Rheological Properties of a Natural Estuarine Mud

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Received: 6.2.2003, Final version: 8.4.2003

Abstract:
In this paper, the linear and non-linear rheological properties of estuarine cohesive sediments were investigated. The density of the sediments has been determined by pycnometry. Creep and oscillatory shear measurements have been performed in order to determine i) the transitions in mechanical response to creep and oscillatory shear and ii) the material properties of these natural fluids as a function of their density. For all samples tested, four different rheological transitions have been determined and all material properties have been shown to be satisfactorily fitted by exponential functions of the density.

Zusammenfassung:
In diesem Artikel werden die linearen und nicht-linearen rheologischen Eigenschaften kohaesiver Sedimente, die aus Flussmuendungen stammen, untersucht. Die Dichte der Sedimente wurde mit Hilfe der Pycnometrie bestimmt. Kriech- und oszillatorische Schermessungen wurden durchgeführt, um i) den Übergang in der mechanischen Antwortfunktion zum Kriechverhalten und zum oszillatorischen Scherverhalten und ii) die Materialeigenschaften dieser natürlich vorkommenden Fluide als Funktion ihrer Dichte zu bestimmen. Für alle untersuchten Proben wurden vier verschiedene rheologische Phasenuebergaenge bestimmt und alle Materialeigenschaften konnten befriedigend mit einer exponentiellen Dichteabhaengigkeit angepasst werden.

Résumé:
Dans cet article, les propriétés rhéologiques linéaires et non-linéaires de sédiments cohésifs estuariens ont été étudiées. La densité des sédiments a été déterminée par pycnométrie. Des mesures en fluage et en cisaillement oscillatoire ont été menées afin de déterminer i) les transitions dans la réponse mécanique en fluage et cisaillement oscillatoire et ii) les propriétés matérielles de ces fluides naturels en fonction de leur densité. Pour tous les échantillons testés, quatre transitions rhéologiques différentes ont été déterminées et on a montré que les grandeurs matérielles étaient décrites, de manière satisfaisante, par des fonctions exponentielles de la densité.

Key Words: cohesive sediments, mud rheology, viscoelasticity, non-Newtonian behavior

1 INTRODUCTION

Cohesive sediments are commonly found in estuaries. They are natural suspensions, mainly composed of clay mineral particles (plus sand and organic matter), with variable physical and mechanical properties depending on numerous parameters : density, mineral and organic content, pH and ionic strength of the medium, temperature [1]. The study of the transport of these natural complex fluids, when subjected to oscillatory shear by water waves, or simple shear by current, is of great interest from both an economical and an ecological point of view. Indeed their accumulation in estuaries may not only hinder navigation but also induce environmental pollution by the presence of contaminants held in sediments. Thus, for coastal management, it is of great importance to know whether planned projects will diminish or increase siltation. In order to make such predictions, hydro-sedimentary models are being used as current tools; IFREMER, in France, has been working on such models for many years. Still getting good predictions from such modeling needs both relevant constitutive laws, and accurate quantitative description of the rheological parameters of these equations, as a function of physical or physico-chemical parameters (e.g. density).

Hydrodynamic, mainly oscillatory, forcing of cohesive sediments was studied in numerous modeling research works, but with sediment rheological properties assumed rather than measured. In these works, the behavior of the sedi-

DOI: 10.1515/arh-2003-0010

ments was assumed to be either purely viscous [2, 3], or purely elastic [4], or viscoelastic [5, 6], or viscoplastic [7], even including thixotopic behaviour [8, 9]. The validity of these theoretical models depends much on the validity of the constitutive laws they assume. Still the experimental rheological characterization of cohesive sediments samples directly taken off from natural sites (e.g. estuarine mudflats) has not yet received much attention in the literature [1, 10 - 12].

On the other hand "model" cohesive sediments, such as concentrated clay water suspensions or aqueous clays/grains mixtures, have been rheologically investigated in many published works, as reviewed in the monograph by Coussot [13]. First it has to be stressed that the rheological investigation of such highly concentrated suspensions is usually a difficult task because of the many experimental problems encountered (edge effects, wall slip, cracks, fracture, ...) [13]. The rheological studies on "model" cohesive sediments mainly concern their linear and non-linear behaviors in steady shear flows; generally speaking, they have shown that these complex fluids usually exhibit two major rheological properties: a yield stress and a shear-thinning behavior at stresses above the yield stress [14]. Thus the flow curve of these "model" cohesive sediments has been shown to be adequately fitted by the Hershel-Bulkley phenomenological model [15]:

$$\tau = \tau_y + K\dot{\gamma}^n$$

where τ_X denotes the yield stress, $\dot{\gamma}$ the shear rate, and K, n are positive parameters.

