



Preferred orientation of calcium aluminosilicate hydrate induced by confined compression



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ABSTRACT

The existing macroscale models of the calcium (alumino)silicate hydrate (C-(A-)S-H), the main binder of concrete, assume that the nanocrystallites maintain random orientation under any loading conditions. However, using synchrotron-radiation-based XRD, we report the development of preferred orientation of nanocrystalline C-A-S-H, from random at ambient pressure to strongly oriented under uniaxial compression with lateral confinement. The *c*-axes of the nanocrystals tend to align with the primary load. This preferred orientation is preserved after removing of external loading. The texture, quantified using a standard Gaussian fiber orientation distribution function (ODF), was used to calculate the averaged bulk elastic tensor of oriented C-(A-)S-H. It changes from isotropic (without texture) to transversely isotropic (with texture). Our results provide direct evidence of the reorientation of nanocrystalline C-(A-)S-H as a mesoscale mechanism to the irreversible deformation of cement-based material. The implications of these results for modeling the mechanical property of C-(A-)S-H at the macroscale are discussed.

1. Introduction

Quantifying the nano- and micro-scale structure of cement-based materials is a key to understanding, designing and, most importantly, controlling their macroscale properties [1]. This endeavor has met great challenges when applied to calcium silicate hydrate (C-S-H, C = CaO, S = SiO₂, H = H₂O in cement chemistry notation), which is the major binding phase in Portland cement (PC) concrete. This is largely due to its hierarchically porous structure, poorly crystalline nature and highly variable chemical composition [2]. For instance, the calcium-to-silica ratio (Ca/Si) of C-S-H ranges from ~0.6 to ~2.0 depending on the initial raw material composition [3,4]. C-S-H also uptakes significant amounts of Al to form C-A-S-H (A = Al₂O₃), as found in PC [5,6] and ancient Roman concrete [7–9] blended with aluminous pozzolanic materials.

Nonetheless, researchers have proposed structural models of C-(A-)S-H that span length scales from nanometers to microns. For

synthesized type I C-(A-)S-H, over the Ca/Si range from 0.6 to 2.0, C-(A-)S-H remains a poorly crystalline layer structure highly analogous to tobermorite minerals, which is composed of a CaO₇ sheet being “sandwiched” by silicate tetrahedra chains [10,11]. This silicate chain, also termed dreierketten chain, is composed of periodically repeated pair silicate tetrahedra, connected by bridging silicate tetrahedra (Fig. 1). As the Ca/Si ratio increases, the bridging silicate tetrahedra are increasingly omitted along with the enrichment of Ca in the interlayer (Fig. 1) [12–17]. C-(A-)S-H usually exhibits a poorly-ordered layer stacking along *c*-axis [18,19]. Such stacking seems to be more disordered when Al is uptaken at low curing temperatures, whereas it is much more ordered at higher curing temperatures due to the Al-induced crosslinking, although the crystallite size is still limited to a few nanometers (Fig. 1) [18,20]. The C-(A-)S-H formed from PC systems has similar layer structure as type I C-(A-)S-H but the layer stacking is more disordered, such that it displays only one broad diffraction peak [21] at the location where type I C-(A-)S-H yields the strongest diffraction, i.e.

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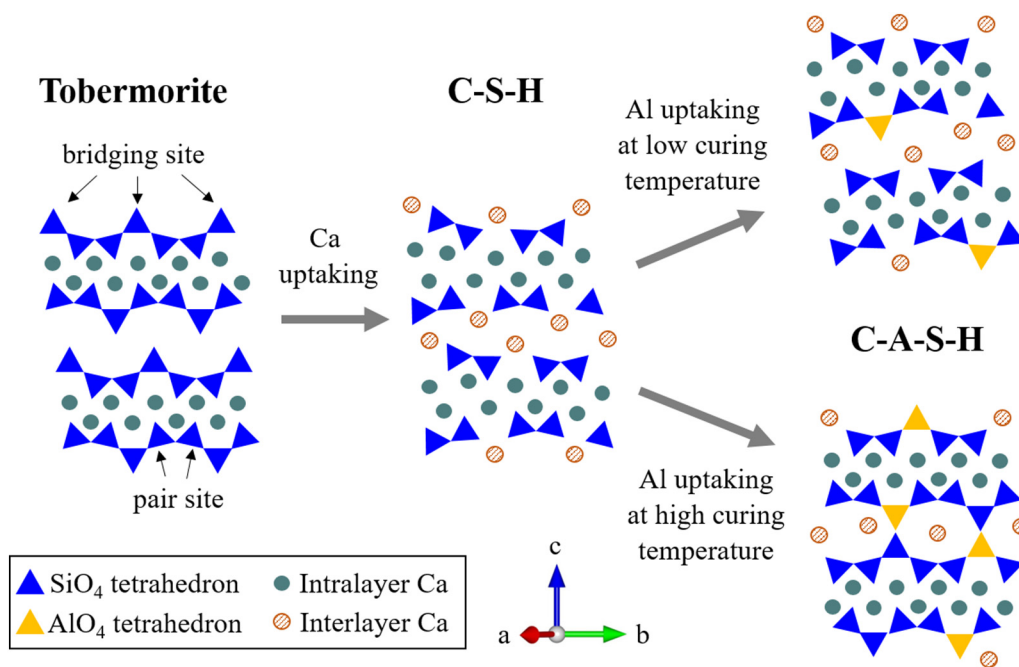


