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*- Technical Paper -*

# A HIERARCHICAL APPROACH TO EVALUATE THE MECHANICAL BEHAVIOUR OF CEMENT PASTE UNDER COMPRESSIVE LOADING

Mylvaganam NITHURSHAN*1, Yogarajah ELAKNESWARAN*2, Yuya YODA*3, and Ryoma KITAGAKI*4

## ABSTRACT

The present paper proposes the integrated multi-stage approach to predict the mechanical properties of cement paste with the function of hydration time. To this purpose, in the initial stage, volume fractions of hydration products are reliably predicted, and representative volume element (RVE) is then developed in MATLAB. Afterwards, the developed RVE is transferred to COMSOL to perform the numerical analysis, where intrinsic mechanical properties are computed. The proposed model results showed good agreement with the existing analytical model and experimental results.

Keywords: multi-stage approach, volume fractions, representative volume element, cement paste

## 1. INTRODUCTION

Concrete has been a first-choice building material due to its low cost compared to its compressive strength [1]. Recent studies revealed that the major components of hydrated cement, such as calcium silicate hydrate (C-S-H), calcium hydroxide (CH), and ettringite contribute strength and stiffness to concrete [2,3]. It has also been reported strength of concrete alters by its microstructure. The expected superior performance of concrete leads to the usage of the additives such as pozzolanic materials (e.g., silica fume and fly ash) that modify the concrete microstructure by converting the weak CH crystals into stiffer C-S-H [4] and fibrous material [5] (e.g., steel, glass and polypropylene) which enhance the cracking-related properties. However, the inclusion of them in the concrete mix raises the cost of construction and using them above certain limits negatively affects the performance of concrete properties under some loading conditions. For instance, even though nano-silica accelerates the early hydration process and thus increases the early strength gain of concrete by generating nucleation sites, the experimental study [6] showed that beyond 3% replacement of nano-silica decreases the compressive strength at all ages (3, 7, 28 and 56 days). This may be attributed to the agglomeration of excess nano-silica, which suppress the hydration reaction of cement particles with increased water demand in producing C-S-H gel [7].

Similarly, Du et al. [8] reported that the inclusion of nano-silica above 0.9% of cement weight did not increase the chloride resisting capacity due to the agglomeration. Hence, carrying out the optimum design of cementitious material is very much needed to improve the performance of concrete structures and lessen the construction cost. However, such an optimum design also demands an appropriate tool that explicitly predicts the mechanical properties of concrete at different stages. Moreover, predicting the evolution of mechanical properties such as compressive strength and young's modulus and poisons ratio of concrete with its curing age is crucial to a structural engineer to analyse or simulate the existing/new concrete structures under different loading (e.g., compression, tension, and shear) and environmental conditions for mitigating the deterioration of structures.

The major challenge in precisely predicting the mechanical properties of concrete is the complex nature of the microstructure of cementitious materials. Hence, over the decades, several methods have been proposed to correlate the macroscopic properties of concrete with the nano/micro properties of cementitious materials. Multi-scale or homogenisation approach is therefore required to pass the nano/micro properties to the following scale, which is the mortar (meso) or concrete (macro) scale [9–11]. For instance, Maekawa et al. [12] proposed a set of constitutive equations that systematically integrates the chemo-physics (heat hydration, temperature, shrinkage, dynamics in pore structures, moisture hygro-thermodynamics etc.) in the microscale and the mechanical properties evolving through multi scales, to predict the macroscopic behaviour of concrete subjected to a wide variety of loading and environmental conditions. In addition, the representative volume element (RVE) for each scale, as shown in Fig. 1, has been proposed by several researchers to simulate the actual microstructure of the cement paste, mortar and concrete [9,13,14]. The present study is a one-step towards that direction, starting from predicting the mechanical properties of cement paste on the micro-scale ($10^{-6}$ m).

