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Multiscale Analysis of Cement Composites

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This paper is devoted to multiscale modeling of cement composites. The need for such an approach is due to the heterogeneous complex internal structure of the composite. A multiscale model of the composite was built and the results of computer simulations for the adopted parameters of the microstructure of the composite were presented, enabling a more detailed analysis of its mechanical and structural properties.

Keywords: multiscale modeling, analysis of heterogeneity of cement composite structure, computer simulations.

1. Introduction

Cement composites, especially concrete, play a key role in construction, being the most frequently used construction material. Their complex structure, resulting from the interaction of component materials such as cement, aggregate, water and chemical additives, and increasingly, various pozzolanic materials, makes predicting their mechanical properties and durability in different operating conditions a challenge for engineers and scientists because even the first scratches or even cracks may occur in the early phase during the hardening of concrete [1]. Traditional macroscopic models, although practical, are often not accurate enough to account for the complex microstructure of cementitious composites, leading to discrepancies between experimental and numerical results. Precise prediction of concrete properties depends mainly on understanding the behavior of hydrated cement paste (HCP). Therefore, computer simulation and prediction of the mechanical properties of HCPs are the main topics of numerical research on cementitious matrix materials. It is generally accepted that the microscale is crucial to the study of HCP because at this level, individual compo-

nents such as calcium silicate hydrate (CSH), calcium hydroxide (CH), cement grains, and various types of pores, which are described in Table 1, can be distinguished. Mechanical models that use microstructure information as input are often used to better understand the relationship between microstructure and macroscopic mechanical properties. Such models are, therefore, considered to be microstructure-based. The analysis of a multiscale model [2] of concrete based on the microstructure of samples was used in the work [3], enabling the prediction of a highly dynamic increase in concrete strength. The authors, adopting the approach of large-scale concrete modeling [4], obtained a more precise determination of the appearance of cracks than macroscopic tests show. In the second approach, the authors use the multiscale thermoelastic analysis of a concrete surface heated by solar energy. In this way, they carry out a sensitivity analysis of the internal moisture content of concrete to assess the risk of microcracks. However, in the following example, the static-strength analysis of a segmented ring of a concrete sleeve shows that multiscale modeling enables a more reliable assessment of the occurrence of displacements and crack openings in this area than macroscopic models. Chaudhuri [5] used the concurrent multiscale method to investigate the nonlinear response of concrete to cracking. In the mesoscopic description, around the concrete cracking zone, the linear elastic behavior of concrete was considered for aggregate.

In contrast, the cohesive cracking method was used for the cement matrix. The area distant from the fracture zone was described on a macroscale. It has been shown that the interface model used has a significant impact on the final results. The extended element-free Galerkin method was used for modeling. A multiscale approach to fiber concrete to determine the cohesive zone for the composite-concrete interface was proposed by Palmieri *et al.* [6], adopting a micromechanical model. This model focused on the description of the cement composite itself and the description of continuous damage for the matrix-aggregate zone and for the cement matrix.

This work is devoted to the multiscale analysis of cement composite. Because the composite is a heterogeneous material structure, multiscale modeling was used. The key issue was to build a representative unit cell that contained representative inhomogeneities at the micro level. The VCCTL (Virtual Cement and Concrete Testing Laboratory) software was selected to simulate the development of the cement microstructure. The multiscale micro-macro problem was solved using a homogenization approach. Computer implementation of the established approach and analysis of the adopted terms are discussed. The application of the developed methodology is presented in a numerical example. This work is the first stage of a broader scope of research, the aim of which is experimental research [7, 8], and multiscale modeling of cement composites with graphene nanoinclusions [9].

2. Multiscale modeling of the composite cement

The cement composite possesses a non-uniform structure, requiring a multiscale approach for its characterization, as shown in Fig. 1. The mesostructure of concrete (spanning sizes from 10^{-3} to 10^{-1} m) is described as a three-phase system comprising large aggregate grains embedded in a mortar matrix, with a transition zone connecting these phases. Within the mortar matrix (ranging from 10^{-4} to 10^{-3} m), the structure is subdivided into fine sand, a transition zone, and a cement matrix that serves as the binding agent. At the level of the cement matrix (10^{-6} to 10^{-4} m), unhydrated clinker particles are dispersed within a matrix of various hydration products. Table 1 provides detailed data on the composition of cementitious materials.

