Experimental report

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Proposal: 1-05-90 **Council:** 4/2021

Title: Chemo-hydro-mechanics of partiallysaturated hydro-sensitive granularmedia

Research area: Engineering

This proposal is a resubmission of 1-05-70

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Samples: C, H, O, Si, Al, Ca

Instrument	Requested days	Allocated days	From	To
NEXT	5	3	31/05/2021	03/06/2021

Abstract:

Several engineering problems originate from the complex chemo-hydro-mechanical interactions of granular media. In food and pharmaceutical industries, most materials (e.g., rice, pasta, sugar, ...) are highly affected by the presence of humidity and/or moisture. While the role of moisture on the mechanical response of impermeable granular media is relatively well understood, organic materials have the additional complexity of the inter- and intra-granular chemical interaction with water (e.g., grain agglomeration because of the production of mucilage, swelling-induced stresses, caking, etc). Purpose of this proposal is to study the spatial distribution of the moisture driving these phenomena (both within and in-between grains). This follows a series of in-operando x-ray experimental campaigns performed by our group and focussing the evolution of the granular skeleton and which allowed the quantification of inter- and intra-granular strains. The combined use of neutron tomography (to study the evolving moisture distribution) and of simultaneous x-ray imaging (to study the evolving granular skeleton) is essential to understand their complex interplay.

Chemo-hydro-mechanics of partially saturated hydro-sensitive granular media

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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 1-50-90 experimental campaign followed the work done within the ESR11 Caliper ITN project (https://caliper-itn.org/), presented in a conference paper by Vego et al. (2021) (5) and in the experimental report UGA-121. As presented in the article, 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.

As explained in the UGA-121 experimental report, thanks to the complementary of neutron and x-ray tomography (6), 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. Furthermore, both the neutron and x-ray techniques highlighted a gradient of water-adsorption/swelling driven by the point of air injection and flow rate.

Purpose of the 1-50-90 campaign is to conduct a similar campaign to the UGA-121, improving the setup so to avoid the so called *beam hardening* effect and capture a complete dilation-compaction response observed by Vego et al. (2021) (5).

2. 1-50-90 experimental campaign

2.1. Material

The material selected for the experimental campaigns was fine couscous. It was specifically selected for this and previous campaigns 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.

2.2. Setup and applied boundary conditions

As mentioned in section 1, the objective was to perform a similar experiment to the UGA-121 one, trying to reduce the beam hardening effect. A new oedometer of $7mm$ diameter was designed and fabricated. Originally, the cylindrical cell was made of Teflon, but it proved not to be rigid enough to sustain the dead-load applying a constant stress of $35kPa$. A new one in aluminium was then made.

The particles were poured in the cell (between two porous stones), with an aimed initial porosity of 50%.

The specimen was then exposed to 97% relative humidity air, injected with a peristaltic pump connected to the bottom of the cell.

