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Key Points:
- We investigated rheological properties of composite smectite-quartz suspensions at variable mixing ratios under the seafloor conditions
- At a constant solid/water ratio, the yield stress and viscosity increase with increasing smectite content in the smectite-quartz mixture
- Smectite can effectively reduce the risk of slope failure and subsequent spreading of sediment gravity flows

Supporting Information:
- Supporting Information S1

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Sensitivity of Clay Suspension Rheological Properties to pH, Temperature, Salinity, and Smectite-Quartz Ratio

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Abstract Understanding the rheological properties of clay suspensions is critical to assessing the behavior of sediment gravity flows such as debris flow or turbidity current. We conducted rheological measurements of composite smectite-quartz suspensions at a temperature of 7°C and a salt concentration of 0.6 *M*. This is representative of smectite-bearing sediments under conditions on the seafloor. The flow curves obtained were fitted by the Bingham fluid model, from which we determined the Bingham yield stress and dynamic viscosity of each suspension. At a constant smectite-quartz mixing ratio, the yield stress and the dynamic viscosity tend to increase as the solid/water ratio of the suspension is increased. In the case of a constant solid/water ratio, these values increase with increasing smectite content in the smectite-quartz mixture. Additional experiments exploring differing physicochemical conditions (pH 1.0–9.0; temperature 2–30°C; and electrolyte (NaCl) concentration 0.2–0.6 *M*) revealed that the influence of temperature is negligible, while pH moderately affects the rheology of the suspension. More significantly, the electrolyte concentration greatly affects the flow behavior. These variations can be explained by direct and/or indirect (double-layer) interactions between smectite-smectite particles as well as between smectite-quartz particles in the suspension. Although smectite is known as a frictionally weak material, our experimental results suggest that its occurrence can reduce the likelihood that slope failure initiates. Furthermore, smectite can effectively suppress the spreading distance once the slope has failed.

1. Introduction

The flow behavior of clay suspensions is of great interest in industrial applications, such as the use of clay minerals in drilling fluids, as ceramic additives, and in cosmetic/pharmaceutical formulations. Smectite is the most extensively studied clay material, and only a few percent of smectite in a suspension can significantly modify its rheology (e.g., Lagaly, 1993). In some cases, 1 vol % or an even lower concentration of smectite can cause a sol-gel transition (Abend & Lagaly, 2000). Aside from the concentration of smectite in the suspension, the flow behavior depends on numerous other factors such as the pH, presence of electrolytes, temperature, particle size, and method of sample preparation (Kelessidis, 2017).

In nature, smectite is widely distributed on the Earth's surface as a product of geologic processes such as rock weathering, the alteration of volcanic ash, and numerous hydrothermal reactions (Meunier, 2005). As smectite is a major constituent of soils and seafloor sediments, accurate knowledge of the flow behavior of smectite-bearing suspensions is important to gain a better understanding of subaerial and submarine sediment gravity flows such as debris flow or turbidity current. In contrast to subaerial sediment gravity flows, submarine flow events show a greater mobility of the flowing material (Hampton et al., 1996), often to the extent of >100 km. Nearshore sediment gravity flows cause direct destruction by triggering tsunamis and damaging structures such as submarine cables (Locat & Lee, 2002). Recent progress in numerical modeling makes it possible to precisely predict the runout behavior of a submarine sediment gravity flow if the suspension rheology is known (Imran et al., 2001; Jiang & LeBlond, 1993).

