EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Granular Matter under Shear: LiquidMorphologies in Real Time	**Experiment number**: MA-187
Beamline: ID 15A	**Date of experiment**: from: 04.10.2006 to: 10.10.2006	**Date of report**:
Shifts: 18	**Local contact(s)**: Marco DiMichiel	*Received at ESRF:* *15*

Names and affiliations of applicants (* indicates experimentalists):

Ralf Seemann*, MPI for Dynamics and Self-Organization, Goettingen, Germany

Mario Scheel*, MPI for Dynamics and Self-Organization, Goettingen, Germany,

Stephan Herminghaus*, MPI for Dynamics and Self-Organization, Goettingen, Germany

Marco Di Michiel, E.S.R.F., Grenoble, France

Report:

It is generally known from everyday experience that the mechanical properties of wet granular material change dramatically if some liquid is added. In general, a stiffening of the granulate is observed. The main reason is the internal cohesion due to capillary forces arising from liquid bridges between the grains [1-7]. While recent years have seen considerable progress in understanding the dynamics of dry granular materials [8-11], the physical mechanisms underlying the properties of wet systems remain largely obscure. This is in part due to the inherent complexity of these highly nonlinear systems, but also due to the fact that only a few experiments have been performed on this topic so far. We thus investigated the network of capillary bridges in a wet granular medium consisting of spherical glass beads [12,13]. When the granulate is sheared, the position of the spheres change. Accordingly the capillary bridges and liquid cluster might break, change their size and shape, or reform. Because liquid bridges need some time to form and grow, the size and number of bridges or clusters, is expected to depend on the shear rate. This results in a shear thinning behavior of wet granulates for high shear rates [13].

In a wide range of liquid content, about six capillary bridges per sphere (coordination number) were found, resulting in a stiff granulate. For larger liquid contents the liquid forms extended liquid clusters and the stiffness of the granulate is expected to be reduced. Whereas in our experiments, the yield stress of the granulates is about constant for a very large range of liquid contents exceeding the regime with coordination number six by far, reaching into the regime where large liquid clusters are present [13]. Hence, the time resolved knowledge of the distribution and shape of liquid bridges and clusters and the corresponding matrix consisting of glass spheres is needed for a quantitative understanding of the mechanical properties of wet granulates.

We explored the dynamic behavior of capillary bridges and liquid clusters within a sheared, wet granulate and how the liquid bridges and clusters change their size and shape as a function of shear rate. The granular material consisted of fairly monodisperse glass beads with a diameter ranging from 250 to 600 µm. The spread in bead size is from a few percent to about 10%, enough to prevent crystallization. As the wetting liquid we used water with a significant amount of dissolved ZnI_2 to increase the X-ray absorption contrast. The different absorption allowed for a clear distinction between the air, the glass beads, and the liquid. Using this system, radiation damage was not observed. Heating effects could be suppressed by reducing the X-ray intensity or giving the sample some time after taking a tomography with high X-ray energy.

The wet granulates were prepared in a small custom made shear cell with an inner diameter of 9 mm, which is lightweight and could be mounted on the rotation stage of the tomography setup. The shear force was applied and measured by computer controlled microfluidic pumps and pressure sensors connected to the shear cell via hydraulic lines. We could shear the granulate using the syringe pumps with a well defined shear volume. The differential pressure in the shear cell could be recorded while keeping the absolute pressure constant. The shear rates were about 0.001 Hz while the corresponding shear velocities were about 1 µm/s. The time resolution to capture a full 3D tomography of about 15 s (~ 4 s operating the CCD camera in binning mode), is sufficient to freeze the motion of the glass spheres, the stretching and reformation of capillary bridges and liquid clusters.

As can be seen in Figure 1, imaging of a sheared granulated worked satisfactory even at reduced beam intensity respectively reduced exposure time to protect the wet granulate from damages due to heating effects. We could clearly detect the liquid and the glass spheres and resolve the breaking and formation of capillary bridge as a function of shear amplitude as shown in greater detail in Figure 2.