Linear and non-linear viscoelastic properties of "model" cohesive sediments have been much less studied : few oscillatory shear measurements have been carried out in order to investigate the viscoelastic response of these materials below the yield stress [16, 17]. Moreover, "model" cohesive sediments have been shown to exhibit time-dependent viscous behavior, that is thixotropy [18, 19]. It is worth noticing that a great majority of the rheological studies of "model" muds focus on the solid volume fraction dependence of the material properties, mainly the yield stress and storage modulus, of these systems [13].

At last we would like to stress that the rheology of such complex systems as natural muds still lacks well-defined unifying physical concepts, like those proposed in the rheology of polymeric systems for instance, though quite recent conceptual approaches are directed to this end [20, 21]. In the present work we report on the linear and non-linear rheological properties of natural estuarine cohesive sediments, submitted to creep and oscillatory shear tests, as a function of their density.

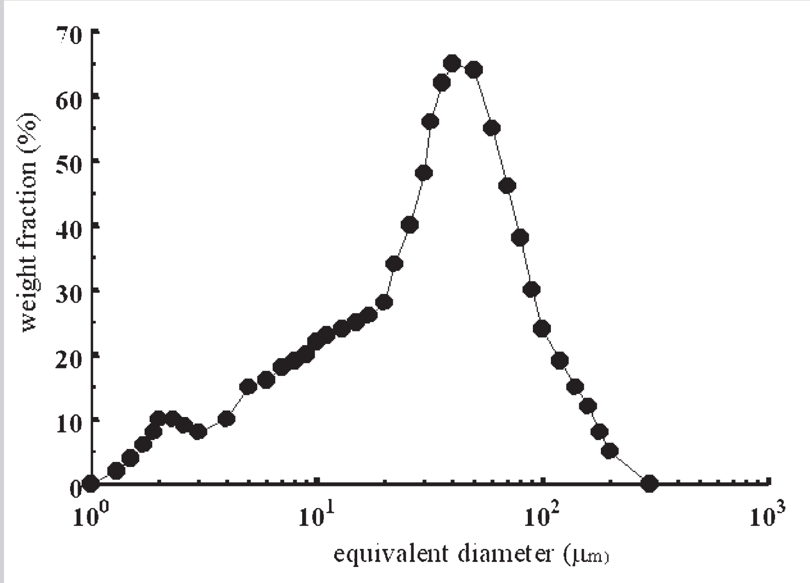


Figure 1: Granulometric profile of the mud: the weight fraction of particles as a function of their equivalent diameter.

2 EXPERIMENT

2.1 MATERIALS

The samples of natural muds studied in this work have been taken off from mudflats of the estuary of La Penzé, a river located in Brittany, North West of the city of Morlaix (Finistère, France). The mud is saturated with sea water from Penzé estuary and kept refrigerated, at a temperature between 2°C and 4°C. It is kept in a Plexiglas column, ready-cut every 2 cm, for four weeks before being tested, so that complete natural consolidation of the mud can be achieved and samples of different densities can be obtained. The particle size distribution has been determined by laser granulometry; the results are plotted in Fig. 1, showing a median particle diameter of about 35μm. Besides, the mud has been shown to contain a very small amount of organic matter: the total organic carbon of the samples has been shown to be less than 1.5 %.

2.2 EXPERIMENTAL METHODS

2.2.1 DENSITY MEASUREMENTS

The density of the samples has been measured using two techniques:

Pycnometry
The pycnometer is a small 50 ml flask with a capillary tube at the top. Knowing the mass of the pyc-

