Mechanism of the Dip in Shear Stress of Blended Electrorheological Fluids

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We propose a theoretical model to explain the dip in shear stress as particles of differing conductivity are blended in an electrorheological fluid, a significant effect that has been experimentally verified and is of great practical importance. We propose that for low concentrations of high conductivity particles ϕ_h , clusters of particles are formed due to the enhanced electrostatic forces. The rigid nature of these clusters leads to cavities forming within the stress transferring columns under shearing deformation. These cavities are the reason for the decrease in shear stress, and the minimum is thought to correspond to the formation of a slip plane composed predominantly of cavities. The rise in shear stress as ϕ_h is further increased is due to the formation of percolating chains of overlapping clusters through the columns, which will enhance the stress transfer ability of the material. It is found that this model shows good qualitative agreement with the experimental results.

Key Words : Electrorheological fluid/Conductivity/Blend

1. INTRODUCTION

An electrorheological fluid (ERF) is a suspension of solid semi-conducting particles in an insulating solvent, which shows a dramatic increase in shear stress upon application of an electric field. The basic mechanism is thought to be the electric polarization of the particles induced by the electric field, with the resulting dipole-dipole interactions leading to thick columns of particles spanning the electrodes. Under shear flow, these columns are constantly breaking and reforming, and enhance the transfer of shear stress across the material.^{1), 2), 3)} This type of controllable rheological behaviour has potential application in tunable vibration dampers for example, and presently much work is being carried out toward device development.⁴⁾

In a previous work,⁵⁾ we investigated experimentally the effect on the rheological properties of blending ERF particles of different conductivity. The shear stress was measured under dc electric field of 2kV/mm and a shear rate of 366s⁻¹. The material used was a suspension of carbonaceous ERF particles in silicone oil, manufactured by Bridgestone Corporation.⁶⁾ We found that in a system of fixed overall concentration, as we increased the proportion of high conductivity particles there is a dramatic drop in the shear stress induced by the electric field. However, as we further increase the proportion of high conductivity particles, the shear stress will rise

and eventually approach the value corresponding to the ERF system with only high conductivity particles. This behaviour is shown schematically in Fig.1. The position of the dip (ϕ_h^*/ϕ_{tot}) was found to be at 20% proportion of high conductivity particle, and the magnitude of the dip ($\Delta\tau/\tau_0$) was 75%. These results are important because they show us that the linear law of mixtures will not hold for

Fig.1 Schematic sketch of the variation of shear stress τ of a blended electrorheological fluid under dc electric field and constant shear rate, as a function of volume fraction ϕ_h of high conductivity particle. The overall suspension volume fraction is ϕ_{tot} .

blended systems, even though the electrorheological effect is known to be more pronounced with higher particle conductivities.

As a possible explanation of this behaviour we postulated that there are “rigid” clusters of particles adhering to each high conductivity particle due to the strong electrostatic interactions.⁵⁾ When the fluid is placed under shear flow this cluster will not deform affinely with the rest of the material, thus preventing the column of particles from achieving the optimum configuration for stress transfer. In the present paper we propose a simple physical model of this concept, and confirm that reasonable qualitative agreement with experiment is obtained.

There have been recently many works treating the effect of particle conduction on ERF behaviour, both theoretical^{7), 8), 9)} and experimental.¹⁰⁾ However, perhaps because of the lack of a suitable electrically homogeneous particle dispersion to act as a base ERF system, Sakurai *et al* only work to date for studying the important practical problem of blending ERF particles of different conductivity,⁵⁾ and there have been no theoretical models presented to explain the observed behaviour.

In this work, we present a model considering the effects that a “rigid” cluster of particles adhering to a high conductivity particle will have on the electrorheological properties. In particular we will examine the changes in the stress transfer and deformation characteristics of the particle column spanning the electrodes. In Section 2, we describe the basic model we will use, and in Section 3 we consider from an electrostatic energy viewpoint the process of deforming a column of particles in which a cluster is imbedded. We find that “cavities” will tend to form in the columns and we estimate the resulting decrease in shear stress. As the concentration of high conductivity particles is increased we use the concept of a percolating chain of overlapping clusters to explain the rise in shear stress. In Section 4, we present a brief discussion of the results, and we give a conclusion in Section 5.

