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Effect of coarse particle volume fraction on the yield stress and thixotropy of cementitious materials

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Abstract

In order to help modelling the yield stress of fresh concrete, we study the behavior of suspensions of coarse particles in a thixotropic cement paste. Our aim is to relate the yield stress of these mixtures to the yield stress of the suspending cement paste, to the time passed at rest, and to the coarse particle volume fraction. We present here procedures that allow for (i) studying an homogeneous and isotropic suspension, (ii) comparing the yield stress of a given cement paste to that of the same cement paste added with particles, (iii) accounting for the thixotropy of the cement paste. We observe that the yield stress of these suspensions of cement paste with coarse particles follows the very simple Chateau-Ovarlez-Trung model [1], consistently with the experimental results of Mahaut *et al.* [2] obtained with many different particles and suspending yield stress fluids. This consistency between the results obtained in various yield stress fluids shows that the yield stress of the suspension does not depend on the physicochemical properties of the suspending yield stress fluid; it only

depends on its yield stress value. This shows that studies of suspensions in model yield stress fluids can be used as a general tool to infer the behavior of fresh concrete. Moreover, we show that the thixotropic structuration rate of the interstitial paste (its static yield stress increase rate in time) is not affected by the presence of the particles. As a consequence, it is sufficient to measure the thixotropic properties of the constitutive cement paste in order to predict the thixotropic structuration rate of a given fresh concrete. This structuration rate is predicted to have the same dependence on the coarse particle volume fraction as the yield stress.

Key words: A. Fresh Concrete, A. Rheology, D. Aggregate, D. Cement Paste, E. Modeling

1 Introduction

Knowing and predicting the flow properties of fresh concrete is a major issue of concrete casting and concrete mix-design. Basically, fresh concretes exhibit a yield stress [3] and have a solid viscoelastic behavior below this yield stress [4]; above the yield stress they behave as liquids, and their steady flow behavior is usually well represented by a Bingham or a Herschel-Bulkley model [3,5]. However, fresh concrete is also known for its evolving rheological behavior. Even, if its steady state flow may be described by the above models, the characteristic time to reach this steady state flow may be rather long [6–9] and, after a long time of rest, the stress that has to be applied to induce a flow may be one or two orders higher than the dynamic yield stress measured when the

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12 material stops flowing i.e. it is thixotropic [10–12]. The static yield stress and
13 its increase rate at rest are actually the most important rheological quantities
14 in terms of potential applications in the case of SCC [13]: it has been shown
15 recently that they determine the formwork pressure [9,14–17], the stability
16 vs. sedimentation of the coarsest particles in SCC [18] and the occurrence
17 of distinct layer casting [19]. As a consequence it is of high importance to
18 understand the role of the various components of a given concrete on this yield
19 stress and its evolution at rest. Moreover, measuring directly the rheological
20 properties of fresh concrete is very difficult [20]; any model providing the yield
21 stress of concrete as a function of the suspending cement paste properties and
22 the properties and the volume fraction of sand and aggregates would then
23 prove to be very useful.

24 The link between concrete mix-design and its flow properties in the fresh state
25 may be studied in the more general framework of suspensions rheophysics
26 [12]. Actually, fresh concretes belong to the wide family of dense suspensions,
27 which often involve a broad range of particle sizes [21] and can be found in
28 many industrial processes (drilling muds, foodstuff transport...) and natural
29 phenomena (debris-flows, lava flows...). All these materials share the same
30 complex features, which originate from the great variety of interactions be-
31 tween the particles (colloidal, hydrodynamic, frictional, collisional...) and of
32 physical properties of the particles (volume fraction, sensitivity to thermal
33 agitation, shape...) affecting the material behavior [22,12]. Basically, in the
34 absence of a contact network of noncolloidal particles (i.e. for moderate non-
35 colloidal particles volume fraction), the yielding behavior originates from the
36 colloidal interactions which create a jammed network of interacting particles
37 [5,12]. Structuration at rest (which has nothing to do with setting) is observed

38 in many aggregating suspensions and colloidal glasses [12]: the evolution of the
 39 behavior of aggregating suspensions at rest may be explained by a progres-
 40 sive and reversible formation of a solid structure by flocculation. Within this
 41 frame, the problem of the influence of coarse particles on the behavior of fresh
 42 concrete may be seen more generally as the problem of the influence of non-
 43 colloidal particles on the properties of yield stress fluids. It is thus of high
 44 importance to clarify the cases where the rheological properties of a suspen-
 45 sion of coarse particles in a yield stress fluid depend only on the rheological
 46 properties of the suspending fluid and on the coarse particle volume fraction
 47 and size distribution. This should provide results applicable to any particles
 48 in any yield stress fluid, in particular to sand and aggregates suspended in a
 49 cement paste. It would allow the use of results obtained in studies performed
 50 e.g. with noncolloidal particles in clay dispersions to predict the behavior of a
 51 mortar or a concrete. On the other hand, any departure from generic results
 52 would be the result of specific physicochemical interactions in the suspensions
 53 (or specific slippage at the paste/particle interface), as e.g. the adsorption of
 54 a fraction of the superplasticizer of the cement paste on the fine aggregates
 55 in SCC [23], and would justify for each material a specific study with the
 56 particular particles and particular paste involved. In this paper, we test the
 57 idea of fresh concrete being a suspension of particles in a yield stress fluid. We
 58 compare the results obtained when suspending particles in a cement paste to
 59 those recently obtained in a broad range of materials (suspensions of various
 60 particles in various yield stress fluids) by Mahaut *et al.* [2].

61 The influence of the aggregates on the rheological properties of fresh concrete
 62 has been studied theoretically and experimentally by de Larrard [24], de Lar-
 63 rard and Sedran [25], Geiker *et al.* [26], Erdogan [27] and Toutou and Roussel

[28]. De Larrard [24] has proposed a model in which concrete is looked as a granular mix in a water suspension. Then, the overall yield stress is the macroscopic counterpart of the friction between solid particles and is interpreted as the stress one needs to apply in order to overcome the intergranular contact forces. The overall yield stress can be estimated from the value of the solid volume fraction and close packing density of the different components of the granular mixture. However, if this model may help understanding the properties of fresh concrete displaying “ordinary” rheology, it is unadapted to the description of modern fluid concrete which contains less coarse particles and where friction between the grains is negligible [29]. Geiker *et al.* [26] have studied experimentally the effect of coarse particle volume fraction on the rheological properties of SCC. They have measured the steady-state flow curves of various materials thanks to the procedure developed in [8]; the dynamic yield stress was then extracted from a fit of the flow curve with a Bingham model. It was found to increase strongly with the coarse particle volume fraction. To model the behavior, they assume that the effect of aggregates on concrete rheological properties can be studied by looking to concrete as a suspension of coarse particles in the mortar seen as a continuum medium. Their experimental data are compared to a model proposed by Nielsen [30] which provides the yield stress of a suspension of ellipsoids as a function of the volume fraction of particles and of the aspect ratio. This model rests on heuristic rules which are not rigorously justified. Nevertheless, the theory can be calibrated in order to accurately describe the data of Geiker *et al.* [26]. Erdogan [27] have studied the effect of aggregate particle shape and surface texture on rheological properties of fresh concrete. Artificial aggregate particles of regular geometric shapes (spheres, cubes and rectangular prisms) with similar centimeter size and volume were prepared. A Couette-vane rheometer (ICAR) was used to

91 measure the dynamic yield stress as the low shear rate limit of a flow curve.
 92 In addition, slump tests were performed. Erdogan has observed that the yield
 93 stress increases slightly when the coarse particle volume fraction increases.
 94 This trend is confirmed by slump experiments: the slump value was clearly a
 95 decreasing function of the coarse particle content, whatever the shape of the
 96 particles is. Toutou and Roussel [28] have studied the flow behavior of mortars,
 97 considered as suspensions of sand in a cement paste, and the flow behavior of
 98 concretes, considered as suspensions of gravel in a mortar. In both cases, the
 99 influence of the coarsest inclusions volume fraction on the suspending paste
 100 properties was investigated. The dynamic yield stress was extrapolated from
 101 the measured flow-curves. The yield stress of the mortar was found to increase
 102 with the sand volume fraction. However, at low volume fraction (below 20%)
 103 yield stresses of mortars were found to be lower than the yield stress of the
 104 suspending cement paste. Toutou and Roussel [28] attributed this feature to
 105 the increased deflocculation of the cement paste due to the presence of the
 106 inclusions during mixing of the suspension, in agreement with Williams *et al.*
 107 [31]. The yield stress of concrete was also found to increase with the gravel
 108 volume fraction. However, Toutou and Roussel [28] found that adding gravel
 109 at a given volume fraction to a mortar yields a much larger increase of the
 110 yield stress than adding sand at the same volume fraction to a cement paste.

