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Quantifying the yield stress of bentonite muds mixed with other clays during drilling operations

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Abstract

In civil engineering, bentonite-water mixtures, commonly referred to as drilling muds, are intensively used in order to lubricate tools, consolidate walls, and help extracting cuttings during drilling operations. The efficiency of drilling muds in each of these tasks lies in their rheological properties depending mainly on the amount of clay materials. During the field works, drilling muds are mixed with the excavated soil materials (e.g., sand, clay, organic matter) that may change drastically the rheological properties of mixtures. With the aim of understanding better the rheology of field drilling muds mixed with other clays, rheological measurements on mono- and binary-clay suspensions were performed using a rotational rheometer equipped with coaxial cylinders, for which the type of clay materials (i.e., bentonite, kaolin and illite), the total clay volume fraction φ_t and the bentonite to clay volume ratio R_b were varied. The contribution of this work is twofold: (i) to highlight the major role of φ_t and R_b independently on the rheology of binary-clay suspensions and (ii) to provide phenomenological models to quantify the dependency of the yield stress on both φ_t and R_b that would be particularly useful for industrial applications.

Keywords: Binary suspension, Clay, Drilling mud, Rheology, Yield stress

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1. Introduction

In the field of civil engineering, drilling muds or slurries, usually composed of bentonite, water and additives (like polymers) are intensively used for underground works with the aim of lubricating tools, consolidating walls thanks to the hydrostatic pressure and the cake formation and helping to extract cuttings during the excavation works. The efficiency of drilling muds is mainly related to their rheological properties depending on many parameters, such as the amount of bentonite (Choo and Bai, 2015), the pH (Brenna et al., 1999; Kelessidis et al., 2007; Choo and Bai, 2015; Gamal et al., 2019), the temperature (Mohammed, 2017), and the type of additives used (Vipulanandan and Mohammed, 2014; Du et al., 2020; Abu-Jdayil et al., 2021). Among them, the amount of bentonite within the drilling muds, commonly referred to as the volume fraction, the mass fraction or the bulk density, is one of major control parameters affecting the rheological properties, especially the apparent viscosity and the yield stress (i.e., the minimum stress to flow) of suspensions.

With the aim of predicting the yield stress of mono-clay suspensions (i.e., composed of one-clay material), several phenomenological models were proposed depending on the clay volume fraction φ_t with fitting parameters allowing to take into account the intrinsic properties of clay materials used (O'Brien and Julien, 1988; Zhou et al., 1999; Flatt and Bowen, 2006; Spearman, 2017). Recently, it was shown that the model type of Zhou et al. (1999) and Spearman (2017) was the most appropriate one to predict the yield stress of non-thixotropic clay suspensions over a wide range of clay volume fractions (Bougouin et al., 2022). This model reads as

$$\hat{\tau}_y(\varphi_t) = \alpha \left(\frac{\varphi_t}{\varphi_m - \varphi_t} \right)^\beta, \quad (1)$$

where α and β are two fitting parameters, and φ_m is the maximum volume fraction at which the yield stress should diverge. Note that this latter one can be found equal to $\varphi_m = 1$ (Zhou et al., 1999) or $\varphi_m = 0.64$ (Spearman, 2017) based on the idea that the maximum volume fraction is equal to the volume fraction of clay sheets on top of the other (i.e., like a package of papers) or the random close packing of spherical, solid and monodisperse clay stacks, respectively. These two values of φ_m are questionable, but it remains a difficult challenge to investigate hyperconcentrated clay suspensions at the laboratory scale.

The rheological behaviour of bentonite suspensions strongly differs from that of suspensions made of other clay materials (e.g., kaolin, illite). Bentonite-

37 water mixtures are usually characterized by thixotropic effects (i.e., time-
38 dependent shear stress) and significantly higher shear stresses under similar
39 conditions, which is attributed to the type of clay minerals contained in-
40 side. Two different properties are usually used to classify clay minerals,
41 namely the swelling behaviour (expanding mineral or not) and the basic
42 crystallographic repeat unit (octahedral/tetrahedral sheets - noted as O and
43 T, respectively, in the following) of the layer structure. Overall, the layer
44 structure of clay minerals is splitted into TO (1:1) - 7Å (kaolinite), TOT
45 (2:1) - 10-20Å (illite, smectite) and TOT:O (2:1:1) - 14Å (chlorite), while
46 only the TOT (2:1) - 10-20Å category is subdivided upon its swelling prop-
47 erty (Chassagne, 2021). In fact, only the low mineral charge of smectites
48 allows hydrated ions to be inserted between the clay mineral layers confer-
49 ring them a swelling property, and thus a peculiar behaviour of bentonite
50 suspensions mainly composed of montmorillonite (a part of smectites) and
51 other minority minerals (e.g., quartz, mica, oxides, zeolites).

