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The squeeze flow behaviour of highly concentrated suspensions of spheres in a Newtonian fluid is studied experimentally. Analysing the evolution of the squeeze force as a function of time for different controlled velocities, the suspension is found to present two main flow regimes. The first regime is characterised by the situation in which the force decreases when the velocity decreases, which corresponds to a viscous flow of the suspension. In the second regime, the force increases when the velocity decreases, which is shown to correspond to a filtration of the solvent through the particle skeleton that behaves then as a deformable porous media. By varying the concentration, the sphere diameter and the solvent viscosity, it is found that the transition between the two regimes is a result of a competition between the viscous shear forces due the flow of the suspension and the damping force caused by the filtration of the fluid through the porous media made up by the particles.

Keywords: Suspensions; Squeeze flow; Flow-induced heterogeneity; Filtration; Pastes

1. Introduction

Squeeze tests are often utilised in practice as a straightforward technique to determine the flow properties of highly viscous materials such as concrete, molten polymers, ceramic pastes etc. The material is squeezed out between two parallel surfaces at either controlled normal force or squeezing speed, and assuming a rheological model, the flow parameters of the material are inferred by fitting the model with the experimental measurements. For small gaps and if the no-slip boundary condition is fulfilled, the flow is dominated by shear and the lubrication-type approach can be used to determine the relationship between the applied force (or velocity) and the rheological parameters of the material. In an early study, Stefan and Sitzungber [1] solved the case of a Newtonian fluid and Scott [2] considered the case of power law fluids. On the other hand, if the material slips along the solid surface, the kinematics is dominated by the extensional component of the flow. This gives a particularly interesting set-up to study the bi-axial

elongation properties of such materials. More recently, Laun et al. [3] considered the case of partial wall slip. In particular, they reported a method, similar to that of Moony [4], for a capillary rheometer, to make a separation between the bulk shear and the wall slip from a single squeeze test.

In all the above studies, the material is supposed to have a pre-defined rheological behaviour. In particular, even complex materials are often assumed to remain homogeneous during the squeezing process and behave as simple power law fluids. Many complex fluids become heterogeneous when subjected to flow; this can induce a radical change in their rheology. For instance, this is the case for polymer melts that partially crystallise in flow fields. To our knowledge, the problem of flow-induced crystallisation in squeeze flows and its consequences on such a rheological technique is still open.

The flow-induced heterogeneities in multi-component systems have also been reported in the literature. Racineux [5] considered the rheological behaviour of highly concentrated ceramic pastes in both capillary and squeeze geometries. He showed that his experimental results could be correctly modelled only by assuming a phase separation in which the particle concentration increases in the low shear regions. However, the physical origins of this phase separation are not well understood.

In our study, squeeze flow experiments are conducted on a model, well-defined fluid–particulate system (suspension). Our objective is to highlight the physical mechanisms responsible for this flow-induced heterogeneity. Concentrated suspensions often become heterogeneous in complex flows. For example, it is well known that non-uniform shear flows induce particle migrations in concentrated suspensions [6]. The origin of this phenomenon is well understood and attributed to irreversible interactions (collisions) among the particles [7]. This process is diffusive [6,8] and is shown to be negligible in our experiments.

The experimental parameters and procedures are described in Section 2. In Section 3, we present the results along with a qualitative interpretation demonstrating a transition between a viscous flow of a suspension and a solvent filtration through a porous media made up by the particles.

2. Experiments

2.1. Squeezing set-up

The suspensions are squeezed out between two parallel discs mounted on a compression–traction machine. The upper disc can be displaced at controlled velocities, while the lower one is maintained stationary. The latter experiences a normal force that is recorded as a function of time for each fixed velocity. In all experiments, the initial disc separation is taken to be 5 mm, which is much smaller than the disc diameter (40 mm). In order to avoid a significant deformation of the particles, the squeezing experiment is stopped when the normal force exceeds 800 N. Since the compression–traction machine used here is designed for studying rather solid materials, the uncertainty in the force measurements is quite high (± 2 N). On the other hand, the disc separation (velocity) is measured with a much higher accuracy, namely ± 1 mm.

In the visualisation experiments, the lower disc is transparent (glass). Hence, owing to a CCD camera associated with an image analyses software (OPTIMAS), the evolution of the microstructure due the squeeze flow can be followed, at least qualitatively, since at high concentrations, the suspensions are opaque.