2. MODEL

2.1 The ERF Suspension

Fig.2 is schematic sketch of the model we use for a blended system

Fig.2 Sketch of a blended suspension of ERF particles.

of high and low conductivity ERF particles suspended in a carrier liquid.

All the ERF particles are assumed to be spheres of radius a . a is typically several microns, so Brownian motion effects can be neglected. The base particles have electrical conductivity σ_b and volume fraction ϕ_b . The higher conductivity particles have conductivity σ_h and volume fraction ϕ_h , and typically $\sigma_h/\sigma_b \approx 10$. There is a low conductivity coating on all particles to prevent direct particle-particle and particle-electrode electrical contact. We will ignore sedimentation effects.

As in the experiments,⁵⁾ the total volume fraction of particles ϕ_{tot} is kept constant at 30%:

$$\phi_{tot} = \phi_b + \phi_h = 0.3 \quad (1)$$

ϕ_b, ϕ_h are varied from 0 to ϕ_{tot} , but always satisfying eq (1) for every sample.

Regarding the carrier liquid, we assume it is a leaky dielectric (nearly insulating) with electric conductivity σ_s and dielectric permittivity ϵ_s , and viscosity η_s . We have of course $\sigma_b \gg \sigma_s$.

To this suspension of blended ERF particles we apply an external dc electric field E_0 . For simplicity, we assume that the field is uniform through electrode gap. At steady state, this electric field induces a dipole in each particle, due to the difference in conductivities between the particle material and the carrier liquid. For the simplest case of an isolated particle, the dipole moment is parallel to the electric field and its magnitude is (for a base particle) :

$$p_b = 4\pi\epsilon_s\beta_{bs}a^3E_0 \quad (2)$$

where β_{bs} is the conductivity mismatch factor given by

$$\beta_{bs} = \frac{\sigma_b - \sigma_s}{\sigma_b + 2\sigma_s} \quad (3)$$

The dipole p_h induced in a high conductivity particle is given by a similar equation to eq (2), but with σ_h for the particle conductivity in the expression for the conductivity mismatch factor eq (3) (here β_{hs}).

The interactions between the induced electric dipoles will cause the particles to aggregate, with a preference for attachment in the direction of the electric field. To the lowest level of approximation, the dipole-dipole interaction energy U_{ij} between two particles i and j with separation vector $\mathbf{r}_{ij}$ is given by the following equation¹¹⁾ :

$$U_{ij} = -\mathbf{p}_i \cdot \mathbf{T}_{ij} \cdot \mathbf{p}_j \quad (4)$$

where $\mathbf{T}_{ij}$ is the dipole-dipole interaction tensor

$$\mathbf{T}_{ij} = \frac{1}{4\pi\epsilon_s r_{ij}^3} \left(\frac{3\mathbf{r}_{ij}\mathbf{r}_{ij}}{r_{ij}^2} - \mathbf{I} \right) \quad (5)$$

The interaction force $\mathbf{F}_{ij}$ is given by

$$\mathbf{F}_{ij} = \frac{\partial U_{ij}}{\partial \mathbf{r}_{ij}} \quad (6)$$

Actually when particles are close there are multipole and

multibody effects which significantly strengthen the interactions between the particles.^{7), 8), 12), 13)}

Due to these dipole-dipole interactions, there will eventually form thick columns of particles between the electrodes. These are generally believed to be the reason for the increase in shear stress under shear flow.^{1), 2), 3)} This picture is valid for low to moderate shear rates, and for high shear rate (eg. higher than $\sim 1000\text{s}^{-1}$), it has been proposed that a “layered” model of ERF structure is a more realistic representation.^{14), 15)} In this work we will focus on lower shear rates (eg. $< 1000\text{s}^{-1}$), under strong electric fields and so will use the column model as our foundation.