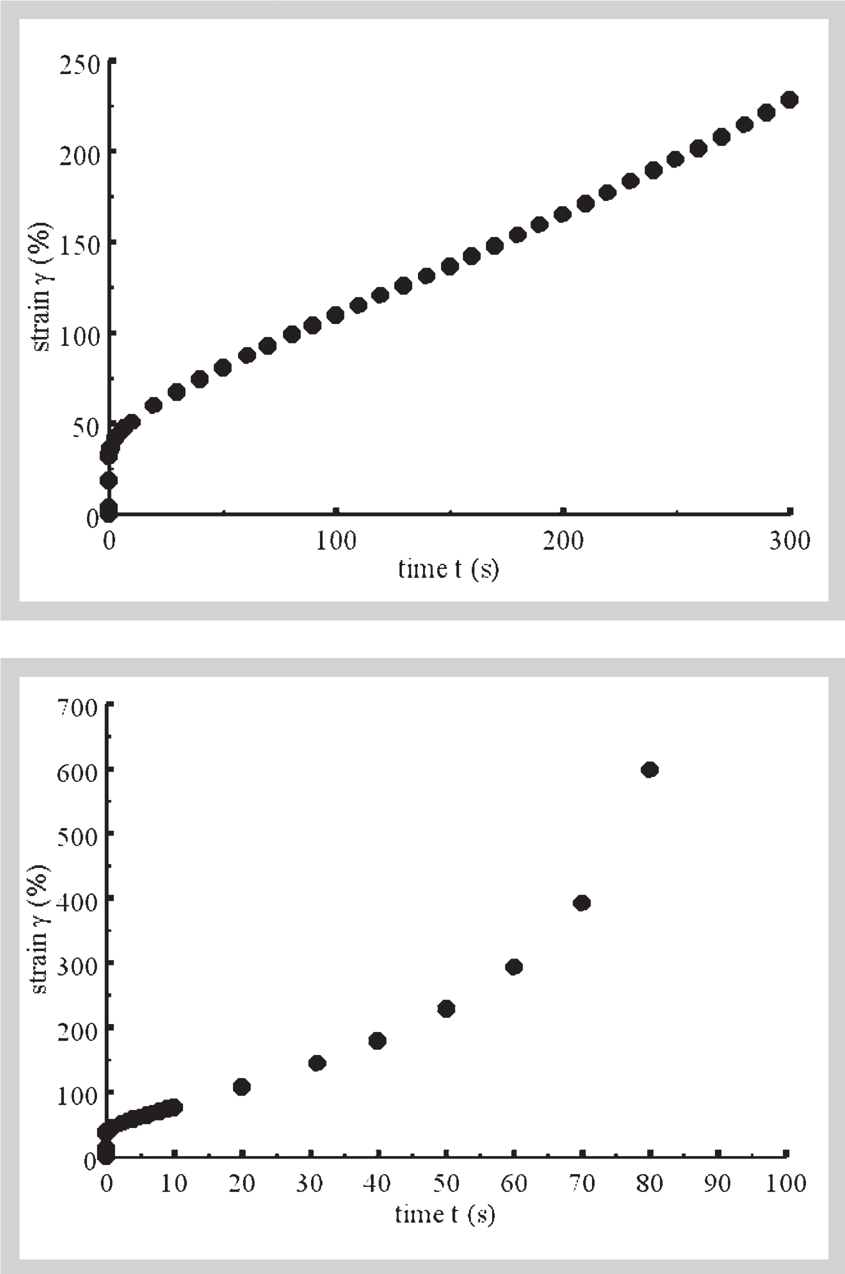


Figure 2: The strain response of a 1380 kg/m³ mud sample to a creep test: a (above): below the yield stress, b (below): above the yield stress.

nometer, M_1, the mass of the pycnometer completely filled with distilled water, M_2, the mass of the pycnometer containing the sample, M_3, and the mass of the pycnometer containing the sample and completely filled with distilled water, M_4, one can determine the mass of the sample:

$$M = M_3 - M_1 \tag{1}$$

and the volume of the sediment, equal to the volume of the water displaced:

$$V = \frac{(M_2 - M_1) - (M_4 - M_3)}{\rho_{H_2O}} \tag{2}$$

ρ_{H2O} denoting the density of distilled water. The density of the mud sample, ρ_{mud}, is then inferred:

$$\rho_{mud} = \frac{M}{V} \tag{3}$$

Using this technique, the absolute error in the determination of ρ_{mud} was estimated to lie within 10 kg/m³.

Drying technique

This technique is based on the determination of the solid volume fraction, Φ, by weighing a given amount of the mud sample before (mass m_{mud}) and after (mass m_{solid}) drying at 105°C until complete evaporation. Knowing the density of the solid content ρ_{solid} and the density of the sea water $\rho_{sea,}$ the volume fraction is determined by:

$$\Phi = \frac{\dfrac{m_{solid}}{\rho_{solid}}}{\dfrac{m_{solid}}{\rho_{solid}} + \dfrac{m_{mud} - m_{solid}}{\rho_{sea}}} \tag{4}$$

The density of the mud sample, ρ_{mud}, is then inferred:

$$\rho_{mud} = \Phi\rho_{solid} + (1 - \Phi)\rho_{sea} \tag{5}$$

Actually, in this work, pycnometry was used to determine the density, ρ_{mud}, of the mud samples, and the drying technique was used to determine the actual density ρ_{solid}, of the solid phase of the samples.

