Imaging non-Brownian particle suspensions with X-ray tomography: application to the microstructure of Newtonian and visco-plastic suspensions

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1 **Imaging non-Brownian particle suspensions with X-ray**
2 **tomography:**
3 **application to the microstructure of Newtonian and visco-plastic**
4 **suspensions**

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Abstract

A key element in the understanding of the rheological behaviour of suspensions is their microstructure. Indeed, the spatial distribution of particles is known to depend on flow history in suspensions, which has an impact on their macroscopic properties. These micro-macro couplings appeal for the development of experimental tools allowing for the rheological characterization of a suspension and the imaging of particles. In this paper, we present the technique we developed to image in three dimensions the microstructure of suspensions of non-Brownian particles, using X-ray computed tomography and sub-voxel identification of particle centers. We also give examples of the information we can get in the case of Newtonian and visco-plastic suspensions, referring to Newtonian and visco-plastic suspending fluid. We compute three dimensional pair distribution functions and show that it is possible to get a nearly isotropic microstructure after mixing. Under shear, this microstructure becomes anisotropic in the shear plane (velocity-velocity gradient plane), whereas it is almost isotropic in the two other planes (velocity-vorticity and velocity gradient-vorticity planes). When changing the plane of shear (from a squeeze flow to a rotation flow), the microstructure reorganizes to follow this change of shear plane. It is known that for Newtonian suspensions the anisotropy is independent on the shear rate; we show here that for a visco-plastic suspension it depends on it. Finally, we study the structuration close to boundaries and we evidence some particle alignment along both solid surfaces and, more surprisingly, along free interfaces.

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I. INTRODUCTION

Many flowing materials encountered in nature (landslides, debris or lava flows, ...) or raw materials produced in industry (concrete, batter, ...) are mixtures of dispersed solids in a liquid. These systems are composed of a wide range of materials of various sizes, which makes them complex to understand. In order to predict the behaviour and optimize the use of these suspensions, a useful step is to identify model systems capable of reproducing the phenomenology of interest and to understand the rheology of controlled suspensions. When length scales between the coarser and the finer dispersed particles of a suspension are well separated, a possible model system is a visco-plastic suspension of solid particles, referring to a visco-plastic suspending fluid, i.e. of nonlinear viscosity with a yield stress. Indeed, from a rheological point of view, a fresh concrete can be considered as coarse particles (sand, gravel) suspended in a visco-plastic fluid (cement paste) [1, 2]. Even more simply, one can consider a Newtonian suspension of solid particles as a model system, referring to a Newtonian suspending fluid [3]. A lot can indeed be learnt from such materials: studying first the interactions of multiple particles in a linear (Newtonian) fluid is crucial to understand their interaction in a nonlinear medium [4, 5]. This knowledge can then be applied to materials closer to applications such as cementitious materials [2, 6].

A key element in the understanding of the rheological behaviour of suspensions is their microstructure [7, 8]. Indeed, the spatial distribution of particles in suspensions is known to depend on the flow history – an anisotropic distribution is induced by shear –, which has an impact on their properties [7, 9–14] – an anisotropic microstructure is at the origin of normal stress differences and of viscosity variations during transient regimes. These micro-macro couplings between microstructure and flow appeal for the development of experimental and numerical tools allowing for the characterization of the rheology of a suspension and the imaging of its microstructure. A lot of numerical studies used Stokesian dynamics equations [8, 15–18]. The existing experimental techniques efficient for non-Brownian suspensions are all based on optics. Non-Brownian particles are indeed too large to use scattering techniques such as X-ray scattering or coherent diffraction imaging. For non transparent particles in a transparent fluid, it is nevertheless possible to image the microstructure only near the free surface [10] or near a transparent boundary. As we will demonstrate here, the proximity of a boundary (a solid wall or a free interface) influences strongly the microstructure and makes

50 it different from the one in the bulk. The bulk microstructure can be obtained only for
 51 specific suspensions, where particles and the fluid have the same optical index; a laser sheet
 52 can then be used to light a plane embedding a fluorescent stain so as to visualize and identify
 53 particles in two dimensions (2D) [11, 12]. Obtaining a three dimensional microstructure
 54 then implies to displace the position of the laser sheet. Confocal microscopy can also be used
 55 for micron-sized particles [18, 19], submitted to Brownian diffusion, that is in competition
 56 with shear flow, quantified by the Peclet number (being the ratio of hydrodynamic over
 57 thermal forces). Here we choose to use non intrusive X-ray techniques as a promising tool
 58 to image in the bulk suspensions in three dimensions (3D).

59 In this paper, we present the technique we developed to image in 3D the microstructure
 60 of suspensions of non-Brownian particles, using X-ray computed tomography (CT) and then
 61 identifying by image analysis the particle centers with a sub-voxel resolution. All the details
 62 to duplicate our technique are given. We give examples of the information we can get in the
 63 case of Newtonian and visco-plastic suspensions. We compute 3D pair distribution functions
 64 (pdf) and show that it is possible to get a nearly isotropic microstructure after mixing. Under
 65 shear, this microstructure becomes anisotropic in the shear plane (velocity-velocity gradient
 66 plane), whereas it is nearly isotropic in the two other planes (velocity-vorticity and velocity
 67 gradient-vorticity planes). When changing the plane of shear (from a squeeze flow to a
 68 rotation flow), the microstructure reorganizes to follow this change of shear plane. While it
 69 is known that for Newtonian suspensions the anisotropy is independent on the shear rate, we
 70 show that for a visco-plastic suspension it depends on it. Finally, we study the structuration
 71 close to boundaries and we evidence particle alignment along both solid surfaces and more
 72 surprisingly along free interfaces.

73 In Sec II, we present the principles of our study and we provide the details on the studied
 74 suspensions and the set-up. Sec III is devoted to the X-ray tomography technique and to the
 75 method we developed to identify particles center and compute pair distribution functions.
 76 In Sec IV, we present various examples of the microstructure of Newtonian and visco-plastic
 77 suspensions.

FIG. 1. X-ray computed micro-tomography set-up (a and b) and dedicated parallel plates set-up (c). The sketch (a) shows the X-ray source (in white), the imaging device (in green), the rotation (in red) and translation (in blue) stages and the parallel plates set-up (in yellow).

II. MATERIALS AND METHODS

A. Goals and general principles

Our goal is to get enough information at the microstructure scale to guide developments of micro-mechanical models for complex suspensions rheology. Thus, we need to image particles in a flow cell and to characterize their spatial distribution for various flow histories.

Our system made of a suspension in a rheometrical set-up should allow for the investigation from small to large space scales, from the length of the dispersed particles to the whole suspension. The flow cell should be large enough to be representative of a macroscopic shear flow, to ensure that the material exhibits a bulk behaviour not influenced by finite size or non local effects [20], and to get enough statistics for studying the microstructure. This leads us to choose a rheometer equipped with a parallel plates geometry (Fig. 1c). In the same time as doing rheology, we want to image the microstructure in 3D, even in the case of non matched optical index materials. This leads us to use X-ray CT imaging, which is efficient for many suspensions and geometries, as long as there is enough X-ray absorption contrast between particles and the fluid.

The sample to image is placed on a rotating stage between an X-ray source and a 2D imaging device (Fig. 1), which monitors the transmission of X-rays through the sample. In our case, the imaging device was a PaxScan2520V flat panel detector from Varian, made of a CSI scintillator and a matrix of 1920x1536 pixels (among which 1840x1456 are effective), with a pixel width of 127 μm . This provides a 2D radiograph, accounting for the total

98 absorption along linear paths through the sample. A series of 2D radiographs for different
 99 rotation angles of the sample (typically over 360° in a lab X-ray CT device) then allows for
 100 the reconstruction of the 3D map of absorption coefficients [21–23]. If particles and the fluid
 101 do not have the same absorption coefficient, we can finally achieve to detect the particles.
 102 This map is discretized over elementary volumes called voxels (i.e. volumic pixels). The
 103 achievable minimum voxel size is given by the ratio of the size of the imaged sample over
 104 the number of pixels in the detector width. The typical width of the CCD detectors used
 105 for X-ray CT is about 2000 pixels. Noting D , the diameter of the parallel plates geometry,
 106 the minimum possible voxel size is $D/2000$. To identify precisely the particles center, their
 107 diameter d should be at least 10 voxels, i.e. $d \gtrsim D/200$. The gap size H between the plates
 108 in the parallel plates geometry should contain at least 10 particles to avoid some finite size
 109 effects, i.e. $H \gtrsim D/20$, which is compatible with the study here but limits the possible
 110 aspect ratios diameter/gap of the geometry to $D/H \lesssim 20$. We will show later that our
 111 home-made algorithms allow us to refine particle centers positions at a sub-voxel resolution
 112 of an order of $d/100$, that ultimately allows for a fine characterization of the microstructure.
 113 Here (Fig. 2a), the diameter of the parallel plates geometry is $D = 2\text{cm}$, the gap of the
 114 geometry is $H = 2\text{mm}$, the size of particles d is between $100\mu\text{m}$ and $200\mu\text{m}$, the size of
 115 voxels is $12\mu\text{m}$ and the final accuracy on the particles centers in most cases presented here
 116 is $6\mu\text{m}$, but it can be decreased down to $1\mu\text{m}$ if necessary.

117 X-ray CT imaging may demand a non negligible time for scanning. The time for one scan
 118 depends of the X-ray set-up, the suspension properties and the chosen spatial resolution (X-
 119 ray flux, number of radiographs, time exposure, ...). With our X-ray CT device and the
 120 chosen spatial resolution, the whole sample can be scanned in 20 minutes but with a poor
 121 signal-to-noise ratio. The results presented here are obtained from 1 hour-scan in order to
 122 enhance the image quality. During the time of a scan, particles must not move within the
 123 suspending fluid (i.e. displacement less than 1 voxel or even less, if subvoxel accuracy is
 124 expected). This leads us to use first a visco-plastic fluid as a suspending fluid ensuring that
 125 particles do not move as long as stresses are smaller than the yield stress. Then we apply to
 126 our suspension a given shear history, then stop the flow and image the material at rest with
 127 a frozen structure, which is supposed to be close to the structure under flow. The underlying
 128 hypothesis is that relaxation of the microstructure at rest is negligible. This flow-arresting
 129 technique was already used before for Brownian suspensions [18]. In a second time, we

FIG. 2. (a) Scheme of the parallel plates set-up: D and H are the plates diameter and gap, d is the particles diameter, Ω and V are the rotational and translational velocities of the top plate. (b) Schemes of the rotational and squeeze shear flows: the suspension velocity $u_\theta(r, z)$ and $u_r(r, z)$ are aligned with the azimuthal and radial directions respectively. (c) A picture processed from X-ray slices showing the rough surface with grooves of the set-up's plates.