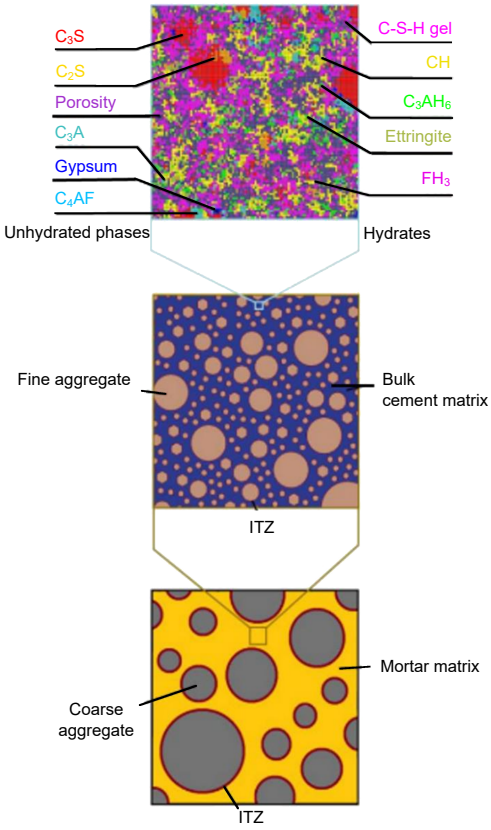
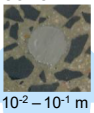
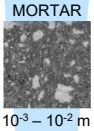
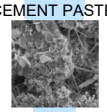
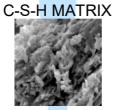
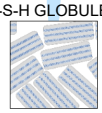


Fig. 1. Individual components and hydration products in cement composites, source: [4]. Adopted notations: C_3S – tricalcium silicate (alite), C_2S – dicalcium silicate (belite), C_3A – cubic tricalcium aluminate, C_4AF – tetracalcium aluminoferrite (brownmillerite), CH – calcium hydroxide of portlandite, $CaSO_4 \cdot 2H_2O$ – calcium sulfate dihydrate (gypsum), AFt – $Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$ (ettringite), C_3AH_6 – hydrogarnet, FH_3 – trihydrogen fluoride, C-S-H – calcium silicate hydrate, ITZ – interphase transition zones.

TABLE 1. Structure and microstructure of concrete, considering porosity and solid phases [10]. Adopted notations: AFt – $Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$ (ettringite), AFm – $C_3A \cdot CaSO_4 \cdot 12H_2O$ (monosulfate).

POROSITY	SOLID PHASES	FEATURE
	Mortar, aggregate and reinforcing steel bars	CONCRETE  $10^{-2} – 10^{-1}$ m
Entrained air	Cement paste with aggregate and ITZ	MORTAR  $10^{-3} – 10^{-2}$ m
Large capillary pores	C-S-H matrix, UNHP and CH crystals	CEMENT PASTE  $10^{-6} – 10^{-3}$ m
Large gel pores (small capillaries), medium capillaries, large capillary pores	LD C-S-H, HD C-S-H, AFt and AFm	C-S-H MATRIX  $10^{-9} – 10^{-6}$ m
Intraglobular pores, small gel pores	C-S-H	C-S-H GLOBULES  $10^{-10} – 10^{-9}$ m

Hardened cement composites containing Portland cement are characterized by a complex, porous microstructure that exhibits heterogeneity in composition and structure across multiple scales. This intricate, multiscale framework, essential for understanding material properties and performance at the macro level, comprises hydration products, unreacted clinker particles, porous regions, and aggregate grains.