2.2.2 RHEOMETRY

Two types of rheometrical tests have been performed: creep tests, corresponding roughly to natural hydrodynamic forcing by currents, and oscillatory shear tests, corresponding roughly to natural hydrodynamic forcing by water waves. Rheological measurements were all performed at 10 ± 0.1°C (average temperature of the mud samples in their natural environment), using mainly a Carri-Med CSL50 controlled stress rheometer, equipped with a parallel-plate geometry, characterized by a plate diameter of 40 mm and a gap of 1.5 mm, which is much larger than the average size of the grains. The two surfaces of the parallel-plate geometry were roughened with waterproof abrasive paper of equivalent roughness of 80 μm, in order to avoid wall slippage. To prevent mud samples, located in the geometry gap, from drying, sea water was regularly put on the air/sample interface.

3 RESULTS

3.1 DENSITY

The density of the mud samples, ρ_{mud}, tested in this work has been shown to lie between 1380 and 1630 kg/m^3, from the top to the bottom of the column, due to natural consolidation.

The use of the drying method described above led to the determination of the average density of the solid phase of the samples, ρ_{solid}. ρ_{solid} was shown to be about 2200 kg/m^3, that is lower than the approximate value, 2700 kg/m^3, of the average density of clays [13]. The lower value of the density of the solid phase tends to show the presence of other minerals than clays, but this statement cannot be confirmed in the absence of analysis of the mineral content of the mud tested.

3.2 CREEP BEHAVIOR

Fig. 2 shows the creep curve of a mud sample with density of 1380 kg/m^3. The results plotted in Fig. 2(a) are quite representative of those obtained for any samples tested below a certain critical shear stress τ_y. In this stress range, the strain response to a constant shear stress exhibits two regions: a viscoelastic region at short times, characterized by a retardation time, and a viscous region on any longer time scales, characterized by a constant shear rate, determined by the slope of the linear strain response.

At shear stresses above τ_y, the strain response is illustrated in Fig. 2(b). At short times, the response is the same as that shown in Fig. 2(a), but at a longer time, the curve presents an inflexion point marking the transition from a low strain rate response to (very) high strain rate levels. For any creep tests performed in this work, sufficient time was left for the final shear rate to attain a steady state value, but no systematic investigation of time-dependent response over very long times was ever performed.

The corresponding flow curve is plotted in Fig. 3. The representation in a shear stress vs. shear rate plot, in Fig. 3(a), clearly shows that the critical shear stress τ_y is an apparent yield stress [22]. The same results, plotted in a viscosity versus shear rate plot, in Fig. 3(b), show that the behavior of the mud sample is Newtonian at very low shear rates, with very high zero-shear viscosity levels η_O, and that, above a critical shear rate $\dot{\gamma}_c$ corresponding to the yield stress τ_y, the mud "liquefies" very rapidly. Moreover, $\dot{\gamma}_c$ was shown to have no significant dependence on the density of the mud: A value of $\dot{\gamma}_c \sim$ a few 10^{-4} s^{-1} was found for all samples tested.

Fig. 4 shows the Newtonian viscosity η_O as a function of the density, for all samples tested. In spite of a fairly great dispersion of the experimental data, this plot can be adequately fitted by an exponential function:

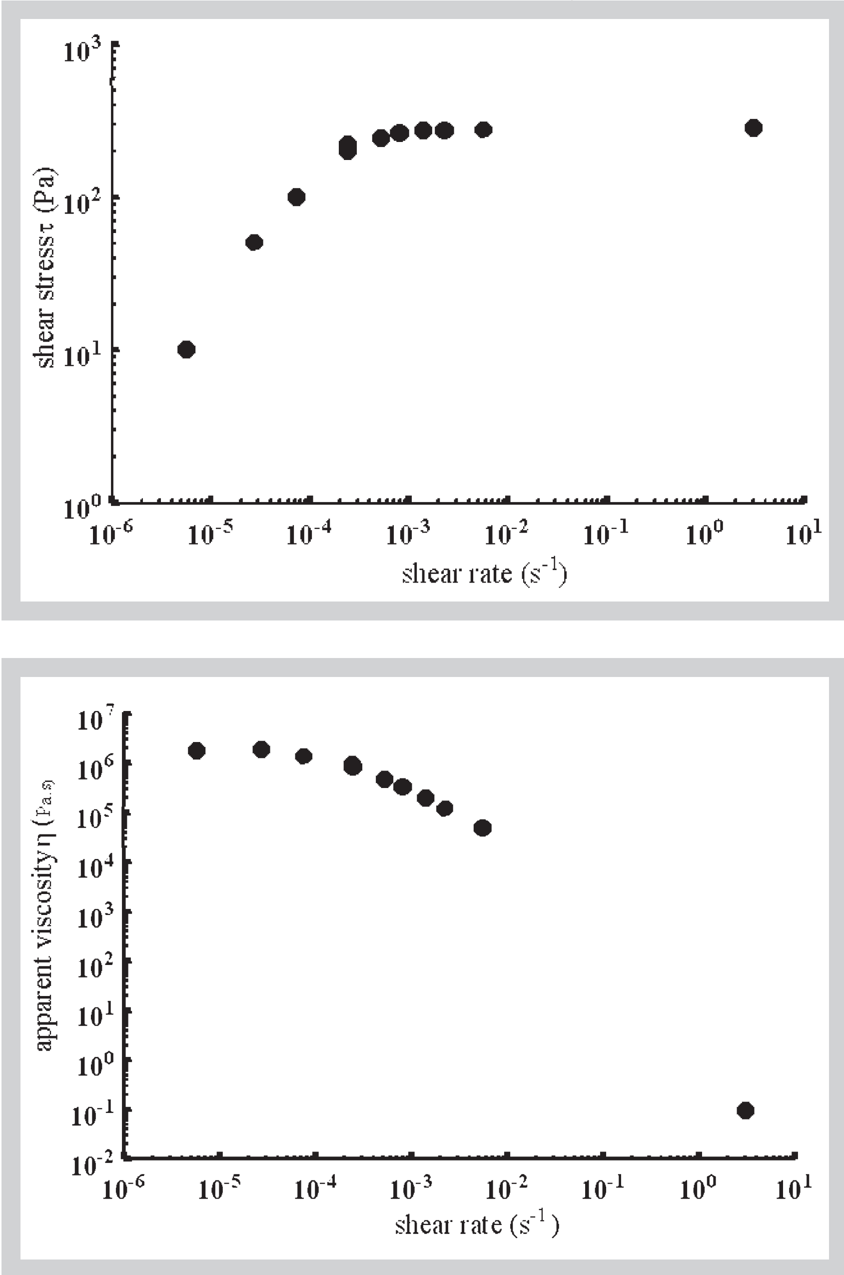
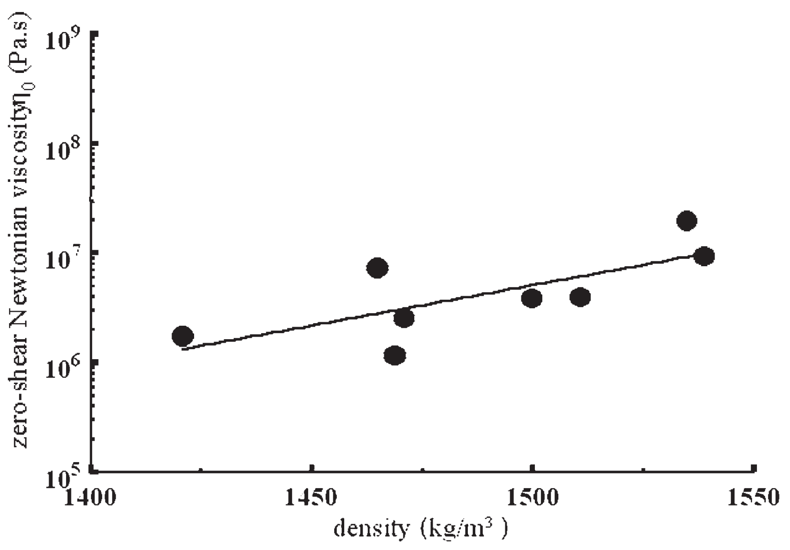


Figure 3: The flow curve of a 1380 kg/m^3 mud sample, as obtained from creep tests: a (above): shear stress as a function of shear rate, b (below): zero-shear Newtonian viscosity as a function of shear rate.