2 · 2 The clusters around high conductivity particles

We now consider the physical reason for the “clusters” that form around the high conductivity particles.

As has been shown by several calculations which take into account the multipole and multibody effects^{7), 8), 12), 13)} the induced electrostatic interactions between spherical particles depend strongly on the degree of mismatch of the electrical properties between the particles and the solvent. Thus, in our present system, the attractive force between a high conductivity particle and a base particle would be much stronger than that between a pair of base particles. Therefore we would expect that, when we externally deform a column of particles, there will appear a cluster of base particles adhering to each high conductivity particle, which will tend to behave as a single “rigid” entity while the surrounding material undergoes deformation, as illustrated in Fig.3. This cluster will not undergo the affine deformation that that particular part of the column would have undergone in the completely uniform case.

We will take a phenomenological approach in this paper, and will model this cluster as a sphere of radius r_{cluster} :

$$r_{\text{cluster}} = \alpha \beta_{hh} a \quad (7)$$

Here α is a dimensionless factor which would depend on the degree of conductivity mismatch β_{hh} , as well as the electric field E_0 .

Fig.3 The “rigid cluster” of particles adhering to the central high conductivity particle within a column due to the enhanced electrostatic forces under the electric field E_0 . Arrows indicate magnitude of polarization.

Chen *et al*⁷⁾ estimate the relationship between particle-particle force and particle-particle distance. They say particle-particle force becomes very small when $\alpha > 2$. So we can estimate that $\alpha \approx 2$.

Finally we note that in the rest of this paper we will refer to this rigid entity in a column as a “cluster”.

3. CALCULATIONS

3 · 1 Cavity formation

In this section we will show that under shear flow conditions, cavities will form around the clusters in the columns, reducing the stress transfer ability of the material. We will use an argument based on the two competing effects of the “elastic” energy cost $\Delta U_{\text{elastic}}$ of a column deforming near an imbedded cluster, and the “surface” energy cost ΔU_{cavity} of a cavity forming around a cluster in a column. Both of these energy concepts arise from the electrostatic energy of the system. There have been several works treating ERF from the point of view of energy, including electric field induced phase transition,¹⁶⁾ energy changes during aggregation,¹⁷⁾ and energy of structures under shear deformation.^{12), 18)} When considering these energy arguments, we should note that strictly speaking, there will also be Joule heating due to the electric current. However, the magnitude of this is estimated to be very small (typically the power density will be $\sim 0.01\text{W/cm}^3$ under 2kV/mm field), and there is no noticeable rise in temperature of the ERF at the end of the 30s measurement. In this paper we will take a continuum mechanics approach and treat the bulk of the column as a continuous medium.

First, we consider the columns and the distribution of the clusters in them. Consider a system where columns have formed across the electrodes, as in Fig.4a. The electrode gap is written as h and the width of each column is r_{col} . The average number of clusters per column is

Fig.4 The formation of the “cavity” around a cluster within a column under shearing deformation. Beginning with the configuration in (a), under small shear strain γ the column locally deforms around the cluster ((b)), until the cavity becomes energetically favourable at larger strains $\gamma > \gamma_c$ ((c)). After the cavity is formed the column’s capacity to transfer stress is significantly reduced.

$$n_{clusters} \sim \frac{\phi_h}{\phi_b} \left(\frac{hr_{col}^2}{a^3} \right) \quad (8)$$

(In this paper we will present mostly scaling arguments with the numerical coefficients removed, and will use the symbol $\sim$ in the equations). Assuming the clusters are uniformly distributed within the columns, the average distance between clusters in a column is

$$r_{av} \sim \left[\frac{hr_{col}^2}{n_{clusters}} \right]^{\frac{1}{3}} \quad (9)$$

