Chapter 17

Electrorheological Fluids

17.1. Introduction

The study of setting dielectric fluids (liquids, suspensions, etc.) in motion and their flow properties under electric fields and mechanical stress constitutes an important section of scientific, technological and practical investigation which has simultaneously caught the interest of researchers and manufacturers. The physical phenomena originating these laws of hydrodynamic behavior can be grouped into three categories: electrokinetic effects, electroviscous effects and electrorheological effects.

17.1.1. Electrokinetic effects

Electrokinetic effects are produced in the following situations: (i) the presence of a porous material immersed in a liquid; and (ii) when colloidal particles are placed in suspension in a liquid [HUN 81]. The electrical double layer, compact and diffuse (Stern and Gouy-Chapman layers), appearing at the solid/liquid interfaces of the initially neutral medium, reacts to the external solicitation (electric field or mechanical constraint) by implementing a reversible "electrokinetic conversion". Thus, the application of an electric field causes a motion in the fluid (displacement of the liquid or the particles): an electrical-mechanical conversion (motor operating) or the setting in motion of the fluid polarizes the electrode, which then takes a voltage U: the mechanical-electrical conversion (generator operating). By taking the previous examples of dielectric media (i) and (ii), we obtain the following associated

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electrokinetic effects: for motor operating, (i) a electro-osmosis or (ii) a electrophoresis, and for generator operating, (i) a flow potential or (ii) a sedimentation potential, also called Dorn effect.

17.1.2. Electroviscous effects

When an isotropic liquid of weak conductivity placed between two metallic electrodes is subjected to a sufficiently high electric field, an agitation appears followed by the liquid being set in motion. This phenomenon is due to a reaction at the electrode/liquid interface whose effect is the creation of charge carriers (injection phenomenon), the establishment of a space charge, then Coulomb forces exerted on the space charge under the action of the electric field. From a certain applied voltage (critical voltage Vc ~ 10 V for a polar liquid and ~ 100 V for a non-polar liquid), an instability regime appears and the liquid, led by the displacement of the charge carriers, is set in motion (electroconvection phenomenon). In the case where the liquid, subjected to a pressure gradient or shear, flows longitudinally between parallel electrodes supporting an electrical potential V, a secondary transversal flow perturbs the principal flow due to the electroconvection and increases the flow losses [ATT 82]. The apparent viscosity of the liquid can thus be controlled by the potential difference applied between the electrodes. For a strongly polar liquid, the increase in viscosity can be very important (in a ratio > 20).

Another situation containing a monophasic homogeneous fluid based on liquid crystal solutions can be encountered. These fluids, whose anisotropic behavior confers a privileged orientation on them during the application of an electric field, cause a strong increase (in a ratio > 30) of the fluid viscosity [YAN 92]. The principal interest of these fluids is their good stability (no sedimentation) but they also have a few disadvantages: their viscosity without a field is quite high (i.e. high viscous losses) and their operating temperature range is relatively limited.

It is important to note that in electroviscous effects, the fluid always remains in the liquid state (only the apparent viscosity of the fluid is modified).

17.1.3. Electrorheological effects

Electrorheology presents a more general character and is applied to suspensions, generally concentrated, called electrorheological (ER) fluids, of thin solid particles in a dielectric liquid [JOR 89]. ER fluids have the remarkable property of being able to "solidify" in the presence of an electric field, then to get their initial flow property back when the field is removed. Another particularity of ER fluids is that they are able to continuously control the flow resistance of the fluid, i.e. its apparent

viscosity, by an electric field. The obtained variation of viscosity is very important (in the "solid" phase, the fluid viscosity is infinite). We can distinguish two kinds of ER effect: the positive ER effect, which presents most interest, characterized by an increase of the apparent viscosity of the fluid (with possibility of "solidification") in the presence of an electric field [CON 92], [TAO 93] and the negative ER effect, which leads to a decrease in fluid viscosity under the action of the electric field¹ [BOI 95], [LOB 99].

17.2. Electrorheology

17.2.1. Electrorheological effect

The implementation of the ER effect is relatively easy. Numerous powders (starch, cellulose, etc.) can constitute the solid phase of an ER fluid, and a standard insulating liquid can be used as the liquid phase [BLO 88]. The most spectacular manifestation of the ER effect is the reversible change of liquid/solid state of the fluid under the action of an electric field (Figure 17.1).

Electrorheology is an interdisciplinary domain of study which catches the attention of numerous specialists: physicists (for modeling of interactions between particles, formation and behavior of organized and distortable media), chemists (particle synthesis with specific characteristics, elaboration of suspensions), electricians (conduction properties under the high electric field of the dielectric materials), fluid mechanics (rheological study of complex fluids), engineers (conception of new applications). In this chapter, after a presentation of the main characteristics and the composition of ER fluids (choice of solid and liquid constituents), we tackle the study of the basic mechanisms of the positive ER effect in more detail by showing the importance of the conduction properties of solid and liquid dielectric materials in the modeling of interactions between particles.