111 The influence of coarse particles on the rheological properties of other yield
 112 stress fluids has been studied by Coussot [32] and Ancey and Jorrot [33]. An-
 113 ceay and Jorrot [33] have suspended coarse particles within a clay dispersion.
 114 They measured the yield stress of the suspension by means of a slump test.
 115 They showed that for well-graded particles, the suspension yield stress does not
 116 depend on the particle characteristics (diameter, material) and that the yield

117 stress diverges when the solid volume fraction value tends toward the maxi-
 118 mum packing fraction. Of course, when the coarse particles are polydisperse,
 119 the value of the maximum packing fraction depends on the size distribution of
 120 the particles, and the yield stress diverges for values of the solid volume frac-
 121 tion depending on this distribution. They observed sometimes that, for low
 122 reduced solid volume fraction, the yield stress can be a decreasing function of
 123 the solid volume fraction of the coarse particle. This effect was ascribed to a
 124 depletion phenomena: the clay particles are supposed to be expelled from the
 125 suspending fluid in the neighborhood of the coarse particles which are then
 126 embedded in a shell of pure water. Then, they cannot contribute to the over-
 127 all yield stress: they behave as voids. Note that this depletion mechanism is
 128 specific to the suspending yield stress fluid studied by Ancey and Jorrot; thus,
 129 it cannot be used to predict what happens when the particles are suspended
 130 in another yield stress fluid.

131 The few existing experimental studies provide very different results; e.g., when
 132 particles having the same shape (spheres) are embedded at a volume fraction
 133 ϕ corresponding to 70% of the maximum packing fraction ϕ_m in a paste,
 134 Geiker *et al.* [26] find that the yield stress of the paste is increased by a factor
 135 50 when the paste is a mortar, whereas Erdogan [27] finds that it is increased
 136 by only a factor 1.3 when the paste is also a mortar, and Ancey and Jorrot [33]
 137 find that, when the paste is a clay dispersion, the yield stress is increased by a
 138 factor 2. Other surprising discrepancies are shown by Toutou and Roussel [28]:
 139 they find that for sand suspended at 70% of ϕ_m in a cement paste the yield
 140 stress is increased by a factor 8 whereas for gravel (of comparable shape and
 141 dispersity) suspended at 70% of ϕ_m in a mortar, it is increased by a factor 25.
 142 As pointed out above, if rigid noncolloidal particles of a given shape and dis-

143 persity were to interact only rheologically with the suspending paste, we would
 144 expect all the results to be roughly consistent as they should not depend on the
 145 paste physicochemical nature. However, the discrepancy between the results
 146 of Geiker *et al.* [26], Erdogan [27], and Ancey and Jorrot[33], and between the
 147 mortar and the concrete case in the work of Toutou and Roussel [28], does not
 148 necessarily imply that there are specific physicochemical interactions between
 149 the different particles and the different pastes involved in these studies, and
 150 that we would fail describing these materials as suspensions of rigid particles
 151 in yield stress fluids. Such discrepancy may indeed find its origin in differences
 152 and shortcomings in the experimental procedures used. Actually, the experi-
 153 ments of Geiker *et al.* [26], Erdogan [27] and Toutou and Roussel [28] involve
 154 a flow of the material. It is then well known that shear-induced migration
 155 of particles towards low shear zones (the external cylinder in coaxial cylin-
 156 ders geometries) is likely to occur [34–36], whatever the care that is taken;
 157 this would cause the material to be heterogeneous inside the measurement
 158 cell, and the measurement to be non-representative of the homogeneous mate-
 159 rial. This is particularly true at high concentrations (above 50% for spherical
 160 monodisperse particles) where it has been shown by Ovarlez *et al.* [36] that
 161 radial migration occurs as an almost instantaneous and unavoidable process in
 162 a Couette geometry. In this case, all the measurements performed in time are
 163 likely to be performed on the same stationary heterogeneous structure: testing
 164 the material at the same rotational velocity at two different times [8] may then
 165 wrongly lead to conclude that there is no shear-induced migration while the
 166 only correct conclusion is that the structure is stationary. Moreover, Geiker *et*
 167 *al.* [26], Erdogan [27] and Toutou and Roussel [28] use a Herschel-Bulkley (or
 168 Bingham) fit of the flow curve to extrapolate the value of the yield stress
 169 instead of a direct measurement. Chateau *et al.* [1] have shown that such

170 an extrapolation generally provides an overestimation of the yield stress of
 171 the suspension, and that this overestimation is more dramatic as the particle
 172 concentration increases. The reason is that the suspension departs from the
 173 Herschel-Bulkley (or Bingham) model at very low shear rate (unaccessible to
 174 most concrete rheometers) and has a lower yield stress than the one extrap-
 175 olated from the measurable flow curve [1]. On the other hand, as the strain
 176 involved in this test is small, there should be no migration, nor extrapolation
 177 problems, in the slump test used by Ancey and Jorrot [33], as long as the yield
 178 stress is high enough to avoid spreading of the material and the correlation
 179 between measured slump and yield stress is suitable to their experiments [37].
 180 Another difference between the procedures is that the particle distribution
 181 after a flow is anisotropic [38–40], whereas the particle distribution is hardly
 182 changed by the slump flow and is thus isotropic in the experiments of An-
 183 cey and Jorrot [33]; as a consequence, the results of Ancey and Jorrot [33] are
 184 not related to the same state of the suspension as the one of Erdogan [27] and
 185 Geiker *et al.* [26]. Finally, note that Ancey and Jorrot [33] and Toutou and
 186 Roussel [28] found in some cases that the suspension yield stress can be lower
 187 than the suspending paste yield stress; as pointed out by Chateau *et al.* [1],
 188 this should not occur if the noncolloidal particle interact only mechanically
 189 with the paste, i.e. these results are likely to apply only to their systems.

190 Finally, it is therefore of high importance to clarify the cases where suspen-
 191 sions can actually be considered as particles in a yield stress fluid, i.e. the cases
 192 where the rheological properties of the suspension depend only on the rheo-
 193 logical properties of the suspending fluid and on the coarse particle volume
 194 fraction, shape and size distribution. With the aim of providing such generic
 195 results, Mahaut *et al.* [2] have recently performed an experimental study on

196 a broad range of materials. They have suspended beads of various sizes and
 197 made of various materials in very different pastes whose common point is to
 198 exhibit a yield stress, and they sought consistency between the results. More-
 199 over, they had a careful look at all the steps of the measurement procedure to
 200 ensure that an homogeneous and isotropic material is studied in all cases. They
 201 showed that the dimensionless elastic modulus $G'(\phi)/G'(0)$ and the dimension-
 202 less yield stress $\tau_c(\phi)/\tau_c(0)$ of such monodisperse suspensions depend on the
 203 bead volume fraction ϕ only (as expected for systems free from specific physico-
 204 chemical interactions or specific slippage at the paste/particle interface). They
 205 found that the elastic modulus/concentration relationship is well fitted to a
 206 Krieger-Dougherty model $(1 - \phi/\phi_m)^{-2.5\phi_m}$ with $\phi_m = 0.57$ for monodisperse
 207 isotropic suspensions. They showed that the yield stress/concentration rela-
 208 tionship is related to the elastic modulus/concentration relationship through
 209 a very simple law $\tau_c(\phi)/\tau_c(0) = \sqrt{(1 - \phi)G'(\phi)/G'(0)}$ in agreement with the
 210 micromechanical analysis of Chateau *et al.* [1], yielding the Chateau-Ovarlez-
 211 Trung model $\tau_c(\phi)/\tau_c(0) = \sqrt{(1 - \phi)(1 - \phi/\phi_m)^{-2.5\phi_m}}$ for the yield stress of
 212 suspensions of monodisperse beads in a yield stress fluid.