130 succeed to use a Newtonian suspension with density-matched particles and fluid, to avoid
 131 any buoyancy effects at rest. In principle, various types of suspensions may be imaged and
 132 studied with our technique, as soon as particles or suspended objects do not move during the
 133 time of a X-ray scan. For particles as small as a few μm , Brownian fluctuations may call for
 134 even quicker X-ray micro-tomograph (e.g. Synchrotron source). In terms of concentration
 135 of particles, we have successfully image in 3D suspensions at solid fractions as high as 50%
 136 and it should be possible to go even higher.

TABLE I. Properties of particles and fluids used for the Newtonian and the visco-plastic suspensions: average diameter d of particles, viscosity η of the Newtonian fluid, elastic shear modulus G' , yield stress τ_y and consistency K of the visco-plastic fluid and density ρ of the fluid and density-matched particles.

PMMA	Newtonian	Density	PS	Yield stress			Density
$d (\mu\text{m})$	$\eta (\text{Pa.s})$	ρ	$d (\mu\text{m})$	$G' (\text{Pa})$	$\tau_y (\text{Pa})$	$K (\text{Pa.s}^{0.5})$	ρ
170 ± 12	1.3	1.19	138 ± 8	250	26	5.1	1.05

B. Suspensions of non-Brownian particles

Two suspensions are studied here (Table I): one is composed of poly(methyl methacrylate) (PMMA) spheres in a Newtonian fluid and the other is composed of polystyrene (PS) spheres in a visco-plastic fluid. In both cases, particles and suspending fluids are density-matched to avoid any buoyancy effects. The PMMA particles average diameter is $170\mu\text{m}\pm 12\mu\text{m}$ and the PS particles average diameter is $138\mu\text{m}\pm 8\mu\text{m}$. These diameters were measured from optical pictures of bead samples. With such a size, particles are non-Brownian and supposed to be non-colloidal. For all of experiments presented in this work, the solid fraction ϕ of the suspensions was observed to be uniform within our rheological set-up.

The Newtonian suspending fluid is an acrylic double matching liquid from Cargille Lab. Its density is 1.19 and its viscosity 1300 mPa.s at 25°C. The natural contrast of X-ray absorption between particles and the fluid is sufficient to separate them in final X-ray CT images.

The visco-plastic fluid, of density 1.05, is a concentrated emulsion of aqueous drops dispersed at a 77.5% volume fraction (volume of dispersed phase over total volume) in a continuous oily phase. Surfactant SPAN80 is added in dodecane oil at 7.5% in mass (SPAN80 mass over dodecane mass) and iodine sodium salt is added in water at 15% in mass (NaI mass over H₂O mass) to stabilize the emulsion and enhance contrast for X-ray imaging. The droplets size is of order of $1\mu\text{m}$, much smaller than the particles size. This ensures that the emulsion is seen as a continuous material – a visco-plastic fluid – by the particles [2, 4, 5]. The behaviour of the visco-plastic fluid is well described by a Herschel-Bulkley law, relating the shear stress τ to the shear rate $\dot{\gamma}$, with $\tau_y = 26\text{Pa}$ and $K = 5.1\text{Pa.s}^{0.5}$:

$$\tau = \tau_y + K\dot{\gamma}^{0.5}, \text{ if } \tau \geq \tau_y, \quad (1)$$

valid in the steady flow regime, i.e. for stresses larger than the yield stress τ_y . The elastic contribution to its behaviour is accounted for by the elastic shear modulus $G' = 250\text{Pa}$.

More details on the rheology of both background (Newtonian and visco-plastic) fluids, with and without particles, in [5] and [24] respectively.

FIG. 3. X-ray radiograph corresponding to the transmitted intensity of X-rays through the sample (the visco-plastic suspension) along one linear path (a), reconstructed horizontal slice encoding for X-ray absorption of voxels in the shear cell at a solid fraction $\phi \simeq 35\%$ (b) and zoom of this slice (c) corresponding to the white rectangle drawn in (b). The scale bar drawn in (c) is 1mm long.

C. X-ray microtomography

In this study, we used the X-ray microtomography device of Laboratoire Navier, made up of an Ultratom scanner designed and assembled by RX Solutions (Annecy, France) on the basis of the specific requests of Laboratoire Navier. Using the commercial software Xact developed by this company, based on the filtered retroprojection algorithm adapted to cone-beam geometry [25], 3D images encoding for the X-ray absorption field are reconstructed from the recorded 2D radiographs. Consistently with the definition of the final 3D images, 1440 radiographs have been recorded over 360° . Note that for each rotation angle, 6 radiographs (with an exposure time for one radiograph of $1/3$ s) have been averaged to improve the signal to noise ratio. The final 3D images had a resolution of $12\mu\text{m}$ and a definition of $1840 \times 1840 \times 170$ voxels.

One of the advantages of this technique is that the spatial resolution can be isotropic, contrary to other scanning techniques, such as confocal microscopy, laser scanning or refractive index matched scanning [12, 26–29]. This allows to get information on the microstructure with the same accuracy in all planes. Figure 3 shows one example of a 2D radiograph of the visco-plastic suspension confined between the two parallel plates and $12\mu\text{m}$ -thick horizontal slices of the reconstructed 3D image for a solid fraction $\phi \simeq 35\%$.

D. Dedicated rheological set-up

The suspension is sheared at the macroscopic scale in a parallel plates geometry (Fig. 2a) of diameter $D = 2\text{cm}$ and gap $H = 2\text{mm}$ (aspect ratio $D/H = 10$), controlled by a rheometer (Kinexus, Malvern). The plates of the shear cell (Fig. 2c) are made rough to prevent slipping of the suspension at solid boundaries. They have been treated as follow: each PMMA plate was roughened thanks to sand-paper of roughness size $150\mu\text{m}$, then a grid pattern of grooves was imprinted thanks to a file with an edge (triangular lime). In the following, we will compare some results with smooth PMMA plates and roughened PMMA plates.

To image a sample in 3D, 2D radiographs have to be recorded for many different view angles, which means that X-rays have to be allowed to propagate through the sample for all directions nearly perpendicular to the rotation axis, which would not be possible with a standard rheometer (its body would block X-rays when placed between the source and the detector). This leads us to build a dedicated parallel plates set-up, made of PMMA to ensure a low X-ray absorption by the set-up (Fig. 1c). This set-up can be inserted in the rheometer for shearing the material and characterizing its rheological response. After a given shear history in the rheometer, it can then be blocked thanks to a chuck, removed from the rheometer and carefully moved (to avoid any disturbance) to the X-ray CT set-up, where it is put on the rotating stage for imaging. During the whole duration of X-ray imaging, the parallel plates set-up is blocked. We checked that our ex-situ experiments (moved from the rheometer to the X-ray CT set-up) lead to the same results as in-situ ones (for which both shear and imaging are on the spot).

In a parallel plates geometry, when rotation is imposed to the top plate at a rate Ω while keeping fixed the bottom plate (Fig. 2b), the force balance equation and boundary conditions (no slip at the two plates) predict a simple shear of the fluid in the orthoradial plane (θz), the single non-zero value of the shear rate tensor being:

$$\dot{\gamma}(r) = 2d_{z\theta}(r) = \Omega r/H, \quad (2)$$

with an azimuthal velocity:

$$u_{\theta}(r, z) = \Omega z r/H, \quad (3)$$

(r, θ, z) being the cylindrical coordinates [30]. The rotational shear is not homogeneous but depends linearly on the radial direction r . This analysis holds not only for Newtonian fluids,

209 but for complex fluids too.

210 In the same geometry of parallel plates, a squeeze flow can be imposed by translating
 211 vertically one plate at a velocity ϑ (Fig. 2b). It can be approximated by a simple shear
 212 flow in the radial plane (rz), in the case of a large aspect ratio D/H and of no slip at
 213 solid boundaries [30–32]. In this case, the shear rate $\dot{\gamma}(r, z) = 2d_{zr}(r, z)$ is inhomogeneous
 214 and a priori unknown: it depends on the rheology of the fluid of interest. Nevertheless,
 215 a convenient approximation of the maximal shear rate, valid for a Newtonian fluid in the
 216 lubrication approximation, is:

$$\dot{\gamma} = 3D\vartheta/(2H^2). \quad (4)$$

217 Note that by symmetry, the shear has a different sign in the upper and lower part of the
 218 suspension through the central surface at $z = 0$.

219 In order to shear the material in a parallel plates geometry, it is first poured on the
 220 bottom plate, then it is squeezed by the top plate to reach the required gap. So even if the
 221 interest is in a rotational shear, the material generally experiences first a squeeze flow during
 222 the loading of the fluid in the geometry, which might affect its initial state. That is why in
 223 the following, we will investigate the impact of both flows on the suspension microstructure.

224 III. IMAGE ANALYSIS

225 An accurate identification of particle centers from the obtained 3D images is crucial for the
 226 computation of pair vectors and pair distribution functions to characterize the microstructure
 227 of the suspension. Thus, we choose to develop home-made algorithms to be able to identify
 228 particles center at a sub-voxel resolution.

229 In all the text, the 3D images I we work with are fields of absorption coefficients encoded
 230 as intensity levels without unit measurement. In practice, 16 bits images have been used
 231 (grey levels between 0 and $2^{16}-1$).

232 Figure 4 shows the typical greylevel profile of a particle: grey levels of all voxels in the
 233 neighborhood of a particle are plotted against the distance r to the voxel considered to be
 234 its center. Assuming the density of the particles to be uniform, one would have expected a
 235 Heaviside-like shape for such plots, with a uniform grey level for r lower than the particle
 236 radius (of the order of 6 voxels), corresponding to the absorption coefficient of PMMA or
 237 PS, and another (larger) uniform grey level for larger r , corresponding to the absorption of

FIG. 4. Absorption levels (without unit measurement, included between 0 and $2^{16} - 1$) of voxels over one particle and its close neighbourhood in the visco-plastic suspension at a solid fraction $\phi \simeq 40\%$, as a function of the distance r to its center, from the raw and filtered images I (a) and I_f (b)

the fluid.