^{1.} The decrease in fluid viscosity (in the case of a suspension whose particles are more insulating than the carrier liquid) is due to a particular configuration of the solid phase. Under the action of the electric field, the particles acquire a charge (the diffuse layer is driven away by the field) and migrate towards an electrode: the suspension then becomes a medium with two separate phases (liquid and solid). Further, when the suspension is set in motion, the electric field can cause a rotation of the particles according to the vorticity axis (Quincke rotation), whose effect is an increase in the flow speed and therefore a decrease in the apparent viscosity of the suspension.



Figure 17.1. Illustration of the reversibility of the electrorheological effect [BOI 96a]. Top row: suspension of polymer grains (size ~ 2mm) in an insulating liquid; bottom row: suspension of cellulose particles (size ~ 20–30 μ m) in a hydrocarbon (insulating liquid for transformer). (a) The ER fluid is liquid; (b) for sufficient E (E > 2 kV/mm) the particles (or the grains) line up with the field and the fluid becomes a "solid" gel; (c) the fluid becomes liquid again

17.2.2. Characterization of electrorheological fluids

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17.2.2.1. Rheological characteristics

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The basic feature of an ER fluid (rheogram) is given by the variation of the mechanical stress (shear stress) as a function of the velocity gradient (shear rate) for various applied electric fields [LEM 92]. The measurement is generally performed in Couette flow and in cylindrical geometry. Subject to special dimensional conditions, this configuration can reduce to the simple case of a shear between two parallel plane plates between which the ER fluid is applied. These plates, which constitute the electrodes subjected to an electric potential difference V, create a constant electric field E = V/d (d, distance between the electrodes) in the fluid. The typical characteristics obtained show that the rheological behavior of an ER fluid is similar to that of a Bingham body (Figure 17.2):

$$\tau (E, \gamma) = \tau_{S} (E) + \eta_{S0} \gamma$$
[17.1]

where τ : shear stress, τ_s : threshold stress or yield stress, γ : shear rate, η_{s0} : dynamic viscosity of the suspension without electric field, E: applied electric field (the field usually ranges from 2 to 5 kV/mm).



Figure 17.2. Typical rheogram of an ER fluid. The behavior of the fluid is of Bingham-body for $E \neq 0$ and Newtonian-body for E = 0. τ ext: external applied shear stress, τ_{51} , τ_{52} : threshold stress of the fluid flow

The value of the yield stress τ_S is the most important characteristics of an ER fluid. In the case of ER fluids based on micrometer-size particles, the maximum value of τ_S is of the order of 5 to 8 kPa (for an applied electric field about 5 kV/mm). Its dependence regarding the electric field E and numerous other parameters (constituent characteristics, temperature, volume fraction, nature and geometry of the particles, fluid structure, experimental conditions etc.) is not established in a precise manner. Nevertheless, an approximate expression allows a few elements of modeling to be brought out:

$$\tau_{\rm S} \propto \varepsilon_{\rm L} \phi^{\rm m} {\rm E}^{\rm n}$$
 [17.2]

where ε_L : permittivity of the liquid carrier, ϕ : volume fraction of the particles (m ~ 1 for $\phi < 10\%$, m ~ 2 to 3 for $\phi > 20\%$), E: applied electric field (n ~ 2 for E < 0.5 kV/mm, n ~ 1 for E > 2 kV/mm).

Further, each solid or liquid state of the fluid possesses specific characterization parameters:

a) With ER fluid in solid state: under oscillatory shear flow of strain amplitude γ and pulsation ω , we obtain [JOR 92], [MCL 91]:

$$\tau(\omega) = [G'(\omega) + iG''(\omega)]\gamma(\omega)$$
[17.3]

where G': storage modulus (elasticity), G'': loss modulus (viscosity).

b) With ER fluid in liquid state: under steady flow with shear stress τ and shear rate $\dot{\gamma}$, the associated apparent viscosity η_{app} is defined by:

$$\eta_{app} = \tau / \gamma = \eta_0 (1 + K/Mn),$$
 [17.4]

with Mn: Mason number and K a constant:

$$Mn = \frac{\text{Hydrodynamic forces}}{\text{Electric forces}} \cong \eta \dot{\gamma} / 2 \varepsilon_{L} E^{2}$$
[17.5]

The Mason number, the ratio between the hydrodynamic and electric forces, expresses the structure level of an ER fluid [MAR 89]. The electric forces, induced by the field, have a structuring effect (formation of fibers and columns) and tend to "solidify" the fluid. Conversely, hydrodynamic forces, due to the flow, have a destructuring effect (break of fibers, separation of particles) and cause the flow of the fluid. It is therefore the competition between these two antagonist forces which determines the actual state (solid or liquid behavior) of the ER fluid.

