge transport, whereas the conductive film model is based on charge migration, typically considered "normal" conduction in an ohmic material.

An experimental approach quite different from any previous technique has been used to investigate small particle surface phenomena [3]. Instead of the usual technique of studying suspensions of many particles, the electrical properties of individual particles and chains were measured using a dielectrophoretic levitator. Levitation of small particles (20 μm to 300 μm in diameter) is accomplished by supplying a dielectrophoretic force equal to the force of gravity. The inverse square of the applied voltage required to levitate a sample with the dielectrophoretic (DEP) force is proportional to the particle's induced moment. This relationship between the levitation voltage and the induced moment allows the particle's electrical properties to be determined over a range of frequencies.

Using the DEP levitator to measure directly the characteristics of single particles eliminates many of the variables and difficulties involved in traditional capacitive cell measurements on suspensions of particles. For example, field distortions from adjacent particles are eliminated, as well as the possibility of forming conducting bridges between contacting particles and between particles and electrodes. In addition, particle size and shape are strictly controlled and measured.

2. BACKGROUND

Moisture on the surface of particles in the fluid is evidently required for most ER fluids because of the strong effect it has on polarization at low frequency. The first comprehensive study of the ER effect in AC fields, following its discovery by Winslow in 1949, was performed by Klass and Martinek on silica dispersions [4]. They found a substantial weakening of the ER effect as the frequency of the applied electric field increased beyond 100 Hz. Measuring a corresponding decrease in the dielectric constant over the same frequency range, they established a connection between polarization and the ER effect. Though they offered no theoretical model themselves, Klass and Martinek referred to the model proposed by Schwarz [5] to explain the polarization mechanism and the Debye relaxation in the dielectric constant. The link between Schwarz's theory and ER fluids is tenuous. Schwarz used electric double layer (EDL) theory to analyze the low frequency dielectric dispersion of colloidal particles and restricted his theoretical model to systems of particles suspended in aqueous electrolytes. The connection to ER fluids (which employ insulating media) arises because of the particle surface moisture.

According to EDL theory, the movement of ions near a surface is governed by the competition between diffusion and electrical potential gradients. In Schwarz's EDL model, a monolayer of ions, held strongly to the surface, responds to the external field by moving tangentially along the surface, in effect, "jumping" between defect sites in the surface structure. He claimed that the surface decreases the mobility of the ions because they must overcome a potential barrier each time they jump from one site to another. Schwarz's assumption that ions move only along the surface and can not move radially has been questioned in subsequent articles [6, 7]. By including radial movement, a diffuse layer of ions surrounding the particle then comes to dominate the surface polarization.

Dukhin reworked the theory developed by Overbeek [8] and Booth [9] which postulates a diffuse cloud of ions surrounding each particle. The analysis of Dukhin predicts a distribution of relaxations in the polarization. His equation describing the mean relaxation time is nearly identical to Schwarz's equation that predicts, instead, a single relaxation. The only difference, other than the expected distribution of relaxations, is the assumption by Schwarz of a decrease in ion mobility due to the proximity of the ions to the surface. In either case the characteristic relaxation time of a sphere's dipole moment is expressed as

$$\tau = R^2/2ukT \tag{1}$$

where k is Boltzmann's constant, T is temperature, R is the radius of the sphere and u is the ion mobility (in Schwarz's model u is reduced by surface interactions).

An alternate approach to modeling surface polarization of dielectric particles is to consider "normal" conduction in a thin ohmic surface layer. This concept of surface conductivity was first applied to particles by Murphy and Lowry [10] and theoretically developed by Miles and Robertson [11], and O'Konski [12]. By solving Laplace's equation for a spherical particle having a thin conducting shell, Miles and Robertson showed that the dielectric behavior of the coated sphere is equivalent to a homogeneous sphere with a modified volume conductivity. The conductive film model, therefore, can be considered as an extension of Maxwell's [1] and Wagner's [2] theories of heterogeneous dielectrics. In the Maxwell–Wagner model, charge migration through the bulk of a lossy dielectric particle was shown to cause a relaxation in its polarization. The relaxation is due to the finite response time of the charge that accumulates at the surface of the particle. This surface charge screens the applied electric field from the bulk of the particle. Miles and Robertson showed that a nonconducting particle will exhibit a similar relaxation if an ohmic surface layer is present. O'Konski reworked the original theory and presented a more physical interpretation than Miles and Robertson while achieving similar results. The characteristic relaxation time was found to be

$$\tau = (\varepsilon_2 + 2\varepsilon_1)/(\sigma_2 + 2\lambda/R + 2\sigma_1) \tag{2}$$

where the particle, having a bulk permittivity ε_2, surface conductivity λ and bulk conductivity σ_2, is suspended in a fluid of permittivity ε_1, and conductivity σ_1. It is important to note that the inclusion of surface conductivity introduces a particle size dependence into the relaxation time of the polarization that does not appear in the Maxwell–Wagner model. This size dependence is linear with particle radius for the simplified case of surface conduction on a nonconducting particle in a nonconducting medium.