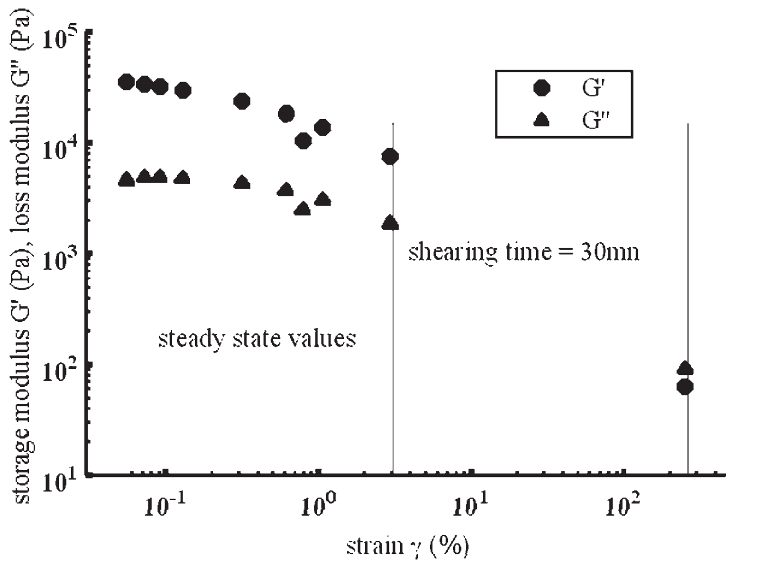


Figure 4 (above): The zero-shear Newtonian viscosity as a function of the density of the mud.

Figure 5 (below): The storage and loss moduli as a function of the strain amplitude at a frequency of 1 Hz (1490 kg/m³ mud sample).

$$\eta_o \cong 8 \cdot 10^{-7} e^{0.02\rho_{mud}} \quad (6)$$

where ρ_{mud} is expressed in kg/m³ and η_o in Pas. From Eq. 6, the dependence of the yield stress τ_y can be inferred:

$$\tau_y \cong \eta_o \dot{\gamma}_c \cong 3 \cdot 10^{-10} e^{0.02\rho_{mud}} \quad (7)$$

where ρ_{mud} is expressed in kg/m³ and τ_y in Pa. The best fit was obtained for $\dot{\gamma} = 4 * 10^{-4}$ s^{-1}. η_o and τ_y can also be expressed in terms of the solid volume fraction, Φ, of the mud using Eq. 5:

$$\eta_o \cong 390 e^{24\phi} \quad (8)$$

$$\tau_y \cong \eta_o \dot{\gamma}_c \cong 0.16 e^{24\phi} \quad (9)$$

3.3 OSCILLATORY SHEAR BEHAVIOR

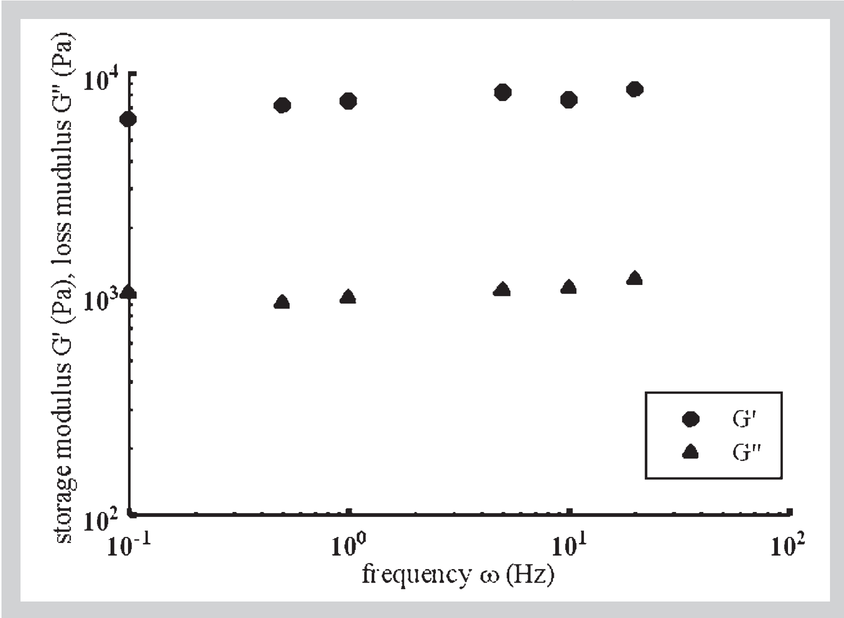
Fig. 5 shows the viscoelastic moduli *G'* and *G''* as a function of the strain amplitude, at a frequency of 1 Hz, of a mud sample with density of 1490 kg/m³. These results are quite representative of those obtained for any samples tested. The viscoelastic response to oscillatory shear exhibits three main regions:

- at small strains, that is for strain amplitudes less than a few hundredths %, the response is linear. The extent of this linear viscoelastic regime, expressed in terms of strain amplitude, does not depend significantly on the density of the sample. Moreover the frequency dependence of the linear viscoelastic moduli, at least in the range of practical interest (frequency range of the wave forcing), is very weak, as illustrated in Fig. 6.