17.2.2.2. Electrical characteristics

Concerning the electrical characteristics of the ER fluids, the most significant parameter for the applications is the average current density ($j_{av} = I/S$), where I and S are the current and the cell surface. A magnitude of current density greater than a few tens of μ A/cm2 must be avoided. Indeed, in the contact zone of the particles, where the current densities are strengthened, a degradation of the materials resulting from local heating is possible.

The dielectric spectroscopy analysis of the suspensions (at weak or high electric field and with or without flow) leads to a correlation of the electrical properties of the materials (conductivity and permittivity) to the rheological behavior of the suspension in static or dynamic flow.

17.2.2.3. Energy assessment

In DC voltage, it is easy to express the density of the control losses P_C (power control consumption) and then, by associating the transmitted power density P_T , to determine the power gain G_P and the efficiency η of the system.

For example, in the case of a device composed of two parallel electrodes of surface S, separated by a distance d (input electrode, index I and output electrode, index O), subjected to a mechanical stress (speed V and force F) and an electrical

stress (voltage U et current I) by inserting an ER fluid, we can define the following mechanical and electrical values:

– Speed gradients: of the ER fluid: $\dot{\gamma} = (V_O - V_I) / d$, of the output electrode: $\dot{\gamma}_O = V_O / d$,

- Mechanical parameters: $\tau_0 = F_0 / S$, $\tau_1 = F_1 / S$, viscous losses: $P_V = (\tau_1 - \tau_0) \dot{\gamma}$,

- Applied electric field: E = U / d, average current density: $j_{av} = I / S$,

– Control loss density: $P_C = E.j_{moy}$, transmitted power density: $P_T = \tau_0 \gamma_0$,

We obtain:

$$G_{\rm P} = P_{\rm T} / P_{\rm C}$$

$$[17.6]$$

$$\eta = P_T / (P_T + P_C + P_V)$$
[17.7]

NOTE: In steady shear flow, we have:

- for ER fluid in solid state, $V_0 = V_1$ and $\tau_0 = \tau_1$ (P_V=0)

- for ER fluid in the liquid state, $V_0 \le V_1$ and $\tau_0 \le \tau_1$ ($P_V \ne 0$).

17.2.3. Composition of electrorheological fluids

An ER fluid is a suspension of solid particles (dispersed phase) immersed in a dielectric liquid (continuous phase). The volume fraction ϕ of the solid phase ($\phi = V_S / (V_S + V_L)$) is about 20 to 40%. In order to avoid the sedimentation of the particles, their density must be as close as possible to that of the liquid².

The choice and the characteristics of the materials making up the suspensions condition the ER fluid performances (value of the yield stress, current density, etc.) widely [BLO 90]. A great number of liquid and solid constituents produce an ER effect but until recent years, all experimental results published in this field led to a

^{2.} This restraint disappears in the case of much reduced-size particles. Indeed, the sedimentation speed is proportional to the square of the radius of the particles and the presence of the Brownian motion can inhibit the gravitational effect resulting from the difference in solid and liquid material densities.

relatively low efficiency for the fluids tested (especially for the yield stress strength).

a) Continuous phase: the choice of a liquid is not the most important issue when making up an ER fluid. The liquid must nevertheless be a good electric insulator, even at high temperature and under high electric field, and if possible, have a weak viscosity ($\eta_L < 20$ mPas) and a moderate permittivity ($\varepsilon_r \sim 3$). The commonly used liquids can be listed in three categories: mineral oils with "standard" properties, silicon oils (good temperature stability but relatively poor dielectric performance) and chlorinated liquids ("high" density and permittivity but moderate insulating power) [VUA 01].

b) Dispersed phase: this is the dispersed solid phase which conditions the performances of an ER fluid, almost on its own. The knowledge of their required characteristics leading to production of a strong ER effect was ignored for a long time. It is only over the past 15 years that research has shown that the electrical conduction properties of the materials were crucial in the implementation of the ER effect³ [AND 92], [DAV 92], [FOU 92].

We list below the various types of solid materials (the particles) used, in three categories:

i) The insulating homogeneous materials (silica, starch, cellulose, etc.) requiring the contribution of an external activator (water, surfactant). These materials are not thermally stable in the temperature range of about 0–100°C [IKA 98], [OTS 92];

ii) The homogeneous materials with intrinsic, ionic or electronic conductivity (aluminosilicates, zeolites, semi-conducting polymers, polyaniline, etc.) [GOW 90], [ISH 95];

iii) The inhomogeneous (multilayer) materials with functionalized surface (silica covered with polyaniline, aluminum covered with an insulating layer, etc.) [AKH 99], [WU 98a].