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*1 Graduate School of Engineering, Hokkaido University, JCI Student Member
*2 Associate Prof., Division of Sustainable Resources Engineering, Hokkaido University
*3 Engineering Researcher, Shimizu Corporation, Shimizu Institute of Technology, JCI Member
*4 Associate Prof., Division of Human Environmental System, Hokkaido University, JCI Member

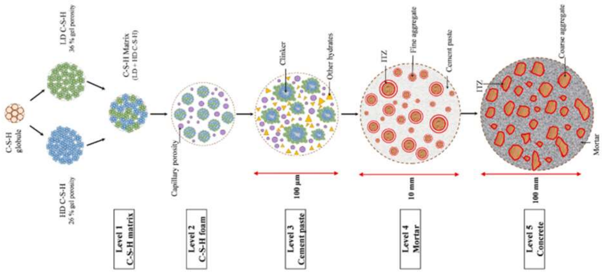

Fig.1 Different length scales of cementitious materials [15]

## 2. MODEL DESCRIPTION

The research aims to predict the compressive strength development characteristics of hydrated ordinary Portland cement (OPC) cement paste. As discussed in the introduction section, the combined effect of chemical-hygral-thermal processes highly influences mechanical properties. Therefore, a coupled thermo-hygral-hydration model developed by Krishnya et al. [16] was chosen to predict the volume fraction of hydration products with hydration time, as illustrated in Fig. 2 (stage 0). The model (Krishnya et al.) was developed based on the widely accepted cement hydration model proposed by Parrot and Killoh [17] combined with Cemdata18 [18] and the PHREEQC thermodynamic database by considering the formation of C-S-H (prime binder) in two different densities; Low density and High density [16]. Moreover, this model can continuously predict the phase changes with hydration time, whereas other hydration models could predict the phases present in the cement paste at a particular age. In stage 1 (Fig. 2), the representative volume element (RVE) was developed based on the results from stage 0 in three-dimensional (3D) and two-dimensional (2D) space to compare the predicted results with computational cost. Finally (in stage 2), numerical analysis was carried out on the RVE to compute the mechanical properties of cement paste. The modelling approach is comprehensively discussed below.

### 2.1 Prediction of volume fractions

The proposed model starts with accurately predicting the volume fraction of phase present in cement paste. It has been reported that determining the volume of different phases available in the cement paste is sufficient to compute the mechanical properties [19]. When water is added to cement, various chemical reactions, called hydration, produce several new chemical components such as typically C–S–H, portlandite, ettringite, monosulfate, hydrotalcite, Fe-siliceous hydrogarnet, capillary pores, etc. However, it is reported in the literature that the hydration products such as C–S–H, portlandite, ettringite and monosulfate contribute to the strength development of the cement matrix [3]. In addition, a mandatory volume reduction, defined as chemical shrinkage, occurs during the hydration process due to the molar volume change between new hydration products and clinker minerals. Hence, to develop an RVE of cement paste in stage 1 (Fig. 2), it was considered that hydrated cement paste comprised three phases: hydrated phase, unhydrated phase and pores.

Unlike in the past studies (in which only C-S-H and/or portlandite were considered to be the hydrated product and other hydrated products were neglected), the hydrated phase is assumed to have consisted of C–S–H, portlandite, ettringite and monosulfate. The unhydrated phase is included unreacted clinker components such as $C_3S$, $C_2S$, $C_3A$ and $C_4AF$, while the pore phase is computed from the chemical shrinkage and capillary porosity of cement paste. Accordingly, the volume fraction of three major phases (hydrated, unhydrated and pore) was predicted from the phase assemblage of $C_3S$, $C_2S$, $C_3A$, $C_4AF$, portlandite, ettringite, monosulfate, chemical shrinkage and capillary porosity. Moreover, the volume fraction of these phases was predicted continuously over the hydration period in this study, whereas in the past, volume percentages of phases were determined at a specific age using experimental results such as (TGA) and X-ray diffraction analysis (XRD) [20,21].

