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Viscosity of an electro-rheological suspension with internal rotations.

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Synopsis

The rheological behaviour of a suspension of insulating particles dispersed in a slightly conducting liquid under the action of a DC electric field is studied. The polarisation of the particles induced by the field is shown to be responsible for a rotation of the particles (Quincke rotation) which, in turn, leads to a drastic decrease of the apparent viscosity of the suspension. The purpose of the paper is to provide a relation between the apparent viscosity of the suspension and the E field intensity. First, the steady state solutions are searched for the angular velocity of a particle subjected to both DC E field and simple shear flow. Since the solutions are multivalued, their stability is studied using a linear stability analysis. Then, the stable solution for the particle angular velocity is used to deduce the value of the apparent viscosity of the suspension. The predictions of the model are compared to experimental data which have been obtained on a suspension of PMMA particles dispersed in a low polar dielectric liquid. The agreement between experiments and theory is rather good even if the model overestimates the viscosity decrease induced by the field.

I. INTRODUCTION

The control of the viscosity of a suspension by the application of an electric field has attracted considerable attention these last years because it can be used in various kinds of industrial devices such as active dampers, clutches or brakes [Phani and Venkatraman (2005), Lim et al. (2005), Tan et al. (2006)]. Usually, the so-called electrorheological (ER) fluids are made of relatively polarizable particles dispersed in a nonconducting liquid.

When the suspension is subject to an electric field, the particles group together in elongated aggregates aligned in the direction of the external field. This structuring of the suspension leads, in the rheology of the ER fluids, to the fast and reversible change from a liquid to a solid state. Most of ER fluids are used with an AC electric field; in this case, it is the permittivity mismatch between the particles and the liquid which is responsible for the polarisation of the particles [Marshall et al. (1989)]. DC electric fields are more rarely employed and when they are, the polarisation of the particles comes from the conductivity difference between the solid and the liquid phases. If the conductivity of the particles is higher than that of the suspending liquid, the dipole moments of the particles are collinear to the E field direction and a particle-chaining happens, exactly like in the AC case [Atten et al. (1997)]. But, if the particles are less conducting than the suspending liquid, the dipole moments of the particles lie in a direction opposite to the field and, if the E field intensity is high enough, this orientation is unstable and can give rise to a rotation of the particles. This particles' spinning is known as Quincke rotation [Quincke (1896)]. In such a case, the particles behave like colloidal motors which drive the suspending liquid and the viscosity of the suspension is lessened by the application of the field so that we can speak about "negative ER effect" [Lobry and Lemaire (1999)].

A negative ER effect has already been reported in the literature but the involved mechanism is quite different: Atten et al (1995) showed that it was possible to decrease the

viscosity of a suspension of electrically charged particles upon the application of a DC electric field. Indeed, the charged particles group together on the electrode of opposite charge. The suspension is therefore composed of two phases: the first one has a high particles concentration and its volume fraction is close to that of the packing while the second phase is only constituted of the suspending liquid. The resulting mean apparent viscosity was shown to be lower than that of the homogeneous suspension (its value is comprised between that of the suspending liquid and that of the suspension).

The possibility of decreasing the viscosity of a suspension by exerting a torque on its particles has been demonstrated on ferrofluids subject to AC or rotating magnetic fields [Bacri (1995)]. The ferrofluids are suspensions of rigid dipoles so that a time-varying external magnetic field is able to exert a torque which results in the particles rotation. Recently, C. Rinaldi et al (2005) demonstrated the possibility of obtaining "negative viscosity" when the ferrofluid is placed in a cylindrical Couette geometry and subject to a uniform rotating field.