Substituting eq (8) into eq (9) yields

$$r_{av} \sim a \left(\frac{\phi_b}{\phi_h} \right)^{\frac{1}{3}} \quad (10)$$

Now let us consider the “elastic” energy cost $\Delta U_{elastic}$ of a column deforming around an imbedded cluster. Let us assume that the top electrode is displaced slightly by a small strain γ as shown in Fig.4b. In this configuration, the column will have a base electrostatic energy which can be treated as a type of elastic energy,^{(18), (19), (20)} and we now consider the increment in the energy due to the extra deformation caused by the presence of the cluster. In the spirit of the previous calculations, we will model the column material between the electrodes as an “elastic body” with an “elastic modulus” Y determined by the interparticle forces between the base particles:

$$Y \sim \beta_{bs}^2 E_0^2 \quad (11)$$

However, due to the presence of the “rigid” clusters, the column must locally deform its preferred affine deformation to go around the clusters, as indicated in Fig.4b. The extra elastic strain energy per column volume due to these local disturbances is

$$\Delta U_{elastic} \sim n_{clusters} \tau_{elastic} \Delta \gamma \quad (12)$$

Here $\Delta \gamma$ is the extra strain (in addition to the externally applied affine strain γ) caused by the disturbances due to the clusters. $\tau_{elastic}$ is the characteristic elastic stress in the column.

From simple geometrical considerations, $\Delta \gamma$ can be approximated by

$$\Delta \gamma \sim \frac{r_{clusters}}{r_{av}} \gamma \quad (13)$$

and $\tau_{elastic}$ can be estimated by

$$\tau_{elastic} = Y \gamma \sim \beta_{bs}^2 E_0^2 \gamma \quad (14)$$

Substituting eqs (7), (10), (13) and (14) into eq (12) gives us

$$\Delta U_{elastic} \sim \alpha \beta_{bh} \beta_{bs}^2 E_0^2 \left(\frac{\phi_h}{\phi_b} \right)^{\frac{4}{3}} \left(\frac{hr_{col}^2}{a^3} \right) \gamma^2 \quad (15)$$

Recall that this quantity $\Delta U_{elastic}$ is the extra energy cost of maintaining the configuration in Fig.4b; that is, the extra elastic

strain energy due to the disturbances caused by the clusters.

We now further increase the strain γ and when it exceeds a certain critical value γ_c (calculated later) a cavity will form around the cluster (Fig.4c), since the total energy of this configuration will become lower.

We now consider the “surface energy” energy cost ΔU_{cavity} of a cavity forming around a cluster in a column. Recalling that the column will have a base electrostatic energy, we now consider the increment in the energy due to the formation of the “cavity” and the newly created interface between the carrier liquid and the ERF particles. The modelling of ERF particle aggregate surfaces by a surface energy deriving from the electrostatics has been discussed and calculated in detail by Toor and Halsey,⁽²¹⁾ Davis,⁽²²⁾ Toor⁽²³⁾ and Clercx and Bossis.⁽²⁴⁾

The magnitude of the surface energy S per surface area⁽²¹⁾ at the interface between the carrier liquid and particle aggregate is

$$S = \beta_{bs}^2 E_0^2 a \quad (16)$$

The surface area A_{cavity} of the cavity formed will be approximately equal to

$$A_{cavity} \sim r_{cluster} w \quad (17)$$

Here w is the width of the strip of particles peeled off to form the cavity. We assume that w is of the order of several a .

Thus the extra surface energy ΔU_{cavity} due to these cavities per column volume V_{col} is

$$\Delta U_{cavity} = n_{clusters} S A_{cavity} / V_{col} \quad (18)$$

Substituting eqs (8), (16), (17) into eq (18), we obtain

$$\Delta U_{cavity} = \frac{\phi_h}{\phi_b} \left(\frac{hr_{col}^2}{a^3} \right) \beta_{bs}^2 E_0^2 a r_{cluster} w \frac{1}{hr_{col}^2} \sim \frac{\phi_h}{\phi_b} \beta_{bs}^2 E_0^2 \alpha \beta_{bh} \quad (19)$$

Let us now compare these two competing processes, by considering the ratio of the energy costs of the two configurations (Fig.4b versus Fig.4c):

$$\frac{\Delta U_{cavity}}{\Delta U_{elastic}} \sim \left(\frac{\phi_h}{\phi_b} \right)^{\frac{1}{3}} \left(\frac{a^3}{hr_{col}^2} \right) \frac{1}{\gamma^2} \quad (20)$$