Actual profiles first differ from ideal profiles because of natural random image noise, which cannot be avoided, especially if fast imaging conditions are required. Such fluctuations are indeed observed near $r = 0$, up to a distance of about 2 to 3 voxels, where grey levels are essentially uniform on average but fluctuate randomly.

The second discrepancy with respect to the ideal situation takes the form of a gradual increase of the average grey levels for r varying from about 3 to 7 voxels. While there are still fluctuations associated to natural image noise, the smooth progressive increase of the average grey level is due to the limited spatial resolution of the CT imaging device. Indeed the grey level of a given voxel in the reconstructed image should be considered as a spatial convolution of the physical absorption coefficient of the material (relative to the spectrum of the polychromatic X-ray beam) with some kernel of small finite size. This kernel characterizes the resolving power of the imaging system. For CT imaging systems, it is sensitive to various parameters, such as the finite spot size of the X-ray source (which can be evaluated to be about $8 \mu\text{m}$ for our imaging conditions), diffusion effects of the scintillator, fill factor of the pixel matrix, blurring effects of electronic device,... A more detailed discussion can be found in [33]. In the present setup, and considering the grey level profile presented in figure 4, the radius of this kernel is likely to be of the order of 2 to 3

TABLE II. Values of the parameters ℓ_{noise} , r_{neigh} , r_{min} and r_{sym} used for image analyses, with d , the diameter of particles

ℓ_{noise}	r_{neigh}	r_{min}	r_{sym}
1 voxel	~ 7 voxels	~ 6 voxels	~ 7 voxels
$\sim d/10$	$0.6d$	$0.5d$	$0.6d$
$\sim 12\mu m$	$\sim 83\mu m$	$\sim 69\mu m$	$\sim 83\mu m$

voxels, which explains that the sharp theoretical particle-matrix interface is degraded into a smooth interface with a thickness of about 5 voxels.

A third discrepancy is for r larger than 7 voxels, where the profile seems to tend on average to a uniform grey level, expected to be related to the absorption coefficient of the fluid, but with much larger fluctuations than within the particles. This might be due to other neighbor particles: the voxels that are close to the latter will exhibit a lower grey level than those further away.

In the following, we will finally take advantage of these (slightly) inhomogeneous grey level profiles of particles in order to determine accurately their position.

Below we describe the successive steps. First, we detect approximately particle centers as the local minimum absorption and we get rid of false and multiple detections. Second, we refine particle center positions at a sub-voxel resolution by using the symmetry of the increasing absorption around the center. Finally, we explain how we compute pair distribution functions.

A. Detection of particles

We process the 3D images with 3D morphological operations within Matlab. We use as few as possible filtering operations and we introduce as few as possible tunable parameters, whose values are not severely chosen, so that our method depends only slightly on the value of parameters and is efficient for various solid fractions without changing the value of any parameter.

First, we smooth the raw 3D image I by filtering high spatial frequency noise with a

FIG. 5. Two dimensional (a, b) and three dimensional (c, d) images of the visco-plastic suspension of length about $\sim 1\text{cm}$ at a solid fraction $\phi \simeq 35\%$: raw images I (a, c) and filtered images I_f (b, d)

277 Gaussian filter of characteristic size $\ell_{noise} = 1$ voxel: $I_f = f * I$ with

$$f(x) = \exp\left(-\left(\frac{x}{2\ell_{noise}}\right)^2\right) / \int_{-\infty}^{\infty} \exp\left(-\left(\frac{x}{2\ell_{noise}}\right)^2\right) dx, \quad (5)$$

278 where the symbol $*$ refers to the convolution operator: $(f * I)(x_0) = \int_{-\infty}^{\infty} f(x)I(x - x_0)dx$,
 279 and I_f is the image we work with in the following steps (Fig. 4 and 5). This filtering
 280 operation has a long-range effect even in the case of a small value for the length ℓ_{noise} , due
 281 to the extended range of integration in the convolution operation; in practice, the integration
 282 is realized over a size of $21 \times 21 \times 21$ voxels, chosen after we checked that a larger size does not
 283 change the value of I_f .

284 Second, we extract the positions of voxels showing a local minimum of absorption as a
 285 first estimation of the particle centers.

286 False detections corresponding to local extrema in the air, the plates or the fluid, as well
 287 as multiple detections (several centers for the same particle) are possible. In practice, they
 288 represent less than 1% of all detections; however, we do get rid of these false and multiple
 289 detections thanks to the two following steps.

- 290 • We first note that X-ray absorption coefficients of air μ_{air} , particles $\mu_{particles}$ and
 291 suspending fluids μ_{fluid} are well separated and sorted according to:

$$\mu_{air} < \mu_{particles} < \mu_{fluid}. \quad (6)$$

292 To get rid of false detections, we build an histogram of the values of absorption of
 293 the detected extrema (Fig. 6). In practice, we do not use the value of the extrema,
 294 but the averaged value over a close neighbourhood around the extrema position, of

FIG. 6. Histogram (in fraction) of absorption values (without unit measurement) averaged over a close neighbourhood of size r_{neigh} around the detected extrema, for the visco-plastic suspension at a solid fraction $\phi \simeq 35\%$, before selection of extrema identified as real particles.

size r_{neigh} taken equal to 60% of the mean diameter of particles (Tab. II), to avoid considering small dust (solid of tiny size). Also, this allows for a better separation in the histogram. On the histogram, we clearly distinguish the absorption values corresponding to particles, fluid and air. We kept only voxels of averaged absorption values between μ_{min} and μ_{max} . In Figure 6, the width of the histogram results from the macroscopic inhomogeneities of absorption in space (due to a phenomenon called beam hardening) and likely bears the signature of particle distribution in space. False detections corresponding to local extrema in the plates have the same absorption value as particles, so a meticulous inspection of slices close to the plates and a manual exclusion may be needed if necessary.

- We compute distances between possible particle center positions to get rid of multiple detections. If two possible centers are closer than a threshold distance r_{min} , we keep only the one of minimal absorption. In the case of perfectly monodisperse particles and non-noisy images of infinite spatial resolution, r_{min} could be taken as 100% of the diameter of particles. Since particles are slightly polydisperse and at this step the accuracy of particle centers is only of ± 1 voxel, we choose r_{min} equal to 50% of the mean diameter of particles (Tab. II). Figure 7a shows the apparent volume fraction ϕ_∞ far from reference particles, computed from the number of neighbours detected by unit volume, for different values of the threshold distance r_{min} . The

FIG. 7. a) Volume fraction $\phi_\infty(r_{min})$ far from reference particles, computed from the number of neighbours detected by unit volume, for different values of the threshold distance r_{min} (minimal distance between two centers). b) Distribution of the interparticle distance for the identified first neighbours for all $r_{min} \leq 9$. The visco-plastic suspension (a, b) has a solid fraction equal to $\phi \simeq 36\%$.

value of r_{min} was varied excessively from 1 to 15 voxels corresponding to $0.1d$ and $1.3d$, with d , the particle diameter. We see that for $r_{min} < 9$ voxel, the volume fraction ϕ_∞ is unchanged, and corresponds to the volume fraction of the prepared particles in the suspension; for larger values of r_{min} , the measured volume fraction ϕ_∞ decreases strongly, showing that some relevant particles are lost. Figure 7b shows the distribution of inter particle distances for the identified first neighbors for all $r_{min} \leq 9$. All distributions are superimposed, but there is clearly an excess of neighbours at small distances for $r_{min} < 6$, which likely corresponds to multiple centers for the same particle. That is why $r_{min} = 6$ finally seems to be a good choice (Table II). Moreover we checked that this choice allows to preserve the quality and quantity of the pdf.

Note that the values of the parameters r_{neigh} , r_{min} , μ_{min} , μ_{max} (Tab. II) are not severely chosen, ensuring no arbitrary exclusion or selection of particles and more importantly a low influence of their values on our results. Finally, we do not observe any forgotten particle when we systematically check some samples of 3D images.

B. Sub-voxel identification of particle centers

At this stage, the selected centers corresponding to exact voxel positions are identified as particles. We then refine their positions at a sub-voxel resolution by using symmetry properties of the absorption signal over a particle around its center (Fig. 4).

First, the filtered image I_f is linearly interpolated at the chosen sub-voxel spatial resolution to give I_{vx} (the resolution we want for the particles centers). The (tri)linear interpolation lays on the following principle: the value of the voxel to interpolate (at the chosen position) is taken equal to the average of the values of the (original) crossed voxels, weighted by the relative partial crossed volumes.

Second, an algorithm searches the position (x_0, y_0, z_0) in a range $\pm(\delta x_0, \delta y_0, \delta z_0)$, minimizing ΔI , built to quantify the asymmetry of the signal around this center, defined as:

$$\Delta I^2 = \sum_{\Delta r \leq r_{sym}} (I_{vx}(x_0 + \Delta x, y_0 + \Delta y, z_0 + \Delta z) - I_{vx}(x_0 - \Delta x, y_0 - \Delta y, z_0 - \Delta z))^2 \quad (7)$$

for values of $\Delta r = \sqrt{\Delta x^2 + \Delta y^2 + \Delta z^2}$ smaller than a threshold r_{sym} , taken as 60% of the mean diameter of particles (Tab. II).

A compromise between precision and computer time leads us to refine successively the centers positions at 1 voxel first, then 1/2 voxel, until $(1/2)^n$ voxel, with n , the integer such that the chosen spatial resolution is reached.

Figure 8a shows the variations of ΔI with the displacement Δx of the center in the x -direction in the range ± 2 voxels, for different couple values $(\Delta y, \Delta z)$ of the displacement of the center in the y - and z -directions: there is indeed a minimum ΔI_{min} allowing to identify the center. The same holds for Δy and Δz in the y - and z -directions. Figure 8b shows the decrease of the minimum ΔI_{min} with n , the step of sub-voxel identification, what confirms that the minimization and the sub-voxel refinement have still a physical sense till $n = 5$, allowing to reach a resolution on the particles center position of $(1/2)^5$ voxel $= 0.375\mu\text{m} \simeq d/100$. Note that $n = -1$ (Fig. 8b) corresponds to the case of no interpolation of the images I_f , so that $\Delta I_{min}(n = -1)$ quantifies the asymmetry of the signal around the minimal absorption voxel. For the results presented in this paper, a spatial resolution of $6\mu\text{m} \simeq d/20$, i.e. $n = 1$ is enough.

