Deformation Behavior of Clay-Water Suspensions

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Deformation behavior of clay-water suspensions from rheological tests

Santagata, M.
School of Civil Engineering, Purdue University, W. Lafayette, IN

Dalmazzo, D., Santagata, E.
Dipartimento di Idraulica, Trasporti, ed Infrastrutture Civili, Politecnico di Torino, Italy

ABSTRACT: The paper investigates the small strain shear stiffness and the reduction of stiffness with shear strain of bentonite-water suspensions prepared at a range of water contents significantly greater than the liquid limit (1.7 - 4 LL). Testing is performed using an advanced rheometer through which the visco-elastic behavior as well as the flow properties can be evaluated. The emphasis in this paper is on the visco-elastic properties which are investigated performing oscillatory tests, to elucidate the effects of shear strain level (0.2%-100%) on the mechanical response of the suspensions.

The results indicate that at small strains the behavior of these high water content systems is predominantly elastic, with an approximately constant value of the storage (elastic) modulus (G′). This behavior extends to fairly large strains (>1%) beyond which there is significant degradation in G′, and the response of the material becomes increasingly viscous. Suspension concentration and degree of structuring resulting from sample stress history determine the magnitude of the initial elastic modulus. Concentration alone appears instead to control the critical shear strain in correspondence to which the behavior transitions from primarily elastic to primarily viscous. Overall, this transition is gradual, evidence of the thixotropy of the material.

1 INTRODUCTION

From a geotechnical perspective the *mechanical* properties of clay water suspensions are relevant in a variety of contexts: clay suspensions occur in nature, for example in mudflows; they form much of the sediment materials dredged from waterways and are encountered in tailing ponds; additionally, they are produced by man to be used as stabilizing fluids for excavations, drilled shafts and tunnels, as lubricants in drilling, microtunneling and pipe-jacking, and alone, or in combination with other materials, for soil improvement. Moreover, recent work on sand-bentonite mixtures (El Mohtar et al. 2008) suggests that the well documented increased cyclic resistance of sands with plastic fines is directly linked to the pore fluid formed: a concentrated clay suspension.

Significant research has been performed over the years on the sedimentation and consolidation properties of these materials, as well as on their flow and strength properties (e.g. Zreik et al. 1997). More recent work (Klein and Santamarina 2005) based on shear wave propagation measurements has provided insight also into the stiffness of these materials.

Great insight into the deformation behavior of these high water content systems can be gained through the use of advanced rheometers, such as the one employed in this study, designed for the characterization of complex fluids. These devices enable the study of these materials under well-established boundary conditions, and allow probing both the flow and the at rest properties.

This is the approach taken in this study, which focused on one particular clay: a Wyoming sodium bentonite which was used to prepare suspensions at water contents corresponding to 1.7 - 4 LL. Oscillatory tests were performed to investigate the visco-elastic properties of these suspensions at both small and large strains. These tests provide insight into fundamental aspects of behavior of clay-water suspensions, as the flow behavior is a very sensitive and direct criterion for particle interaction.

2 EXPERIMENTAL PROGRAM

2.1 *Materials and Equipment*

A commercially available bentonite was used for the tests presented in this paper. This material is a high grade sodium Wyoming bentonite, free of all

additives and chemical treatments, with 80% finer than 75 μm (#200 sieve), commercialized by Baroid Industrial Drilling Products. It is a traditional, general use bentonite used in a range of trenchless technology applications.

Concentrated suspensions were prepared from this bentonite using deionized deaired water, in a 500 ml stainless steel cup. Mixing was performed for 20 minutes using a Hamilton Beach model 936 mixer with mixing speed of 10,000 RPM (over the first few minutes mixing was halted 4-5 times to scrape the material from the sides of the cup and ensure the homogeneity of the samples). Data for three bentonite concentrations are presented in this paper: 4%, 6% and 9% by mass of the water. These concentrations correspond to water contents of 1111%, 1667% and 2500%, which fall in the 1.7 – 4 LL range (the liquid limit, LL, of the bentonite used in this study is 630%). Immediately after mixing the suspensions were stored in sealed plastic containers and tested after 24 hours to ensure full hydration of the bentonite.

