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MORPHOLOGICAL MODELING OF CEMENT BASED MATERIALS AND HYDRATION PROCESS

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Abstract

In this work we present a new morphological model for heterogeneous media that is well suited for cement based materials. The main application addressed here is a model for the evolution cement paste with respect to the hydration degree, containing only anhydrous cement, water, and hydrated products. Secondly, those morphologies are projected on FE trusses, which allow us to predict the evolution of any chosen material characteristic (elastic properties and diffusivity) with the evolution of the hydration degree.

1. INTRODUCTION

Cement based materials modeling has always been a great challenge. For nuclear or construction industry, a reliable predictive approach is still an undergoing challenge. A new approach based on random field's thresholding is presented here, in order to generate microstructures of heterogeneous materials. This mutli-scale morphological modeling framework is particularly adapted to cement based materials since it is capable to represent easily a material with multiple concentrical phases.

Following the work of R.J. Adler [2], the idea is to yield phases from random fields' excursion sets. The advantages of such a method are various. First, contrary to most of the numerical hydration models, we will not be dealing with spheres but random shapes of inclusions. Secondly, the creation of such morphologies is much lighter than average, since we will need only three parameters (two for the random field, and one for the threshold) for an entire morphology (instead of four parameters for each inclusion).

Eventually, one can generate various random fields, with chosen characteristics. By thresholding them, various morphology can be created, controlling parameters such as volume fraction or specific surface area, and thus create a heterogeneous model for any material.

The focus is made here on anhydrous cement within a "matrix" of water. By changing the thresholds, a *n*-concentrical-phases material can be created. A simple analytical hydration model [10] has been chosen, considering only three phases: anhydrous cement, hydration products (including gel water) and free water. The link that has been made between these

volume fractions and the thresholds allows us to make evolve the initial morphology, and thus model the hydration process for a cement paste.

Once the morphologies have been created, at any hydration degree, they are projected on a FE mesh with embedded discontinuities [3]. It gives us the possibility to predict properties such as the Young modulus, the Poisson's ratio, the effective diffusivity, or any transport properties, using a sequenced multi-scale approach.

2. MORPHOLOGY CREATION

In this part we explain shortly how the morphologies are created. With first the generation of random fields with chosen characteristics, then the thresholding, and finally the link between the random fields variables and the geometric characteristics of the excursion sets.

2.1 Random fields

The first step in the framework we present here is the generation of random field realisations. We give here only the main ideas and present the main tools used for the generation process. Since the whole process relies on correlated random fields, the main issue was the numerical implementation of their generation.

First, we used the orthogonal decomposition [5], which allows, for Gaussian correlated fields, a separation between the space and random variables. In a second time, the Karhunen-Loève decomposition [6] is used to determine the space variables by solving an eigenvalues problem. Demonstration can be found in [2]. The numerical implementation relies here on a Finite Element Method. Finally, the last step is to generate a field with such space functions. For the random variables, a simple random number generator can be used. However, such multi-dimensional, large size, generation processes are very demanding in memory storage. In this regard, we used here the Turning Band Method [8], which allows creating mutli-dimensional fields from several one-dimensional ones.

2.2 Excursion sets

The second step is the thresholding of those realisations. Hereafter, we explain the principle of excursion sets. We shall consider random fields as random functions:
$\gamma(x,\omega) : M \subset R^N \rightarrow R^k$, where M is a N dimensional Euclidian space. In the framework we present here, we are dealing with Gaussian (or Gaussian related) correlated random functions γ, which are defined in a cube of size T, with value in R. An excursion A_u of such a function is defined by the points of M where the values of γ are above a threshold u (equation (1)):

$$A_u \equiv A_u(\gamma,M) = \{ x \in M : \gamma(x) \geq u \} \quad (1)$$

An example is shown on Figure 1, for a one dimensional realisation γ, defined over M. A second example (Figure 2) shows the variety of excursion sets yielded for different values of the threshold u, from sponge (left side) to meatball like (right side), with all the intermediaries.