Although numerous rheological experiments have been performed on smectite suspensions, none has focused on the effect exerted by the presence of other rock-forming minerals, despite this being the typical situation in real geologic environments. To examine the effect of such minerals on the flow behavior of natural soils and sediments, we conducted rheological experiments on composite smectite-quartz suspensions, which are a comparable system to smectite-bearing seafloor sediments. We particularly focus on how the smectite-quartz ratio affects the rheology of the bulk suspension. To this end, experiments were conducted with a wide range of smectite-quartz ratios. We also conducted flow experiments with varying physicochemical conditions (pH, temperature, and ionic strength) because the flow behaviors of smectite

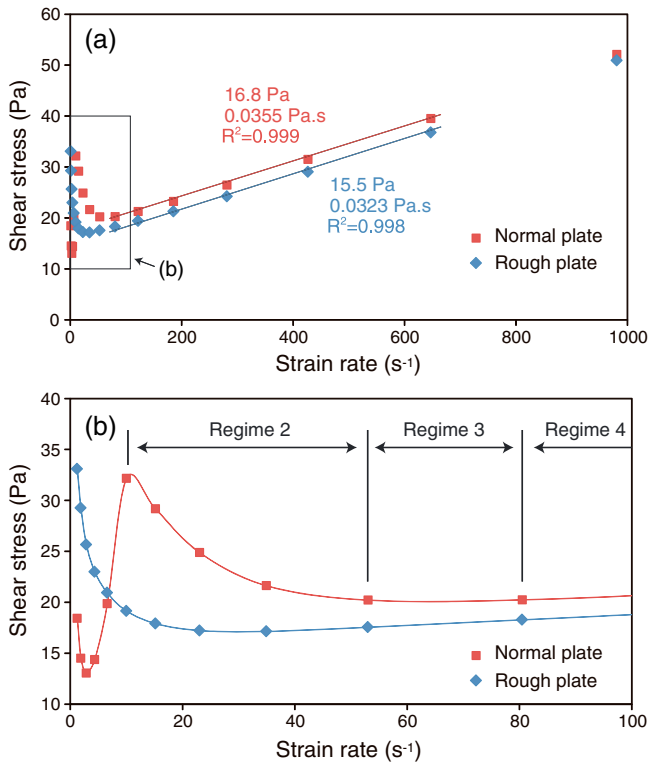

Figure 1. (a) Typical flow curves (strain rate versus shear stress) for the 50 *w/v*% suspension containing 25 wt % smectite in the solid component. The red and blue symbols indicate data obtained using the normal and rough plates, respectively. The yield stress and dynamic viscosity were determined by fitting the data using the Bingham fluid model (solid lines). (b) An enlarged portion of Figure 1a for strain rate between 0 and 100 s^{-1}. The flow stages (Regimes 2–4) were defined after Pignon et al. (1996).

suspensions are strongly influenced by these factors due to modifying smectite surface properties (Kelessidis, 2017), and thus, these experiments can give important implications of the mechanism controlling the suspension rheology. Based on our experimental results, we discuss the significance of smectite abundance on the runout behavior of submarine debris flows or turbidity currents.

2. Methods

2.1. Materials and Sample Preparation

We prepared composite smectite-quartz powders by mixing a commercially available Na-smectite (Kunipia-F; Kunimine Industries) and crystalline quartz (silicon dioxide 199–00625; Wako Pure Chemical Industries) at smectite weight fractions of 10, 25, 40, 60, 80, and 100 wt %. Kunipia-F is a high-purity montmorillonite powder containing only 1 wt % quartz and is produced from natural bentonite from the Tsukinuno mine, Japan. Although the specific density of the montmorillonite is 2,880 kg/m^3 under dry conditions, the montmorillonite specimen occurs with interlayer water to form its one-layer hydration state (d_{001} = 12.7 A) at ambient conditions (Colten-Bradley, 1987), as revealed by X-ray diffraction (XRD) analysis (Figure S1 in the supporting information). This implies that its specific density is ~2,500 kg/m^3 as the samples are being prepared. More information on the chemical properties of Kunipia-F can be found in Tachi and Yotsuji (2014). The quartz powder we used is composed of angular crystals with sand-sized grains (average grain size of ~100 μm; Oohashi et al., 2015), which is relevant for turbidity currents. No impurity was detected by XRD measurements (Figure S1).

