ALBA users / AUSE meeting 2024

**Title: "4D nanoimaging of Portland cement hydration"**

**Authors:** Shiva Shirani, Ana Cuesta, Ana Diaz, Pavel Trtik, Mirko Holler, Alexander Rack, Bratislav Lukic, Emmanuel Brun, Miguel A. G. Aranda*

g_aranda@uma.es

Departamento de Química Inorgánica, Cristalografía y Mineralogía, Universidad de Málaga, 29071 Málaga, Spain.

Laboratory for Macromolecules and Bioimaging, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland.

Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland.

ESRF-The European Synchrotron, 71 Rue des Martyrs, 38000 Grenoble, France.

Université Grenoble Alpes, Inserm UA7 STROBE, 38000 Grenoble, France.

**Abstract (2500 characters' maximum):**

Portland cements are environmentally contentious, accounting for ≈8% of the anthropogenic $CO_2$ emissions. The understanding of the cement hydration reactions (dissolution and precipitation processes) is important to contribute to develop cements with lower $CO_2$ footprints.

We are pushing 4D (3D+time) cement hydration nanoimaging within a multiscale framework. Full-field laboratory X-ray micro Computer Tomography (μCT) is widely used to study cement hydration but the best spatial resolution is about 2 μm for a Field of View (FoV) of ≈1×2 mm (H×V) with polychromatic measurements taking hours. Moreover, the contrast between the different components is poor. Full-field propagation-based phase-contrast synchrotron X-ray μCT can study similar FoVs ≈1×2 mm with better spatial resolution, ≈0.50 μm. The monochromatic measurements are fast, i.e. 5-10 minutes. Unfortunately, the contrast is only slightly better. Cement hydration can be studied with much better contrast and spatial resolution by scanning near-field ptychographic nano-computed tomography (nCT). In this case the FoV could be ≈200×30 μm with spatial resolution, close to 250 nm, and excellent component contrast. Even air and water can be differentiated. However, these nCTs takes about 3-4 hours in optimized beamlines (BL) at third generation synchrotrons.

We will present here our latest 4D nanoimaging results (Shirani et al. (2023) Nature Comm. 14:2652) from data taken at cSAXS beamline of SLS-PSI for a commercial Portland cement 52.5R with a water-to-cement mass ratio of 0.40. In the period 4-19h, the alite spatial dissolution rate, for particles smaller than 3 microns, has been measured as ~100 nm/h. In the same period, alite particles with average sizes larger than 10 microns dissolved with a spatial dissolution rate of ~33 nm/h, i.e. three times more slowly. In the period 19-47h, the alite spatial dissolution rate, for particles larger than 10 microns, is ~25 nm/h. Moreover, in the same 19-47 h time interval, the etch-pit growth rate, for particles larger than 10 microns, was ~40 nm/h. These values highlight the importance of particle size for determining the kinetics of cement hydration processes.

Moreover, in the period 4-19h, the C-S-H gel growth rate has been estimated as ~33 nm/h. C-S-H gel shell densifies with time and its average electron density evolves from 0.47 $e^{-}Å^{-3}$ at 19 h to 0.53 $e^{-}Å^{-3}$ at 47 h. Some other details will be discussed.