52 During the excavation operations, bentonite drilling muds are mixed
53 with the excavated soil materials (e.g., sand, clay, organic matter) that may
54 affect drastically the rheological properties of suspensions. In particular,
55 different types of clay soils may be encountered depending on the geological
56 context and the early or intermediate stages of weathering. Then, bentonite
57 drilling muds may be less efficient than as expected, requiring to antici-
58 pate such modifications with the type and the amount of excavated soil
59 materials. The addition of sand within mono-clay suspensions, and more
60 generally, that of coarse grains within yield stress fluids, has already been
61 intensively studied through laboratory experiments and theoretical mod-
62 els (Ancey and Jorrot, 2001; Mahaut et al., 2008a,b; Chateau et al., 2008;
63 Pantet et al., 2010; Ovarlez et al., 2015; Hafid et al., 2016; Pellegrino and Schippa,
64 2018; Nguyen et al., 2018; Vu et al., 2018; Bougouin et al., 2022). Overall,
65 the presence of coarse grains contributes to increase viscous dissipation in
66 the system promoting larger apparent viscosities, larger yield stresses and
67 larger elastic modulus of suspensions. From a purely mechanical point of
68 view, Chateau et al. (2008) proposed predictive models to estimate the de-
69 pendency of the rheological parameters of suspensions on the coarse volume
70 fraction based on a nonlinear homogenization approach. In the field, how-
71 ever, the contribution of coarse grains can be significantly reduced by de-
72 sanding recycled and re-used drilling muds, which is a more complex process
73 with clayey cuttings. In the case of binary-clay suspensions (i.e., composed
74 of two-clay materials), both the total of clay content and the relative pro-
75 portion of clay materials modify the rheological behaviour of suspensions
76 (Neaman and Singer, 2000; Chemedda et al., 2014; Shakeel et al., 2021a). In

77 contrast to coarse-grained clay suspensions, however, no predictive mod-
 78 els exist to quantify the relative influence of these two parameters on their
 79 rheological properties, and more particularly on their yield stress.

80 In this general context, the present study addressed the rheological be-
 81 haviour of binary-clay suspensions with the aim of helping the modelling
 82 of drilling muds mixed with other clays for the excavation operation suc-
 83 cess. More specifically, rheological measurements of bentonite-kaolin and
 84 bentonite-illite suspensions were performed using a rheometer equipped with
 85 a coaxial cylinders, for which both the total clay volume fraction and the
 86 bentonite to clay volume ratio were varied independently.

87 2. Rheometer device and clay materials

88 Rheological measurements of clay suspensions were performed using an
 89 Anton Paar RheolabQC rheometer equipped with coaxial cylinders (outer
 90 radius: $R_o = 14.5$ mm; inner radius: $R_i = 13.4$ mm; height of the inner
 91 cylinder: $h = 40.0$ mm). The rheological procedure of measurements was
 92 defined as follows. First, a pre-shearing was applied to the suspension at 100
 93 s^{-1} , for 30 seconds. Then, it was left at rest to obtain a structural reference
 94 state, for 120 seconds. Finally, the suspension was sheared by increasing
 95 and then decreasing the shear rate $\dot{\gamma}$ logarithmically between 1 s^{-1} and
 96 10^3 s^{-1} , referred to as up and down logarithmic ramps, respectively, in the
 97 following. Note that other measurement protocols drawn from Shakeel et al.
 98 (2021b), were considered without changing the following results. Moreover,
 99 some specific cautions were taken to avoid a misinterpretation of results. In
 100 order to anticipate a viscous-to-inertial transition within the suspensions,
 101 the Taylor number was estimated as

$$T_a = 2\pi N \frac{\rho_s R_i (R_o - R_i)}{\eta_s} \left(\frac{R_o - R_i}{R_o} \right)^{1/2}, \quad (2)$$

102 where N is the rotational speed (in rev.s^{-1}), R_o and R_i are the outer and
 103 inner radius of coaxial cylinders, respectively, and ρ_s and η_s are the bulk
 104 density and the apparent viscosity of suspensions, respectively. According
 105 to Matignon et al. (2014), the rheological measurements satisfying $Ta \geq 40$
 106 were disregarded because of inertial effects within the suspension. More-
 107 over, at low shear rate, any measurement that deviated surprisingly from
 108 the expected trend (e.g., abrupt change in slope, decrease of the shear stress
 109 with increasing shear rate) was also ignored attributed to experimental arte-
 110 facts, such as wall slip (Meeker et al., 2004; Ballesta et al., 2012) and shear

111 banding (Ovarlez et al., 2009; Kaci et al., 2009; Schall and Van Hecke, 2010;
112 Ovarlez et al., 2013).

113 Suspensions were made of tap water mixed with one or two types of
114 clay materials, referred to as mono- and binary-clay suspensions, respec-
115 tively, in the following. Tap water was characterized by a pH of 7.5 ± 0.1 ,
116 an electrical conductivity of $0.58 \pm 0.05 \text{ mS.cm}^{-1}$, an hardness of 24 ± 2
117 mg/l as CaCO_3 and an alcalinity of 20 ± 2 mg/l as CaCO_3 . The types of
118 clay materials used were bentonite (bentonite 1 - 35% calcite, 33% mont-
119 morillonite, 32% quartz; bentonite 2 - montmorillonite, interstratified illite-
120 montmorillonite, kaolinite, halloysite; bentonite 3 - montmorillonite, inter-
121 stratified illite-montmorillonite, kaolinite, halloysite), kaolin (kaolin 1 - 67%
122 kaolinite, 17% quartz, 16% illite; kaolin 2 - 56% kaolinite, 38% biotite, 6%
123 quartz) and illite (47% illite, 47% calcite, 6% quartz), corresponding to min-
124 eral mixtures with a high content of montmorillonite, kaolinite and illite,
125 respectively. The term ‘illite’ is commonly used to define both mineral and
126 clay material, and it would be referred only to the clay material in the follow-
127 ing. Recall that X-ray diffraction (XRD) provides only a semi-quantification
128 of the mineralogical composition of clay materials, and these measurements
129 allowed only to identify major minerals within the clay materials used. On
130 the other hand, bentonites 2 and 3 corresponded to industrial samples and
131 they had probably undergone specific treatments. For this reason, the de-
132 termination of the crystallographic structure was much more complex with
133 a standard analysis from X-ray diffraction, and the mass proportion of min-
134 erals was therefore not provided here. The particle size of each clay material
135 was measured with a Malvern Mastersizer 2000 analyser, and it was found as
136 $d_b = 34_{-30}^{+125} \mu\text{m}$ (bentonite 1), $d_b = 28_{-16}^{+75} \mu\text{m}$ (bentonite 2), $d_b = 52_{-28}^{+96} \mu\text{m}$
137 (bentonite 3), $d_k = 7_{-5}^{+16} \mu\text{m}$ (kaolin 1), $d_k = 11_{-9}^{+208} \mu\text{m}$ (kaolin 2), and
138 $d_i = 4_{-3}^{+10} \mu\text{m}$ (illite), where the given values corresponds to the size of clay
139 stacks for which 10%, 50%, and 90% of the total volume of particles is be-
140 low. The particle densities of bentonite, kaolinite and illite were estimated
141 as $\rho_b = 2650 \text{ kg.m}^{-3}$, $\rho_k = 2550 \text{ kg.m}^{-3}$ and $\rho_i = 2500 \text{ kg.m}^{-3}$, respectively,
142 from direct measurements (Bougouin et al., 2022) and values given in the
143 literature (Coussot, 1997).