In the present paper, we develop a model to predict the viscosity reduction induced by the application of a DC electric field to a suspension. This work is an extension of a previous study [Cebers (2000)] where the direction of the axis of rotation of the particles was assumed fixed. This assumption was responsible for a divergence of the apparent "negative viscosity" of the suspension at low shear rate. Here, we show that, if the direction of the rotation axis of the particles *is* not necessarily chosen along the vorticity direction, the "negative viscosity" divergence disappears and the model is able to fit the experimental data. The paper begins with a presentation of the model which allows the prediction of the viscosity as a function of the applied electric field (part II). The first subsection is dedicated to a discussion of the mechanisms involved in Quincke rotation and of the conditions for such a DC electro-rotation to take place. In the second subsection, the Quincke rotation of particles in the presence of a shear flow is analysed and, at last, we state how the electro-rotation of the particles modifies

the apparent viscosity of the suspension. The following section (part III) is experimental, both the samples and the experimental procedure are described. The last part is devoted to the presentation of the results and to their analysis.

II. THEORY

A. Mechanism of Quincke rotation.

Even though the rotation of small particles dispersed in a dielectric liquid and subject to a DC electric field, \mathbf{E}_0 , was observed as early as 1896, it was only at the end of the 20th century that a correct analysis of the phenomenon was proposed. The analysis proposed by Melcher (1974) is based on the role played by the free electrical charges on the particle surface and the necessary condition for the Quincke rotation to appear deals with the relative magnitude of the charge relaxation times in the liquid and in the particle. The charge relaxation time is given by the ratio of the dielectric permittivity to the conductivity: $\tau_i = \epsilon_i / \gamma_i$, $i=l,p$ refers to the liquid and the particle respectively. Indeed, if the charge relaxation time is shorter in the liquid than in the particles, when the E field is applied, the superficial charge which builds at the interface between the particle and the liquid is equivalent to that of a dipole moment, \mathbf{P} , which is in the direction opposite to the DC field (fig.1a). Any angular deviation of the particle produces a torque, $\mathbf{\Gamma}^E = \mathbf{P} \times \mathbf{E}_0$, which tends to increase its rotation further and the particle begins to rotate with an angular velocity ω .

The dipole moment \mathbf{P} is named "retarded dipole moment" and is given by a relaxation equation [Pannacci (2007a)]:

$$\frac{\partial \mathbf{P}}{\partial t} = (\boldsymbol{\omega} \times \mathbf{P}) - \frac{1}{\tau} (\mathbf{P} - (\chi^0 - \chi^\infty) \mathbf{E}_0) \quad (1)$$

where the characteristic relaxation time for the polarisation to relax is the Maxwell-Wagner time:

$$\tau = \frac{\epsilon_p + 2\epsilon_1}{\gamma_p + 2\gamma_1} \quad (2)$$

and χ^0 and χ^∞ are the polarisability factors of the particle (radius a) at low and high frequencies respectively:

$$\chi^0 = 4\pi\epsilon_1 a^3 \frac{\gamma_p - \gamma_1}{\gamma_p + 2\gamma_1} \text{ and } \chi^\infty = 4\pi\epsilon_1 a^3 \frac{\epsilon_p - \epsilon_1}{\epsilon_p + 2\epsilon_1} \quad (3)$$

When the particle is rotating at angular velocity $\boldsymbol{\omega}$, it undergoes a viscous resistant torque, Γ^η :

$$\Gamma^\eta = -\alpha(\boldsymbol{\omega} - \boldsymbol{\omega}_0) \quad (4)$$

where $\boldsymbol{\omega}_0$ is the angular velocity of the surrounding fluid (half the vorticity) and $\alpha = 8\pi\eta a^3$, the rotational friction coefficient of the spherical particle, η being the viscosity of the liquid.

The dynamics of the particle is therefore determined upon solving Eq.(1) coupled with the angular momentum principle:

$$I \frac{d\boldsymbol{\omega}}{dt} = \mathbf{P} \times \mathbf{E}_0 - \alpha(\boldsymbol{\omega} - \boldsymbol{\omega}_0) \quad (5)$$

where I is the moment of inertia of the particle. In a previous paper, we have shown both experimentally and theoretically that a particle undergoing Quincke rotation in a quiescent fluid could exhibit a complex dynamical behaviour. In particular, when the field intensity is sufficiently high, the rotor can exhibit a chaotic behaviour which is exactly described by the Lorenz equations [Peters (2005)].