In this polarization model, O'Konski did not include the effects that characterize Schwarz's model, namely, the displacement of ions and the competing diffusion mechanism. On the other hand, Schwarz did not account for the ohmic conduction resulting from charge migration upon which O'Konski's model is based. Schurr [6] was able to combine the two concepts in a general model. Using new boundary conditions, Schurr's solution predicts two relaxations, one closely related to that of Schwarz and the other equivalent to that of O'Konski. To account for a few questionable assumptions made by Schurr, more rigorous treatments have been published [7, 13] but the relaxation equations do not change.

Dukhin [7] pointed out that Schurr's theory considered only the case in which the conductivity of the surface is much less than that of the medium. Dukhin modified Schurr's theory to model a moist particle in a nonconductive medium and concluded that a single relaxation occurs under these conditions. The equation for the relaxation time simplifies to Eq. (1) for very small particles and to Eq. (2) for large particles. However, when the thickness of the water layer is comparable with the particle radius this theory no longer holds. Dukhin suggested that the diffuse ion model, which predicts a distribution of relaxations, is appropriate under this condition.

It seems apparent that some form of surface polarization is responsible for the ER effect. However, little experimental work has been directed toward testing relevant polarization models. Most, if not all, published studies concern experiments on suspensions or particle beds having relatively high volume fraction suspensions (greater than 0.05).

Working with an ER fluid composed of cellulose particles ($\sim 5\,\mu m$) dispersed in a chlorinated insulator oil, Uejima [14] measured a relaxation in the fluid's effective dielectric constant and found that it shifted to higher frequency as the amount of adsorbed water was increased. Uejima offered no explanation for the distribution of relaxations he measured in the dielectric constant of the ER fluid, nor any information about particle shape or size range. However, he presented evidence indicating that a correspondence exists between surface polarization and the strength of the ER effect by showing the influence of the adsorbed water of the apparent viscosity of the fluid when an electric field is applied.

A study by Takashima and Schwan [15] on bulk crystalline powders of glycine, tyrosine, glycylglycine, and ovalbumin measured the effect of moisture content on the dielectric constant for frequencies ranging from 20 Hz to 200 kHz. They found that the effective dielectric constant at low frequencies increases sharply with water content up to 5%, then levels off and becomes constant. As the researchers have pointed out, such data are difficult to interpret because the amount of water that penetrated into the crystals was unknown. They did find that while particle size has little influence on the magnitude of the dielectric constant, it substantially alters the relaxation time.

A considerable amount of literature exists which concerns the low-frequency polarization of aqueous electrolyte-based colloids [16–19]. However, because of material differences it is difficult to apply any of these findings to ER fluids, which generally consist of larger particles, adsorbed water layers, and nonconducting fluids.

3. EXPERIMENT

The experiments described here were conducted with individual spherical particles ($30\,\mu m$ to $150\,\mu m$ in diameter) suspended in silicone oil (Dow Corning 200$^{®}$, kinematic viscosity = 10 cSt). A constant relative humidity, used to control the amount of physically adsorbed water on the particles, was established in sealed chambers with saturated aqueous salt solutions containing excess solids. Particles were heat-dried at 250°C for 20 minutes before being placed in the humidity chambers. An effective relative humidity of 0% was achieved by heat-drying the particles in this manner then quickly covering them with oil.

The electrical characteristics of individual particles were measured with the DEP levitator by observing the DEP force over a range of frequencies (more information on the experimental apparatus can be found in Ref. [20]). An expression relating the real part of the relative polarization of the particle ($K = \mathrm{Re}[\underline{K}]$) to the applied electric field needed for levitation and other known parameters is found by equating the dielectrophoretic and gravitational forces on the particle.

