# Modeling Mechanical Properties of Cement-based Materials from their Microstructure

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## 1. Abstract

The evolution of mechanical and physical properties of the cement paste is the consequence of hydration and physical changes in the microstructure. To predict the elastic properties of hydrating tricalcium silicate ($C_3S$) at early age, based on its microstructure and the characteristics of constituents, homogenization techniques are used. The evolving microstructure is generated by a continuum model based on hydration kinetics.
The estimation of effective properties of heterogeneous materials is made through the application of different kind of boundary conditions applied at the boundary of a Representative Volume Element (RVE). The effect of boundary conditions and the effect of microstructure size have been shown and the results discussed.
Furthermore, an ultrasonic transmission method was used to measure the evolution of elastic properties of hydrating cement paste with different water to cement ratios. The results obtained were correlated to microstructural parameters of the cement paste through the hydration model and numerical simulation of the mechanical properties.

Key words**:** elastic properties, homogenization techniques, hydration model, ultrasonic technique, size and boundary conditions effects.

## 2. Introduction

Cement-based materials, such as concrete, are generally complex heterogeneous composite material, with a random microstructure at different length scales ranging from the nanometer scale to the macroscopic decimeter scale. The evolution in time of their mechanical properties is strongly related to the hydration reactions in the cement paste.

Cement paste contains micro-pores and variety of capillary pores, which vary in volume and size with water-cement ratio, and the degree of hydration. Any mathematical or numerical model representing mechanical behaviour of such materials, which would claim to be realistic, has to take into account the heterogeneities at different micro-structural levels. In the past decades, different numerical models have been proved to be efficient tools in modelling physical and mechanical properties of cement-based materials at both the meso-level (mortars or concrete) [1-3] and the micro-level (cement paste) [4-7]. One of the models [6], called IPKM (Integrated Particle Kinetics Model), a continuum mode in which the effect of inter-particle contacts and the accessibility of water on the rate of hydration and on the structure formation are taken into account explicitly. In this simulation the anhydrous particles of $C_3S$ are considered to be spherical and possess a similar particle size distribution as Portland cement particles [7]. The microstructure parameters including the overall degree of hydration, pore volume, the contact surfaces, solid phases content can be simulated by the model.

Moreover, ultrasonic wave methods have been used for long time to as NDT technique to characterise the evolution of hardening and setting of cementitious materials [8]. The elastic properties of an isotropic elastic solid are fully described by two independent elastic constants, bulk modulus and shear modulus. Acoustic wave velocities (compressional and shear velocities) are functions of the two elastic constants. Therefore, measurement of both compressional and shear wave velocities is necessary to fully provide the two elastic constants.

The work described in this paper aims t o characterize the early age properties of cement paste by numerical and experimental techniques and thus to establish a correlation between mechanical properties development and some microstructure parameters.

## 3. Ultrasonic wave testing

In order to follow the evolution of the properties of cement paste at early ages, non destructive ultrasonic wave propagation technique is used. The base of this technique is the evaluation the velocity of both longitudinal and transverse waves by measuring the fly time through the specimen. By considering the cement paste as isotropic and the wavelength larger than the size of the largest inhomogeneity (the largest pore or the largest grain), the Young's modulus E and the shear elastic modulus G are determined.
To avoid any entrapped air, the samples were vacuum pumped for 20 minutes. In fact the presence of air bubbles in the mix cause a very high attenuation of the compressional waves mainly in the early age (Fig.1 and Fig.2) and thus influences the of the mechanical properties calculation of the cement paste.

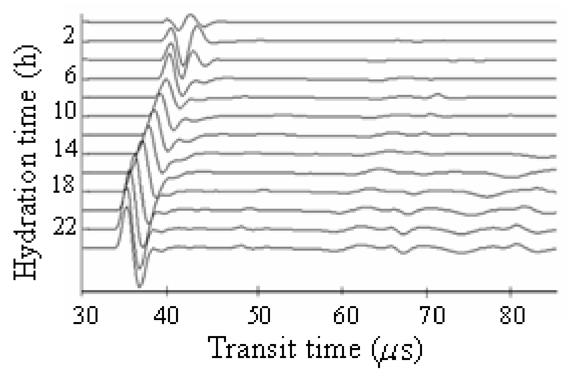


Fig.1: Waterfall plot of compressional waves at increasing hydration time for de-aerate paste

