Interaction between a falling sphere and the structure of a non-Newtonian yield-stress fluid

Nicolò R. Sgreva^{a,*}, Anne Davaille^a, Ichiro Kumagai^b, Kei Kurita^c

^a*Université Paris-Saclay, CNRS, FAST, 91405, Orsay, France.*

^b*School of Science and Engineering, Meisei University, Hino, Tokyo 191-8506, Japan*

^c*Earthquake Research Institute, University of Tokyo, Tokyo, Japan*

Abstract

We present an experimental study using mixtures of aqueous superabsorbent polymers (SAP) where we systematically investigate the influence of the size of grains that make up the fluid structure on the effective rheology and its domain of validity. In water, these polymer powder grains swell up to 200 times and form gel grains whose size can be controlled by controlling the size of the initial powder. The rheology of this mixture (water and touching grains) combines viscous, elastic and plastic aspects and can be characterized using the free-fall of spheres of different diameters (between 3 and 25 mm-diameter) and densities (from 2200 to 15000 kg/m³). As the typical size of the gel grains was varied between 1 and 8 mm, there is a range where it becomes comparable to the size of the falling spheres. We observe five different motion regimes. (1) A linear regime where the sphere has a rapid and linear fall and reaches a constant terminal velocity. (2) An irregular regime where the sphere's velocity varies around a constant value. (3) An intermittent regime where periods of no-motion and periods of irregular falls follow one another.

*nicolo.sgreva@u-psud.fr

(4) A slow fall regime where the sphere's speed progressively decreases in a logarithmic way. And (5) a no-motion regime when spheres are not heavy enough to overcome the yield stress of the mixture, or are too small compared to the grain size. Besides these five regimes, we find that the effective yield stress and effective consistency of the mixtures always increase with the grain size, suggesting that the effective rheology depends on the size of the grains which constitute the fluid structure. Moreover, the critical Yield number (Y_c) above which there is no motion decreases as the sphere to grain diameters ratio becomes smaller than 2. This shows the role of the fluid structure also on the critical condition of motion under which the sphere is trapped.

Keywords: Yield stress, Falling sphere, SAP, Irregular motion, Intermittent motion

1. Introduction

Yield-stress fluids are a broad category of fluids which have been intensively studied in the last decades due to their key roles both in industry (e.g. drilling fluids, cement pastes, foams) and in natural phenomena (e.g. lava and mud flows, avalanches, landslides). Very often, the material consists typically of a suspension of particles in a liquid. When the particle volume fraction increases, they become in close contact with one another, and the material can face jamming. In this jammed state, the material can support stresses without flowing. A so-called yield-stress fluid does not flow if the applied stresses are not high enough to unjam the structure, but it does flow when the stresses become larger than a threshold value, the yield stress (σ_Y). The latter introduces a non-linearity into the material rheology, which

13 strongly impacts on its dynamics [1, 2, 3, 4]. An example of these materi-
14 als are soft particle glasses. Above the yield stress, their flow results from
15 the concurrence of particles elastic deformations and plastic events during
16 which particles rearrange [5]. Plastic local events can influence each other
17 and lead to non-local elastic relaxation. This collective rearrangement can
18 be the reason of spatial variation of viscosity in an homogeneous stress field
19 in emulsions [6, 7]. Other finite size effects to the bulk rheology of a jammed
20 system appear due to concentration gradients of particles in non-Brownian
21 particle suspensions [8] or due to structural heterogeneities in polymers [9].

22 Several rheological models have been proposed to describe the rheology
23 above the yield stress, such as the Bingham and the Herschel-Bulkley "HB"
24 models. One issue is the domain of validity of these rheology models: how
25 well do they describe the fluid behavior close to the yield stress and how much
26 larger than the microstructure typical length scale must be the flow length
27 scale? For example, if we take the simple (and well known in Newtonian
28 fluids) problem of a solid sphere settling in a particulate fluid, when and how
29 will the fluid microstructure interact with the sphere motion? There are two
30 end-members depending on the size ratio between the fluid microstructure
31 d_g (e.g. the size of the particles in suspension in the liquid) and the falling
32 sphere d_s . When $d_g/d_s \gg 1$, the sphere will fall through a porous medium,
33 moving along the liquid channels between particles, and when $d_g/d_s \ll 1$,
34 the sphere will see a continuum with an effective rheology (e.g. "HB" model).
35 But what happens for intermediate d_g/d_s ?

36 The $d_g/d_s \ll 1$ case has been well studied for viscoplastic fluids, such
37 as Carbopol [2]: the sphere reaches a steady-state downward motion only

38 if a critical force is overcome, otherwise the fall is prevented [10, 11, 12].
 39 However, the accurate value of this critical force is still debated and it varies,
 40 for example, considering or not the elasticity of the material [13]. Once the
 41 sphere is set in motion, crucial aspects are the determination of the drag
 42 coefficient [10, 11, 14, 15] and the determination of the shape and position of
 43 the yield surface that separates the solid-like region from the fluid-like one.
 44 The yield surface and the drag force on the sphere have first been determined
 45 numerically and analytically for a Bingham fluid by Beris *et al.* [10] and
 46 subsequently for an HB fluid numerically [14, 16, 15] and experimentally by
 47 Tabuteau *et al.* [11] in Carbopol. Within the liquid-like region, the flow in
 48 Carbopol is confined nearby the sphere, within an area which size depends
 49 on the yield stress of the fluid [17], and can be either symmetric [18] or
 50 asymmetric [17], depending on the sample preparation. Similar results
 51 had been found for Laponite, a very thixotropic colloidal suspensions [19],
 52 where the asymmetry of the flow pattern increases with the age of the fluid
 53 through the appearance of a negative wake (i.e. upward fluid motion in
 54 the sphere's wake [20]) and in viscoelastic aqueous polyacrylamide solutions
 55 [21]. Another peculiar characteristic of a falling sphere in yield stress fluids
 56 or in viscoelastic materials is that its velocity can depart from a constant
 57 value and show oscillations and irregularities. One of the first observations
 58 of an oscillating particle settling has been in hydroxyl propyl guar (HPG)
 59 polymer gels [22], a yield-stress power-law family of fluids [23]. In this work,
 60 the authors suggested that the irregularities in the descending motion were
 61 linked to the elasticity of the gel. Something similar is reported in entangled
 62 wormlike micellar fluids [24, 25], in Laponite [26], where the vertical velocity

