X-ray Total Scattering techniques and the nanoscale structure of hydraulic binders

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Although two thousand years have passed since the first production of hydraulic binders (the cement of the Romans), many questions and ambiguities remain on the understanding of the atomic scale structure that controls the physical properties of cement at the macro-scale [1]. The physical properties of ancient and modern cement largely depend on formation of insoluble calcium silicate hydrates (indicated as C-S-H) and in particular on their compositional and structural characteristics. C-S-H typically exhibits a broad X-ray diffraction signal reflecting its atomic ordering confined only at the nanoscale [2]. The structural and microstructural characterization of nanosized material is a challenging task, and this explains why C-S-H in cements has continued to escape detailed and direct structural analysis. The atomic-scale structural description of relevant binding phases is further complicated when the cement content is progressively replaced by different types of supplementary cementitious materials (SCM), such as metallurgical slags, calcined clays and other industrial by-products (blended cement), driven by the upcoming challenge of realising high-performance hydraulic binders within a circular economy and meeting the goals of zero net carbon emissions [3]. In slag-based cement, or other blended cement, the reaction products consist mainly of calcium aluminium silicate hydrates (C-A-S-H gel) with variable stoichiometry and a nanoscale structural order. Achieving the detailed structural description of these poorly crystalline phases requires the application of advanced analytical techniques, specifically devised for the characterization of nanostructured and disordered materials [4]. In this project, an innovative approach based on Small Angle X-ray Scattering and Wide Angle X-ray Total Scattering (WAXTS) measurements coupled to modelling techniques based on the Debye Scattering Equation (WAXTS-DSE method) will be developed to investigate the stoichiometry, structure, size, shape and defects in poorly crystalline binding phases to be correlated to the functional properties of alternative and sustainable construction materials.

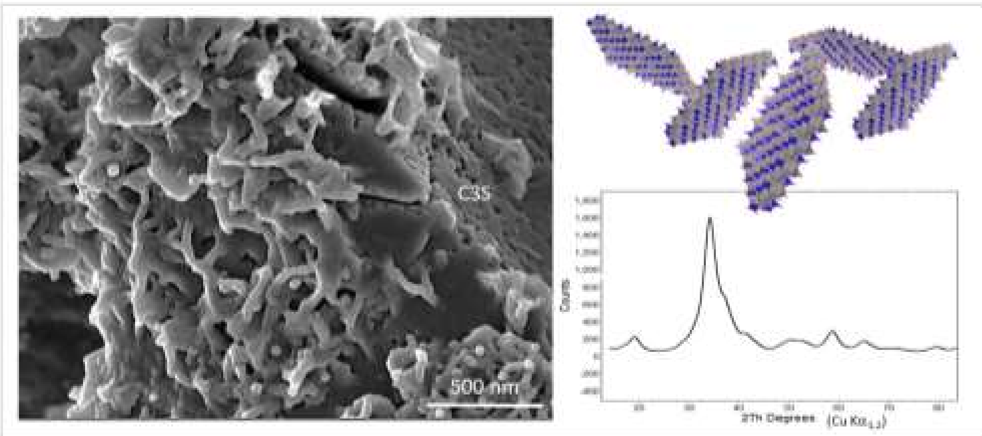

Figure 1. Left: Secondary Electron SEM image of a cement particle covered with calcium silicate hydrate C-S-H. Top right: atomistic modeling of C-S-H nanoparticles. Bottom right: measured X-ray diffraction pattern of C-S-H.

The proposed research project aims to accomplish the following goals:

- developing experimental and modelling tools based on small-angle and WAXT-DSE methods aiming at describing nanometer-scale features of C-S-H and C-A-S-H;
- achieving detailed knowledge on chemical factors governing the growth and atomic ordering of C-S-H nanoparticles;

- achieving a detailed comprehension of the chemical parameters (Ca/Si ratio and Na content) that control slag reactivity in cement or alkali-activated systems;
- achieving detailed knowledge of the factors controlling the rheology of fresh paste in alkali activated systems based on in-situ X-ray diffraction measurements and small amplitude oscillatory shear (SAOS) testing.

The research project is part of the current research activity on hydraulic binders at the Circe Center-Geoscience Department, University of Padua. Collaborations with the OPIGEO s.r.l, IGG-CNR of Padua, the Total Scattering Laboratory (To.Sca.Lab) at University of Insubria and Technische Universität Wien, - TU Wien are foreseen.
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References

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