Once weighed, the solid powders were dispersed in solutions with controlled pH and electrolyte (NaCl) concentrations (pH 1.0–9.0 and 0.2–0.6 *M*; representative seawater pH and NaCl concentration are ~8.0 and 0.47 *M*, respectively; Stumm & Morgan, 1996). A 50 mL polypropylene conical tube was used to prepare suspensions with different solid weight concentrations, ranging from 10 to 80 *w/v*% (i.e., the weight of the solid component (*g*) divided by the volume of the suspension (cm^3)). This is equivalent to a porosity range of ~65–95%. The pH and electrolyte concentration of the fluid were controlled by adding 0.1 N HCl solution and known weights of NaCl, respectively, to the suspension. The tubes were shaken for ~10 min to homogenize the samples before the experiments.

2.2. Rheological Measurements

All the rheological tests were conducted using an HR-2 rheometer (TA Instruments) with a parallel plate geometry. The geometry is composed of an aluminum rotational upper plate, with a 40 mm diameter, and a stationary lower Peltier plate, which controls the temperature (normally kept at 7°C, except when investigating the influence of temperature). There was a 1 mm gap between the plates. The suspensions were loaded between the plates, and their rheology was tested by measuring the shear stress τ under the applied strain rate $\dot{\gamma}$, which was increased as the upper plate was continuously accelerated from 0 to 1,300 s^{-1} (sampling interval is 5 points per decade for 10 min).

3. Experimental Results

Figure 1 shows a typical flow curve (shear stress versus strain rate) for the 50 *w/v*% suspension containing a solid mixture of 25 wt % smectite and 75 wt % quartz dispersed in a 0.6 *M* solution (red symbols in Figures 1a and 1b). The flow curve shows an initial stress peak at a strain rate of ~10 s^{-1}, followed by a gradual decrease in stress up to a strain rate of ~50 s^{-1} (Figure 1b), followed by a further rise with increasing strain rate. When carrying out rheometric experiments on suspensions, care must be taken to a strain discontinuity within the

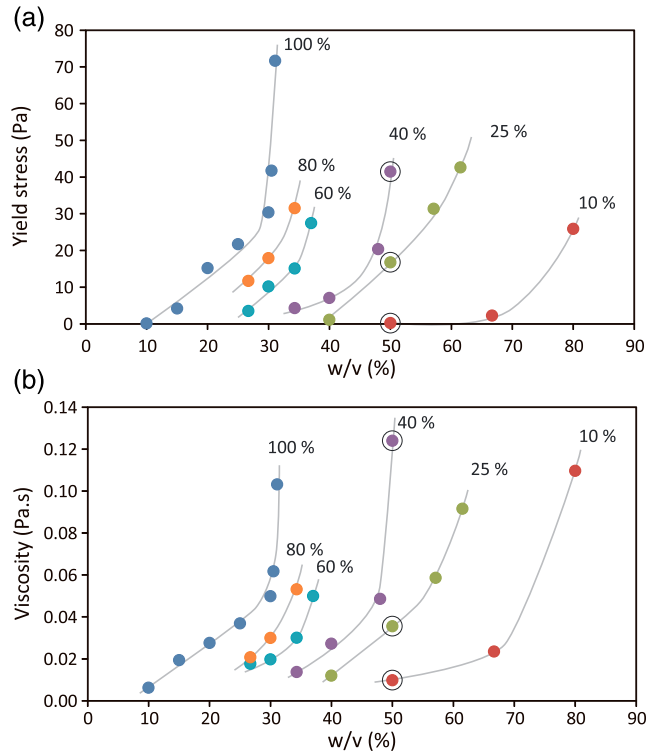

Figure 2. (a) Summary plots of yield stress against *w/v*%, and (b) viscosity against *w/v*%. Relationships are shown as a function of the smectite fraction in the solid component. The data adopted for runout calculations (50 *w/v*%) are highlighted by double circles.