NOTE:

- The materials cited above, which possess a suitable electric conductivity, produce an ER effect in DC voltage but practically no effect in AC voltage. However, some ferroelectric materials (barium titanate, etc.) produce an ER effect under high frequency AC voltage;

^{3.} This finally justifies the observations made for decades on the necessity of using slightly hydrated solid particles to produce an ER effect (anhydrous or hydrophobic particles were without effect) and that particles too hydrated reduced the effect (it is actually water adsorbed at the surface of the particles which supplies sufficient conductivity).

- The composition and geometry of the solid phase (mono or polydispersed particles, particle aspect ratio, etc.) also have an impact on the rheological behavior of ER fluids [QI 02]. However, the size of the particles and their surface functionalization are the most important factors which contribute to producing a very strong ER effect [SHI 94], [WEN 04], [WU 05];

- The dependence of the ER effect on temperature is explained by its role on liquid viscosity, the conductivity of the materials and the surface polarization of the particles. Experiments show that the ER effect (value of the yield stress) passes through a maximum for temperatures ranging approximately between 60 to 80°C [WU 98b].

17.2.4. Applications of electrorheological fluids

The applications of ER fluids turn to good account the ability to control the structure of suspensions by an electric field E. An ER suspension can thus pass from a homogeneous and isotropic dispersion (where E = 0) to an inhomogeneous medium having a strong orientation anisotropy (where $E \neq 0$). The two behaviors of the fluid can operated: with a reversible change in liquid/solid state (switching-mode) or a continuous control of the flow (modulation-mode). The first applications appeared in the world of mechanics (for shock absorbers, clutches, brakes, etc.) but the performance of ER fluids and the powers brought into play make the implementation of such applications difficult [FUR 99], [HAR 91], [MON 97], [STA 92]. Other types of application require little power and turn to good account the easiness of miniaturization of ER devices include micro-wave transmitters, microfluid displays, etc. [FAN 02], [NIU 05], [WEN 05].

17.3. Mechanisms and modeling of the electrorheological effect

Discovered in the 1940s by W. Winslow [WIN 49], the ER effect then gave rise to numerous projects and led to successive advancements concerning the elaboration and characterization of new fluids and new approaches to ER effect mechanisms [BLO 88], [HAV 95], [KLA 67], [UEJ 72], [WEN 03]. However, because of the insufficiency of ER fluid performances (yield stress and temperature range too weak) and a relative lack of knowledge about basic ER effect mechanisms, no decisive advancement regarding application could be realized in the past. The recent discovery of the giant ER effect (see section 17.5.) strongly revived interest in these fluids and permits us now to envisage the implementation of operational ER devices in the near future.

17.3.1. Forces exerted on and between the particles

In the absence of an electric field, the particles immersed in a liquid are subjected to several types of forces: a gravitational force due to the difference of densities in solid and liquid phases (this effect goes hand in hand with the size of the particles) and could lead to their sedimentation; Brownian motion (not very important for particles of size > 1 μ m and for temperatures T < 50°C); and hydrodynamic force (generated by the liquid flow). Further, two types of interaction between particles can also be taken into consideration: the van der Waals forces play a part at nanometric distances which have an actual effect only for particles of very small size (< 0.1 μ m), and electrostatic repulsion forces due to diffuse layers surrounding the particles.

In the presence of an electric field, a dielectrophoretic force (dipole subjected to a field gradient) [POH 78] resulting from the polarization of particles and an electrostatic interaction force (interfacial charges) appears between the particles. Finally, an electrophoretic force (Coulomb force) exists if the particles are charged (with an intrinsic charge or solid/liquid interface effect due to the electrical double layer). In the case of particles more insulating than the liquid, the application of a DC electric field induces electrophoretic forces, which drain the particles towards an electrode: the separation of the solid (particles) and liquid phases leads to a decrease in the apparent viscosity of the suspension (negative ER effect).

NOTE: For ER suspensions composed of micrometer-size particles (common ER fluids), only two force categories must be taken into consideration (the other forces are negligible) to explain the behavior of the fluid: electric (dielectrophoretic and electrostatic) and hydrodynamic forces.

17.3.2. Mechanisms of the electrorheological effect

An ER fluid not subjected to an electric field is a concentrated suspension of solid particles randomly spread out in a liquid and relatively near one another (for a volume fraction of 20%, the average distance between mono-dispersed spherical particles is about 0.7 times the radius of the particles).

The mechanisms leading to the (positive) ER effect are as follows:

1 - From the application of an electric field, the particles get polarized then attract or repel one another, according to their relative position.

2 - The particles which attract each other group together to form a multitude of chains, or elementary fibers, parallel to the field lines (response time of a few milliseconds). The electric current which is established throughout the fibers ensures their preservation and provides them some rigidity. The ER fluid then presents itself as a solid gel.

3 – The application of an external force (shear or squeeze force) distorts the fibers and induces a reaction force from the fluid depending on the distortion level of the fibers and the value of the electric field. For example, in the case of a plane shear, the reaction force increases with the inclination θ of the fibers and reaches its maximal value (yield stress) for $\theta \sim 20^{\circ}$ [CON 92].