As for the stationary behaviour of a particle undergoing Quincke rotation in a quiescent liquid ($\boldsymbol{\omega}_0 = \mathbf{0}$) has been studied in detail [Jones (1984)] and it is well known that the particle rotation depends on a threshold value of the field, E_c and that, above this critical field,

the particle rotates around itself with an axis pointing in any direction perpendicular to the DC field. The modulus of its angular velocity is given by:

$$\omega = \frac{1}{\tau} \sqrt{\left(\frac{E_0}{E_c}\right)^2 - 1} \quad \text{with} \quad E_c = \sqrt{\frac{-\alpha}{(\chi^0 - \chi^\infty)\tau}} \quad (6)$$

If the liquid vorticity is non-zero, the resolution of Eqs. (1) and (5), even in the stationary case, is much more complicated. This problem is solved in the following sub-section.

B. Quincke rotation in the presence of a simple shear flow

Let us consider a suspension of spheres (volume fraction ϕ) subject to a DC electric field applied in z -direction. If the liquid is at rest, the particles start rotating around themselves in any direction perpendicular to the field and so the average spin rate of the suspension is zero (figure 2.a). But, when a velocity gradient is applied along the E field direction, the particles rotation axes will be favoured in the vorticity direction (x -direction). Therefore, the degeneracy of the rotation direction will be lifted giving rise to a non zero spin rate of the ensemble of particles (figure 2.b).

The angular velocity of the suspension, ω_s , viewed like a macrocontinuum, is given by the volume average of the angular velocity over the particles and the suspending liquid [Brenner (1970)]:

$$\omega_s = \frac{1}{2} \nabla \times \mathbf{v} = -\frac{\dot{\gamma}}{2} \mathbf{e}_x = \phi \omega + (1 - \phi) \omega_0 \quad (7)$$

where $\mathbf{v} = v(z)\mathbf{e}_y$ is the suspension velocity field and $\dot{\gamma}$, the macroscopic shear rate. For the following, we have to keep in mind that $\dot{\gamma}$ is the only external parameter of Eq.(7) while ω and ω_0 are coupled variables.

Upon using Eq. (7) to express the suspending fluid angular velocity, ω_0 , in terms of $\dot{\gamma}$ and ω , the projections of Eqs. (1) and (5) on the axes Ox, Oy and Oz (see figure 2) give rise to a system of five scalar equations:

$$\begin{aligned}
\frac{dX}{dt^*} &= \text{Pr} \left(Y - \frac{X + \dot{\gamma}\tau/2}{1-\phi} \right) \\
\frac{dY}{dt^*} &= -X(Z-r) - Y \\
\frac{dZ}{dt^*} &= XY - Z - \psi\xi \\
\frac{d\xi}{dt^*} &= \psi(Z-r) - \xi \\
\frac{d\psi}{dt^*} &= -\text{Pr}(\psi + \xi)
\end{aligned} \tag{8}$$

where the variables are dimensionless:

$$\begin{aligned}
X &= \omega_x \tau ; \quad \psi = \omega_y \tau ; \quad t^* = \frac{t}{\tau} \\
\xi &= P_x \frac{E\tau}{\alpha} ; \quad Y = P_y \frac{E\tau}{\alpha} ; \quad Z = P_z \frac{E\tau}{\alpha} + r
\end{aligned}$$

and where we have introduced the ratio of the electric field intensity to the threshold value:

$$r = \left(\frac{E}{E_c} \right)^2$$

and the ratio of the Maxwell-Wagner time to the characteristic mechanical time:

$$\text{Pr} = \frac{\alpha\tau}{I}$$

In stationary state, this system of equations admits 5 solutions which can be divided into two groups. The solutions of the first group correspond to a rotation of the particles in x-direction while the few intuitive solutions of the second group correspond to zero-vorticity of

the suspending liquid ($\omega_0=0$) and a non-zero value for the y-component of the angular velocity. Let us give some details about these 5 solutions:

