

Rempei Gotoh<sup>1</sup> and Kiyoshi Shimizu<sup>2</sup>

## Abstract

It was shown that the shear moduli of the hydrogels of bentonite clay vary with the concentration of the clay, giving the relationship similar to that for polymer gels. Over the whole range of concentration (8-80 %) examined, a linear relation was obtained between the shear modulus ( $G$ ) and the critical shear stress ( $\sigma$ ) where the characteristic slip fracture occurs, the critical shear strain ( $\gamma$ ) taking values between  $\frac{1}{10}$  and  $\frac{1}{30}$ .

## Introduction

Gotoh and Hirai (1) found that a hydrogel of bentonite clay under a critical compression shows a distinct slip pattern which inclines at  $45^\circ$  to the direction of force as shown in Photo. 1. : this direction of slip is that of maximum shear in the specimen. Such a slip pattern is not observed for the bentonite dispersed in organic medium nor for that dehydrated by heating at temperatures higher than  $800^\circ\text{C}$ . In other words, it seems that the slipping occurs in the lyophilic bentonite, but not in the lyophobic one.

Aida, Hanai and Gotoh (2) investigated the visco-elastic behavior of the bentonite gel with a cone and plate viscometer. It was found that the bentonite gel showed an instantaneous elastic deformation followed by a viscous flow or yielding under a relatively low shear stress, while an abrupt slip of the cone was observed above a critical shear stress. A typical example of the visco-elastic behavior and slipping of bentonite gels is shown in Fig. 1. which was obtained by the present authors.

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1. Institute for Chemical Research, Kyoto University (Takatsuki, Osaka)  
Professor
  2. Hokkaido Gakugei University (Hakodate)  
Professor

It may reasonably be assumed that the critical shear stress at which the slip pattern appears under compression corresponds to that at which the cone slips. The critical shear stress can be measured by the shear testing box as well as by the above two kinds of methods.

In this paper, the critical shear stresses or the shear strength ( $\sigma'$ ) of bentonite gels are measured by the three methods and the shear moduli (G) by the compression method and the cone and plate viscometer over a wide range of concentration. The relations between  $\sigma'$  and G are discussed.

### Experimental

#### 1. Preparation of the specimen

Finely ground Wyoming bentonite (400g) was dispersed in 20 l of water. After the suspension was kept standing for a week and coarse particles were settled, the upper part (13 l) was taken and evaporated to various concentrations at a temperature below 95°C. Exact concentrations of each specimen were determined by drying at 105°C after the experiments. The gels thus obtained were molded with a brass cylinder and used for the compression method. The height and radius of the specimens were both 2.5 cm. So far as the height of the specimen was equal to or larger than the radius, the numerical result was found to depend mainly upon the concentration and not upon the size of the specimen.

#### 2. Measurements of the shear strength

The critical shear stress or the shear strength of the bentonite gels was measured by the following three kinds of methods:

(1) Compression method: The cylindrical specimen was placed on a table balance and compressed vertically with a screw device (Fig. 2). The critical point of compression (W) was detected by the backward motion of the pointer of the scale board when the slip pattern appears on the surface of specimen. The critical shear stress or and shear strength in the direction of slip pattern is given by (W/2). With this method, Young's moduli (E) of bentonite gels were measured and the shear moduli (G) obtained by using the relation  $E = 3G$ , the Poisson's ratio being assumed to be 0.5.

(2) Cone and plate method: The bentonite gel was placed between the cone and plate, and the cone was rotated by torques

caused by various loads through a pulley. The viscoelastic behavior was observed by the deflection of a mirror attached to the axis of rotation of the cone. The deflection of the mirror was recorded by an optical tracer. As is shown in Fig. 1, critical shear stresses for slipping of bentonite gels were determined at various concentrations. The shear moduli of gels can be obtained from the instantaneous deformations on loading or unloading.

(3) Shear testing box method: A two-surface shear testing box was used to measure the shear strength under various normal pressures. It was confirmed that the shear strength was independent of the normal pressure but depended upon the concentration of the gel.

### Results and Discussion

#### 1. Dependence of the elasticity of the gel on the concentration

Curve I in Fig. 3 shows the relation between the shear modulus and the concentration of bentonite gels. The experimental fact that the shear moduli (G) obtained by two different methods fall on the same line suggests that Poisson's ratio of the gel is nearly equal to 0.5. Curve I consists of the three parts having different slopes. This is the general feature of polymer gels as was pointed out by Hirai (3), who ascribed it to the rubber like network structure of polymer gels. Such an elastic feature suggests that bentonite gels also have network structures.

