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Simulation of the Influence of Intrinsic C-S-H Aging on Time-Dependent Relaxation of Hydrating Cement Paste

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Abstract

The viscoelastic/viscoplastic behavior of cement paste may occur due to intrinsic calcium silicate hydrate (C-S-H) viscoelasticity/viscoplasticity and cement grain dissolution during the hydration process. A numerical model that combines a microstructure model and a finite element calculation model has been developed to predict the time-dependent behavior of cementitious materials based on these two mechanisms, while incorporating C-S-H intrinsic aging. The simulation results from the model suggest that when considering C-S-H aging, the time-dependent properties of C-S-H are capable of generating the aging effect of cement paste, and can become a significant mechanism leading to the overall relaxation of cement paste.

Keywords

Cement Paste; Viscoelastic; Aging; Dissolution; Finite Element Analysis; Microstructure

1. Introduction

Decades of determined research efforts have been dedicated to studying the mechanisms leading to the viscoelastic/viscoplastic (VE/VP) behavior of cementitious materials, yet the primary mechanism is still debatable. Inherent time-dependent properties of the calcium silicate hydrate (C-S-H) constituent has been historically accepted as dominating the long-term, load-induced, time-dependent behavior of the cement paste [1]. Many theories, such as the seepage theory [2, 3] and the viscous shear theory [2, 4], have been proposed to explain the viscoelasticity/viscoplasticity of the C-S-H phases. Another proposed mechanism that leads to relaxation/creep behavior of the cement paste is the dissolution of solid, load-bearing constituents [5, 6], leading to stress redistribution inside cement paste [7–12].

It is difficult to experimentally evaluate the contribution of each of the aforementioned to the overall time-dependent creep/relaxation behavior of cement paste. Relaxation or creep tests on long-term VE/VP behavior of cementitious materials suffer from long test durations and

high costs. During these tests, multiple mechanisms likely occur simultaneously in cement paste composites, so that it is virtually impossible to distinguish and evaluate each mechanism separately. To overcome these experimental limitations, the development of computational methods are strongly motivated such that, in a computational model, the examination of the creep/relaxation induced by inherent C-S-H viscoelasticity/viscoplasticity and load bearing phase dissolution can be carried out concurrently or separately. Recently, this goal was achieved by a computational scheme that couples a microstructure evolution model and a time-stepping finite element routine. This method is able to calculate the overall time-dependent mechanical properties of cement composites [9, 10], and its results suggested that the dissolution of cement grains during the hydration process is likely a significant source of relaxation of early-age cement paste. The simulations also suggested that C-S-H viscoelasticity/viscoplasticity likely is not the major mechanism leading to cement paste creep/relaxation at early ages. This is due to the fact that, if C-S-H relaxation is considered as the dominant mechanism of cement paste relaxation, the simulations suggested that one should observe greater relaxation when cement pastes are stressed or deformed at later ages; this is in direct contrast from the observed aging of relaxation effects in cement paste and concrete that older specimens yield slower relaxation. As suggested in [9, 10], unless the VE properties of the C-S-H dramatically change with age (i.e., C-S-H itself ages), C-S-H viscoelasticity/viscoplasticity cannot be the primary mechanism of cement paste relaxation. Therefore, the major objective of this paper is to test the hypothesis that intrinsic C-S-H aging combined with C-S-H viscoelasticity is sufficient to account for the overall aging viscoelastic behavior of cement paste.

To meet the objective, we revisit the model derived in [9, 10], and modify the constitutive relations for the C-S-H phase in order to account for intrinsic aging within that phase. It has been suggested that C-S-H may age due to increased polymerization with age [13], which could be exacerbated by drying effects [14]) or increased packing density of C-S-H globules with age [15]. Here, we utilize the time-shift approach to quantify the aging in the constitutive function of C-S-H; this approach has been commonly utilized to model aging of polymers due to increased polymerization with time (see, e.g., discussion in [16]) and has been applied sparingly to model cement paste aging [17, 18]. Through virtual experimentation using the newly modified model, the contribution of inherent C-S-H time-dependent behavior (including C-S-H viscoelasticity/viscoplasticity and intrinsic C-S-H aging) towards the overall time-dependent creep/relaxation behavior of the cement paste can be examined more accurately and thoroughly.

