Study of hydration of sulphoaluminate cement by *in situ* synchrotron diffraction

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The hydration of calcium sulphoaluminate cement mixtures was studied in situ by synchrotron X-ray diffraction at the XRD1 beamline at the Laboratório Nacional de Luz Síncrotron (LNLS – Campinas). The specimens were analyzed in borosilicate glass capillary tubes of 0.7 mm and imbued with deionized water. As the hydration reaction is very fast, the data collection was started after two minutes of mixing with water. The X-ray wavelength chosen to get an adequate flux for these short time acquisitions was 1.033258 Å (12 keV), determined with a corundum standard. Diffraction patterns were collected every 35 seconds at temperatures ranging from 40 °C to 55 °C with accuracy better than 0.1 °C attained with a hot air blower. The diffracted signal was collected with an array of 24 Mythen detectors [1]. The diffraction patterns accumulated had appropriate statistics to determine the kinetics of the reaction either by quantitative Rietveld analysis or by fitting isolated diffraction peaks to Gaussian curves as a function of time. The most important phases involved in the hydration are Klein´s salt, also known as Ye'elimite, $Ca_4(AlO_2)_6SO_4$, and gypsum, $CaSO_4.2H_2O$ to yield Ettringite, $Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$, phase responsible for the mechanical properties. These studies show the potential of XRD1 beamline to investigate at controlled temperatures in situ fast reactions involving crystalline phases with time resolutions inferior to one minute, which is ideal for the hydration of cementitious mixtures.

The series of diffraction patterns collected for one and half hour are shown in Figure 1(a) and (b). In Figure 1(c) the evolution of the relative proportions of crystalline phases can be seen. Non-crystalline phases such as CSH gel or $Al(OH)_3$ (gibbsite) are not taken in consideration [2].

The figures clearly show that the ettringite formation happens in two stages, the second one starting after approximately forty minutes. It is also interesting to notice that this stages seem to coincide with the depletion of Ca_2SiO_4 (C2S) for the first stage and the depletion of Ca_3SiO_5 (C3S) for the second one.

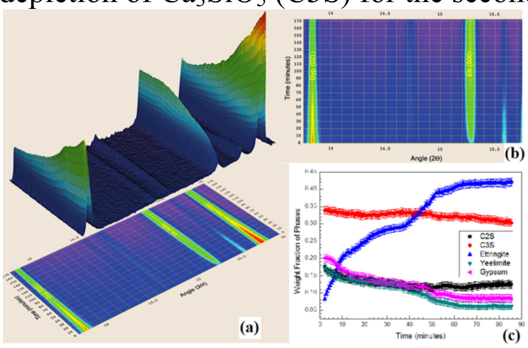

Figure 1: Sequence of diffraction patterns in a pseudo- 3D fashion as a function of time on a narrow angular domain to illustrate the evolution of the main crystalline phases involved in hydration: (a) 3D assembly of patterns; (b) projection as 2D contour map; (c) weight fractions of the crystalline phases plotted as determined by quantitative Rietveld analysis with GSAS. Some other non-crystalline products such as gibbsite and CSH gel are not taken in consideration.

[1] A. M. G. Carvalho et al.; J. Synchrotron Rad. 23 (2016) 1501.
[2] C. M. Rossetto, R. U. Ichikawa, L. G. Martinez, Carezzato, G. L., A. M. G. Carvalho and X. Turrillas. In situ hydration of sulfoaluminate cement mixtures monitored by synchrotron x-ray diffraction. Submitted to Mat, Sci. Forum (2017).