63 can oscillate in bursts and, again, in HPG [27]. In cornstarch suspensions
 64 [28, 29], the sphere velocity never approaches a steady terminal velocity but
 65 instead it oscillates, decreases with time, to reach a series of stop-go cycles
 66 as the sphere becomes close to the experimental tank bottom. In wormlike
 67 micellar fluids, the cause of non-transient oscillations is to be found in the
 68 formation and successively breakage of flow-induced structures; in cornstarch
 69 suspensions, in the formation and dissolution of a jammed layer that, under
 70 sufficient stress, increases drag and slows the sphere down; while the irregular
 71 motion in laponite is due to the existence of flow instabilities and shear
 72 banding.

73 In this paper, we consider viscoplastic fluids that are constituted of an
 74 aqueous suspension of soft gel grains. To do so, we place ourselves in between
 75 the two end-members described above, using macroscopic super absorbent
 76 gels. Varying independently the size of the gels grains and the size of the
 77 falling spheres allows us to investigate the influence of the particle size on the
 78 effective rheology of the fluid and on the motion of spheres that are falling
 79 through it. The paper is organized as follow: in section 2 we describe the
 80 experimental set-up and the fluids used during the experiments. Section 3
 81 presents the different regimes of motions that were observed. We close with
 82 section 4 and section 5 where results and their implications are discussed and
 83 summarized.

84 2. Experimental set-up and fluids

85 2.1. *The fluids*

86 According to the product company (Omiya Green Service Co., Ltd), the
87 fluid used is a superabsorbent polymer (polyacrylamide) made by copoly-
88 merization of acrylic acid and acrylamide. It is part of the family of aqueous
89 dispersions of superabsorbent polymers (SAPs), polymeric materials with
90 large capacity of water absorption [30, 31] which are used in many differ-
91 ent water absorbing applications in agriculture [32], in health care industry
92 (e.g. in sanitary pads and baby diapers), in sealants and in air-fresheners
93 [33, 34]. The dry material is a white and granular powder of particles with
94 different irregular shapes. In water, the original small particles can swell up
95 to 200 times and form gel grains whose size can be controlled by controlling
96 the size of the initially dry grains. Beside the general behaviour of such
97 materials that combines viscoelastic [31] and shear-thinning aspects [35], the
98 measurement of appropriate rheological properties at the swollen state is still
99 quite complicated as typical commercial SAPs have large (up to few millime-
100 ters) and irregular swollen particles that render classical rheological methods
101 difficult to use [31].

102 Starting from the provided raw material as the largest grains size end-
103 member, we gradually ground the dry particles to form four thinner powders.
104 All the final gel samples were then obtained by letting 17.5 ± 0.1 g of dry
105 product react with 3.5 l of distilled water for one night. The resulting swollen
106 mixture have been gently stirred (100 rpm) for 2-3 days with an electric stirrer
107 in order to remove air bubbles and to homogenize the mixture. Careful weight
108 measurements of a given volume of the final aqueous suspension before and

Figure 1: (a) Experimental set up. The cylindrical vessel containing the fluid is 50 cm high (H) and 10 cm wide (D). Videos are recorded in the tube section between the two black dotted lines (h). (b) Black and white image of the fluid structure illuminated by a laser sheet. The gel grains are in dark grey and the thin water film around them is in light grey.

109 after it was completely drained from the free water allowed to estimate a
 110 particle volume fraction of $62 \pm 2\%$.

111 As it was for the starting dry powders, the five final fluids under scrutiny
 112 differ from each other only in the size of the grains that make up their
 113 structure. To obtain the grains size distribution of the swollen gel samples,
 114 we analyzed several high resolution images (4288×2848 pixels) of a laser-
 115 illuminated vertical cross-section of the experimental tank (Fig.1) by using
 116 Multispec, an image analysis software [36]. The sample was stirred between
 117 each image, so that each image shows a different organization of the fluid

name	Mean diameter (mm)	std (mm)	Median (mm)	Max diameter (mm)
Gel A	1.4	0.3	1.4	2.3
Gel B	2.1	0.5	2.1	3.5
Gel C	3.3	0.9	3.0	5.8
Gel D	4.8	1.2	4.5	9.5
Gel E	5.7	1.2	5.5	9.6

Table 1: Results from the imaging analysis of our five gel samples. Here we report the mean diameter of the grains, the standard deviation (std), the median and the maximum diameter. The grain equivalent diameter (d_g) of a specific gel sample refers to the mean grain size value.

118 structure. Fig.1b shows an example of how the fluid structure looks like.
119 The arrangement of the grains is easily recognizable and the sampling of
120 each of them during the images analysis can be automatically or manually
121 done directly from black and white images without needing more complex
122 adjustments. Once the outline of the grains is defined from the image, the
123 grain surface comes by counting the number of pixels of which it is composed.
124 We describe each grain by the equivalent diameter of a circle with the same
125 area as the measured surface. The final grains size distributions are reported
126 in Fig.2 where data are normalized to a probability density function and in
127 Table 1 where the characteristics of the distributions are listed.

