X-RAY MICROTOMOGRAPHIC STUDIES OF LC^3 AND RELATED BINDERS

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Extended Abstract

LC^3 type cements do not need introduction in this conference, neither the importance of studying the microporosity features (amount, size(s), connectivities, etc.). However, analysing the porosity in an accurate, quantitative and robust way is far from easy/well-established. Here, we intend to contribute by analysing selected microporosity features and their evolution using synchrotron (and laboratory) X-ray microtomography (μCT) with the final goal of gaining a deeper insight into the microstructures of LC^3 binders. Nowadays, synchrotron μCT allows obtaining tomograms in binders with voxel sizes of 0.3 μm and true spatial resolution close to 1.0 μm [1]. Additionally, for this work, the detector was placed at 5.5 mm from the samples and therefore, phase-propagation increased the visibility between components with similar attenuations to the X-rays. The Paganin phase retrieval algorithm [2] was employed which resulted in augmented contrast. Moreover, another advantage is that fast acquisition times are possible, i.e. a full high-resolution tomogram can be recorded in 5 minutes.

Synchrotron μCT data were taken at TOMCAT beamline (Swiss Light Source, PSI) at 7 and 28 days of hydration for two systems: (i) PC-MK-Cc-G which contains 52 wt% of a commercially available Portland cement (PC-42.5R), 30 wt% of pure kaolinite calcined at 800ºC (MK), 15 wt% of limestone and 3 wt% of gypsum; and (ii) PC-MK-G system which contains 67 wt% of the same PC, 30 wt% of MK and 3 wt% of gypsum. The outputs for these unpublished series will be related to our recent publication [3] where a LC^3 binder was studied in identical experimental tomographic conditions. This LC^3 binder was formulated with 52 wt% of PC, 30 wt% of a calcined clay (containing ~80 wt% of kaolinite), 15 wt% of limestone, and 3 wt% of gypsum. The pastes were hydrated for 7, 28, and 60 days. Moreover, complementary techniques, such as laboratory X-ray powder diffraction have also been employed.

To illustrate the gain in information by using optimised phase-contrast synchrotron μCT, Figure 1 displays orthoslices of the LC^3 binder, hydrated for 60 days, with the standard, i.e. absorption contrast reconstruction, and with the Paganin (propagation-based) reconstruction. In the histogram plots, it can be seen that the 15 wt% of calcite is easily disentangled in the phase-contrast data but strongly overlapped in the absorption data. This observation opens the way to follow calcite reactivity (and possible activation) which is very important in LC^3 systems because of the synergistic effect [4] of limestone with the aluminate-rich fraction of the SCMs. Limestone is difficult to follow by powder diffraction because of its preferred orientation along [104] direction and also due to the possible accidental carbonation of the paste(s). *In situ* X-ray imaging allows, at least in theory, to follow the partial dissolution/reaction of the pristine limestone particles in the starting binder, where neither accidental carbonation nor preferred orientation effects should contribute. Moreover, the capillary porosities up to the spatial resolution attained ~1.0 μm, can also be followed. For this LC^3 binder, it was shown in [3] that the measured total porosities were 16.6, 10.0 and 2.4 vol% at 7, 28 and 60 days of hydration, respectively. Chiefly, the porosity connectivities decreased from 92% at 7 hydration days to 9% at 60 days, contributing to explain the good durability properties of these binders. Similar synchrotron μCT porosity studies will be presented for PC-MK-Cc-G and PC-MK-G. A drawback of the employed approach is that capillaries with different specimens (mixing the same initial powders) are scanned and hence a larger degree of variability has to be accepted. Conversely, with laboratory μCT the same capillary/specimen can be followed with time, given more robust results, but with worse spatial resolution. In this context, the strength and advantages but also the weaknesses and drawbacks of the microtomographic approach (both synchrotron and laboratory) will be discussed.