The time-averaged DEP force on a particle in a sinusoidal electric field E is

$$\langle F_{\mathrm{DEP}} \rangle = \frac{1}{2}\,\mathrm{Re}[\underline{p}_{\mathrm{eff}} \cdot \nabla E^{*}] \tag{3}$$

(asterisk * indicates complex conjugate). Consider a single uniform sphere of radius R, permittivity ε_2, and conductivity σ_2, suspended in a fluid of permittivity ε_1, and conductivity σ_1. The induced dipole moment of the sphere due to an alternating electrical field of radian frequency ω is given by the Maxwell–Wagner theory as

$$\underline{p}_{\mathrm{eff}} = 4\pi\varepsilon_1 R^3 E\underline{K} \tag{4}$$

where the complex Clausius–Mossotti factor is

$$\underline{K} = (\underline{\varepsilon}_2 - \underline{\varepsilon}_1)/(\underline{\varepsilon}_2 + 2\underline{\varepsilon}_1) \tag{5}$$

and $\underline{\varepsilon}$ represents complex permittivity: $\underline{\varepsilon} = \varepsilon - j\sigma/\omega$ (underscore _ signifies a complex quantity, and $j = \sqrt{-1}$) [24]. The conductive film model [10–12] can be used to account for a thin ohmic surface layer by letting

$$\underline{\varepsilon}_2 = \varepsilon_2 - j(\sigma_2 + 2\lambda/R)/\omega \tag{6}$$

By equating the DEP force and the gravitational force, the experimentally-measured value of the real part of $\underline{K}$ is calculated

$$(K)_{\mathrm{exptl}} = \frac{2g(\gamma_2 - \gamma_1)}{3\varepsilon_1 |\partial E^2_{\mathrm{rms}}/\partial z|} \tag{7}$$

where $g = 9.81\ \mathrm{m/s^2}$; γ_2 and γ_1 are the mass densities of the particle and medium, respectively. Equation (7), along with field information relating the measured voltage to E^2_{rms}, is used to convert DEP voltage spectra into a more informative format: $(K)_{\mathrm{exptl}}$ *versus* frequency [3]. This conversion introduces a systematic error no larger than 4% due to the uncertainty in the densities and a measurement error of approximately 1% due to the uncertainty in the position of the levitated particles.

The frequency spectrum of the effective moment, $\underline{p}_{\mathrm{eff}}$, is conveniently represented by the Debye equation [21]. Expressed in terms of the high frequency (K_∞) and low frequency (K_0) limits of K, the Debye equation is

$$K = K_\infty + \frac{K_0 - K_\infty}{1 + (\omega\tau)^2} \tag{8}$$

where

$$K_\infty = (\varepsilon_2 - \varepsilon_1)/(\varepsilon_2 + 2\varepsilon_1) \tag{9}$$

$$K_0 = (\sigma_2 + 2\lambda/R - \sigma_1)/(\sigma_2 + 2\lambda/R + 2\sigma_1) \tag{10}$$

Note that K has the following limits: $-0.5 \le K \le 1.0$. Figure 1 shows a typical Debye relaxation curve that has a characteristic relaxation frequency of 1 kHz. A more convenient expression than τ, the relaxation frequency, is the frequency at which $K = (K_0 + K_\infty)/2$ and is defined as

$$f_r = \frac{1}{2\pi\tau} \tag{11}$$

From the measurement of the DEP force over a range of frequencies the particle's relaxation time, surface conductivity, and permittivity can be determined. A nonlinear least-squares curve fit of the DEP spectra (K *versus* frequency), based on the Debye equation, yields the measured relaxation times of the levitated particles. The effective permittivity of a particle, ε_2, is obtained from the measured value of K_∞ and knowledge of the permittivity of the medium, ε_1. Similarly, K_0 determines the effective surface conductivity, λ. However, for the