17.3.2.1. Polarization and bringing the particles closer

A non-fibrated ER suspension can be perceived as a sequence of solid (particle) and liquid materials. The electrical behavior of such a medium then comes down to that of two dielectric layers in series (a solid layer, of index s and a liquid layer, of index L).

Reasoning for a comparable system but with a simpler geometry: the presence of two plane layers with the same surface, constant thicknesses d_S and d_L , conductivities σ_S and σ_L and permittivities ε_S and ε_L , the response of the system to a voltage V(t) shows the presence of a density of charge q_S at the solid/liquid interface (interfacial Maxwell–Wagner polarization) and defines the distribution of the electric field in the medium.

Thus, for example, the response to an applied voltage of magnitude V leads to:

- The initial state (t = 0⁺): $q_s = 0$, the electric field distribution depends on the value of the permittivities: $\varepsilon_s E_s = \varepsilon_L E_L$, avec $d_s E_s + d_L E_L = V$;

– The final state $(t \rightarrow \infty)$: $q_s = (\epsilon_s \text{ Es} - \epsilon_L \text{ E}_L)$, the electric field distribution depends on the value of the conductivities: $\sigma_s \text{ Es} = \sigma_L \text{ E}_L$.

NOTE: The time constant value τ of the system which corresponds to the temporal limit between the initial state and the final state is:

$$\tau = \frac{\varepsilon_{\rm s} d_{\rm L} + \varepsilon_{\rm L} d_{\rm s}}{\sigma_{\rm s} d_{\rm L} + \sigma_{\rm L} d_{\rm s}}$$
[17.8]

For t $\ll \tau$ (case of high frequency AC voltage): the polarization mechanism is dominant and the field distribution depends on the permittivities of the materials.

For t >> τ (case of steady state in DC voltage): the conduction mechanism is dominant and the field distribution depends on the conductivities of the materials.

Let us now consider a spherical particle of radius R, with conductivity σ_s and permittivity ϵ_s , immersed in a liquid of conductivity σ_L and permittivity ϵ_L . The application of a uniform electric field E_o polarizes the particle and induces an equivalent dipole p:

$$p = 4\pi \epsilon_{\rm L} R^3 \beta E_0$$
 [17.9]

 β depends on the difference in the electrical characteristic values of the materials.

By reasoning in the AC and DC conditions previously defined, we respectively obtain $\beta = \beta(\varepsilon)$ for the AC regime at high frequency and $\beta = \beta(\sigma)$ for the DC regime at steady state, with:

$$\beta(\varepsilon) = \frac{\varepsilon_{\rm s} - \varepsilon_{\rm L}}{\varepsilon_{\rm s} + 2\varepsilon_{\rm L}} \quad \text{and} \quad \beta(\sigma) = \frac{\sigma_{\rm s} - \sigma_{\rm L}}{\sigma_{\rm s} + 2\sigma_{\rm L}} \quad [17.10]$$

The introduction of a second particle, identical to the previous one, at a distance s from the first particle (s >> R), the line Δ passing through the centers of the particles and the direction of the electric field forming an angle θ , the component F_{Δ} of the dipolar interaction force between the particles is expressed by:

$$F_{\Delta}(s,\theta) = 12\pi R^{2} \varepsilon_{L} \beta^{2} E_{0}^{2} \left(\frac{R}{2R+s}\right)^{4} (3\cos^{2}\theta - 1)$$
[17.11]

Equation 17.11 (point-dipole approximation) [AND 94], [KLI 91a] shows that for $\theta <55^{\circ}$ the particles attract each other and they repel one another for $\theta >55^{\circ}$. Consequently, during the application of an electric field to an ER suspension, the nearby particles will either attract (those which are localized in an attraction cone at angle $\theta \cong 55^{\circ}$), or repel, and form rows of chains or fibers separated by a liquid⁴ zone.

For weaker separation distances s (s < R) the previous dipolar model is no longer valid (the polarization charges concentrate in the zone near the particles). The multipolar approach, which allows this situation to be taken into account, leads to a

^{4.} The local and microscopic behavior between the particles (attraction or repulsion) generates a diphasic macroscopic structure in the fluid (the solid fibers composed of particles alternate with the liquid layers).

reinforcement of the interaction between the particles. Then the replacement of the two isolated particles by a set of particles structured in fiber (multi-body system) allows the electrostatic interaction force of a chain of particles to be calculated [CLE 93], [HAL 90], [KLI 91b]. However, this approach which only takes into account the polarization effect of the materials (polarization model of the ER effect) and which is based on the hypothesis that the solid and liquid dielectrics used are perfect insulators (conductivity is zero) leads to theoretical values of the interaction force between particles and the yield stress much less than those experimentally obtained. Furthermore, when the particles get in contact (formation of chains), the calculations show that the electric field in the liquid near the contact becomes infinite.

