UDC 691.542

MODELLING OF THE VOXEL-BASED MICROSTRUCTURE OF THE CEMENT PASTE

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Abstract

Microstructural modelling of the cement paste is a hard task due to its extremely complex and heterogeneous structure. At the same time, accurate predicting of early age properties of the cement paste is directly dependent on how close the microstructural model is to the real structure.

There are two approaches in modelling microstructure development of the cement paste: vector and discrete, both generate a structure of the cement paste with randomly distributed phases. The voxel-based discrete approach is best suited for generating and processing random morphology of cement in the cement paste, in contrast to the vector approach limited by its regular geometric shape. A major challenge the voxel-based discrete approach faces is to reconstruct the real random distribution of cement in the fresh cement paste.

The paper presents a voxel-based microstructural model of the cement paste based on the Gaussian blur method to generate the initial spatial distribution of phases in it.

Keywords: cement paste, microstructural model, voxel, Gaussian blur, pore size distribution.

МОДЕЛИРОВАНИЕ МИКРОСТРУКТУРЫ ЦЕМЕНТНОГО КАМНЯ НА ОСНОВЕ ВОКСЕЛЬНОЙ МОДЕЛИ

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Реферат

Моделирование микроструктуры цементного камня является достаточно сложной задачей из-за его чрезвычайно сложной и неоднородной структуры. В то же время точность прогнозирования свойств цементного камня в раннем возрасте находится в прямой зависимости от того, насколько модель микроструктуры близка к реальной структуре.

Существует два подхода к моделированию развития микроструктуры цементного теста: векторный и дискретный, оба позволяют генерировать структуру цементного камня, в которой фазы распределены случайным образом. Дискретный подход на основе воксельной модели лучше всего подходит для генерации и обработки морфологии цемента случайной формы в структуре цементного камня, в отличие от векторного подхода, ограниченного правильной геометрической формой. Основная задача, решаемая в рамках дискретного воксельного подхода, заключается в воссоздании начального распределения фазы цемента в структуре цементного камня максимально приближенного к реальному распределению.

В статье представлена воксельная модель микроструктуры цементного камня, основанная на методе размытия по Гауссу для создания начального пространственного распределения фаз в его структуре.

Ключевые слова: цементный камень, микроструктурная модель, воксель, размытие по Гауссу, распределение пор по размерам.

Introduction

The primary phase of cement-based composites is a cement paste, which is normally regarded as a matrix. It holds dispersed phases of aggregates and shares a load with them. Being a matrix, the cement paste has a significant impact on basic properties of cement-based composites. That is why, researching the microstructure of the cement paste is an important objective in concrete technology.

The cement paste is a porous solid with an extremely complex and heterogeneous structure formed during hydration process. It causes the fact, that microstructural modelling can be a hard task, depending on what properties of the cement paste need to be evaluated.

Accurate predicting of early age properties of the cement paste requires a microstructural model that is as close as possible to its real structure. To achieve this goal, two approaches are currently used:

1) A vector approach [1, 2] based on the random placement of cement particles in a representative volume. Typically, cement particles have a geometrically regular shape in the form of a sphere. The number of spheres and their radii are determined by the particle size distribution of cement. During hydration, the volume of each sphere decreases, while a layer of hydrates around it increases.

2) A discrete approach [3] based on splitting up of a representative volume into cubic cells with a certain edge length, called voxels. Each voxel represents a specific phase of the cement paste, for instance, at the initial time it can only be marked as water or cement. During hydration, a voxel either updates its phase state, and is marked as hydrated or remains the same.

Both of these approaches have the same feature. They are computationally complexity. This is a price to pay for getting closer to a real microstructure of the cement paste.

The discrete approach looks to be a more preferable due to easy spatial distribution of phases in an arbitrary shape and easy of computations. Basically, it suffers from two limitations:

1) The model resolution. Since, it is limited by the voxel size, some fine cement particles cannot be taken into consideration.

2) The initial spatial distribution of cement. It is extremely difficult to reconstruct the real random distribution of cement in the fresh cement paste.

The main complaint about the vector approach are as follows:

1) The regular (spherical) shape of cement particles.

2) Numerical techniques used to evaluate the pore size distribution are extremely computationally expensive. In consequence of this, even vector models use discrete techniques to approximate the pore size distribution [2].