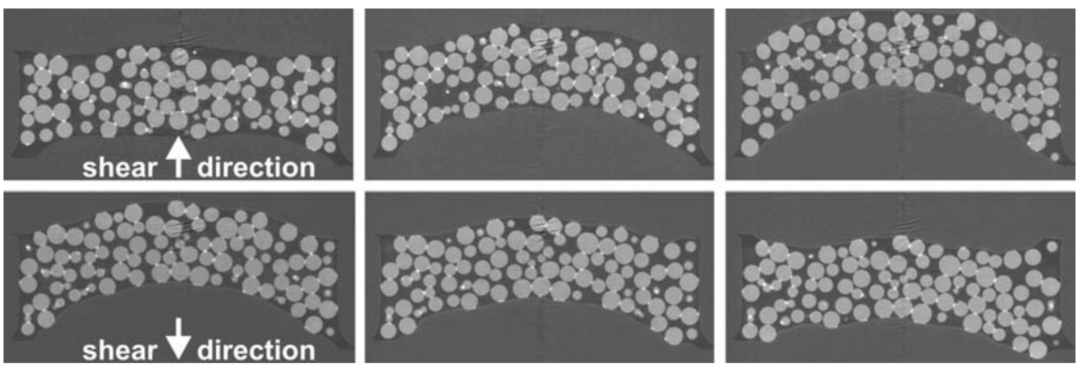

Figure 1 Time series showing 2d slices of 3d X-ray tomography images of a sheared wet granulate. Every fivth image during half a shear cycle is displayed. The sphere diameter was about 500 µm, the liquid content was W = 0.01. The rubber membrane to apply the shear force is about 70 µm thick and is hardly visible in the images.

For a precise measurement of the differential pressure (and thus the yield stress) of the sheared granulate during the X-ray experiments we applied the shear force via very thin rubber membranes. But as it turned out from the reconstructed 3D images, the thin rubber membranes, used in our experiment, were not stiff enough to guarantee a parabolic shear profile in all experiments and thus to prevent ‘avalanche effects’ at the side walls of the shear cell. This effect could not be detected optically from “outside” and was only visible in the X-ray tomography images. Furthermore, the thin rubber mebranes changed their elastic modulus during X-ray exposure – an effect that was never observed with thicker membranes. A slight change in the design of the shear cell allows us to replace the thin rubber membranes by thicker ones stiff enough to guarantee a parabolic shear profile. We measured sheared granulates with different liquid contents between W = 0.01 and W = 0.1, different shear rates, and different sphere sizes.

First analysis of the results at low shear rate indicate that there is a significant number of capillary bridges, which break or form when the packing geometry changes when a wet granulate is sheared. This is demonstrated in Figure 2a with close up views of 2d slices of a sheared sample with a liquid content of W = 0.03. But even that the liquid bridges reorganizes during the shear cycle, the average number of capillary bridges within the granulate remains constant and does not differ from the number of bridges in a static sample as displayed in Figure 2b for one complete shear cycle. In contrast to a static sample, where the capillary bridges have time to equilibrate and the volume of the individual capillary bridges is almost identical, the volume of the individual liquid bridges seem to vary significantly. But because the attractive force a capillary bridge exerts on the spheres is largely independent on its volume it is clear that yield stress of a granulate at low shear rates is largely independent on the shear.

