

LINEAR VISCOELASTIC BEHAVIOR OF BENTONITE-WATER SUSPENSIONS

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ABSTRACT:

Bentonite are extensively used materials in a wide range of applications. Creep and oscillatory shear experiments in the linear viscoelastic domain were carried out on bentonite-water suspensions at different solid fractions. It was found that bentonite dispersions exhibit important viscoelastic behavior which could be represented by the generalized Kelvin-Voigt mechanical model. It is well known that an exhaustive study of colloidal dispersions may require the determination of its viscoelastic properties over a wide frequency scale. Unfortunately, due to microstructure changes, the experiments are limited in time. In order to avoid such limitation, oscillatory data were deduced from creep curves - without actually vibrating the clay dispersions - because a periodic experiment at frequency ω is qualitatively equivalent to a creep test at time $1/\omega$. That is, it was possible to complete the dynamic response in the low-frequency range using data obtained from the transient response in creep.

ZUSAMMENFASSUNG:

Bentonit ist ein häufig verwendetes Material mit breitem Anwendungsspektrum. Dehnungs- und oszillatorische Scherexperimente wurden im linearen viskoelastischen Bereich für Bentonit-Wasser-Suspensionen mit unterschiedlichen Feststoffgehalten durchgeführt. Wir finden, dass diese Materialien ein viskoelastisches Verhalten aufweisen, das sich durch das generalisierte Kelvin-Voigt-Modell beschreiben lässt. Es ist wohlbekannt, dass die erschöpfende Untersuchung kolloidaler Suspensionen die Kenntnis ihrer viskoelastischen Eigenschaften über einen grossen Frequenzbereich erfordert. Unglücklicherweise, bedingt durch mikrostrukturelle Umordnungen, sind die Experimente nur während eines beschränkten Zeitintervalls durchführbar. Um diese Beschränkung zu umgehen, wurden Schwingungsdaten von den Ausdehnungskurven abgeleitet – ohne die Dispersionen in Schwingungen zu versetzen – da ein periodisches Experiment bei Frequenz ω einem Zeitstandsversuch zur Zeit $1/\omega$ qualitativ gleichwertig ist. Das heisst, es war möglich, die dynamische Resonanz im Niederfrequenzbereich zu ermitteln, indem Daten verwendet werden, die aus der zeitabhängigen Antwort während der Ausdehnung erhalten werden.

RÉSUMÉ:

Les bentonites sont des argiles couramment utilisées dans diverses applications industrielles. Les expériences en fluage et en régime dynamique ont permis de mettre en évidence le caractère viscoélastique des suspensions de bentonite. On a notamment montré que ces propriétés viscoélastiques peuvent être correctement décrites par un modèle analogique de type Kelvin-Voigt. Il est admis qu'une étude complète des suspensions colloïdales nécessite la connaissance des propriétés viscoélastiques sur une gamme de fréquences très large (souvent de 10 à 12 décades). En raison des modifications structurales intervenant pendant la mesure, les expériences sont limitées dans le temps. Pour contourner ce problème, et en se basant sur le postulat qu'une expérience en dynamique à une fréquence ω est qualitativement équivalente à une expérience en fluage à un temps $1/\omega$, il a été possible de calculer les grandeurs dynamiques, aux faibles fréquences, à partir des mesures faites en fluage, sans passer par l'expérience.

KEY WORDS: bentonite suspensions, viscoelastic properties, creep curve, storage and loss moduli

$$J''(\omega) = \sum_{i=1}^n J_i \frac{\omega \theta_i}{1 + \omega^2 \theta_i^2} + \frac{t}{\eta_N} \quad (5)$$

Values for G' and G'' may be deduced using Eqs. 6 and 7:

$$G' = \frac{J'}{J'^2 + J''^2} \quad (6)$$

$$G'' = \frac{J''}{J'^2 + J''^2} \quad (7)$$

The results in terms of variation of the storage modulus G' with frequency are illustrated in Fig. 5. The high frequency values are derived from oscillatory measurements with the Carri-Med rheometer (solid symbols) and the low frequency values are obtained by transforming creep data (open symbols). One may observe that the transformed dynamic functions compare well with the functions obtained directly from oscillatory measurements and that the observable frequency scale increases from two to four decades. This method was applied elsewhere in the case of other complex fluids like foams [34].

4 CONCLUSION

The viscoelastic properties were investigated in terms of creep flow and dynamic tests. It was found that the dispersions exhibit pronounced viscoelastic properties for concentrations higher than 6% by weight which can be represented by the mechanical Kelvin-Voigt model. It was necessary to combine transient and oscillatory measurements to determine the viscoelastic properties over a sufficiently wide frequency range: The dynamic response in the low frequency range (long time) was completed using data from the transient response in creep. The remarkable feature revealed by this method is that the transformed dynamic functions compare well with functions obtained directly from oscillatory measurements.