2.2. Suspensions

The suspensions consist of hard spherical particles dispersed in a Newtonian fluid. The particles are polymethylmethacrylate (PMMA) (kindly supplied by the company 'GAZECHIM matières plastiques').
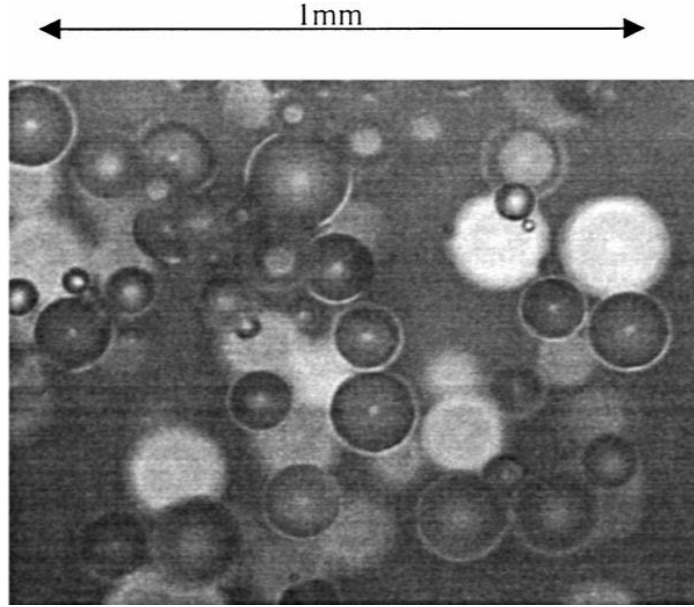
Fig. 1. Visualisation of a sample of the particles using an optical microscope.

A photograph of a sample of the particles is represented in Fig. 1. The shape of the particles is fairly spherical, but there is quite a strong polydispersity in their diameter distribution (Fig. 1). Four batches of particles are used, A, B, B1 and B2, corresponding to different average diameters, respectively: 117, 79, 69, and 89 μm. Batches B1 and B2 are obtained by sieving batch B; they are then less polydisperse. Samples of about 100 particles are analysed, and the particle size distribution functions are found to present approximately a Gaussian shape. The breadth of this distribution is about 50% in the case of batches A and B, and 20% in the case of the two other batches.

Three different suspending fluids are used: a 20%/80% water/glycerol solution, pure glycerol and silicone oil. Their viscosities are measured using a Couette device (Stresstech from Rheologica). The viscosity of the solution water/glycerol is 0.085 Pa s. At 25°C, the glycerol and the silicone oil are found to be Newtonian up to a shear rate of 10^2 s^{-1}, with a viscosity of 0.86 and 12.5 Pa s, respectively. The density of the particles is sufficiently close to that of the solvents so that we can ignore sedimentation effects. Indeed, in the worst case (highest sedimentation rate), the characteristic mean sedimentation time (the time required for a particle to sediment over its diameter) can be estimated as [14] $\tau_s = (18\mu/(\Delta\rho g d))$ $(1-\phi)^{-6.55} \approx 2000$ s. In this expression, μ is the viscosity of the solvent, $\Delta\rho$ the difference between the density of the particle and that of the solvent, d the average diameter of the particles, and where ϕ is the volume fraction. This sedimentation time is higher than the largest corresponding flow characteristic time (the time required for the particle to move over its diameter due to the flow) involved in our experiments, which can be estimated as $\tau_f = d/U \approx 7$ s. U is the squeezing velocity. Finally, the largest particle Reynolds number in our experiments is $Re = (\rho U d/\mu) \approx 2.5 \times 10^{-3} \ll 1$; inertia is then negligible.