- Calcium silicate hydrate (C-S-H) is a primary component of hydrated Portland cement, comprising 50% and 70% of its structure. This phase is crucial for forming strong bonds between the grout ingredients. The properties of hardened cement paste are mainly unaffected by the specific characteristics or spatial distribution of C-S-H.
- Calcium hydroxide (CH), or portlandite, is a byproduct of the hydration process alongside C-S-H. It forms a crystalline structure with plate-like shapes, typically a few microns in size and over one μm wide. CH occupies the porous regions of the cement matrix and accounts for up to 10% of the total volume of the hydrated cement paste.

- Ettringite (AFt) and monosulfate (AFm) are hydration products resulting from the reaction of aluminates and ferrites in the cement. While these compounds form in the hardened cement paste, they have a limited influence on its macroscale properties. Ettringite is characterized by its elongated needle-like crystals, ranging from approximately 250 nm to several dozen microns.
- Porosity in a typical hardened cement paste makes up about 20% of the total composite volume. It exists across a wide range of scales, from less than 1 nm to 1 mm, forming the complex and interconnected three-dimensional pore network.
- Unhydrated cement particles (UNHP) are remnants of cement clinker, usually constituting around 5% of the total volume of the hydrated cement paste. Their presence is influenced by factors such as the water-to-cement ratio.

Criteria derived from atomistic simulations evaluate the mechanical properties of individual phases in cementitious materials. Techniques such as ab initio methods, Monte Carlo simulations and molecular dynamics allow these criteria to be used in a variety of applications, enabling precise and comprehensive analysis of cement-based systems.

The initial phase of multiscale modeling for concrete composites involves nanoscale simulations at the molecular level. The elastic properties obtained at this stage serve as input for the subsequent microscale analysis, which examines a cement paste composed of the C-S-H phase, portlandite, ettringite, air voids, and unhydrated cement particles. This iterative approach continues through the scales until the mechanical properties of the representative volume element at the macroscale, including cement paste and aggregate, are determined.

The molecular model of C-S-H depicted in Fig. 2 includes the following components:

- Green and black spheres symbolize oxygen and hydrogen atoms within water molecules,

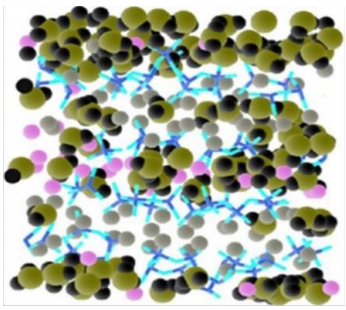


Fig. 2. C-S-H molecular model [11].

- Pink and gray spheres denote calcium ions located in the interlayer and intralayer positions, respectively,
- Blue and violet rods represent silicon and oxygen atoms forming the tetrahedral silica structure.

3. Simulation of microstructure development in cement paste

Due to its advantages in modeling the development of the microstructure of cement slurry made of Portland cement (Fig. 3) and the possibility of including the results of the cement hydration process from isothermal calorimetry, the VCCTL software was used to simulate the development of cement microstructure. The parameters of cement 140 used for analysis in the VCCTL software are shown in Fig. 3.

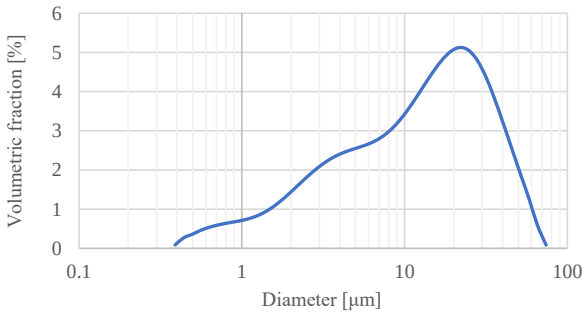


Fig. 3. Cement 140 from the VCCTL program accepted for analysis.

For cement 140 from the VCCTL database (Fig. 4), hydration was carried out assuming a cube with dimensions of $100 \times 100 \times 100$ microns.