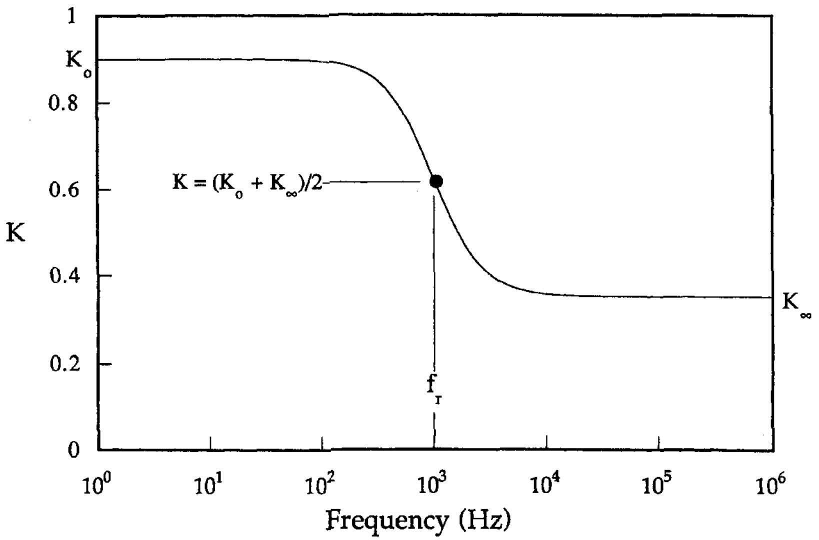

FIGURE 1 Typical Debye relaxation spectrum. The characteristic relaxation frequency, f_r = 1 kHz, is the frequency at which $K = (K_0 + K_\infty)/2$.

system studied here, that is when σ_1 and σ_2 are much less than $2\lambda/R$, the surface conductivity can not be accurately determined in this manner because K_0 is very insensitive to λ/R. Instead, we assume that the conductive film model applies and use the relaxation time given by Eq. (2), which is adequately sensitive to λ/R, to calculate the surface conductivity

$$\lambda = (\varepsilon_2 + 2\varepsilon_1)R/2\tau \tag{12}$$

4. RESULTS

Initial measurements were performed on untreated, spherical, soda-lime glass particles having no apparent physical differences other than size. The untreated particles, obtained from the same batch and not modified in any way, exhibit relaxations which are broadly distributed; characteristic relaxation frequencies range from below 10 Hz to greater than 10 kHz. This wide range is illustrated in Figure 2, which shows

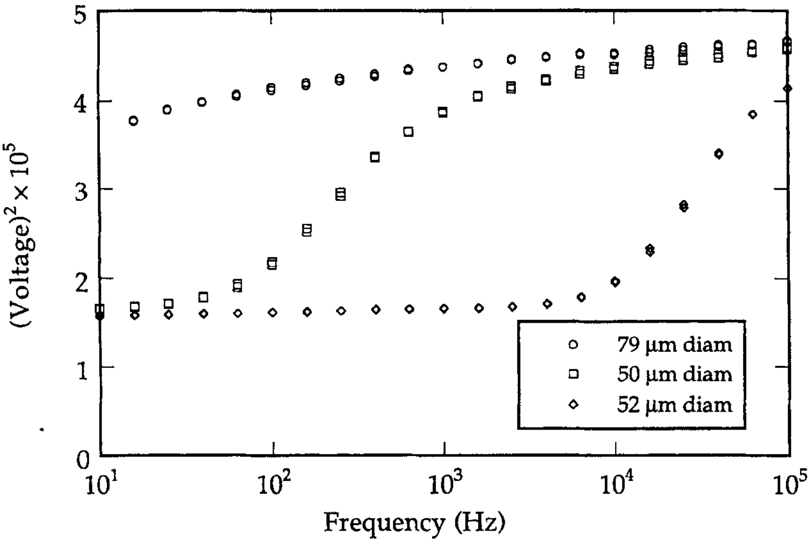

FIGURE 2 Untreated soda-lime glass particles levitated in silicone oil exhibit relaxations which are broadly distributed; characteristic relaxation frequencies range from below 10 Hz to greater than 10 kHz.

the DEP voltage spectra of three typical untreated glass spheres. The DEP voltage spectrum is the square of the voltage needed to levitate the particle at a fixed position as a function of the frequency of the applied voltage.

Adsorbed water greatly affects the frequency-dependent polarization of glass spheres-relaxation times change by more than 5 orders of magnitude depending on humidification. Although various particle treatments produce differing DEP spectra, certain similarities in the measured DEP spectra are evident. For example, the majority of the measured spectra of single particles reveal a well-defined Debye relaxation and all particles exhibit the same value for the relative polarization at high frequencies: $K_\infty = 0.35$. The dielectric constant, obtained from Eq. (9) and the measured value of K_∞, matches the particle manufacturer's specifications, $\varepsilon_2/\varepsilon_0 = 6.9$. Another similarity in the DEP spectra is that the low frequency value of K is nearly equal for all particles, that is $K_0 \approx 0.92$.