2. Computational method

2.1 Model from previous research

A computational model was developed in previous research by the authors [9, 10]. This model is comprised of two parts: a microstructure evolution model and a time-stepping finite element analysis model. The microstructure model utilized in the research is called THAMES (Thermodynamic Hydration And Microstructure Evolution) [19, 20]. The original measured clinker constituent mass fractions, the particle size distribution of the cement and the water to cement mass ratio (w/c) are input into THAMES at the sub-particle level. Based

on empirically derived equations for clinker reaction rates [21], THAMES integrates the dissolution and precipitation processes associated with hydration forward in time using discrete time steps. At each time step, THAMES first calculates the pore fluid composition as a result of dissolved cement grains. Then based on the calculated fluid composition, a thermodynamic engine GEMS (Gibbs Energy Minimization) [22, 23], which has been implemented in THAMES, will calculate the equilibrium solution speciation in the pore fluid and the precipitated solid constituents. The newly formed hydration products are then spatially located in the microstructure model based on many rules, such as the local geometric information and the growth habit of the constituent. Based on thermodynamic equilibrium calculations algorithm implemented in GEMS as well as the empirically derived dissolution kinetics [19] utilized in the hydration model, THAMES produces 3D snapshots (collections of voxels) of hydrating cement paste at different ages. The simulated microstructures obtained at different ages are in the form of a 3D array of voxels, where each voxel consists of a single phase. More detailed description of the microstructure model can be found in [9, 10, 19].

The second major part of the computational scheme is the finite element calculation. To predict the time-dependent VE/VP behavior of cement composite, the whole computational scheme was discretized in time. At each time step, the finite element routine receives a 3D microstructure snapshot from THAMES, which is directly mapped to a finite element mesh. In the 3D snapshots, each voxel is an eight-node tri-linear cubic element consisting of a single phase, and the phases are assigned with isotropic elastic or VE properties in the finite element calculation. Averages of Voigt-Reuss bounds are used for anisotropic phases. Through minimizing the total mechanical energy stored in the whole microstructure under an external applied periodic strain-controlled displacement boundary condition, the boundary value problem at each time step can be solved [24]. To calculate the time-evolving mechanical properties of the whole composite, the time-evolving stress field history under the specific applied boundary strain are averaged spatially over the whole composite volume at all time steps [16, 25]. The virtual work principle is used in the scheme with the viscoelastic model to calculate the mechanical energy of the viscoelastic phases (C-S-H^a) inside the microstructure [9, 26, 27]. In the computationally implemented model, the time-dependent VE/VP bulk and shear moduli of cement paste are first obtained under periodic volumetric and shear boundary conditions. Laplace transformation analysis is then applied on the resulting data to determine the VE/VP Young's modulus and VE/VP Poisson's ratio.

One major challenge for the VE/VP moduli prediction scheme is to account for the cement grain dissolution process associated with the hydration reaction. During the hydration reaction, cement grain phases dissolve and solid hydrates precipitates under applied stress/strain. The stress and strain distribution inside cement composite would change as a result of this dissolution and precipitation process. To simulate this process, one assumption was made in the program that stress would redistribute much faster than any chemical process. When the phase component inside one voxel changes between two neighboring time steps, stress would redistribute instantaneously from the dissolved voxel to the surrounding solid

^aCement chemistry notation is used to designate phase compositions, where C = CaO, S = SiO₂, H = H₂O, and C-S-H designates a poorly ordered calcium silicate hydrate phase with variable composition.