144 The preparation of suspensions required several specific stages. First,
145 each type of clay materials was weighed and mixed manually with water.
146 Then, the suspensions were left at rest at least one day before the rheological
147 measurements to ensure a proper hydration of clay materials. In presence of
148 bentonite materials, a second step of mixing was carried out with a propeller
149 for 5 minutes after a dozen hours of hydratation to eliminate sustainable

150 aggregates. Finally, the suspensions were remixed manually for 10 minutes
 151 before to set up within the cup of the rheometer. Note that the pH of
 152 suspensions was found to be roughly constant in the range 9 – 10. To
 153 characterize the suspensions, both the total clay volume fraction φ_t and the
 154 bentonite to clay volume ratio R_b were defined as

$$\varphi_t = \frac{(\rho_{k/i}m_b + \rho_b m_{k/i})\rho_w}{\rho_{k/i}\rho_w m_b + \rho_b \rho_w m_{k/i} + \rho_b \rho_{k/i} m_w}, \quad (3)$$

155

$$R_b = \frac{\rho_{k/i}m_b}{\rho_{k/i}m_b + \rho_b m_{k/i}}, \quad (4)$$

156 where m_b , $m_{k/i}$ and m_w are the masses of bentonite, kaolin or illite and
 157 water, respectively, and $\rho_w = 1000 \text{ kg.m}^{-3}$ is the water density. In the
 158 present study, both the total volume fraction φ_t and the bentonite to clay
 159 volume ratio R_b were varied over a wide range summarized in Table 1.

Table 1: Summary table of mono- and binary-clay suspensions with the type of clay materials used, the total clay volume fraction φ_t (Eq. 3) and the bentonite to clay volume ratio R_b (Eq. 4).

		Type of clay materials	Total clay volume fraction φ_t [-]	Bentonite to clay volume ratio R_b [-]
Mono-clay	suspensions	Bentonite 1	[0.03 : 0.097]	1
		Bentonite 2	[0.015 : 0.099]	1
		Bentonite 3	[0.016 : 0.08]	1
		Kaolin 1	[0.052 : 0.301]	0
		Kaolin 2	[0.1 : 0.301]	0
		Illite	[0.1 : 0.302]	0
			0.06	[0.3 : 0.9]
Binary-clay	suspensions	Bentonite 1	[0.05 : 0.15]	0.5
		Kaolin 1	[0.091 : 0.292]	[0.13 : 0.61]
		Bentonite 1	[0.061 : 0.15]	[0.31 : 0.9]
		Kaolin 2	[0.06 : 0.248]	[0.19 : 0.8]
		Bentonite 2	[0.066 : 0.17]	[0.15 : 0.74]
		Kaolin 1	[0.063 : 0.159]	[0.1 : 0.95]
		Bentonite 3		
		Kaolin 1		

160 **3. Rheological results**

161 *3.1. Mono-clay suspensions: bentonite vs. kaolin/illite*

162 The rheological behaviour of mono-clay suspensions, i.e. composed of
163 one-clay material, was first addressed to quantify the influence of both the
164 type and the total volume fraction φ_t . Hence, the bentonite to clay volume
165 ratio R_b was reduced to 0 and 1 for kaolin or illite and bentonite suspensions,
166 respectively (see Table 1).

167 Figure 1 shows the shear stress τ (inset : the apparent viscosity η_s) as
168 a function of the shear rate $\dot{\gamma}$ for (a) bentonite suspensions and (b) kaolin
169 suspensions with increasing the total clay volume fraction φ_t . Regardless
170 of the type and the volume fraction of clay materials, the rheology of sus-
171 pensions was characterized by a yielding and shear-thinning behaviour suc-
172 cessfully described by the Herschel-Bulkley model (lines), as expected for
173 mono-clay suspensions (Coussot and Piau, 1994). Quantitatively, the rheo-
174 logical properties of suspensions were strongly dependent on both the type
175 and the volume fraction φ_t of clay materials. In particular, at lower φ_t and
176 at a given $\dot{\gamma}$, bentonite suspensions exhibited much larger values of shear
177 stresses τ and apparent viscosities η_s compared to kaolin suspensions. The
178 same conclusions were also drawn between bentonite and illite suspensions
179 (not shown here). This was attributed to the higher level of cohesiveness
180 of the montmorillonite-containing bentonite materials in comparison with
181 kaolin and illite. Moreover, the increase of the total clay volume fraction φ_t
182 also promoted larger shear stresses τ and apparent viscosities η_s , for a given
183 type of clay materials. The addition of clay materials within the suspension
184 reduced the mean distance between clay stacks strengthening cohesion and
185 increasing viscous dissipation by decreasing the size of interstices, in which
186 water was sheared. Finally, one observed that the flow curves of bentonite
187 suspensions mismatched between up (upward triangles) and down (down-
188 ward triangles) logarithmic ramps, which was hardly reported for kaolin and
189 illite suspensions. Such an observation of lower down flow curves than up
190 ones was the sign of thixotropic effects attributed to a longer timescale to
191 reach the microstructural steady-state compared to the time of measure-
192 ments (Paumier, 2007). Although thixotropy is an important property of
193 bentonite suspensions, the associated variations on the yield stress studied
194 in the following, were much lower than those due to the type and the wide
195 range of volumes fractions of clay materials considered (see Supplementary
196 Material, available online). Thus, thixotropic effects of suspensions contain-
197 ing bentonite were disregarded in the following. In the same way, it could
198 be mentioned that, for kaolin and illite suspensions, up flow curves were