3) Limited details of the microstructure. Normally, such a microstructure of the cement paste includes the only aggregated phases: unhydrated cement, hydration products, and pores.

The serious problem of the vector approach is the particle size distribution of cement, which is used to generate the initial structure of the cement paste. Commonly, the Rosin–Rammler distribution correlated with cement fineness is used. Since, the cement paste is made by mechanical mixing cement and water, one must wonder whether we consider that the particle size distribution of cement in the fresh cement paste is the same as before mixing.

The paper presents the microstructural model of the cement paste based on the voxel discrete approach. It is aimed at obtaining pore characteristics of the cement paste, which can be used in modelling the moisture transport in porous media. Meanwhile, the model has limitations and simplifications for reducing its computational complexity in numerical simulations, enabling to be incorporated into complex models to predict mechanical properties of concrete.

https://doi.org/10.36773/1818-1112-2024-133-1-14-18

Microstructural model

Model limitations and simplifications

1) The microstructure of the cement paste consists of four aggregated phases: unhydrated cement, hydrated products, water, and void space occupied with air.

2) The voxels marked as unhydrated cement and hydrated products represent solid phase of the cement paste.

3) The interaction of adjacent voxels occurs along the normal direction.

4) Porosity is a space between solid phase i.e., occupied with voxels marked as water and air. This type of porosity is characterized mainly by capillary pores in which a concave meniscus occurs between water and solid.

5) Stoichiometry of hydration reactions is used to find the volume fractions of the cement paste phases during hydration.

6) The evolution of the microstructure is considered in sealed conditions i. e., without the loss of water to the environment.

3D Microstructure generation

The discrete model of the microstructure of the cement paste is a 3D array[1] consisting of a set of voxels, each of which represents one of the four phases at an arbitrary time step. By voxel in this context is meant a numeric value in the 3D array associated with a certain phase.

At the initial time (t=0), only voxels representing unhydrated cement and water are randomly distributed in the 3D array. There are several ways to spatial distribute voxels in the 3D array: from the simplest random shuffle to complex reconstruction with the help of correlation functions that convert 2D microstructure images under an electron microscope to 3D. The last way is used in the well-known discrete model CEMHYD3D [3].

However, if there is no need to separate cement into clinkers phases, the Gaussian blur method can be directly applied to generate the initial distribution of cement and water in the cement paste. It is often applied to generate the porous media where parameters of the microstructure need to be controlled. The flowchart of a generation process using the Gaussian blur method is presented in Figure 1 [4].

The standard deviation of the Gaussian distribution normally controls the morphology of a solid in the porous media. In the case of the cement paste, the standard deviation can be correlated with the fineness of cement, which will ensure control of the size and number of cement particles in the generated array.

Model resolution

A discrete model of a random system has three sources of error: statistical fluctuation, finite size effect, and resolution [5]. Statistical fluctuation error occurs due to the modelled system is random, while finite size effect error occurs due to the model covers a very small part of the real object, which in the theory of composite materials is called a representative elementary volume (abbreviated REV).

Statistical fluctuation error is unavoidable, while finite size effect error directly depends on the REV size. In most of conducted researches, REV of the cement paste was generally considered no more 100×100×100 μm in size.

The 3D image representing REV of the cement paste is dimensionless (independent of physical sizes of REV) and is characterized by a resolution expressed by the number of voxels it contains.

Resolution error occurs when the random system contains elements that are smaller than the voxel size. Finite size error and statistical fluctuation are not important for a cement hydration model with a resolution of 1 μm^3/voxel and REV size of 100×100×100 μm. A further increase in the REV size does not produce to significant accuracy in the hydration model prediction [5].

The following relationship can be given to determine the number of voxels (n_{vox}), which helps to convert a dimensionless 3D image to real sizes of REV and vice versa:

$$n_{vox} = \frac{V_{RVE}}{res} \quad (1)$$

where V_{RVE} – is the physical volume of RVE, μm^3;

res – is the model resolution, μm^3/voxel.

Consequently, a 3D image should contain of 1000000 voxels for REV of 100×100×100 μm in size with a resolution of 1 μm^3/voxel to avoid expected errors.

