



Calcium silicate hydrate from dry to saturated state: Structure, dynamics and mechanical properties

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Abstract

Calcium silicate hydrate (C-S-H) is the most important hydration product of cement-based materials. In the nanostructure of C-S-H, structural water molecules are distributed in the interlayer region and determine the mechanical performance of C-S-H gel. In this study, C-S-H gels with different water contents expressed as the water/Ca ratio are characterized in the light of molecular dynamics. In order to study the influence of the water molecules, the structures of 12 C-S-H gel samples with water/Ca ratios from 0 to 0.95 are investigated. It is found that the penetration of water molecules transforms the C-S-H gel from an amorphous to a layered structure by silicate depolymerization as the water content gradually increases. The structures are then tested for mechanical properties by simulated uniaxial tension and compression. The mechanical tests associated with structural analysis reveal that the structural water molecules can greatly weaken the stiffness and the cohesive force by replacing the ionic-covalent bond with unstable H-bond connections. By studying the tensile failure mechanism of C-S-H gels at different humidity levels, the disconnecting role of the structural water molecules is comprehensively interpreted. Because the interlayer water molecules prevent reconstruction of the bonds between the Ca_w and the silicate chains, the plasticity of the C-S-H gels is reduced significantly in the change from a dry state to a saturated state. In addition, the compressive strength of a C-S-H gel in the saturated state is much larger than the tensile strength. This provides molecular evidence for the tensile weakness of cement paste.

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1. Introduction

When Portland cement reacts with water, cement hydrates are produced. Calcium silicate hydrate (C-S-H) gel is the dominant cement hydrate, and hence it determines the binding force of the cement paste. For the whole service life of a cement paste/concrete, the interaction between water and C-S-H gel determines the mechanical properties and durability of cement-based materials. On the one hand, the initial water content of the cement paste, or water-to-cement ratio, influences the microstructure of

the cement hydrates [1]; on the other hand, the water molecules, as essential elements of C-S-H gel, are sensitive to the surrounding environment and humidity [2]. For example, when immersed in water, C-S-H gel is very likely to reach a saturated state, while the structural water of the C-S-H may be missing in the case of a fire.

In order to understand the role of water in C-S-H gel, it is necessary to investigate the intrinsic structure and mechanical performance of the gel at different humidity levels. The molecular structure of C-S-H has been studied for more than half a century, particularly using a variety of experimental techniques, including nuclear magnetic resonance (NMR) imaging [3], X-ray diffraction [4] and small-angle neutron scattering (SANS) [5], and C-S-H is now widely believed to be the analogue of the layered

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minerals tobermorite [6,7] and jennite [8]. Based on experimental data, Pellenq et al. [9] employed the molecular dynamics method to construct a C-S-H model that can describe well the structural, dynamical and mechanical properties of cement at the nanoscale. Because the model is based on previous experimental results, it has been widely accepted in the nanoscale simulation field and is defined as a “realistic model”. The “CSHFF force field” [10] was developed to describe the interaction between atoms in cement systems, and this force field demonstrated good transferability in simulating cement-based materials.

Water molecules, essential constituents in C-S-H gel, have been investigated since the birth of the “realistic model”. Pellenq et al. [9] simulated the stress–strain relation of C-S-H gel in resisting shear force. By comparing the mechanical performance of dry and wet C-S-H gels, it was concluded that, during the loading process, large displacements of water molecules in wet C-S-H gels weaken the shear strength and result in unrecovered deformations. Youssef et al. [11] investigated the structural and dynamic features of water molecules in C-S-H gels. Due to the highly hydrophilic nature of calcium silicate sheets, H-bonds constructed between the non-bridging oxygen and water are very strong. Therefore, water molecules constrained in the gel demonstrate a glassy nature: the tetrahedral spatial structure is distorted and the diffusion rate is significantly reduced. Ji et al. [12] utilized five classic water models, SPC, TIP3P, TIP4P, TIP4P05 and TIP5P, to simulate calcium silicate hydrate. They concluded that SPC and TIP5P accurately describe the intrinsic properties of C-S-H gel, with SPC (flexible) being more computationally efficient. Recently, Bonnaud et al. [13] interpreted the cohesive force of C-S-H gel by analyzing the fluid pressure of the water molecules and the counter-ions in the interlayer region. Under different humidity conditions, they found that the cohesive force mainly results from negative pressure caused by the interaction between interlayer calcium atoms and the calcium silicate sheets. Even though this work took the amount of water in the C-S-H gel into consideration, the molecular structure of the calcium silicate sheet was assumed to remain unchanged under different humidity conditions. This assumption, ignoring the structural evolution due to the water effect, needs to be further investigated.