NOTATIONS:

G' : Shear storage modulus [Pa]
 G'' : Shear loss modulus [Pa]
 $f(t)$: Over-all creep compliance
 $L(\theta)$: Retardation spectrum

J_0 :	Instantaneous elastic compliance [Pa ⁻¹]
J_i :	Retarded elastic compliance of the i th component [Pa ⁻¹]
J' :	Storage compliance [Pa ⁻¹]
J'' :	Loss compliance [Pa ⁻¹]
t :	Time [s]
δ :	Phase angle between the stress and the strain (loss angle) [rad]
$\dot{\gamma}$:	Shear rate [s ⁻¹]
γ_0 :	Strain amplitude [-]
η_i :	Viscosity of the i th component [Pas]
η_N :	Newtonian viscosity [Pas]
θ_i :	Retardation time [s]
τ_0 :	Stress amplitude [Pa]
ω :	Frequency of oscillation [Hz]

REFERENCES

- [1] Akhatov ISH, Khasanov MM and Khusainov: Movement stability analysis of a pipe string in a thixotropic fluid, J. Engineering Physics and Thermophysics 66 (1994) 353-359.
- [2] Schmitt L, Ghnassia G, Bimbenet JJ and Cuvelier G: Flow properties of stirred yogurt: calculation of the pressure drop for a thixotropic fluid, Journal of Fluid Engineering 37 (1998) 367-388.
- [3] Clark. CR and Carter LG: Mud displacement with cement slurries, J. Petroleum Tech. 1 (1973) 775-783.
- [4] Speers RA, Tung M.A and Williamson DT: Rheological determination of peptizing agents in bentonite clays, Rheol. Acta 27 (1988) 561-564.
- [5] Van Olphen H : An Introduction to Clay Colloid Chemistry, Wiley, New York (1963).
- [6] Luckam PF and Rossi S: The colloidal and rheological properties of bentonite suspensions, Adv. Colloid Interface Sci. 82 (1999) 43-92.
- [7] Durán JDG, Ramos-Tejada MM, Arroyo FJ and González-Caballero F: Rheological and electroki-

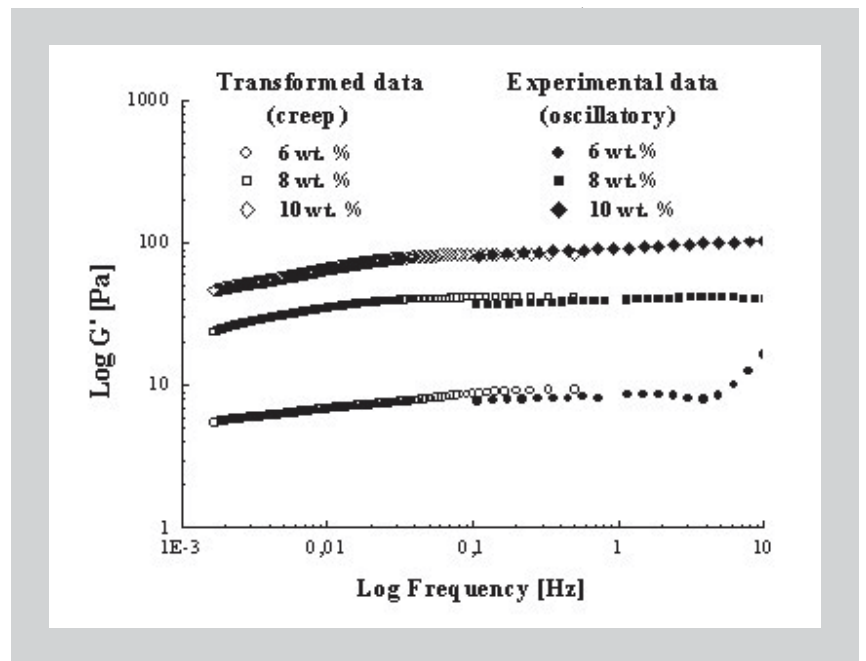


Figure 5: Unified plots of the variation of the storage modulus G' with frequency for three bentonite suspensions (6, 8 and 10wt%). Low frequency values are derived by transforming creep data (open symbols), and high frequency values are measured by oscillatory measurements with a Carri-Med rheometer (solid symbols).