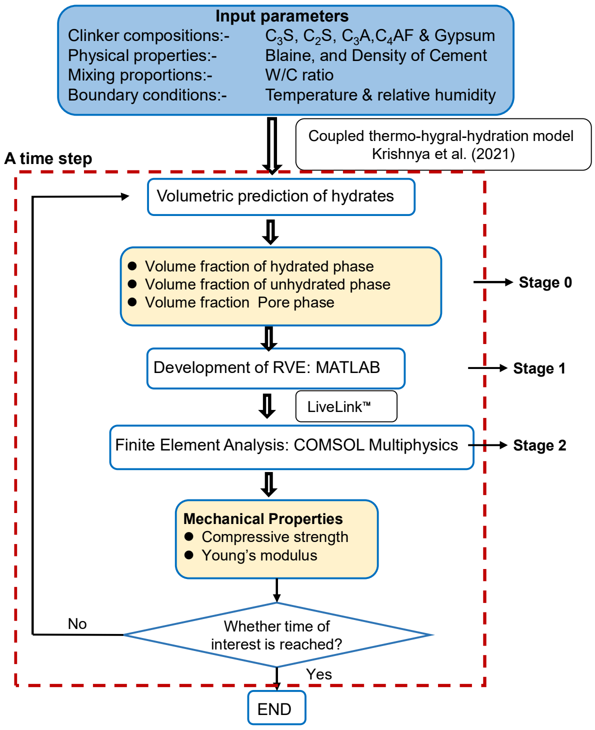

Fig.2 Structure of the proposed multi-stage model

### 2.2 Developing RVE geometry

Reproducing the exact microstructure of cement paste is unattainable with the existing computing power at a reasonable processing time. For instance, the gel pores and the smallest capillary pores in the paste are generally on a nanometer scale (0.5-10 nm), and depicting those phases on the micro-scale leads to computational complications. However, microstructural models, which simplify the microstructure of pastes, have been proven to successfully analyse the mechanical properties of cement paste [22]. The representative volume element (the smallest volume adequately represents the behaviour of that whole material) was therefore developed in MATLAB based on the predicted volume fractions from the previous step (stage 0). In addition, to maintain the cement paste's heterogeneity, the matrix material was assumed to be the hydrated phase, and the other two phases (anhydrous and pore) were randomly dispersed in the matrix. The anhydrous and pore were considered as spherical particles [23] for 3D to easily represent them, including their location and shape. Codes were written in MATLAB for the execution of random distribution and non-overlapping of the particles in the RVE using the "take and place" method [14]. To more precisely simulate the hydration process and the microstructure development, the size of the spherical particles that represent the unhydrated phase was considered to be the function of the degree of hydration, as shown in Eq. 1 [24].

$$R_t = R_i(1-\alpha)^{1/3} \tag{1}$$

where, $R_t$, $R_i$ and $\alpha$ are the radii of a particle at time t, the initial radii of the particle, and the degree of hydration of cement, respectively.

The actual particle distribution of clinker and pores varies widely from nanometer to micrometre, and the depiction of the natural distribution challenges the computational effort. The initial diameter of the anhydrous particles was therefore chosen as 10 µm; for pores, two sizes (6 and 4 µm) were considered according to the literature [1,9]. Subsequently, the total number of particles for both phases was calculated from the diameter and volume fraction of phases computed in stage 0. Moreover, the length of the RVE was chosen to be 100 µm as many studies witnessed the size suitability for the cement paste with regards to homogenisation and material characteristics. The developed geometry for RVE for a typical 28-day aged cement paste is shown in Fig. 3.

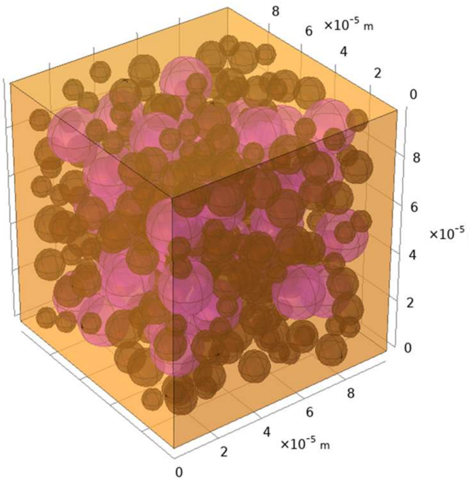

Fig.3 Developed RVE of cement paste (orange: hydrated phase, pink: unhydrated particles, and brown: pores)

### 2.3 Numerical analysis of the RVE

The generated geometry of RVE in MATLAB (in stage 1) was transferred to COMSOL Multiphysics 6.0 via LiveLink™ for MATLAB® to perform finite element analysis on it. The structural Mechanics Module was used to simulate the compressive behaviour of the cement paste.