After confirming that the wide range in relaxation frequencies exhibited by otherwise identical glass spheres (as seen in Fig. 2) was due to water adsorption, consistency in the relaxation frequencies of measured DEP spectra was attained by careful control of the humidity to which particles were exposed. The water adsorbed on the surface of the glass particles was altered by heat and humidity treatments. Heat-drying the glass particles at 250°C for 20 minutes to remove physically-adsorbed water consistently decreases the measured relaxation frequency to less than 10 Hz. Exposing the particles to a water-saturated environment increases the relaxation frequency several orders of magnitude. Figure 3 shows a typical example of the effects on the relative polarization of particles after either of two treatments: heat-drying or constant humidity. As outlined above, the voltage data are converted to the relative polarization, K, and a curve is fitted (for the moisture-treated case) based on the Debye equation, Eq. (8). This data presentation format, K *versus* frequency, is maintained throughout the remainder of this article. The slight rise in K as frequency is decreased below 100 Hz, seen in Figure 3, is typical for all soda-lime

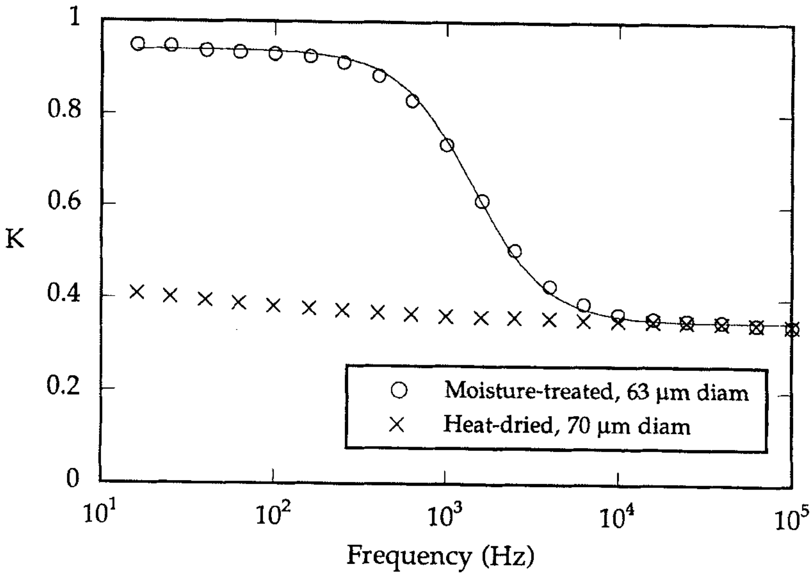

FIGURE 3 A relaxation appears in the relative polarization of soda-lime glass particles suspended in silicone oil when exposed to a humid atmosphere. Heat-dried particles show no relaxation. A nonlinear least-squares curve fit using the Debye equation (8) is shown for the moisture-treated case (f_r = 1.4 kHz) [26].

particles that are heat-dried. This increase is consistent with independent measurements of the dielectric constant of soda-lime glass in this frequency range [22].

DEP spectra obtained for particles exposed to various values of fixed relative humidity (after heat-drying at 250°C for 20 minutes) show an increase in relaxation frequency with increasing relative humidity. Figure 4 shows representative DEP spectra for particles treated at various *rh*, while Figure 5 graphs the relaxation frequency *versus* percent relative humidity for all the data. A large increase in f_r occurs between 15% and 32% *rh*, while a smaller increase, one order of magnitude, occurs between 32% and 90% *rh*. The 6 data points at 0% *rh* in Figure 5 represent particles that were heat-dried at 250°C for 20 minutes. The spectra of these particles do not exhibit any relaxation phenomena within the frequency range of the DEP levitator. A somewhat arbitrary relaxation frequency of 5 Hz was assigned to these data and to the 15% *rh* data; this value represents an upper limit, actual values may be much lower.