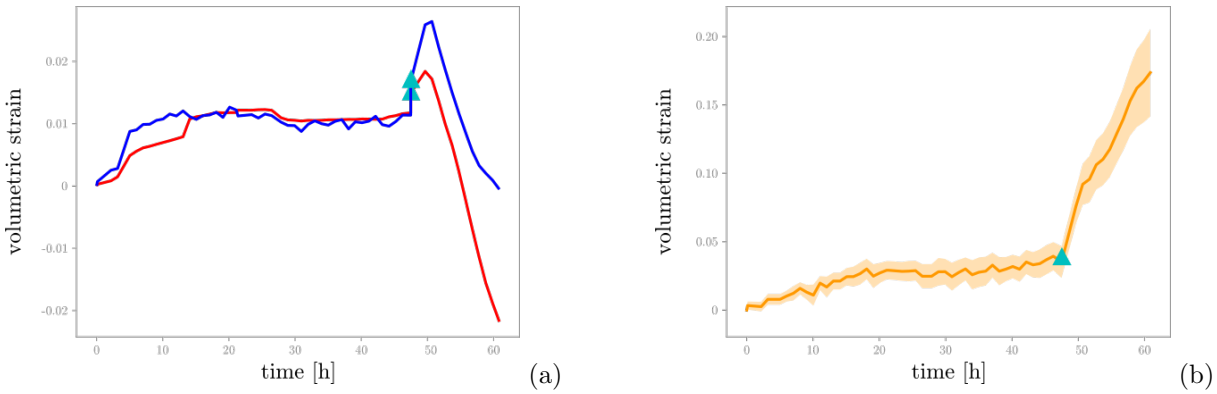

Figure 1: Volumetric response of couscous particles during hydration. On the left (a), the macroscopic volumetric strain measured from both the x-ray (red) and neutron (blue) sets of images. After an initial dilation, no significant variations were measured. Therefore, the flow rate was increased and the specimen dilated due to the particles swelling, but then compacted as they also lost mechanical capacity.
On the right (b), the evolution of the average particle volumetric strain shows how initially the particles swelled up to 2.5% and then after increasing of flow rate they swelled even faster, even during the macroscopic compaction stage.

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 1200 projections, and it was about $1h$ long. The aimed voxel-size was of $15\mu m$ for the neutron images and $25\mu m$ for the x-rays ones, in order to have respectively about ≈ 50 and ≈ 30 voxels across a grain. The temporal resolution was controlled by the intrinsic rate of the processes.

To be noted that after accidental crashes of the camera of the neutron detector, both the detectors (x-rays and neutrons) where in turn re-calibrated.

2.4. Major results

All the analyses if the acquired images were performed using 'spam' (7).

The volumetric evolution of the sample was measured in real-time. As shown in figure 1a, the digital correlation of neutron and x-rays images revealed that after an initial macroscopic dilation of about 1%, for many hours nothing significant was happening. This was likely due to some defects of the new aluminium cell, which led to infiltration of lower RH air in the cell, and the particles did not swell as much as in previous the previous campaigns. To bypass the infiltration problem, it was decided to increase the flow rate from the peristaltic pump by a factor of 10 (from $0.015L/min$ to $0.15L/min$).

The response to this new conditions was immediate. The specimen dilated up to about 2% and then compacted under the constant stress applied as the particles became less and less stiff (see figure 1a).

All the 523 particles comprising the sample were tracked and their kinematics and deformation measured. In figure 1b is shown the evolution of the average particle volumetric strain. As for the macroscopic scale, the particles stopped swelling after about $18h$ and then, as the flow rate was increased, they started again to increase in volume, even during the macroscopic compaction stage.

The particles volumetric strain was compared to the greyvalue (i.e., attenuation) variation of neutrons and x-rays (see figure 2). As the particles swelled their density slightly decreased and so did the x-ray grey-value (the absorbed water contribution to the overall x-ray attenuation is practically negligible). On the other hand the neutron attenuation increased together with the swelling.

3. Conclusion

Thanks to the complementary of neutron and x-ray tomography, it was possible to investigate the volumetric response along with the variation of hydrogen (thus, water) in the sample.

The two initial goals of having the experimental campaign (images less affected by beam hardening and full dilation-compaction response) were achieved.

Despite the infiltration of lower RH air in and the several re-calibration (and consequent effect on the greyvalues), it was possible to measure the macroscopic volumetric strain from both sets of images and finally all the particles were tracked and their kinematics and deformation measured. The response at the macroscopic-

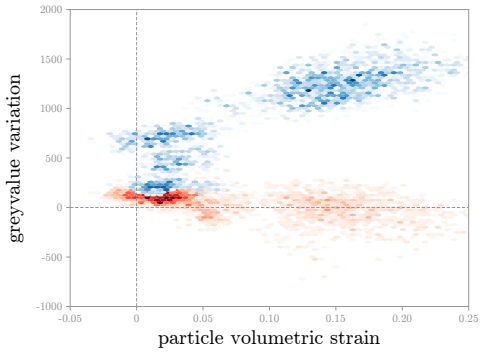

Figure 2: Volumetric response of couscous particles during hydration. On the left (a), the macroscopic volumetric strain measured from both the x-ray (red) and neutron (blue) sets of images. After an initial dilation, no significant variations were measured. Therefore, the flow rate was increased and the specimen dilated due to the particles swelling, but then compacted as they also lost mechanical capacity.
On the right (b), the evolution of the average particle volumetric strain shows how initially the particles swelled up to 2.5% and then after increasing of flow rate they swelled even faster, even during the macroscopic compaction stage.

and particle-scale was found to be coherent with previous experimental campaigns and variations of boundary conditions.

The particles volumetric strain was compared to greyvalue variation of both neutrons and x-rays. From this comparison, important information could be acquired to obtain a distribution of the water content inside the sample and understand how it evolves with time.

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.