- at moderate strains, that is for strain amplitudes between a few hundredths and a few %, the behavior is slightly non-linear but does not exhibit any time-dependent response, so that steady state *G'* and *G''* can be measured.

- at higher strains, the response exhibits thixotropic features, characterized by pronounced time-dependent behavior, as displayed by the continual decrease of *G'* and *G''* with time, illustrated in Fig. 7. The onset of this time-dependent behavior does not depend significantly on the density of the sample: it appears above strain amplitudes of ~ 5 %. Applying this critical strain amplitude to the sample leads, after a certain shearing time, to its “liquefaction”, defined by the *G'*, *G''* crossover point, above which the mud is more dissipative than cohesive. For the sample considered in Fig. 5, one has to wait about 30min before the mud “liquefies” when submitted to a strain amplitude of ~ 4 %. Moreover the order of magnitude of the strain at which “liquefaction” occurs was shown to be ~ 100 %, whatever the sample considered.

Fig. 8 shows the linear viscoelastic moduli, G'_o and G''_o, as a function of the density, for all samples tested. In spite of a fairly great dispersion of the experimental data, these plots can be adequately fitted by exponential functions:



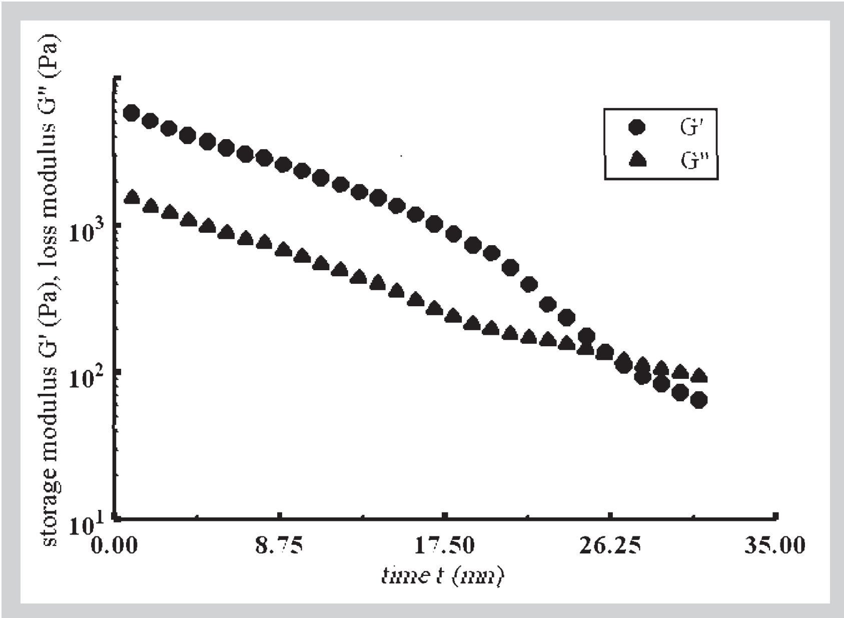


Figure 6 (above): The storage and loss moduli as a function of the frequency at a stress amplitude of 5 Pa (1440 kg/m³ mud sample).

Figure 7 (below): The time-dependent behavior of the storage and loss moduli at a stress amplitude of 280 Pa and a frequency of 1 Hz (1490 kg/m³ mud sample).

$$G'_o \cong 10^{-9} e^{0.02\rho_{mud}} \tag{10}$$

$$G''_o \cong \frac{G'_o}{10} \cong 10^{-10} e^{0.02\rho_{mud}} \tag{11}$$

where ρ_{mud} is expressed in kg/m³ while G'_O and G''_O are given in Pa. G'_O and G''_O can also be expressed in terms of the solid volume fraction, Φ, of the mud using Eq. 5:

$$G'_o \cong 0.5 e^{24\phi} \tag{12}$$

$$G''_o \cong \frac{G'_o}{10} \cong 0.05 e^{24\phi} \tag{13}$$

It is worth noticing that i) the exponential dependence on density for all material properties $F_O \equiv G'_O, G''_O$, and η_O is quite similar ($F_O \sim e^{0.02\rho mud}$) and ii) such a qualitative and quantitative dependence was obtained with other natural estuarine muds [12].