phases, which increases the stress they must support. Meanwhile, the newly precipitated hydrate phases form in the deformed configuration conforming to the pre-existing deformation, while neglecting all the previous memory of the historical response. At the moment of formation, the new phases are unable to conduct linear momentum associated with external tractions or displacements previously applied to the composite material. Since we are explicitly considering here the time dependent response to externally imposed tractions or displacements (e.g., creep and relaxation) and are not simulating free deformation associated with crystal growth pressures, we do not consider internally balanced stresses. Thus, phases are necessarily formed in a stress-free state. In this way, apparent strain was produced inside the microstructure, further complicating the finite element calculation [9]. Apparent strain is defined in the paper as the strain that exists independent of the state of stress. Unlike free strains (e.g., strain induced by changes in temperature or moisture state of the material), the apparent strain here does not involve a change in the atomic or molecular spacing from the reference configuration. However, from a book-keeping perspective, apparent strain is treated in the same fashion as free strains. This whole dissolution process occurs simultaneously with the creep/relaxation behavior of the VE phases (C-S-H). In the simulations, the utilized cement pastes were partially saturated, and water was assumed to be able to flow freely among the nearby void voxels, which means that even if the aqueous solution voxels got pressurized, the pressure will relax with time. Because the flow distance of the fluid is relatively short in the utilized cement paste microstructures, this pressure-release process of the fluid voxels are assumed to occur much faster than the stress redistribution process. The term “apparent VE/VP” moduli is reserved in the paper for modeling the constitutive time-dependent VE/VP behavior of cement paste that occur due to the generation of apparent strain as a result of the dissolution and precipitation process during the hydration reaction. The inherent viscoelasticity/viscoplasticity of the VE/VP phases (C-S-H) inside cement paste does not contribute to the apparent VE/VP moduli. Note that even if no phase had inherent VE/VP behavior, the hydration process under the above assumptions would produce apparent VE/VP effects.

One major advantage of this newly developed computational scheme is that it allows one to investigate and evaluate the influences of different mechanisms concurrently or individually on the overall viscoelasticity/viscoplasticity of cement paste. To evaluate the contribution of intrinsic C-S-H viscoelasticity/viscoplasticity, time-evolving constitutive properties could be assigned to any given phase while analyzing constant unchanging cement composite microstructure. To evaluate the significance of microstructure evolution on the overall creep/relaxation of the cement paste, all the phases inside cement paste could be assigned with purely elastic properties while the cement paste microstructures evolve with time. By such simulations, these two different mechanisms could be analyzed and compared in the computational program. More detailed descriptions of this computationally implemented model are included in [9, 10].

2.2 Including intrinsic C-S-H aging

It has often been suggested that the aging effect of concrete is as a result of intrinsic aging of C-S-H properties, which can occur due to either polymerization [13] or densification [15]. Meanwhile, as hydration progresses with time, the amount of C-S-H inside cement paste

also increases. Many modeling approaches (such as the solidification theory [28, 29] and solidification theory incorporating intrinsic C-S-H aging [30]) have been developed to predict the aging behavior of concrete. However, according to the creep/relaxation prediction model [9, 10], increases in C-S-H volume fraction yields more creep/relaxation simply because a larger volume fraction of C-S-H requires a greater fraction of the composite stress to be transmitted through C-S-H phases (and thus more creep/relaxation associated with C-S-H phases). In the traditional modeling approaches, it is assumed that cement grains are incapable of carrying stresses and the averaged stress transmitted by C-S-H *decreases* as the volume fraction of C-S-H increases. Thus, a modeling approach (such as existing analytical approaches) that neglects the fact that cement grains carry stress is inadequate to capture the effect of C-S-H aging on macroscale time-dependent properties of the cement paste, motivating the utilization of the above-mentioned model to predict the overall creep/relaxation of cement composites.

In the previous creep/relaxation prediction model, all the hydrated and unhydrated phases (except C-S-H) were assigned age-independent linearly isotropic elastic moduli. As intrinsic C-S-H aging properties would lead to less overall relaxation in cement paste, to include this mechanism into the computational scheme, the finite element calculation, where the moduli of each phase were assigned, was modified in such a way that C-S-H phases formed at different ages or different time steps were treated as different phases. Each of these phases was assigned different VE/VP mechanical properties based on the initial non-aging VE/VP properties of C-S-H, the aging function of C-S-H, and the time when a specific C-S-H particle is formed. THAMES and finite element code can easily handle the many phases necessary.