Previous research on C-S-H gels can be categorized into two classes: molecular structure analysis and thermodynamics investigation. Few efforts have been made to investigate the evolution of mechanical properties with water content variation. In particular, the tensile strength of C-S-H gels at the nanoscale, considered as the most essential property of construction material, has not been studied so far. The aim of this paper is to bridge the relationship between the morphology and the mechanical performance of C-S-H gels in different humidity conditions. On the one hand, the structural feature evolution can be characterized by analyzing the silicate chain morphology and chemical bonds in different water contents. On the other

hand, the mechanical properties, including the stiffness and cohesive force, can be achieved directly from uniaxial tension/compression simulation. Combining the structural and mechanical evolution, the water molecules’ role in loading resistance can be determined and assessed.

2. Simulation method

The CSHFF force field [10], developed for cement-based materials, is utilized to simulate C-S-H gels at different humidity states. The force field approach has been widely used in C-S-H simulations and has been proven to be able to describe the structure, energy and mechanical properties of various calcium silicate phases satisfactorily [9,11–14]. The force field parameters of Ca, Si, O and H can be obtained from the literature [15].

2.1. Dry and saturated model

The C-S-H model in the present study is constructed based on the procedures proposed by Pellenq et al. [9]. Firstly, the layered analogue mineral of C-S-H, tobermorite 11 Å without water, was taken as the initial configuration for the C-S-H model [16,17]. Silicate chains were then broken to match the Q species distribution obtained from NMR testing [18]. The dry disordered structure is plotted in Fig. 1. Secondly, Grand Canonical Monte Carlo simulation of the water adsorption is operated on the dry disordered C-S-H structure at 300 K. The adsorption is conducted from 0 to 100 million steps to obtain 12

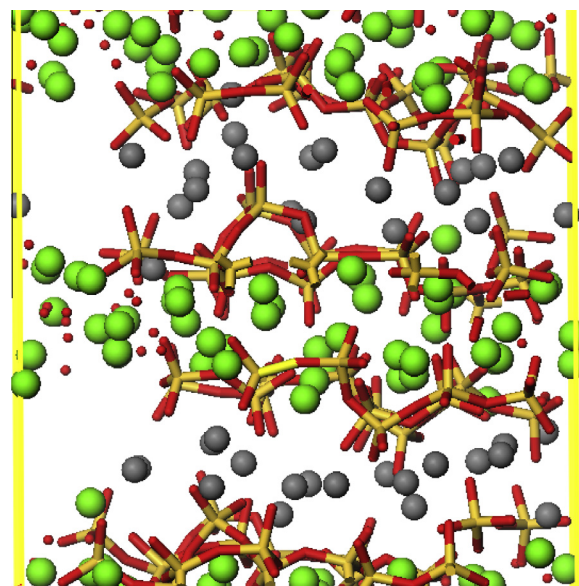


Fig. 1. Initial dry C-S-H structure for water adsorption simulation. Simulation box size: $a = 21.3$, $b = 21.2$, $c = 21.9$; $\alpha = 90^\circ$, $\beta = 90^\circ$, $\gamma = 90^\circ$. Yellow and red bonds represents the silicate chain (Si–O_s); the gray and green balls correspond to the interlayer calcium atoms (Ca_w) and the layered calcium atoms (Ca_s), respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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