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Molecular dynamics study on axial mechanical properties of calcium silicate hydrate

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Abstract

The axial mechanical properties of calcium silicate hydrate during uniaxial tension or stress have been investigated by molecular dynamic simulations. Based on the classic calcium-silicate-hydrate model, the influence of calcium to silicate ratio and temperature are studied in detail. It demonstrated that calcium silicate hydrate with different Ca/ Si ratio have similar stress-strain curves under uniaxial compression, but greatly difference on the uniaxial tension behaviors. The uniaxial tensile properties was affected by temperature that the strain rate in the low-temperature is higher than that in high temperature. The work in this paper reveals the mechanical properties of calcium silicate hydrate at the nanoscale and establishes the foundation explore the failure mechanism of cement or cement-based materials.

1. Introduction

Cement, as an important building material, is the most commonly used materials and has a significant role in national construction. Calcium-silicate-hydrate (C–S–H) is the main cement hydration product, consisting of ordered-disordered calcium silicate skeletons layers. As the most important binding phase in cement-based material, C–S–H determines the strength and modulus of cement concrete materials [1]. Meanwhile, the crystalline structure of C–S–H plays a significant role in the mechanical properties of concrete and cement materials. Recently, high purity C–S–H have been successfully fabricated via hydrothermal method, but the research of C–S–H mainly concentrates on the macro-micro level. It is still a challenge to study the structural properties of C–S–H at the micro-level, mainly due to the low degree of crystallization and chemical composition instability. Meantime probing into the complex system of cement at molecular-level is difficult to resolve with current experimental technical. The molecular dynamics (MD) simulations, which play important role to analyze the composition and structure performance of C–S–H, provides a useful approach for understanding and investigating the chemical and physical behavior of cement or cement-concrete materials. As a remarkable affecting factor to cement materials, the research on mechanical property of C–S–H has been the extensive investigation topic, which is not only important for the theoretical researches on concrete but also the applications of cement materials in construction engineering.

The main product of the hydration of Portland cement is calcium silicate hydrate (or C–S–H). It is primarily responsible for the strength in cement-based materials. C–S–H is a nano sized material, determining the mechanical properties of the hardened cement paste as well as macroscopic properties of the cement-based materials such as hardening, shrinkage and cohesion [2]. The underlying atomic structure of C–S–H is similar to the naturally occurring mineral Tobermorite. It has a layered geometry with calcium silicate sheet structure separated by an interlayer space. The silicate anion layers have a net negative charge and are held together by Ca$^{2+}$ cations in the interlayer region. C–S–H is the dominant hydration product of cement-based materials. Its compositions and structures have great impact on the micro-structural property of C–S–H and the failure mechanism of cement-based materials. Researches showed that the mechanical strength of C–S–H specimens is

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not only determined by calcium to silicon (Ca/Si) ratios, but also the siliceous material as well as the specific surface area [3]. Through the study on the micro structure and size distribution of the cement hydration products, Taylor found that C–S–H appeared by bimodal distribution for the difference of Ca/Si ratio [4]. Due to the discrepancy of hydration degree result by the environmental temperature, the variation of Ca/Si ratio is caused in the crystal of C–S–H as a result generally. By means of the transmission electron microscopes (TEM) tests, Zhang et al concluded that the Ca/Si ratio of C–S–H is between 0.6 and 2.3 [3]. Through the cement paste tested, Vandamme et al [6, 7] found the elastic modulus of C–S–H gel was significantly affected by their density. And the density could divide into high-density and low-density state, among that the average of high-density C–S–H was 1.7 [2]. By the Nuclear Magnetic Resonance (NMR) Spectroscopy tests [8], it is found that the degree of polymerization of C–S–H was determined by Ca/Si ratio, which also had influence on the structure of C–S–H [9]. Cong stated that the degree of polymerization of silicate chains in C–S–H structure showed an increased tendency with temperature [10]. Bauchy et al computed the fracture toughness and obtained the tensile stress-strain curve of the fracture at the atomic scale [11]. C–S–H is the amorphous material, but its internal basic composition keeps atomic short-range regular arrangement [12]. According to the researches of the transmission electron microscopes (TEM) tests and x-ray diffraction (XRD) tests, there are two mineral crystal analogue of C–S–H widely accepted by the researchers, tobermorite and jennite.