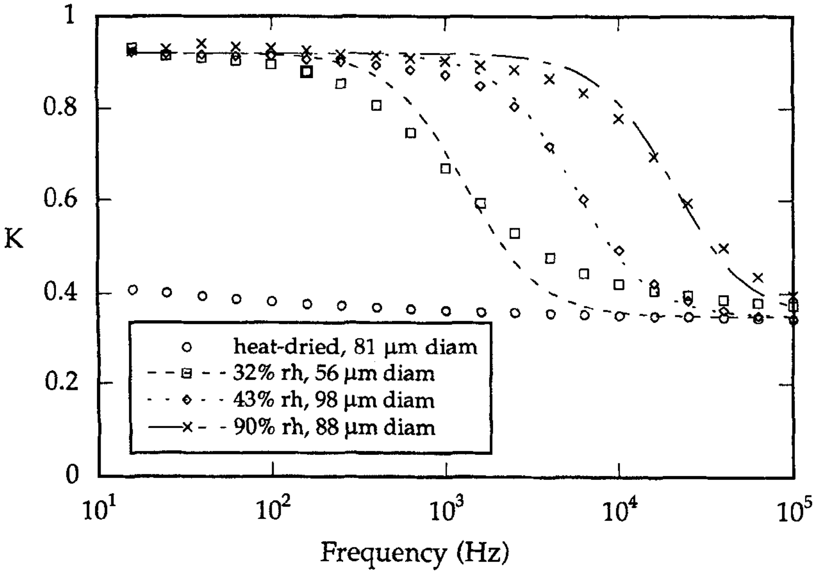

FIGURE 4 Representative DEP spectra for soda-lime glass particles treated at various relative humidities. Curves are fitted to the Debye equation.

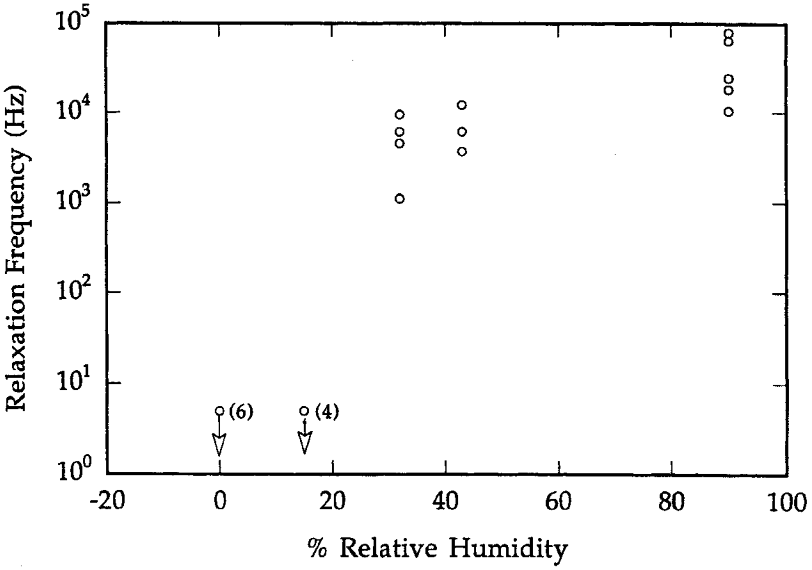

FIGURE 5 Relaxation frequencies of soda-lime glass particles treated at different relative humidities (diameters range from 54 µm to 78 µm).

Although stringent measures were taken to assure that the particles were all treated the same, some scatter appears in the data. In addition to the precautions described above, particle size and shape were strictly monitored by visual inspection with a microscope. Only particles that appeared clean and perfectly spherical were tested. Samples having shape deformities or surface contamination were discarded. Apparently, undetectable particle-to-particle differences in the surface structure are responsible for the scatter shown in Figure 6. The surface characteristics of alkali glass, known to vary greatly from sample to sample, strongly influence the effect that moisture has upon surface conductivity. According to the conductive film model (Eq. (9)) these influences are reflected in the relaxation time.

Contrary to the predictions of the polarization models considered above, no correlation is found between particle size and relaxation time. The absence of any discernible size dependence in the relaxation time is attributed to the scatter in the data and the limited range in the particle size (30 to 150 µm diameter). A linear radius dependence, if present, might be obscured by the scatter (approximately 1 order of