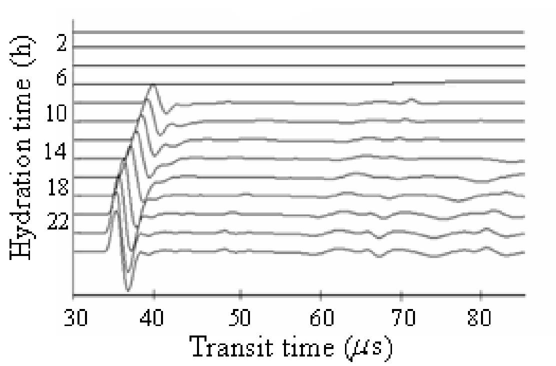


Fig.2: Waterfall plot of compressional waves at increasing hydration time for no de-aerate paste

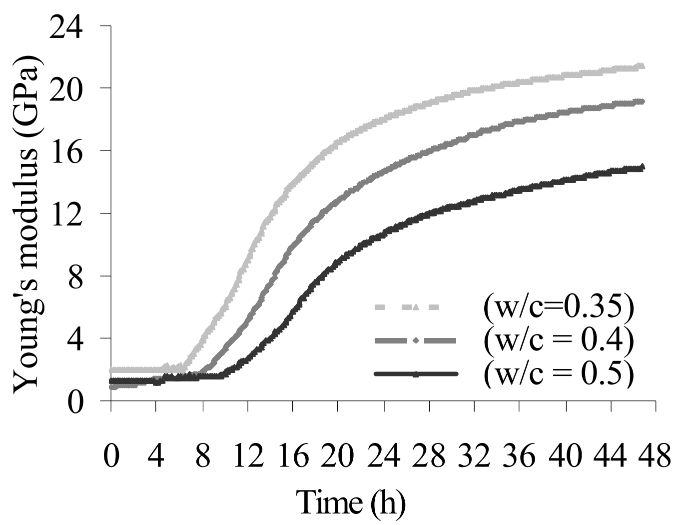


Fig.3: Evolution of Young’s modulus for different water to cement ratios

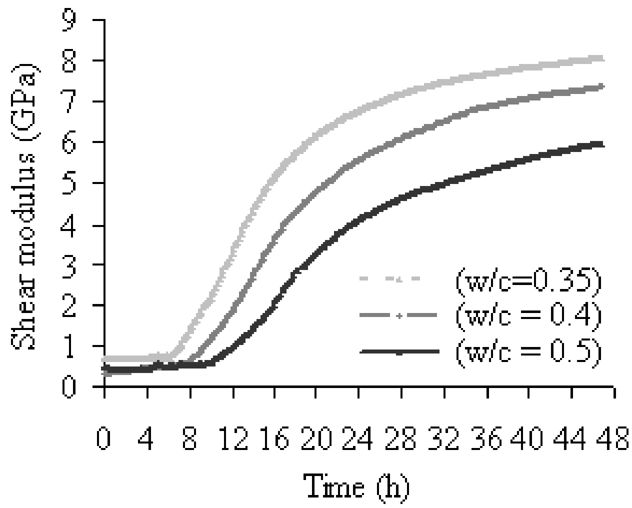


Fig.4: Evolution of Shear modulus for different water to cement ratios

In this study, Portland cement pastes with different water/cement ratios were investigated. The compressional and shear wave signals are recorded every 10 minutes during 48 hours. The Young's and Shear modulus for different water-cement ratios are plotted in Fig.3 and Fig.4 respectively.

These moduli increase strongly at early ages but progress slowly during later stage. The w/c ratio affect the development of Young's modulus as it is shown in Fig.3 and Fig.4. As expected, the lower the water to cement ratio, the stiffer the material.
It was also possible to relate the development of mechanical properties to the degree of hydration (Fig.6) by measuring the degree of hydration using isothermal calorimetry ( Fig.5). The degree of hydration $\alpha$ is the ratio $Q/Q_{max}$, where $Q$ is the heat produced at a given time and $Q_{max}$ the heat that would be produced in a complete hydration of cement. The value of $Q_{max}$ is calculated from the enthalpies of the hydration reactions and the proportion of the constituents.