Figure 9 shows the original (non interpolated, i.e. at 1 voxel resolution, $n = -1$) absorption signal I_f (without unit measurement) over one particle as a function of distance from

FIG. 8. a) Variations of ΔI (Eq. 7) with the displacement Δx of the center in the x -direction for the first step of sub-voxel identification $n = 0$. Each line corresponds to different couple values of the displacement of the center ($\Delta y, \Delta z$) in the y - and z -directions. b) Variations of $\Delta I_{\min}$ with the number of steps of sub-voxel identification n . $\Delta I(n = -1)$ quantifies the asymmetry of the signal around the minimal absorption voxel, i.e. without any interpolation of the image I_f . Note that here ΔI was normalized by the number of voxels for which it has been computed to allow for a comparison at different sub-voxel resolutions

the center, for the initial center (determined from the local extrema) (Fig. 9a) and for the centers refined successively with $n = 0, 1$, and 2 in Figure 9b, c and d respectively. We see that for $\Delta r \leq r_{\text{sym}}$ the scattering of data (or asymmetry of the signal around its center) decreases at each step of sub-voxel identification, that is quantified by ΔI . Even at 1 voxel resolution (for $n = 0$), we already note that the center of symmetry (which is used to plot Figure 9b) is not always the minimal voxel of absorption (which is used to plot Figure 9a).

C. Pair distribution functions

From particle positions within the parallel plates geometry, we are able to study how particles are spatially distributed in the suspending fluid. In addition to information on the structure of the suspension at the macroscopic scale from global positions of particles within the geometry, we do get information on the microstructure of the suspension at the particle scale from relative positions of particles (relative to each other) through the computation of pair distribution functions [7].

The pair distribution function $g(\vec{r})$ is the probability of finding a particle pair separated

FIG. 9. Absorption levels (without unit measurement) of voxels over one particle and its close neighbourhood in the visco-plastic suspension at a solid fraction $\phi \simeq 40\%$ as a function of the distance Δr to its center, through the different steps ($n = -1, 0, 1, 2$ in a, b, c, d) of sub-voxel identification based on symmetrisation

by the vector $\vec{r}$ normalized by the mean particle density, so that the asymptotic value of g for large values of $||\vec{r}||$ is:

$$g(||\vec{r}|| \rightarrow \infty) = 1, \quad (8)$$

in the absence of macroscopic structure or long-range correlations. Equivalently, $g(\vec{r})$ is a normalized probability of finding a (test) particle located at $\vec{r}_0 + \vec{r}$ with another (reference) particle located at $\vec{r}_0$. This last definition suggests naturally how to compute $g(\vec{r})$: N_{ref} particles are subsequently chosen to be the reference particles; we identify the N^{pair} test particles separated by $\vec{r}$ from each reference particle, corresponding to the number of pairs characterized by $\vec{r}$ in the region of interest.

In the following, $\vec{r}_0 = (r_0, \theta_0, z_0)$ and $\vec{r}_0 + \vec{r} = (r, \theta, z)$ are the cylindrical coordinates (attached to the axis of the rheometer circular plates) of a reference and of a test particle

FIG. 10. Histogram (in fraction) of the interparticle 3D (solid lines) and 2D (dashed lines with crosses) distances for the first neighbours identified in slices of different thicknesses $\Delta = d/8$, d and $2 * d$, with d , the particles' diameter. The visco-plastic suspension has a solid fraction equal to $\phi \simeq 38\%$.

located in M_0 and M .

To avoid any finite size or geometry effect on $g(\vec{r})$, that would introduce some bias – for example underestimating the statistics in the direction of a boundary – test particles shall be selected around their reference particle within an isotropic region (e.g. that does not cross any boundary). To be able to study the microstructure even close to boundaries (solid plates or free interfaces), we use adaptative spheres of variable radius, equal to the minimal distance between the reference particle and the closest boundary, as the area where searching test particles around a reference particle. In this case, we have to update the number of reference particles N_{ref} that is no more constant for all values of $\vec{r}$, especially for large values of $||\vec{r}||$. This choice is an alternative to exclusion of regions of investigation as in [14] and to a geometry-dependent correction introduced in the computation of $g(\vec{r})$ as in [11].

The 3D pair distribution function $g(\vec{r})$ is a function of 3 scalar variables of size $\sim N^3$ in the discretized space, with N , the number of voxels in a given direction of space. However, for visualization, we generally plot the values of $g(\vec{r})$ taken in three orthogonal slices (data of size $\sim N^2$). As relevant slices, we choose the cylindrical ‘planes’ attached to the axis of the rheometer circular plates, as naturally suggested by the cylindrical symmetry of the set-up and the flow. So, to reduce computation time, we generally compute only three 2D pair

401 distribution functions: g_r in the orthoradial ‘plane’ (θz) normal to r (being a toroidal ring or
 402 a cylinder), g_θ in the radial plane (rz) normal to θ and g_z in the horizontal plane (θr) normal
 403 to z . So, the region where searching for test particles can be reduced to the intersection of
 404 a sphere and the elementary cylindrical ‘plane’ of interest attached to the reference particle.
 405 In practice, defining the elementary planes as strictly 2D slices of cylindrical coordinates
 406 $r = r_0$, $\theta = \theta_0$ or $z = z_0$ for a reference particle in $M_0(r_0, \theta_0, z_0)$ does not allow to sample
 407 enough test particles and get enough statistics, so we affect elementary thicknesses to the
 408 cylindrical ‘planes’. Whereas the definition of the thicknesses Δz of horizontal planes (θr)
 409 and Δr of orthoradial ‘planes’ (θz) is obvious, the thickness of radial planes (rz) offers
 410 several choices (see Appendix A). We choose the definition of:

$$\Delta h = r \sin(\theta - \theta_0), \quad (9)$$

411 corresponding to a constant euclidean thickness (or minimal distance of M from the ra-
 412 dial direction OM_0), because it prevents a non symmetric pattern on the pair distribution
 413 function g_θ (see Appendix A). We checked the influence of the value Δ of the thicknesses
 414 of the planes $\Delta = \Delta z = \Delta r = \Delta h$ on our results. Figure 10 compares two distributions:
 415 the 3D distances (real distance) and the 2D distances (projected in the slice) between first
 416 neighbours of particles in an elementary cylindrical plane for different thicknesses Δ be-
 417 tween $d/8$ and $2 * d$. The 3D distances are always equal or larger than d with a maximum
 418 probability for d . But the 2D distances are not, because they do not represent well real 3D
 419 distances in slices of thicknesses larger than d , as demonstrated by the distributions that do
 420 not super-impose for $\Delta > d$. This leads us to choose for the thickness of elementary cylin-
 421 drical ‘planes’ $\Delta = d$, with d the particle diameter, as a compromise between accuracy and
 422 statistics. Moreover we checked that this choice allows to preserve the quality and quantity
 423 of the pdfs.

424 Different choices for the characterization of the pair vector $\vec{r} \equiv \overrightarrow{M_0 M}$ (relative coordinates
 425 between test and reference particles located in M and M_0) are possible (see Appendix A).
 426 Euclidean coordinates can in principle be chosen but we believe that non euclidean coor-
 427 dinates, curvilinear projected along the cylindrical ‘planes’ aligned with the circular flow
 428 streamlines, are more relevant to characterize the microstructure induced by a simple shear
 429 flow. We choose for coordinates of the pair vector $\vec{r} = (\rho, \ell, \xi)$:

$$\rho = r - r_0, \quad \ell = r(\theta - \theta_0), \quad \xi = z - z_0, \quad (10)$$

FIG. 11. Two particles $M_0(r_0, \theta_0, z_0)$ and $M(r, \theta, z)$ in the global cylindrical framework and definition of the pair vector $\vec{r} \equiv \overrightarrow{M_0 M}$ characterizing their separation in curvilinear coordinates (along circular flow lines): $\vec{r} = (\rho, \ell, \xi) = (r - r_0, r(\theta - \theta_0), z - z_0)$

as shown in Figure 11. This choice does not induce any bias in the symmetry of the pair distribution functions g_r , g_θ or g_z (see Appendix A). Note that in the following, when showing a 2D pdf g (g_r , g_θ and g_z), we will use alternatively cartesian coordinates (couples among ρ , ℓ or ξ) or polar coordinates (ρ_{2d}, ϕ_{2d}) for a pair of particles in the plane of interest. In the (θz) , (rz) and (θr) planes, cartesian and polar coordinates are respectively related through $(\rho_{2d} \cos \phi_{2d}, \rho_{2d} \sin \phi_{2d}) = (\ell, \xi)$, (ρ, ξ) and (ρ, ℓ) .