The time-age superposition method [16], a conventional way to model the age-evolving properties of aging VE/VP materials, was used to model the aging time-dependent moduli of C-S-H phases. According to the time-age superposition method, the effect of different ages is incorporated by simply shifting the retardation or relaxation times without changing the shape of the compliance or moduli functions of the materials on the log time scale. By selecting one age to be the reference age or the ‘master’ age, the retardation or relaxation times for the other ages can be simply shifted by a constant time shift factor a_j [30]

$$a = (t/t_{ref})^f, \quad (1)$$

where t is the age of C-S-H. The aging factor f in Eq. (1) is a constant (constitutive) factor throughout each simulation, and controls the aging rate of C-S-H. By simply changing the value of the aging factor f , all of the time shift factors at different ages change simultaneously. Note that as aging and relaxation occurs simultaneously on the same time scale, the constitutive equation for aging C-S-H can be written as [31]

$$\sigma_{11}(t; f, t_{ref}) = \int_0^t E(\xi(t) - \xi(s)) \frac{d\varepsilon_{11}(s)}{ds} ds, \quad (2)$$

where $E(t)$ is the reference age viscoelastic Young's modulus, $\sigma_{11}(t; f, t_{ref})$ is the axial stress function, $\varepsilon_{11}(t)$ is the axial time-dependent strain, and $\xi(t)$ is a pseudo-time expressed as

$$\xi(t) - \xi(s) = \int_s^t \frac{ds'}{a(s')}. \quad (3)$$

From the above three equations, one can see that when $f=0$, the C-S-H phases are non-aging materials whose mechanical properties do not evolve with age. As the value of f increases the time shift factor also increases, yielding a faster and more obvious aging effect on the resultant C-S-H viscoelasticity. Thus, by comparing the simulation results with different input values of f , the influence of intrinsic C-S-H aging towards the overall creep/relaxation behavior of the cement paste can be evaluated.

While one could devise an analytical composite model using a simplified geometry (parallel, composite spheres, etc.) to simulate an aging, viscoelastic, composite material, the computationally implemented model described herein allows one to simultaneously capture effects induced by phase dissolution and to assign particular constitutive behavior to each single phase. Thus, the results from these simulations provide enhanced insight into aging, creep, and relaxation beyond that attainable from classical, analytical models.

3. Simulation results & discussions

Several virtual experiments were conducted in the paper to evaluate the influence of intrinsic C-S-H aging towards the overall creep/relaxation behavior of the cement paste at early ages (from 1 d to two months). The microstructures analyzed are all of 100^3 voxels, where the volume of each cube-shaped voxel is $1 \mu\text{m}^3$. A list of these microstructures were utilized in the computational model to predict the time-evolving apparent VE/VP moduli of cement composites. All microstructures utilized in the virtual experiments were generated by THAMES on simulated cement pastes of the ASTM Cement and Concrete Reference Laboratory (CCRL: <http://www.ccrl.us/>) Proficiency Sample Cement 168 [32] under isothermal, sealed conditions of 25°C (298 K), and the elastic moduli of each phase (*e.g.*, C_3S , C-S-H) were taken from previous studies [33].

One thing to note about the utilized microstructures is that, in reality, for the volume of 1 cubic micron voxel size, multiple phases might exist within this volume rather than one single phase. For the voxels of C-S-H phase analyzed in the microstructure model, nanosclae porosity as well as nanoscale CH could reside within this volume, intermixed with the predominant C-S-H. In the computational model, when assigning mechanical properties to

these $1 \mu\text{m}^3$ C-S-H voxels, the “mean field” properties of this volume of “C-S-H phases” are utilized, which means that the assigned mechanical properties for each C-S-H voxel inherently account for the porosity or nanoscale CH that might reside within this $1 \mu\text{m}^3$ volume; this is accomplished by utilizing mechanical properties obtained from experimental investigations probing a similar length scale (e.g., nanoindentation tests that probe a roughly 1 micron cube interaction volume). Thus, in this paper, the “intrinsic properties of C-S-H” refers to the “mean field” properties of C-S-H. These intrinsic properties of the C-S-H phases could age with time (due to either polymerization [13] or densification of C-S-H [15]), and this aging process of C-S-H is accounted for in the aging functions discussed in section 2.2.