213 In this paper, we study suspensions of coarse spherical particles in a thixotropic
 214 cement paste. We measure the static yield stress of the suspensions as a func-
 215 tion of the resting time and of the particle volume fraction. We design new
 216 procedures that allow for comparing the yield stress of a given cement paste
 217 to that of the same cement paste added with particles. We also take care of
 218 designing a procedure that allows for properly accounting for thixotropy of
 219 the paste, independently of any irreversible change in the paste behaviour. In
 220 Sec. 2, we present the materials and the experimental setup. In Sec. 3, we
 221 present the procedure we developed to ensure comparing properly the yield

222 stress of the suspensions to the yield stress of the suspending cement paste, as
 223 a function of the resting time. We present the results in Sec. 4 and compare
 224 the yield stress obtained with particles suspended in a cement paste with this
 225 procedure to the ones obtained on model materials by Mahaut *et al.* [2], and
 226 to the Chateau-Ovarlez-Trung model [1].

227 2 Materials and methods

228 2.1 Pastes and particles

229 We performed our experiments with a thixotropic cement paste. White Ce-
 230 ment CEM I/52.5 N CE CP2 NF “SB” from Gargenville Calcia was used
 231 to prepare all the cement pastes. Its specific gravity is 3.01. Its compressive
 232 strength is 62 MPa at 28 days according to NF EN 196-1 test. The size dis-
 233 tribution was measured in water using a laser granulometer (according to NF
 234 ISO 13320-1 test) for different amount of superplasticizer and is given in Fig. 1.
 235 The specific area determined using a BLAINE permeameter, according to NF
 236 EN 196-6 test, is 4117 cm²/g. The cement chemical constituents are summa-
 237 rized in Tab. 1. The Water to Cement ratio W/C studied here was 0.35. A
 238 Superplasticizer (Glenium 27) and a nanosilica slurry (Rhoimat CS 60 SL,
 239 Rhodia) were added to the mixture with a Superplasticizer to cement mass
 240 ratio of 1% and a nanosilica slurry to cement mass ratio of 2%. The fluids
 241 (water + superplasticizer + nanosilica) were first mixed together to obtain an
 242 homogeneous suspension, and then added to the cement powder before a 5
 243 minutes mixing phase in a planetary Controlab mixer: the velocity was first
 244 set to 140rpm during 2min, and then to 285rpm during 3min. All the exper-

245 iments were performed on the fresh cement paste, less than 75min since the
246 constituents were mixed together. Before any measurement, the cement paste
247 was presheared again in the mixer at 285rpm during 2min in order to always
248 start the experiments on a paste in an initially destructured state.

249 The particles suspended in the cement paste are spherical monodisperse glass
250 beads of 2 mm diameter. This ensures that the particle size is much larger
251 than the paste microstructure, so that the particles may “see” the cement
252 paste as a continuum medium.

253 We chose to compare the results obtained with the suspensions of particles in
254 a cement paste to the one obtained by Mahaut *et al* [2] where particles are
255 suspended in various other yield stress fluids: emulsions, colloidal suspensions,
256 and a physical gel (see Mahaut *et al.* [2] for details on the preparation of these
257 materials). The emulsions are water in oil emulsions, in which the origin of
258 the yield stress is the surface tension between the droplets [5]. The colloidal
259 suspensions are bentonite suspensions, made of clay particles of length of or-
260 der $1\mu\text{m}$ and thickness 10nm. The yield stress then originates from colloidal
261 interactions between the particles. The physical gel is a Carbopol dispersion.
262 Basically, the polymers arrange in roughly spherical blobs which are squeezed
263 together [41,42]; this yields a yield stress. The particles used in the Mahaut *et*
264 *al.* study are spherical monodisperse beads. They are either polystyrene beads
265 of density 1.05, or glass beads of density 2.5., of various particle diameters: 80,
266 140, $315\mu\text{m}$ in the case of the polystyrene beads, and 140, 330 and $2000\mu\text{m}$
267 in the case of the glass beads. The beads are washed in an ultrasound bath
268 during 30 minutes and then dried. This is particularly important for experi-
269 ments performed in Carbopol gels: when the unwashed beads are embedded
270 into a Carbopol gel, it actually results in a lower yield stress than when the

271 washed beads are suspended, indicating residual surface effects [2]; such resid-
 272 ual surface effects may be due to colloidal impurities at the particle surface
 273 (or residual surfactant at the particle surface when polystyrene particles are
 274 used [2]). A single washing is enough to ensure a reproducible state. All ma-
 275 terials were prepared (i) to ensure that the particle size is much larger than
 276 the paste microstructure size, (ii) to check that the results depend only on
 277 the mechanical properties of the paste i.e. that they are independent of the
 278 physicochemical origin of the yield stress, (iii) to check that the results are
 279 independent of the noncolloidal particles size (when the particles are monodis-
 280 perse and have constant shape and surface texture), (iv) to check that there
 281 are neither particle/particle nor particle/paste physicochemical interactions.
 282 Moreover, by varying the suspending paste yield stress, it was checked that the
 283 dimensionless yield stress depends only on the particle volume fraction (when
 284 the particle are monodisperse). If we obtain the same behavior with suspen-
 285 sions prepared with all materials, including the cement pastes, and whatever
 286 the particle size, this ensures that there is no contribution from specific parti-
 287 cles/material physicochemical interactions and that the results we obtain can
 288 be applied to the case of any other particles in any other yield stress fluid (in
 289 particular to any cement paste formulation).

290 The insertion of air is unavoidable. The effect of air on the yield stress is not
 291 negligible [2], it should thus be checked that its content is negligible: it changes
 292 not only the continuous phase mechanical properties [5] but also the effective
 293 bead volume fraction, which is a sensitive parameter at high volume fractions.
 294 However, methods such as centrifugation to remove the bubbles cannot be used
 295 if we want to ensure that the materials remain homogeneous as explained in
 296 Sec. 3. We thus chose to work with a constant volume of material in order to

297 check that the air content is always lower than 1%.

298 All the measurements we present in this paper were performed on suspensions
299 of coarse particles embedded in pastes at a volume fraction ϕ ranging between
300 0 and 55%, with an air content lower than 1%.

301 2.2 *Rheological tools*

302 Most rheometric experiments are performed within a vane in cup geometry
303 (inner radius $R_i = 22.5\text{mm}$, outer cylinder radius $R_e = 45\text{mm}$, height $H =$
304 45mm) on a commercial rheometer (Bohlin C-VOR 200) that imposes either
305 the torque or the rotational velocity (with a torque feedback). In order to
306 avoid wall slip [43,12], we use a six-blade vane as an inner tool, and we glue
307 sandpaper on the outer cylinder wall. For the small particles in model yield
308 stress fluids, we use another six-blade vane in cup geometry (inner radius
309 $R_i = 12.5\text{mm}$, outer cylinder radius $R_e = 18\text{mm}$, height $H = 45\text{mm}$). Working
310 within these wide-gap geometries allows for studying easily coarse particles
311 and to ensure that, for all the materials studied, there are enough particles in
312 the gap to consider that we measure the properties of a continuum medium
313 (the suspension).

