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Bentonite dispersions: Transition from liquid-like to solid-like behavior and cracking

D.D. Pelot [a], S. Jun [a], A.L. Yarin [a,b,*]

[a] Department of Mechanical and Industrial Engineering, University of Illinois at Chicago, 842 W. Taylor St., Chicago, IL 60607-7022, United States
[b] College of Engineering, Korea University, Seoul, South Korea

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ABSTRACT

Cylindrical samples of 11–13 wt% aqueous bentonite dispersions with a range of resting times after preparation (0–72 h) were subject to axial squeezing and revealed a transition from a Bingham liquid-like behavior to solid-like behavior. The liquid-like behavior is characterized by viscous spreading arrested by the yield stress, whereas the solid-like behavior is characterized by stasis at lower loads followed by discontinuity of the displacement and cracking above some critical level of load. A mechanical response, dependent on resting time, implied irreversible aging as a result of slow swelling of the clay crystallites, i.e. the internal structure being built over time in the material at rest. Dispersions of 11 wt% and 12 wt% transitioned from liquid-like to solid-like behavior in a single experiment at a resting time of 24 h and 3 h, respectively. At higher concentrations (18–22 wt%) the material always behaves as a solid. The solid-like behavior is distinguished from the liquid-like behavior by the appearance of cracks. The additional bending and buckling experiments conducted with dispersions of 18–22 wt% revealed Young's moduli of 330–500 kPa, yield stresses of 9–15 kPa, cracking stresses of 15–25 kPa, and cracking strains of 8–10%. For all concentrations explored, a linear dependence of the yield stress on concentration was found. It is demonstrated for the first time using uniaxial compression that liquid bentonite dispersions at a concentration of 11–12 wt% reveal transition to a solid in a single experiment where they become capable of developing surfaces of discontinuity of the displacement.

1. Introduction

Bentonite is mainly composed of montmorillonite, a plate or leaflet shaped mineral that gives bentonite its unique properties such as the ability to swell. Its structure was first uncovered in Hoffman et al. [1]. Marshall and Krinbill [2] described montmorillonite as two silica sheets that sandwich an alumina sheet. Within the structure, rearrangement is also possible, that is, magnesium atoms can replace aluminum atoms and aluminum atoms can replace silicon atoms, as described by Mering [3] and Siguín et al. [4] who gave a general chemical formula. According to Marshall [5] the plates organize themselves with nearby particles in quasi-parallel groups. Then, Mering [3] attempted to describe various levels of organization of the platelets from small groups of platelets to parallel platelets joint laterally, as well as combinations of the two to create an organized structure within the material. When bentonite is dispersed in water, cations leave the structure through dissociation and enter the water layer between sheets causing repulsion between the platelets resulting in swelling of the material [3,4,6,7].

According to Clem and Doehler [8], bentonite has many industrial applications, in particular, as a binding agent, plasticizer, and suspending agent but can also be used as a filter or barrier. As a binding agent, bentonite is commonly used in foundry molding sand, in rock wool for insulation, pellets for animal feed, and pelleting of finely divided magnetite concentrates. It can be used as a plasticizer in ceramic, concrete products, other construction materials, paints, greases, lubricants, and cosmetics. As a suspending agent it is used as a viscosity modifier, drilling mud, fire retardant gel, and media for ingesting medicine. Due to the platelet shape of bentonite, it has a high surface to volume ratio, which makes it an ideal carrier for chemicals, such as insecticides, or as a filter for decolorizers of oil, clarifying agents in wine and beer, paper coatings, paint, pharmaceuticals, cosmetics, and metals from the environment [8–10]. In addition, it can also be used as a containment barrier as in cutoff walls, geosynthetic clay liners, landfill liners, overpack for radioactive canisters, and for borehole and shaft sealing [11–14]. It is not surprising then that in 2005 over

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[*] Corresponding author at: Department of Mechanical and Industrial Engineering, University of Illinois at Chicago, 842 W. Taylor St., Chicago, IL 60607-7022, United States.

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3.5 million metric tons of bentonite were used in the United States according to the U.S. Geological Survey [15].

Although there are a wide variety of applications for bentonite, each application has a unique range of concentrations that is most appropriate for its purpose. For example, pelletizing iron ore concentrates use a concentration of less than 1 wt% bentonite, whereas drilling mud can be as low as 3 wt% bentonite, and molding sands in foundries can be in the range of 5–12 wt% bentonite [16,17]. In outside applications, such as liners or back fill, bentonite will be subjected to rain, humidity, or evaporation (desiccation) where the change in concentration could alter the yield stress, flow properties, and cracking stress, therefore changing the permeability of the material. Furthermore, the resting time of bentonite dispersions can also have an effect on the yield stress as was shown in [18]. It is in this regard that this study was undertaken. We aimed at understanding how small changes in concentration or extended resting periods affect the mechanical behavior of bentonite. In particular, we were interested to establish and measure mechanical properties of bentonite dispersions in a range of concentrations that reveal both liquid and solid behavior and understand the cracking mechanism.