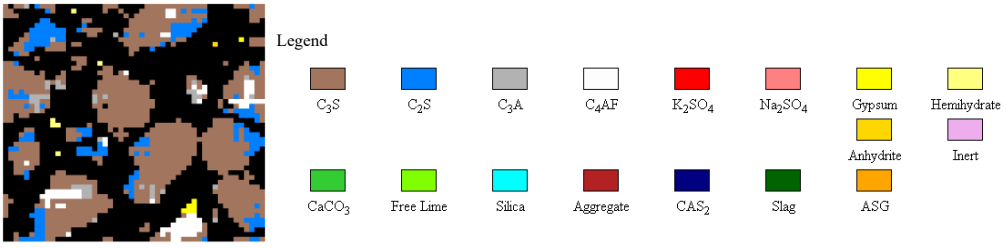


Fig. 4. Example of a section of the cement microstructure from the VCCTL database with a legend. Adopted notation: C_3S, C_2S, C_3A, C_4AF – explanations in Fig. 1, K_2SO_4 – potassium sulfate (arcanite), Na_2SO_4 – sodium sulfate (thenardite), $CaCO_3$ – calcium carbonate, CAS_2 – calcium aluminosilicate glass (in fly ash), ASG – slag grain, Hemihydrate – semi-aqueous gypsum, Anhydrite – calcium sulfate, Inert – inert materials.

Table 2 presents the statistics of the porosity phase as well as C_3S, C_2S, C_3A, C_4AF, K_2SO_4, Na_2SO_4 adopted for further analysis.

TABLE 2. Cement phase statistics.

Phase	Pixel volume	Pixel surface area	Fraction volume	Fraction surface area	Mass fraction
Porosity	74473	0	0	0	0
C_3S	32919	17672	0.68676	0.68509	0.67948
C_2S	7666	4129	0.15993	0.16007	0.16168
C_3A	3916	2070	0.0817	0.08025	0.0756
C_4AF	3433	1924	0.07162	0.07459	0.08234
K_2SO_4	0	0	0	0	0
Na_2SO_4	0	0	0	0	0
Total	47934	25795	0.94868	1	0.95776

The distribution of cement phases resulting from hydration after 28 days is shown in Fig. 5.

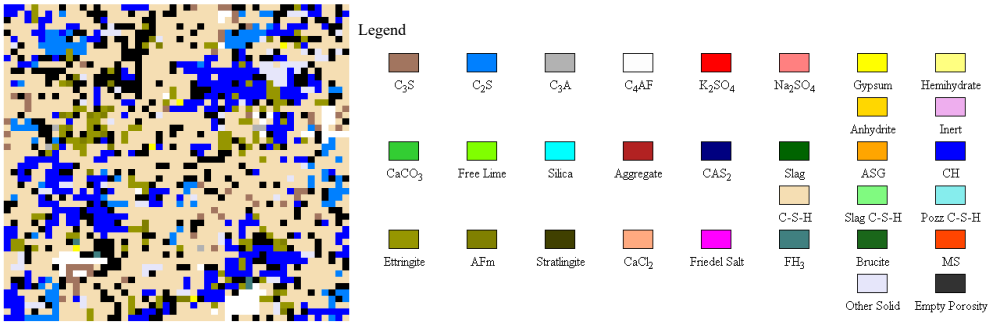


FIG. 5. Example section of cement microstructure after 28 days with legend. Notations as previously adopted.

The data used for computer simulations are presented in Tables 3 and 4. Table 4 provides the shares of individual cement phases. Less significant products (16%) have been omitted due to their negligible strength properties.

TABLE 3. Cement porosity with a total pore volume of 26.288 μm^3 and a density of 3.1 g/cm^3.

Diameter [μm]	Number	Fraction
1.000000	26252	0.998631
3.000000	36	0.001369
5.000000	0	0.000000
7.000000	0	0.000000
9.000000	0	0.000000
11.000000	0	0.000000

TABLE 4. Proportion of individual phases in cement/cement matrix.