314 We measure the yield stress $\tau_c(\phi)$ of the paste as a function of the volume
315 fraction ϕ of coarse particles embedded in the pastes. In a wide gap geometry,
316 the shear stress τ continuously decreases within the gap: the shear stress at
317 a radius R is $\tau(R) = \frac{T}{2\pi HR^2}$. Therefore, one has to choose a definition of
318 the shear stress that is measured in a given rheological experiment. Here,
319 we want to perform yield stress measurements; whatever the measurement

method we choose, yield first occurs where the stress is maximal i.e. along the inner virtual cylinder. As consequence, we define the shear stress measurement as $\tau(R_i) = \frac{T}{2\pi HR_i^2}$, so that the yield stress τ_c is correctly measured (any other definition of the shear stress would provide an underestimation of the yield stress). Anyway, we will focus on the evolution of the dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ with the bead volume fraction ϕ , which should be independent of the definition of τ .

3 Experimental procedure

In this section, we present the procedure aiming at showing the influence of the inclusion of coarse particles on the yield stress of cement pastes. We first show that the choice of the sample preparation and of the yield stress measurement procedure is critical to know how the particles are distributed in the suspension. We then establish a new procedure to ensure a good knowledge of the interstitial paste properties in the suspension.

3.1 Preparation and yield stress measurement

First, we need to define precisely the state of the materials we want study. Three points are actually important: (i) we want to perform our yield stress measurement on a homogeneous suspension, otherwise the measurement would have no meaning, (ii) we want to control the microstructure of the suspensions (i.e. the distribution of the neighbors of the coarse particles) to ensure that all measurements deal with the same state of the suspension, and can be compared and modelled, (iii) we need the interstitial cement paste to be

342 initially destructured in order to study thixotropy.

343 These three points impose severe restrictions about the preparation and yield
344 stress measurement procedures, as shown by Mahaut *et al.* [2]. They showed
345 that measurements involving an important flow of the material (a large strain)
346 pose several problems. First, a flow causes particle migration towards the low
347 shear zones (the outer cylinder in coaxial cylinder geometries) i.e. creation of
348 a heterogeneous structure. This migration phenomenon is well documented
349 for suspensions of noncolloidal particles in Newtonian fluids [34–36] but is
350 still badly known in yield stress fluids. As it needs a large strain to occur
351 for moderate volume fraction [34,35], it may be avoided in these cases by
352 performing only short duration experiments. However, for volume fractions
353 of the order of 50% and more, migration is a critical phenomenon: it seems
354 unavoidable as it is almost instantaneous as shown by Ovarlez *et al.* [36].
355 Another problem when suspensions flow is that an anisotropic microstructure
356 of the particles is created by the flow, as observed in suspensions of particles
357 in Newtonian fluids [38–40]. It is also a critical phenomenon: Mahaut *et al.* [2]
358 showed that suspensions of isotropic and anisotropic microstructure have very
359 different rheological properties.

360 These problems imply that we cannot preshear our materials with the rheome-
361 ter and that we cannot use a yield stress measurement method based on a shear
362 flow such as shear rate [8] or shear stress ramps [44] and creep tests [45]; we
363 then have to measure the static yield stress. On the other hand, as the static
364 yield stress of thixotropic materials depends on the time passed at rest in
365 the solid state [46], the measurements have to be performed on a well defined
366 state of the paste, i.e. the material needs to be first strongly presheared to get
367 a destructured initial state. However, as pointed out above, we cannot apply

368 a controlled preshear with the rheometer to the system after its preparation.
 369 That is why, before loading the material in the measurement cup, we first pres-
 370 sheared the cement paste alone during 2 minutes with the mixer at 285rpm;
 371 this ensures that the cement paste is initially in a destructured state. Then,
 372 the particles and the paste are mixed together in the measurement cylinder,
 373 and the loaded suspension is strongly stirred by hand in random directions to
 374 disperse the particles; this random stirring should ensure keeping the material
 375 in a destructured state while avoiding particle migration and anisotropy. Af-
 376 terwards, the vane tool is inserted in the cup, and we perform our yield stress
 377 measurement after a given resting time with the vane method [47,48]: a small
 378 rotational velocity, corresponding to a shear rate of 0.01s^{-1} is imposed to the
 379 vane tool. Note that we checked that we observe the same effect of the parti-
 380 cles on the yield stress whatever the low velocity that is chosen to drive the
 381 vane tool. Fig. 2 shows the shear stress vs. strain for yield stress measurement
 382 experiments performed in a cement paste. There is an overshoot, followed by
 383 a slow decrease of the shear stress: the peak defines the static yield stress, the
 384 decrease corresponds to destructuration of the material under the shear flow;
 385 the suspension structure at yield should then be isotropic and homogeneous.
 386 Then, any new yield stress measurement requires a new sample preparation
 387 or a new random manual preshear in the cup: it has been shown by Mahaut *et*
 388 *al.* [2] that the small strain of order 1 induced by the whole measurement
 389 procedure is sufficient to change the material state (it is sufficient to change
 390 the suspension microstructure or to induce migration): the suspension states
 391 before and after the yield stress measurements are characterized by different
 392 rheological properties.

394 As we are interested in the influence of the particles on the yield stress of
 395 cement pastes, we will need to compare the suspension yield stress and the
 396 cement paste yield stress. It is thus important to ensure that we have a good
 397 knowledge of the properties of the interstitial paste state in the suspension.
 398 The procedure developed to ensure this measurement is presented in detail in
 399 the Appendix A. We present here the main steps.

400 First, we have to note that it is very difficult to achieve a good reproducibility
 401 of a cement paste mechanical behavior (see Appendix A). That is why we
 402 chose to work on the same batch for the measurement of the properties of the
 403 paste alone and for the suspension.

404 Then, for a given cement paste batch, we observe that yield stress measure-
 405 ments performed in the same conditions as regards thixotropic effects (i.e. for
 406 a 2 minutes resting time after a strong stirring of the paste) provide values
 407 that depend on the time t_{age} elapsed since the constituents of the cement paste
 408 were mixed together (Appendix A). This means that one cannot know what
 409 is the yield stress of the interstitial paste in a suspension if the yield stress of
 410 the cement paste alone is not measured at exactly the same time after mixing
 411 as the yield stress of the suspension. That is why we chose to measure simul-
 412 taneously the yield stress of the suspension and the yield stress of the cement
 413 paste alone in exactly the same conditions (same age t_{age} after mixing the
 414 constituents of the cement paste, same time t_{rest} after the end of the strong
 415 stirring), with the help of 2 rheometers that perform their measurements in
 416 parallel.

417 We have also shown that when the same suspension sample, after a first resting
418 period and a first measurement, is stirred again in the measurement cup,
419 its interstitial cement paste is not in the same state of destructurement as
420 the cement paste alone stirred with the same procedure (Appendix A). This
421 means that the suspension and the cement paste cannot be compared anymore.
422 A solution to this problem is to perform only a single measurement on a
423 suspension, for a given resting time after its preparation.

424 A key point of the comparison between the suspension and the cement paste
425 is actually that the cement paste is initially strongly presheared in the mixer
426 for both samples: this defines an initial destructured state of the paste that is
427 the same both for the interstitial cement paste and for the cement paste alone.
428 After this preshear, the cement paste is loaded alone in one measurement cup,
429 and with the particles in another cup. Both samples are then strongly stirred
430 by hand during 30s in random directions: this ensures an homogeneous dis-
431 persion of the particles in the suspension, while keeping the cement paste in
432 a destructured state in both samples. Then, the stirring is stopped simultane-
433 ously for both samples: this defines the beginning of the resting period. With
434 this procedure, we have shown that the paste alone and the interstitial paste
435 have the same history and thus the same behavior (see Appendix A).

436 Finally, as the cement paste is thixotropic, its static yield stress increases as
437 a function of the time t_{rest} elapsed since the end of the stirring. However,
438 we showed that the yield stress value also depends on the time t_{age} elapsed
439 since the constituents were mixed together, even at short times. This would
440 mean that a characterization of thixotropy would only have a meaning for this
441 age t_{age} , and it would make the study of the impact of the coarse particles
442 on this thixotropy difficult. Nevertheless, we have shown that the irreversible

phenomena can be separated from the reversible phenomena. The thixotropic (reversible) increase of the yield stress is actually the same whatever the paste age t_{age} : it depends only on the time t_{rest} elapsed since the end of a preshear (Appendix A). The increase of the yield stress of our cement paste due to thixotropy is basically linear in t_{rest} : it reads $\tau_c(t_{rest}) = A_{thix} t_{rest}$ with an increase rate $A_{thix} = 12\text{Pa/min}$.