First group of solutions: $\omega=\omega e_x$, $X=X_{1,2,3}$

$\psi=\xi=0$ and the variables Y, Z can be expressed as a function of X which is the solution of the following third order equation:

$$X^3 + \frac{\dot{\gamma}\tau}{2} X^2 + [1-(1-\phi)r]X + \frac{\dot{\gamma}\tau}{2} = 0 \quad (9)$$

When the shear rate tends toward zero, the particles rotate only if the E field intensity is higher than a critical value, E^* :

$$E^* = \frac{E_c}{\sqrt{1-\phi}} \quad (10)$$

When the electrical field intensity is lower than E^* , Eq.(9) admits only one solution which corresponds to a rotation of the particles in the direction of the vorticity (X_1 in figure 3). For higher field intensities, there exist either one or three solutions to equation (9) (see figure 3), depending on the value of $\dot{\gamma}$. X_1 , which is always a solution, corresponds to a rotation of the particles in the vorticity direction while the other two possible solutions represent a rotation in the opposite direction (X_2 and X_3 in figure 3).

Second group of solutions: $\omega_y \neq 0$, $X=X_4$

$$\begin{aligned} X = Y &= -\frac{\dot{\gamma}\tau}{2\phi} \\ Z &= r - 1 \\ \psi = -\xi &= \pm \sqrt{-\left(\frac{\dot{\gamma}\tau}{2\phi}\right)^2 + r - 1} \end{aligned} \quad (11)$$

For ψ and ξ to be real, this double solution exists only if :

$$\dot{\gamma}\tau < \dot{\gamma}^* \tau = 2\phi\sqrt{r-1} \quad (12)$$

The x-component of the angular velocity of the particles does not depend on the E field intensity and, according to eq.(7), this non intuitive solution corresponds to a zero vorticity of the suspending liquid ($X_0=\omega_0\tau=0$) . Then, the modulus of the particles' angular velocity is equal to the value we have defined previously when there was no shear flow (Eq.6). At last, it should be stressed that this solution is compatible with the flow geometry wherein the y-component of the velocity gradient is zero. This requirement is achieved if the y-component of the angular velocity correspond to clockwise rotation ($\psi<0$) for half of the particles and counter-clockwise ($\psi>0$) for the other half.

All the X-solutions of the system of equations (8) are plotted in figures 4.a and 4.b. In appendix I, we study the stability of these solutions and we show that the solution X_4 is always a stable solution. When the shear rate is higher than the critical value $\dot{\gamma}^*$, X_4 does not exist any more but X_1 becomes stable. In figures 4.a and 4.b, the thick lines represent the stable solutions.

C. Influence of electric driving torque on the value of the apparent viscosity of the suspension.

The zy-component of the total stress tensor in the suspension, σ_{zy} is obtained upon adding its symmetric and anti-symmetric parts. Its symmetric part is the viscous stress while its anti-symmetric part is equal to the half of the x-component of the volume torque exerted on the suspension, γ^E :

$$\sigma_{zy}^T = \eta(\phi)\dot{\gamma} + \frac{1}{2}\gamma_x^E \quad (13)$$

where $\eta(\phi)$ is the viscosity of the suspension in the absence of an electric field.