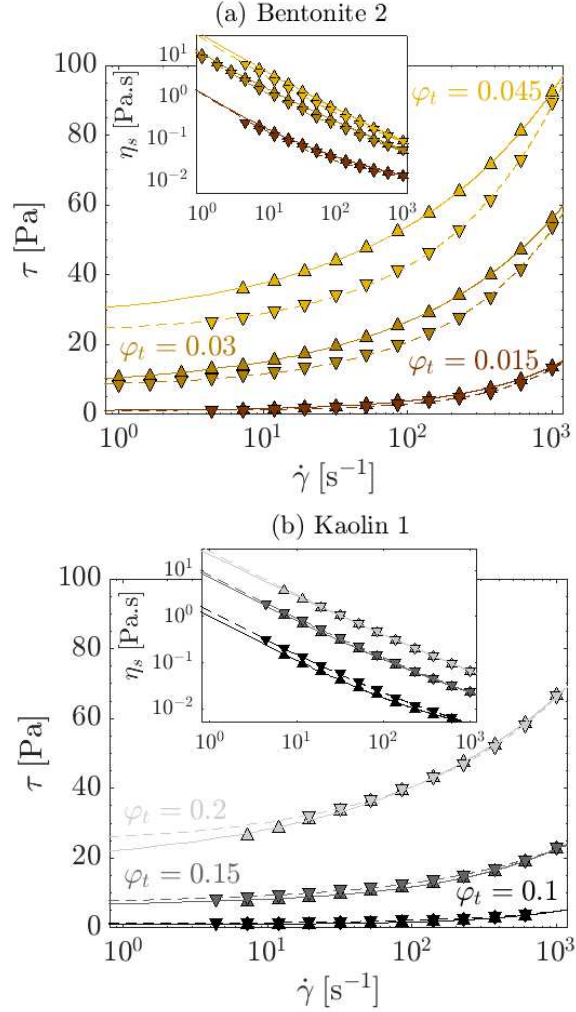


Figure 1: Shear stress τ (inset: apparent viscosity η_s) as a function of the shear rate $\dot{\gamma}$ during up (upward triangles and solid lines) and down (downward triangles and dashed lines) logarithmic ramps, for mono-clay suspensions with various φ_t . Lines correspond to the best fit of the Herschel-Bulkley model $\tau = \tau_y + K\dot{\gamma}^n$ (inset: $\eta_s = \tau_y/\dot{\gamma} + K\dot{\gamma}^{n-1}$), with the yield stress τ_y , the consistency K and the flow index n .

199 slightly lower than down flow curves probably due to a modification of the
 200 internal structure of suspensions. In any case, the variations between up
 201 and down flow curves were considered by adding error bars in the following
 202 when larger than the size of symbols.

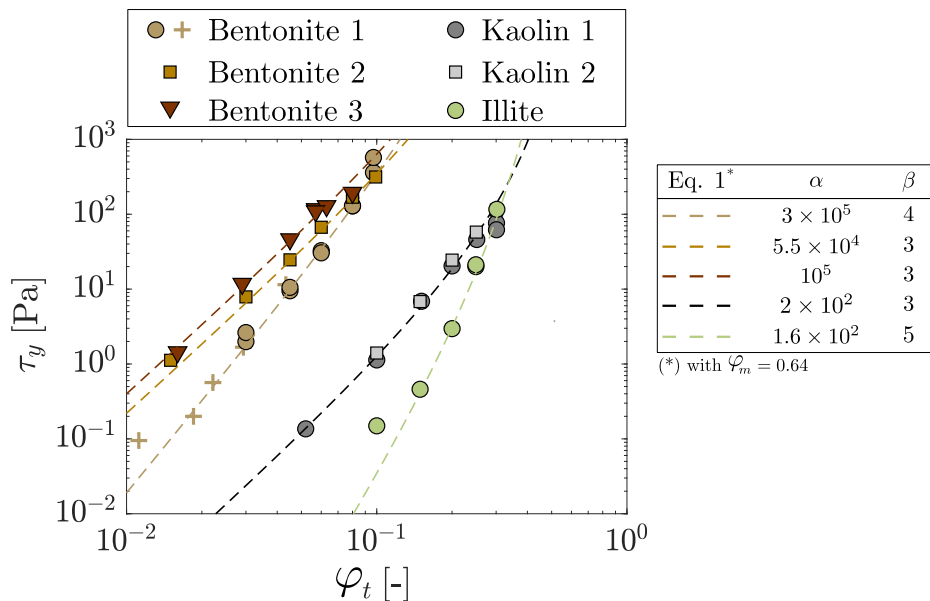


Figure 2: Yield stress τ_y of mono-clay suspensions as a function of the total clay volume fraction φ_t . (---) Spearman’s model (Eq. 1, with $\varphi_m = 0.64$). Crosses correspond to additional experiments of suspensions made of bentonite 1 using other procedures of preparation and measurement than defined in Sec. 2.