4 CONCLUDING DISCUSSION

The rheological response of natural estuarine mud samples to constant and oscillatory shear stress was investigated as a function of their density, with the aim of studying the linear and non-linear rheological behaviors of a mud under hydrodynamic forcing by currents and by water waves. Four different mechanical transitions in the rheological behavior of the mud have been defined:

a) a transition in the response to a constant stress, at a critical shear rate $\dot{\gamma}_c$ of a few 10^{-4} s^{-1}, marking the passage from a Newtonian behavior, with very high viscosity levels η_O, to a strongly non-Newtonian behavior. This transition can also be seen as the signature of an apparent yield stress $\tau_y = \eta_O \dot{\gamma}_c$, but $\dot{\gamma}_c$ seems to be more relevant than τ_y insofar as it does not depend significantly on the density of the mud sample, contrary to τ_y. This transition could be attributed to the shear rate governed rupture of the clay particle network in the cohesive sediment when submitted to a constant shear stress.

b) three transitions in the response to oscillatory shear stress at a frequency of 1 Hz. All these transitions have to be preferably defined in terms of strain amplitudes instead of stress amplitudes, since these strain transitions do not depend significantly on the density of the mud sample, contrary to the corresponding stress transitions.

At a strain amplitude γ_{lin} of a few hundredths %, the viscoelastic response becomes non-linear. Such a very small value of γ_{lin} is quite classical for concentrated non-colloidal suspensions, where non-linear effects appear for very small relative displacements of neighboring particles. Still this transition is not really relevant regarding the rheological behavior of muds under natural hydrodynamic forcing: the viscoelastic behavior of muds has to be considered as non-linear in most cases.

At a strain amplitude $\gamma_{destruct}$ of a few %, a strong time-dependent non-linearity appears, manifested by the continual decrease of the viscoelastic moduli as time elapses. This thixotropic-like behavior is the signature of a microstructural kinetics governing the destruc-

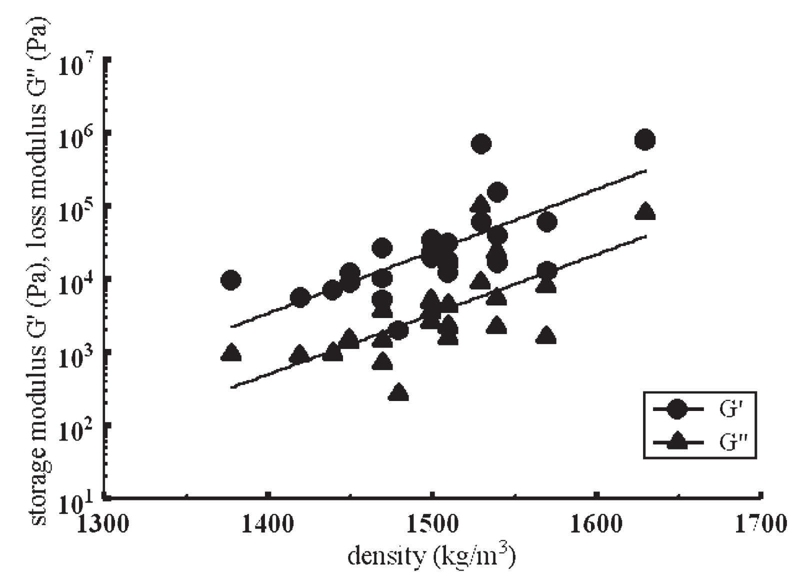


Figure 8: The linear viscoelastic moduli, at a frequency of 1Hz, as a function of the density of the mud.

turation of the mud above critical relative displacements of neighboring particles. This destructuration process finally leads to a liquefaction ($G'' > G'$) of the mud at a strain amplitude γ_{liquif} of about 100%, above which the sediments progressively lose their cohesive character. The transitions $\gamma_{destruct}$ and γ_{liquif} are certainly the most relevant one regarding the rheological response of natural muds. Indeed strain amplitudes of a few % are commonly encountered in natural hydrodynamic forcing [12], therefore liquefaction is expected to occur quite frequently, even for sediments which are very dense and cohesive at rest. At last, it is worth noticing that the order of magnitude of γ_{liquif} was found for another type of estuarine sediments, namely Seine estuary muds [12].

The mechanical transitions defined above mainly concern non-linear rheological behaviors of the mud. As far as linear rheological properties of this material are concerned, this work shows that the density dependence of the Newtonian viscosity η_0, and the linear viscoelastic moduli G'_0 and G''_0 can be satisfactorily fitted by an exponential function. Such exponential dependence of material properties with density, or volume fraction, was observed for other natural estuarine muds [12] and, more generally, for many other clay-water systems [13], even though the microstructural interpretation of such an exponential increase is not yet clear.

The qualitative and quantitative rheological results presented and discussed in the paper will be compared with those obtained on many different natural estuarine cohesive sediments in a near future in order to study the influence of mineral and organic content, granulometry and sea water properties on the rheological transitions and material properties studied in this work.

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