The consideration of the actual characteristics of the materials and of the experimental conditions (DC or AC voltage) therefore turns out to be very necessary. As we have previously underlined, in a DC electric field and steady state, the distribution of the electric field within the suspension (diphasic dielectric medium) is determined by the conduction properties of the materials.

17.3.2.2. Formation of chains and conduction current

Considerations of a general nature show that under a DC field, the attraction of particles and the formation of stable chains leading to the electrorheological effect require two conditions [BOI 96a]:

a) Conductivity of particles greater than that of the liquid:
$$\Gamma = \frac{\sigma_s}{\sigma_L} > 1$$

NOTE: The induced dipole p (equation [17.9]), with $\beta = \beta(\sigma)$, must be in the direction of the electric field. In the opposite case, the particles get polarized, come closer then turn back simultaneously and separate.

b) Conductivity of particles limited: $\sigma_{S max} \cong 10^{-7} \text{ S/m}$

NOTE: The contact time of the particles (t_{mc}) must be less than the relaxation time of the charges between two particles (t_{rl}) . In this case, chains form between the electrodes and the current which flows through them, holds the superficial charges and induces an electrostatic force of attraction between the particles. It is this force which determines the rigidity of the chains (or the fibers) and leads to the value of the yield stress.

$$t_{mc} \cong \frac{\eta_L}{\epsilon_L E_0^2}$$
, $t_{rl} \cong 10 \frac{\epsilon_L}{\sigma_s}$, with η_L : dynamic viscosity of the liquid.

NOTE: This double condition on the conductivity value of the particles justifies the necessity of using properly hydrated powders to produce an ER effect. It is the

presence of water molecules at the surface of the particles which confers on these latter an appropriate conductivity.

The almost instantaneous structure formation of the solid phase in a chain network evolves and transforms little by little into thick fibers or columns composed of a group of chains (characteristic time of a few seconds to a few minutes) [GIN 95]. In this case, chains inside the columns most often form a body-centered-tetragonal lattice. This structure in columns, which only appears with chains or very extended elementary fibers (with a size of the particles much smaller than the interelectrode distance) reinforces the rigidity of the ER fluid in the "solid" state [GUL 93], [KLI 93], [TAO 91].

17.3.2.3. Flow resistance

The state of the structure of an ER fluid simultaneously subjected to an electric field and a mechanical stress (represented by the Mason number Mn) determines the rheological behavior of the fluid. Three situations can be envisaged:

i) Mn \rightarrow 0 (high E and $\tau_{ext} \cong$ 0): the fluid behaves like a solid gel, it can transmit a force (or torque) without speed slipping (synchronous transmission);

ii) Mn $\rightarrow \infty$ (E \cong 0 and high τ_{ext}): the fluid is in a liquid state and its viscosity, constant, is near that of the suspension in the absence of electric field. The force is transmitted with speed slipping (asynchronous transmission);

iii) Intermediate Mn (moderate E and τ_{ext}): the fluid is in the liquid state and its viscosity is variable, it depends on E and τ_{ext} (controlled asynchronous transmission).

NOTE: Measurements performed on a model ER fluid (suspensions of cellulose powder in mineral oil) showed the direct connection between the structure level of the fluid and, the rheological (apparent viscosity) and electrical (current density) responses of the fluid [FOU 96].

17.4. The conduction model

The conduction model, valid in a DC field or in low frequency AC field, is based on the consideration of the electrical conduction properties of the solid and liquid constituents and the liquid permittivity of an ER fluid to determine a variation law of the attraction force between nearby particles or particles in contact, as a function of the applied electric field and the characteristics of the materials [ATT 94], [ATT 02], [BOI 96b]. This approach, which relies on the results of the electrostatics and electrodynamics of the continuous media, sheds a new light on the understanding of the electrorheological effect and was experimentally verified in a large scale set-up (with an interaction between two millimeter-sized hemispheres) [FOU 94].

17.4.1. The bases of the conduction model

Let us consider the case of a chain formed by spherical particles in contact, immersed in a liquid and subjected to a DC electric field. The cohesive force of the chain is directly related to the attraction force between the particles themselves and, therefore, for symmetrical reasons, to that of two half-spheres in contact (Figure 17.3). Particles are assumed to be more conductive than the liquid ($\sigma_s > \sigma_L$).