All rheological tests conducted for this study were performed using the Physica MCR 301 rheometer (Fig. 1a), an air bearing, stress-controlled device manufactured by Anton Paar, which can also operate in strain-rate controlled mode through a feedback control loop. The Physica MCR 301 is equipped with a permanent magnet synchronous drive (minimum torque = 0.1 μNm, torque resolution <0.1 μNm), and an optical incremental encoder for measurement of the shear strain (resolution < 1 μrad). The rheometer can be used to perform a variety of oscillatory and monotonic, stress-controlled, and strain-rate controlled tests including amplitude sweeps, frequency sweeps, time sweeps, rate ramps, stress ramps and creep and recovery tests.

A 50 mm diameter, cone and plate geometry (Fig. 1b) was selected for all the tests presented here. The cone plate geometry was selected for this study as it ensures a constant shear rate across the specimen, an issue of particular significance when testing materials such as bentonite suspensions that are known to exhibit shear rate dependent behavior. The cone has a 1° inclination and a truncated tip. A gap (between truncated tip and plate) equal to 0.046 mm, corresponding to a sample volume of 0.57 ml, was used for all the tests.

At the time of testing, a sample of the suspension is obtained from the appropriate container using a small spatula and placed in the middle of the base plate. The cone – connected to the coupling in the measuring head – is then advanced at a controlled slow speed until it reaches a distance slightly greater than the gap height. A small spatula is used to "trim" the sample, and the final gap height is then reached.

The rheometer is equipped with a Peltier temperature control system that can control the temperature in the -40 to 200 °C range. The hood of the Peltier system, which is lowered over the sample after sample setup is completed, also houses a ring which functions as an evaporation blocker (Sato and Breedveld 2005). While temperature control was not a major concern in this study (all tests presented were conducted at 25°C, close to room temperature), minimizing evaporation was found to be critical. Tests conducted without the hood were found to generate unreliable results particularly if the test duration exceeded 15'-20' minutes.
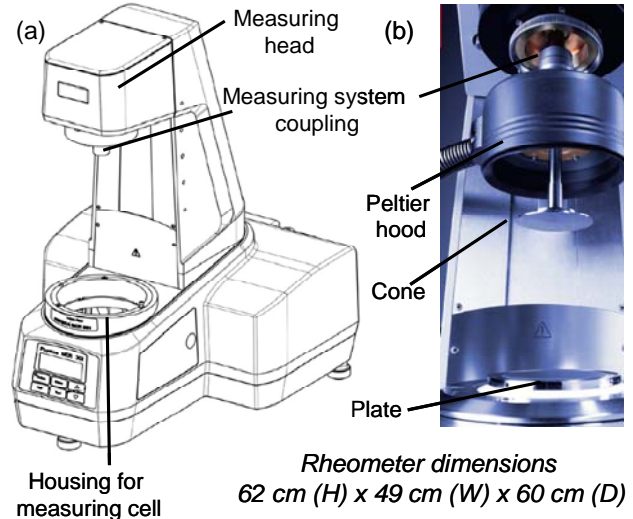
Figure 1: (a) Schematic of Physica MCR 301 rheometer, and (b) detail of measuring system and Peltier hood.

2.2 Testing Program

The primary focus of this study was the investigation of the deformation behavior of concentrated bentonite suspensions. For this purpose amplitude sweeps were performed (e.g. see Barnes et al. 1989). These are tests in which the material under investigation is subjected to an oscillatory shear strain in general following a sinusoidal time function:

$$\gamma = \gamma_0 \sin(\omega t) \qquad \text{eq.1}$$

with γ_0 the maximum amplitude and ω the angular frequency.