The phase volume contained in each voxel is computed as $\frac{1}{n_{vox}}$ μm^3/μm^3.

Mechanism of microstructure initialization and evolution

The basic steps of microstructure modelling are listed below:

1) The number of voxels in REV (n_{vox}) is computed. The parameters of V_{RVE} and res are adjusted to provide a cubic volume with a domain size of $\sqrt[3]{n_{vox}}$ (in voxels).

2) The initial volume fraction of cement ($V_{c,0}$) and water ($V_{w,0}$) is computed by the following equation:

$$V_{c,0} + V_{w,0} = V_{c,0} \cdot \left(1 + \frac{\rho_c \cdot w/c}{\rho_w}\right) = 1 \quad (2)$$

where w/c – is the water to cement ratio;

ρ_c and ρ_w – is the density of the cement and water, respectively, kg/m^3.

3) The initial microstructure of the cement paste is generated using the Gaussian blur method. The porosity threshold is taken equal to the value of $V_{w,0}$. The output is a binary 3D image represents each voxel by either a zero (for the water voxels) or one (for the cement voxels).

4) Since, the microstructural model processes the volume change of the phases, the hydration reactions are expressed in term of volumes per unit volume of the clinker phase rather than masses. For instance, the hydration reaction of C_3S presented in [7]:

$$2 \cdot C_3S + 10{,}6 \cdot H \rightarrow C_{3,4}S_2H_8 + 2{,}6 \cdot CH \quad (3)$$

can be rewritten in term of volumes as:

$$1 \cdot V_{C_3S} + 1{,}34 \cdot V_H \rightarrow 1{,}52 \cdot V_{C_{3,4}S_2H_8} + 0{,}61 \cdot V_{CH} \quad (4)$$

where V_{C_3S}, V_H, $V_{C_{3,4}S_2H_8}$, and V_{CH} – is the volume fraction of alite, water, CSH gel, and portlandite per unit volume of reacting C_3S, respectively.

5) The following algorithm of the microstructure evolution is applied at each time step (t):

5.1) Based on stoichiometric calculations, the volume fractions of clinker minerals, hydration products, and water are determined.

5.2) The volume fractions of the aggregated phases of the cement paste are determined as:

$$V_c = \sum_i V_{cl,i};$$
$$V_{hyd} = \sum_j V_{hyd,j};$$
$$V_{air} = 1 - V_c - V_{hyd} - V_w \quad (5)$$

where V_c, V_{hyd}, and V_{air} – is the volume fraction of the unhydrated cement, hydration products, and air, respectively.

5.3) The number of consumed voxels of cement and water and newly formed voxels of hydrated products and air are computed.

5.4) At first, the voxels of cement adjacent to voxels of water are randomly selected. Voxels with the most neighbours have the highest selection priority i.e., voxels with six neighbours have the highest selection priority, while voxels with one have the lowest. Each selected voxel of cement is combined with a randomly selected adjacent voxel of water.

5.5) The phase state of each pair of voxels «cement-water» is updated to hydration products.

5.6) The number of voxels of water, hydration products, and air is balanced. This means that some voxels of water are additionally updated as hydration products and air to balance the phase volume. For clarification, we can transform the equation (4) into a simpler form:

$$1 \cdot V_c + 1{,}34 \cdot V_w = 2{,}13 \cdot V_{hyd} + 0{,}21 \cdot V_{air} \quad (6)$$

It follows that after step 5.5 it is still necessary to update $(0{,}34 \cdot V_w)$ voxels of water to $(0{,}13 \cdot V_{hyd})$ voxels of hydration products and $(0{,}21 \cdot V_{air})$ voxels of air.

5.7) After, the step 5.4 cannot be completed (there are no longer cement voxels adjacent to water voxels), voxels of cement with at least one adjacent o voxels of hydration products are randomly selected. Each selected voxel of cement is combined with the nearest voxel of water found along the normal to a randomly selected adjacent voxel of hydration products.