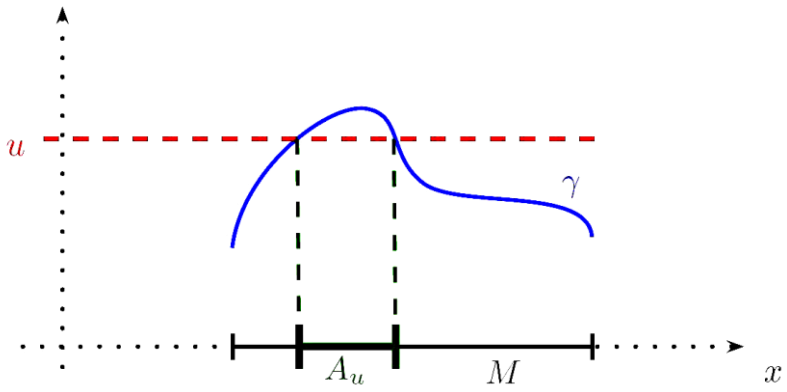


Figure 1 : Example of a one dimensional excursion set (A_u)

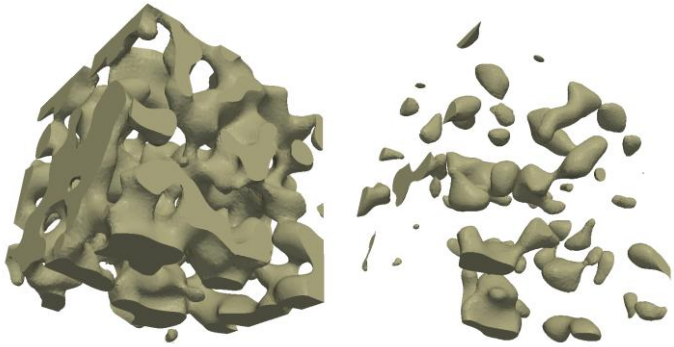


Figure 2 : Examples of a three-dimensional excursion sets

2.3 Characterization

The whole point of using this framework lies in the work of R.J. Adler [1]. He established a probabilistic link between the random fields' quantities (L_c and σ) and the threshold on one side, and the geometrical and topological quantities of the excursions sets on the other side.

To be able to describe the morphologies, one needs a measure. It has been chosen here to use the Lipschitz-Killing curvatures (denoted LKC's). In an N dimensional space, there are N+1 such curvatures. So here, for a three dimensional geometry, we have four LKC's. Three of them are geometrical, and the last is topological. They are defined by:

- L_3 (A_u) is the volume of (A_u)
- L_2 (A_u) is half the surface area of (A_u)
- L_1 (A_u) is twice the calliper diameter of (A_u)
- L_0 (A_u) is the Euler characteristic of (A_u), which is the only topological measure, that can be calculated by:

$$L_0(A_u) = \#\{\text{connected components in } A_u\} - \#\{\text{“handles” in } A_u\} + \#\{\text{“holes” in } A_u\} \qquad (2)$$

Moreover, a positive Euler characteristic reflects a disconnected morphology, and a negative one reflects a connection between the components.

The link previously mentioned, is so-called probabilistic since its only gives us expectations of the LKC's. However, since we have only three parameters as inputs

(correlation length, variance and threshold), we choose to keep only three parameters as outputs: the volume fraction V_v, the volumic surface area S and the number of aggregates N.

So in the end, we will have a link that can be expressed as following:

$$\begin{aligned} &E\{L_3\}(u,\sigma) = V_v.T^3 \\ &E\{L_2\}(u,\sigma,L_c) = \tfrac{1}{2}.S.\ T^3 \\ &E\{L_0\}(u,\sigma,L_c) = N \end{aligned} \quad (3)$$

where T is the size of the excursion. For further details about those equations, please refer to [2] for the explicit formulae, and [1] for the demonstrations.

The only issue here is that those equations are highly non-linear. So most of the time, the previous system of equation does not have solutions. Typically, so far we cannot yield excursions with more than 15% of volume fraction using Gaussian fields, and no more than 30% using chi-square distributions. It still isn't very realistic regarding concrete mixes used nowadays (it would represent cement pastes with w/c ratios higher than 0.7), but it is the best that we can do so far.

A last point worth mentioning, is that excursion sets are not necessarily defined by "all the points of M where values of γ are above a given threshold u". We can also define them by "all the points of M where values of γ are between two thresholds u and v", which will allow us to create several "concentrical" phases.

3. HYDRATION PROCESS

In this part we present an application to the cement paste hydration process, using a simple analytical model to create initial morphologies, and make them evolve.