We substitute the typical values $\phi_b/\phi_h \sim 10$ and $h \sim 100a$. We also note from the simulations of column formation reported by Toor⁽²³⁾ that $r_{col} \sim 10a$, indicating that we can estimate $a^3/hr_{col}^2 \sim 10^{-4}$. If we now consider the critical state of cavity formation and set $\Delta U_{cavity}/\Delta U_{elastic} \approx 1$, eq (20) gives us for the critical strain

$$\gamma_c \sim 10^{-\frac{11}{6}} \quad (21)$$

which is a very small strain indeed. So we would expect cavities to form in ERF's under steady or oscillatory shear unless the shear strain is extremely small.

Thus under constant shear conditions, we see that there will be a

strong tendency for cavities to form in blended systems, and the shear transfer strength of the columns will be consequently reduced.

3·2 Decrease in shear stress at low ϕ_h

Here we consider the physical mechanisms for the reduction of shear stress with small amounts of high conductivity particles, due to these cavities. We model the decrease in shear stress by modifying a model often used in ceramics to estimate the effect of air pores on ceramic mechanical strength.

First of all we note that the presence of the cavities in the columns will reduce the ability of the column to transfer shear stress. There are a number of possible mechanisms for this:

- (a) The maximum stress that a column will be able to transfer will be reduced since there would be a weakening of the dipole-dipole interactions through the cross section of the column containing the cavity. This is because of the reduction of the multibody reinforcing effect of the polarised particles in close proximity.
- (b) Since the maximum stress of the column is reduced, there will also be a reduction of the critical strain at which the column will rupture. Thus these columns will spend less time on average participating in stress transfer compared to columns without cavities (i.e. the uniform particle case).

As ϕ_h is increased, we would expect both of these effects to be more pronounced and consequently a larger reduction in stress transfer through the ERF.

We now quantify this reduction in shear stress with increasing ϕ_h . One approach would be to base our calculations on the theoretical microscopic model proposed by Gilman²⁵⁾ and Ahlquist,²⁶⁾ which relates the mechanical strength of crystalline solids to the dislocation concentration. This model is also based on considerations of electrostatic energy. However in this paper we will modify and use a different model, which has been widely used to explain the mechanical properties of ceramics. This model relates the mechanical strength of ceramics to the amount of air cavities present,^{27), 28), 29)} the essential idea being that the cavities will accelerate crack propagation and so reduce the overall strength of the material.

We should note of course that there are significant differences between the fracture of a ceramic, and the deformation and rupture of a column of ERF particles, the most obvious difference being the brittleness of the ceramic material as opposed to the fluidic nature of the ERF aggregates. Nevertheless, we believe that the basic physical picture of a material containing cavities under mechanical stress, is still applicable when we consider the instantaneous strength of a column of ERF particles. We will also make the reasonable assumption that this reduced strength of the columns will translate directly to reduced shear stress of the ERF. Thus, using the model of the air cavities in ceramics,^{27), 28), 29)} we can write for the reduction in shear stress $\Delta\tau/\tau_0$:

$$\frac{\Delta\tau}{\tau_0} = 1 - \exp(-\kappa\phi_{cavities}) \quad (22)$$

Here $\phi_{cavities}$ is the volume fraction of cavities in the column (with respect to volume of column). κ is a numerical constant, which is approximately $\kappa \approx 5$ from the results of Spriggs.²⁸⁾ If cavities exist in ceramics of original modulus of E_0 , its shear modulus E is obtained

as $E = E_0 \exp(-\kappa\phi_{cavities})$ under the fabrication technique.

Using this value of k and an experimentally typical value of $\phi_h^*/\phi_{tot} = 0.2$, we find from eq (22) after a simple calculation

$$\frac{\Delta\tau}{\tau_0} = 0.5 \quad (23)$$

which is of similar magnitude as the dip curve measured experimentally. We thus see that although eq (22) was originally developed for modelling the effect of air cavities on ceramic mechanical properties, it also appears to provide a reasonable picture for the reduction in shear stress of the ERF due to cavities as ϕ_h is increased.

