Portland cement (PC) production accounts for about 7% of the anthropogenic CO₂ emissions to our atmosphere. There are several proposals for sustainable, widely-available, economically-feasible low-carbon cements but they all have slow hydration kinetics at early ages, 3 days or less. Therefore, to study Portland cement hydration is important to rationally accelerate cement hydration which will help to decrease their CO₂ footprints.

Here, we have used near-field ptychographic X-ray computed tomography (PXCT) to acquire 4D (3D+time) data in a record-thick capillary of ∼160 µm, using a relatively high photon energy, 8.93 keV. This configuration, and the iterative algorithms that allowed the reconstructions, now meet simultaneously the four stringent requirements which are necessary for relevant cement hydration imaging: (i) water to cement mass ratio (w/c) close to 0.50, (ii) submicrometer spatial resolution, (iii) good contrast to be able to identify the different evolving components (more than eight), and (iv) relatively large scanned volume to allow hydration to progress with appropriate particle averaging; the particle sizes of commercial PCs have $D_{v,50} \approx 10-20$ µm. The overall current data collection time, between 3 to 4 hours, is modest but they are expected to decrease in the years to come.

The hydration of PC, in relevant conditions, has been measured with unprecedented spatial resolution and contrast, see Figure 1. This nanoimaging work shows known features like the alite (the main component of Portland cements) dissolution and Ca(OH)₂ (portlandite) growth. However, this study has revealed other nano-features of the dissolution-precipitation processes like the calcium silicate hydrate (C-S-H) gel shells surrounding every alite grain which does not preclude diffusion.

The measured alite spatial dissolution rates, $\sim 100$ nm/h for small grains in the acceleration period and $\sim 25$ nm/h for large particles in the deceleration stage impose constraints on the cement hydration models. Etch-pit growth rate, $\sim 40$ nm/h, and coalescence have also been measured but better spatial resolution is required for etch-pit quantification.

**Figure 1.** Enlarged views of the PXCT data at the studied hydration ages showing the hydration evolution of the PC-52.5-w/c$\sim 0.40$ paste which includes: i) dissolution of alite particles [$i_a$ denotes a particle which hydrates to yield inner C-S-H gel and $i_b$ highlights an alite particle which dissolves to leave water porosity at 47 h and then it dries at 93 h, i.e. air porosity], ii) calcium hydroxide (portlandite) growth, iii) C-S-H gel shell surrounding the alite particles at 19 h, iv) chemical shrinkage, mainly at 93 h due to water consumption, and v) etch-pit development with time.

More technical and scientific details will be presented at the meeting.

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