3.3 Summary

As a summary we present in Fig. 3 a sketch of the whole procedure used to study the influence of coarse particles on the yield stress of cement pastes.

This procedure ensures (i) that an homogeneous material is studied; (ii) that we study a well defined state of the material: we chose to study the case of isotropic distributions of particles; (iii) that the interstitial cement paste is well characterized; (iv) that the initial destructured state of the interstitial cement paste is well defined; (v) that thixotropy is accounted for and separated from irreversible phenomena; (vi) that the results obtained with cement pastes can be compared to measurements performed in other yield stress fluid.

4 Experimental results

In this section, we summarize the results of the yield stress measurements performed on the suspensions with the procedure presented above. We compare the results obtained with the cement pastes to the results obtained by Mahaut *et al.* [2] with various yield stress fluids, and to the Chateau-Ovarlez-Trung model [1].

466 In Fig. 4 we plot the dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ vs. the volume
 467 fraction ϕ of coarse particles embedded in the cement paste, when the yield
 468 stresses are measured with the procedure developed in Sec. 3 for various times
 469 t_{rest} after the end of a strong stirring.

470 We first observe that the yield stress increases when the coarse particle volume
 471 fraction is increased. This increase is quite limited for volume fraction lower
 472 than 45%: in this case, the yield stress is increased by a factor less than 3.
 473 However, the yield stress is found to increase sharply at the approach of a
 474 60% volume fraction. E.g., the yield stress of a suspension of 55% particles is
 475 20 times higher than the yield stress of the interstitial cement paste.

476 We also observe in Fig. 4 that the same evolution of the yield stress with the
 477 particle volume fraction is found whatever the time t_{rest} passed at rest before
 478 the measurement. This means that the yield stress of suspensions of coarse
 479 particles embedded at a volume fraction ϕ in a thixotropic cement paste of
 480 time-dependent yield stress $\tau_c(0, t)$ reads

$$\tau_c(\phi, t) = \tau_c(0, t)g(\phi) \quad (1)$$

481 This feature is expected if the coarse (i.e. noncolloidal) particles have only a
 482 mechanical interaction with the cement paste [2]: in this case, they should not
 483 interfere with the physical process at the origin of thixotropy. Then, at time
 484 t the interstitial paste has naturally the same yield stress $\tau_c(0, t)$ as if it had
 485 not been in contact with the coarse particles. Finally, as the relative increase

486 of the yield stress due to the monodisperse particles should be a function
 487 of their volume fraction ϕ only, independently of the value of the interstitial
 488 fluid yield stress, the yield stress of the suspension at time t is expected to be
 489 equal to $\tau_c(0, t)$ multiplied by some function $g(\phi)$ whatever $\tau_c(0, t)$, as observed
 490 experimentally.

491 Eq. 1 has an interesting consequence: it means that it is sufficient to know
 492 how the interstitial cement paste evolves in time to predict the suspension
 493 evolution at rest. This is important for fresh concrete as their behavior is
 494 hard to measure: our results show that the knowledge of the cement paste
 495 structuration rate at rest is sufficient to predict the fresh concrete structuration
 496 rate. As found on the cement paste we studied (see Sec. 3.2) the yield stress
 497 evolution at rest after a preshear of a cement paste usually reads [19]:

$$\tau_c(0, t) = \tau_c(0) + A_{thix}t \quad (2)$$

498 where A_{thix} is the structuration rate of the paste. In this case, Eq. 1 reads:

$$\tau_c(\phi, t) = \tau_c(0)g(\phi) + A_{thix}g(\phi)t \quad (3)$$

499 As a consequence, if the mechanical impact of the coarse particles is to increase
 500 the yield stress by a factor $g(\phi)$, then their impact on the structuration rate of
 501 the paste is to increase it also by a factor $g(\phi)$. It is thus sufficient to measure
 502 the cement paste yield stress evolution in time (i.e. A_{thix}) and to measure the
 503 increase of the yield stress with the volume fraction (i.e. $g(\phi)$) for a single
 504 resting time t_{rest} to infer the value $A_{thix}g(\phi)$ of the structuration rate of the
 505 suspension (and more generally of fresh concrete).

507 In Fig. 5, we plot a summary of the dimensionless yield stress measurements
 508 $\tau_c(\phi)/\tau_c(0)$ performed on all the materials by Mahaut *et al.* [2], together with
 509 the results obtained with cement pastes.

510 We find that all the results are consistent: the dimensionless yield stress
 511 $\tau_c(\phi)/\tau_c(0)$ is independent of the physicochemical origin of the material yield
 512 stress, of the bead material and of the bead size, and of the paste yield stress;
 513 it is a function of the volume fraction only. This means that the particles have
 514 a purely mechanical contribution to the paste behavior, which is indepen-
 515 dent of the physicochemical properties of the materials: the only important
 516 matter is the value of the yield stress of the paste. This also validates our
 517 approach: as long as the coarse particle size is much larger than the cement
 518 paste microstructure, a suspension of coarse particles in a cement paste can
 519 be considered more generally as a suspension of rigid noncolloidal particles in
 520 a yield stress fluid.

521 This result helps proposing a method than can be applied to obtain quickly
 522 the effect of particles of any kind (any shape, any size distribution) on the yield
 523 stress of a cement paste. Actually, preparing a model yield stress fluid of stable
 524 and reproducible rheological properties, showing no setting nor thixotropic
 525 effects (e.g. an emulsion), is quite easy, and measurements are much easier to
 526 perform on these materials. Then a great amount of accurate experiments can
 527 be performed to measure the properties of suspensions of particles in this yield
 528 stress fluid. Finally, the result of the measurement of the dimensionless yield
 529 stress $\tau_c(\phi)/\tau_c(0) = g(\phi)$ as a function of the volume fraction ϕ of particles

530 in this yield stress fluid should hold if the interstitial paste is a cement paste.
 531 Moreover, we have shown that the knowledge of the structuration rate A_{thix} of
 532 a cement paste is sufficient to infer the structuration rate of the suspension of
 533 particles in this cement paste (it is equal to $A_{thix}g(\phi)$). A measurement of the
 534 cement paste structuration at rest plus the measurement $\tau_c(\phi)/\tau_c(0) = g(\phi)$ in
 535 a model yield stress fluid then provides everything that is needed to infer the
 536 behavior of mortars or concretes. Note however that these results apply only
 537 as long as the particle size is much larger than the cement paste microstructure
 538 typical size so that the particles see the yield stress fluid as an homogeneous
 539 material. This should not be true otherwise: if the particles were to be sensitive
 540 to the cement paste microstructure, then the behavior should depend on the
 541 exact details of the specific microstructure of each paste. E.g., in the case of
 542 particles suspended in a foam, Cohen-Addad *et al.* [49] found that the behavior
 543 of the suspension depends on the particle size for particles of size lower than
 544 5 times the bubble size in the foam. Note finally that another important
 545 requirement is that the fraction of superplasticizer adsorbed at the surface
 546 of the aggregates suspended in the paste is negligible. The study of Hammer
 547 and Wallevik [23] suggests that in some cases (it may depend strongly on the
 548 cement paste composition) this may be true only if the aggregates are larger
 549 than 0.25 to 0.5mm; in such cases, our approach would then be valid for SCC
 550 only if the suspending yield stress fluid includes the fine aggregates (of size
 551 lower than 0.25 to 0.5mm in the study of Hammer and Wallevik).