3.1 Analytical model

The Powers and Brownyard model [10] has been implemented here, but in a simple version. Usually, this model considers five volumic fractions: anhydrous cement, hydration products, gel water, free water and chemical shrinkage. We decided to use only three for this first application, taking into account only anhydrous cement, hydration products (including gel water) and free water . We also decided not to take into account the chemical shrinkage (its volume is taken into account within the volume of water). Considering these assumptions, we also decided that the hydration degree should be able to reach one (due to the high w/c ratio, this hypothesis is realistic) at the end of the process in order not to underestimate the volume of hydration products (20% of the initial volume of anhydrous cement is included in the pores at the final stage). The simplified equations can be resumed as follows:

$$p = \frac{w/c}{w/c + \rho_w/\rho_c} \quad (4)$$

$$V_{anh} = (1-p)(1-\alpha)$$

$$V_h = 2.12(1-p)\alpha$$

$$V_w = 1 - V_{anh} - V_h$$

where p is the initial porosity, α the hydration degree, and V_{anh} , V_h , V_w respectively the volume fractions of anhydrous cement, hydration products and water.

3.2 Initial morphology

The geometrical and physical description of the anhydrous cement is simple for a classical Portland cement, without any additions. If we consider a CEM-I cement, the usual average density is $\rho_c \approx 3.1\ g.cm^{-3}$, with an average mass surface S_{mass}=4000 $cm^2.g^{-1}$. This gives us a specific surface of S_{sp}=12000 $cm^2.cm^{-3}$. Moreover, if we assume a disconnected morphology, we will set a positive Euler characteristic, and finally, we will set the size of the cube, $T = 100\mu m$ (it is the usual size of a representative element of volume (*REV)* in such modelisations, as it can be found in [4] or [11]).

Setting one by one the previous parameters, we can deduce the ratio u/σ^2 of the random field needed and the correlation length L_c. In an ideal way, we should also set the volume fraction, which would lead to setting the variance σ but the strong non-linearity of equation (3) does not allow us to do so. So far we are limited to a maximum volume fraction around 30%, which for a cement paste represent a *w/c* of 0.7. We have therefore to assume arbitrary a variance $\sigma = 1$.

With all these parameters, we are now able to generate a realisation of a random field (Figure 3, left), and by thresholding it, we will create an excursion set (Figure 3, right) that will represent anhydrous cement. The cube's complementary will be set as water.

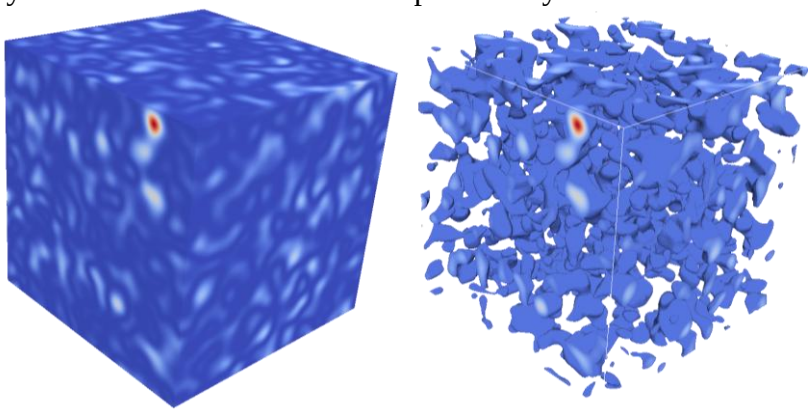


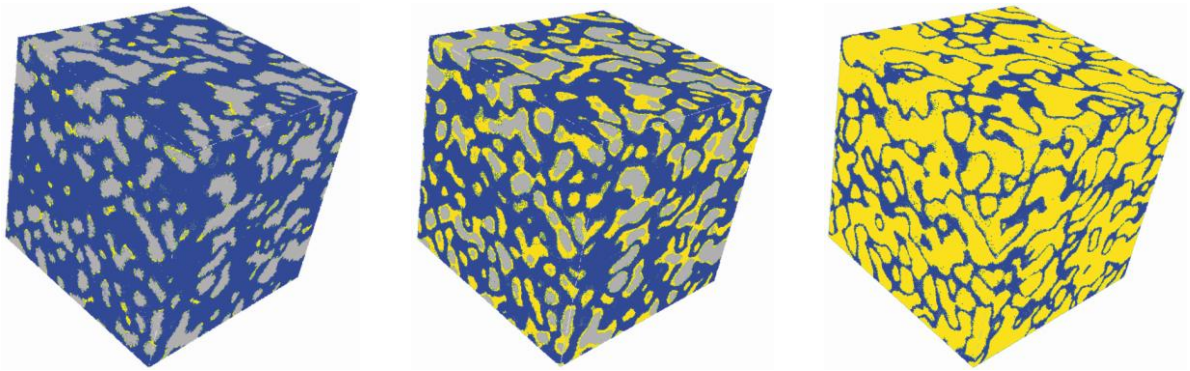
Figure 3 : Random field (left) and Excursion set (right, representing anhydrous cement)

3.3 Hydration process

Once an initial morphology is created, we use equation (3) to make evolve the threshold according to equation (4). So for a given hydration degree, we will know the exact volume fraction of each phase, and thus the thresholds we have to apply to yield the different phases. As we can see on Figure 4, we can follow the evolution of the morphology according to the hydration degree.