#### 2. Dependence of the shear strength on the concentration

The relation between the shear strength and the concentration is shown by Curve II in Fig. 3. Almost all values of the shear strength obtained by the three methods lie on the same line over a wide range of concentration. Moreover, Curve II shows a similar tendency to Curve I, thus suggesting that there holds a relation of proportionality between shear strengths ( $\sigma'$ ) and shear moduli (G). Fig. 4 shows this relationship. It is found that the values of shear strength lie in general within the range between  $\frac{G}{10}$  and  $\frac{G}{30}$ . In other words, we have a relation  $\sigma' = \gamma G$  where  $\sigma'$  is the critical shear stress and  $\gamma$  the critical shear strain which takes nearly constant values between  $\frac{1}{10}$  and  $\frac{1}{30}$ . G varies with the concentration of bentonite.

#### 3. Structure of gels and shear strength

Gel may be reasonably defined as a state of colloidal

dispersion which shows an elastic behavior. Naturally this property leads to the concept of the framework structure of gels as is mentioned above. According to light-scattering, birefringence and viscoelastic studies by M'Ewen (4,5) et.al., bentonite leaflet particles are aligned edge-to-edge in the form of flat ribbons at low concentrations, and make a three dimensional network structure based on a system of cross-linked ribbons at higher concentrations. Van Olphen (6) proposed a card-house structure for clay gels which is idealized as an edge-to-face associated cubic network of clay particles. In short, the bentonite gel has a network structure analogous to that of polymers, although bentonite itself consists of leaflet particles.

On the other hand, it is well known that the hydrogel of bentonite shows typical thixotropy and apparent viscosity varies with the rate of shear. Further, the gels yield or creep under very low rate of shear as is shown in Fig. 1. These facts suggest that the force of linking between bentonite particles is relatively weak. Thus it is expected that the framework of the gel is very brittle.

Now, the experimental results in Fig. 4 shows that the shear strength of the gels takes values ranging from  $\frac{G}{10}$  and  $\frac{G}{30}$ . These values agree fairly well with the theoretical one proposed by Mackenzie (7) for the shear strength of ideal crystals. Bragg and Lomer (8) demonstrated that the observed shear strength of a perfect two-dimensional crystal of bubbles agrees with the theoretical value,  $\frac{G}{30}$ . Further, Buchdahl (9) pointed out that the ratio of the modulus and the yield or breaking strength for a variety of amorphous polymer in the glassy state has the same order of magnitude as the theoretical value.

It is concluded from these facts that the proportional relationship between the shear strength and the shear modulus predicted by Mackenzie generally holds for brittle and relatively soft materials, and the critical strain lies between  $\frac{1}{30}$  and  $\frac{1}{10}$ . As is well known, however, this relationship does not hold for most of actual hard crystals.

Furthermore, an instantaneous change in birefringence was detected for a bentonite gel sheared between two transparent glass plates as is shown in Photo. 2. This fact indicated that the bentonite leaflets are oriented on shearing.

Thus following mechanism is proposed for the slip fracture of bentonite gel: under a critical shear stress, the card-house of the bentonite gel falls down one upon another starting

from some point of stress concentration and leaflets of the gel are oriented in the direction of the maximum shear, when the slipping occurs.

## References

1. R. Gotoh and N. Hirai : Bull. Inst. Chem. Res. Kyoto Univ., 20, 48 (1950).
2. H. Aida, T. Hanai and R. Gotoh : Nippon Kagaku Zasshi, 83, 536 (1962).
3. N. Hirai : Bull. Inst. Chem. Res. Kyoto Univ., 33, 21 (1955).
4. M. B. M'Ewen and M. I. Pratt : Trans. Farad. Soc., 53, 535 (1957).
5. M. B. M'Ewen and D. L. Mould : ibid. 53, 548 (1957).
6. H. van Olphen : " Clay Colloid Chemistry " Interscience Pub. New York, 103 (1963).
7. J. K. Mackenzie : " Dislocations and Plastic Flow in Crystals " by A. H. Cottrell. Oxford Univ. Press, London, 8 (1963).
8. W. L. Bragg and W. M. Lomer : Proc. Roy. Soc., A 196, 171 (1953).
9. R. Buchdahl : J. Polymer Sci., 28, 239 (1958).