The rheometer then measures the resulting time dependent stress, which can be expressed as the sum of an elastic component in phase with the strain and a viscous component 90° out of phase with the strain (eq. 2):

$$\tau = \gamma_0 \left[G' \sin(\omega t) + G'' \cos(\omega t) \right] \qquad \text{eq.2}$$

where G' is the storage (elastic) modulus, and G'' is the loss (viscous) modulus. The ratio of the two moduli provides the phase angle δ ($\tan\delta = G''/G'$), i.e. the offset between the applied oscillatory strain and the measured shear stress.

In practice each of the tests conducted on the suspensions tested in this study involved several separate stages as summarized below:

- Stage 1 - Amplitude sweep
  (γ=0.2-100%, f=1 Hz)
- Stage 2 - Pre-shear
  (60" with $d\gamma/dt$=900 s^{-1})
- Stage 3 - Rest period with monitoring of G'
  (duration t= 3' or 10' [most tests])
- Stage 4 - Amplitude Sweep
  (γ=0.2-100%, f=1 Hz)

The *first amplitude sweep* involved the application of oscillatory cycles (f=1Hz) with amplitude γ increasing from 0.2% to 100% in 20 increments equally spaced on a logarithmic scale, allowing a minimum of 7 oscillations at each shear strain. Note that the response measured during this first amplitude sweep cannot be considered representative of the intact behavior of the suspension at 1 day, as the sample setup operations produce significant disturbance and actually cause the material to flow (when the suspension is squeezed between the plate and the cone). The degree of destructuring associated with these operations is unknown.

During the *pre-shear stage* a high (900 s^{-1}) shear rate was applied for a period of 60". As a result of this stage the suspensions are destructured in a manner that is considered consistent and reproducible for all samples, eliminating the effects of differences in the setup operations.

In the *third stage* the sample was allowed to rest for a controlled period of time – equal to 10' for the majority of the tests presented in this study (in a limited number of tests the rest period lasted 3'). The goal of stages 2 and 3 was to control and render uniform the history of all the samples, to facilitate comparisons between the results of the subsequent stage on different samples. Similar pre-test procedures have been employed by other researchers investigating the rheology of clay suspensions.

During the rest period the small strain behavior of the suspension was "monitored" applying oscillatory cycles at a constant frequency (f=1 Hz), with amplitude sufficiently small that the material remains in the linear visco-elastic regime. In the tests presented in this paper a constant maximum amplitude, γ_0=0.2%, was employed for all samples. Measuring the variation of the small strain values of G' and G'' over the rest period provides the means to monitor the structuring processes that occur over time in a clay.

Finally, the *last amplitude sweep (stage 4)* was performed following the same procedures outlined for stage 1. With the pre-test (pre-shear and rest stages) standardized, the results of these tests serve to evaluate the effects of the testing variables under consideration on the deformation behavior of the bentonite-water suspensions examined.

Note that in a limited number of tests, following the second amplitude sweep, stages 2-4 were repeated.

3 RESULTS

## 3.1 *Elastic nature of behavior of clay-water suspensions*

Figure 2 shows plots of the storage modulus (G') the loss modulus (G'') and the phase angle (δ) measured as a function of shear strain in the final amplitude sweep performed on two samples of the most concentrated (9% bentonite by mass of water) of the suspensions examined in this study.

The following observations can be made based on the plots shown in the figure:

- the data from the two tests conducted on independent samples are very consistent (with less than 5% difference in the small strain value of G');
- at small strains the storage modulus (G') greatly exceeds the loss modulus (G''); this is evidence of an essentially elastic behavior;
- the elastic nature of the behavior of the suspension at small strain is reflected by the very small values of the phase angle (<2° for both samples);
- the storage (elastic) modulus remains essentially constant up to shear strains exceeding 1%; the arrow on the picture indicates a threshold linear shear strain (γ_{lin}) of approximately 2% chosen in correspondence to $G'/G'max = 0.98$ (as done by other authors, e.g. Santagata et al. 2007). This magnitude of the linear threshold exceeds the values reported for even very plastic soils by over an order of magnitude (e.g. Vucetic and Dobry 1991, Lo Presti 1989);
- as the shear strain increases beyond γ_{lin}, significant non-linearity in the behavior is observed;
- as G' decreases with increasing shear strain, G'' increases, and the behavior of the material becomes increasingly viscous; this is reflected in the increase of the phase angle;
- in correspondence to a critical shear strain (γ_{crit}), identified in the figure by the dashed line, the

cross-over between storage and loss modulus is observed, i.e. the behavior becomes predominantly viscous; for both tests shown in Figure 2 $\gamma_{crit}\sim 14\%$;

- at high shear strains the value of the phase angle is close to 90°, i.e. the material essentially behaves as a liquid.

(a)

(b)
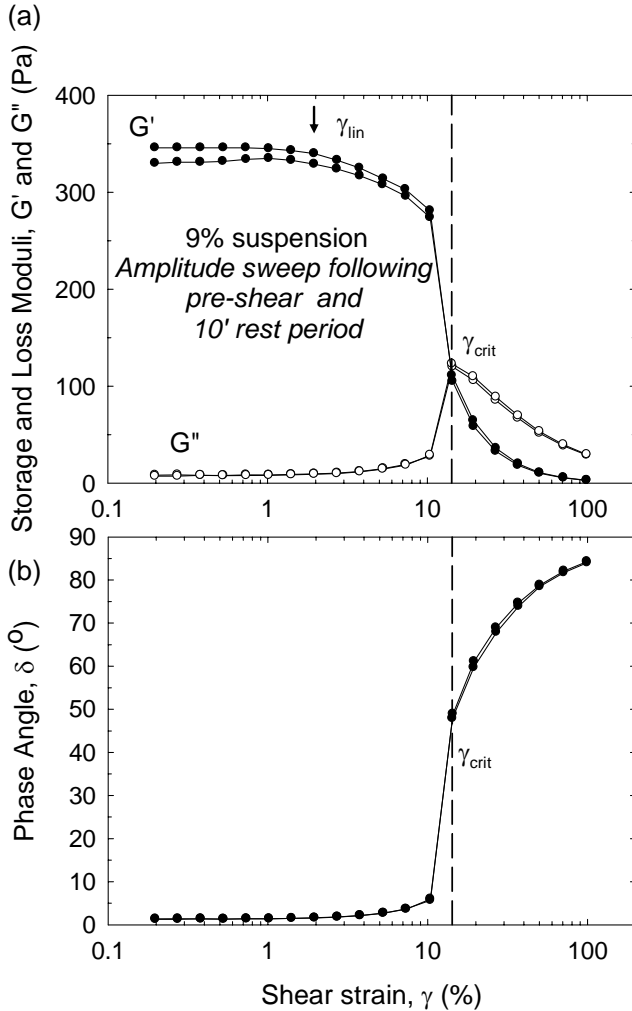
Figure 2: (a) G′, G″ and (b) δ as a function of shear strain from amplitude sweeps conducted on 9% suspension following pre-shear and 10' rest period.

Note also the value of the small strain elastic modulus (G′) – approximately 350 Pa. While the effective stress state of the sample is not known, this modulus is a few orders of magnitude smaller than stiffness values reported even for very soft soils.