Figure 2: Grains size distributions for the five gels used in the experiments. Histogram representation of grains equivalent diameters with a normal distribution curve on top for gel A (a), gel B (b), gel C (c), gel D (d) and gel E (e). (f) All gels together.

128 2.2. Setup

129 All the spheres are released in a 50 cm deep (H) and 10 cm wide (D)
130 cylindrical Plexiglas vessel, previously filled with approximately 3.5 liters of
131 fluid (Fig.1a). In order to reduce optical distortions during the recording
132 of images illuminated laterally by a laser sheet, the vessel was placed in a
133 rectangular tank filled with water. A vertical cross-section of the tank is
134 then illuminated by a 532 nm laser sheet, and images are recorded using a
135 video camera (Canon Legria HF S21 1080p) with frame rate of 25 frames/s.
136 The position of the center of the sphere on each frame was determined by
137 plotting the spatio-temporal evolution of a vertical pixel line centered on the
138 sphere, or by using the blob analysis method in Matlab [37] (fig.4 and 5a).
139 Both methods allow the derivation of the local vertical velocity of the sphere
140 through time. And the blob analysis also provides the measurements of the
141 horizontal position and velocity of the sphere.

142 We run experiments with spheres of different materials and sizes. Their
143 properties (i.e. density and diameter) are summarized in Table 2. All the
144 steel spheres (density of 7980 and 7970 kg/m³) have been painted with black
145 spray paint to avoid strong reflections, while the other spheres have been left
146 with their original surfaces. The use of spray paint could have increased the
147 surface roughness that in turn can affect the velocity field in the fluid around
148 the object as well as the shape of the yielded region [38]. However, we did
149 not observe any discrepancies between the painted and the bare spheres.

150 Before each experiment, we stirred gently the fluid to remove any possible
151 preferential path that might have been formed during previously runs. SAP,
152 in fact, shows a quite strong hysteresis where the fluid structure does not

ρ_s (kg/m^3)	d_s (mm)
14952	6.00
7980	6.00; 12.00; 14.00; 16.00; 18.00
7970	3.00; 4.00; 5.00; 7.00; 8.00; 10.00; 20.00; 22.00; 25.00
7799	6.00
7782	14.00
7621	8.00
7519	3.20
3227	6.00
2200	15.66; 15.81

Table 2: Spheres density ρ_s and diameter d_s .

153 recover completely its original state after it has been deformed. This aspect
154 is observed when a second sphere is dropped from the same position as the
155 previous one without stirring the fluid in between the two runs. For identical
156 spheres, we have always measured higher velocities for those released later
157 and falling through an already deformed path. Stirring the fluid between runs
158 prevents this issue and allowed us to get reproducible velocities for identical
159 spheres.

160 3. Experimental results

161 As first step of our investigation, it is useful to test the reproducibility
162 and the accuracy of the experimental techniques and possible limits arising
163 from the dimension of our setup. To do so, we measured the terminal ve-

164 locities (v_y) of spheres settling in a Newtonian fluid at $T_c=15.2\pm0.3$ °C. The
 165 fluid is glucose syrup (Glucor 60/80) with $\rho=1394.66$ kg/m³ and $\eta=12.3\pm0.7$
 166 Pa.s, density and viscosity at T_c , respectively. Spherical objects falling in a
 167 Newtonian fluid at low Reynolds number reach a terminal velocity given by
 168 the Stokes velocity

$$v_{Stokes} = \frac{2}{9} \frac{R^2(\rho_s - \rho_{fluid})}{\eta} g, \quad (1)$$

169 where R is the radius of the spherical object and g the acceleration due to
 170 gravity. In Fig.3 we plot the velocities measured for spheres of same density
 171 and diameters in between 3 and 18 mm. Larger spheres are subjected to wall
 172 effects and in this case the sphere terminal velocity can be corrected by a
 173 coefficient that writes as

$$K_1^{-1} = 1 + \alpha_1 \left(\frac{d_s}{D}\right) + \alpha_2 \left(\frac{d_s}{D}\right)^3 + \alpha_3 \left(\frac{d_s}{D}\right)^5, \quad (2)$$

174 where $\alpha_1 = -2.104443$, $\alpha_2 = 2.08877$, $\alpha_3 = -0.94813$ and D is the tube
 175 diameter [39]. Experiments carried out in syrup show that the measured
 176 settling velocities are in good agreement with relative corrected Stokes ve-
 177 locities, that is $v_y/(v_{Stokes} K_1^{-1}) = 1$, ensuring that our measurement system
 178 is accurate (Fig.3).

179 We now move to the case where the fluid through which spheres fall is one
 180 SAP sample described in the previous section. In Fig.4 we report a selection
 181 of falling profiles in gel C (grains mean diameter $d_g=3.3$ mm) for four falling
 182 spheres with same density but different diameters. The same type of plot
 183 is also showed in Fig.5a where this time we track spheres having same size

Figure 3: Measured terminal velocities, v_y , in syrup (circles) and Stokes velocities, v_{Stokes} , (crosses and dashed line) as function of spheres squared radius, R_s^2 . Uncertainties for Stokes velocities come from the uncertainties in determining the syrup viscosity whereas the error for v_y is 0.25 mm/s, smaller than the symbols. Star symbols are the ideal velocities considering the wall correction, eq. (2).