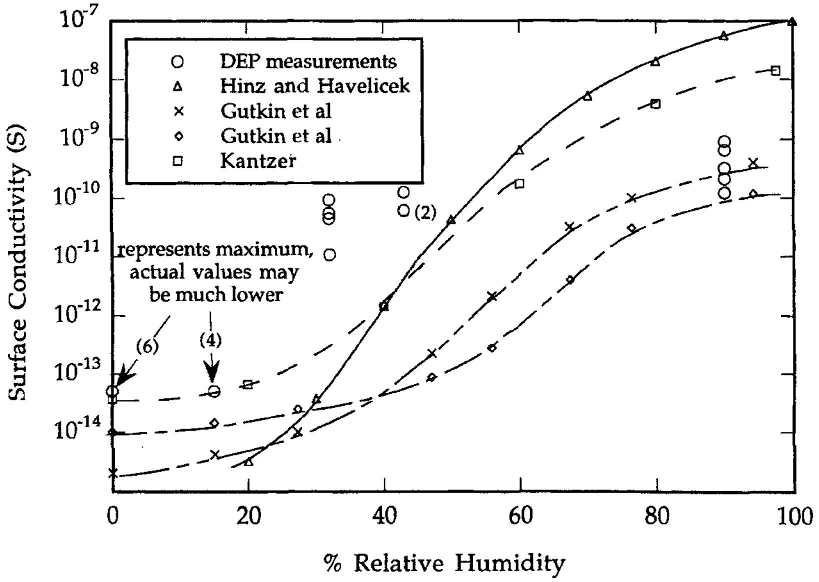

FIGURE 6 The effective surface conductivity, calculated using the conductive film model, for particles treated at different relative humidities (DEP measurements). Also shown are DC measurements of glass plates [27, 28] and glass fibers [29] performed in air.

magnitude) because a 1 order of magnitude change in radius would produce a 1 order of magnitude change in f_r. However, even with an order of magnitude of scatter, a square radius dependence, if present, should have been detectable because f_r would then change by almost 2 orders of magnitude over the range of the particle sizes investigated.

Furthermore, as shown in the following calculation, the characteristic relaxation time of the EDL model fails to predict the measured relaxation frequencies. First, Eq. (1) is written in terms of the characteristic relaxation frequency and the diffusion coefficient, D, of the mobile ions in the surface layer

$$f_r = D/\pi R^2 \qquad (13)$$

The diffusion coefficient changes with concentration and reaches a peak value as the concentration approaches zero. Although the concentration in the surface layer is probably significant, the maximum value for D is used here to illustrate that Dukhin's EDL model is not the correct choice for a polarization model. Using a typical particle radius of 30 μm, and the highest value for the diffusion coefficient of $CaCl_2$, $1.34 \times 10^{-9}\,m^2/s$, Dukhin's model predicts a relaxation frequency of 0.47 Hz – several orders of magnitude lower than the measured relaxation frequencies of particles treated at 32% *rh* and higher.

The conducting film model, which predicts a linear radius dependence, may be used to calculate the effective surface conductivity from the measured values of relaxation frequency (Eq. (13)). Figure 6 graphs the surface conductivity *versus* percent relative humidity for the treated soda-lime glass particles along with published values of the DC surface conductivity for glass fibers and glass plates. It is important to note that the published measurements on the glass plates and fibers were conducted in air. Unfortunately, no published values of surface conductivity for moist glasses in oil are available. Furthermore, all values derived from the DEP experiments were obtained from AC measurements over a range of frequencies. In addition, the chemical compositions and surface attributes of the glass plates and fibers differ from the soda-lime glass used in the DEP measurements. Nevertheless, the DEP measurements (represented by the circles in Fig. 6) are roughly comparable with the DC measurements performed in air. These encouraging results lend support to the hypothesis that the conductive film model may, to a first approximation, describe the surface polarization phenomena of the particles.

The rapid increase in surface conductivity with relative humidity for the DC measurements reflects the commonly accepted view that a monomolecular layer of physically adsorbed water forms on glass at approximately 40% *rh*. On the other hand, the DEP measurements show a rise in λ somewhere between 15% and 32% *rh*. This difference may be due to the different method used here to measure λ. In the DC measurements on the glass plates and fibers, the measured conductivity increases rapidly once a continuous film forms a conducting path along the surface between measurement probes. In the present research, an AC polarization technique is employed to measure λ, thus, capacitive losses are included and non-continuous patches of water on particle surfaces at the lower humidities may increase the effective AC surface conductivity in the DEP measurements.

When left in the silicone oil for more than 2 hours the measured relaxation frequencies of moisture-treated particles begin to shift gradually to lower values. The "time-in-oil" effect is shown in Figure 7 for particles treated at different *rh*. Note that each data point in the figure represents a complete DEP spectrum; a Debye curve fit of the spectrum yields f_r. In addition, each matching symbol on the plot is the same particle at different times, that is, all the data presented in Figure 7 represent only 3 different particles. The curves in Figure 7, which are identical in shape, show that the absorption of water by the oil occurs at the same rate in each of the 3 cases. The data labeled "☐ H-glass, 20 mM $CaCl_2$, 90% *rh*" shows the time-in-oil effect for a particle heated in distilled water at 50°C for 90 minutes to remove surface ions in the bulk near the surface then heated in an oven at 250°C for 20 minutes to densify the surface. The particle was then immersed in a 20 mM solution of $CaCl_2$ to intentionally deposit ions on the surface. Next the particles were heat dried before being exposed to 90% *rh* [23].

