Quantification and analysis of amorphous components in cements by synchrotron ptychographic X-ray tomography

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Building materials have complex hierarchical microstructures. To fully understand their main properties a sound description of their spatially-resolved contents is compulsory. Developing this knowledge is challenging as about half of the volume is amorphous/nanocrystalline. Furthermore, more than one amorphous component can coexist which is even more challenging to quantify with standard techniques.

On the other hand, ptychographic X-ray computed tomography (PXCT) is a non-destructive X-ray imaging technique which retrieves the full refractive index of the sample, 3D electron density – rho_e– (very related to mass density) and attenuation coefficient –mu– distributions, with a 3D isotropic resolution, nowadays slightly better than 100 nm for cement pastes. Crystallinity is not required and spatial analysis can be carried out by a set of tools including segmentation of the different material phases in the 3D dataset(s). Acknowledging the intrinsic lower spatial resolution in the reconstructed absorption tomograms, the importance of having both (rho_e and mu tomograms) will be highlighted for studying complex samples with amorphous components.

Here, we will review our ongoing efforts to contribute to the understanding in cement science at the mesoscale by analysing and quantifying the amorphous component(s). This is key in cements as volume stability of buildings is key and mass density values (and their spatial distribution) must be known, which is far from trivial for amorphous components intermixed with crystalline phases.

Firstly, we will discuss our work on Portland cement [1] where three unaltered samples: neat Portland cement (PC) paste, PC-calcite and PC-fly ash blended pastes, were analysed. For the neat PC paste, the PXCT study gave densities of 2.11 and 2.52 gcm^{-3} and contents of 41.1 and 6.4 vol% for nanocrystalline calcium silicate hydrate (C-S-H) gel and poorly crystalline iron-siliceous hydrogarnet, respectively. Furthermore, it was possible to differentiate inner product and outer product C-S-H gels. Amorphous fly ash content was also analysed. Further details will be provided.

Secondly, we will elaborate on the use of PXCT for characterizing ye'elimite-based ecocements [2]. These cements are interesting as an alternative to Portland cements to decrease the CO_2 footprint of cement production. It was possible to discriminate between an aluminium hydroxide gel and calcium aluminium monosulfate, which have close electron density values. Specifically, the composition and mass density of two aluminium hydroxide amorphous gel agglomerates were determined: $(CaO)_{0.04}Al(OH)_3 \cdot 2.3H_2O$ with 1.48 gcm^{-3} and $(CaO)_{0.12}Al(OH)_3$ with 2.05 gcm^{-3}.

Finally, desired performances for post-EBS (10-30 keV) PXCT will be sketched.

Acknowledgements. This work has been supported through BIA2014-57658 and BIA2017-82391-R research grants, which are co-funded by FEDER.

Data accessibility. All reconstructed 'raw' tomograms (tiff format) were deposited, to be freely accessed, on Zenodo; dois: 10.5281/zenodo.2533863 & 10.5281/zenodo.3202557, for OPC and ye'elimite-based pastes, respectively.

References:

[1] - A. Cuesta, et al., "Quantitative disentanglement of nanocrystalline phases in cement pastes by synchrotron ptychographic X-ray tomography" *IUCrJ,* **2019**, 6, 473-491.

[2] - A. Cuesta, et al., "Chemistry and Mass Density of Aluminum Hydroxide Gel in Eco-Cements by Ptychographic X‑ray Computed Tomography" *J. Phys. Chem. C*, **2017**, 121, 3044−3054.