We now move on to consider what happens at the bottom of the “dip” (i.e. the minimum of shear stress). The essential idea is that when ϕ_h increases, a “slip layer” will form with most column breakages occurring in a shearing layer predominantly occupied with cavities. That is, at this value of ϕ_h , there are enough cavities to almost occupy a shearing layer at some height through the columns. Since there are mostly cavities in this slip layer, re-attachment of columns and their reformation to the optimum configuration for stress transfer will be impeded.

We will now estimate the volume fraction ϕ_h at which this slip layer will form. As shown in Fig.5, we would expect a slip layer when there is a planar region parallel to the electrodes of height of order $r_{cluster}$ in which almost all columns would have a cavity. Clearly, from simple geometrical considerations, this will occur when the distance between cluster centres r_{av} within a column is of the order $r_{cluster}$. So we set

Fig.5 The formation of the “slip plane” when cavities predominantly occur in one shear plane through the columns. The ability of the fluid to transfer shear stress is minimal when this occurs, and this corresponds to the volume fraction ϕ_h^* in Fig.1.

$$r_{av} \approx r_{cluster} \quad (24)$$

Using eqs (7), (10) and substituting typical values ($\alpha = 2$ and $\beta_{bh} = 0.75$) gives us $\phi_h^*/\phi_{tot} \sim 0.22$ for the slip layer to form. This is of the correct order of magnitude for the dip position.

Here in Figs 4 and 5, only one cluster is shown for brief consideration of the effect of one cluster on the column. Actually there exist a large number of clusters in one column randomly distributed, but each cluster is expected to be in a situation as depicted in Figs 4 and 5.

3.3 Increase in shear stress at higher ϕ_h : Percolation of clusters

We now propose a model for the increase in the shear stress above a certain value of ϕ_h (i.e. the reason for the dip). The basic idea is that for higher values of ϕ_h we will get percolation of clusters through a column which will lead to increasing strength (Fig.6a). Thus the columns achieve larger stress transfer and consequently have a larger strain at rupture, leading to increased overall shear stress. Note that this model is also able to explain the continuous increase of shear stress with larger ϕ_h .

Fig.6 With increasing volume fraction ϕ_h of high conductivity particles, a percolating chain of overlapping clusters will form and the shear stress transferred will rise ((a)). As ϕ_h is further increased, more percolating clusters are formed and the shear stress continuously rises to the value of the suspension with only high conductivity particles ((b)).

To check that this model qualitatively agrees with experiment, we will estimate the volume fraction ϕ_h^{perc} at which the percolation of clusters will occur. It is known from percolation analysis³⁰⁾ that the threshold volume fraction for percolation ϕ_{sphere}^{perc} for a system of randomly placed spheres is

$$\phi_{sphere}^{perc} \approx 0.15 \quad (25)$$

Although eq (25) is for infinite (or near-infinite) systems, let us use this result to estimate ϕ_h^{perc} by considering the percolation of clusters in a column.

Noting that the clusters' volume fraction inside the column is given by the total volume of the clusters ($\sim n_{clusters} r_{clus}^3$) divided by the column's total volume ($\sim hr_{col}^2$), and using eqs (7), (8) with (25), we have

$$\frac{\phi_h^{perc}}{\phi_h} \alpha^3 \beta_{bh}^3 \approx 0.15 \quad (26)$$

This is the volume fraction at which the clusters will form a

percolating chain through the column. Note that for the strengthening effect of the percolating clusters, we will require that there be some overlap between the clusters, which indicates we set $\alpha \approx 2$. Substituting other typical values into eq (26) yields $\phi_h^{perc}/\phi_{tot} = \phi_h^*/\phi_{tot} \approx 0.05$. This figure is somewhat lower than the figure of $\phi_h^*/\phi_{tot} \approx 0.2$ measured in our experiments, but may be the best we can expect from such a crude model. Here we estimate the distribution of clusters at the dip position of ϕ_h/ϕ_b . We used particles of $a = 3.5\mu\text{m}$ giving $r_{cluster} = 5.25\mu\text{m}$, so the volume fraction of clusters in the column will be 68% for $\phi_h/\phi_{tot} = 0.2$, which is of the expected magnitude. We also should keep in mind that we are assuming only that the dip position will scale as the percolation threshold, and that there need not be precise quantitative agreement. Nevertheless, this idea of percolation of the clusters leading to stronger columns is able to neatly explain the position of the dip in shear stress at lower volume fractions ϕ_h .

