Experimental report

15/02/2022

Proposal:	**UGA-121**	**Council:**	10/2020
Title:	Fluid distribution in partially saturated hydro sensitive aggregates		
Research area:			

This proposal is a new proposal

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Samples: Wheat grains (cous cous), Ch2Oh, in 1mm granules (not powder)

Instrument	Requested days	Allocated days	From	To
D50 T	2	2	22/02/2021	24/02/2021

Abstract:

Fluid distribution in partially saturated hydro sensitive aggregates

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1. Introduction

The behaviour of hygroscopic granular materials is highly influenced by chemo-hydro-mechanical interactions. Of particular interest for food or pharmaceutical industry, such materials can undergo many processes caused by water-content increase, which consequently severely affects the materials functionality, and resource loss (1; 2; 3; 4).
There are several phenomena that can occur at the grain scale, such as swelling, agglomeration, loss of stiffness and strength, which can ultimately lead to caking, a deleterious phenomenon in which the particles stop behaving discretely, become "rubbery", agglomerate and induce to deformations and lower flowability (1; 3; 4). Despite the importance of caking in industrial processing, very little is known about it, specifically about the link between the microscopic processes and the macroscopic material response (4).
The UGA-121 experimental campaign followed the work done within the ESR11 Caliper ITN project (https://caliper-itn.org/), and partially presented in the conference paper by Vego et al. (2021) (5). In this study, a number of x-ray in-operando campaigns were performed to study the evolution of the granular skeleton, and developed a number of image analysis tools (based on Discrete Digital Image Correlation) to follow each of several thousands grains across more than a hundred tomographies (4+ days). It was possible to study the evolution of the inter- and intra-granular strains, the porosity and to follow the evolution of the granular contacts. The key missing ingredient to complete the understanding of the processes is therefore the spatial distribution of moisture driving the phenomenon. Purpose of this campaign was to use the unique combination of high neutron flux/high resolution and of the combined x-ray imaging available at NeXT (6) to study at once the evolving distribution of moisture and the evolution of the granular skeleton.

2. UGA-121 Experimental campaign

2.1. Material

The material selected for the experimental campaigns is fine couscous. It was specifically selected because it presents several of the processes characterising hygroscopic materials; notably it can swell, it can develop agglomeration, and its mechanical properties depend on water-content. Additionally, the size of fine couscous (about $0.8mm$ equivalent diameter) is well within the optimal parameters for imaging.

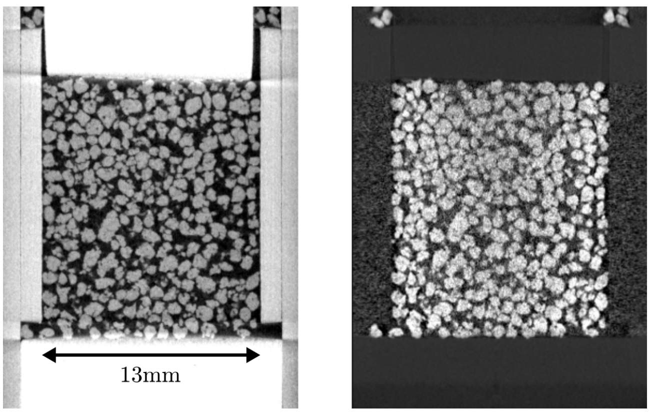

Figure 1: Vertical slices of 3D grey-scale images of the couscous specimen at dry conditions ($t = 0.0h$). On the left the attenuation map from x-ray tomography, while on the right the one from neutron tomography.

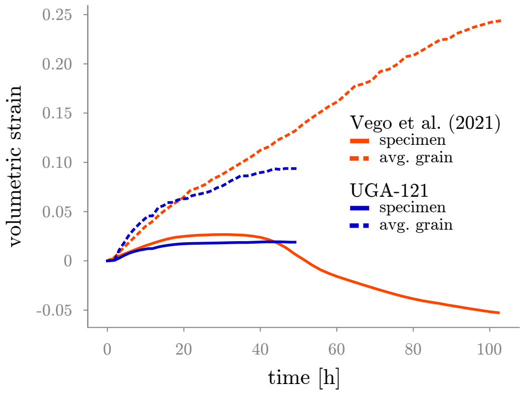

Figure 2: Comparison between the evolution of macroscopic and average grain volumetric strain of both the UGA-121 and the Vego et al. (5) experimental campaigns.

2.2. Setup and applied boundary conditions

The objective was to reproduce a real-case scenario, such as one that could be found in a storage silo. A sample of couscous was tested under oedometric conditions, presumably the best one to reproduce that state. Meanwhile, the material was exposed to 97% relative humidity air, injected with a peristaltic pump at the bottom of the cell. A constant stress of $35kPa$ was applied over the specimen. The sample diameter was about $13mm$, while its height $16mm$. The initial aimed porosity was 50%.

2.3. Tomography settings

A first tomography was acquired before starting the injection as a reference dry state for the following analyses. Then, the peristaltic pump was turned on and tomographies were acquired continuously.
Each x-ray or neutron tomography comprised 992 projections, and it was about $1.5h$ long. The aimed voxel-size was of $32\mu m$, in order to have about ≈ 30 voxels across a grain, while the temporal resolution was controlled by the intrinsic rate of the processes.
A vertical slice of the first reconstructed image from x-ray and neutron tomography in showed in figure 1.