Altogether, with the mean particle density (or number of particles per unit volume):

$$n_{0v} = N/V = \phi/(\pi d^3/6), \quad (11)$$

with the total number of particles N , the total volume of the suspension V , the particle concentration (or solid fraction) ϕ , the particle diameter d , the formula for the 2D pair distribution functions g_r , g_θ and g_z in the (θz) , (rz) and (θr) planes respectively (for spatial resolutions $\delta\rho$ and $\delta\ell$, $\delta\xi$) are:

- $g_r(\ell, \xi) = P_r(\ell, \xi)/n_{0v}$, with $P_r(\ell, \xi)\delta V_r = N_r^{pair}(\ell \pm \delta\ell/2, \xi \pm \delta\xi/2)/N_{ref}$,
- $g_\theta(\rho, \xi) = P_\theta(\rho, \xi)/n_{0v}$, with $P_\theta(\rho, \xi)\delta V_\theta = N_\theta^{pair}(\rho \pm \delta\rho/2, \xi \pm \delta\xi/2)/N_{ref}$,
- $g_z(\rho, \ell) = P_z(\rho, \ell)/n_{0v}$, with $P_z(\rho, \ell)\delta V_z = N_z^{pair}(\rho \pm \delta\rho/2, \ell \pm \delta\ell/2)/N_{ref}$,

with N_{ref} , the number of reference particles, and (ρ, ℓ, ξ) , the coordinates of a pair vector defined in Eq. (10). While g refers to a pair distribution function, P refers to a probability of finding a pair of particles and N^{pair} to a number of pairs of particles. More precisely, $N_z^{pair}(\rho \pm \delta\rho/2, \ell \pm \delta\ell/2)$ is the number of pairs of coordinates $(\rho \pm \delta\rho/2, \ell \pm \delta\ell/2)$ in cylindrical

horizontal planes (θr) of thickness Δz or equivalently the number of test particles in the same horizontal plane than the reference particles (at z_0), i.e. particles at $z_0 \pm \Delta z/2$; the same holds for $N_r^{pair}(\ell \pm \delta\ell/2, \xi \pm \delta\xi/2)$ and $N_\theta^{pair}(\rho \pm \delta\rho/2, \xi \pm \delta\xi/2)$ with the thickness Δr and Δh of orthoradial plane (θz) and radial plane (rz) respectively. The elementary sampling volumes (defining the spatial resolution for g in 2D) are equal to:

$$\delta V_r = \Delta r \delta \ell \delta \xi, \quad \delta V_\theta = \delta \rho \Delta h \delta \xi, \quad \text{and} \quad \delta V_z = \delta \rho \delta \ell \Delta z. \quad (12)$$

Here, the thickness of the ‘planes’ are chosen as $\Delta z = \Delta r = \Delta h = d$ and usually the spatial resolution for pair distribution functions g is chosen as $\delta\rho = \delta\ell = \delta\xi = 1/2$ voxel. Note that these spatial averages may lead to some underestimate of peak values of g whether there are large variations of g , that may lead to some discrepancies between theoretical and experimental values. Finally, in three dimensions, the expression for the pdf g is:

$$g(\vec{r} \pm \delta\vec{r}) = N_\infty^{pair}(\vec{r} \pm \delta\vec{r}) / N_\infty^{pair} \quad \text{with} \quad N_\infty^{pair} = N_{ref} n_{0v} \delta V, \quad (13)$$

N_∞^{pair} being the number of pairs separated by a vector $\vec{r}$ such that $||\vec{r}|| \rightarrow \infty$ in the sampling volume δV in the space of pair vectors delimited by $\vec{r} \pm \delta\vec{r}$.

In the following, the analysis of the microstructure will be generally realized, unless specified otherwise, far enough from the free interface and from the solid plates, to ensure not to mix possible structurations near boundaries to bulk microstructure. In order to characterize the microstructure for a roughly homogeneous simple shear, it has to be computed at a fixed radial position R_0 in the parallel plates geometry. In practice, to get a good precision of the pdf, a toroidal region of sufficient thickness ΔR has to be analysed. As an example (see Appendix B for details), if we want a precision $\Delta g = 10^{-2}$ on $g(\vec{r})$ and a spatial resolution for the 2D pdf $\delta V = \Delta \delta^2$ with $\Delta = d = 140\mu\text{m} = 12\text{vx}$ and $\delta = 1\text{vx} = 12\mu\text{m}$, for a solid fraction $\phi = 35\%$ ($n_{0v} = 244\text{mm}^{-3}$), we need at least $2 \cdot 10^4$ reference particles in the region of interest. This leads us to choose $R_0 = 0.72D/2 \pm 0.12D/2$, unless specified.

In the following, we now illustrate the potential of our technique by analyzing the microstructure of our suspensions near the horizontal solid plates, near the orthoradial free interface, and in the bulk separately.

FIG. 12. a) X-ray radiograph of a non sheared drop of a visco-plastic suspension at a solid fraction $\phi \simeq 35\%$. b) 3D plot of centers of all detected particle (in blue) and reference particles (in green) for the characterization of the microstructure

IV. MICROSTRUCTURE

A. Bulk microstructure of a non sheared visco-plastic suspension

First, we characterize the initial microstructure of a visco-plastic suspension poured on the bottom plate of the rheometer, but before any loading and shear history. A suspension is prepared by simply mixing ‘by hand’ the particles and the fluid in a cup with a spatula, with the goal of achieving a mixing close to chaotic mixing. With this procedure, we aim to prepare a material that is homogeneous and isotropic. It is then degassed to remove possible air bubbles (in a vacuum or in a centrifuge). Upon pouring, the visco-plastic suspension has the shape of an irregular drop due to its yield stress (Fig. 12). Here, we analyze the microstructure in a toroidal region ($R_0 = 0.36D/2 \pm 0.30D/2$ and $Z_0 = 2.4H \pm 1.2H$) inside the drop far from solid surfaces and free interfaces (Fig. 12b).

In its initial configuration, we observe that the suspension has at first order an isotropic microstructure: the pair distribution function $g(\vec{r})$ does not depend much on the direction $\vec{r}/||\vec{r}||$ (Fig. 13 and 19). Figure 13 shows $g_r(\ell, \xi)$, $g_\theta(\rho, \xi)$ and $g_z(\rho, \ell)$: they are roughly the same in the three orthogonal 2D ‘planes’, with a circular symmetry in each 2D ‘plane’. $g(\vec{r})$ mostly depends only on the distance $||\vec{r}||$. Figure 14b shows the 1D scalar pdf $g(||\vec{r}||)$: $g(||\vec{r}||) = 0$ for (and only for) distances $||\vec{r}|| \lesssim d$; it has a maximal value for $||\vec{r}|| \simeq d$ and a second local maxima for $||\vec{r}|| \simeq 2d$; then g tends to 1 for larger values of $||\vec{r}||$.

Note that a closer inspection of the 2D pdfs (Fig. 13b) shows that they are not exactly the same and not exactly isotropic: the averages of g_r and g_θ for distances ρ_{2d} close to its maxima ($\rho_{2d} = d \pm d/6$) as a function of ϕ_{2d} (represented in a polar plot) are not perfectly

FIG. 13. a) Three dimensional microstructure of a visco-plastic suspension in a non sheared configuration (drop) at a solid fraction $\phi \simeq 35\%$: 2D pair distribution functions $g_r(\ell, \xi)$, $g_\theta(\rho, \xi)$ and $g_z(\rho, \ell)$. b) Polar plots of $\langle g(\phi_{2d}) \rangle_{\rho_{2d}=d \pm d/6}$ in the three planes of interest (θz) , (rz) and (θr)

FIG. 14. Numerical microstructure from a simulation of finite-size particles following a naive rule of exclusion and redistribution for a solid fraction $\phi \simeq 36\%$: a) 2D pair distribution function $g(\ell, \xi)$ and b) 1D scalar pair distribution function $g(\rho_{2d})$ super-imposed with the experimental 1D pdf for a non sheared suspension

495 circular but bear a signature of a slight over-population of pairs of particles roughly aligned
 496 with the gravity. This slight micro-structuration is not visible in the (θr) plane (on g_z).
 497 This may be attributed to vertical flows of the suspension when it is poured on the plate.

498 This almost isotropic experimental pdf is now compared to the numerical pdf obtained
 499 when simulating a random distribution of finite-size particles, with a naive rule of exclusion
 500 and redistribution (Fig. 14). The finite-size effect introduced here, arbitrarily governed
 501 (i.e. without introducing physical forces), has to be seen as a minimal steric constraint. A

FIG. 15. Three dimensional microstructure of a visco-plastic suspension after loading and squeezing in a parallel plates geometry at a solid fraction $\phi \simeq 35\%$: 2D pair distribution functions $g_r(\ell, \xi)$, $g_\theta(\rho, \xi)$ and $g_z(\rho, \ell)$ for $z < 0$ (a) and for $z > 0$ (b)

random distribution of points (representing particles of zero size) gives obviously a constant pdf equal to 1 everywhere. Adding an effect of a finite size by taking away over-lapping particles (separated by a distance smaller than their diameter d) and separating them by their diameter d in the initial direction (and repeating this until no more particles over-lap [34]) leads to a pdf dependent on $||\vec{r}||$ (Fig. 14) with the same features as the experimental pdf we measure in 3D in a non sheared visco-plastic suspension, in particular local minimum and secondary maximum at the same positions. The main visible difference is the width and the value of the first peak of g maybe due to the difference of granulometry of the particles, to the arbitrary rule of redistribution of over-lapping particles and to a specific spatial distribution of particles in a non sheared suspension. Finally, the fluid does not seem to have any significant impact on the microstructure during the mixing.

B. Bulk microstructure of a visco-plastic suspension after a continuous squeeze

When a suspension is loaded into a parallel plates geometry, it is first poured on the bottom plate. The top plate is then translated downwards, thus squeezing the visco-plastic suspension between the two plates. The flow induced by this squeeze flow is an inhomogeneous simple shear flow, which has been thoroughly described in the literature [30–32] and is briefly discussed in part IID.

The technique developed here for the 3D characterization of microstructure allows us

FIG. 16. Pair distribution function $g_\phi(\rho, \xi)$ after a squeeze flow in the shear plane for $z < 0$ (a) and for $z > 0$ (b)

FIG. 17. Pair distribution functions g_r , g_θ and g_z averaged for distances $\rho_{2d} = d \pm d/6$ plotted as a function of the angle ϕ_{2d} in polar coordinates after a squeeze flow for $z < 0$ (a) and for $z > 0$ (b) for a visco-plastic suspension at a solid fraction $\phi \simeq 35\%$

520 to investigate if and how the simple shear in the (rz) plane induced by this squeeze flow
 521 changes the microstructure of the suspension, initially nearly isotropic as seen above. The
 522 plane in the middle of the gap, defined by $z = 0$, is an axis of symmetry of the flow. To take
 523 into account the different sign of the shear rate (velocity gradient) in the upper and lower
 524 part of the suspension, we analyse here the microstructure separately in both semi-planes
 525 $z < 0$ and $z > 0$.