In the following, we shall use the expression of the viscosity proposed by Brenner (1970) since, as it can be seen in figure 5, for rather low volume fractions of particles, it captures the variation of the viscosity with the particles' concentration:

$$\eta(\phi) = \eta \frac{1 + \frac{3}{2}\phi}{1 - \phi} \quad (14)$$

The volume torque is given by adding the torques, Γ^E , exerted on each particle contained in a unit volume:

$$\gamma_x^E = \frac{\phi}{\frac{4}{3}\pi a^3} \Gamma_x^E = \frac{\phi}{\frac{4}{3}\pi a^3} P_y E \quad (15)$$

The apparent viscosity of the suspension is then given by the ratio of the total tangential stress in the suspension to the mean shear rate:

$$\eta_{\text{app}} = \frac{\sigma_{zy}^T}{\dot{\gamma}} = \eta(\phi) + \frac{\phi}{\frac{8}{3}\pi a^3} \frac{P_y E}{\dot{\gamma}} \quad (16)$$

and the apparent viscosity normalised by the viscosity of the suspension when no field is applied is given by:

$$\eta_{\text{app}}^* = \frac{\eta_{\text{app}}}{\eta(\phi)} = 1 + \frac{3\phi}{1 + \frac{3}{2}\phi} (1 - \phi) \frac{Y}{\dot{\gamma}\tau} \quad (17)$$

Since the y-component of \mathbf{P} (see figure 2.b) is negative, it appears clear that the apparent viscosity is lowered by the application of a DC electric field. It should be stressed that expressing P_y in terms of ω_x , Eq. (17) is formally equivalent to the expression for the viscosity of a suspension of rotating spheres proposed by Brenner:

$$\eta_{\text{app}} = \eta(\phi) + \eta_r(\phi) \quad \text{with:} \quad (18)$$

$$\eta(\phi) = \eta \frac{1 + \frac{3}{2}\phi}{1 - \phi} ; \quad \eta_r(\phi) = \eta \frac{\frac{3}{2}\phi}{1 - \phi} \frac{\dot{\gamma} + \omega_x}{\frac{\dot{\gamma}}{2}}$$

where η is the suspending liquid viscosity.

The variation of η_{app}^* with normalised shear rate is shown in figures 6.a and 6.b for various volume concentrations and field intensities. As mentioned before, the value of the low shear rate viscosity plateau does not depend on the field intensity but only on the particle volume fraction. In particular, when ϕ is lower than 1/6, the value of the plateau is negative.

III. EXPERIMENTS

A. Materials

We studied the rheological behaviour of a suspension of PMMA particles (poly-methyl-methacrylate, ICI Acrylics, grade 2041) dispersed into a mixture of transformer oil (dielec S, Hafa, France) and Ugilec (Elf Atochem). The resulting suspending liquid is such that its density is the same as that of the particles, $\rho = 1.18 \cdot 10^3 \text{ kg/m}^3$ and its viscosity at 25°C is 13 mPa.s. Its dielectric constant was measured to be $3.69 \epsilon_0$ and its conductivity has been raised to $1.5 \cdot 10^{-8} \text{ S/m}$ upon addition of AOT salt (Sigma Aldrich, sodium dioctyl-sulfosuccinate). Although Quincke rotation effect is not expected to depend on the particles size, the particles were sieved between 50 and 63 μm . Their dielectric constant is $2.6 \epsilon_0$ and their conductivity is about 10^{-14} S/m so that, compared to the suspending liquid, they are

regarded as insulating. The volume fraction is varied between 5 and 20%. All these physical properties give the following values for the parameters introduced previously:

$$\tau=3 \text{ ms}; E_c=830 \text{ V/mm}; Pr= 720.$$

B. Rheological measurements

The rheological measurements were carried out with a controlled stress rheometer, Carrimed CSL 100, in Couette flow geometry. The coaxial cylinders also serve as electrodes so that the E field is applied along the velocity gradient direction (radial direction). A high voltage is applied to the outer stator (radius R_1) while the inner rotor (radius R_2) is grounded to the earth through an electrode dipping in a mercury reservoir situated at the top of the rotation axis of the rheometer. The gap between the two cylinders (1 mm) is narrow compared to their radii ($R_2=14.5$ and $R_1=15.5$ mm) so that the flow can be approximated by a simple shear flow, $\dot{\gamma}$, where the shear rate is related to the angular velocity of the inner cylinder Ω by:

$$\dot{\gamma} = \frac{\Omega R_1}{R_2 - R_1} \quad (19)$$

Furthermore, since the gap is narrow, the shear stress, σ , can be considered as homogeneous in the suspension and given by:

$$\sigma = \frac{M}{4\pi h} \frac{R_1^2 + R_2^2}{R_1^2 R_2^2} \quad (20)$$

where M is the mechanical torque exerted on the inner cylinder. Then from the measurement of the variation of Ω with M , using relations (19) and (20) the experimental value of the viscosity is determined. The rheological measurements are made at equilibrium: first, in the absence of an electric field, a constant shear stress is applied as long as necessary (≈ 10 s) to record a constant viscosity value, $\eta(\phi)$. Then the E field is applied and the value of η_{app} is

estimated when the shear rate has become constant. After that, the viscosity in the absence of the electric field is again measured to verify that the suspension has not been heated by the electric current which passes through the suspension when the electric field is present.

IV. RESULTS AND DISCUSSION

The variation of η_{app}^* with the normalised shear rate is shown in figures 7.a, 7.b and 7.c, for three volume fractions, 5, 10 and 20%, respectively. The solid symbols and dashed lines correspond to the experimental results while the open symbols and the solid lines represent the theoretical predictions given by Eq.(18) wherein Y has been substituted by its stable stationary value. The qualitative agreement between model and experiment is quite good since there is no free parameter. In particular, for the highest fields and the highest volume fractions, the data display a clear viscosity plateau at low shear rates. Whatever the field intensity, the concordance between experimental data and theoretical values of the viscosity is satisfactory at high shear rates. A discrepancy appears for low values of the shear rate, especially in the case of low field intensities for which the experiments show that the field is responsible for an increase of the viscosity ($\eta_{app}^* > 1$) while the theory predicts a decrease. In a previous paper [Pannacci (2007b)] we have attributed this discrepancy to a structuring of the suspension which occurs at low shear rates and for moderate E field intensities. We have shown in this paper that the E field had two opposite effects on the structure of a suspension. Indeed, it induces dipole-dipole interactions between particles, which tend to structure the suspension but, because of particles' spinning, the E field also gives rise to hydrodynamic forces which tend to break the structures. These two antagonistic actions of the E field explain why no structuring of the suspension takes place for the highest

field intensities, contrary to what would have happened in a classical ER fluid. These theoretical predictions have been confirmed by experimental observations of the suspension structuring carried out with semi-transparent cylinders mounted on the rheometer which allow the observation of the structure of the suspension when an electric field is applied.

For the highest field intensities, the low shear rate viscosity plateau is observed as expected from the model and that's an important point since the presence of this plateau substantiates the existence of the solution X_4 (and associated values of X_0 , Y , Z , ψ and ξ). The model presented in section II and the stability analysis reported in appendix I predicts that the solution X_4 is stable and so is observable for shear rates lower than a critical value, $\dot{\gamma}^*$ which is an increasing function of the particle volume fraction and of the field intensity (Eq.(12)). Even though it is difficult to determine an accurate value for $\dot{\gamma}^*$ from the experimental rheological curves, the low shear rate plateau seems to be longer the higher the field intensity and the particle volume fraction are. Despite this rather good qualitative agreement, it is worth noting that the viscosity decrease is overestimated by the theory. This disagreement could originate, as already said, in the dipolar forces but also in the hydrodynamic interactions between particles which could lead to a dispersion in their spin velocities. There exists a third possible source of the discrepancy which lies in a tendency of the suspension to separate into two phases, one constituted by only the suspending liquid while the other contains a high concentration of particles. This kind of phase separation is illustrated by the picture in figure 8 which represents the Couette cell filled with a suspension containing 10% of particles after application of an electric field of 2000V/mm and of a shear stress of 5 Pa for 5 minutes. Since the value of the viscosity plateau is an increasing function of the spinning particle concentration, the phase separation of the suspension is responsible for an increase of the measured apparent viscosity. We have observed that the demixing was the most pronounced for the most diluted suspension so that the discrepancy of the

experimental and theoretical values of the viscosity plateau is the greatest in the case of the less concentrated suspensions.