Fig. 1. Schematic layer-structural model of C-(A-)S-H and its evolution after uptaking Ca and Al. The structural disorder in C-S-H and low-temperature C-A-S-H is illustrated. The Al-induced crosslinking in high-temperature C-A-S-H improves the structural order. The interlayer water content is a function of the drying conditions and is not shown for viewing convenience.

of Laue indices (020) and (022) as refined by the 11 Å tobermorite structure (monoclinic, $a = 6.735 \text{ \AA}$, $b = 7.385 \text{ \AA}$, $c = 22.487 \text{ \AA}$, $\gamma = 123.25^\circ$) [10,11,20,21]. Note that several tobermorite-based structural models can be used to refine the C-(A-)S-H lattice parameters and yield almost identical results [20]. Throughout the paper, C-(A-)S-H will be used to denote the tobermorite-like nanocrystalline C-S-H, either with or without Al-incorporation. C-A-S-H is specifically used to denote the Al-incorporated C-S-H, for example the sample being studied in this work.

In general, all the hotly debated nanoscale models of C-(A-)S-H in PC concrete [2,22–24] agree on the poorly-crystalline nature of C-(A-)S-H, whose smallest coherent domain size is estimated to be smaller than 5 nm. This coherent domain size is consistent with pair distribution function studies [25] and nanoindentation results [26]. Historically, several names were used to denote such coherent domain in C-(A-)S-H, e.g. nano-globule [2], nano-grain [25], nano-platelet [27] and nanocrystal [13,14]. In the present work, the term ‘nanocrystal’ will be used throughout to denote the coherent region that exhibits a layered poorly-crystalline structure similar to tobermorite and often with the smallest dimension along the direction perpendicular to the layer. Atomistic modeling has been intensively applied in the past decade to correlate the chemical composition with the mechanical properties of C-(A-)S-H [28–33]. It is only recently that such correlations were directly verified by experimental data at molecular scale [18,20]. The bulk modulus of C-(A-)S-H is greatly controlled by the compliance along its c -axis. The interlayer densification and the Al-induced crosslinking are the dominant driving force for C-(A-)S-H stiffening, whereas the bridging silicate omission is irrelevant to the overall stiffness [18,20].

The orientation distribution of grains in polycrystalline materials, often referred to as “texture”, markedly determines the anisotropy of properties at the macroscale [34]. To our knowledge, the existing cement models, either based on hydration chemistry [35–37], or colloidal packing behavior [38], assume a random orientation of the C-(A-)S-H nanocrystals, which leads to isotropy of the matrix at the macroscale. It is therefore of interest to quantify the orientation of the nanocrystalline C-(A-)S-H and to determine its influence on the anisotropy of C-(A-)S-H properties at the macroscale.

One of the most efficient ways to quantify grain orientation distributions is by studying the variation of intensity along the azimuthal angle of Debye rings obtained from synchrotron X-ray diffraction

experiments. Once the texture is measured, homogenization models can be used to estimate the anisotropic properties at the macroscale [39–42]. The primary goal of the present work is to investigate whether nanocrystalline C-(A-)S-H develops preferred orientation under deviatoric stresses, to measure its evolution with increasing stresses, and to model how the texture influences the macroscopic elastic properties of C-(A-)S-H aggregate. Our previous work used synchrotron X-ray diffraction at high pressure (HP-XRD) to measure the mechanical properties of the C-(A-)S-H nanocrystals in a diamond anvil cell (DAC) under hydrostatic pressure [18,20]. However, the development of texture in the polycrystalline material often requires the presence of deviatoric stress [43]. In the present work a hydrostatic pressure medium was not used in the DAC. The study of texture in diffraction experiments requires a sufficient crystallinity, therefore a C-A-S-H sample synthesized at 80 °C was selected because it was found to have the most ordered crystal structure among reported C-(A-)S-H samples. Using the texture and single crystal elastic constants as input, anisotropic elastic properties of C-(A-)S-H at the macroscale were calculated. Note that the term “texture” in cement research community often refers to “micro-structure” and is irrelevant to crystal preferred orientation [38–44]. In this work, “texture” specifically refers to the orientation distribution of nanocrystalline C-(A-)S-H.

2. Materials and methods

2.1. Materials

Details of C-(A-)S-H synthesis are reported elsewhere [20,45]. Stoichiometric amounts of SiO₂ (Aerosil 200, Evonik), CaO (obtained by burning CaCO₃ (Merck Millipore) at 1000 °C for 12 h) and CaAl₂O₄ were mixed with water (water/solid ratio of 45), such that the initial Ca/Si and Al/Si molar ratios were 1.0 and 0.1, respectively. The suspensions were then stored in a Teflon bottle at 80 °C for 8 weeks. The reaction product was vacuum filtered and freeze-dried for 7 days. All the above processes were carried out in a N₂-filled glove box to avoid carbonation. Following the notation in a previous publication [20], the sample is named hereafter Al10.

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