Cement phase	Tricalcium silicate (C_3S)	Dicalcium silicate (C_2S)	Calcium hydroxide (CH)	Calcium silicate hydrate (C-S-H)	Porosity
Volume of the fraction phase in cement	0.007152	0.030200	0.163872	0.445040	0.201416
Bulk modulus	0.248817	0.990407	3.805238	6.583145	0.637753
Bulk modulus fraction	0.016595	0.066054	0.253786	0.439056	0.042534
Shear modulus	0.149860	0.590719	1.966467	3.776670	–
Shear modulus fraction	0.018591	0.073281	0.243949	0.468512	–
Young's modulus	0.374412	1.478260	5.032504	9.511191	–
Young's modulus fraction	0.018257	0.072083	0.245394	0.463784	–

Figure 6 shows the proportions of various cement phases and porosity within a $50 \times 50 \times 50$ μm element. The data and distribution of components in the $50 \times 50 \times 50$ μm sample are used to construct a numerical model in the ANSYS program.

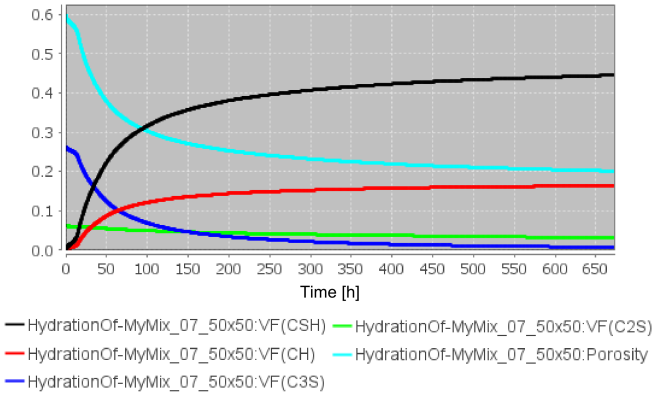


FIG. 6. Volumetric proportion of components in the sample.
Adopted notations: C_2S – dicalcium silicate, C_3S – tricalcium silicate, CH – calcium hydroxide, C-S-H – calcium silicate hydrate, VF – volumetric fraction.

Data obtained from computer simulations conducted using the VCCTL program were utilized to construct the model of a cubic sample, which serves as the basis for homogenization at the microscale. In the first step, output data from the VCCTL program were transformed using an algorithm in Matlab into a script for the ANSYS APDL program, the finite element method software. As a result, a topology matrix was created with the automatic assignment of properties for the four main components of the cement matrix, including the

consideration of pores. The obtained results of the model construction are presented in Fig. 7: (a) combined model, (b) C-S-H – calcium silicate hydrate, (c) CH – calcium hydroxide, (d) C_3S – tricalcium silicate, (e) C_2S – dicalcium silicate. The numerical analysis performed using the ANSYS program solved a three-dimensional boundary value problem for a $50 \times 50 \times 50$ μm cube using SOLID185 finite elements. Compression and shear simulations were conducted on the constructed model, consisting of individual pixels in the form of cubic fi-