sheared material (Coussot & Piau, 1994). To check for the possible influence of wall slip, a shear experiment was also conducted using a rough upper plate, which was prepared by attaching waterproof sandpaper of #100 (φ = 125 μm) to the conventional flat plate (blue symbols in Figure 1). Comparison of the two curves indicates that the effect of wall slip is not significant at higher strain rates (>100 s^{-1}), although the stress peak appears at the onset of shearing when using the rough plate. A flow experiment with preshearing (at 1,000 s^{-1} for 1 min) shows that the stress peak was reduced by this treatment (Figure S2) despite the almost comparable flow curves at higher strain rates. This observation implies that such a high stress at the lowest strain rate may be caused by an insufficient normal stress relaxing during the sample loading between the plates (Heymann et al., 2002). The flow characteristics at strain rates of greater than >100 s^{-1} seen in our experiment have also been observed in prior work on colloidal suspensions of synthetic smectite (Pignon et al., 1996). From visual observations of the strain field within the sheared suspension, Pignon et al. (1996) documented that the stress drop is due to the onset of discontinuous and localized deformation (Regime 2) and that homogeneous strain (Regime 4) is achieved after a stage of growth/thickening of the localized shear zone (Regime 3). Particle settling, which may also affect rheology (Scotto di Santolo et al., 2010), is expected to be more significant at lower strain rates. Based on these findings, we analyzed the flow curve over the portion where the stress successively increases as the strain rate increases (i.e., Regime 4 from Pignon et al., 1996; Figure 1). The relationship between stress and strain rate over this portion exhibits an almost linear correlation and was reasonably fitted by the following equation for a Bingham fluid:

$$\tau = \tau_y + \mu\dot{\gamma} \tag{1}$$

where τ_y is the Bingham yield stress and μ is the dynamic viscosity. The values for these parameters obtained from the smooth and rough plates are comparable; therefore, all the data were collected using the conventional plate geometry. It is also noted that the suspensions commonly show a thickening behavior at much higher strain rates (e.g., >700 s^{-1} for the experiments shown in Figure 1), and thus, this portion was also precluded from data fitting.

Figures 2a and 2b show variations in the Bingham yield stress and the dynamic viscosity of the suspensions as a function of *w/v*%, respectively (also summarized in Table S1). Figure 2 shows data for the same smectite-quartz mixing ratio, marked with identical symbols. Both the yield stress and dynamic viscosity tend to increase with an increase in the *w/v*% for a fixed smectite-quartz mixing ratio. The yield stress and viscosity measured for the pure smectite suspension at 10 *w/v*% are 9.43×10^{-2} Pa and 6.12×10^{-3} Pa s, respectively. These values show a progressive increase up to 30.3 Pa and 4.99×10^{-2} Pa s at 30 *w/v*%. At higher solid concentrations in the suspension, the values dramatically increase even for a small increase in *w/v*%. Similar relationships were observed for every series of experiments with a fixed smectite-quartz ratio. However, when we compare the effect of different mixing ratios, the yield stress and dynamic viscosity show a systematic decrease with increasing quartz content.

We also examined the influences of pH, temperature, and electrolyte concentration on the flow behavior of the suspension. The initial pH of the suspension (25 wt % smectite, 50 *w/v*%) was 8.91, and we successively added HCl solution to progressively shift the pH toward more acidic conditions. The flow behavior was moderately affected by the pH of the suspension; that is, the yield stress and dynamic viscosity gradually decreased at lower pH (Figure 3a). We conducted a series of measurements at different temperatures ranging from 2 to 30°C (40 wt % smectite content, 40 *w/v*% suspension) but did not find any apparent change in