4. APPLCATIONS

We present here some possible applications for this morphological framework, with first the transition from morphology to FE truss, then an application aimed to estimate the Young modulus evolution over the hydration degree increase, and finally the estimation of an effective diffusion coefficient.



α=0.1 α=0.5 α=1

Figure 4: Morphologies for different hydration degrees (water, hydration products, anhydrous cement)

4.1 Embedded discontinuities

Once the morphologies are created, we need to "project" them on a FE truss, in order to make any kind of calculation. It has been chosen to use not adapted meshes, not to constrain nodes spatial position by the evolving morphology. It means that both mechanical and geometrical properties have to be handled inside some interface elements. These elements that are between two phases are split in two parts, each having different material properties by enhancing them with strain (weak) discontinuities [9]. We choose to use the Embedded Finite Element Method (E-FEM), to achieve this enhancement.

For the initial morphologies, we have two different materials, which mean we have three types of elements: the ones only in anhydrous cement, the ones only in water, and the ones between the two (cut elements). Following, for a given hydration degree, we will have three different materials, and three different cut elements.

To set properly the materials properties, a projection of the different excursion sets are made onto the trusses. That allow us to have a multiphase material, and each phase has its own material properties.

4.2 Young modulus evolution

One possible application of this framework is presented here, within a sequenced multi-scale approach. For a given cement paste, we can follow a property over hydration. For example, we estimate here the Young modulus evolution. We enter properties at a microscopic scale (each material is then different), and we are computing for a macroscopic property. With the material properties set in Table 1, and by doing first a tri-tension test, and then a pure shear test, for each step of hydration, we can estimate the Young modulus and Poisson's ratio. Timoshenko's bi-phasic beam elements are used, in order to take into account tension, compression and shear.

As we can see on Figure 4, the macroscopic Young modulus is growing continuously over hydration degree. The overall shape of the curve is quite representative for such *w/c* ratio [121]. A slight raise of the slope can be noticed after α=0.4. The final value is not very representative. However, one have to remember that such a cement will never go further than α=0.8, and that the volume fraction of hydration products is overestimated by all the assumptions made earlier. Besides, the percolation threshold seems too high compared to available experimental results [12].

Table 1: Material properties [12]

Property	E (MPa)	v[1]	D_e ($m^2.s^{-1}$)
Anhydrous cement	135000	0.25	$1{,}00.10^{-14}$
Hydration products	25000	0.25	$1{,}00.10^{-11}$
water	1	0.27	$1{,}00.10^{-07}$

The Young modulus values for anhydrous cement and hydration products are weighted mean between all the chemical components (Portlandite, Afm, Aft and C-S-H for a CEM-I cement) obtained by nano-indentation.

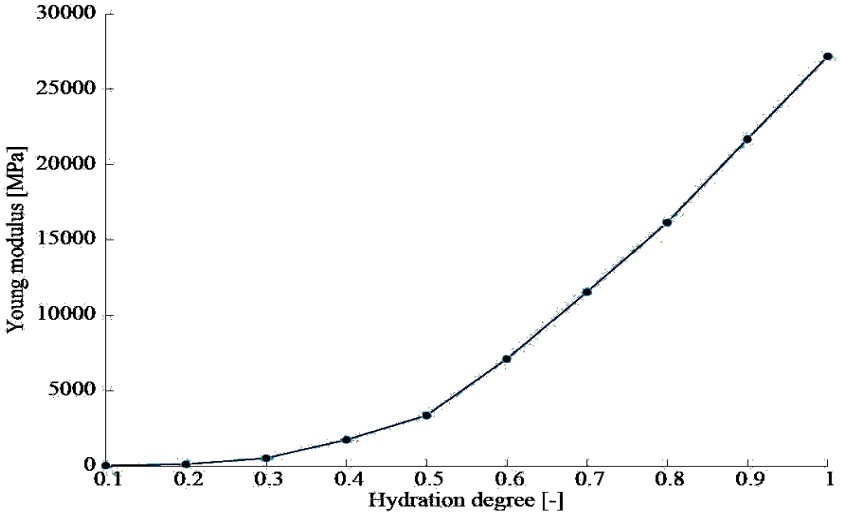


Figure 4 : Young modulus evolution for a cement paste (w/c = 0.7)

4.3 Effective diffusion coefficient

Another type of application is to focus on hydrated cement paste, and estimate transport properties. An application is presented here for the diffusion coefficient.