Faucon et al [13] employed the molecular dynamics simulation using tobermorite crystal structure model with Ca/Si ratio of 0.66 and 0.83 to investigate the instability of C–S–H structure as well as the fracture mechanism of silicon chain. Hou et al [14–16] employed the molecular dynamics simulation using the tobermorite 11 Å model to investigate the axial mechanical properties of C–S–H through the axial tension simulation. Zaoui et al [17] adopted the Buckingham potential [18] and the core–shell potential that usually applied in modeling the zeolite material to compute the elasticity modulus of tobermorite 11 Å structure under different pressure with several Ca/Si ratio. It demonstrated that the elastic property of structure has significant variation under the 20 GPa of hydrostatic pressure, but the elastic modules of tobermorite with different Ca/Si ratio tended to develop to the same value as the hydrostatic pressure increases. Palkovic et al investigated the constitutive response of C–S–H under combined loading [19]. Bauchy et al [20] utilized the stiffness theory to study the interactions between C–S–H gels, and found that the hardness and stiffness of the C–S–H decrease with the increasing Ca/Si ratio. Meanwhile the C–S–H gel showed certain flexibility when the Ca/Si ratio is 1.7. Pelisser et al [21] utilized the nanoindentation technique measured the hardness and young’s modulus of C–S–H increases with the decreasing Ca/Si molar ratio. Meanwhile another study adopted both the simulations and experiments methods indicated a significant decrease of the average indentation elastic modulus with increasing Ca/Si ratio [22]. Eftekhar et al investigated the compressive tensile and shear property of the CNTs reinforced C–S–H cement materials using MD simulations [23]. Qomi et al utilized the Reax–FF force field [24] to describe the chemical reaction of water molecules, and found the physicochemical characteristics of water molecule was altered by Ca/Si ratio of the C–S–H gel [25]. Alizadeh reported the dynamic mechanical properties of the C–S–H compacted samples was determined by the Ca/Si ratio [26]. Recent literatures [27–30] investigated the mechanical properties of C–S–H gel using the COMPASS force field [31] in MD simulations, which is found to be suitable for describing the interaction of hydrolysis product of cement.

C–S–H has important contribution to the cement concrete strength and plays an significant role in concrete material mechanical property. Meanwhile the mechanical property of C–S–H gel is difficult to research at the atomic scale due to the amorphous state and the complicated structures. As for the C–S–H crystal structure, two crystalline mineral model (tobermorite [32] and jennite [33]) are widely employed in MD simulation. However the mechanical performance of C–S–H with different calcium to silicon (Ca/Si) ratios has not been investigated thoroughly so far. It is necessary to reveal the mechanics performance of C–S–H on the nanoscale and probe the failure mechanisms of cement-based materials. In this work, we investigated the axial mechanical properties of C–S–H via the molecular dynamics simulations using the traditional original crystal structure of tobermorite 11 Å [34] model without deleting a segment of silicon atom and water molecule. The uniaxial tensile and compressive mechanism properties of C–S–H with different calcium to silicon (Ca/Si) ratio are systematically studied with the COMPASS force field. The effects of temperature on the mechanical performance of C–S–H during axial loading are also analyzed.