3.1 Different time shift factors

From previous research, the main reason that C-S-H viscoelasticity was determined unlikely as the dominant mechanism leading to creep/relaxation of the cement paste is the fact that this mechanism gives conflicting simulation results of the cement paste aging effect in comparison to experimental evidence [17, 34]. Larger relaxation is observed in older cement paste specimens when C-S-H viscoelastic properties do not age because of the higher C-S-H volume fractions in older, more fully hydrated specimens. However, myriad experimental data indicate that older specimens exhibit lower rates of relaxation than younger specimens. Previous simulations assigned a non-aging VE/VP constitutive function for C-S-H. Incorporating the intrinsic C-S-H aging effect into the computational model so that the overall time-dependent properties of C-S-H evolve with age may yield different early age cement paste aging effects.

As will be discussed in section 3.2, changing the values of the aging factor f enables one to evaluate by virtual experiments the effect of intrinsic C-S-H aging on the overall creep/relaxation behavior of the cement paste. In this paper, the reference age t_{ref} in Eq. (1) was set to be equal to 7 d, and the VE Young's modulus of C-S-H at the reference age in Eqs. (2) and (3) was prescribed as $E(t) = 11.2 \text{ GPa} + 11.2 \exp(-0.2t) \text{ GPa}$, where t denotes the loading time of each specific C-S-H voxel, while the Poisson's ratio of C-S-H was assumed to have a time-independent value of 0.25. The choice of reference age is arbitrary, so long as one is consistent in shifting all other ages to the relaxation function determined for the reference age. The aging factor was assigned to be $f = 0.7, 1.4, 2.1$ or 2.8 to simulate different aging rates. All the other phases were assumed to be purely elastic with constant mechanical properties.

Figure 1 shows the simulation results for a 0.40 w/c cement paste with different C-S-H aging factors. These simulation results were generated using a fixed microstructure to eliminate any relaxation effects due to hydration, so that relaxations occur solely by C-S-H time-dependent properties. The non-aging, 1 d curve was generated at the loading age of 1 d, and the other curves were generated at the loading age of 7 d.

Figure 1 shows that faster C-S-H aging (*i.e.*, higher f) results in less relaxation in the apparent VE/VP Young's modulus of the composite. When the aging factor is increased to a sufficiently large value, 7 d specimens potentially can exhibit slower relaxation than the 1 d old specimen, especially for the case when $f = 2.1$ and $f = 2.8$. As already described,

simulations suggested that C-S-H viscoelasticity/viscoplasticity could not be the dominating mechanism leading to cement paste creep/relaxation at early ages because that mechanism alone implies greater overall relaxation in older specimens, in conflict with experimental data. However, the new simulations shown here demonstrate that the well-known aging effect can be recovered by introducing intrinsic aging of the C-S-H constitutive properties.

3.2 Different loading ages

Through utilizing unchanging microstructures at each specific age, Figure 2 shows the simulation results of the 0.40 w/c cement paste subject to loading at ages of 1 d, 7 d, 14 d and 28 d when assigned with different aging factors $f = 0$ (no C-S-H aging), 0.7, 1.4 or 2.1. The C-S-H phase was assigned a VE Young's modulus prescribed as $E(t) = 11.2 \text{ GPa} + 11.2 \exp(-0.2t) \text{ GPa}$ at the reference age of 1 d, and the Poisson's ratio was assigned the same value as in section 3.1 for the phase C-S-H. All the other phases are treated as purely elastic with constant mechanical properties. Apparent VE/VP Young modulus of the cement paste composite, which are normalized by the instantaneous elastic Young's modulus of each cement paste composite, are shown in Figure 2.