[1] The term «image» is also often come across

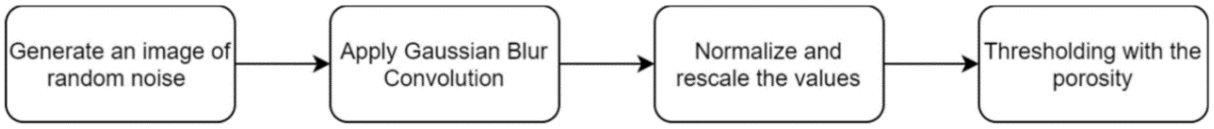

Figure 1 – Flowchart of porous media generation process

Pore size distribution

One of the obvious advantages of the discrete approach is a possibility to approximate the pore size distribution. Existing approximation ways are mainly based on the pixel-erosion method of a binary image.

However, they have two serious weaknesses:

1) The high computational complexity, superimposed on the inherent complexity of the discrete approach.

2) The resulting distribution is quite difficult to use in practice, since it is normally presented in the form of a histogram, which requires additional effort to fit a distribution function.

For this reason, the different approach is used here, including the following:

1) The distribution function is taken in advance.

2) The parameters of the distribution function are estimated based on the porosity data extracted from the microstructural model.

The unimodal Raleigh–Ritz distribution function is taken to provide the pore size distribution of the cement paste [6]:

$$\phi(r) = 1 - \exp(-B \cdot r) \quad (7)$$

where $\phi(r)$ – is the porosity fraction up to pore radius r;

B – is the pore structure parameter corresponding the peak of porosity distribution on a logarithmic scale, 1/m.

The pore structure parameter can be approximately computed by two ways.

1) Assuming that representative pore radius (r_{peak}) can be expressed as $r_{peak} = \frac{2 \cdot V_{p,peak}}{S_p} = \frac{2 \cdot \phi(r_{peak}) \cdot V_p}{S_p}$, which together with the expression (7) leads to the following relationship:

$$B = -\frac{S_p}{V_p} \cdot \frac{\ln\left(1-\phi(r_{peak})\right)}{2 \cdot \phi(r_{peak})} \quad (8)$$

2) Using the relationship presented in [6], which has been obtained by fitting a large number of data-sets which relate B as a function of $\frac{S_p}{V_p}$:

$$B = \exp\left[\left(\frac{\ln\left(\frac{S_p}{V_p}+a\right)}{b}\right)^c\right] \quad (9)$$

where $V_{p,peak}$ – is the pore volume up to pore radius r_{peak}, m^3;

S_p – is the total pore surface, m^2;

V_p – is the total pore volume, m^3;

a, b, and c – are the fitting constants.

Hence, in order to determine pore structure parameter B, it just needs values of S_p and V_p obtained from the microstructural model. Actually, these values are pretty easy to extract for any random structure using a voxel discrete model.

The value of V_p is a sum of the voxel volumes marked as water or porosity, while S_p is a sum of the voxel surfaces marked as water or porosity, and adjacent to the voxels marked as unhydrated cement or hydrated products.

Modelling results

The Portland cement paste with parameters reported in Table 1 was used for the simulation.

Table 1 – Parameters of the cement paste

Mix proportions, kg/m^3		Water to cement ratio	Density of cement, kg/m^3	Fineness of cement, m^2/kg	Mineral composition of cement (mass %)
Portland Cement	Water				
370	185	0,5	3150	345	C_3S: 54,5; C_2S: 17,3; C_3A: 8,9; C_4AF: 7,6; Gypsum: 5

Sets of hydration reactions and phase characteristics of Portland cement contained in [7] were used to carry out the stoichiometry calculations.

The Parrot and Killoh model [8] was taken for evaluate hydration kinetics of Portland cement.

The actual temperature was assumed to be 20 °C.

The model resolution of 1 μm^3/voxel was considered. A 3D image of 1000000 voxels requires significant computational resources to generate and process the microstructure. An edge size of REV was considered in the range of 30, 40 and 50 μm to reduce the computational cost.

The standard deviation of the Gaussian distribution (σ) as a function of the blobiness was computed as follows:

$$\sigma(b) = \frac{\sqrt[3]{n_{vox}}}{40 \cdot b} \quad (10)$$

where b – is the blobiness.

The blobiness is the variable that controls the morphology of cement. By reducing its value, the standard deviation increases, and smaller blobs of cement are expected in the microstructure. The value of $b = 1$ was used in the simulation.

The pore structure parameter was approximated by the relationship (8), where $\phi(r_{peak}) = 0{,}5$.