V. CONCLUSION

The study of the rheological behavior of a suspension of insulating particles suspended in a slightly conducting liquid submitted to a DC electric has shown that it was possible to drastically reduce the apparent viscosity. The model we use to predict the viscosity reduction is based on the calculation of the angular velocity of the particles subject to a DC field in a shear flow. We have been able to qualitatively explain the viscosity decrease and to derive a model which captures the essential variation of the suspension viscosity with the E field intensity and with the shear rate. In particular, we have shown that if the direction of the axis of rotation of the particles is not fixed, the low shear rate viscosity is expected to be constant and independent of the electric field intensity. These predictions are corroborated by the experimental observation of a low shear rate viscosity plateau.

From an application point of view, this electric field-induced viscosity reduction should develop in an interesting way since it is easy to achieved and the viscosity changes are quite important. We have already demonstrated the possibility of using DC electrorotation to increase the flow rate in a capillary [Cebers (2002)]. Besides these applications in microfluidics, these fluids should be used as smart fluids in a variety of applications, including damping or vibration control.

APPENDIX A: *Stability analysis of the solutions*

A linear stability analysis of system of equations (8) is conducted to discriminate the stable solution from the unstable ones. All the 5 variables, v , of the system (8) are perturbed around their stationary values:

$$X=X_S+\delta X e^{\lambda t^*}, Y=Y_S+\delta Y e^{\lambda t^*}, Z=Z_S+\delta Z e^{\lambda t^*}, \psi=\psi_S+\delta \psi e^{\lambda t^*}, \xi=\xi_S+\delta \xi e^{\lambda t^*}$$

where v_S represents the equilibrium values (solutions of eq.(9) and associated values of the other variables and eq.(10)) and $\delta v e^{\lambda t^*}$ the first order perturbation with λ , the normalised growth rate of the perturbation.

Around v_S , the system of equations (8) is written:

$$\begin{aligned} (\lambda+\text{Pr}/(1-\phi)) \delta X - \text{Pr} \delta Y &= 0 \\ (Z_S-r) \delta X + (\lambda+1) \delta Y + X_S \delta Z &= 0 \\ -Y_S \delta X - X_S \delta Y + (\lambda+1) \delta Z &= 0 \\ (\lambda+1) \delta \xi - (Z_S-r) \delta \psi &= 0 \\ \text{Pr} \delta \xi + (\lambda+\text{Pr}) \delta \psi &= 0 \end{aligned} \tag{A1}$$

To have nontrivial solutions, the determinant of the system of 5 equations has to be zero or:

$$\begin{aligned} \{(\lambda+\text{Pr})(\lambda+1)+\text{Pr}(Z_S-r)\} \left\{ (\lambda+\frac{\text{Pr}}{1-\phi}) [X_S^2+(\lambda+1)^2] \right. \\ \left. + \frac{\text{Pr}}{1-\phi} \left[-\frac{X_S+\frac{\dot{\gamma}\tau}{2}}{X_S} (\lambda+1) + X_S(X_S+\frac{\dot{\gamma}\tau}{2}) \right] \right\} = 0 \end{aligned} \tag{A2}$$

In particular, the determinant is zero if the first factor of the left-hand side of Eq.(A2) is zero:

$$\lambda_{1,2} = \frac{-(\text{Pr}+1) \pm \sqrt{(\text{Pr}+1)^2 - 4\text{Pr}(Z_S-r+1)}}{2} \tag{A3}$$

So, if $Z_{S-r+1} < 0$, or equivalently from Eq.(7) and the first two equations of Eq.(8) if $\frac{X_0}{X} < 0$, there exists a positive real solution for λ and the perturbation grows. As a result, if the directions of the angular velocity of the particles and of the vorticity of the suspending liquid are opposite, the solution can not be stable. Therefore, X_2 and X_3 are always unstable solutions. Now let us find a condition for which X_1 and X_0 have opposite signs. The instability criterion implies that X_1 becomes unstable when X_0 changes its sign. Cancelling X_0 and expressing X_1 in terms of $\dot{\gamma}$ in Eqs. (8) we obtain that X_1 is unstable if $\dot{\gamma}\tau < \dot{\gamma}^* \tau = 2\phi\sqrt{r-1}$.