As ϕ_h is further increased, more of these percolating chains of clusters will form, leading naturally to stronger columns (Fig.6b). Thus there will be a smooth transition to the state with all high conductivity particles.

Note that in this model the volume fraction of particles for the dip is assumed to depend on the ratio of σ_h/σ_b , since $r_{cluster}$ will vary with σ_h/σ_b . For example, when σ_h/σ_b is relatively large, $r_{cluster}$ will be larger and the volume fraction for the dip will be smaller. Thus we expect the position of the dip will not be symmetric with respect to the proportion of high and low conductivity particles. The relationship between the dip position and σ_h/σ_b will be studied in more detail in a future work.

4. DISCUSSION

We have seen that this model, based on cavity formation at low ϕ_h and percolation of clusters at high ϕ_h in the columns of ERF particles, is able to explain the dip in shear stress in blended systems which has been measured experimentally.⁵⁾ Let us now discuss the experimental conditions under which this model would be applicable.

First, we should note we have been assuming strong electric fields and low to moderate shear rates. In terms of the non-dimensional quantity the Mason number ($Mn = \eta\dot{\gamma}/2\epsilon_s\beta_{hs}^2E_0^2$) used by many researchers (see for example the review of Zukoski³¹⁾), we are considering the low Mn regime. We require the electric field effects to be strong to ensure that thick columns of particles will form between the electrodes, as has been confirmed by computer simulations.²³⁾

We now consider the effect of increasing the shear rate. If we were to keep the electric field constant and raise the shear rate, the system will undergo a transition to a "layered" structure, with solid layers of particles adhering to the electrodes and a free-flowing layer in the centre where all the flow is concentrated.^{14), 15)} It has been confirmed that even within the framework of this "layered" structure model, the dip in shear stress for blended suspensions can be explained.³²⁾ Thus the concept of columns is not essential for the understanding of this dip behaviour. However, we should also keep in mind that as we increase the shear rate, we are increasing the plastic viscosity contribution of the shear stress. If we recall that the rheological

response of ERF is usually modelled as a Bingham fluid, we would expect the relative magnitude of the dip to decrease with shear rate since the dip is essentially due to the induced dipole interactions between the particles, corresponding to the Bingham yield stress contribution. Measurements of the shear rate dependence of the dip are reported in our other work.³³⁾

5. CONCLUSIONS

We have proposed a simple model to explain the “dip” in electric field induced shear stress when ERF particles of differing conductivity are blended.

The essential idea is the rigid “clusters” of particles forming around each high conductivity particle, due to the enhanced electrostatic forces, which prevent affine deformation of the columns under shear deformation. At low concentrations of high conductivity particles, considerations of elastic versus surface energies (both of which arise from the electrostatic interaction forces), show that a cavity will form around each cluster, which will severely weaken the column and thus the stress transfer. We find that this effect can be quantified by modifying the model often used for the effect of air cavities in ceramics. The minimum of the shear stress dip is thought to correspond to the formation of a slip zone predominantly containing cavities in a single shearing plane.

The reason for the increase in shear stress as the concentration of high conductivity material is further raised is well explained by the formation of a percolating chain of clusters in the columns between the electrodes. These chains will increase the stress transfer capability of the columns. Further increase in concentration will cause the formation of even more percolating chains, increasing the strength even further, until it approaches the value of the ERF composed of only high conductivity particles.

The degree of uniformity of electrical properties of ERF particles is of great practical importance. This paper has attempted to clarify the mechanism of the dip in shear stress in blended suspensions.

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