The modeling results are presented in Figures 2–7.

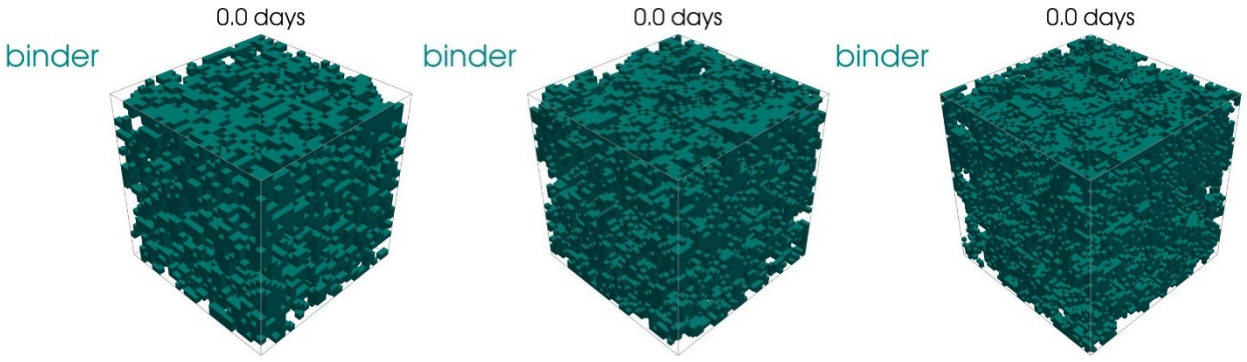

Figure 2 – The comparison of the initial spatial distribution of cement in the cement paste
(Left: REV size of 30 μm; Middle: REV size of 40 μm; Right: REV size of 50 μm)

https://doi.org/10.36773/1818-1112-2024-133-1-14-18

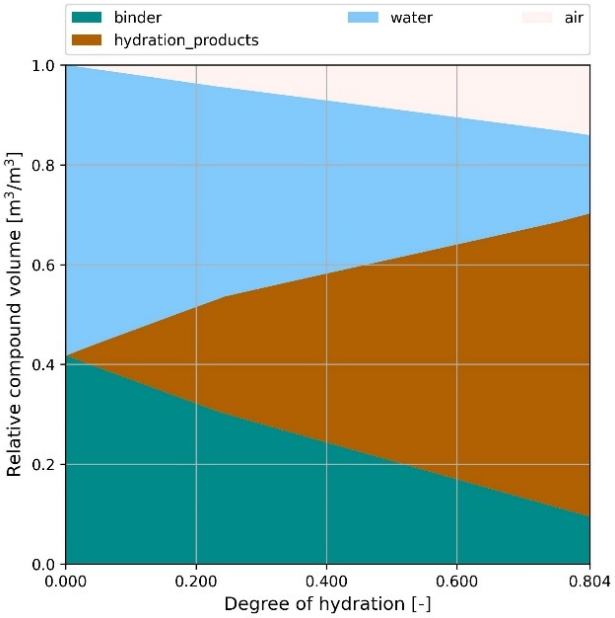

Figure 3 – The predicted volumetric composition of the cement paste over 100 days

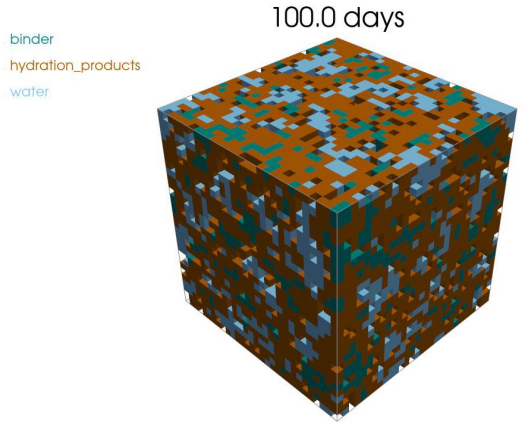

Figure 4 – The microstructure evolution of the cement paste with the REV size of 30 μm
(Empty cells refer to air voxels)

Figure 5 – The microstructure evolution of the cement paste with the REV size of 40 μm
(Empty cells refer to air voxels)