2. Physical model and molecular dynamics simulation

2.1. The physical model of C–S–H

With the rapid development of computer science and technology, the materials micro-scale studies of cement material investigated upon the theoretical analysis and numerical simulation is growing, among which the molecular dynamics simulation is one of the most effective numerical method for nanoscale mechanical properties investigation of calcium-silicate-hydrate. The molecular dynamics models of tobermorite are divided
into three types due to the difference of interlayer space. The tobermorite 11 Å model possesses an interlayer structure (liked sandwich). As the skeleton of tobermorite 11 Å is constructed by silica tetrahedrons while the calcium ions and water molecules are distributed in the interlayer region. It is proposed that the calcium silicate layers of tobermorite 11 Å were linked by the chemical bond. Besides, Hamid et al. demonstrated a modified model of tobermorite structure, of which the layer structure was considered to be individual from each other. The modified model proposed by Hamid et al. allows the tobermorite structure possessing different Ca/Si ratio.

The monoclinic crystal tobermorite 11 Å is chosen as the research model to study the axial mechanical properties of C–S–H. Monoclinic structure is the primitive crystal structure of C–S–H, and often used in mechanical performance simulation analysis. The orthorhombic structure can be obtained by coordinate transformation from monoclinic structure, and is often used as C–S–H model in the ionic adsorption and diffusion simulation. For studying the influence of Ca/Si ratio on the mechanical properties of C–S–H, the Hamid tobermorite 11 Å model is adopted in this article. Table 1 displays the lattice parameters of Tobermorite 11 Å models employed in this work, and the kinds and amounts of atoms in each simulated system are shown in figure 1. According to the crystal parameters reported by Merlino and Hamid et al., the C–S–H structures model with four different types of Ca/Si ratio (0.67, 0.83 and 1, respectively) are constructed. These models have the same amount of silicon atoms and water molecules with similar single silicon chain skeleton. The simulation parameters of tobermorite 11 Å models are shown in table 2.

### 2.2. Molecular dynamics simulation

The molecular dynamics simulation models of tobermorite 11 Å atomic structures with different kinds of Ca/Si ratio are built by utilizing the Materials Studio software package (https://www.3ds.com/products-services/biovia/products/molecular-modeling-simulation/biovia-materials-studio/). Before the simulations, the unit

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**Table 1.** The lattice parameter of two types of tobermorite structures.

| Structure       | Tobermorite 11 Å Monoclinic crystal | Hamid tobermorite 11 Å Monoclinic crystal |
|-----------------|-------------------------------------|------------------------------------------|
| Lattice parameter | Crystal system | Space group | B11 m | P21 |
| a/Å             | 6.732 | 6.69 |
| b/Å             | 7.369 | 7.39 |
| c/Å             | 22.68 | 22.77 |
| α/°             | 90 | 90 |
| β/°             | 90 | 90 |
| γ/°             | 123.18 | 123.49 |

**Figure 1.** Number of atoms in C–S–H model with different types of Ca/Si ratio.
cell of C–S–H is experienced the geometry optimization to keep initial structural stability. Then a (4 × 3 × 1) supercell of tobermorite 11 Å is made by stacking the unit cells. Then the tobermorite 11 Å is undergone a 60 ps relaxation at 298 K utilizing the smart algorithm to obtain the energy minimization. The relaxation time of 60 ps for C–S–H is is enough for the model to achieve the energy optimization. Meanwhile the temperature and pressure are maintained to simulating the indoor enviroment using the Andersen [37] and Berendsen [38] function method respectively. The figure 2 shows the supercell of tobermorite 11 Å which reached equilibrium state after the relaxing process. The uniaxial tension or compression behaviors of monoclinic C–S–H was performed by applying uniaxial pressure loading along the z-direction. During the simulation, each energy minimized structure of C–S–H was subjected to uniaxial pressure in NPT ensemble of 500 ps duration (200 ps equilibration and 300 ps data collection) with timestep of 1 fs, as the pressure was controlled using Souza-Martins function method. After the loading is applied, the bond length and angle of the model change uniformly during 200 ps dynamic simulation. After that, the bond length and angle reach stability, 300 ps dynamic simulation is employed for data collection. The C–S–H model was stretched (or compressed) along the z-direction, the pressure in the other two orthogonal directions (the x, y-direction) was controlled to be zero. The length variation of model box was recorded along the z-direction and the model axial strain $\varepsilon$ was given by equation (1)

$$\varepsilon = \frac{c_1 \times \cos(90^\circ - \alpha) \times \cos(90^\circ - \beta)}{c_0}$$

(1)

(where the $c_1$, $\alpha$, $\beta$ are the lattice parameters of monoclinic crystal after 200 ps, $c_0$ is the initial z-directional length of supercell model).