3. Experimental results and discussion

3.1. Viscous flow behaviour

Fig. 2 represents the evolution of the squeezing force versus the gap distance h for different velocities V. This corresponds to the case of a suspension in silicone oil with a volume concentration of 56.6%.
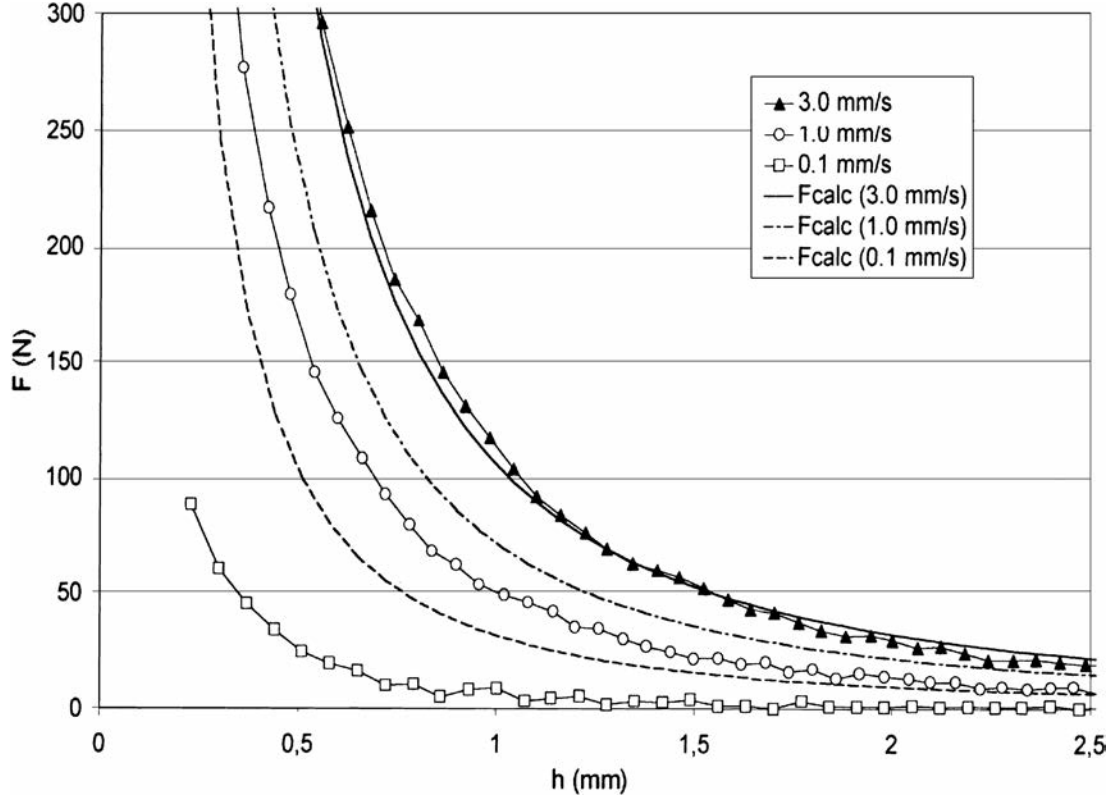
Fig. 2. Temporal evolution of the normal force for different velocities in the case of a concentrated suspension (56.6%) for a relatively high viscous fluid (silicone oil of 12.5 Pa s). The experimental results (dotted lines) are compared to a theoretical calculation assuming that the suspension has a power law fluid behaviour.

The particles are taken here from Batch B1. The suspension behaves as a viscous fluid: the force is an increasing function of the velocity for a given gap thickness and diverges when the gap approaches 0. Nevertheless, the behaviour of the suspension is not Newtonian, contrary to that one would expect for non-Brownian hard spheres in a Newtonian fluid undergoing only hydrodynamic interactions. Moreover, the suspension can not be modelled as a simple power law with the same exponent for all the squeeze speeds considered. Indeed, in Fig. 2, we also represent the predicted temporal evolution of the force for the Oswald de Waele power law fluid. We used the Scott result for the normal force [2]:

$$ F = 2\pi \left(\frac{2m + 1}{m} \right)^m \frac{A}{m + 3} \frac{V^m}{h^{2m+1}} R^{(m+3)}. $$

