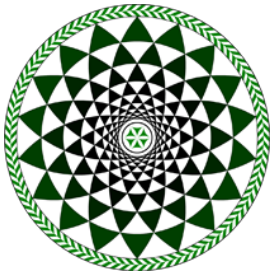

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Multi-scale investigation of MgO-based cements by NMR spectroscopy and relaxometry

Tuesday 4 July 2017 11:30 (30 minutes)

In the last years, MgO-based cementitious materials have attracted great interest as eco-sustainable alternatives to traditional Portland (CaO-based) cement, the dominant form of cement used worldwide. In particular, MgO-based formulations have been proposed with the aim of both reducing the environmental footprint associated with the production of Portland cement and developing cements to be used in the immobilization of nuclear or metal containing waste [1].

The binder phase of these cements is magnesium silicate hydrate (M-S-H), the amorphous phase that forms from the reaction of MgO with a source of silica (SiO_2) and water. Although a significant quantity of literature exists concerning the structure and nature of the M-S-H gel [2-8], a full comprehension of properties, such as hydration kinetics, the nature of the hydrated products and their multi-scale structure and organization, is still lacking. The investigation of these properties, as well as the research for new formulations with improved performances, is fundamental to achieve the industrial breakout of these materials.

In this work, novel MgO-based cements obtained by hydration of a 1:1 molar mixture of MgO and fumed silica (MgO/SiO_2) and mixed-formulations containing different amounts of MgO/SiO_2 and Portland cement were developed. The multi-scale structural properties and the hydration kinetics of these systems were investigated in detail by means of solid-state NMR spectroscopy (SSNMR) and relaxometry, which already proved to be very powerful for the study of traditional cement [9-12], with the support of complementary techniques such as X-Ray Diffraction, thermogravimetry, IR spectroscopy, Scanning Electron Microscopy, and Differential Scanning Calorimetry.

The nature and the structure at the sub-nanometric scale of the formed hydrated phases, as well as the kinetics of their formation, were investigated at different hydration times on freeze-dried samples by means of multi-nuclear high-resolution SSNMR experiments [13,14]. In particular, ^{29}Si MAS experiments allowed the different silicon sites, Q^1, Q^2 and Q^3, characterized by different connectivity to -OSi, -OH, -OMg or -OCa groups, to be identified and quantified. In the case of mixed systems, it was also possible to distinguish between M-S-H and C-S-H (calcium silicate hydrate arising from hydration of alite and belite present in Portland cement) domains, and to study their properties and relative amounts as a function of composition and hydration time. The state of water and the evolution of the solid matrix during the hydration process were investigated as a function of time directly on the cement pastes by means of ^1H T_1 Fast Field-Cycling relaxometry and measurement of ^1H T_2 relaxation times at low magnetic field (proton Larmor frequency of 20 MHz) [15].

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