In this paper, the atomic interaction in C–S–H model is described using COMPASS force field, which is useful to simulate the product of cement hydration and investigate the mechanical performance of C–S–H gel at the nano-scale. Meanwhile the COMPASS force field contains the energy calculation of atomic and molecular interaction in silicate materials, that is suitable for inorganic covalent bonds system and improving the simulation accuracy. For the investigation of mechanical behaviors of C–S–H at a different temperature. The C–S–H model,

| Structure | Tobermorite 11 Å | Hamid Tobermorite 11 Å |
|-----------|-----------------|------------------------|
| Calcium-Silicate ratio | 0.75 | 0.67 | 0.83 | 1 |
| Density g/cm$^{-3}$ | 2.82 | 3.268 | 3.409 | 3.667 |
| Chemical formula | $\text{Ca}_4(\text{SiO}_4\text{(OH)})\cdot5\text{H}_2\text{O}$ | $\text{Ca}_4[\text{Si}_4\text{O}_{10}(\text{OH})_2]\cdot2\text{H}_2\text{O}$ | $\text{Ca}_6[\text{Si}_6\text{O}_{14}(\text{OH})_4]\cdot2\text{H}_2\text{O}$ | $\text{Ca}_6[\text{Si}_6\text{O}_{18}]\cdot2\text{H}_2\text{O}$ |

Figure 2. Supercell of tobermorite 11 Å after the relaxation processes (Yellow: Si; Green: Ca; Red: O; White: H).
has four types Ca/Si ratio (0.75, 0.67, 0.83 and 1, respectively), are applied the axial tension or compression loading during 500 ps. The simulation temperature are set to 100 K, 200 K, 300 K, 400 K and 500 K, respectively, through the Andersen thermostat. Meanwhile kept other parameter setting remained unchanged. The stress-strain curve is obtained at different temperatures condition.

3. Results and discussion

3.1. Uniaxial mechanical property of C–S–H

The crystalline tobermorite 11 Å model with four types of different Ca/Si ratio (0.75, 0.67, 0.83 and 1, respectively) are adopted to investigate the Ca/Si ratio effects on uniaxial mechanical property of C–S–H. The environmental temperature is maintained at 300 K and the uniaxial loadings are applied along z-axis direction.

The figure 3 shows the compressive stress-strain curve of C–S–H with different Ca/Si ratio at 300 K. For all Ca/Si ratio conditions, the stress-strain relation present similar changing process. It is found that the Ca/Si ratio affects the compressive properties of C–S–H in a considerate way. When the strain does not exceed 20%, the structure of crystal almost unchanged. Most of atoms fluctuates around the original locations and the short-range order arrangement is maintained. Meanwhile, the variation of model lattice parameter reduces slightly. But a turning point occurs in the stress-strain curve when the strain exceed 20%. The stress-strain curve trend becomes steep and the stress increased apparently with the strain. Meanwhile the length of model box in x-direction is elongated gradually and the γ angle is decreased. As the tetrahedral SiO₄ structure is compressed, the water molecules and calcium ions start to escape from interior structure and the atoms arrangement is disordered and in irregular state. Once the compression strain εₚ increases to 60%, parts of tetrahedral SiO₄ has been divorced from the interior structure. At the same time, calcium ions and water molecules continue escaping from interior structure and the silicon-oxygen bond starts to cleavage. The crystal structure is distorted and actually in a destructive state. Comparing the emergence of turning point in stress-strain curves with different Ca/Si ratio, the emergence of turning point in Ca/Si ratio of 0.67 and 0.75 are earlier than the ones in Ca/Si ratio of 0.83 and 1.0, which implies the positive correlation between the compression deformation capability of the Ca/Si and the Ca/Si ratio.