#### 2.3.1 Material model for phases

The hydrated phase was considered to behave according to the concrete material model based on the Bresler–Pister criterion used to predict the strength of concrete under multiaxial stresses. This failure criterion is expressed in Eq. 2.

$$F_y = \sqrt{J_2} + k_1 I_1^2 + k_2 I_1 + k_3 \tag{2}$$

Where $J_2$, $I_1$, and $I_2$ are stress invariants, and $k_1$, $k_2$, and $k_3$ are parameters depending on the uniaxial compressive strength ($\sigma_{cs}$), the uniaxial tensile strength ($\sigma_{ts}$), and the biaxial compressive strength ($\sigma_{bc}$). The compressive strength of the C-S-H matrix, which consisted of LD and HD C-S-H, was computed from the multi-scale model proposed by [16] as the C-S-H matrix mainly resists the compressive stress [3]. In addition, tensile strength ($\sigma_{ts}$) and the biaxial compressive strength were assumed to be 1/8- and 1.2 times compressive strength because there is no appropriate method to compute those strengths of the C-S-H matrix. The assumption is from the Griffith strength model [25], which was adopted in similar studies [1]. The effective modulus of elasticity was calculated based on the volume fraction of components (C–S–H, portlandite, ettringite and monosulfate) for the hydrated phase, and the Poisson's ratio was assumed to be 0.24.

The unhydrated phase, which contains unhydrated clinker, was considered to behave in a linear elastic manner as used in other studies. Since the material properties of clinker do not change with hydration time, the modulus of elasticity and Poisson's ratio were considered to be 130 GPa and 0.3, respectively. The pore phase was incorporated as voids immersed within the hydrated phase.

#### 2.3.2 Boundary conditions for the geometry of RVE

The boundary conditions that coincide with the actual uniaxial compressive strength test setup were simulated in COMSOL. The bottom surface of the RVE was constrained in the x and y direction, and the prescribed displacement was applied on the top surface in the negative y direction. Periodic boundary conditions were also applied to two opposite vertical edges to ensure the accuracy of the solution. Four-node tetrahedral elements have been used to mesh the RVE for finite element analysis. However, pores were excluded in messing but considered as embedded voids within the hydrated phase.

Average stress was calculated as the reaction forces at the bottom surface over the cross-sectional area, and the average strain was measured as the applied displacement over the initial height of the RVE, as shown in Eqs. 3 and 4 [26].

$$\sigma_z = \frac{R_z}{A} \tag{3}$$

$$\varepsilon_z = \frac{\delta_z}{L} \tag{4}$$

where $R_z$ is the sum of the reaction forces, $A$ is the cross-sectional area of the bottom surface, $\delta_z$ is the prescribed displacement, and $L$ is the height of the RVE.

The stress-strain curve was then drawn from Eqs. 3 and 4, the compressive strength was obtained as the maximum stress in the stress-strain curve.

## 3. RESULTS AND DISCUSSIONS

### 3.1 Proposed model prediction efficiency

The volume fraction of the three phases was computed for a given OPC, which contains 62.2% of $C_3S$, 18.3% of $C_2S$, 5.6% of $C_3A$ and 9.8% of $C_4AF$, and the physical properties of Blaine (311 $m^2/kg$), water to cement ratio (0.4) and temperature (20 °C), as described in section 2.1. Table 1 summarises the volume fraction of phases corresponding to 28 days of hydration age. Subsequently, the numerical analysis was performed on the 3D RVE, and the stress-strain curve was drawn, as shown in Fig. 4. However, the computational time was immensely high, as reported in past studies [1]. Therefore, 3D RVE was transformed into an equivalent two-dimensional (2D) planner element, as depicted in Fig. 5, because it has been proven that such a transformation exhibits a negligible effect on the homogenised mechanical behaviour with less computational cost compared to a full 3D RVE [13].