The rheological parameters m and A involved in this expression are related to the constitutive equation for a power law fluid which can be written in the case of a shear dominated flow as $\tau = A\dot{\gamma}^m$, where τ is the shear stress and $\dot{\gamma}$ the shear rate. The best fit of Scott's equation with the experimental results gives $A = 594.6$ Pa s^m and $m = 0.5$. For a Newtonian fluid, $m = 1$ and A is its Newtonian viscosity; since $m < 1$, the suspension behaves as a shear thinning fluid. The shear thinning effect can not be attributed to the solvent since the silicone oil is Newtonian in the shear rate interval involved in this experiment ($\dot{\gamma} < 5$ s^{-1}). This can rather be attributed to non-hydrodynamic close contact interactions among the particles. As we can see in Fig. 2, the experimental results for the other velocities can not be fitted using the same rheological parameters. The suspension can not be then modelled as a simple power law fluid. Other squeeze experiments have been conducted at smaller velocities for which similar behaviour has been obtained.
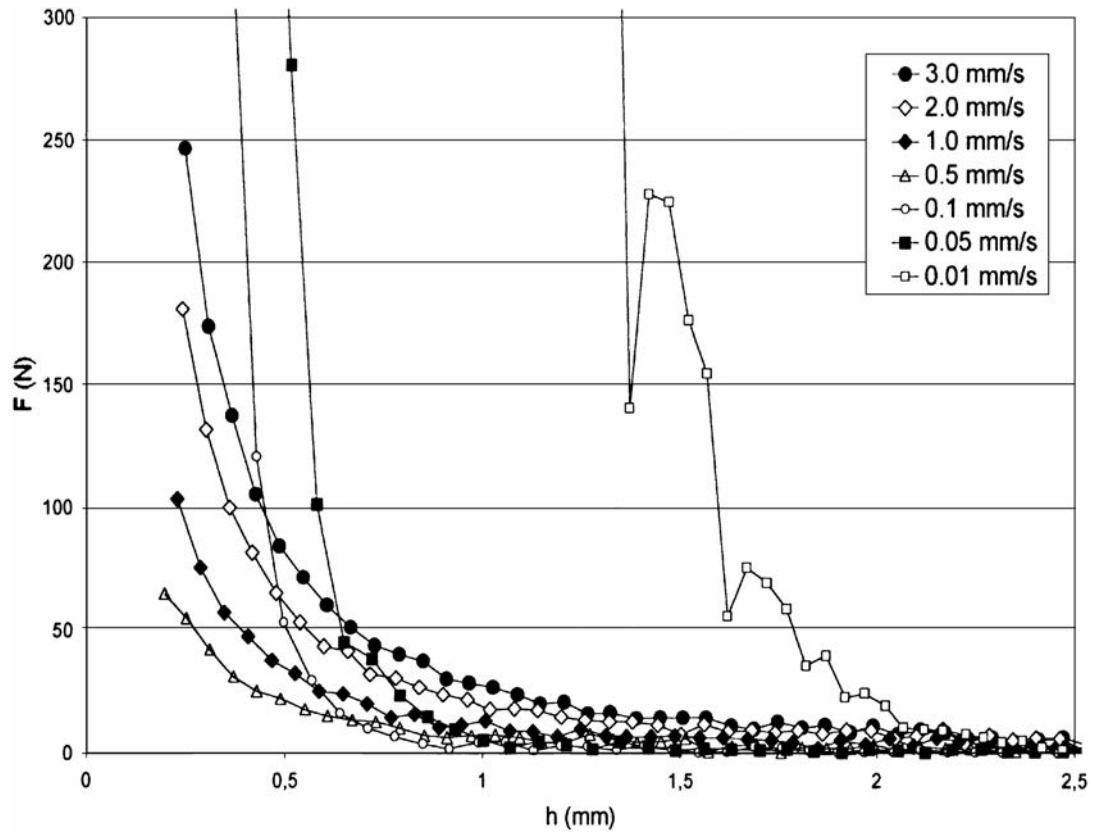
Fig. 3. Temporal evolution of the normal force for different velocities, in the case of a concentrated suspension (56.6%) in a less viscous fluid (glycerol) than in Fig. 2.

3.2. Filtration

In Fig. 3, the temporal evolution of the normal force is represented for different velocities, for the same batch (B1) and concentration as that given above, but with a less viscous solvent (glycerol). The squeeze behaviour of the suspension is radically different from that with silicone oil (see Fig. 2). Above a certain velocity ($U_c = 0.5 \, \text{mm s}^{-1}$), the results are similar to those obtained with a suspension in silicone oil: for a given gap thickness, the force increases with the velocity. Below U_c, the trend is reversed: the force strongly increases when the velocity decreases. Note also the appearance of very large force fluctuations at small velocities. This suggests that, in this very low velocity flow regime, the suspension deforms rather like a dry granular material.

3.3. Interpretation

The most significant parameters that control the transition reported above can be inferred by assuming a diphasic behaviour of the suspension. A model based on such an assumption has been successfully applied to describe the rheological behaviour of ceramic pastes [9,10]. In this approach, the suspension is assumed to deform with two flow fields v_p and v_f, associated, respectively, with the particles and the fluid. This vision is different from what is generally assumed for flow of suspensions, in which the particles are supposed to move affinely with the fluid. The diphasic approach is rather similar to the Brinkman model [11], but with a deformable porous media.