203 The yield stress τ_y is one of the main physical properties of clay suspen-
 204 sions and it could be easily extracted from the best fit of the Herschel-Bulkley
 205 model. In Fig. 2, the yield stress τ_y of mono-clay suspensions was shown as
 206 a function of the total clay volume fraction φ_t , for each type of clay mate-
 207 rials used. Firstly, one clearly observed that the yield stress τ_y of bentonite
 208 suspensions was much larger than that of kaolin and illite suspensions, at a
 209 given φ_t . As discussed above, it was attributed to the high level of cohesive-
 210 ness of montmorillonite-containing bentonite materials in comparison with
 211 the other types of clay materials, i.e. kaolin and illite. Moreover, the yield
 212 stress τ_y was strongly dependent on the total clay volume fraction φ_t , which
 213 was well predicted by the Spearman’s model (Eq. 1, with $\varphi_m = 0.64$) as
 214 shown by the dashed lines. The relevance of this model quantifying the φ_t -
 215 dependency of the yield stress has already been reported for kaolin and illite
 216 suspensions over a wide range of volume fractions (Bougouin et al., 2022),
 217 while it was shown here to be also appropriate in the case of bentonite sus-
 218 pensions. The two fitting parameters (α, β) were dependent on the type of

219 clay materials used, and thus directly related to their intrinsic properties.
 220 Here, it was found that the prefactor α was in the order of magnitude of 10^2
 221 and $10^4 - 10^5$ for kaolin/illite and bentonite materials, respectively, while the
 222 exponent β was included in the range [3 : 5]. Although different batches of
 223 bentonite were used, the specific behaviour of the major clay mineral (i.e.,
 224 montmorillonite) contained inside allowed to distinguish clearly bentonite
 225 suspensions from others. Nevertheless, the variability of the couple (α, β)
 226 in the case of bentonite suspensions suggested that other parameters, such
 227 as the presence of other minority minerals and additives, powder quality
 228 or activation process, should also be considered to predict totally the yield
 229 stress of suspensions.

230 3.2. Binary-clay suspensions: bentonite-kaolin and bentonite-illite

231 The rheological behaviour of binary-clay suspensions made of bentonite
 232 and another type of clay materials, namely kaolin or illite, was then ad-
 233 dressed by varying both the total clay volume fraction φ_t and the bentonite
 234 to clay volume ratio R_b independently.

235 In this way, Fig. 3 shows the shear stress τ (inset: the apparent viscos-
 236 ity η_s) as a function of the shear rate $\dot{\gamma}$ during up and down logarithmic
 237 ramps, at (a) constant $R_b = 0.5$ and φ_t varied and (b) constant $\varphi_t = 0.06$
 238 and R_b varied. As expected, the increase of the total clay volume fraction
 239 φ_t promoted larger shear stresses τ and larger apparent viscosities η_s , as
 240 already discussed for mono-clay suspensions. Additionally, at constant φ_t ,
 241 the bentonite to clay volume ratio R_b also affected significantly the rheo-
 242 logical behaviour of suspensions due to different properties of clay minerals
 243 contained in bentonite and kaolin/illite. Thus, both parameters should be
 244 considered to fully describe the rheological properties, and in particular the
 245 yield stress, of binary-clay suspensions.

246 Beyond this idea, a phenomenological model taking into account the de-
 247 pendency of the yield stress τ_y of binary-clay suspensions on both the total
 248 clay volume fraction φ_t and the bentonite to clay volume ratio R_b was de-
 249 veloped. First, it was argued that the yield stress of binary-clay suspensions
 250 was bounded by the upper yield stress $\tau_y(\varphi_t, R_b = 1) = \tau_y^b$ of mono-clay sus-
 251 pensions made of bentonite and the lower yield stress $\tau_y(\varphi_t, R_b = 0) = \tau_y^{k/i}$
 252 of mono-clay suspensions made of kaolin or illite (more details given in
 253 Supplementary Material, available online). Moreover, both quantities τ_y^b
 254 and $\tau_y^{k/i}$ could be well predicted by the Spearman's model (Eq. 1, with
 255 $\varphi_m = 0.64$) as shown in Sec. 3.1, referred to as $\hat{\tau}_y^b$ and $\hat{\tau}_y^{k/i}$ for bentonite
 256 and kaolin/illite suspensions, respectively, in the following. Finally, the