Figure 2(a)–(b) show that older specimens exhibit larger and faster relaxation than younger specimens when f is small (little aging effect). When the aging factor for C-S-H is higher, as shown in Figure 2(c)–(d), older specimens relax slower than younger ones, which is in agreement with myriad experimental data regarding aging viscoelasticity in cement paste. Thus, we have confirmed the hypothesis that the aging VE/VP behavior of cement paste may be accounted for strictly by time-dependent behavior of the C-S-H, provided that the viscoelastic properties of the C-S-H exhibit strong enough aging. This finding should not be construed to conclude that strong C-S-H aging is *necessary* for cement paste to exhibit aging viscoelastic behavior, only that it is theoretically *sufficient*. To investigate more thoroughly how cement paste at different loading ages responds to the different aging rates of C-S-H phases, or to investigate how sensitive cement pastes at different loading ages are to changes in f , Figure 2 shows the normalized apparent VE/VP Young's modulus of cement paste (which are normalized by the instantaneous elastic Young's modulus of each cement paste composite) after 56 d of loading versus the change in the aging factor f . This figure uses the same data as in Figure 2, plus one more set generated with $f = 2.8$; if the cement paste is loaded at 14 d, the normalized VE/VP Young's modulus after 56 d of loading, which is at an actual age of 70 d, was used in the figure.

When the aging factor is small ($f < 0.7$), Figure 3 indicates that cement paste loaded at an earlier age, such as 1 d, has a shallower slope than paste loaded at a later age, meaning that cement pastes are less sensitive to the change in the intrinsic C-S-H aging rate when loaded at earlier ages. The major reason for this phenomenon is that younger specimens have a lower C-S-H volume fraction. When the aging effect is minor (i.e., f is small) and dissolution effects are excluded, the volume of C-S-H dominates the behavior, leading to overall less relaxation in younger specimens, as shown in Figure 2(b). As the aging factor increases, the slopes of the earlier age curves gradually become steeper than later age curves (especially when the aging factor is greater than 1.4), meaning that earlier age cement paste becomes more sensitive to the intrinsic C-S-H aging effect. This phenomenon can be

explained from Eqs. (1)–(3). When substituting Eq. (1) into Eq. (3), under the same reference age t_{ref} the pseudo-time function has a t^{1-f} dependence, which is more sensitive to changes in f at early ages with smaller values of t , and thereby the C-S-H VE Young's modulus, compared to an older specimen. Thus, from this aspect, younger cement pastes are more sensitive to the change in the values of f , and thus to the intrinsic aging of C-S-H phases.

3.3 Different w/c

Additional virtual experiments were also conducted to examine the influence of intrinsic C-SH aging on cement paste with different w/c. Cement pastes with w/c varying from 0.35 to 0.5 were subjected to loading at 7 d with one of four different aging factors, $f = 0, 0.7, 1.4$, or 2.1. The C-S-H phase was assigned the same properties as in section 3.2. Figure 4 shows the time evolving apparent VE/VP Young's modulus of the cement paste composites, normalized by the instantaneous elastic Young's modulus, using different values of the aging factor. Figure 4 shows the normalized apparent VE/VP Young's modulus after 56 d of loading versus the change in the aging factor f to demonstrate the sensitivity of different w/c cement pastes towards intrinsic C-S-H aging.

From Figure 4 and 5, one can see that in all cases, cement pastes with a lower w/c value relax faster and are more sensitive to intrinsic C-S-H aging due to their higher C-S-H volume fractions at early ages [10]. Under the same loading age and the same aging factor, the relative amount of C-S-H phases is the only factor affecting the relative relaxation of cement paste under the mechanism of C-S-H time-dependent properties.