A numerical study of the second term of the determinant shows that, for the experimental system which will be described hereafter ($Pr \approx 700$, $0 \leq r \leq 20$, $0 \leq \dot{\gamma}\tau \leq 2$), the values of λ which cancel this term are always negative if $\frac{X_0}{X} > 0$ so that the stability criterion defined before is still valid.

As a summary, the stable solutions are X_4 for $\dot{\gamma} < \dot{\gamma}^*$ and X_1 for $\dot{\gamma} > \dot{\gamma}^*$. At last, it should be stressed that if the rotation axis of the particle is fixed ($\boldsymbol{\omega} = \omega \mathbf{e}_x$), the solution X_4 does not exist and the solutions X_1 and X_2 are both stable. This is reported in Cebers (2000) and studied in detail in a recent paper by Dolinsky and Elperin (2007).

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Figure captions

Figure 1: Representation of the particle dipole, \mathbf{P} , coming from the interfacial charge distribution. If the charge relaxation time is higher in the particle than in the host liquid ($\tau_1 < \tau_2$), \mathbf{P} is anticollinear to the field direction (a). If the E field intensity is high enough this situation is unstable and the particle rotates either in clockwise or counter-clock-wise direction. In such a case, the rotation of the particle at constant velocity and the tilt of \mathbf{P} correspond the stationary equilibrium of the system.

Figure 2: Representation of the suspension subject to a DC E field. If the suspension is quiescent, the particles rotate around themselves with any axis perpendicular to the field so that the average spin rate is zero. When a shear flow is applied, all the particle rotation axes may be aligned in the direction of the mean vorticity of the suspension and the collective movement of the particles drives the suspending liquid in such a way that the apparent viscosity of the suspension is lessened.

Figure 3: Variation of the driving electric torque (solid lines) and of the opposite of the viscous resistant torque (dashed lines) exerted on a particle with its angular velocity. The electric torque has been drawn for two intensities of the electric field and the viscous one for two values of the average shear rate in the suspension. Depending both on the values of the field intensity and of the mean shear rate, there exists either one or three solutions for the angular velocity of the particle.

Figure 4: Normalised average angular velocity of the particles of a suspension ($\phi=10\%$). (a): variation with the square of the normalised field intensity for an average normalised shear rate of 0.2. (b): variation with the normalised shear rate for $r=2$. The thick lines represent the stable values of the angular velocity.

Figure 5: Variation of the viscosity of a suspension of PMMA particles dispersed in neutrally buoyant mixture of dielec S and Ugilec with the particle volume fraction. The solid line represents the values of the viscosity predicted by Brenner (1970).

Figure 6: Variation of the normalised apparent viscosity of the suspension with the average normalised shear rate. (a): for different values of the E field intensity and for $\phi=10\%$. (b): for different particle volume fractions and $r=2$.

Figure 7: Comparison between experimental (solid symbols and dotted lines) and theoretical results (open symbols and solid lines) for the variation of the apparent viscosity of a suspension of PMMA particles dispersed in a neutrally buoyant mixture of dielec S and Ugilec with the shear rate for different E field intensities: squares 500 V/mm, triangles 1000 V/mm, circles 2000 V/mm, diamonds 3000 V/mm . (a): $\phi=5\%$, (b): $\phi=10\%$, (c): $\phi=20\%$.

Figure 8: Phase separation in the Couette geometry. The suspension has been subject to a shear stress of 10 Pa and to a DC electric field of 2000V/mm for 5 minutes.