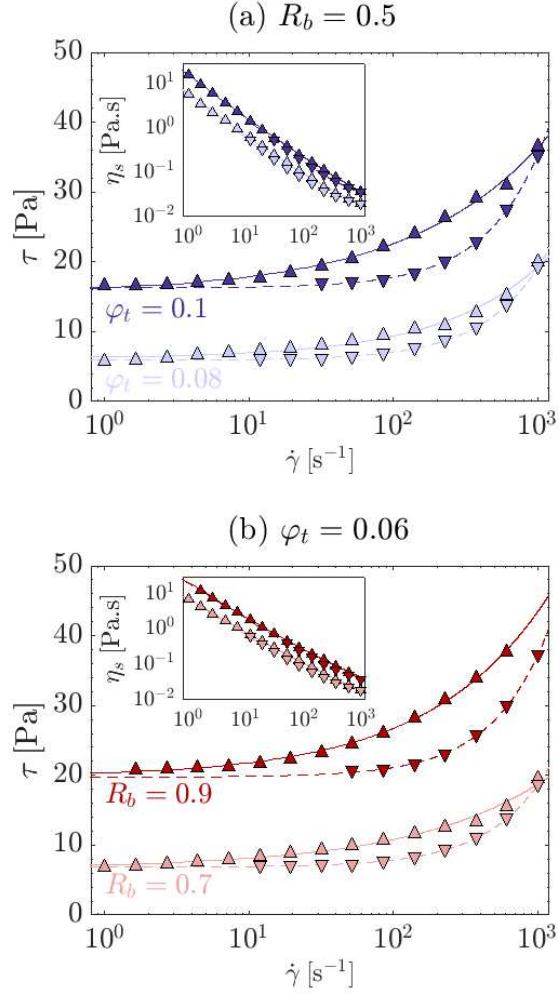


Figure 3: Shear stress τ (inset: apparent viscosity η_s) as a function of the shear rate $\dot{\gamma}$ during up (upward triangles and solid lines) and down (downward triangles and dashed lines) logarithmic ramps, for binary-clay (bentonite 1 - kaolin 1) suspensions at (a) constant $R_b = 0.5$ and $\varphi_t = [0.08, 0.1]$ and (b) constant $\varphi_t = 0.06$ and $R_b = [0.7, 0.9]$. Lines correspond to the best fits of the Herschel-Bulkley model $\tau = \tau_y + K\dot{\gamma}^n$ (inset : $\eta = \tau_y/\dot{\gamma} + K\dot{\gamma}^{n-1}$), with the yield stress τ_y , the consistency K , and the flow index n .

257 trend of the yield stress τ_y with R_b could be addressed through the quan-
 258 tity $(\tau_y - \hat{\tau}_y^{k/i})/(\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ bounded between 0 and 1 when $R_b = 0$ and 1,
 259 respectively.

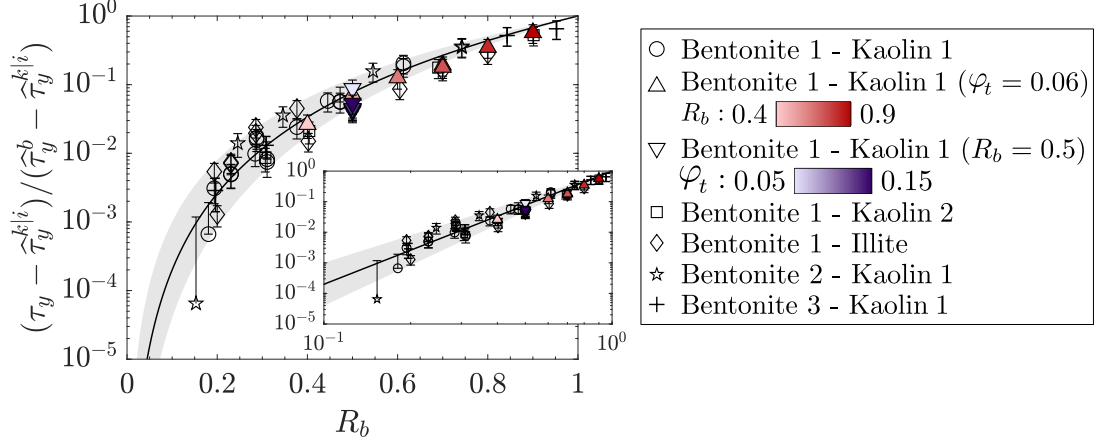


Figure 4: Quantity $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ as a function of the bentonite to clay volume ratio R_b for the whole set of binary-clay suspensions, where $\hat{\tau}_y^{k/i}$ and $\hat{\tau}_y^b$ are the yield stresses of mono-clay suspensions made of kaolin/illite and bentonite, respectively, estimated by the Spearman's model (Eq. 1, with $\varphi_m = 0.64$) as fitted in Fig. 2. The solid line and gray area correspond to $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i}) = R_b^\lambda$ with $\lambda = 3.7 \pm 0.7$.

260 Figure 4 shows the quantity $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ as a function of the
 261 bentonite to clay volume ratio R_b for the whole set of binary-clay suspen-
 262 sions considered in this study. Overall, the quantity $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$
 263 increased with increasing the values of R_b until reaching a value of 1, as ex-
 264 pected. Moreover, the trend of $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ with R_b was broadly in-
 265 dependent on the type and the total volume φ_t of clay materials. To confirm
 266 such an observation, downward and upward triangles correspond to the trend
 267 of $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ at constant $R_b = 0.5$ and constant $\varphi_t = 0.06$ with
 268 increasing φ_t and R_b , respectively. This means that, at least at first order,
 269 the yield stress of binary-clay suspensions was mainly controlled by the type
 270 and the amount of clay materials contained inside, without physicochemical
 271 interactions between different minerals. More quantitatively, one obtained
 272 a master curve given by $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i}) = R_b^\lambda$ with $\lambda = 3.7 \pm 0.7$
 273 (solid line and gray area). To conclude on, the yield stress of binary-clay
 274 suspensions may thus be quantified by the relation

$$\tau_y(\varphi_t, R_b) = R_b^\lambda \hat{\tau}_y^b(\varphi_t) + (1 - R_b^\lambda) \hat{\tau}_y^{k/i}(\varphi_t), \quad (5)$$

275 where $\hat{\tau}_y^b$ and $\hat{\tau}_y^{k/i}$ are the yield stresses of mono-clay suspensions made
 276 of bentonite and kaolin/illite, respectively, estimated by the Spearman's

277 model (Eq. 1, with $\varphi_m = 0.64$), and $\lambda = 3.7 \pm 0.7$ is an empirical constant
 278 parameter. Note that, for $R_b < 0.2$, the accuracy of measurements could
 279 lead to negative values of $(\tau_y - \widehat{\tau}_y^{k/i})/(\widehat{\tau}_y^b - \widehat{\tau}_y^{k/i})$, i.e. $\tau_y < \widehat{\tau}_y^{k/i}$, and they
 280 were therefore disregarded.