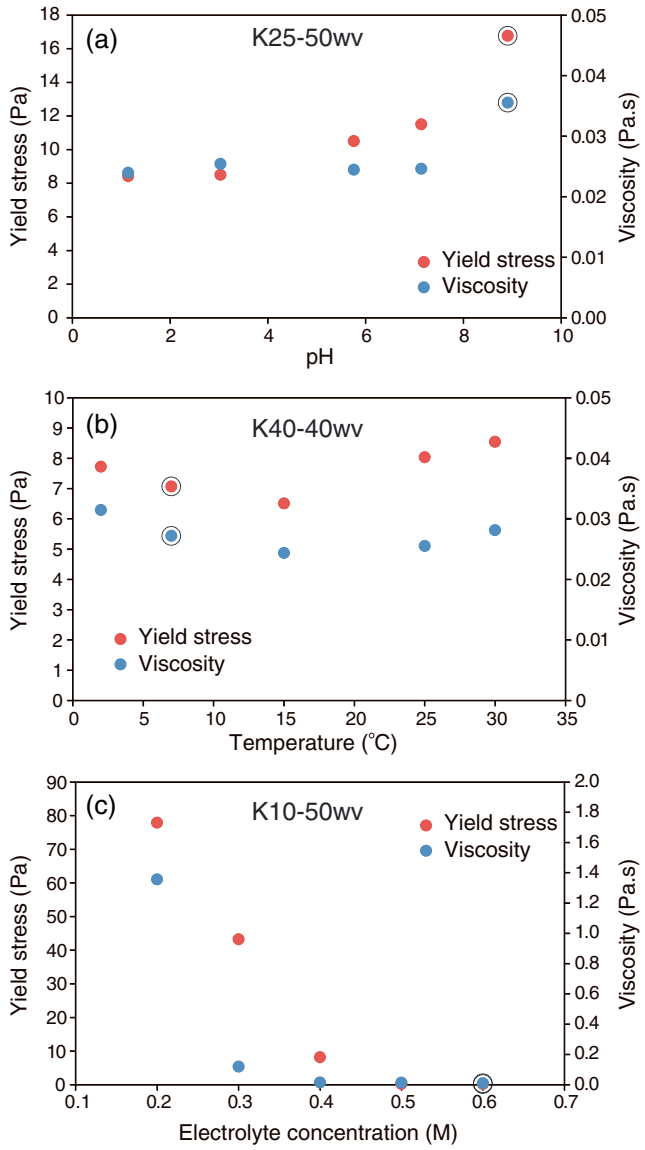

Figure 3. Variations in yield stress and viscosity for different physicochemical conditions. (a) pH, (b) temperature, and (c) electrolyte concentration. The reference data plotted in Figure 3 are highlighted by double circles.

rheological parameters over the tested range (Figure 3b). In contrast, the electrolyte concentration greatly affects the suspension rheology (10 wt % smectite content, 50 *w*/*v*% suspension), and both the yield stress and the dynamic viscosity gradually increase with decreasing electrolyte concentration from 0.6 to 0.4 *M* and then dramatically increase with further decreases (Figure 3c).

4. Discussion

4.1. Factors Affecting the Rheological Properties of Composite Smectite-Quartz Suspensions

Our experiments demonstrate that the rheological properties of composite smectite-quartz suspensions are strongly affected by certain parameters. For a fixed smectite-quartz mixing ratio, an increase in the solid concentration effectively strengthens the suspension, increasing both the yield stress and dynamic viscosity (Figure 2). In a suspension containing fine clay particles, a complex network may form via direct and/or indirect (i.e., double-layer) interactions between the particles (Coussot et al., 1993). The increase in the solid concentration enhances the intensity of such interactions, and this is likely seen as a higher yield stress and viscosity of the suspension (Coussot & Piau, 1994). Furthermore, the present experiment also showed that the increase in smectite content can significantly strengthen its yield stress and viscosity even for a constant concentration of solids. This may be due to enhancement of double-layer interactions between smectite particles. Several studies have investigated the effect of silica nanoparticles (amorphous silica, ~20 nm diameter) on the rheological behavior of smectite suspensions (Hilhorst et al., 2014; Kleshchanok et al., 2012; Landman et al., 2014). These works found that small quantities of silica nanoparticles can significantly modify the transitional behavior of the liquid crystal phases in smectite suspensions. Landman et al. (2014) went further and reported that the yield stress and viscosity show a progressive reduction with increasing silica fraction. They suggested that a reduction in double-layer interactions due to the addition of silica spheres could reduce these values. Although the grain size is quite different (~100 μm), the present data suggest that the addition of quartz crystals may also alter and reduce the interparticle interactions between the smectite platelets in the suspension.