The uniaxial tensile stress-strain curves with different Ca/Si ratio are shown in figure 4. It manifests that the Ca/Si ratio affect the tensile stress-strain curves of C–S–H, which indicates the great influence of Ca/Si ratio on the mechanical properties of C–S–H in uniaxial tension. It can be deduced from stress-strain curves of Hamid Tobermorite 11 Å models that as the Ca/Si ratio increases, the tensile mechanical property of C–S–H is strengthened. With Ca/Si ratio of 0.75, the C–S–H has the highest tensile strength as the simulation model is Tobermorite 11 Å, of which the crystalline structure differs from the Hamid Tobermorite 11 Å models (with Ca/Si ratio of 0.67, 0.83 and 1.0). In the Tobermorite 11 Å crystalline structure, the the calcium silicate layers are linked by the chemical bond, wherein the calcium ions and water molecules are distributed in the interlayer region. Meanwhile, the degree of silicon–oxygen tetrahedral polymerization is affected slightly when the uniaxial stress increases. As a result, the C–S–H with Ca/Si ratio of 0.75 has better elasticity and deformation capability, indicating the optimal Ca/Si ratio of C–S–H for high mechanical property.

Figure 3. Curve of C–S–H model with different Ca/Si ratio under axial compression at 300 K.
3.2. Effect of temperature

The stress-strain curves under the axial compression of different Ca/Si ratio at temperatures are shown in figure 5. It is observed that stress-strain curves shows an upward trend as the temperature increases. It indicates that the mechanical properties of monoclinic crystal tobermorite 11 Å are affected by temperature. For the convenience of discussion, the 300 K is defined as the limit. Temperatures below 300 K was called low-temperature (100 K and 200 K), and the ones above 300 K was called high-temperature.
It can be found that the compression performance of the structure is influenced greatly by temperature. The turning point occurs in the curves at the low-temperature. Before the turning point, the stress increased rapidly with the increasing strain, then the curves enter to plastic stage. For different Ca/Si ratio cases, all the curves have the same variation tendency in the different temperature, but the characteristic strain values corresponding to the linear transformation points on different curves are quite different. At the high-temperature, the stress-strain curves are almost in elastic stage as the strain increases. It indicates that the compressive properties of C–S–H are enhanced with the rising temperature. As the temperature increases, the atomic motion becomes active that allow the chemical bond easily twist and deform. As a result, the C–S–H perform bearing capability to compression at high temperature.

As shown in figure 6 are the stress-strain curves of C–S–H model at different temperature. It is observed that the mechanical properties of C–S–H in axial tension was affected by temperature obviously as well as the Ca/Si ratio. It shows that as the temperature increases, the tensile modulus decreases. In the low-temperature, the slope of curves gradually decreases with the increasing strain. In the high-temperature, the stress increases slowly as the strain is loaded. When the temperature is rising, the tensile proportional limit of the C–S–H increases. However, with different Ca/Si ratios the variation trend of tensile stress-strain curves over temperature are distinctive. With Ca/Si ratio of 0.75 as the Tobermorite 11 Å simulation model, the nonlinearity of tensile stress-strain curve is weakened by the increasing temperature, indicating that linear extent is expanded as the temperature rises. For Hamid Tobermorite 11 Å models, it can be observed that, with the increasing Ca/Si ratio the temperature effect on the linearity stress-strain curve is reduced. There is a negative correlation between the polymerization degree of the tetrahedral SiO₄ with the Ca/Si ratio of C–S–H. As the temperature increases, the volume of crystal structure expands. Meanwhile the reduction of the average chain length of silicon chain affects the tensile properties of C–S–H, resulting the coupling effect between temperature and Ca/Si ratio.
4. Conclusions