2.4. Major results

All the analyses if the acquired images were performed using 'spam' (7).
The evolution of the average grain volumetric strain was compared to the macroscopic response of the sample. The swelling of the grains induced a macroscopic expansion, a response similar to the one measured by Vego et al. (2021) (5) (see figure 2). It was not possible to observe likewise a compaction stage, both due to limited allocated time and particles that obstructed the descent of the oedometer's ram (figure 1).
From the x-ray images dataset, all the 3014 grains of the sample were tracked and their kinematics and deformation computed. A swelling vertical gradient was detected. Particles at the bottom, closer to the injection point of high RH air increased in volume sooner than others, as shown in figure 3a.
Along with the volumetric strain study, it was possible to also observe the phenomena also from the neutron set of images. Knowing the position of the particle, the average neutron grey-value was computed for each grain and compared with the its initial value at dry conditions. Despite the effect of beam hardening, a vertical gradient was again observed, coherently with the volumetric strain evolution (figure 3b).
A general increase of inter-particle contact areas was also detected from the measurements of the x-ray images.

3. Conclusions and future developments

Thanks to the complementary of neutron and x-ray tomography, it was possible to investigate some of the microscopic processes that lead to caking.
Couscous grains adsorbed water so to reach an equilibrium with the imposed environmental conditions. It was possible to detect the observe the water-content increase from the neutron images, an increase that also triggered the swelling, which was quantitatively measured. Both the neutron and x-ray techniques highlighted a gradient of water-adsorption/swelling driven by the point of air injection and flow rate.
The data collected during these campaigns is uniquely valuable, but the experiment could be still improved. A smaller diameter cell could be used, to decrease the beam hardening effect, as well as remove the grains that

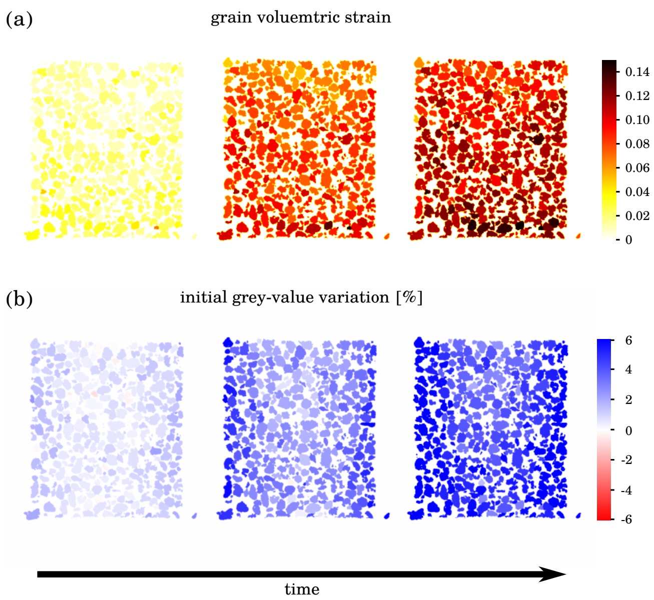

Figure 3: Maps of single grains volumetric strain (a) and neutron grey-value variation (b) at different time-steps. A vertical gradient of particle swelling was detected, since those to the bottom were closer to the high RH source. A gradient was detected as well from neutron images although affected by beam hardening that make look the inner particle less attenuating.

obstructed the eventual sample compression or further expansion.
On the other hand, from the acquired images, it would be of great significance to gather a water-content vs. neutron attenuation relation, which would consequently be linked to volumetric strain of the grain. This information would be crucial for future calibration of numerical models, which would allow to explore more elaborated conditions, which are difficult to impose experimentally on such small samples.

References

[1] J. Aguilera, J. del Valle, and M. Karel, "Caking phenomena in amorphous food powders," *Trends in Food Science & Technology*, vol. 6, no. 5, pp. 149–155, 1995.

[2] K. Brockbank, B. Armstrong, and J. Clayton, "Measurement and quantification of caking in excipients and food products with emphasis on the non-homogeneous interaction with ambient moisture," *Particuology*, vol. 56, pp. 75–83, 2021.

[3] J. J. Fitzpatrick and L. Ahrné, "Food powder handling and processing: Industry problems, knowledge barriers and research opportunities," *Chemical Engineering and Processing: Process Intensification*, vol. 44, no. 2, pp. 209–214, 2005.

[4] U. Zafar, V. Vivacqua, G. Calvert, M. Ghadiri, and J. S. Cleaver, "A review of bulk powder caking," *Powder Technology*, vol. 313, pp. 389–401, 2017.

[5] I. Vego, A. Tengattini, E. Andò, N. Lenoir, and G. Viggiani, "X-ray tomographies of a water-sensitive granular material (couscous) exposed to high relative humidity: an experimental study," in *EPJ Web of Conferences*, vol. 249, p. 08012, EDP Sciences, 2021.

[6] A. Tengattini, N. Lenoir, E. Andò, B. Giroud, D. Atkins, J. Beaucour, and G. Viggiani, "Next-grenoble, the neutron and x-ray tomograph in grenoble," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 968, p. 163939, 2020.

[7] O. Stamati, E. Andò, E. Roubin, R. Cailletaud, M. Wiebicke, G. Pinzon, C. Couture, R. C. Hurley, R. Caulk, D. Caillerie, *et al.*, "spam: Software for practical analysis of materials," *Journal of Open Source Software*, vol. 5, no. 51, p. 2286, 2020.