Electrolyte concentration is another parameter that greatly affects the rheological behavior of a smectite-quartz suspension. In general, the presence of inorganic salts reduces the dispersivity of smectite particles and/or destroys their flocculations due to compression of the diffuse double layers of individual smectite platelets (Mitchell & Soga, 2005). Several studies have shown that only a few percent mass of smectite can cause a noticeable yield stress of the suspension (Kelessidis, 2017), but in this experiment we observed the apparent increase of the yield stress from 10 *w*/*v*% (curve for 100 wt % of smectite; Figure 2). This difference may be due to a high salinity of the present suspensions (0.6 *M*). In fact, the yield stress and viscosity of smectite suspensions decrease with increasing electrolyte concentration (Abu-Jdayil, 2011; Kelessidis et al., 2006), which is consistent with the present results. Moreover, our data demonstrate that as little as 10 wt % of smectite in the solid can effectively modify the bulk suspension rheology in response to changes in the electrolyte concentration.

It is known that pH has a significant influence on smectite suspensions, because smectite platelets have two types of surfaces (edge and face) exhibiting differing pH-dependent surface charge properties, and thus, the association mode of the platelets varies with pH (e.g., Kelessidis, 2017). Kelessidis et al. (2007) investigated the dependence of rheological behavior on pH for bentonite suspensions and observed a progressive increase in

the yield stress from pH ~7.6 to ~9.0, which then began to decrease at higher pH. On the other hand, Benna et al. (1999) found that the yield stress of a Na-smectite suspension is at its minimum around a neutral pH and then greatly increases with a decrease in pH, down to ~3–5. Our experiment showed that the yield stress decreases with decreasing pH from ~9.0 to ~1.0 (Figure 3a). This is comparable to the results of Kelessidis et al. (2007) for neutral to weakly alkaline conditions, while it differs greatly from the findings of Benna et al. (1999) for acidic conditions. These various results can be explained by the presence/absence of quartz grains, as Kelessidis et al. (2007) used a bentonite sample, which is dominated by montmorillonite but contains quartz and feldspar, while Benna et al. (1999) used a suspension free from such grains. The surface charge of quartz is governed by the dissociation manner of silanol groups (Dove & Rimstidt, 1994), and the positive-to-negative change of the quartz surface charge occurs at pH ~2–3. Under such acidic conditions, electrostatic association between the positively (or close to neutral) charged quartz and negatively charged smectite may hinder the smectite-smectite (edge to face) interactions, which could cause a weaker yield stress as shown in Figure 3a, but this was not found by Benna et al. (1999) due to the lack of quartz particles.

Coussot and Piau (1994) investigated the effect of temperature on the rheology of fine clay suspensions but found no apparent influence within a range of 0–20°C, except for one clay sample that exhibited a slightly higher value of yield stress at 0°C due to the formation of small ice crystals. In our experiment, there was no apparent change in the rheological parameters within the temperature range of 2–30°C, which is consistent with the results of Coussot and Piau (1994). At higher temperatures, of up to 80°C, Vryzas et al. (2016) reported a gradual strengthening in the yield stress and reduced viscosity of sodium montmorillonite dispersions. The authors argued that thermally activated Brownian motion may cause flexures and strong associations between the smectite platelets, which may in turn cause a higher yield stress at higher temperatures.