In this paper, the axial mechanical behaviors of calcium silicate hydrate (C–S–H) during uniaxial compression and tension are studied by applying pressure load to the monoclinic crystal tobermorite 11 Å via the molecular dynamics (MD) simulations. The mechanical properties and behaviors of C–S–H under the axial tension and compression stress are analyzed at the atom scale. The influences of Ca/Si ratio and temperature on the axial mechanical properties of C–S–H are also investigated. The results show that the uniaxial compression property of C–S–H is affected by Ca/Si ratio slightly, but the compression deformation capability of the model was positive correlation with the Ca/Si ratio. The uniaxial tensile property of C–S–H is greatly influenced by Ca/Si ratio, meanwhile the best tensile mechanical property of C–S–H is obtained as the Ca/Si ratio is 0.75. The mechanical properties of C–S–H are affected by temperature. Especially at the low-temperature, the temperature effect on stress-strain curves is obvious. The mechanical modulus of C–S–H under tension and compression are decreased with the increasing temperature. The results imply the strong coupling effect between temperature with Ca/Si ratio on the mechanical property of C–S–H, which plays a significant role in the micro-mechanical properties of C–S–H and the mechanical performance of cement-based materials. The results in this paper have guiding significance to designing high performance cement and its engineering application.

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References

[1] Taylor H FW 1997 Cement Chemistry (London: Thomas Telford Publishing) pp. 114–41
[2] Allen AJ, Thomas J J and Jennings HM 2007 Composition and density of nanoscale calcium-silicate-hydrate in cement Nat. Mater. 6 311–6
[3] Taylor WH, Coled W F and Moorehead R 1964 High-strength calcium silicate hydrate Nature 201 918–9
[4] Taylor H FW 1993 Nanostructure of C–S–H: current status. Adv. Cem. Based Mater. 1 38–46
[5] Zhang X et al 2008 Nanostructure of calcium silicate hydrate gels in cement paste J. Am. Ceram. Soc. 83 2600–3
[6] Vandamme M and Ulm F J 2009 Nanogranular origin of concrete creep PNAS 106 10352–7
[7] Vandamme M and Ulm F J 2013 Nanoindentation investigation of creep properties of calcium silicate hydrates. Cem. Conc. Res. 52 38–52
[8] Beaudoin J J, Raki L and Alizadeh R 2009 A Si-29 MAS NMR study of modified C–S–H nanostructures. Cem. Concr. Compos. 31 585–90
[9] Klar I et al 1998 C–S–H Structure Evolution with Calcium Content by Multinuclear NMR Nuclear Magnetic Resonance Spectroscopy of Cement-Based Materials (Berlin, Heidelberg: Springer) pp 119–41
[10] Cong X and Kirkpatrick R J 1995 Effects of the temperature and relative humidity on the structure of C–S–H gel. Cem. Conc. Res. 25 1237–45
[11] Bauchy M et al 2015 Fracture toughness of calcium-silicate-hydrate from molecular dynamics simulations. J. Non-Cryst. Solids 419 58–64
[12] Skinner LB et al 2010 Nanostructure of calcium silicate hydrates in cements. Phys. Rev. Lett. 104 195502
[13] Faucon P et al 1997 Study of the structural properties of the C–S–H(I) by molecular dynamics simulation. Cem. Conc. Res. 27 1581–90
[14] Hou D et al 2014 Mechanical properties of calcium silicate hydrate (C–S–H) at nano-scale: a molecular dynamics study. Mater. Chem. Phys. 146 503–11
[15] Hou D et al 2014 Calcium silicate hydrate from dry to saturated state: structure, dynamics and mechanical properties. Acta Mater. 67 81–94
[16] Hou D et al 2014 Molecular dynamics study on the mode I fracture of calcium silicate hydrate under tensile loading. Eng. Fract. Mech. 131 557–69
[17] Zaoui A 2012 Insight into elastic behavior of calcium silicate hydrated oxide (C–S–H) under pressure and composition effect. Cem. Conc. Res. 42 306–12
[18] Schröder K-P et al 1992 Bridging hydrotalcite layers in zeolitic catalysts: a computer simulation of their structure, vibrational properties and acidity in protonated faujasites (H Y zeolites) Chem. Phys. Lett. 188 320–5
[19] Palkovic S D, Yip S and Buyukozturk O 2017 Constitutive response of calcium–silicate-hydrate layers under combined loading. J. Am. Ceram. Soc. 100 713–25
[20] Bauchy M et al 2014 Nanoscale structure of cement: viewpoint of rigidity theory. J. Phys. Chem. C 118 12485–93
Pelisser F, Paul Gleize P J and Mikowski A 2012 Effect of the Ca/Si molar ratio on the micro/nanomechanical properties of synthetic C–S–H measured by nanoindentation. J. Phys. Chem. C 116 17219–27