Figure 17.3. Half-spheres in contact immersed in a dielectric liquid and brought to a voltage +U and -U, respectively. The current I passes through the half-spheres for $x > \delta$ and through the liquid for $x < \delta$

The distribution of the current between the solid (half-sphere) and the liquid depends on the values of their respective conductances C_S and C_L (C = σ S/l), σ , S and l representing the conductivity, the surface and the length of each element, respectively. Figure 17.3 shows that the passage of the current between the positive electrode and the negative electrode (diametral planes of half-spheres brought to the voltage +U and - U) distinguishes two zones:

Zone 1: $x > \delta$. The conductance of the solid is high (high values of σ_s and S_s) and that of the liquid is weak (weak value of σ_L and high l_L). All of the current passes through the solid. The conductance of the solid being high, the decrease in voltage V(x) due to the passage of current is negligible: the half-sphere can therefore be considered as equipotential: V(x) = U. The electric field $E_1(x)$ in the liquid is in this case equal to:

$$E_1(x) = 2U/y = 2RE_0/y \cong 2R^2E_0/x^2$$
 [17.12]

Zone 2: $x < \delta$. The conductance of the solid is weak (high σ_s but very weak S_s) and that of the liquid is high (weak σ_L but very weak l_L). All of the current passes through the liquid.

In this zone the voltage of the half-sphere decreases and passes from U (for $x = \delta$) to 0 (for x = 0) and the distance y also decreases, passing from x^2/R (approached value) to 0.

The electric field $E_2(x)$ in the liquid is considered constant and equal to:

$$E_{\delta} = 2RU/\delta^2 = 2R^2 E_0/\delta^2$$
 [17.13]

17.4.2. Attraction force between half-spheres

The calculation of the attraction force between the half-spheres is obtained by integrating the electrostatic pressure in the electric contact zone $(0-\delta)$ and the external zone $(\delta-R)$:

$$\mathbf{F} = \int_{0}^{R} \frac{1}{2} \boldsymbol{\varepsilon} E^{2} d\mathbf{x} = 4\pi \mathbf{R}^{2} \boldsymbol{\varepsilon}_{\mathrm{L}} \mathbf{E}_{0}^{2} \left(\frac{R}{\delta}\right)^{2}$$
[17.14]

For $x = \delta$ the current changes of medium (passage from solid to liquid) and the conductance of the liquid C_L (in the external zone, $x > \delta$) is equal to the conductance of the solid C_S (in the electric contact zone, $x < \delta$). δ is deduced from the relation $C_S = C_L$.

The conductance C_S of the solid spheres can be approximated by assuming that the current passes through disks of radius δ (the electrical contact zone). This result can be obtained from the classic capacitance expression, $c = 8\epsilon.\delta$ of a conducting disk of radius δ isolated in an infinite medium of permittivity ϵ . This gives c' = c/2for the capacitance of one face of the disk relative to the corresponding half-sphere of infinite radius. The resistance of the medium lying between a disk face and the corresponding half-sphere of infinite radius is r such that the relaxation time is r.c' = ϵ/σ . We thus obtain the equivalent conductance $C_S = 1/2r$ of the two halfsphere in series.

$$C_{\rm S} \cong 2\sigma_{\rm S}\delta$$
 (conductance of an electric contact between two solids) [17.15]

The conductance of the liquid C_L is obtained by integration of the conductance dC_L of the liquid layer between cylinders of radii x and x+dx: $dC_L = 2\pi\sigma_L x.dx/y \cong 2\pi R\sigma_L dx/x$.

$$C_{\rm L} = 2\pi R \int_{\delta}^{R} \sigma_{\rm L} \frac{dx}{x}$$
[17.16]

The conductance C_L depends on the value of the liquid conductivity [TOB 96]. Further, Equation [17.12] reveals a significant enhancement of the electric field in the liquid for values of x near δ (contact zone). In practice, the field E_{δ} can reaches several tens of kV/mm. For such fields, the liquid conductivity is strongly increased and depends enormously on the value of the field [ONS 34].

Consequently, the calculation of the attraction force between two particles, given by the conduction model (a two zone model), leads to two expressions:

i) In a low applied field ($E_0 < 0.1 \text{ kV/mm}$)

The field in the contact zone remains limited ($E_{\delta} < \sim 3-4$ kV/mm), the liquid conductivity is constant (liquid conductivity in low field $\sigma_{L0} \sim 10^{-12}$ S/m) and δ is independent of E_0 .

$$F = 4\pi R^2 \varepsilon_L K_{\Gamma}^2 \Gamma^2 E_0^2, \text{ with } K_{\Gamma} = [\pi \ln(R/\delta)]^{-1}$$
[17.17]

This law shows that the force depends on the square of the applied electric field E_0 and the square of the conductivities ratio Γ .

ii) In a high applied field ($E_0 > 1 \text{ kV/mm}$)

The field in the contact zone is very high ($E_1 > \sim 8-10$ kV/mm), the conductivity of the liquid is increased and the electric contact zone gets wider (δ increases with E_0).

$$F = 2\pi R^2 \varepsilon_L E_c E_0 \left[\ln \left[(10 \ \Gamma/\pi) (2 \ E_0 / \ E_c)^{1/2} \right] \right]^2$$
[17.18]

The force varies quasi-linearly with the applied electric field and does not depend much on the conductivities ratio Γ .