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Since the particles and the fluid do not necessarily have the same velocity, the rate of the viscous energy dissipation can be divided into two terms. The first term is related to the flow of the suspension as a whole (the particles have the same velocity as the fluid), and the second term is related to the damping force caused by the filtration of the fluid through the porous media made up by the particles. Hence, this rate of energy dissipation can be written as

$$W = \int_V \left(\sigma \dot{\gamma} + \frac{\eta_0}{k} (v_\mathrm{f} - v_\mathrm{s})^2 \right) \mathrm{d}V.$$

In the above expression, k is the permeability of the porous media formed by the particles and σ the shear stress experienced by the suspension (which is a function of its effective viscosity η_s). The integration is made over the volume of the sample.

The permeability scales with the particle size squared, and is a rather weak function of the concentration (cf. the Kozeny–Carman relationship [12]). This is in contrast to the effective viscosity (shear stress), which is a strong function of the concentration but independent of the particle size. As discussed above, the suspension presents a shear thinning flow behaviour; thus, the shear stress can be written as $\sigma = A\dot{\gamma}^m \approx A\,(U/h)^m$, where m is the shear thinning index, and A is the consistency index of the suspension.

The 'critical' velocity for which the filtration competes with the suspension flow can be estimated by assuming that this occurs when the two terms of the viscous dissipation are equal:

$$U_\mathrm{c} = \left(\frac{Ak}{\eta_0 h^{m+1}} \right)^{1/(1-m)}.$$

According to this expression, the non-Newtonian character of the suspension is required to obtain a filtration/suspension flow transition.

The permeability of the porous media made up by the particles scales with the particle diameter squared; we expect then that the appearance of the filtration would depend strongly on the particle size, in particular for highly shear thinning suspensions. For example, we found experimentally that increasing slightly (from 0.069 to 0.089 mm) the mean particle diameter, and keeping the other suspension parameters unchanged, the velocity for which the filtration starts up is multiplied by a factor of 2.

The influence of the particle volume fraction results in two opposing effects. The effective suspension viscosity is an increasing function of the concentration ($\eta_\mathrm{s} = \eta_0(1 - (\phi/\phi_\mathrm{max}))^{-2}$ for a Newtonian hard sphere suspension [13]). On the other hand, the permeability is a decreasing (although slightly) function of the concentration ($k \propto ((1 - \phi)^3/\phi^2)$, according to the Kozeny–Carman model [12]). The resulting effect of the concentration increase would be the increase in the squeeze velocity for which the filtration (phase separation) appears. This is confirmed experimentally by conducting squeeze experiments for different volume fractions. For instance, increasing the volume fraction from 53.6 to 56.6% results in an increase in the transition velocity by a factor of 5 (0.1 to 0.5 mm s^{-1}).

Due to the quite narrow interval of the suspension's parameters accessible in our experiments, it is not possible to go farther in the comparison between our qualitative model and the experiments. Moreover, the main objective of this model is only to bring to the fore the main parameters controlling the appearance of the phase separation. Numerical simulations similar to those of Racineux and Poitou [10] are needed to make quantitative predictions.

It is to be noted that the origin of this flow-induced concentration heterogeneity can not be attributed to the well-studied shear-induced particle migration in non-homogeneous shear flows [6,8]. Indeed, this phenomenon is diffusive and occurs on time scales (hours) [6] much larger than those involved in our
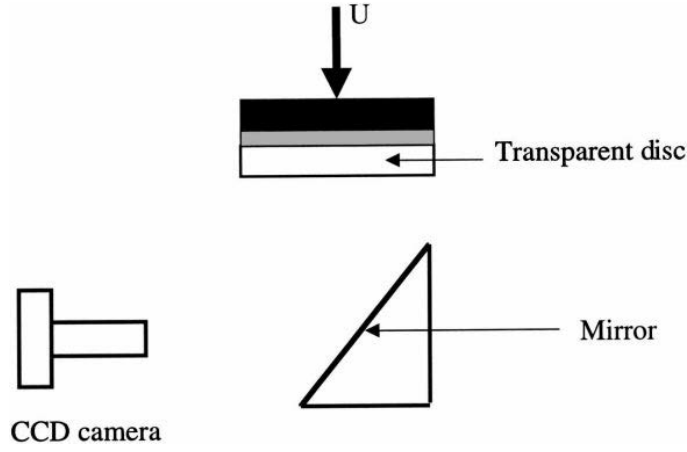
Fig. 4. Sketch of the set-up for the visualisation experiments.

experiments (seconds or few minutes). The length scale of the flow-induced heterogeneity observed in our experiment is the disc radius R, and the diffusion coefficient for shear-induced particle migrations can be estimated as $a^2\dot{\gamma}$ [8]. The characteristic time corresponding to this phenomenon can be written then as $R^2/a^2\dot{\gamma}$. This diffusive time is much larger than the squeezing time $(1/\dot{\gamma})$ which is actually required to induce the observed heterogeneity.