526 In Figures 15 and 16, we observe that an anisotropic microstructure develops in the shear
 527 plane (rz) (velocity-velocity gradient plane), while it remains approximately isotropic in the

two other cylindrical planes (θz) and (θr) where the suspension does not experience any shear. Contrary to the nearly isotropic microstructure observed previously in the initial configuration, the pdf $g(\vec{r})$ is no more simply a scalar function of $||\vec{r}||$ but depends both on the distance $||\vec{r}||$ and the direction $\vec{r}/||\vec{r}||$. As in shear-induced structuration observed in the literature in the case of the rotational shear of a Newtonian suspension [7, 9–14], we observe here a depletion of particles in close contact ($\rho_{2d} \simeq d$) located in the extensional area close to the direction of the velocity (near the angles $+15^\circ$ and $+195^\circ$ for $z < 0$ and $+165^\circ$ and $+345^\circ$ for $z > 0$). Moreover, Figure 19 shows the 1D scalar plot of $g(||\vec{r}||)$: its maxima is larger and tightens (spreads less) in comparison with the case of the non sheared suspension.

As previously seen, a closer inspection of the 2D pdfs (Fig. 17 and 19) by looking at the averages of g_r , g_z and g_θ for distances ρ_{2d} close to its maxima ($\rho_{2d} = d \pm d/6$) as a function of ϕ_{2d} shows a secondary structuration in addition to the depletion of particles in the (rz) plane: g_r and g_z exhibit an ‘hexagonal’ shape, that may be the signature of a more complex flow occurring in the parallel plates than a simple shear flow as approximated in the framework of lubrication when D/H is large (the flow has also a velocity component aligned with the direction of translation of the bottom plate, which might not be negligible).

As a consequence, a suspension initially characterized by a nearly isotropic microstructure in 3D, develops at first an anisotropic microstructure when loaded in the parallel plates geometry, even before any imposed shear history. Such impact of loading on the microstructure of suspension should be observed in most rheometrical devices (cone-and-plate, Couette, Poiseuille, ...). If the characterization of an isotropic structure is needed, a possible rheometrical tool is the vane in cup geometry, classically used to study gels: isotropy can be achieved by chaotic mixing in the cup, and then the insertion of the vane into the cup should not affect the material’s structure.

C. Bulk microstructure of a visco-plastic suspension after a rotational shear flow

After the loading of the visco-plastic suspension in the parallel plates geometry, a simple shear flow in the (θz) plane is imposed thanks to the rotation of the top plate until a stationary state is reached in terms of shear stress.

As already observed for a Newtonian suspension (numerically and experimentally) [7, 9–14] and as reported in Ovarlez *et al.* [5] for a visco-plastic suspension, the microstructure

FIG. 18. a) Three dimensional microstructure: pair distribution functions $g_r(\ell, \xi)$, $g_\theta(\rho, \xi)$, $g_z(\rho, \ell)$ after a steady rotational shear at a shear rate $\dot{\gamma} = 10^{-2} s^{-1}$ of a visco-plastic suspension at a solid fraction $\phi \simeq 35\%$. b) Pair distribution function g_r , g_θ and g_z averaged for $\rho_{2d} = d \pm d/6$ plotted as a function of the angle ϕ_{2d} in polar coordinates.

558 becomes anisotropic in the (θz) shear plane (velocity-velocity gradient plane), while it is ap-
 559 proximately isotropic in the two other ‘planes’, as shown in Figure 18 and 19: this anisotropy
 560 is referred to in the literature as a fore-aft asymmetry. In 3D, it means that the positions
 561 of particle pairs previously correlated in the (rz) plane due to the squeeze flow, decorrelate
 562 thanks to the rotational shear and the particles reorganize themselves relatively to each
 563 other, leading to some new correlations in the (θz) plane.

564 These correlations between particles develop at the scale of particle pairs in close contact,
 565 leading to a depletion of particles in the extensional area, in contrast with the compressional
 566 area, as well as a primary over-population roughly aligned with the direction of the flow
 567 (+170°). This anisotropy can be quantified thanks to the plots of $g(\phi_{2d})$ (the average of
 568 $g(\rho_{2d}, \phi_{2d})$ for $\rho_{2d} = d \pm d/6$) plotted in polar coordinates in Figure 18b: $g(\phi_{2d})$ has the
 569 shape of a ‘butterfly’.

570 More precisely, Figure 18 shows that for distances ρ_{2d} close to the value of the particles
 571 diameter, g_r has minimal values for the angles +30° and +210°, corresponding to a decrease
 572 of the number of pairs in the extensional stress domain; and it has maximal values for the
 573 angles +170° and +350°, corresponding to an increase of the number of particle pairs aligned
 574 roughly with the flow. These two principal extrema can be referred to as an extensional-
 575 depletion of particle pairs (for the minima of g) and a flow-alignment of particle pairs (for
 576 the maxima of g). Whereas the extensional-depletion of particle pairs was already reported

FIG. 19. Scalar pair distribution functions $\langle g(\rho_{2d}) \rangle_{\phi_{2d}}$ averaged for all values of ϕ_{2d} and $\langle g(\phi_{2d}) \rangle_{\rho_{2d}=d\pm d/6}$ averaged for distances $\rho_{2d} = d \pm d/6$ plotted as a function of the distance ρ_{2d} and the angle ϕ_{2d} respectively in cartesian coordinates for a visco-plastic suspension at a solid fraction $\phi \simeq 35\%$. In the first, second and third lines, $\langle g(\rho_{2d}) \rangle_{\phi_{2d}}$ and $\langle g(\phi_{2d}) \rangle_{\rho_{2d}=d\pm d/6}$ are computed from g_z , g_θ and g_r in the (θr) , (rz) and (θz) planes respectively. Light blue lines correspond to a non sheared suspension, dark blue ones to a suspension sheared by a squeeze flow (in the two bottom and top half-planes $z < 0$ and $z > 0$) and red lines correspond to a suspension sheared by rotation.

in the literature for non-Brownian suspensions, the flow-alignment of particle pairs was not, as well as the presence of secondary maxima and minima. By contrast, a flow-alignment was observed in numerical simulations of Brownian suspensions at small and moderate Peclet numbers, even when which Brownian forces contribute preponderantly over hydrodynamics force [8, 15–18].

To our knowledge, our measurements of 3D microstructure of visco-plastic suspensions are the first ones reported in the literature. We have previously presented some of them in [5]; we recall briefly their main features in the next section.

D. Microstructure of the visco-plastic suspension at different shear rates

Unlike a Newtonian suspension, whose shear-induced microstructure does not depend on the value of the shear rate for steady states [24], we could expect a different behaviour for a visco-plastic suspension, due to its visco-plastic rheological behaviour. For comparison, the microstructure of a Brownian suspension depends strongly on the shear rate through the Peclet number (comparing hydrodynamic shear with thermal forces), as well as its rheology, that exhibits both shear-thinning, Newtonian and shear-thickening behaviours [8, 15–18].

Here, we compare the pdfs in the shear plane (θz), where an anisotropic microstructure develops when the suspension is rotationally sheared, for two different shear rates: 10^{-2}s^{-1} and 10s^{-1} . For the visco-plastic fluid we used, this corresponds respectively to Bingham numbers 1.02 and 1.62 (ratio of the shear stress over the yield stress) and to effective viscosities 51Pa.s and 1.6Pa.s (computed as $(\tau - \tau_y)/\dot{\gamma}$). At first order, in both cases, the microstructure is anisotropic showing an extensional-depletion of particle pairs and a flow-alignment of particle pairs. At second order, we do observe another kind of spatial variations of g and a fine difference for the two different shear rates: the depletion of particle pairs in close contact is enhanced by high shear rates (Fig. 20), counter-balanced by a growing tail of large values of g appearing at a distance close to the particles diameter ($\xi \simeq d$) from the direction of the flow in the extensional stress domain (for $\ell \gtrsim d$). This tail is quantified and compared for the two shear rates in Figure 20c, where two profiles of the pdf $g_r(\ell)$ at different positions ($\xi = +d$ and $\xi = -d$) are drawn. These fine differences of microstructure may have strong consequences on macroscopic rheology; more precisely, it was shown that the shear stress at the onset of flow (“static” yield stress) strongly depends on the preshear

FIG. 20. a-b) Pdf in the shear plane (θz) for a steady rotational shear of a visco-plastic suspension at a solid fraction $\phi \simeq 35\%$ for a low ($\dot{\gamma} = 10^{-2} \text{s}^{-1}$, a) and a high ($\dot{\gamma} = 10 \text{s}^{-1}$, b) shear rate. c) Pdf averaged for $\rho_{2d} = d \pm d/6$ as a function of the angle ϕ_{2d} in polar coordinates for the two shear rates. d) Some profiles of the pdf at different positions: $g_r(\ell)$ for $\xi = -d$ and $\xi = d$ for the two shear rates

history [5]. This was attributed to the difference in the microstructures jammed at rest after the preshear. As a conclusion, the shear-thinning rheology of the suspending visco-plastic fluid, the presence of a yield stress or its elasticity may be responsible of the influence of the shear rate on the microstructure and the related rheology.

E. Bulk microstructure of a Newtonian suspension after a rotational shear flow

We now analyse the 3D microstructure of a Newtonian suspension in our parallel plates geometry.

It is already known from experiments and numerical simulations that an anisotropic microstructure develops in sheared Newtonian suspensions [7, 10, 14]. However, in the

FIG. 21. 3D pair distribution functions in the shear plane in a parallel plates geometry for a Newtonian suspension of particles: for $\phi \simeq 36\%$ (a) and $\phi \simeq 44\%$ (b)

literature, most pdfs are 2D, in the shear plane. The more reliable data were obtained in a Couette geometry, which presents a stress inhomogeneity in the shear plane, whereas the stress inhomogeneity in the parallel plates geometry we use is in the velocity gradient-vorticity ($z\theta$) plane. Another difference between these two geometries is that shear stresses and shear rates are imposed in a Couette geometry and in a parallel plates one respectively. Endly, whereas the microstructure in [10, 14] is imaged under a flow, we are using a flow-arresting technique[18], so there may be some relaxation of the microstructure. In the following, we compare our 3D pdf obtained in our parallel plates set-up with 2D data obtained in a Couette geometry by Blan *et al.* [14] for the same Newtonian suspension.