Same way as previously, we can set material parameters for each phases, here effective diffusivity (D_e in Table 1). This time we use bi-phasic bars with one degree of freedom. Then by applying unitary gradients in each direction of the *REV*, we can deduce the diffusivity matrix, which should be symmetric, and isotropic.

Using the Fick's law:

$$\underline{Q} = \underline{\underline{D}} \,.\, \underline{C} \qquad (5)$$

$\underline{C_1}$ = [1 0 0] ; $\underline{C_2}$ = [0 1 0] ; $\underline{C_3}$ = [0 0 1]

we can compute the resulting flux $\underline{Q}$, for each $\underline{C_i}$, and thus reconstruct matrix $\underline{\underline{D_e}}$:

$$\underline{\underline{D_e}} = \begin{pmatrix} 5.03 & 1.02\ 10^{-1} & 1.73\ 10^{-1} \\ 7.94\ 10^{-2} & 4.63 & 1.38\ 10^{-1} \\ 5.07\ 10^{-2} & 3.54\ 10^{-1} & 4.96 \end{pmatrix}.10^{-11}$$

[1] There is a numerical limitation for v ($0.1<v<0.275$), due to the beam elements.

We have, according to theory, a matrix (and thus a *REV*) more or less isotropic, with an order of magnitude between diagonal and extra-diagonal terms.

5. CONCLUSIONS

This work presents a morphological framework for modeling cement paste like materials within a sequenced multi-scale approach. Morphologies are yielded by thresholding realization of random fields. Then, by their projections, we present two different applications relying on non adapted meshes. As we pointed it out, efforts have still to be made on order to have more realistic morphologies (volume fractions), and the analytical model for the hydration process needs to be more accurate, taking into account more phases.

REFERENCES

[1] Adler, R.J. Some new random field tools for spatial analysis. *Stochastic Environmental Research and Risk Assessment* (2008) **22**:809-822Sterman, S. and Marsden, J.G., 'Silane coupling agents', *Ind. Engng. Chem.* **58 (**3) (1966) 33-77.

[2] Adler, R.J. and Taylor, J.E. Random Fields and Geometry (2007) *Springer*, Boston

[3] Benkemoun, N. Failure of heterogeneous materials: 3D meso-scale FR models with embended discontinuities. *Int. J. Num. Meth. in Engng.* (2010) **82**:1671-1688

[4] Kamali-Bernard S., Bernard F., Prince W., *Computational Material Science* (2009) **45** (2):528-535

[5] Karhunen, K. Über lineare Methoden in der Wahrscheinlichkeitsrechnung. *Ann. Acad. Sci. Fennicae. Ser. A. I. Math. – Phys.* (1947) **37**:1-79

[6] Loève, M. Probability theory. *Graduated Texts in Mathematics* (1978) Vol. II, 4th ed.

[7] Mantalgou, A. And Taylor, J.L. The Turning Bands Methods for Simulation and Random Fields using Line Generation with a Spectral Method. *Water Ressources Research* (1982) Vol. II, **2**:129-149

[8] Matheron, G. The intrinsic random functions and their application. *Advances in applied probability* (1973) **5**:439-468

[9] Ortiz, M., Leroy, Y. And Needleman, A. A Finite Element method or localized failure analysis *Computer Methods in Applied Mechanics and Engineering* (1987) **61**:189-214

[10] Powers, T.C. & Brownyard, T.L. Studies of the physical properties of hardened Portland cement paste. *J. Am. Concr. Inst.* (1947) **43**:101-132,249-336,469-505,845-880,933-992

[11] Šmilauer V., Bittnar Z. Microstructured-based micromechanical prediction of elastic properties in hydrating cement paste. *Cement and concrete Research* (2006) **36**: 1708-1718

[12] Stefan L., Benboudjema F., Torrenti J.-M., Bissonnette B., Prediction of elastic properties of cement pastes at early ages, *Computational Materials Science* (2010), Vol 47, **3**:775-784

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