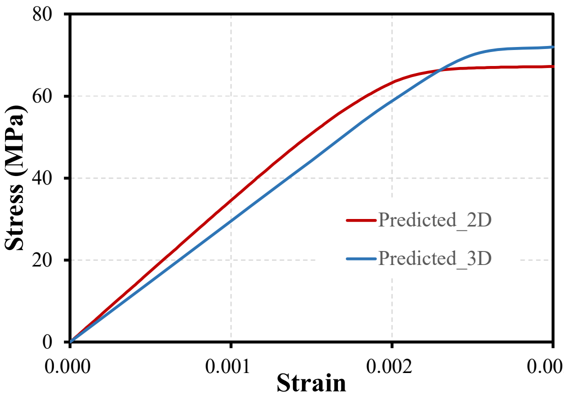

Fig.4 Comparison of stress-strain response in 3D and 2D RVE

Table 1 Volume fraction of phases at 28 days of hydration age

| Phases | Volume fraction |
|---|---|
| Hydrated phase | 0.782 |
| Unhydrated phase | 0.082 |
| Pore phase | 0.135 |

Furthermore, the variation of von mises stress of the 2D RVE at the compressed displacement of 0.3 μm is shown in Fig. 6. It could be visualised that the von-mises stress is relatively high between the pores and parallel to the applied displacement load, which represents the propagation of cracking and vertical failure planes. A similar was observed in the experimental study in [27].

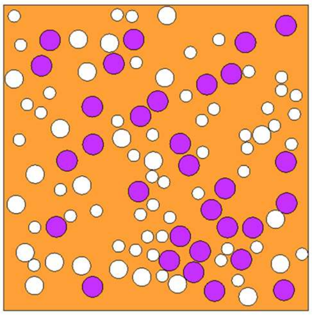

Fig.5 2D RVE of cement paste (orange: hydrated phase, pink: unhydrated particles, and brown: pores)

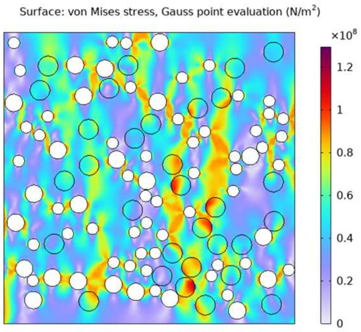

Fig.6 Variation of Von mises stresses at 0.3 μm displacement.

### 3.2 Compressive strength prediction and verification

The predicted compressive strength in 3D and 2D for the same input data are compared in Fig. 4. It can be seen that the stress-strain curve in 2D reasonably matched with 3D results, and there is no significant deviation in the predicted compressive strength. Hence, hereafter, reported mechanical properties such as compressive strength and young's modulus in this study correspond to 2D RVE if not mentioned otherwise.

The compressive strength of cement paste was predicted up to 100 days of hydration age. The input parameters, such as material properties, boundary conditions etc., for the proposed model were chosen from the experimental study by Maruyama et al. [28]. The predicted results were compared with the experimental results and the existing analytical model [16], as depicted in Fig. 7. It should be noted that the size of the RVE (100 μm) ensured the size compatibility of the actual specimen size (100 mm in height and 50 mm in diameter) used for the compressive strength test. It can be seen that the proposed model reasonably reproduces the strength of the w/c ratio of 0.4 as a function of hydration time. However, the proposed model slightly underestimates the strength gain of cement paste, especially in the early stage (< 28 days). This may be attributed to the rapid hydration of cement and the semi-solid state of the cement paste, whose material behaviour has not been accurately captured by the concrete material model, which is mainly applicable to solid-like material.