553 Proposing a theoretical value for the dimensionless yield stress is challenging.
 554 However, it has been shown by Chateau *et al.* [1] that it is possible to give
 555 a general relationship between the linear response of the materials (e.g. its
 556 dimensionless elastic modulus $G'(\phi)/G'(0)$ as probed under the yield stress)
 557 and the dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ of a suspension of rigid parti-
 558 cles in a yield stress fluid. This estimate is based on the following hypotheses:
 559 the particles are rigid and noncolloidal; there are no physicochemical interac-
 560 tions between the particles and the paste; the distribution of the particles is
 561 isotropic. This is what we have managed to perform experimentally, therefore,
 562 our experiments are fitted to provide a test of these theoretical predictions.
 563 Chateau *et al.* [1] find

$$\tau_c(\phi)/\tau_c(0) = \sqrt{(1 - \phi)G'(\phi)/G'(0)} \quad (4)$$

564 Mahaut *et al.* [2] have measured the elastic modulus of all the suspensions
 565 studied above, and found a Krieger-Dougherty model to apply $G'(\phi)/G'(0) =$
 566 $(1 - \phi/\phi_m)^{-2.5\phi_m}$ for the dimensionless elastic modulus. Combining this equa-
 567 tion and the theoretical expression Eq. 4 thus yields for the yield stress the
 568 Chateau-Ovarlez-Trung model [1]

$$\frac{\tau_c(\phi)}{\tau_c(0)} = \sqrt{\frac{1 - \phi}{(1 - \phi/\phi_m)^{2.5\phi_m}}} \quad (5)$$

569 which should be valid for any isotropic suspension of rigid spherical noncol-
 570 loidal particles in yield stress fluids with no physicochemical interactions be-
 571 tween the particles and the paste.

Our experimental data are compared to Eq. 5 on Fig. 5. We find a remarkable agreement between our data and this model with a best fit for $\phi_m = 0.56$; note that this value of 0.56 is valid only for the case of monodisperse spherical particles we studied.

Note however that Eq. 5 can *a priori* be easily modified to account for polydispersity and for complex shapes of the particles when studying more complex suspensions. Actually, Eq. 4 should hold in all cases. It is then sufficient to know what is the linear response of a suspension made with the studied particles to infer the yield stress value. This linear response can be measured with the method presented in this paper (it is the dimensionless elastic modulus $G'(\phi)/G'(0)$); it can also be inferred from the huge amount of dimensionless viscosity data from the literature dealing with suspensions of particles in Newtonian fluids (with the same particles): the problem of the elasticity of a suspension of rigid particles in a linear elastic material is actually formally similar to the problem of the viscosity a suspension of rigid particles in a Newtonian (thus linear) material.

4.4 Comments

Our results are naturally close to the Ancy and Jorrot [33] ones, as they have chosen to measure the yield stress of the suspension by means of a slump test which ensures avoiding migration of particles and anisotropy of the material. On the other hand, we find very different values from Geiker *et al.* [26], Erdogan [27], and Toutou and Roussel [28]. As pointed out in Sec. 1 and Sec. 3, this is due to the shortcomings of their experimental procedure which is based on a flow and an extrapolation of the dynamic yield stress from a flow curve. Their

materials are then likely to be heterogeneous and anisotropic. Moreover, an extrapolation from a flow curve provides an overestimation of the yield stress of the suspension [1] because the suspension departs from a Herschel-Bulkley (or Bingham) model at very low shear rate (unaccessible to most concrete rheometers) and has a lower yield stress than the one extrapolated from the accessible flow curve, even if the suspending yield stress fluid has a Herschel-Bulkley (or Bingham) behavior.

Finally, note that in most papers the results are presented vs. ϕ/ϕ_{max} where ϕ_{max} is the maximum packing fraction (taken at about 0.65 for monodisperse particles) and the yield stress divergence is expected to occur for $\phi/\phi_{max} = 1$. This is not correct: the maximum volume fraction ϕ_m for the yield stress sharp increase should not be taken as the maximum volume fraction one can reach by packing particles together (which is the definition of the maximum packing fraction ϕ_{max}). The maximum volume fraction for the yield stress sharp increase is rather the one at which direct contacts become important, which is the limit of application of models including only hydrodynamic interactions between the particles, and also the limit between SCC and ordinary rheology concretes [29]. This explains why we find the yield stress to diverge at around 56% while the maximum packing fraction is of about 65% for spheres.

5 Conclusion

We have studied the behavior of suspensions of coarse particles in a thixotropic cement paste. We managed to design procedures that allow for (i) studying an homogeneous and isotropic suspension, (ii) comparing the yield stress of a given cement paste to that of the same cement paste added with particles,

(iii) accounting properly for the thixotropy of the cement paste. We observed that the yield stress of these pastes follows the very simple Chateau-Ovarlez-Trung model [1] $\tau_c(\phi)/\tau_c(0) = \sqrt{(1-\phi)(1-\phi/\phi_m)^{-2.5\phi_m}}$, with $\phi_m = 0.56$ for monodisperse spherical particles, consistently with the experimental results of Mahaut *et al.* [2] obtained with many different suspensions. This supports the fact that the yield stress of the suspension is independent of the physicochemical properties of the yield stress fluid, and depends only on its yield stress value. This shows that studies of suspensions in model yield stress fluids can be used as a general tool to infer the behavior of fresh concrete. Moreover, we showed that the thixotropic structuration rate of these pastes (their static yield stress increase rate in time) is not changed by the presence of the particles. This shows that it is sufficient to measure the cement paste yield stress evolution in time and to measure the increase of the yield stress with the volume fraction of coarse particles for a single resting time to predict the value of the structuration rate of fresh concrete. For a linear increase of the cement paste yield stress with a rate A_{thix} , we predict a linear increase of the suspension with a rate $A_{thix}\sqrt{(1-\phi)(1-\phi/\phi_m)^{-2.5\phi_m}}$.

A Characterization of the interstitial paste

In this appendix, we detail the arguments that have led to develop the procedure presented in Sec. 3. This new procedure is built to ensure a good knowledge of the mechanical properties and of the state of structuration of the interstitial cement paste in the suspension.

First, we have to note that it is very difficult to achieve a good reproducibility of a fresh cement paste mechanical behavior. In Fig. A.1a we show the result

644 of the yield stress measurements performed (apparently) exactly in the same
 645 conditions in 2 cement pastes having the same composition. We observe that
 646 the uncertainty on the yield stress of the cement paste we get is of order 25%.
 647 This means that, if we want to measure accurately the ratio of the suspension
 648 yield stress to the interstitial cement paste yield stress, we cannot compare the
 649 properties of suspensions of particles in a cement paste to the properties of a
 650 cement paste having the same composition but being from a different batch.
 651 That is why we chose to work on the same batch for the measurement of the
 652 properties of the paste alone and for the suspension.

653 Then, for a given cement paste batch, we could propose to first measure the
 654 cement paste yield stress and then the suspension yield stress. For the results
 655 to be comparable, one would then just have to perform the experiment in the
 656 same conditions as regards thixotropy (i.e. for the same resting time after a
 657 strong preshear). In order to check the validity of this method, we performed
 658 yield stress measurements several times in the same conditions (i.e. for a 2
 659 minutes resting time after a strong stirring of the paste) on a single cement
 660 paste batch. The results of this experiment are depicted in Fig. A.2 as a
 661 function of the time t_{age} elapsed since the constituents of the cement paste
 662 were mixed together. We observe that due to various irreversible chemical
 663 interactions in the material, the cement paste yield stress, measured in the
 664 same conditions as regards thixotropic effects, evolves (non-monotonously) as
 665 a function of the time t_{age} elapsed since the constituents of the cement paste
 666 were mixed together. This means that one cannot know what is the yield
 667 stress of the interstitial paste in a suspension if the yield stress of the cement
 668 paste alone is not measured at exactly the same time after mixing as the yield
 669 stress of the suspension. That is why we chose to measure simultaneously the

670 yield stress of the suspension and the yield stress of the cement paste alone
 671 in exactly the same conditions (same age t_{age} after mixing the constituents of
 672 the cement paste, same time t_{rest} after the end of the strong stirring), with
 673 the help of 2 rheometers that perform their measurements in parallel. We
 674 show in Fig. A.1b that, as expected, this method yields a very low uncertainty
 675 when the measurements are performed on the same cement paste (without
 676 particles).