A rheological study of dispersed bentonite at a concentration of 10 wt% was conducted in the recent work of the present group in Pelot et al. [19]. Pelot et al. [19] employed uniaxial compression with a constant force to samples of a given volume and demonstrated that such materials behave as Bingham liquids, i.e. approximately follow the phenomenological constitutive equation determined by a viscosity and yield stress.

In their seminal but practically forgotten work, Gotoh and Shimizu [20] also used uniaxial compression to study "slip lines" of bentonite dispersions after a resting time of seven days. They removed coarse particles, which influence flow behavior [21], from their samples by using suspension settling and created different concentrations through evaporation. Due to their sample preparation, they were able to produce "slip lines" at unusually low concentrations (<10 wt%). Therefore, they did not investigate the transition from flowing to discontinuity of the displacement of bentonite samples through an increase in concentration or resting time, i.e. the transition from the liquid-like to solid-like behavior. Since bentonite has multiple applications which utilize different concentration ranges and have a wide range of stresses imposed, understanding how, when, and what occurs during a transition of behavior has significant importance. This is one of the motivations of the present work.

Other authors have investigated transitional behavior of bentonite dispersions. The work of Coussot et al. [22] identifies different ranges of thixotropic material behavior by observing the motion of bentonite dispersion at different concentrations and resting times moving down a ramp positioned at different angles. The height of the material provides a hydrostatic pressure which results in a stress at the wall. If this shear stress is greater than the yield stress, due to the angle of inclination, concentration and resting time, the material is sheared at the wall and flows down the ramp. It is not discussed but would be interesting to measure the height of the fluid where the "tail" and "front" separated since this may represent a similar normal stress that is employed in the present work.

Raynaud et al. [23] have also investigated the influence of concentration and resting time on flows of bentonite dispersions. Using nuclear magnetic resonance imaging (NMRI) they were able to measure the velocity profile within the flow between concentric cylinders. They concluded that the steady-state interface between sheared and un-sheared material is influenced by the resting time, concentration, and rotational velocity of the inner cylinder. Specifically, the sheared region decreases with increasing resting time, increasing concentration, and decreasing rotational velocity.

A perspective on the unique behavior of complex fluids, such as bentonite dispersions, was given by Ovarlez et al. [24,25]. This work describes discontinuity of the displacement as "the shear rate profile in a flowing material exhibits an apparent discontinuity…the shear rate takes two significantly different values in two adjoining regions…" [25]. A specific example is given of cement pastes [26] where an interface exists between a sheared region with a constant shear rate and an un-sheared solid-like region. Furthermore, when a sheared region is confined between unyielded regions, this is attributed to surfaces of discontinuity of the displacement.

In the present work, the transition of liquid-like to solid-like behavior of bentonite dispersions is investigated using uniaxial compression. Liquid-like behavior is defined by material flowing axisymmetrically under compression, whereas solid-like behavior is defined by the presence of surfaces of discontinuity of the displacement created by high shear stress which causes the material to irreversibly slip along a certain surface. The transition from liquid-like to solid-like behavior is revealed through an increase in concentration as well as resting time.

The experimental methods are described in Section 2, the experimental results are presented in Section 3, discussion is contained in Section 4, and conclusions are drawn in Section 5.

2. Experimental

2.1. A material preparation

Bentonite powder (100 wt% pure clay) was obtained from NOW FOODS Inc. and mixed with DI water to prepare 11 wt%, 12 wt%, 13 wt%, 18 wt%, 20 wt%, and 22 wt% bentonite dispersions. The bentonite powder is a sodium type bentonite according to the manufacturer and has a particle size of 5–10 μm. Bentonite dispersions were prepared by mechanical mixing using a handheld kitchen mixer (Hamilton Beach) with the standard mixer attachment initially for 10 min, left for at least 3 days to ensure hydration, and again mixed before each test, except those which required a resting time.

Air bubbles were not entrained during the creation of the bentonite dispersions by taking great care not to fold the material while stirring (several stirrers were tried) as well as forcefully banging of the plastic container on the table which caused the bubbles to rise to the surface during and immediately after stirring. The prepared dispersions were discarded if any significant air pockets were observed when they were dispensed.

The 11 wt%, 12 wt%, and 13 wt% bentonite dispersions were chosen based on their ability to reveal transitional flow behavior under stress due to compression. If the concentration was decreased further, to 10 wt% or less, the material does reveal any transitional behavior given the allotted resting time. This is corroborated by our earlier study in Pelot et al. [19]. If the concentration of bentonite is 13 wt%, then discontinuity of the displacement occurs without any resting time and is related to the initial concentration that results in an immediate solid-like response after stirring. In some cases 12 wt% dispersions without any resting time did reveal discontinuity of the displacement but only near the plates which squeeze the sample (see Section 2.3 below), not through the bulk of the sample.

The higher concentrations of bentonite (18 wt%, 20 wt%, and 22 wt%) were chosen due to their ability to retain the sample shape for bending experiments (see Section 2.4 below). The bending experiments were chosen to find the mechanical behavior because Thusyanthan et al. [27] demonstrated that when materials are too weak to be clamped their mechanical properties can be found by a bending test. The higher-concentration samples were also tested in