The behavioral features outlined above are typical of materials termed by some gels (e.g. Abend and Lagaly 2000), and by others "soft jammed systems" (Coussot 2005), in which each of the "elements" (the colloidal size bentonite particles in this case) interacts with a number of the other elements that surround it. Coussot (2005) suggests that collectively the forces that result from these interactions determine a "potential well"

corresponding to the equilibrium condition (Fig. 3). When the system is perturbed two things can happen: if the perturbation is small the element cannot overcome the potential barrier (ΔE) and returns to the bottom of the well (solid like behavior); if the element is able to jump out of its well, we have the transition from solid to liquid behavior. This transition is associated with a critical deformation (γ_{crit}) (Fig. 2), which Coussot (2005) suggests is "related mainly to the average distribution of particles in space."
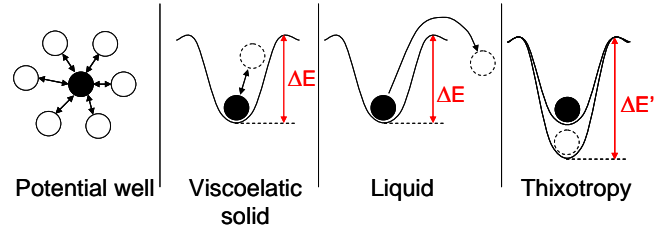
Figure 3: Potential well and relationship to macroscopic rheological characteristics (adapted from Coussot 2005).
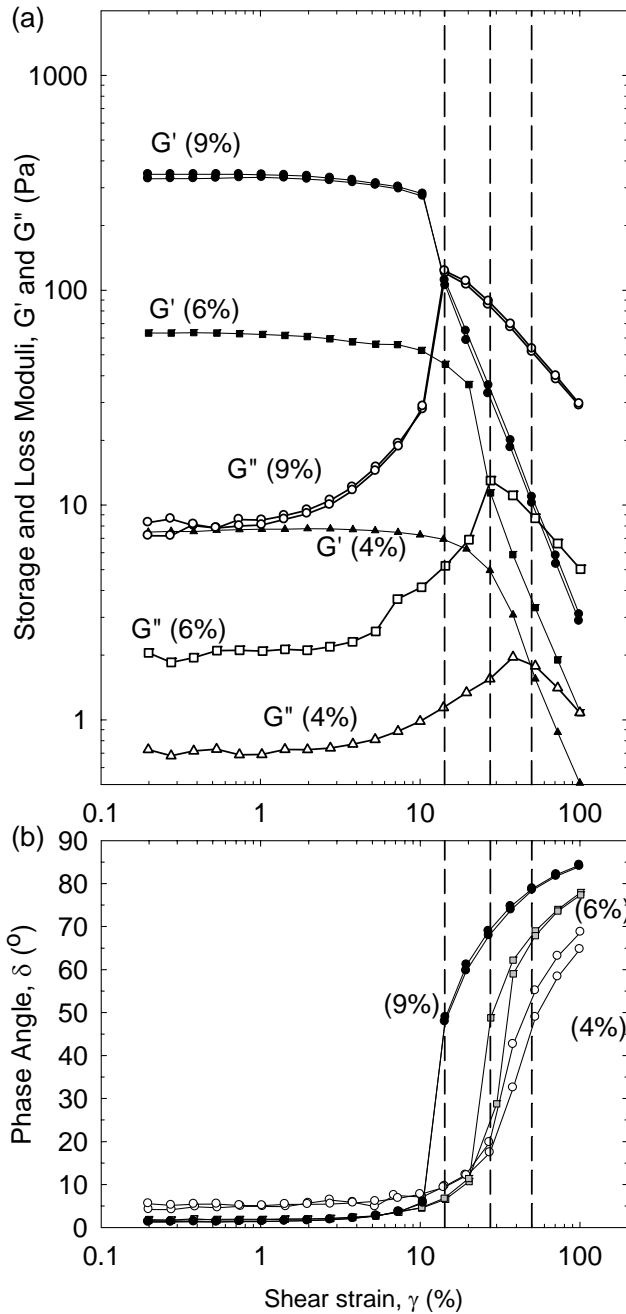
As shown in Figure 2b, the solid-liquid transition is not instantaneous. The gradual nature of this transition is evidence of the thixotropic nature of the suspension, as a result of which the "depth" of potential well increases with time (Fig. 3).