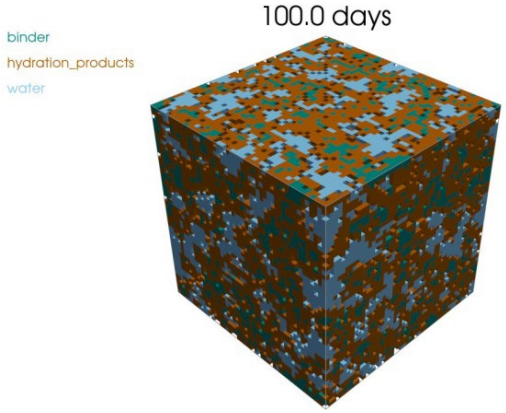

Figure 6 – The microstructure evolution of the cement paste with the REV size of 50 μm
(Empty cells refer to air voxels)

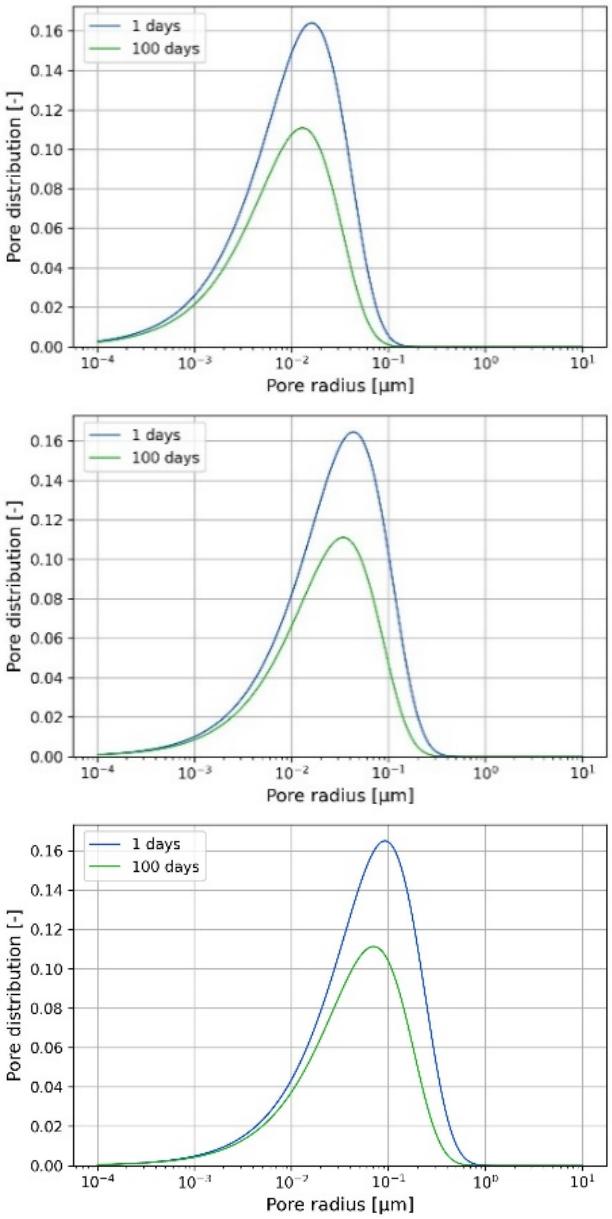

Figure 7 – The predicted size distribution of capillary pores of the cement paste
(Left: REV size of 30 μm; Middle: REV size of 40 μm; Right: REV size of 50 μm)

Conclusions

1. The presented discrete model of microstructure of the cement paste is computationally complex, but provides the evolution of random structures generated by the Gaussian blur method. Such structures are as close as possible to real structures, which in practice should lead to more accurate estimates of their parameters, for instance, porosity parameters.

2. The crucial parameter of the model is a resolution expressed by the number of voxels, which determines amount of computation and its accuracy.

3. To control the cement morphology in the generated structure depending on its fineness, it is necessary to pre-establish a relationship between the fineness of cement and the standard deviation of Gaussian distribution.

4. The presented approximation of the pore size distribution may appear simplistic, but its advantage is that the only two easily extracted porosity parameters are needed to produce acceptable results at a lower computational cost.

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Material received 30/11/2023, approved 24/01/2024, accepted for publication 24/01/2024

https://doi.org/10.36773/1818-1112-2024-133-1-14-18