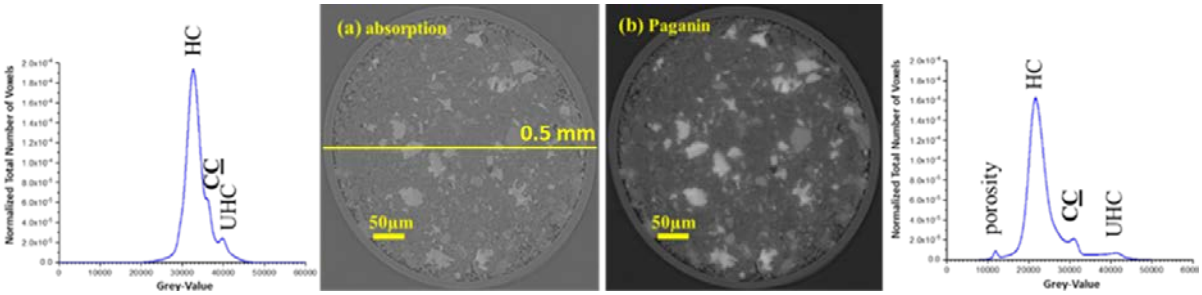
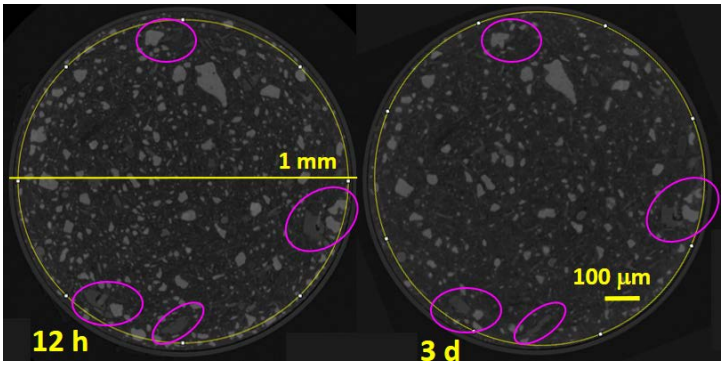


Figure 1. Synchrotron µCT orthoslices (a) attenuation- (b) Paganin- reconstructions for a LC³ binder after 60 hydration days. The histogram traces (blue) show the increase of contrast in (b): i.e. limestone (CC) is shown as a shoulder in the absorption plot but resolved in the Paganin one (HC: hydrated cement products, UHC: unhydrated cement particles), adapted from [3].

In addition, easily accessible lab-µCT [in our case a Bruker SKYSCAN 2214 equipment, taking data in capillaries of 1 mm diameter which allows reproducible sample preparation] can be used to follow the reaction of amorphous phases in 3D. The additional advantage is that MoKα_1 powder diffraction data can be collected in the same regions of the same capillaries [5]. Powder diffraction alone is very limited to characterise the pozzolanic reaction where MK reacts with portlandite to yield C-A-S-H. This reaction involves two nearly amorphous phases in the presence of relatively large quantities of (pristine) nearly-amorphous C-S-H. However, if the same region is 3D scanned with time, the MK particles can be followed, see Figure 2, and the volume-based degree of reaction can be quantified. Merging powder diffraction and tomographic data could provide more robust pozzolanic reaction characterization.



However, because the attenuation coefficient of MK is close to those of hydrated components, global thresholding segmentation is not possible. A more sophisticated approach is required where the size and shapes of the particles are used. Here, Machine Learning (ML) segmentation could be very advantageous.

Figure 2. Laboratory µCT data for a paste of 67 wt%-PC, 30 wt%-MK, and 3 wt%-gypsum with a w/c ratio of 0.50. (Left) Data collected 12 hours after water mixing. (Right) Data collected after 3 days of hydration. Selected reaction of MK particles (darker than the original cement particles) is highlighted in pink. µCT data have been also collected at 1, 2, 7 and 34 days, and ML segmentation with IPSDK (Image processing) software is being attempted. Details of our ongoing efforts will be presented.

Keywords: synchrotron microtomography, laboratory µCT, microstructure, porosity, amorphous

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