3.2 *Effect of bentonite concentration*

Figure 4 combines the results shown in Figure 2 with similar data obtained following the same pre-shear and 10' rest period for suspensions prepared at lower bentonite concentrations: 4% and 6%. Due to the large range over which the stiffness values fall, G′ and G″ are plotted using a logarithmic scale. Overall, the observations made above for the 9% suspensions, apply to the two lower concentration suspensions, i.e. the suspensions exhibit essentially elastic behavior at small strain, and gradually transition to fluid-like behavior with increasing strain. Over the range of concentrations examined, the small strain values of both moduli (G′ and G″) vary by over an order of magnitude.
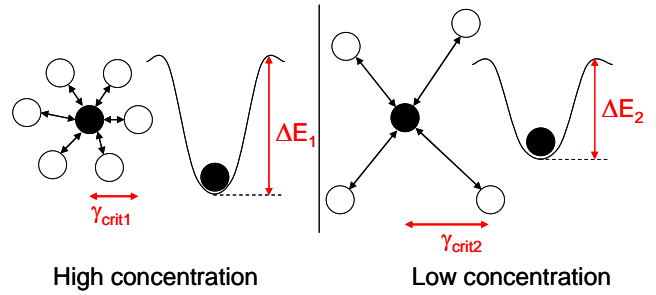
The curves presented in Figure 4 also indicate a slight increase in the linear threshold (γ_{lin}) with a reduction in bentonite concentration.

Figure 4 also shows that with decreasing bentonite content the critical shear strain (γ_{crit}) (identified in the figure by the dashed lines) increases from 14%, to 28% and 50% for the 6% and 4% bentonite suspension, respectively. This indicates that with decreasing bentonite concentration the shear strain required for the material to transition from solid to liquid increases. These results can be interpreted within the behavioral framework proposed by Coussot (2005) as shown in Figure 5.

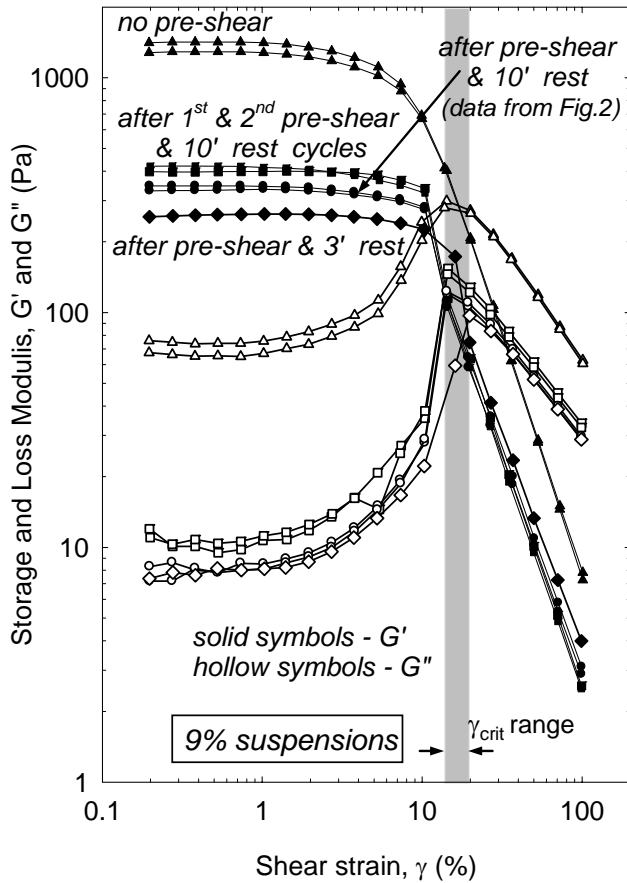**Figure 5:** Potential well for high and low concentration suspensions.