3.4 Combining aging C-S-H relaxation with cement grain dissolution effect

Figure 6 shows the simulated results for 0.40 w/c cement paste with loading ages of 1 d or 7 d when the cement grain dissolution is included in the model together with C-S-H viscoelasticity and intrinsic C-S-H aging. The C-S-H phase was assigned the same properties as in sections 3.2 and 3.3. To generate the data in Figure 6, microstructures from four different ages varying from 1 d to 18 d were utilized in the program to account for microstructure evolution.

The non-aging data in both plots of Figure 6 were generated when considering the mechanisms of C-S-H viscoelasticity and dissolution of cement grains, while the aging lines are generated when including the mechanism of intrinsic C-S-H aging besides the aforementioned two mechanisms. The plots show that for cement paste loaded at 1 d, the contribution of the intrinsic C-S-H aging towards the overall relaxation of cement composite is relatively limited at very early ages, especially at ages less than 5 d. The rapid dissolution rate of cement grains (due to the active hydration reaction) still dominates the early age relaxation behavior of cement paste [9, 10]. As cement paste ages, with the increase in the amount of C-S-H constituents, intrinsic C-S-H aging becomes a more significant mechanism affecting relaxation - depending on the chosen values of the aging factor - as shown in the later age part of Figure 6(a) and the whole of Figure 6(b).

4. Conclusion

A numerical model, which couples 3D digital microstructure evolution algorithms for tracking phase change with a time-stepping finite element method, was modified in this paper to account for and evaluate the intrinsic aging effect of C-S-H phases in cement paste as a mechanism influencing the time-dependent behavior of cement composites. From the simulation results, it shows that when intrinsic C-S-H aging operates together with C-S-H viscoelasticity, the modeled time-dependent VE/VP properties of cement paste are consistent with the experimentally observed aging effect on cement paste relaxation rates. Although the mechanism of cement grain dissolution likely dominates the time-dependent creep/relaxation behavior of the cement paste at early ages, as the volume fraction of C-S-H increases inside cement paste with age, the effect of C-S-H aging towards the overall creep/relaxation behavior of cement paste also likely increases.

Experiments are needed to directly measure the aging of C-S-H VE/VP properties, in order to test and help improve the accuracy of the aging function used in this model. Due to a lack of reliable data, the simulation results presented herein highlight only the *potential* for C-S-H aging to strongly contribute to the overall aging of cement paste creep and stress relaxation. Reliable experimental data that probe explicitly the age dependency of C-S-H creep or relaxation are needed to further evaluate the *extent* to which aging of the C-S-H plays a role in the aging, time-dependent behavior of the cement paste.

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Figure 1.

Apparent VE/VP Young's modulus for 0.40 w/c at the loading age of 1 d and 7 d under different intrinsic C-S-H aging conditions. The non-aging $E(t)$ due to loading at 1 d is included in this figure as a comparison.

Figure 2.

Apparent VE/VP Young's modulus for 0.40 w/c cement paste at different loading ages with different aging factors of (a) $f=0$ (non-aging), (b) $f=0.7$, (c) $f=1.4$ and (d) $f=2.1$

Figure 3.

Normalized apparent VE/VP Young's modulus of 0.4 w/c cement paste after being subjected to 56 d of constant controlled strain applied at the age indicated in the legend versus the aging factor f . The normalization was made by instantaneous elastic Young's modulus. The data were selected from the data utilized in generating Figure 2 and one more set of data when the aging factor $f = 2.8$.

Figure 4. Normalized apparent VE/VP Young's modulus of cement paste with different w/c under the loading age of 7 d with different intrinsic C-S-H aging factors of (a) $f=0$ (nonaging), (b) $f=0.7$, (c) $f=1.4$ and (d) $f=2.1$, where normalization was made by the instantaneous elastic Young's modulus.

Figure 5.

Normalized apparent VE/VP Young's modulus of cement pastes with different w/c after 56 d of loading (loading age of 7 d) versus the aging factor f , where normalization was made by instantaneous elastic Young's modulus. The data was selected from the data utilized in generating Figure 4.

Figure 6.

Apparent VE/VP Young's modulus for 0.40 w/c under different loading ages of (a) 1 d and (b) 7 d with different aging factors.