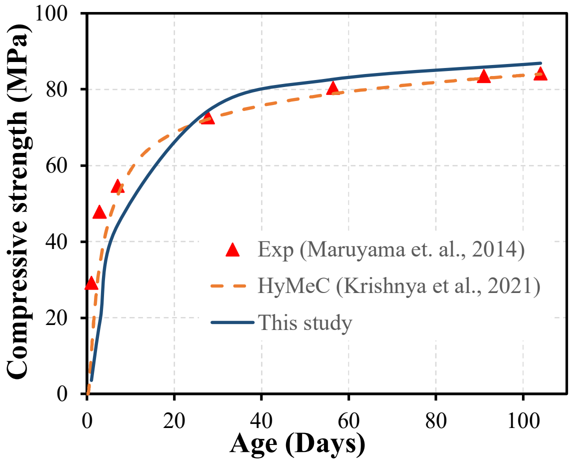

Fig.7 Compressive strength development with hydration time

### 3.2 Young's modulus prediction and verification

Averaged modulus of elasticity was calculated from the initial slope of the computed stress-strain curve. Moreover, the predicted modulus of elasticity was compared with experimental results of cement paste corresponding to a w/c ratio of 0.4, as shown in Fig. 8. It can be seen that the predicted results agree well with the experimental results, such as ultrasonic tests and compression loading tests found in [28].

It should also be noted that the chosen size of RVE is pretty much enough to prevent the size compatibility issues in predicting the young's modulus of the paste, as several studies reported that the minimum size of RVE could be 50 µm during the entire hydration period for any size of experimental specimen [29]. Therefore, it is pretty reasonable to use results from different experimental setups with a specimen size of 40 × 40 × 160 mm for the ultrasonic test for the validation. Moreover, the evolution of young's modulus follows a similar tendency of compressive strength gain over the hydration period. In addition, young's modulus initially increases exponentially and then reaches a steady state with decreasing increase rate, and this similar tendency was reported in the literature [30].

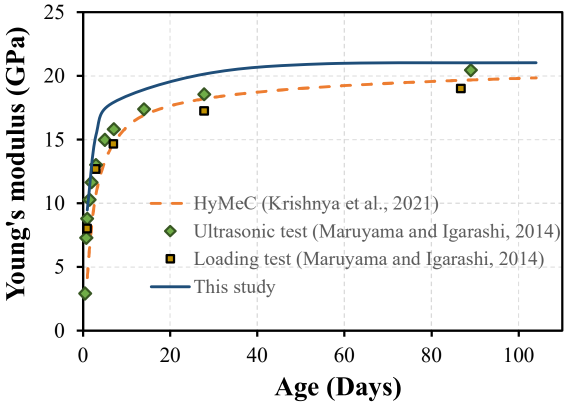

Fig.8 Young's modulus with hydration time

## 4. CONCLUSIONS

In this study, an integrated multi-stage model has been proposed to predict the compressive strength and young's modulus of hydrated OPC cement paste from the hydration process and microstructure development with the function of time. In the initial stage (stage 0), the volume fraction of hydration products were predicted by giving special consideration to the formation of LD and HD C-S-H. Afterwards, a representative volume element (RVE) for cement paste was developed in MATLAB based on the results from the previous stage (Stage 0). Finally, in stage 2, the developed geometry of RVE was transferred to COMSOL Multiphysics via the LiveLink tool to carry out the numerical analysis. Based on the results, the proposed integrated model reasonably predicted the compressive strength and young's modulus of cement paste with time and these values are well matched with the experimental results for the w/c ratio of 0.4. However, the proposed model needs to be verified with different w/c ratios (0.3,0.5,0.6 etc.) to generalise the model. In addition, since the integrated proposed model showed its robustness in predicting the mechanical properties of the cement paste, the modelling approach can be extended to predict the mechanical properties of mortar and concrete. Moreover, the proposed model can predict the mechanical properties of cementitious materials (e.g., fly ash, silica fume, etc.) with appropriate modifications in the input parameters for the volumetric prediction of hydration products.

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