The above also explains the phase angle data shown in Figure 4(b). At very small strains the greater the concentration, the deeper the potential well, the more elastic the nature of the behavior. This is reflected in the slight increase in the initial value of δ with decreasing concentration (for the 4% suspensions δ~4-6° compared to 1-2° for the other two suspensions). At the maximum shear strain (100%) reached in the test shown in Figure 4, this trend is reversed with δ increasing with concentration. This is due to the differences in the values of γ_{crit} discussed above. For the 9% suspension at this shear strain γ_{crit} has been greatly exceeded; hence the behavior is almost completely fluid. For the lower concentration suspension, due to the greater value of γ_{crit} the behavior is still visco-elastic. Higher shear strains would have been required to reach a phase angle close to 90°.

3.3 Degree of structuring and uniqueness of γ_{crit}

Figure 6 reports curves of G′ and G″ versus shear strain obtained from amplitude sweeps conducted following different pre-test procedures, all on 9% suspensions. Specifically, the figure includes: the data already reported in Figure 2 for two sweeps performed after the pre-shear and the 10' monitoring stage; the results from a test conducted with a reduced rest period (3' instead of 10'); two curves from the amplitude sweeps conducted immediately after sample setup, i.e. with no pre-shear; and, finally, data from two sweeps conducted on the same sample: one following the usual pre-shear and 10' rest period, the other following a second pre-shear and 10' rest period stage.

The figure shows a six fold increase in the G′max values measured (~250-1400 Pa) which, given that all suspensions have equal concentration, reflects solely the varying degree of structuring reached as a result of different pre-shear stress histories. The initial values of G″ show a similar range of variation, and for all samples the initial phase angle falls in a very tight range (1.5°-3°).

Figure 4: (a) G′, G″ and (b) δ as a function of shear strain from amplitude sweeps conducted on 4%, 6% and 9% suspensions following pre-shear and 10' rest period.

The figure suggests that at higher concentrations, when the particles are very close to each other a small relative displacement (related to γ_{crit1}) is sufficient to enable the transition to a new arrangement of particles. When the particles are further from each other (lower concentration), this displacement is larger ($\gamma_{crit2} > \gamma_{crit1}$). Note that the shear stress required to produce γ_{crit} would be greater in the more concentrated suspensions, for which, due to the larger number of interactions between the particles, a deeper potential well is produced (Fig. 5).

Figure 6: G′ and G″ as a function of shear strain from amplitude sweeps on 9% bentonite suspensions with different stress histories.

Figure 6 also identifies the range of variation (gray band) of the critical shear strain (γ_{crit}). It is observed that, despite the different stress histories of the samples, γ_{crit} varies within a fairly narrow range (14%-20%) (compare increase in γ_{crit} due to reduction in concentration to 6% which translated in similar variation in G′max). This is consistent with the assumption (Coussot 2005) that the critical shear strain is related primarily to the average distribution of particles in space, which is controlled by the sample concentration but not by the state of the structure. The latter will, of course, affect the stiffness of the suspension and the depth of the potential well, and, hence, determine the shear stress required to produce γ_{crit}. Note also that the limited data around γ_{crit} (due to the logarithmic increase of the shear strain amplitude in the sweep) probably contributes to increase the range of variation of this parameter.

3.4 *Restructuring processes and thixotropy*

As discussed in section 2.2, measurements of the storage and loss modulus were conducted over the rest period following the pre-shear stage to monitor the processes of restructuring of the material.