Figure 4: Depth of spheres settling through gel C as function of time. Spheres have constant density ($\Delta\rho=6980\pm10$ kg/m³). Symbols refer to the diameter of the sphere: $d_s=10$ mm ($\blacktriangle$), $d_s=8$ mm ($\bullet$), $d_s=7$ mm ($\blacksquare$), $d_s=4$ mm ($\blacktriangledown$). Varying sphere's diameter, one can recognize the following motion regimes: linear regime (upward-pointing triangles), irregular motion superimposed to a linear trend (circles), logarithmic regime (squares) and no-motion (downward-pointing triangles). Regimes (1), (2), (4) and (5) in the text, respectively. The insert shows the distance as function of time for regime (4) with a semi-log scale. The color line is the fit $y(x)=a\log(bx)$, where $a=98.34$ and $b=0.66$. The correlation coefficient is $R=0.980$.

184 but different densities settling in gel D ($d_g=4.8$ mm). In both figures, the
185 starting position along y-axis, $h=0$, does not refer to the top fluid surface
186 but to the upper limit of the camera view at which movies were recorded
187 (Fig.1a). This is a consequence of the fact that all movies have been taken
188 for a magnified portion of tube $h=300$ mm, starting from 70 mm under the
189 surface. Camera position and therefore size of the view are kept constant for
190 each run done in the same fluid.

Figure 5: (a) Depth of spheres settling through gel D as function of time. Here spheres have constant diameter ($d_s=6 \text{ mm}$) but different densities. As in Fig.4, we report the case of linear regime (upward-pointing triangles), no-motion (downward-pointing triangles) and, in addition, the intermittent regime (circles). (b) Sequence of snapshots every 100 frames showing the irregular motion in gel D for a sphere with $d_s=7 \text{ mm}$ and $\rho_s=7970 \text{ kg/m}^3$.

191 For each gel, depending on the diameter and density of the sphere, we
192 distinguish the following five regimes (Fig.4 and Fig.5). (1) A linear regime:
193 rapid and linear fall in which spheres reach a constant terminal velocity; (2)
194 an irregular regime superimposed to a linear one where spheres never stop
195 during their way down ($v_y > 0$) but their local velocity fluctuates around
196 a constant value; (3) an intermittent regime: another irregular regime but
197 here periods of no-motion, where the sphere's vertical velocity goes to zero,
198 and periods of irregular falls follow one another; (4) a logarithmic regime
199 where the sphere's speed progressively decreases (inset in Fig.4); and (5)
200 a no-motion regime in which spheres do not move at all from their initial
201 positions.

202 Regimes (1) and (5) are characteristic of a yield stress fluid [11, 26, 40]
203 and one can interpret them based on the state of the medium that surrounds
204 the object. In the linear regime (1) the fluid around the sphere has a liquid-
205 like behaviour that allows spheres to go down, whereas in regime (5) local
206 stresses are below the yield stress (σ_Y) and the material around the sphere
207 remains in the solid state, leading to the no motion of the object. **Here the**
208 **stress field is generated by the weight of the sphere minus the buoyancy** and
209 the no-motion regime is always observed for smallest diameters d_s (Fig.4) or
210 density contrast $\Delta\rho$ (Fig.5a, as in [11]). We have had no-motion whether the
211 sphere was placed on the surface of the fluid column, or it was gently pushed
212 within it and released deeper in the fluid. In both cases, we left the spheres
213 in that position for days (up to a week for one run) and they never moved
214 away from their starting positions.

215 Although the distinction between the two end-member cases (1) and (5)

216 is easy to establish, to discern quantitatively between the first steady-state
 217 motion regime (1) and the more irregular ones is more delicate. We sum-
 218 marize how we define the different motion regimes in Table 3. From the
 219 falling profiles one can notice that, although sometimes it is still possible to
 220 identify an almost constant falling speed superimposed on fluctuations (cir-
 221 cle symbols both in Fig.4 and Fig.5a), in some other cases, spheres seem to
 222 slow down logarithmically during their way to the bottom (square symbols
 223 in Fig.4), making impossible the determination of a constant terminal veloc-
 224 ity. We report this aspect in Fig.6a by plotting the mean squared variations
 225 (MSE) obtained by fitting the falling profiles with a linear or a semilog fit
 226 as function of spheres buoyancy. The logarithmic regime (4) is defined for
 227 $MSE_{linear} > MSE_{log}$, that is when falls are better represented with a logarith-
 228 mic fit. Hence, this regime describes a continuous decrease in sphere's speed
 229 which never reaches the steady-state. Although in the case of Carbopol [11]
 230 velocity decreases following a power law, in most of our cases it best fits
 231 a logarithmic curve. Although from Fig.6a is evident that the logarithmic
 232 behaviour appears more often at lower buoyancies, the distinction between
 233 linear and non-linear regime is not straightforward. In a few cases, indeed,
 234 at same buoyancy the two behaviours coexist.