In the experiments presented here, we focus on steady simple shear flows controlled by the rotation of the top plate of the parallel plates rheometer. Figure 21 shows the microstructure through pair distribution functions in the three cylindrical ‘planes’ $g_r(\ell, \xi)$, $g_\theta(\rho, \xi)$ and $g_z(\rho, \ell)$: it is not the same in the three planes. $g_r(\ell, \xi)$ becomes anisotropic in the shear plane, while it is still nearly isotropic in the two other ‘planes’. Figure 21 shows the microstructure of two Newtonian suspensions for two different solid fractions $\phi \simeq 36\%$ and $\phi \simeq 44\%$: they are the same.

We can show an extensional-depletion of particle pairs and a flow-alignment of particle pairs. Secondary maxima and minima are observed too here and (experimental and numerical) results from Blanc [14, 24] show that these two local maxima and minima are visible for solid fractions larger than $\phi \simeq 30\%$. For comparison, Figure 22 reports two 2D pdfs measured by Blanc *et al* for $\phi \simeq 35\%$ and $\phi \simeq 45\%$ for the same suspension but in a different

FIG. 22. Pair distribution functions in the shear plane from the literature (Blanc et al [14]) in a Couette geometry (co-centric cylinders) for a Newtonian suspension of particles: in the bulk for $\phi \simeq 35\%$ (a) and $\phi \simeq 45\%$ (b). Here a is the particles radius, whereas d is the diameter. Note the different color scales in a and b.

FIG. 23. Scalar angular pdf in the shear plane: (a) from Blanc et al in a Couette geometry for $\phi \simeq 35\%$ and $\phi \simeq 45\%$ and in our parallel plates geometry for $\phi \simeq 36\%$ and $\phi \simeq 44\%$ for the same Newtonian suspension; (b) in our parallel plates geometry for a Newtonian suspension at $\phi \simeq 36\%$ and a visco-plastic suspension at $\phi \simeq 37\%$ at a low ($10^{-2}s^{-1}$) and high ($10s^{-1}$) shear rate

637 geometry: they found that the angle of the minima θ_{min} of g changes from 21° to 34° . More
638 generally, they observed a strong influence of the solid fraction ϕ on θ_{min} : 4.5° for $\phi = 5\%$
639 and 37° for $\phi = 56\%$ [14].

640 We now compare the values of $g(\rho_{2d}, \phi_{2d})$ averaged close to the particle diameter ($\rho =$
641 $d \pm d/6$) as a function of ϕ_{2d} in the shear plane for Newtonian suspensions and visco-plastic

suspensions in Figure 23. The maxima and minima described above are clearly visible in this representation. The shape and the position of the maxima/minima in the Blanc experiments and in our experiment are in very good agreement. However, there are some quantitative differences. We checked that they are not due to the radial thickness Δ over which data are averaged nor to the width $\delta\rho = d/6$ for the average $\langle g(\phi_{2d}) \rangle_{\rho_{2d}=d\pm\delta\rho}$, neither the position R_0 of the toroidal region where we compute our pdf. To this date, we cannot explain these differences.

We also note that the angular dependence of the pdf $\langle g(\phi_{2d}) \rangle_{\rho_{2d}=d\pm\delta\rho}$ close to contact of particles in visco-plastic fluids is very close to that in Newtonian suspensions. We note however that the amplitude of the minimum and of the maximum change with the shear rate, which is specific to a nonlinear suspending fluid. It seems that more depletion can be obtained in a visco-plastic fluid at a high shear rate than in a Newtonian fluid.

As a conclusion, in our geometry the main difference of microstructure between Newtonian and visco-plastic suspensions is the dependence on the shear rate for visco-plastic and the independence on the shear rate for Newtonian. However, in all cases (visco-plastic or Newtonian, parallel plates or Couette geometry), the microstructure becomes anisotropic in the shear plane characterized by both an extensional-depletion of particle pairs and a flow-alignment of particle pairs at similar angles. Finally, there may be a difference of quantitative values of the pdf (and of the microstructure) of a suspension in a Couette geometry and in a parallel plates one, where shear stresses and shear rates are imposed respectively and where a stress inhomogeneity is in the shear plane and in the velocity gradient-vorticity ($z\theta$) plane respectively. Endly, whereas the microstructure in [14] is imaged under a flow, here we are using a flow-arresting technique[18], so there may be some relaxation of the microstructure.

F. Microstructure near solid walls: macroscopic layering

In our parallel plates geometry, the suspension is confined between two plates and the gap is not so large compared to the size of particles ($H/d \simeq 14$ for the visco-plastic suspension and 12 for the Newtonian one) – this aspect ratio being common for such set-ups. These solid boundaries induce some layering of particles, as shown by the solid fraction profiles as a function of z for smooth and rough surface plates (Fig. 24). Even when the surfaces

FIG. 24. Vertical concentration profiles close to a solid boundary for smooth (a) versus rough (b) surfaces after a rotational shear of $\approx 5s^{-1}$ for a visco-plastic suspension at a solid fraction $\phi \simeq 40\%$.

FIG. 25. Pair distribution function g_r in the shear plane (θz) computed near a smooth solid wall located in the bottom (a) and in the bulk far from any solid wall (b).

672 have been roughened (Fig. 2c and 24b), we may observe a few alternate layers of high and
 673 low solid fraction values, suggesting about 1 layer of aligned particles laying parallel to the
 674 walls. In comparison with the case of smooth surfaces (Fig. 24a), the layering is reduced for
 675 rough surfaces (Fig. 24b), and is localized close to the solid boundaries, whereas layering
 676 comes deep inside the gap for smooth surfaces.

677 This macroscopic layering close to solid boundaries is also visible on pair distribution
 678 functions in the shear plane, when comparing pdfs computed close to solid boundaries with
 679 pdfs computed in the bulk (far from any solid surface) for smooth solid plates (Fig. 25). A
 680 periodic pattern of strips is superimposed on the bulk microstructure. This effect was already
 681 reported by [24, 35] for Newtonian suspensions, so this layering does exist for suspensions
 682 of different rheology.

683 As a conclusion, studying the bulk microstructure imposes to be far enough from solid

FIG. 26. Pair distribution function g_r in the orthonormal plane (θz) computed in the bulk (i.e. $R_0 < 0.60D/2$, $1.2H < Z_0 < 2.2H$) (a) and near the orthonormal free surface (i.e. $0.60D/2 < R_0$, $1.2H < Z_0 < 2.2H$) (b) in the non sheared configuration (drop). The later pdf (b) is not observed anywhere else in the suspension than in the upper part of the suspension drop.

FIG. 27. a-b) Pair distribution function g_r in the orthonormal plane (θz) computed in the bulk (i.e. $R_0 < 0.60D/2$, $0.5H/2 < Z_0$) (a) and near the orthonormal free surface (i.e. $0.60D/2 < R_0$, $0.5H/2 < Z_0$) (b) in the loaded and squeezed configuration. The later pdf (b) is not observed anywhere else in the suspension than in the quarter cylindrical part of the suspension near the top plate.

FIG. 28. Pair distribution function g_r in the shear plane (θz) far from (a, c) and close to (b, d) the orthoradial free interface for a Newtonian (a, b) and a visco-plastic suspension (c, d)

G. Microstructure near free interfaces: alignment of particle doublets with free interfaces

A more subtle structuration is observed here near the orthoradial free interface, probably enhanced when coupled with a shear flow (or with a flow in general). This structuration is observed both in non sheared (drop) and sheared (by squeeze and rotation flows) suspensions (Fig. 26, 27 and 28): this is an alignment of particle doublets (two particles in close contact) with the free interface along $\phi_{2d} \simeq 0^\circ$ and $\phi_{2d} \simeq 180^\circ$. However, whereas this alignment is systematically observed for rotational shear experiments, it is less pronounced and more localized in experiments of squeeze shear and in non loaded drop. This suggests that the occurrence of secondary flows in the squeeze flow (see part IV B) and when pouring the non loaded drop (see part IV A) may play a role in this structuration, due to a coupling between free interfaces and flows. The pair distribution function exhibits a larger maxima in the orthoradial plane $g_r(\ell, \xi)$ for values of $\ell \simeq d$ and $\xi \simeq 0$ when computed near the free

FIG. 29. Radial profiles of density (number per unit volume) of particle pairs aligned with the local flow ($\vec{e}_\theta$) $\phi_{pair}(R_0)$ for a Newtonian ($\phi \simeq 45\%$, a) and a visco-plastic suspension ($\phi \simeq 38\%$, b), for uniform radial profiles of solid fractions $\phi(R_0)$.

interface (Fig. 26, 27 and 28), that is not visible on pdf computed in the bulk (far from the orthoradial free interface). This maxima of the pdf corresponds to an over-population of particle doublets in close contact in the azimuthal (flow) direction; it is counter-balanced by the apparition of a minima (drop, squeeze) or by a decrease of the secondary minima (rotation), resulting in a modification of the shape of the pdf. This feature may recall the flow-induced structural transition (from disorder to order to disorder as the Peclet number is increased) observed in numerical simulations of Brownian suspensions [8, 15–18].

This can be observed at the macroscopic scale by computing ϕ_{pair} , the density (number per unit volume, normalized by the solid fraction) of particle doublets in close contact aligned with $\vec{e}_\theta$ in toroidal regions at different radial positions R_0 (Fig. 29). This density is found to increase close to the free interface in a region of width about $D/8$ (Fig. 29), despite some spatial fluctuations of ϕ_{pair} . We may wonder if this local alignment is due to a steric constraint of the free interface or to capillary stresses due to the contact lines between air, particles and the fluid, and a coupling with solid walls or with some flows.

We observe this specific microstructure near the free interface both for Newtonian and for visco-plastic suspensions in a parallel plates geometry (Fig. 28, Fig. 29). Interestingly, this allows us to provide an explanation for the observation of a peak in the flow direction for the first experimental pdfs of Newtonian suspensions reported in the literature [10] and reprinted here (Fig. 30), which has long been a puzzle. Indeed, pdfs from Parsi *et al.* [10] were computed from optical observations of the first layer of particles on top of

FIG. 30. Pair distribution functions from the literature in a Couette geometry (co-centric cylinders) for a Newtonian suspension of particles: a) close to the top free surface for $\phi = 40\%$ from [10], b) far from the top free surface for $\phi \simeq 35\%$ from [14]; the top free surface being also the shear plane.

a Couette geometry (co-centric cylinders), that is from the observation of particles close to the horizontal free interface, which is also the shear plane. Their results contrast with the pdfs reported by [14], computed from the particle positions in the bulk of a Couette flow and reprinted here (Fig. 30), in which such a peak is absent. The same conclusion can thus be drawn from these studies (near and far from the free interface) as from our observations: some peak appears in the pdf in the shear plane corresponding to an overpopulation of particle doublets in close contact in the flow direction near the free surface (Fig. 28, Fig. 30). Note that in both cases (parallel plates and Couette geometry) the free surface is orthogonal to vorticity; no prediction can yet be made from our observations for a free surface orthogonal to the velocity gradient, as in a flow along an inclined plane.