The solid symbol curve in Figure 7 shows the increase in G′ measured during the rest period in one particular test (similar results were obtained from all rest stages). It is observed that G′ increases rapidly over the 10' rest period (by a factor of 2.5-2.8 for the six independent tests performed on 9% bentonite suspensions), with the rate of increase decreasing with time. Note that no significant change in the loss modulus, G″, is observed over the 10' rest stage. This translates into a continuous reduction in the phase angle with time.
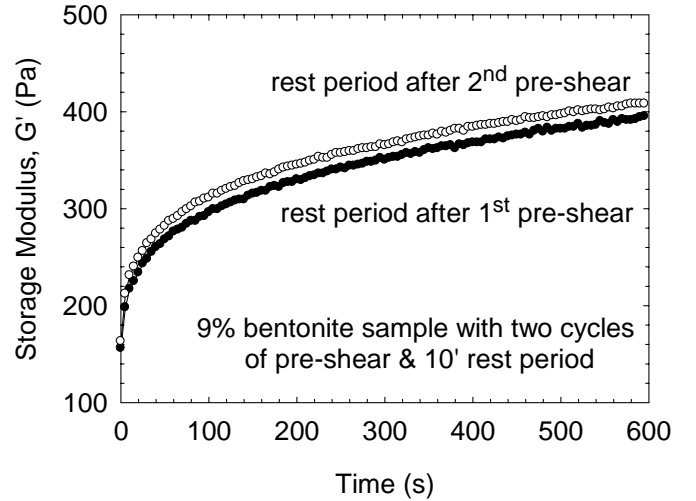
Figure 7: Increase in G′ during rest period following pre-shear stage

Similar results are derived from all other 10' monitoring stages, although the increase in G′ over this period increases significantly with decreasing bentonite concentration.

In the test shown in Figure 7, a second pre-shear and rest period with monitoring were performed following the first rest period and the subsequent amplitude sweep. The hollow symbol curve in Figure 7 shows the data for the second monitoring stage. The figure shows that the initial G′ values for the two stages are very consistent, and that overall the curves remain close through the monitoring stage (see also results for subsequent amplitude sweeps shown in Figure 6). This indicates that approximately the same level of destructuring is reached following the two pre-shear stages, and demonstrates the thixotropic nature of the material.

The results for this test presented in Figures 6 and 7 also suggest that the effects of sample evaporation on the amplitude sweeps presented in this paper are negligible.

4 CONCLUSIONS

This paper has addressed the deformation behavior of bentonite suspensions prepared at

concentrations (4%, 6% and 9% by mass of the water) corresponding to water contents greatly exceeding the liquid limit (1.7 - 4 LL).

The study relied on oscillatory tests performed using a Physica MCR-301 rheometer to describe the dependence of the following visco-elastic properties on shear strain (0.2%-100%): storage modulus G', loss modulus G'', and phase angle δ.

All suspensions exhibit essentially elastic behavior at shear strains up to 1-2%, as reflected by values of δ of below 5°. The value of the elastic (storage) modulus in this region increases with concentration; at a given concentration G' is controlled by the degree of structuring which is determined by the stress history of the sample. For all the tests performed G' falls in the 8–1500 Pa range, well below values reported in the literature for soft soils and sediments.

With increasing shear strains there is significant degradation in G', as G'' increases. The transition from solid-like to liquid-like behavior is gradual, indicating the thixotropic nature of the material. A critical strain (γ_{crit}) identifies the cross-over between G'' and G'. γ_{crit} appears to be controlled by the average distribution of particles in space, which is controlled by the sample concentration. Hence, it increases with decreasing bentonite concentration; at a given bentonite concentration it is observed instead to be independent of the sample stress history and the degree of structuring.

The study also provides insight into the rate of structure (and stiffness) buildup in these high water content systems, which is found to increase with decreasing bentonite concentration.

The rheological data presented in the paper contribute to the understanding of the fabric/microstructure of the clay-water systems investigated. The behavior of all suspensions examined in this study is found to be consistent with the formation of a gel, a "single particle association that fills an entire space" (van Olphen 1977).

The experimental techniques and procedures utilized in this study can be extended to the investigation of bentonite (and other clay mineral) water systems under a more broad range of testing conditions (e.g. modifying the ionic strength and pH of the pore fluid or the testing temperature) to develop "fabric maps" (e.g. Palomino and Santamarina 2005) that describe in a systematic manner the modes of particle interaction that occur as a function of the properties of the pore fluid and the environment. Such work is significant, for example, to the understanding of sediment formation, and the stability and performance of drilling and tunneling fluids in different environments.

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