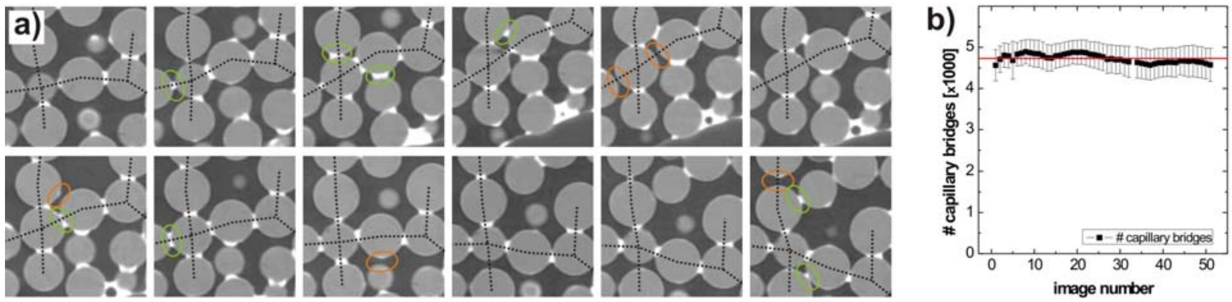

Figure 2 a) Time series showing close up 2d slices of 3d X-ray tomography images of a sheared wet granulate. The sphere diameter was about 500 µm, the liquid content was W = 0.03. The first four images are sheared from bottom to top, the remaining images are shered from top to bottom. Every fourth image during ~2/3 of a shear cycle is displayed. The center of some spheres are connected to visualze the motion of the spheres relative to each other. Points where a capillary bridges was just formed are indicated by a greed ellipse, points where a capillary bridge just ruptured are displayed in orange. b) Absolute number of capillary bridges in the sheared granulate during a complete shear cycle.

Parallel to the rupturing and forming of capillary bridge in a sheared granulate, we performed tests to record the formation and rupturing of capillary bridges while vapour condensing/evaporating liquid in the granulate. We used dibrommethane as a liquid with reasonably large vapour pressure and large X-ray absorption. The sample consisting of a dry granulate on top of a liquid reservoir was mounted on a hot plate with a diameter of about 1.2 cm diameter. When slightly heating the sample, the liquid evaporated from the reservoir and condensed in the granulate forming liquid bridges between individual spheres predominantly at points of physical contact. When reducing the temperature of the reservoir the liquid bridges slowly evaporated. Using the same tomography setup and similar imaging parameter as used for the shear cell experiments we could image the capillary bridges for increasing and decreasing volume *in-situ*.

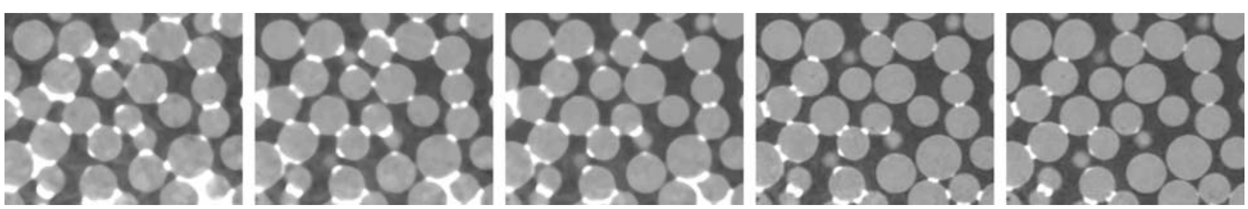

Figure 3 Time series showing close up 2d slices of 3d X-ray tomography images of a granulate where dibrommethane was evaporated. Every fivth image of the cooling cycle is displayed. The sphere diameter was about 500 µm.

[1] D. J. Hornbaker, R. Albert, I. Albert, A.-L. Barabasi, and P. Schiffer, *Nature* 387, 765 (1997)
[2] T. C. Halsey and A. J. Levine, *Phys. Rev. Lett.* 80, 3141 (1998)
[3] L. Bocquet, E. Charlaix, S. Ciliberto, and J. Crassous, *Nature* 396, 735 (1998)
[4] N. Fraysee, H. Thomé, and L. Petit, *Eur. Phys. J. B* 11, 615 (1999)
[5] R. R. Hartley and R. P. Behringer, *Nature* 421, 928 (2003)
[6] G. Ovalez, E. Kolb, and E. Clement, *Phys. Rev. E* 64, 060302(R) (2001)
[7] D. Geromichalos, M. M. Kohonen, F. Mugele, and S. Herminhaus, *Phys. Rev. Lett.*, 90, 168702 (2003)
[8] G. H. Ristow, *Pattern Formation in Granular Materials* (Springer, Berlin, 2000)
[9] H. M. Jaeger, S. R. Nagel, and R. P. Behringer, *Rev. Mod. Phys.* 68, 1259 (1996)
[10] L. P. Kadanoff, *Rev. Mod. Phys.* 71, 435 (1999)
[11] Aleksandar Donev at al., *Science* 203, 990 (2004)
[12] M. M. Kohonen, et al., *Physica A*, 339, 7 (2004)
[13] Z. Fournier, et al. *J. Phys.: Condens. Matter* 17, S477 (2005)