The conduction model also permits the attraction force F between particles to be expressed as a function of the current I flowing through the fluid and the applied voltage U to the fluid. By using the relation $I = C_s U$ and removing δ between Equations [17.14] and [17.15], we obtain:

$$F = 4\pi\epsilon_{\rm L} (\sigma_{\rm S} R)^2 U^4 / I^2$$
[17.19]

This new expression, which appears in the form of a "universal law", independent of the value of the applied electric field was experimentally verified in a wide range of the conductivities ratio (variation of Γ from 3 to 1,500) [ATT 94]. This law also reveals the correlation between the force (and the yield stress) and the current passing through the ER fluid, and confirms the validation of the conduction model of the ER effect in DC voltage.

NOTE: The conduction model presented above only considers the volumic conduction properties of the liquid. Furthermore, the high field enhanced conduction phenomenon (Onsager effect), due to the dissociation of neutral species, assumes the existence of a "neutral species reservoir" sufficiently important to permanently supply the dissociation. This hypothesis is verified in the case of a large-scale system (e.g. millimetric half-spheres) or for ER fluids with particles of medium size (> 50 μ m) but cannot be kept in the other cases. For ER fluids based on micrometric, and, particularly, nanometric particles, the conduction process of the liquid under high electric fields can be explained by the presence of charge injection phenomena at the particle/liquid⁵ interface [ATT 02].

17.5. Giant electrorheological effect

The giant ER effect discovered in 2003 by researchers from the University of Hong Kong [WEN 03] is characterized by a very strong mechanical response. Measurements carried out on these fluids showed that a yield stress τ_s of the order of 150 kPa could be reached for a 5 kV/mm DC electric field. Such an advance in performance allows operational applications in the domain of mechanical transmissions with quite higher power levels to be envisaged. Other work then confirmed the presence of an intense ER effect ($\tau_{s} \sim 100$ kPa) with nanosuspensions [SHE 05a], [YIN 05]. We can note that this giant ER effect appears with fluids based on nanoparticles (size ~ 50 to 100 nm) functionalized on the surface. In the case of the fluid elaborated by Wen et al. [WEN 03], the suspension is composed of barium titanyl oxalate nanoparticles (size ~ 60 nm, volume fraction $\phi = 30\%$) coated with 5 nm of urea, immersed in a silicone oil. The other characteristic features of these fluids are: an high current consumption ($j \sim 50$ to 100 μ A/cm² for E_(DC) ~ 4 kV/mm), a fairly high intrinsic viscosity (without electric field): $\eta_0 > 1$ Pa.s, and a quasi-linear dependence of the yield stress on the electric field.

The nanometric dimension of the particles and their functionalization (with the presence of a strong surface polarization) can explain the existence of the giant ER effect. At this reduced scale, the Van der Waals forces appearing between the particles in contact [KRU 52], [SHE 05b] on the one hand, and the very high electric field prevailing in the liquid near the contact zone (on the other hand) probably contribute to the strengthening of the attraction force between the particles and as a consequence of the yield stress of the fluid. Indeed, it is well-known that a dielectric liquid can support an electric field even higher when its volume (surface and

^{5.} Charge injection phenomena in the presence of metallic electrodes are fairly well-known [DEN 82], but this is not the case for electrochemical processes relative to organic or inorganic material interfaces (the case of particles in ER suspensions) and insulating liquids.

thickness) is restricted; the Paschen law for gases is the most famous example. We therefore expect a similar result for a liquid [LES 02] but with different behavior laws (existence or absence of charge carriers in the case of extreme liquid confinements, injecting ability of the electrodes, etc.). At the experimental level, the effect of the size of the particles on the rheology of ER nano-suspensions is confirmed: the viscosity increases when the size of the particles decreases [GUE 06], [WEN 04]. This can also explain the high value of the intrinsic viscosity of nano-suspensions.

17.6. Conclusion

The study of the ER effect has seen a renewed interest over the past few years, owing to spectacular advancements with regard to the performance of new fluids based on nanoparticules and functionalized dielectric materials. Operational applications turning the possibility of controlling the structure level of an ER suspension from an electric field to good account are now conceivable. The mechanical sector (for couplers, shock absorbers, etc.) is the most active, but other areas of applications have already been explored: electromagnetic wave transmitters, display devices, etc. The nanometric size of particles allows tremendous yield stress under electric fields to be obtained but also causes, in the absence of a field, strong interactions between particles, whose effects are a high viscosity of the suspension limiting the use of these fluids for applications which only bring into play weak motion speeds. Other advancements which lead to the elaboration of better ER fluids, characterized by an intrinsically weak viscosity, a strong rigidity of the suspension in solid state and a limited current consumption, therefore still remains to be realized but the important point is that the issue resulting from the yield stress weakness of the old ER fluids ($\tau_{\rm S} < 10$ kPa) is now solved.

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