281 To go further on, such an approach could be envisaged for predicting the
 282 yield stress of mono-clay suspensions by considering the rheology of mono-
 283 clay mineral suspensions, i.e. the rheology of suspensions made of each clay
 284 mineral contained in the clay material. However, some caution should be
 285 exercised because it will require to determine the rheological behaviour of
 286 each component contained in the clay material, while the present model was
 287 only established for two-component suspensions.

288 3.3. Towards the industrial applications

289 In the industrial field, the bulk density of drilling muds is usually pre-
 290 ferred to the total clay volume fraction φ_t because of its ease of estimate
 291 in the fieldwork activities. Less rigorously, the previous models could then
 292 be expressed in this sense. In particular, both the total clay volume frac-
 293 tion φ_t and the bentonite to clay volume ratio R_b were substituted for the
 294 bulk density ρ_s and the density ratio $(\rho_s^* - \rho_w)/(\rho_s - \rho_w)$, where ρ_s^* is the
 295 bulk density of bentonite-water mixtures (i.e., without kaolin and illite), ρ_s
 296 is the bulk density of binary-clay suspensions, and ρ_w is the water density.
 297 Note that both ρ_s and $(\rho_s^* - \rho_w)/(\rho_s - \rho_w)$ were proportional to φ_t and R_b ,
 298 respectively, because of the low variation in density of natural clay materi-
 299 als, i.e. $\rho_c \sim 2650 \pm 150 \text{ kg.m}^{-3}$, and the low variation of the volume ratio
 300 $0.4 \lesssim (V_b + V_w)/(V_b + V_w + V_{k/i}) \leq 1$, with $V_b, V_w, V_{k/i}$ the volumes of ben-
 301 tonite, water and kaolin/illite, respectively. To finish with, both parameters
 302 ρ_s^* and ρ_s could be interpreted as the bulk densities of drilling muds before
 303 and after they were mixed with extruded clays.

304 Figure 5(a) shows first the increase of the yield stress τ_y with the bulk
 305 density ρ_s of mono-clay suspensions, as expected. The dependency of τ_y
 306 with ρ_s could be fairly well described by the ρ_s -dependent Spearman's model
 307 defined as

$$\widehat{\tau}_y(\rho_s) = \alpha \left(\frac{\rho_s - \rho_w}{\rho_m - \rho_s} \right)^\beta, \quad (6)$$

308 where α and β are two fitting parameters, and $\rho_m = \varphi_m \rho_c + (1 - \varphi_m) \rho_w =$
 309 2056 kg.m^{-3} (with $\varphi_m = 0.64$ and $\rho_c = 2650 \text{ kg.m}^{-3}$) is the maximum bulk
 310 density. Note that the clay density ρ_c was kept constant here to maintain
 311 only two fittings parameters in Eq. 6, i.e. the couple (α, β) , for the simplicity
 312 of industrial applications. This assumption led however to a slight increase

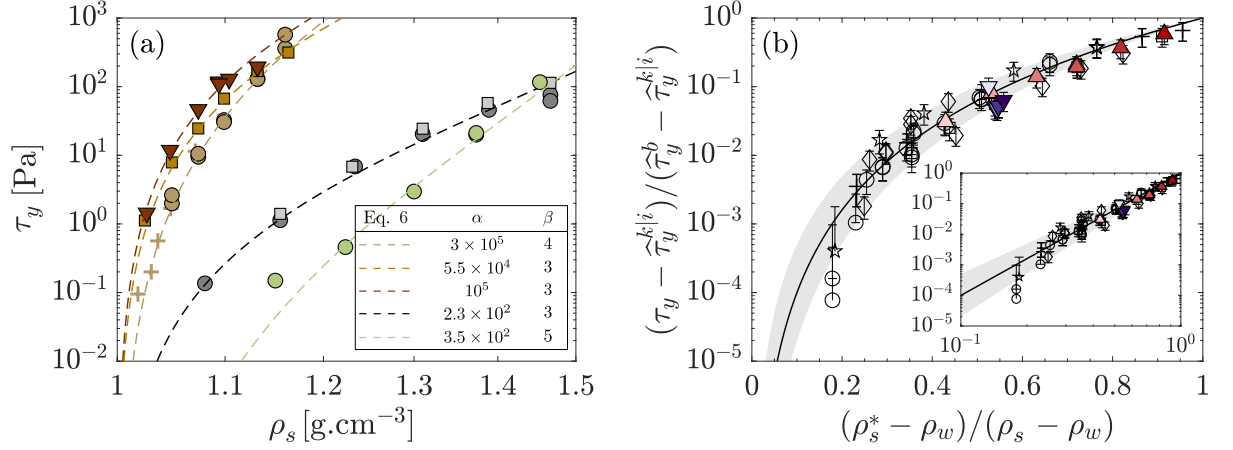


Figure 5: (a) Yield stress τ_y as a function of the bulk density ρ_s for the same set of mono-clay suspensions shown in Fig. 2. (---) ρ_s -dependent Spearman's model (Eq. 6). (b) Quantity $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$ as a function of the density ratio $(\rho_s^* - \rho_w) / (\rho_s - \rho_w)$ for the same set of binary-clay suspensions shown in Fig. 4. Here, ρ_s^* and ρ_s are the bulk densities of bentonite-water mixtures (i.e., without kaolin/illite) and binary-clay suspensions, respectively, ρ_w is the water density, and $\hat{\tau}_y^{k/i}$ and $\hat{\tau}_y^b$ are the yield stresses of mono-clay suspensions made of kaolin/illite and bentonite estimated by the ρ_s -dependent Spearman's model (Eq. 6), as fitted in Fig. 5(a). The solid line and gray area correspond to $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i}) = [(\rho_s^* - \rho_w) / (\rho_s - \rho_w)]^\lambda$ with $\lambda = 4.0 \pm 0.7$.