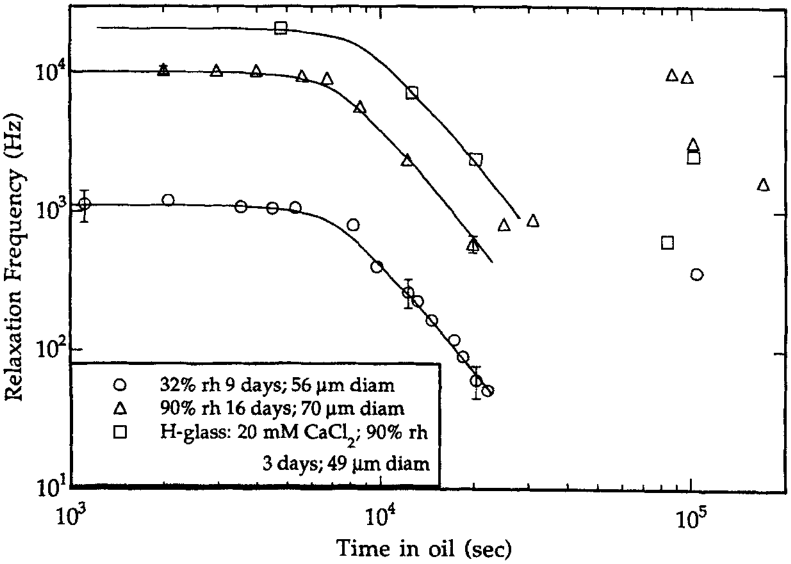

FIGURE 7 Time of particle exposure to oil *versus* the relaxation frequency, which is obtained from curve-fitting the particle's spectra. The curves shown in this figure are identical in shape, thereby indicating similar rates of water absorption by the oil up to approximately 2 × 10^4 sec. Error brackets indicate 99% confidence intervals that result from the Debye curve fits.

Silicone oil is capable of absorbing significant amounts of water (up to 0.1% by weight) and, apparently, the oil slowly absorbs the surface moisture on the particles. A decrease in the thickness of a particle's water layer reduces the surface conductivity, thereby decreasing its relaxation frequency.

The scatter in the data that appears after 2×10^4 seconds (about 6 hours) is most likely due to changes in ambient conditions in the laboratory; the relative humidity of the laboratory was not controlled and the levitation chamber was not sealed. To avoid any variability in subsequent experiments from the time-in-oil effect, measurements of the DEP spectra were performed only if the sample had been in the silicone oil for less than 2 hours. Furthermore, the silicone oil was replaced after each measurement.

5. CONCLUSIONS

A new technique, dielectrophoretic levitation, has been employed in the study of the dielectric surface properties of small particles in the frequency range between 10 Hz and 100 kHz. A Maxwell–Wagner relaxation in the dipole moment of glass particles is clearly evident in measured DEP spectra. In most cases, this relaxation can be very closely modeled by the Debye equation. Models for the surface polarization of small particles have been reviewed and the conductive film model has been found to yield the best predictions of measured relaxation frequencies. According to the conductive film model, the characteristic relaxation frequency of the dipole moment is proportional to both the amount of adsorbed water and the density of ions within the water layer.

The polarization of glass particles was found to be strongly influenced by surface moisture. Adsorbed water from constant humidity treatments introduces a high surface conductivity on a particle that acts to screen applied electric fields, thereby greatly increasing the particle's effective dipole moment at low frequencies. Indeed, depending on the type of surface treatment, the characteristic relaxation times of soda-lime glass particles change by more than 5 orders of magnitude.

While untreated particles exhibit unpredictable characteristic relaxation frequencies that vary from one particle to the next over several orders of magnitude, heat-dried particles exhibit no relaxation in the range of the DEP levitation instrument. Drying the particles lowers their relaxation frequency well below 10 Hz and almost no frequency variation is found in the effective dipole moment of the particles throughout the range of the DEP levitator. On the other hand, exposing the heat-dried particles to a constant humidity for 3 or more days consistently increases their characteristic relaxation frequency. These experiments show that the relaxation time of glass particles is directly related to the relative humidity of the particle treatments.

The experimental results presented here begin to fill a gap that exists in our understanding of the dielectric response of small particles. In particular, the study of ER fluids should benefit from these findings. The investigation of single-particle dielectric phenomena may be used to broaden our understanding of the electrical properties of a wide range of materials [24], *e.g.*, biological cells [25] and toner particles of electrophotographic machines.

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