Experimental report

01/12/2023

Proposal: **7-01-590** **Council:** 4/2023

Title: Nuclear quantum effects in cement hydration and setting

Research area: Materials

This proposal is a resubmission of 7-01-570

Main proposer: **Felix FERNANDEZ-ALONSO**

Experimental team: Cristina MACIA CASTELLO
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Local contacts: Monica JIMENEZ RUIZ
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Samples: Ordinary Portland Cements

Instrument	Requested days	Allocated days	From	To
IN1 LAG	4	4	20/11/2023	24/11/2023
D1B	2	0		

Abstract:

Cement and concrete are amongst the most widely used synthetic materials, yet they are also under increasing scrutiny in terms of their sustainability. Their large-scale production continues to be on the rise on a global scale, and it is currently responsible for about 10% of total anthropogenic carbon-dioxide emissions to the atmosphere. In the context above, our research efforts to date have looked at the relatively untapped possibility of achieving chemical speciation of the hydrogenous species present in these materials, using INS in combination with computational modelling. Encouraged by these results, we have also begun exploring hydration and setting phenomena, and have found strong nuclear quantum effects that merit further investigation using IN1-LAGRANGE and D1B. These experiments will give us important insights into the underlying microscopic mechanisms. And, on a more practical front, these effects can be used to tune hydration rates by adjusting the amount of light and heavy water in the primordial mix. To the best of our knowledge, this strategy represents a new means of controlling and tailoring the properties of this important class of materials.

EXPERIMENTAL REPORT

Proposal: 7-01-590 **Scientific council:** 2023-4

Title: Nuclear quantum effects in cement hydration and setting

Type of proposal: New proposal

Research area: Neutron scattering, cement-based materials

Main proposer: Felix FERNANDEZ-ALONSO (Materials Physics Center, San Sebastián, Spain)

Experimental team: Cristina MACIA-CASTELLO (Materials Physics Center, San Sebastián, Spain)

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Samples: Tricalcium silicate (C3S, Ca_3SiO_5), Ordinary Portland Cement (OPC)

Instrument: IN1 **Allocated days:** 4 **From:** 20/11/2023 **To:** 24/11/2023

Abstract:

Cement and concrete are amongst the most widely used synthetic materials, yet they are also under increasing scrutiny in terms of their sustainability. Their large-scale production continues to be on the rise on a global scale, and it is currently responsible for about 10% of total anthropogenic carbon-dioxide emissions to the atmosphere. In the context above, our research efforts to date have looked at the relatively untapped possibility of achieving chemical speciation of the hydrogenous species present in these materials, using INS in combination with computational modelling. Encouraged by these results, we have also begun exploring hydration and setting phenomena, and have found strong nuclear quantum effects that merit further investigation using IN1-LAGRANGE. These experiments will give us important insights into the underlying microscopic mechanisms. And, on a more practical front, these effects can be used to tune hydration rates by adjusting the amount of light and heavy water in the primordial mix. To the best of our knowledge, this strategy represents a new means of controlling and tailoring the properties of this important class of materials.

Experiment report

Inelastic neutron scattering experiments were performed on several cement samples using IN1 at ILL to study the hydration evolution and isotopic effects in cements. The measured specimens were anhydrous and hydrated C3S and Ordinary Portland Cement (OPC) with a water to cement ratio of w/c = 0.4. In the case of the hydrated samples, we studied 3 different H2O/D2O concentrations corresponding to pure H2O, pure D2O and an equimolar mixture. In all cases, we looked at two situations of relevance in concrete research: at long times, when the kinetics of hydration have reached a quasi-stationary steady state (2 week-aged samples); and the situation whereby the reaction of these samples with water have formed a percolated solid 3D skeleton (setting point).

Neutron vibrational spectra were recorded at T = 5 K on IN-Lagrange spectrometer along the full energy range (from 8 to 4000 cm-1) to obtain as much information as possible from the sample. Figure 1 shows the spectrum of anhydrous C3S. From the spectrum, we can see clear bands are formed in the phonon region, as a consequence of the bonds between Calcium and Oxygen. In figure 2 two different scenarios are shown; one for the hydrated aged sample, and other for the one at the setting point. Both cases show similar spectra, but in the low energy region some discrepancies appear as the lattices are very different, which is an indicator of the different hydration stages and kinetics the two of them reflect.

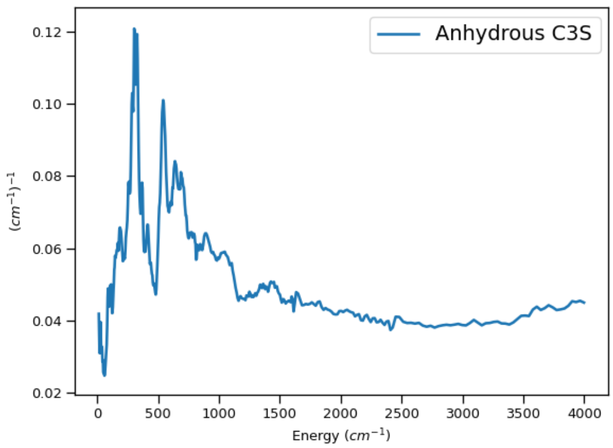

Figure 1: Anhydrous C3S spectrum recorded at 5 K over the full energy range

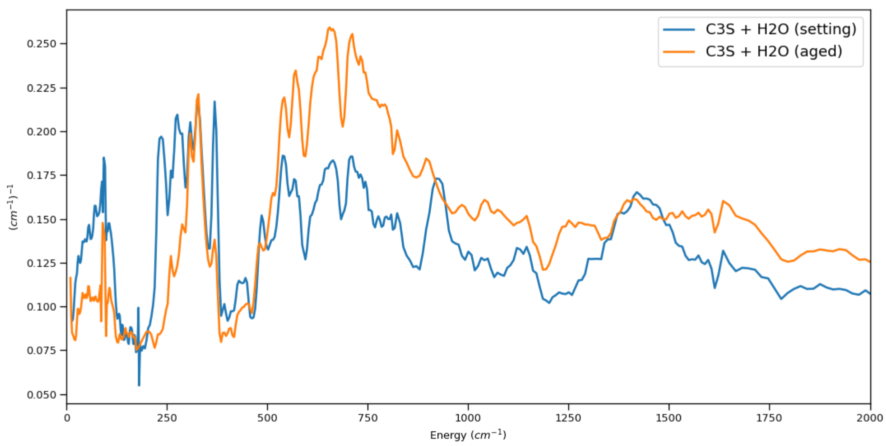

Figure 2: Hydrated C3S + H2O spectra. The two scenarios mentioned before are shown: Aged sample (orange) and setting point (blue).