Abdolhosseini Qomi M J et al 2014 Combinatorial molecular optimization of cement hydrates. Nat. Commun. 5 4960–4960

Eftekhar M and Mohammadi S 2016 Molecular dynamics simulation of the nonlinear behavior of the CNT-reinforced calcium silicate hydrate (C–S–H) composite. Composites Part a-Applied Science and Manufacturing. 82 78–87

Pellenq HMSMFVDF-JURI-M. 2012 Confined water dissociation in microporous defective silicates: mechanism, dipole distribution, and impact on substrate properties. JACS 134 2208–15

Qomi M J A et al 2014 Anomalous composition-dependent dynamics of nanoconfined water in the interlayer of disordered calcium-silicates. J. Chem. Phys. 140 054513

Alizadeh R, Beaudoin J J and Raki L 2011 Mechanical properties of calcium silicate hydrates. Mater. Struct. 44 13–28

Al-Ostaz A, Wu W, Cheng A H D et al 2010 A molecular dynamics and microporomechanics study on the mechanical properties of major constituents of hydrated cement. Composites Part B-Engineering. 41 543–9

Hajilar S and Shafei B 2015 Nano-scale investigation of elastic properties of hydrated cement paste constituents using molecular dynamics simulations. Comput. Mater. Sci. 101 216–26

Li K, Shui Z H and Dai W 2012 Molecular dynamic simulation of structural and mechanical properties of cement hydrates: from natural minerals to amorphous phases. Mater. Res. Innovations 16 338–44

Lai M H et al 2020 Dilatancy mitigation of cement powder paste by pozzolanic and inert fillers. Structural Concrete 21 1164–80

Sun H 1998 COMPASS: an ab initio force-field optimized for condensed-phase applications-overview with details on alkane and benzene compounds J. Phys. Chem. B 102 7338–64

Merlino S, Bonaccorsi E and Armbuster T 2001 The real structure of tobermorite 11 Angstrom: normal and anomalous forms, OD character and polytypic modifications Eur. J. Mineral. 13 577–90

Bonaccorsi E, Merlino S and Taylor H F W 2004 The crystal structure of jennite, Ca₉Si₆O₁₈(OH)₆·8H₂O Cem. Concr. Res. 34 1481–8

Pellenq R J M et al 2009 A realistic molecular model of cement hydrates. PNAS 106 16102–7

Hamid S A 1981 The crystal structure of the 11 A natural tobermorite Ca₂₂(Si₆O₁₈)(OH)₆·1H₂O Zeitschrift für Kristallographie - Crystalline Materials. 154 189–98

Dharmawardhana C C et al 2013 Role of interatomic bonding in the mechanical anisotropy and interlayer cohesion of CSH crystals. Cem. Concr. Res. 52 123–30

Andersen H C 1980 Molecular dynamics simulations at constant pressure and/or temperature J. Chem. Phys. 72 2384–93

Berendsen H J C et al 1984 Molecular dynamics with coupling to an external bath J. Chem. Phys. 81 3684–90