677 Now, by performing these simultaneous measurements of the suspension yield
 678 stress $\tau_c(\phi)$ and of the cement paste yield stress $\tau_c(0)$ several times, at various
 679 ages t_{age} after mixing the constituents of the cement paste, we should observe
 680 the same effect of the particles on the yield stress whatever the age of the
 681 cement paste. In Fig. A.3 we present the dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$
 682 as a function of the time t_{age} elapsed since the constituents of the cement paste
 683 were mixed together. We observe that $\tau_c(\phi)/\tau_c(0)$ is not constant in time. This
 684 means that the interstitial paste is not in the same mechanical state as the
 685 paste alone although they have apparently the same history. The only differ-
 686 ence stands in the preshear procedure: before performing each measurement,
 687 the paste and the suspension are presheared to ensure a reproducible destruc-
 688 tured initial state. As pointed out above, the preshear has to be manual to
 689 avoid migration and anisotropy. Our results show that this preshear is not as
 690 efficient in the suspension as in the cement paste. It is harder to shear the sus-
 691 pension, thus an experimentalist cannot shear the suspension the same way as
 692 the paste alone. It can be noted that, in the case of a strong mechanical pres-
 693 hear in a mixer, an opposite result has been obtained by Toutou and Roussel
 694 [28] due to the mixing effect of the particles. As a result of these imperfect
 695 and perturbing preshears, the differences between the structuration state of

696 the paste alone and of the interstitial paste in the suspension increases with
 697 time. The suspension and the cement paste cannot be compared anymore, and
 698 the function $\tau_c(\phi)/\tau_c(0)$ is no more correctly measured by this means. A solu-
 699 tion to this problem is to perform only a single measurement on a suspension,
 700 for a given resting time after its preparation. As pointed out in Sec. 3.2, a
 701 key point of the comparison between the suspension and the cement paste is
 702 then that the cement paste alone is first initially strongly presheared in the
 703 mixer for both samples before being loaded (and eventually mixed with the
 704 particles) in the measurement cups: this defines an initial destructured state
 705 of the paste that is the same both for the interstitial cement paste and for
 706 the cement paste alone. The manual stirring in the measurement cup then
 707 ensures an homogeneous dispersion of the particles in the suspension, while
 708 keeping the cement paste in a destructured state in both samples. With this
 709 procedure, one ensures that the paste alone and the interstitial paste have the
 710 same history and thus the same behavior. We show actually in Fig. A.3 that,
 711 in these conditions, the same value of $\tau_c(\phi)/\tau_c(0)$ is found within the measure-
 712 ment uncertainty whatever the time t_{age} elapsed since the constituents of the
 713 cement paste were mixed together.

714 Finally, as the cement paste is thixotropic, its static yield stress increases as
 715 a function of the time t_{rest} elapsed since the end of the stirring. However,
 716 if we want to account properly for the (reversible) thixotropic behavior of
 717 the cement paste, and to check what the influence of the particles on this
 718 thixotropic behavior is, we face a problem. We showed that the value of the
 719 yield stress measured 2 minutes after a strong stirring evolves as a function
 720 of the time t_{age} elapsed since the constituents were mixed together, even at
 721 short times. This would mean that in order to characterize the increase of

the yield stress of cement pastes due to structuration at rest, as a function of the resting time t_{rest} after a strong stirring, we would need to perform all the yield stress measurements only at a same given age t_{age} after mixing the constituents of the cement paste. And this characterization of thixotropy would only have a meaning for this age t_{age} . However, we show in the following that the thixotropic increase of the yield stress is actually the same whatever the paste age. In Fig. A.4a, we plot the yield stress of a cement paste as a function of the age t_{age} of the paste for 3 different times t_{rest} after a strong stirring; note that as we have only 2 rheometers, these measurements had to be performed on 2 batches, so that the uncertainties may be rather large (as in Fig. A.1a).

We observe the same evolution of the paste behavior as a function of t_{age} whatever t_{rest} . An important consequence is that the irreversible effects can be separated from the reversible effects by writing

$$\tau_c(t_{rest}, t_{age}) = \tau_c(t_{age}) + \tau_c(t_{rest}) \quad (\text{A.1})$$

where $\tau_c(t_{age})$ is the yield stress that would be measured just after a preshear, which depends on the time t_{age} elapsed since the constituents were mixed together, and $\tau_c(t_{rest})$ represents the increase of the yield stress due to thixotropic effects, which depends only on the time elapsed since the end of a preshear. This is shown in Fig. A.4b: all data are superposed when shifted by a constant value that depends only on t_{rest} . From the superposition of data in Fig. A.4b, we find that $\tau_c(t_{rest}=4\text{min}) - \tau_c(t_{rest}=2\text{min}) = 24\text{Pa}$ and $\tau_c(t_{rest}=6\text{min}) - \tau_c(t_{rest}=2\text{min}) = 48\text{Pa}$. This is consistent with the simple law proposed by Roussel [19] i.e. the increase of the yield stress due to thixotropy

745 is basically linear in t_{rest} : it reads $\tau_c(t_{rest}) = A_{thix} t_{rest}$ with an increase rate
 746 $A_{thix} = 12\text{Pa}/\text{min}$. Finally, as the absolute increase of the yield stress due to
 747 thixotropic effects is the same at any time t_{age} (lower than 90 min) since the
 748 constituents were mixed together, this shows that studies of thixotropy and
 749 of the effect of the coarse particles on this thixotropy performed at different
 750 times t_{age} can be compared together and provide relevant information on the
 751 thixotropy of the suspensions.

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Constituents	% by mass
SiO ₂	20.95
Al ₂ O ₃	4.08
TiO ₂	0.14
Fe ₂ O ₃	0.22
CaO	65.55
MgO	0.49
Na ₂ O	0.12
K ₂ O	0.20
SO ₃	2.60
RI	1.47
PAF	3.36

Table 1

Cement chemical constituents.

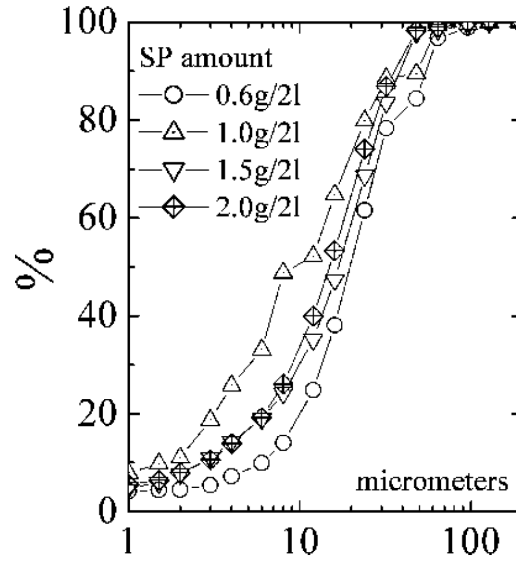


Fig. 1. Cement size distribution curve for various superplasticizer (SP) amount.

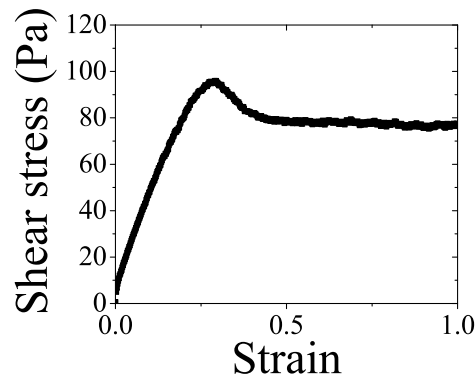


Fig. 2. Shear stress vs. strain when slowly shearing a cement paste at 10^{-2}s^{-1} 2 minutes after a strong stirring of the paste.