235 Once the separation between stoppage cases (logarithmic regime (4) and
 236 no-motion (5)) and the other cases is established, one can identify and rec-
 237 ognize diversities between the steady-state motion (regime (1)) and the more
 238 irregular and chaotic regimes (2) and (3). Depending on the buoyancy of
 239 the object and hence on its velocity, the irregularities show up as fluctua-
 240 tions in the sphere local vertical velocity and in a progressive increase of its

Figure 6: Determination of the different regimes of motion. In (a), (c) and (e) data refer to experiments carried out in gel E. In (b), (d) and (f) data are for all fluids and colours indicate the grain size as in Fig.2. (a) best fit (MSE_{linear}/MSE_{log}) of the falling profiles. ○ indicates a better linear fit whereas ◇ a better semilog fit (logarithmic regime (4), in the text). (b) mean falling velocities ($\bar{v}_y$) for those falling profiles that have been linearly fitted. The error for v_y is 0.25 mm/s, smaller than the symbols. We do not report the standard deviation for v_y here (whereas it is in (c) and (d)) as being also linked to the physical fluctuation of the sphere, for small values of velocity it would make the plot unreadable. In (c) and (d) we separate those experiments that show a more irregular and chaotic motion (□) from those that do not show any irregularities during the fall (○) by plotting $std(v_y)/\bar{v}_y$. The dashed line is for $std(v_y)/\bar{v}_y = 0.15$, from the Newtonian case. In (e) and (f) we separate the intermittent regime (3) (×) from the other irregular regime (2) (□) based on the amount of time t_s the sphere remains at $v_y \leq err(v_y)$. t_0 is the total time the sphere takes to descend h . The dashed line indicates $t_s/t_0=5$.

241 horizontal motion (Fig.5b). Regimes (2) and (3) show some similarities: in
 242 both of them, spheres have a long term constant falling velocity on which
 243 is superimposed a more complex and irregular pattern of fluctuations. The
 244 latter are characterized by accelerations and subsequent decelerations in a
 245 short period of time and, in some cases, they can result in a succession of
 246 stops of the sphere. In the most drastic cases, we observe the complete arrest
 247 of the sphere after it has passed through most of the fluid (Fig.5a).

248 To determine the boundaries between the irregular regimes (2) and (3)
 249 and regime (1), one can look, as a first approximation, at the ratio between
 250 the standard deviation (*std*) of the local falling velocity and its mean value.
 251 Variations of the local velocity for spheres in irregular regimes are much
 252 larger than the experimental uncertainty (Fig.6c-d). The latter is due to
 253 the precision of the technique we used to detect the sphere position in time
 254 (hereafter $err(v_y)$). It is estimated to be 0.25 mm/s from the syrup-case mea-
 255 surements and from local velocity measurements in the no motion regime. In
 256 the Newtonian case, $std(v_y)/\bar{v}_y$ always remains smaller than 0.15. We define
 257 the linear regime (1) using this value, that is for $std(v_y)/\bar{v}_y < 0.15$ (dashed
 258 line in Fig.6c-d). This limit represents therefore a boundary between the
 259 steady-state motion and what shows a more spread distribution of velocities.

260 Once the linear regime is bounded in this way, we distinguish between
 261 the irregular regime (2) and the intermittent regime (3) as follows. In the
 262 intermittent case the sphere remains during a certain cumulative amount of
 263 time, t_s , at $v_y \leq err(v_y)$. We then compare t_s to the total time, t_0 , the sphere
 264 takes to descend 30 cm. The intermittent regime is defined as $t_s/t_0 \geq 5\%$.
 265 The remaining irregular regime (2) is therefore defined as $t_s/t_0 < 5\%$ and

	Regimes				
	(1)	(2)	(3)	(4)	(5)
$\bar{v}_y = 0 \pm err(v_y)$					x
$MSE_{lin} > MSE_{log}$				x	
$MSE_{lin} < MSE_{log} \ \& \ std(v_y)/\bar{v}_y < 0.15$		x			
$MSE_{lin} < MSE_{log} \ \& \ std(v_y)/\bar{v}_y \geq 0.15$			x	x	
$MSE_{lin} < MSE_{log} \ \& \ t_s/t_0 \geq 5\%$				x	

Table 3: Summary of the observed motion regimes and the way they are classified

266 $std(v_y)/\bar{v}_y > 0.15$ (Fig.6e-f).

267 Fluctuations, accelerations and subsequent decelerations and the result-
268 ing general complex set of behaviours of the moving sphere, characterize the
269 experiments both in the direction of fall and in the one perpendicular to it.
270 In fact, the irregular motion we have observed is not only present and visible
271 in the vertical direction but it also affects the horizontal motion of spheres in
272 a such a way that, along this direction, they collect a certain amount of trav-
273 eled distance (L_x) with respect to their initial position, depending on their
274 diameter (Fig.7a), on the gel grains size (Fig.7b), and on the vertical falling
275 distance h (Fig. 8a). Fig. 8b shows that for each run, L_x is roughly propor-
276 tional to h . Fig.7 shows that small spheres change frequently their horizontal
277 position, oscillating continuously around the vertical axis, and traveling for
278 larger distances L_x than the large spheres. If we normalize L_x by h and d_s

Figure 7: Total distance traveled in the horizontal direction (L_x). (a) L_x as function of the sphere diameter d_s . Dashed line indicates the uncertainty in position. (b) Same plot with y-axis normalized by the vertical falling distance h and x-axis by the gel grain size d_g . Colours indicate the different fluids as indicated in the legend. Symbols refer to the different motion regimes: ○ for the linear regime (1); □ for an irregular regime superimposed to a linear one (2); × for the intermittent regime (3) and ◇ for the logarithmic regime (4). Empty symbols are for steel spheres while the symbol with a point inside refers to $\rho_s = 7799 \text{ kg/m}^3$ and those filled with a plus sign are for $\rho_s = 14952 \text{ kg/m}^3$.

by the diameter of the gel grains (d_g), we achieve a good collapse for all our data along a line with slope -2. That is

$$L_x/h = C_{exp} (d_s/d_g)^{-2}. \quad (3)$$

where $C_{exp} = 0.49 \pm 0.26$. In this way the total travelled distance in horizontal direction turns to be a good candidate to underline the strong interaction of the sphere and the gel grains that make up the fluid structure when those two have similar sizes.