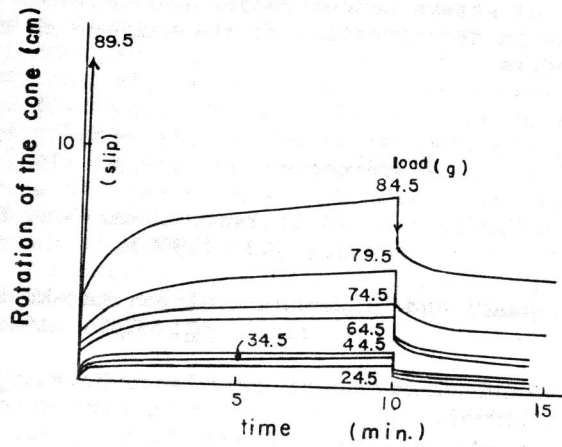


Fig. 1. Viscoelastic behavior and slip of a bentonite gel ( cone and plate method )

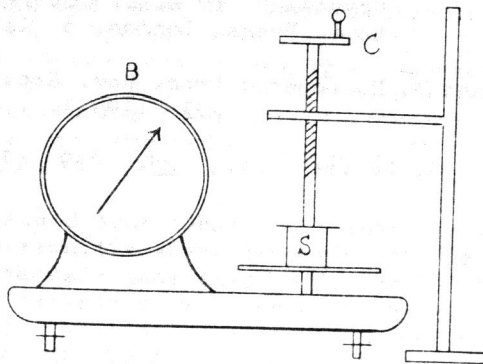


Fig. 2. Compression Method

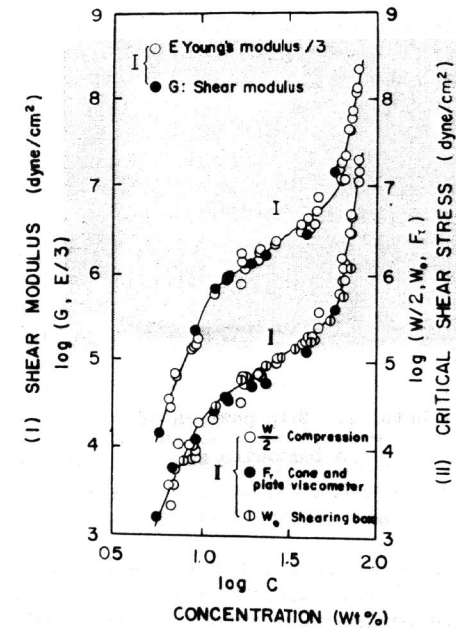


Fig. 3 Relation among shear modulus critical shear stress, and concentration

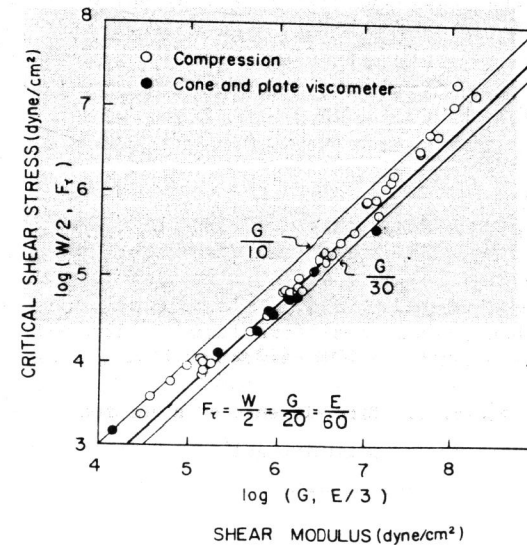


Fig. 4 Relation between critical shear stress and shear modulus

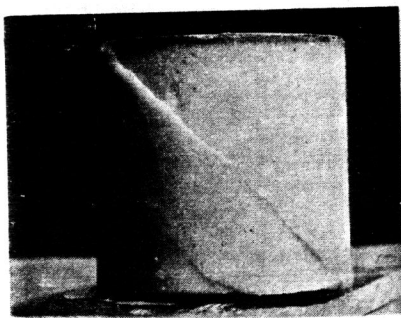


Photo. 1. Slip pattern of a bentonite gel

Intensity of birefringence

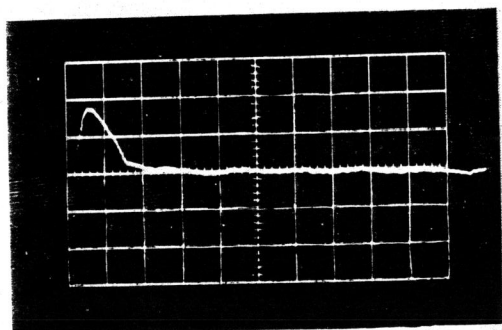


Photo. 2. Birefringence of a sheared bentonite gel.