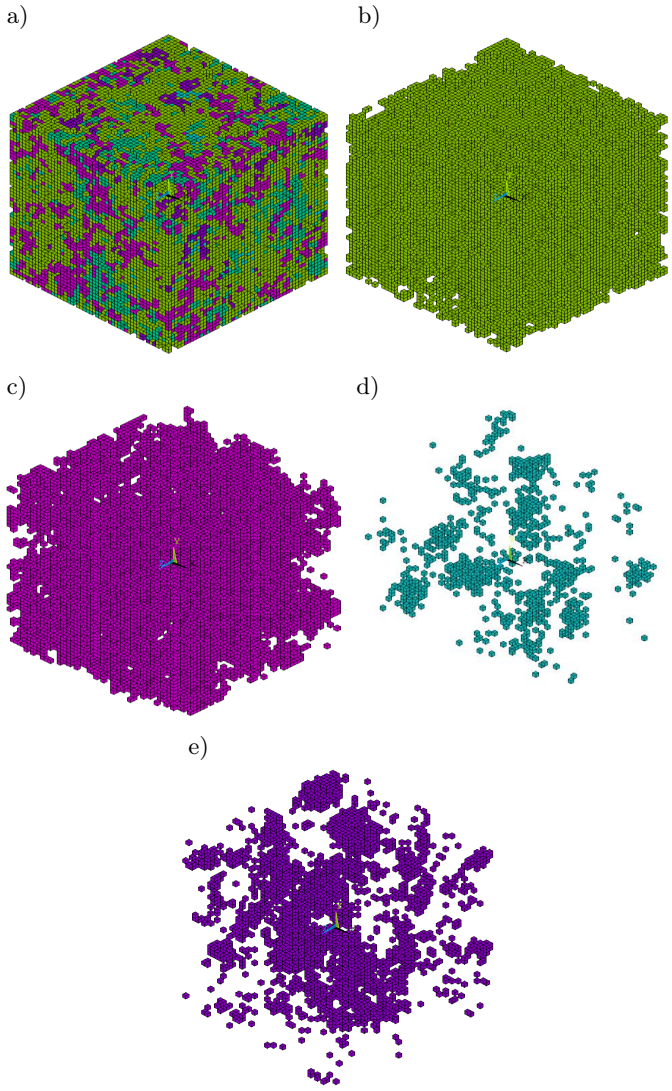


Fig. 7. Simulation results modeled using the ANSYS program: a) initial model – combined, b) C-S-H – calcium silicate hydrate, c) CH – calcium hydroxide, d) C_3S – tricalcium silicate, e) C_2S – dicalcium silicate.

nite elements with individually assigned mechanical properties. This yielded the global mechanical properties of the studied element (Table 5), which can be used to develop a "higher-order" structural model based on the described sample.

Table 5. Mechanical and material properties of cement paste.

Cement paste elastic moduli	
Bulk modulus	14.993862 GPa
Shear modulus	8.060981 GPa
Young's modulus	20.507815 GPa
Poisson's ratio	0.272042

This model can also be utilized to analyze the impact of additives, such as various types of fibers, and to incorporate mechanical models of friction force transfer, similar to the fiber-reinforced cement paste with steel or polypropylene fibers. Furthermore, this model can serve for detailed analysis of the fiber interface, i.e., the so-called ITZ (interfacial transition zone).

4. Final remarks

The conducted research yields several important conclusions:

- The analysis utilizing a virtual cement matrix constructed using the VCCTL software allows the incorporation of the actual structure of the cement paste into the analysis.
- The selection of properties for a representative element with a 50 μm edge can facilitate using VCCTL data for analyzing the microstructure of a cement composite with nano-inclusions.
- The simulation confirmed the global mechanical properties determined through the VCCTL.
- Data obtained from numerical analysis can be directly applied with homogenization for higher scales.
- The adopted model can be extended to nonlinear analysis and homogenization methods for higher scales (mesoscale, macroscale).

The obtained results provide a solid foundation for formulating directions for further research, incorporating nanofibers into the basic model. Currently, nanoscale admixtures, such as carbon nanotubes (CNTs) and graphene nanoplatelets (GPs), are increasingly being used to enhance the mechanical properties of cementitious composites. Aglan *et al.* [12] and Shebl *et al.* [13] have confirmed the effectiveness of cement paste reinforcement. By creating numerous nucleation sites in the suspension, nanoinclusions lead to densification of the structure and

improvement in the material's mechanical properties, increasing its resistance to crack formation and propagation.

Multiscale modeling of concrete composites with nanoparticles enables an assessment of the effects of added nanoparticles across various scales and allows predictions regarding the outcomes of the process [14]. A thorough understanding of the impact of nanoinclusions on the interparticle relationships within the composite will facilitate purposeful control of their application, in terms of type and quantity, to enhance the properties of cementitious or concrete composites. Multiscale modeling offers the potential to design new composite materials with desired properties.

Preliminary studies on multiscale modeling of cement composites with nanoinclusions have already been undertaken. The results of these studies are presented in [9]. Experimental research on cement composites with nano-inclusions has also been conducted.

Using nanoindentation, the effects of nano admixtures, such as XG graphene flakes, graphene coated with monosilane (GN+Si), oxidized graphene coated with monosilane (GN-OX+Si), and carbon nanotubes (CNTs) in amounts of 0.05% and 0.10% by weight, on the time-dependent properties of cement paste nanocomposites were investigated. Improvements in mechanical properties were observed, and the results of these studies are presented in [7] and [8].

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