Figure 8: Distance traveled in the horizontal direction in gel C. (a) Distance traveled in different 15-cm-vertical-sections of h , $L_x[Sect]$, as function of spheres buoyancy. Sections are $Sect_A=[h=0 \text{ cm}, h=15 \text{ cm}]$; $Sect_B=[h=15 \text{ cm}, h=30 \text{ cm}]$; $Sect_C=[h=7.5 \text{ cm}, h=22.5 \text{ cm}]$. (b) Ratio between the total distance traveled in 30 cm, L_x , and the distance travelled in 15 cm, $L_x[Sect]$.

4. Discussion

4.1. Determination of the SAP effective rheology

The fall of a spherical object within certain boundary conditions is described by the combination of a proper set of equations of motion and the constitutive relation of the fluid through which the sphere is falling. For yield stress fluids a commonly used rheological model that relates stress and strain in a non-linear way is the Herschel-Bulkley model [2, 12, 41]:

$$\begin{cases} \sigma = \sigma_Y + K_v \dot{\gamma}^n & \text{if } \sigma > \sigma_Y \\ \dot{\gamma} = 0 & \text{if } \sigma \leq \sigma_Y \end{cases} \quad (4)$$

where σ is the stress, σ_Y the yield stress, $\dot{\gamma}$ the shear rate, K_v the consistency and n the shear-thinning index. Considering a fluid which flows following

295 such model, for small Reynolds numbers ($Re \ll 1$), the problem of a settling
 296 particle needs two parameters to be characterized [11, 42]. They are the
 297 Bingham number

$$Bi = \frac{\sigma_Y}{K_v (v_y/d_s)^n}, \quad (5)$$

298 that compares yield stress with viscous stresses, and the yield number

$$Y = \frac{3\sigma_Y}{gd_s \Delta\rho} \quad (6)$$

299 that compares the yield stress with the buoyancy stress.

300 As described in section 1, most of the experimental work done to study
 301 this problem is carried out by using Carbopol. The main difference between
 302 experiments done with Carbopol and the present work regards the typical
 303 size of the fluid structure compared to the dimension of the object that
 304 is moving through it. Carbopol corresponds to the case where the fluid
 305 structure (grains) is much smaller than the spheres. In this case, only regimes
 306 (1), (4) and (5) were reported (e.g. [11]) while neither the irregular regime
 307 (our regime (2)) nor the intermittent regime (3) were reported; and the fall
 308 of the sphere is always reported to be free of any sort of chaotic motion
 309 [11, 17, 38].

310 In regime (1), that is when a constant terminal velocity can be deter-
 311 mined, Tabuteau *et al.* [11] showed that their data are well-fitted by the
 312 following equation

$$\frac{1}{Y} = 7 + \frac{8.52}{Bi} \quad (7)$$

313 with $n=0.5$, and that there is a critical value of Y , $Y_c=0.145$, above which
 314 there is no motion.

315 Taking into account the differences between SAP and Carbopol, one can
 316 use eq.(7), defined for $d_s \gg d_g$, and check whether it applies for a fluid with
 317 a much larger structure. Thus, assuming a framework in which eq.(7) is valid
 318 also in our case, we can rewrite it as

$$\frac{gd_s\Delta\rho}{3} = 7\sigma_{Y(eff)} + 8.52K_{v(eff)} \left(\frac{v_y}{d_s} \right)^n, \quad (8)$$

319 where $\sigma_{Y(eff)}$ and $K_{v(eff)}$ represent the effective yield stress and consis-
 320 tency of the SAP through which the sphere is falling. In eq.(8), v_y is the
 321 average falling velocity defined for those experiments which do not show a
 322 decrease in speed, that is those in regime (1), (2) and (3). By plotting the
 323 buoyancy stress ($gd_s\Delta\rho$) as function of the effective strain rate, i.e. $(v_y/d_s)^n$,
 324 Fig. 9 shows for SAP the same linear relationship showed for Caropol [11].
 325 Therefore the slope of the linear fit is related to an effective consistency
 326 through

$$K_{v(eff)} = \left[\frac{gd_s\Delta\rho}{25.56} - 0.86\sigma_{Y(eff)} \right] \left(\frac{v_y}{d_s} \right)^{-n}. \quad (9)$$

327 and, similarly, an effective yield stress is related to the intercept at zero
 328 effective strain rate through

$$\sigma_{Y(eff)} = \frac{gd_s\Delta\rho}{21}. \quad (10)$$

329 In this way, by using the falling sphere system as reometer we can define
 330 an effective rheology, which has to have same form of eq.(7), for each of our
 331 gel sample. Fits in Fig.9 depend on the shear thinning exponent n used to

332 calculate the effective shear rate. Minimum mean squared values for linear fit
 333 are found for n in between 0.5 and 0.7 (Fig.11). Unless otherwise indicated,
 334 in what follows we will use $n=0.5$ since it is with this value that eq. (7)
 335 is defined. Interesting, experiments in both regime (1), (2) and (3) can be
 336 linear fitted by the same line. Moreover, effective yield stress and consistency
 337 increase with the gel grains size (Fig.10).