4.2. Implications for the Mobility of Smectite-Bearing Deposits at the Seafloor

When a stress exerted on an inclined seafloor exceeds the shear strength due to processes such as earthquake-induced ground oscillation or pore pressure elevation (Hampton et al., 1996; Stigall & Dugan, 2010), shear failure can occur along a horizon beneath the slope, and materials above this horizon will start to move down the slope. If smectite occurs in such slope-forming materials, even a small stress fluctuation can potentially trigger slope failure because, as a number of experimental works have shown (e.g., Kenney, 1967; Summers & Byerlee, 1977), smectite is one of the weakest materials in terms of shear strength. Several experiments have further demonstrated that the shear strength of artificial smectite-quartz mixtures shows a progressive decrease with increasing smectite content (Brown et al., 2003; Logan & Rauenzahn, 1987; Oohashi et al., 2015; Takahashi et al., 2007). Therefore, the occurrence of smectite has been often postulated as a cause of slope destabilization (e.g., Piteau & Peckover, 1978; Calcaterra et al., 1998). However, our experimental data indicate that a higher fraction of smectite progressively increases both the yield stress and viscosity of the resulting suspension. Interestingly, this property could rather inhibit slope failure, which is consistent with the recent findings of Ikari and Kopf (2015) that large cohesion in clay-rich sediments is less likely to fail under certain conditions than quartz sediments. Furthermore, the occurrence of smectite in the suspension could significantly suppress the mobility of the sediment gravity flows. To examine the effect of this mineralogical property on runout behavior, we here carried out 1-D numerical modeling of downslope spreading using the BING program developed by Imran et al. (2001). In this program, the governing equations for mass and momentum conservation in the plug and the shear layers are solved numerically. Although the modeling approach adopted in this program significantly overestimates the flow velocity for a sediment gravity flow, due to neglecting the hydrodynamic drag effect, it precisely predicts the runout distance (Huang & Garcia, 1998). The input parameters for the calculations were determined from the measurements described above. We considered the situation where an initial deposit of 100 m length × 10 m height (with a parabolic shape) is flowing down a gently dipping (2°) slope. We compared three suspensions of 50 *w/v*% with different smectite fractions in the solid mixture (10, 25, and 40 wt %). For simplicity, we assume that the density of seawater is 1,000 kg/m^3. The density of the suspension is then calculated to be ~1,300 kg/m^3 based on the mixing ratio between smectite (specific density of 2,500 kg/m^3) and quartz (2,650 kg/m^3). The reference strain rate for each suspension is calculated by dividing the yield stress by the viscosity (Imran et al., 2001).

The parameters adopted for the modeling and the calculation results are summarized in Table 1. The suspension containing 40 wt % smectite continued to flow for ~5 min, and the eventual runout distance was

Table 1
Rheological Constants Used in the BING Modeling and Calculation Results

Run	Yield stress (Pa)	Viscosity (Pa s)	Reference strain rate (s^{-1})	Mud density (kg/m^3)	Fluid density (kg/m^3)	No. of nodes	Artificial viscosity	Runout distance (m)	Runout time (min)
K4050wv	41.48	0.12	334.55	1,300	1,000	21	1.0×10^{-3}	2,078	5.39
K25-50wv	16.76	0.036	471.62	1,300	1,000	21	1.0×10^{-3}	5,280	10.47
K10-50wv	0.23	0.0098	23.22	1,300	1,000	21	1.0×10^{-3}	>100,000	>720

2,078 m. The suspension containing 25 wt % smectite reached 5,280 m and stopped flowing after ~10 min. In the case of the suspension containing 10 wt % smectite, the runout distance exceeded 100 km and movement had not ceased at the end of the experiment at 720 min. As expected from the rheological parameters obtained in our experiments, these results demonstrate that an increase in the smectite fraction can significantly suppress the runout distance of a submarine debris flow or turbidity current.

It has been argued that the shape, size, frequency, and distance of mass transport of submarine sediment gravity flows are affected by numerous factors, such as the geologic environment, sediment type, and flow mechanics (Hampton et al., 1996; Locat & Lee, 2002). Our experimental results indicate that the smectite content in the sediments is also a fundamental factor to control runout behaviors of submarine flow events. In fact, smectite is commonly found in seafloor sediments worldwide, but its abundance varies from one locality to another (Biscaye, 1965; Griffin et al., 1968). Recent efforts of the International Ocean Drilling Program, in particular at active convergent margins, have provided more quantitative data on the absolute smectite abundance in bulk samples (e.g., Kameda et al., 2015; Underwood, 2007). These mineralogical data will help to assess not only slope stability (Ikari & Kopf, 2015; Ikari et al., 2011) but also the postfailure behavior of potential submarine sediment gravity flows.

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