313 in values of α compared to those estimated in Fig. 2, for kaolin and illite
 314 suspensions.

315 Then, Fig. 5(b) shows the dependency of the quantity $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i})$
 316 $\hat{\tau}_y^{k/i}$) on the density ratio $(\rho_s^* - \rho_w) / (\rho_s - \rho_w)$ for the same set of binary-
 317 clay suspensions shown in Fig. 4. Here, the yield stresses $\hat{\tau}_y^{k/i}$ and $\hat{\tau}_y^b$ of
 318 mono-clay suspensions made of kaolin/illite and bentonite, respectively, were
 319 estimated by the ρ_s -dependent Spearman's model (Eq. 6). As previously
 320 concluded, data collapsed on a master curve independently on the type and
 321 the total volume fraction φ_t of clay materials. More specifically, it was found
 322 $(\tau_y - \hat{\tau}_y^{k/i}) / (\hat{\tau}_y^b - \hat{\tau}_y^{k/i}) = [(\rho_s^* - \rho_w) / (\rho_s - \rho_w)]^\lambda$ with $\lambda = 4.0 \pm 0.7$ (solid line
 323 and gray area) consistent with the value reported in the previous section.
 324 Thus, the yield stress of binary-clay suspensions could also be predicted less
 325 rigorously from the bulk density as

$$\tau_y(\rho_s, \rho_s^*) = \left(\frac{\rho_s^* - \rho_w}{\rho_s - \rho_w} \right)^\lambda \hat{\tau}_y^b(\rho_s) + \left[1 - \left(\frac{\rho_s^* - \rho_w}{\rho_s - \rho_w} \right)^\lambda \right] \hat{\tau}_y^{k/i}(\rho_s), \quad (7)$$

326 where ρ_s^* and ρ_s are the bulk densities of bentonite-water mixtures (i.e.,
327 without kaolin/illite) and binary-clay suspensions, respectively, $\rho_w = 1000$
328 kg.m^{-3} is the water density, $\hat{\tau}_y^{k/i}$ and $\hat{\tau}_y^b$ are the yield stresses of mono-clay
329 suspensions made of kaolinite/illite and bentonite, respectively, estimated
330 by the ρ_s -dependent Spearman's model (Eq. 6), and $\lambda = 4.0 \pm 0.7$ is an
331 empirical constant parameter.

332 Conclusion

333 In civil engineering, bentonite drilling muds are commonly used with
334 the aim of lubricating tools, consolidating walls, and helping extracting
335 cubbings for the drilling operation success. During the excavation works,
336 bentonite drilling muds are progressively mixed with the excavated soil ma-
337 terials (e.g., sand, clay, organic matter) that may change drastically their
338 rheological properties, and thus their efficiency in the achievement of their
339 tasks. However, there is still a lack in predicting the modification of the rhe-
340 ology of drilling muds when they are mixed with other clays during drilling
341 operations. In this objective, the rheological behaviour of mono- and binary-
342 clay suspensions made of various types of clays materials (i.e., bentonite,
343 kaolin, illite), was investigated using a rotational rheometer equipped with
344 coaxial cylinders. Overall, the rheology of suspensions was characterized
345 by a shear-thinning and yielding behaviour successfully described by the
346 Herschel-Bulkley model $\tau = \tau_y + K\dot{\gamma}^n$, from which the yield stress τ_y , the
347 consistency K and the flow index n could be deduced. More quantitatively,
348 the rheological properties of suspensions were mainly controlled by the type
349 of clay materials, the total clay volume fraction φ_t and the bentonite to clay
350 volume ratio R_b . From the rheological measurements on mono-clay suspen-
351 sions, it was first shown that the dependency of the yield stress τ_y on a wide
352 range of the total clay volume fraction φ_t was fairly well described by the
353 Spearman's model, for which the two fitting parameters were only depen-
354 dent on the type of clay materials used. Then, the influence of both the total
355 clay volume fraction φ_t and the bentonite to clay volume ratio R_b on the
356 rheology of binary-clay suspensions was investigated independently. This
357 approach allowed to propose a phenomenological model taking into account
358 both φ_t and R_b for predicting the yield stress τ_y of binary-clay suspensions
359 as $\tau_y(\varphi_t, R_b) = R_b^\lambda \hat{\tau}_y^b(\varphi_t) + (1 - R_b^\lambda) \hat{\tau}_y^{k/i}(\varphi_t)$, where $\hat{\tau}_y^b$ and $\hat{\tau}_y^{k/i}$
360 were the yield stresses of mono-clay suspensions made of bentonite and kaolin/illite pre-
361 viously estimated by the Spearman's model, respectively, and $\lambda = 3.7 \pm 0.7$
362 was an empirical constant parameter. Finally, the different predictive mod-
363 els were then expressed in light of the bulk density of suspensions usually

364 used in the industrial applications. These predictive models would be par-
365 ticularly useful for quantifying the yield stress of bentonite drilling muds
366 mixed with other clays for the excavation operation success.

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