338 For experiments involving large spheres, it is opportune to check whether
 339 wall corrections are needed or not. For sufficiently small sphere-to-tube di-
 340 ameter ratio, d_s/D , we do expect no reductions of sphere's terminal velocity.
 341 This means that the measured velocity, v_y , is equal to the ideal terminal
 342 velocity of a sphere falling in an unbounded medium, v_∞ . On the other
 343 hand, if d_s/D exceeds a critical value, $(d_s/D)_{crit}$, velocity decreases leading
 344 to $v_y/v_\infty < 1$. The critical diameter ratio for a yield stress fluid writes as [43]

$$(d_s/D)_{crit} = 0.055 + 1.114 Y \quad \text{if } Y \leq Y_c. \quad (11)$$

345 In our case, d_s/D is smaller than $(d_s/D)_{crit}$ as long as $d_s/D < 0.1$. The
 346 influence of walls causes a reduction in speed such that [43]

$$\frac{v_y}{v_\infty} = 1 - 1.7 [(d_s/D) - (d_s/D)_{crit}] \quad \text{if } d_s/D > (d_s/D)_{crit}. \quad (12)$$

347 For $0.1 \leq d_s/D \leq 0.17$ experiments are above critical conditions and, ac-
 348 cording to eq. (12), wall effects lead to $0.8 < v_y/v_\infty < 1$, that is a maximum
 349 decrease of v_y/v_∞ of less than 20%.

350 Considering only the experiments with $v_y/v_\infty > 0.8$ and ignoring the
 351 others, we recalculate the effective rheology in the same way described above

Figure 9: **Sphere buoyancy as function of effective shear rate, $(v_y/d_s)^n$** . Following Tabuteau *et al.* [11] approach, v_y is the average falling velocity for those experiments where a steady decrease of the speed has not been observed (i.e. we do not consider regime (4)) and $n=0.5$ the power-law index. Colours represent the fluids (same as Fig.7) while symbols the regimes (circles for regime (1), squares for regime (2) and crosses for regime (3)). Empty symbols for steel sphere; symbol with a point inside for $\rho_s=7799$ kg/m³; with a plus sign for $\rho_s=14952$ kg/m³ and filled symbols for $\rho_s=2200$ kg/m³. Solid lines are the linear data fits.

Figure 10: Effective yield stress (σ_Y) and effective consistency (K_v) as function of gel grains size.

Figure 11: Power law index (n) that best linearly fits data in Fig.9 for all gel samples. y -axis represents the mean squared errors (MSE) of the linear fit normalized by the maximum MSE found. The best range of n is in between 0.50 and 0.70.

Figure 12: Influence of the velocity reduction due to wall effects on the effective rheology. $n=0.5$. (a) The new fit that considers only experiments with $v_y/v_\infty > 0.8$ (solid lines) is compared with the previous fit from Fig.9 (dashed lines). Colours and symbols here are those described in Fig.9. (b) Effective yield stress and consistency as function of gel grains size from the new fit (blue symbols). Red symbols are those in Fig.10.

(Fig.12a). The new values of $\sigma_{Y(eff)}$ and $K_{v(eff)}$ do not differ much from the previous ones (Fig.12b); which means that our set up is big enough.

Once we got the yield stress and the consistency of each sample, we plot Y^{-1} as function of Bi^{-1} in Fig.13. For values of $Bi^{-1} > 0$ we achieve a good collapse along eq.(7) for experimental data that belong to regimes (1), (2) and (3). In this way, even considering the irregular regime (2) and the intermittent (3) where the interaction between moving objects and fluid structure results in highly irregular falls of objects themselves, an effective

360 rheology following the Hershel-Bulkley model with a classical shear-thinning
 361 index of 0.5 well represents the majority of our data.

362 4.2. Entrapment conditions

363 Fig.13 also shows that for $Bi^{-1}=0$ spheres get entrapped also for $Y^{-1} >$
 364 Y_c^{-1} . From the experimental work of Tabuteau *et al.* [11], the critical value
 365 of the yield number (Y_c) above which spheres do not move in a simple yield
 366 stress fluid is 0.145. This value is very close to 0.143, predicted in previous
 367 works of Beris *et al.* [10] and Blackery and Mitsoulis [14]. On the other hand,
 368 a more recent study [13] points out how the critical value may increase as
 369 function of the Deborah number if elastic effects are taken into account. In
 370 our case, however, we observe sphere arrest also for $Y < Y_c$. For example,
 371 we observed arrest even for $Y = 0.042 \approx Y_c/3$ (4 mm steel sphere).

372 As we highlighted at the beginning of this section, equation (7) was de-
 373 rived for Carbopol when the size of the moving object is much larger than the
 374 size of the particles than make up the fluid structure. Inside this assumption,
 375 the critical condition is well predicted in our case too. In gel E, for example,
 376 we had no-motion for a large glass sphere ($d_s/d_g > 2.5$ and $\Delta\rho=1200 \text{ kg/m}^3$)
 377 for $Y=0.478$ (Fig.14), greater than $Y_c=0.145$. It is only when the diameter
 378 of the sphere becomes comparable to the diameter of the gel particles that Y_c
 379 moves to lower values and the stoppage regime (5) cannot be anticipated any
 380 more from eq.(7). For the same gel E, a smaller steel sphere ($d_s/d_g < 2$ and
 381 $\Delta\rho=6970 \text{ kg/m}^3$) is in stoppage for $Y=0.13 < Y_c$. We attribute this to the
 382 interaction between spheres and the gel structure: when $d_s \rightarrow d_g$, the spheres
 383 "see" obstacles on their way and the effective rheology breaks down.

Figure 13: $1/Y$ as function of $1/Bi$. Colours are for the different fluids. The dashed line indicates $Y_c^{-1}=0.145^{-1}$ [11]. Symbols are filled in a different way to distinguish between spheres of different materials: full-filled symbols are for light spheres ($\rho_s = 2200 \text{ kg/m}^3$), open-symbols for $\rho_s = 7970 \text{ kg/m}^3$, point-filled for $\rho_s = 7799 \text{ kg/m}^3$, asterisk-filled for $\rho_s = 3227 \text{ kg/m}^3$ and plus-filled for $\rho_s = 14952 \text{ kg/m}^3$. We show the good collapse of our data along eq.(7) (solid black line). The inset shows the data zoomed around the critical conditions in order to better display the entrapment conditions.

