Experimental report

15/02/2020

Proposal: **6-07-54**

Council: 4/2019

Title: Water self-diffusion in the nanopores of C-(A)-S-H, the binding phaseof Portland cement and of low CO2 cement formulations

Research area: Materials

This proposal is a new proposal

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Samples:
3(CaO)SixAl1-xO2·nH2O
3(CaO)SiO2·nH2O

Instrument	Requested days	Allocated days	From	To
IN16B	6	2	03/02/2020	05/02/2020

Abstract:

At the global scale, cement is the single most manufactured material, and responsible of 5-7% of the global emissions of CO2 to the atmosphere. Among all the cement components, calcium silicate hydrates (C-S-H) is the main binding phase. New formulations of low-CO2 cements include Al substitutions, yielding C-A-S-H. In spite of the widespread use of cement materials, the poorly crystalline character of C-(A)-S-H has prevented a detailed characterization of its structural and dynamical characteristics, preventing as well the characterization of the structure and dynamics of the different water components. Only few studies hae addressed water dynamics in C-S-H nanopores but without an specific care of the sample preparation. Here, we propose QENS experiments to probe the water dynamics in C-S-H and C-A-S-H under different water humidities, using a controlled sample preparation protocol combined with water vapor isotherms, TGA and other characterization. The results will be key to characterize water dynamics in the different pores of these materials. Water dynamics influence carbonation and creep processes, which are responsible for the strength loss in cements.

ILL Experimental report

Title: **Water self-diffusion in the nanopores of C-(A)-S-H, the binding phase of Portland cement and of low CO_2 cement formulations**

Experiment number **6-07-54**

Experimental team

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Dates: **03/02/2020 to 04/02/2020**

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1. *Goal of the experiment*

New formulations of cement, made with the goal of decreasing its CO_2 footprint, include Al-containing admixtures which make that the structure and composition of the C-S-H binding phase is modified, including Al substitutions (C-A-S-H). In spite of the widespread production and use of cement for many years, the poorly crystalline character of C-(A)-S-H has prevented a detailed characterization of its structural and dynamical characteristics, preventing as well the characterization of the structure and dynamics of the different water components.
The goal of the proposal is to study the diffusional properties of water in the different pores of C-(A)-S-H following the same sample preparation strategy used for high-energy inelastic neutron scattering and for neutron diffraction experiments.

2. *Results*

Samples of C-S-H (main binder in Portland cement) and of C-A-S-H with two Ca/Si ratios (C/S = 1, 1.2) with adsorbed water (i.e., no capillar bulk-like water) were prepared by subjecting synthetic powders to a 55% relative humidity for 3 months.
Fixed window scans at different energy transfers ($\Delta E = 0$, 3 and 6 μeV) were performed as a function of temperature from 2 to 340 K at IN16B. The results for $\Delta E = 0$ and 3 μeV are shown in Figure 1. An decrease of the elastic intensity is observed from T ~ 75K, which is steeper for samples containing higher Ca/Si.

The main difference between the results of the elastic fixed window scans comes from the fact that samples containing higher Ca^{2+} seem to have a faster decrease of the elastic intensity as a function of T, meaning that some dynamic modes are activated at lower T with higher Ca^{2+}.
On the contrary the inelastic fixed window scans show a very similar behavior up to 250 K. From that point on, a plateau is observed, probably meaning that some dynamics outside of the energy range explored here are activated at these T. The intensity only decays for the sample with higher Ca/Si.

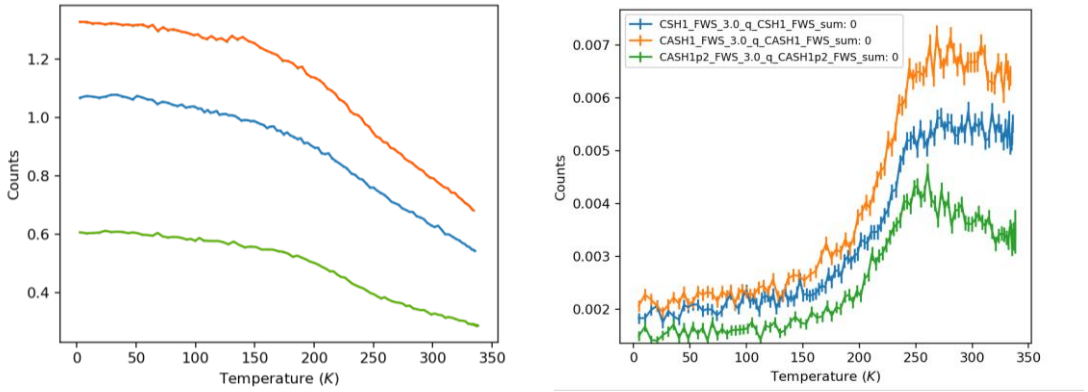

Figure 1. Elastic (left) and inelastic ($\Delta E = 0$, 3 µeV, right) fixed window scans for C-S-H (Ca/Si=1) and C-A-S-H (Ca/Si = 1 and 1.2).

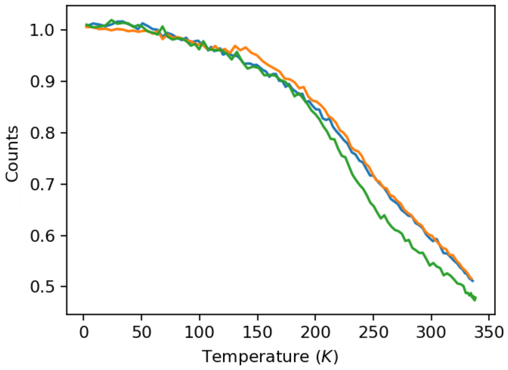

Figure 2. Normalized elastic FWS. The sample with higher Ca/Si shows a faster decrease of the elastic intensity.

3. *Perspectives*

Data analysis for the different Q values is ongoing, with the aim to determine if the dynamics are dispersive or not. In addition, these data will be combined with information from water adsorption isotherms to try to understand the different components of water whose dynamics could be activated at different temperatures.

A proposal will be submitted in a future call to complement these measurements with QENS data at different temperatures.

Two other samples of amorphous carbonates (considered as ‘natural cements’) have also been studied during this experiment with the aim of obtaining preliminary data.