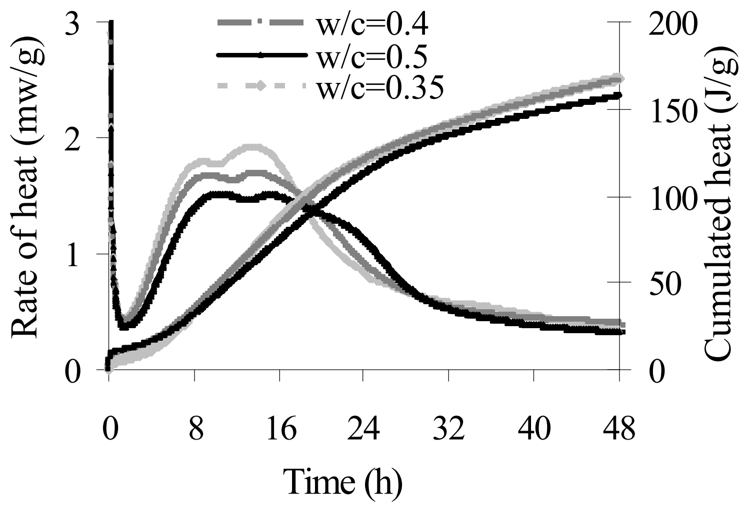

Fig.5: Isothermal calorimetry of Portland cement paste at 20°c

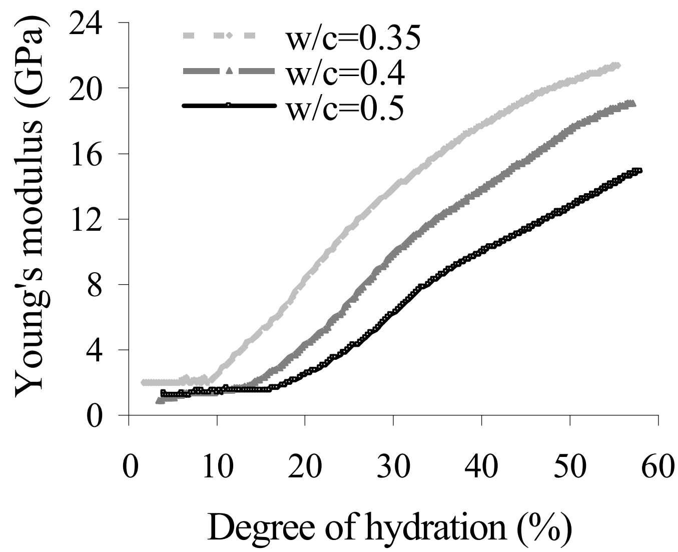

Fig.6: Evolution of Young's modulus of cement paste for different water to cement ratio

## 4. Numerical evaluation of elastic properties of hydrated $C_3S$ paste

### 4.1 Cement Hydration model

To determine numerically the effective properties of cement paste, the IPKM model was used to simulate the hydration of the tricalcium silicate ($C_3S$) - the main component of the cement. In this model, the anhydrous $C_3S$ particles are considered spherical and they are placed randomly in a periodic volume according to a particle size distribution and a chosen water-cement ratio. Three different mechanisms; nucleation and growth, phase boundary reaction and diffusion; control the evolution of hydration of $C_3S$. At the start of the hydration process there are two phases: $C_3S$ and water. After hydration, we have five phases among them three solid phases ($C_3S$, CSH, and CH) and tow fluid phases (water and air) which form the capillary porosity (see Fig.9).

During hydration, each particle reacts with water and interacts with the neighboring particles. At each step of hydration the overlap of each particle with the other particles is checked and if there is an overlap the contact surface is calculated directly from the real microstructure generated by the hydration model as shown in Fig. 9(a), [7]. In the second step, the same contact surface is calculated from the discretized microstructure as shown in Fig. 9(b). Fig.7 shows the contact surface for each step of hydration calculated in a computational volume 50 x 50 x 50 µm with a voxel size of 3.33 µm (regular mesh) and compared to the one obtained directly by the microstructure model. The difference between the two the results reveals the effect of the mesh ing technique used on contact area calculation. Thus, using a regular mesh seems to overestimate the real contact area. This can be avoided by a deep mesh refinement - which will increase the computational time -or by non-regular mesh technique which can take into account the topology of both anhydrous particles and hydration products. Moreover, the computational size seems to have a weak effect on the value of the contact area (Fig.8).