Figure 14: Range of validity of the effective rheology. Colours are for the different fluids. Symbols are those used in Fig.13. The dashed line indicates $Y_c^{-1}=0.145^{-1}$ [11]. For d_s/d_g approaching the value of 2, we have no motions (triangles), even for yield numbers lower than the critical one commonly used.

384 4.3. Grains rearrangement and elasticity

385 The emergence of a yield stress in SAP can be related to the jamming of
386 soft grains. Gel grains act as athermal elastic particles that interact by gliding
387 and compressing each other. The collective behaviour of a fluid made up by
388 these elastic grains or particles can show elasto-viscoplastic responses [5, 44].
389 Therefore, one can expect that also the observed irregular motion regimes
390 can be linked to the jamming nature of the fluid. Within this framework,
391 going from the linear to the intermittent regime by decreasing the sphere
392 size makes the elasticity and the yield stress seen by the sphere no longer
393 the collective one of the bulk material, but rather the one of few gel grains.
394 Once this occurs, to allow the continuous fall of the sphere, grains in front
395 of it must undergo rearrangement. The mechanics of the flow appears then
396 similar to the one of soft glassy materials. In this case the flow occurs through
397 a sequence of elastic (reversible) deformations and local plastic (irreversible)
398 rearrangement events which induce a long ranged relaxation of stress [6,
399 45, 46]. Grains rearrangements will be all the more common the higher
400 the local shear stresses are (e.g. in the case of a larger sphere). Since the
401 number of plastic events for unit of time controls fluid flow and relaxation
402 [6], a sufficient high number of them leads the system to be more fluid. The
403 resulting sphere motion would be in the linear regime (1); whereas irregular
404 and intermittent regimes might be the result of a less fluid system where
405 the local rheology seen by the sphere becomes more relevant. In this way,
406 the observed deformation induced by the sphere falling in the linear regime
407 is mainly the elastic interaction between the grains, as it is for flow of soft
408 particle glasses [5].

409 Fluid elasticity have been also linked to a peculiar characteristic of the
 410 fluid flow around the sphere, namely its asymmetry. The rise of a fore-arc
 411 asymmetry and the formation of a negative wake at the sphere's tail has
 412 been detected experimentally in Carbopol [e.g. 17, 38] and associated with
 413 fluid elasticity [13, 17] or the emergence of thixotropy [18]. In SAP, we
 414 have observed a fore-arc asymmetry in the velocity field collected from PIV
 415 measurements. Indeed, Fig.15 shows that a negative wake in the region at
 416 the sphere back can be recognized at least for the linear motion regime (1).
 417 Although to discern the cause that breaks the fore-arc symmetry in SAP is
 418 not straightforward, its formation is more probably due to elasticity. In fact,
 419 between SAP grains there are not polymer chains that form system-spanning
 420 networks and the fluid, during a single experiment, does not undergo aging.
 421 More details about will be given in a coming work where the fluid velocity
 422 field will be properly analyzed and explained.

423 Moreover, a further analysis on how the properties of a single elastic grain
 424 could influence $\sigma_{Y(eff)}$ and $K_{v(eff)}$ might be relevant to evaluate the role that
 425 its viscoelasticity plays for the bulk rheology. However, the extreme softness
 426 of the material makes it very difficult. After several attempts to measure
 427 forces and penetration depth by using a microindenter with a minimum nor-
 428 mal force of 20 mN, we have not been able to get any reliable measurements
 429 since slip or rupture of the grain always occurred even for such small values
 430 of normal force.

Figure 15: Fluid flow from PIV measurements. We display the vertical component of the right half part of instant velocity field (v_z) for a sphere ($d_s=19$ mm) falling in the linear regime (1) through SAP.

431 5. Conclusion

432 In this work we have experimentally investigated the influence that the
433 size of particles which build up the structure of an yield stress fluid has on
434 the dynamics of a spherical intruder falling under gravity through it. As the
435 typical size of the gel grains was varied between 1 and 6 mm and the sphere
436 diameter was varied between 3 and 25 mm, there exists a range where they
437 become comparable.

438 We have been able to expand from three (in Carbopol) to five (in SAP)
439 the observed regimes of motion of the sphere: (1) a linear regime where the
440 sphere has a steady-state motion; (2) an irregular regime superimposed to a
441 linear one where spheres never stop during their way down but their velocity
442 varies; (3) an intermittent regime in which periods of no-motion and periods
443 of irregular falls follow one another; (4) a logarithmic regime where the sphere
444 velocity progressively decreases; (5) no-motion. Beside the classical steady-
445 state motion (1) and stoppage regimes (5), typical of viscoplastic fluids, the
446 interaction between moving objects and the fluid structure results then in
447 two additional regimes ((2) and (3)) where the motion becomes more chaotic.
448 The same strong interaction between the sphere and the gel grains is also
449 observed from the sphere total travelled distance in the horizontal direction.

450 Moreover, we have shown that the effective rheology of SAP follows the
451 Herschel-Bulkley model with a power index $n=0.50$, making this simple set-
452 up a good rheometer for this type of fluids.

453 We also found that both yield stress and consistency of the gel increase
454 with the gel grain size, suggesting that the effective rheology, due to the
455 material heterogeneous properties, depends on the size of the particles which

456 constitute the fluid structure. However, it is remarkable that the overall
457 sphere motion predicted by the Herschel-Bulkley model begins to "see" the
458 texture only for sphere diameters less than twice the SAP grain size.

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462 **References**

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