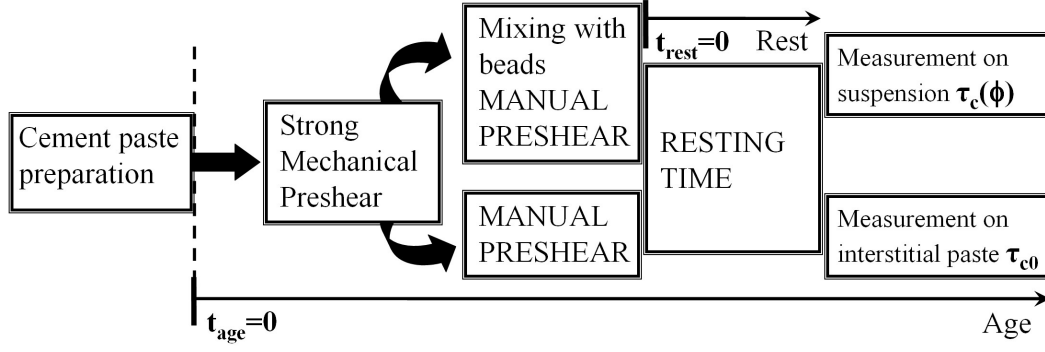


Fig. 3. Sketch of the procedure designed to study the evolution of the dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ with the volume fraction ϕ of particles in the suspension.

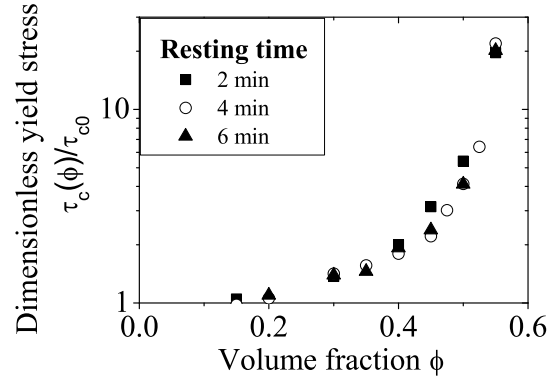


Fig. 4. Dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ vs. the bead volume fraction ϕ for suspensions of 2mm glass beads in a cement paste, measured with the procedure developed in Sec. 3 for various times t_{rest} after a strong stirring of the suspension.

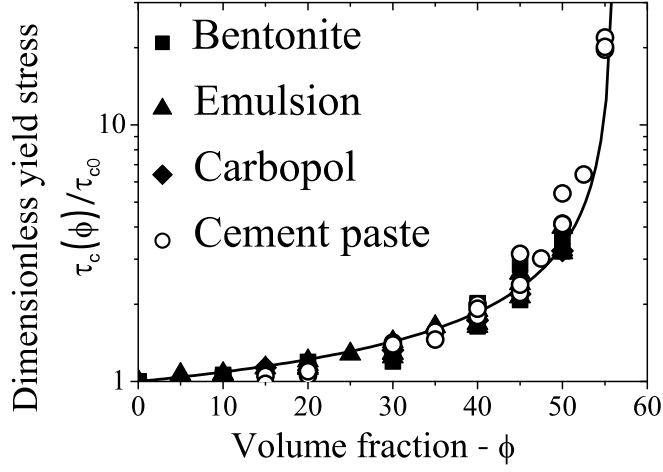


Fig. 5. Dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ vs. the bead volume fraction ϕ for suspensions of 80, 140, and 315 μm polystyrene beads and 140 μm , 330 μm and 2mm glass beads in various bentonite suspensions, emulsions and Carbopol gels (results from Mahaut *et al.* [2]), and for 2mm glass beads suspended in a cement paste. The solid line is the Chateau-Ovarlez-Trung model $\sqrt{(1-\phi) \times (1-\phi/\phi_m)^{-2.5\phi_m}}$ with $\phi_m = 0.56$.

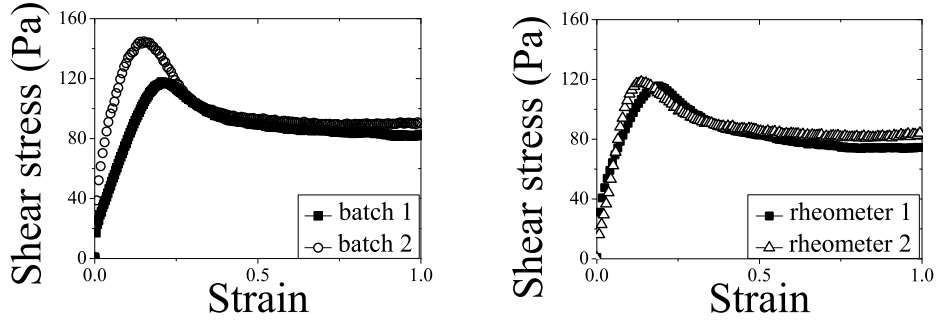


Fig. A.1. a) Shear stress vs. strain when slowly shearing two batches of a cement paste at 10^{-2} s^{-1} 2 minutes after a strong stirring of the paste. b) Shear stress vs. strain when slowly shearing simultaneously on 2 rheometers a cement paste from a single batch at 10^{-2} s^{-1} 2 minutes after a strong stirring of the paste.

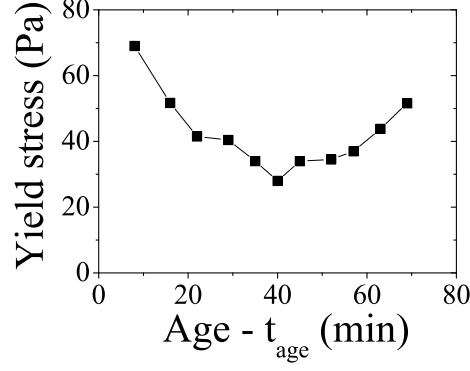


Fig. A.2. Yield stress of a cement paste measured 2 minutes after a strong stirring of the paste vs. the time t_{age} elapsed since the constituents of the cement paste were mixed together.

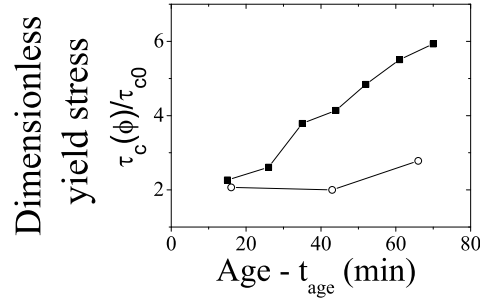


Fig. A.3. Dimensionless yield stress $\tau_c(\phi)/\tau_c(0)$ measured 2 minutes after a strong stirring of the suspension vs. the time t_{age} elapsed since the constituents of the cement paste were mixed together (with a volume fraction of coarse particles $\phi = 40\%$), in two cases: when the same suspension of particles is used for all measurements (squares); when the particles are mixed with the cement paste just before each measurement (open circles).

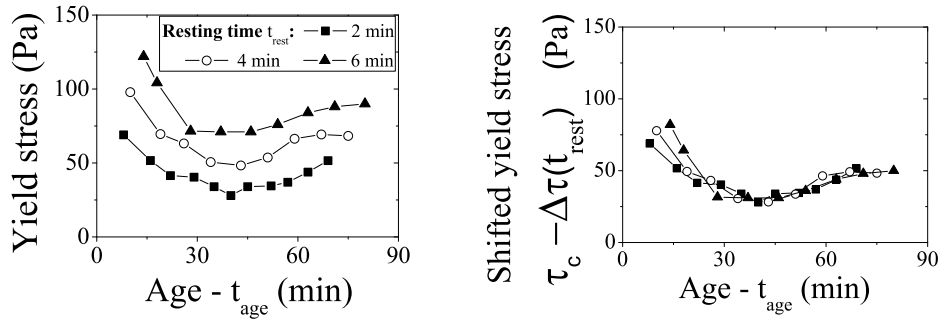


Fig. A.4. a) Yield stress of a cement paste measured 2, 4 and 6 minutes after a strong stirring of the paste vs. time t_{age} elapsed since the constituents of the cement paste were mixed together. b) Data of Fig. A.4a rescaled by shifting the yield stress values by a function $\Delta\tau_c(t_{rest})$ of the time t_{rest} elapsed since the end of the strong stirring.