As a conclusion, studying the bulk microstructure imposes to be far enough from both free interfaces and solid boundaries; this precludes optical studies on non-index-matched suspensions.

V. CONCLUSION

In this paper, we present a technique we developed to image in 3D hard particle suspensions with X-ray tomography. It allows for the three dimensional characterization of the microstructure in space – in different planes and at different positions in the set-up

735 – from particle center positions identified at a sub-voxel resolution, thanks to home-made
736 algorithms for image processing. An accurate characterization of anisotropic microstructure
737 is possible, even for particle pairs in close contact.

738 The dedicated set-ups used here for imaging have been built so that they can also be
739 used in a rheometer to perform rheological measurements. It is thus possible to correlate
740 the 3D-microstructures and the rheological properties for a same shear history (see [5] for
741 an example).

742 To illustrate the technique presented here, we apply it to two materials: a visco-plastic
743 suspension and a Newtonian suspension, meaning that our non-Brownian particles are sus-
744 pended in a visco-plastic and a Newtonian fluid respectively. The Newtonian suspension
745 is the same as used by Blanc *et al.* [14], but it is loaded here in a different geometry and
746 characterized in 3D, both in the bulk and near the interfaces.

747 In principle, all the details of our technique are given so that it should be possible to
748 reproduce them. Our method should work for many suspensions (with a contrast of X-ray
749 between particles and the fluid), as soon as particles do not move during the time of a X-ray
750 scan. This may prevent us from imaging Brownian particles ($d \lesssim 1\mu m$) with a conventional
751 X-ray tomograph but may demand a quicker X-ray source.

752 After a rotational shear, we find approximately the same shear-induced anisotropic pair
753 distribution function (in the shear plane) in the bulk of the Newtonian suspension as in
754 Blanc *et al.* [14]. Moreover, we find that it is nearly isotropic in the two other planes.

755 The visco-plastic suspension is characterized in different configurations. Its microstruc-
756 ture is found to be nearly isotropic in 3D after preparation, showing that it is possible to
757 design such a nearly isotropic suspension. It becomes anisotropic in the shear plane what-
758 ever the shear is (rotational shear or continuous squeeze); loading in any geometry (except
759 the vane-in-cup) thus leads to an anisotropic initial state. The shear-induced pair distribu-
760 tion function for the visco-plastic suspension is found to be similar to that for a Newtonian
761 suspension, except at high shear rates. Indeed, as a consequence of the non-linear nature of
762 the visco-plastic fluid (either the yield-stress or the shear-thinning viscosity), the shear rate
763 changes the anisotropy of the microstructure.

764 For the two hard non-Brownian particles suspensions used here (Newtonian and visco-
765 plastic suspensions), we find some layering close to solid walls, especially if they are smooth,
766 and a more surprising layering close to the free interface. This latter is not as obvious as

FIG. 31. Two particles M and M_0 in the global cylindrical framework and four examples for the definition of the pair vector $\vec{r} \equiv \overrightarrow{M_0 M}$ characterizing their separation: euclidean ($\vec{r} = \overrightarrow{M_0 M}$) or curvilinear – along circular flow lines – coordinates, with orthogonal or not orthogonal axis

the former because it is not strikingly visible on macroscopic quantities. It is visible at the scale of some particle pairs only: it induces a flow-alignment of particle doublets at close contact, in regions of quite large extent close to free interfaces. This effect is likely to be a consequence of a coupling between free interfaces and shear; it allows to explain previous data of the literature [10].

Appendix A: Possible geometrical choices to compute pair distribution functions

Pair distribution functions are computed as normalized probabilities of finding a test particle M located at (r, θ, z) from another reference particle M_0 located at (r_0, θ_0, z_0) in the cylindrical framework attached to the axis of the rheometer circular plates (see Part III C). There are several choices for the characterization of the pair vector $\vec{r}$ characterizing their separation: euclidean ($\vec{r} = \overrightarrow{M_0 M}$) or curvilinear (along circular flow lines) coordinates, orthogonal or not orthogonal axis (unit vectors dependent on M_0 only or both on M_0 and M). Four examples of possible definitions are shown in Figure 31 : $(r \cos((\theta - \theta_0)/2) - r_0, r \sin(\theta - \theta_0), z - z_0)$, $(r - r_0, 2r \sin((\theta - \theta_0)/2), z - z_0)$, $(r - r_0, r_0(\theta - \theta_0), z - z_0)$ and $(r - r_0, r(\theta - \theta_0), z - z_0)$. Euclidean coordinates can in principle be chosen but we believe that non euclidean coordinates, but curvilinear coordinates projected along the cylindrical ‘planes’ aligned with the circular flow streamlines, are more relevant to characterize the microstructure induced by a simple shear flow. We choose for coordinates of the pair vector $\vec{r} = (\rho, \ell, \xi)$: $\rho = r - r_0$, $\ell = r(\theta - \theta_0)$, $\xi = z - z_0$. This choice does not induce any bias in the symmetry of the pair distribution functions g_z , as well as of g_θ or g_r , as shown by the

FIG. 32. Influence on the pair distribution function g_z computed in (θr) planes (in 2D (a), and in 1D (b)) of the definition of the pair vector $\vec{r}$ (for the four definitions written in the text). Our choice, Eq. (10) does not induce any bias in the symmetry of the pair distribution functions between negative and positive values of ρ

comparison in Figure 32 of g_z computed in (θr) planes for reference particles in a toroidal ring at a small radius for the four previous different definitions of $\vec{r}$.

In order to get enough statistics, we need to define a thickness for the elementary planes where we compute 2D pair distribution functions (see Part III C). Whereas the definition of the thicknesses Δz of horizontal planes (θr) and Δr of orthoradial ‘planes’ (θz) is obvious, the thickness of radial planes (rz) offers several choices. Different possible choices for the thickness of radial planes (rz) are: constant euclidean thickness $r \sin(\theta - \theta_0)$, constant wedge angle $\theta - \theta_0$, or constant arc length $r(\theta - \theta_0)$. Figure 33 shows the pair distribution function g_θ computed in (rz) planes (in 2D and in 1D) for the three previous different definitions of the thickness. Whereas the definitions of a constant wedge angle and of a constant arc length show a bias (there is an asymmetry between the left and right sides), the definition of a constant euclidean thickness does not. That is why we choose to work with radial planes (rz) of thickness $\Delta h = r \sin(\theta - \theta_0)$.

FIG. 33. Influence on the pair distribution function g_θ computed in radial (rz) planes (in 2D (a) and in 1D (b)) of the definition of their thicknesses (for the three different definitions written in the text). The choice of a constant euclidean thickness $\Delta h = r \sin(\theta - \theta_0)$, Eq. 9, does not induce any bias in the symmetry of the pair distribution functions between negative and positive values of ρ

Appendix B: Statistical precision of pair distribution functions

Here we discuss the statistics in terms of number of particles required for fine sampling and good precision of the pdf g from its following expression (see Part III C), where $N^{pair}(\vec{r} \pm \delta\vec{r})$ is the number of pairs of particles separated by $\vec{r} \pm \delta\vec{r}$ and N_{ref} is the total number of the reference particles:

$$g(\vec{r} \pm \delta\vec{r}) = N^{pair}(\vec{r} \pm \delta\vec{r})/N_\infty \quad (\text{B1})$$

with:

$$N_\infty = N_{ref} n_{0v} \delta V, \quad (\text{B2})$$

the number of pairs separated by a vector $\vec{r}$ such that $||\vec{r}|| \rightarrow \infty$ in the sampling volume δV in the space of pair vectors delimited by $\vec{r} \pm \delta\vec{r}$. Note that $n_{0v} \delta V$ is the number of particles in the sampling volume δV . The precision Δg on $g(\vec{r})$, or the minimal detected variation, corresponding to the variation of the integer $\Delta N^{pair} = 1$, is:

$$\Delta g = 1/N_\infty = (N_{ref} n_{0v} \delta V)^{-1}. \quad (\text{B3})$$

FIG. 34. Scalar pair distribution function $g(||\vec{r}||)$ for different numbers of reference particles N_{ref} increasing from arbitrary small values to larger values (but for constant numbers of reference particles with $||\vec{r}||$, i.e. non adaptative spheres).

We will usually compute the pdf on toroidal rings of thicknesses ΔR and ΔZ centered on R_0 and Z_0 (of volume $\vartheta = 2\pi R_0 \Delta R \Delta Z$), so that the number of reference particles is approximately:

$$N_{ref} \simeq n_{0v} \vartheta. \quad (\text{B4})$$

For a given density n_{0v} and chosen precisions on the spatial resolution δV and on the pdf Δg , this prescribes a minimum number of reference particles from Eq. (B3):

$$N_{ref} = (n_{0v} \delta V \Delta g)^{-1}, \quad (\text{B5})$$

or equivalently a minimum volume ϑ for the toroidal ring from Eq. (B4).

As a numerical example, if we want a precision $\Delta g = 10^{-2}$ on $g(\vec{r})$ and a spatial resolution for the 2D pdf $\delta V = \Delta \delta^2$ with $\Delta = d = 140\mu\text{m} = 12\text{vx}$ and $\delta = 1\text{vx} = 12\mu\text{m}$, for a solid fraction $\phi = 35\%$ and $d = 140\mu\text{m}$ ($n_{0v} = 244\text{mm}^{-3}$), we need at least $2 \cdot 10^4$ reference particles in the region of interest. Otherwise, we may see the discrete nature of variations of $g(||\vec{r}||)$ (Fig. 34). This effect of discretization may appear anyway for large inter-particle distances, because of the use of adaptative spheres. As a compromise, these spatial averages may lead to some underestimate of peak values of g , whether there are large variations of g .

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