- netic properties of sodium montmorillonite suspensions. I. Rheological properties and interparticle energy of interaction, *J. Colloid Interface Sci.* 229 (2000) 107-117.
- [8] Ramos-Tejada MM, Arroyo FJ, Perea R and Durán JDG: Scaling behavior of the rheological properties of montmorillonite suspensions: correlation between interparticle interaction and degree of flocculation, *J. Colloid Interface Sci.* 235 (2001) 251-259.
- [9] Ramos-Tejada MM, de Vicente J, Ontiveros A and Durán JDG: Effect of humic acid adsorption on the rheological properties of sodium montmorillonite suspensions, *J. Rheol.* 45 (2001), 1159-1172.
- [10] Gao D, Heimann RB, Williams MC, Wardhaugh LT and Muhammad M: Rheological properties of poly(acrylamide)-bentonite composite hydrogels, *Journal of Materials Science* 34 (1999) 1543-1552.
- [11] Barry BW: Viscoelastic properties of concentrated emulsions, *Adv. Colloid and Interface Sci.* 5 (1975) 37-75.
- [12] Benzing DW, Russel WB: *J. Colloid Interface Sci.* 83 (1981) 178-190.
- [13] Mitaku S., Ohtsuki T. and Okano K: *Jap. J. of Applied Physics* 19 (1980) 439-448.
- [14] Legrand C and Da Costa F: The effects of vibrating on the rheological behaviour of bentonite mud. Comparison with cement pastes, *Materials and Structures* 22 (1989) 133-137.
- [15] Legrand C and Da Costa F: The effects of shearing on the rheological behaviour of thixotropic muds, *Materials and Structures* 23 (1990) 126-130.
- [16] Grandjean J: Note: Water sites at a clay interface, *J. Colloid and Interface Sci.* 185 (1997) 554-556.
- [17] Bekkour K, Ern H and Scrivener O: Rheological characterization of bentonite suspensions and oil-in-water emulsions loaded with bentonite, *Appl. Rheol.* 11 (2001) 178-187.
- [18] Coussot P, Leonov AI and Piau JM: Rheology of concentrated dispersed systems in a low molecular weight matrix, *J. Non New. Fluid. Mech.* 46 (1993) 179-217.
- [19] Ancey C., Coussot P. and Evesque P: A theoretical framework for granular suspensions in a steady simple shear flow, *J. Rheol.* 43 (1999) 1673-1699.
- [20] Benna M, Kbir-Arigoib N, Magnin A and Bergaya F: Effect of pH on rheological properties of purified sodium bentonite suspensions, *J. Colloid and Interface Sci.* 218 (1999) 442-455.
- [21] Franck AJP: A Rheometer for Characterizing Polymer Melts and Suspensions in Shear Creep and Recovery Experiments, *J. Rheol.* 29 (1985) 833-850.
- [22] Bonifas JL: Contribution à l'étude de la viscosité de suspensions de bentonite, thèse de Doctorat, Université Louis Pasteur de Strasbourg, France (1998).
- [23] Piau JM: Crucial elements of yield stress fluid rheology. In: Adams MJ, Mashelkar A, Pearson JRA and Rennie AR (eds.), *Dynamics of Complex fluids*, Imperial College Press, The Royal Society, London (1998) 351-371.
- [24] Yoshimura A and Prud'homme RK: Wall slip for Couette and parallel disk viscometers, *J. Rheol.* 32 (1988) 53-67.
- [25] Alfrey TT and Gurnee EF: Dynamics of viscoelastic behavior, in: *Rheology, Theory and Applications*, Volume 1 (1956) 387-429, Eirich FR ed., Academic Press, New York.
- [26] Ferry J.D: Experimental techniques for rheological measurements on viscoelastic bodies, in: *Rheology, Theory and Applications*, Volume 2 (1958) 433-473, Eirich FR ed., Academic Press, New York.
- [27] Leaderman H: Viscoelastic phenomena in amorphous high polymeric systems, in: *Rheology, Theory and Applications*, Volume 2 (1958) 1-61, Eirich FR ed., Academic Press, New York.
- [28] Marchal P, Moan M and Choplin L: Rheological properties of clay-polymer suspensions, Proc. IXth European Rheology Conference, Sevilla (Spain), Steinkopff Verlag, Darmstadt (1994) 669-671.
- [29] Inokuchi K, *Bull. Chem. Society, Japan* 20 (1955) 453.
- [30] Warburton B and Barry BW: Concentric cylinder creep investigation of pharmaceutical semi-solids, *J. Pharm. Pharmacol.* 20 (1968) 255-268.
- [31] Ferry JD: *Viscoelastic Properties of Polymers*, Wiley and Sons, New York (1980).
- [32] Tschoegl NW: Time Dependence in Material Properties: An Overview, *Mechanics of Time-Dependent Materials* 1 (1997) 3-31.
- [33] Tschoegl NW: The phenomenological theory of linear viscoelastic behavior, Springer-Verlag, Heidelberg (1989).
- [34] Bekkour K and Scrivener O: Time-Dependent and Flow properties of Foams, *Mechanics of Time-Dependent Materials* 2 (1998) 171-193.