3.4. Visualisation experiments

Visualisation experiments are carried out in order to examine, at least qualitatively, the temporal evolution of the concentration during a squeeze experiment. The experimental set-up is depicted in Fig. 4. In these experiments, a colouring is added to the suspending fluid in order to allow observations. In Fig. 5a, we represent a sequence of two images corresponding to the beginning ($h=5$ mm) and the end
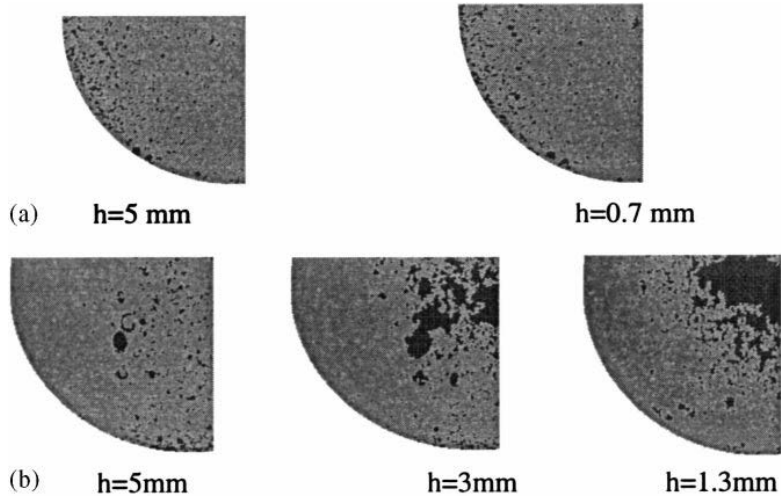

Fig. 5. (a) Visualisation of the initial and the final state of the suspension for a squeeze velocity ($U=3$ mm h^{-1}) for which phase separation is not expected according to the force behaviour. The concentration remains homogeneous during the experiment. (b) A sequence of images showing the increase in the particle concentration in the central part of the discs. The dark regions correspond to high particle concentration. In this experiment, the velocity is 0.02 mm s^{-1}, and the phase separation is expected to appear at $U=0.5$ mm s^{-1} according to the force measurements.

(h=0.7 mm) of the experiment, at a velocity (U=3 mm s^{-1}) for which phase separation is not expected. Qualitatively, it can be seen that the concentration remains homogeneous on sufficiently large scales.

In Fig. 5b, we represent an image sequence corresponding to a velocity (U=0.02 mm s^{-1}) for which phase separation is expected according to the force behaviour. Clearly, it can be seen in Fig. 5b that the concentration significantly increases in the central region of the discs, a result of a phase separation between the particles and the fluid.

4. Conclusion

The squeeze test is often used in practice to determine the rheological properties of highly viscous materials. Such a rheometer is quite simple in its design, but the interpretation of its measurements is not straightforward since the materials it is used for are in general complex fluids. In this work, the flow of concentrated model suspensions in a squeeze rheometer has been considered. Our purpose was to help in understanding the results reported in the literature regarding the rheological behaviour of ceramic industrial pastes [5]. Our squeeze force results were qualitatively similar to those obtained with industrial pastes. At relatively high squeeze speeds, the suspension presented a usual, although non-Newtonian, viscous flow behaviour. At low squeeze velocities, the suspension responded much like a sponge-form media. This behaviour has been attributed to a phase separation between the fluid and the particles due to the competition between the shear flow of the suspension as a whole and the solvent filtration through the particles. This is confirmed by visualisation experiments, which clearly made evidence of a concentration increase in the central part of the squeeze device at sufficiently low velocities. Our investigation was rather qualitative, and more quantitative studies in order to test models based on the above assumptions are clearly needed, including for instance the concentration measurements in refractive index-matched suspensions.

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