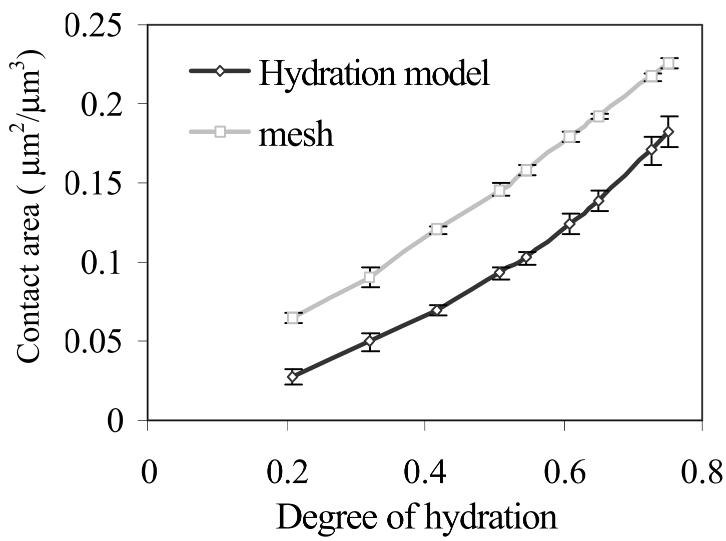


Fig.7: Evolution of specific contact area before and after meshing

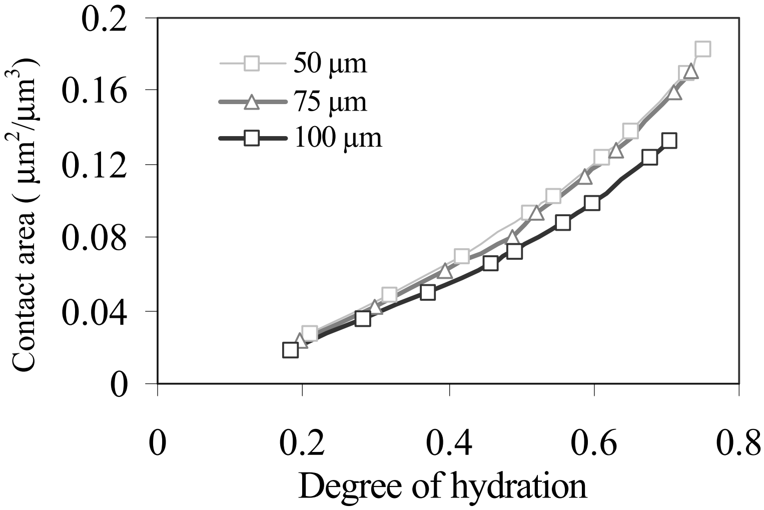


Fig.8: Effect of RVE size on contact area

4.2 Numerical testing

The homogenisation process is made on a three different computer-generated microstructures with w/c=0.4 and the same particle size distribution with appropriate truncation. The diameter of particles varies from 0.4 to 40 µm. Fig.9 shows a 2D cross-section after 96 hours of hydration of $C_3S$ in a computational volume of 100 x 100 x 100 µm.

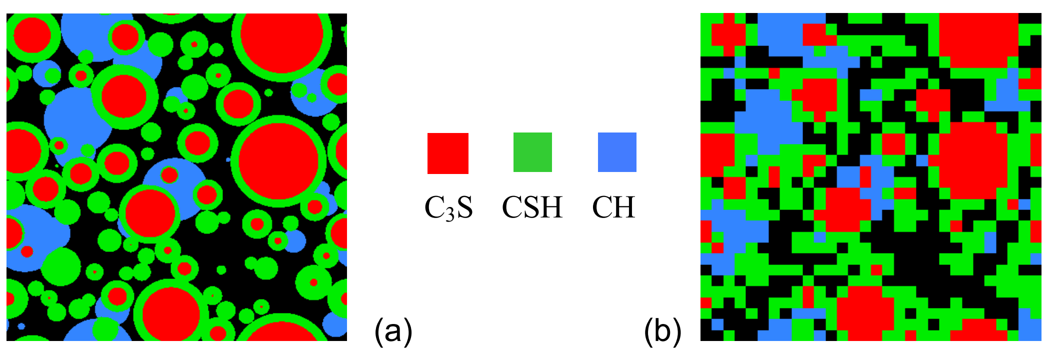


Fig.9: Computer generated microstructure (2D cross section) before (a) and after meshing (b).

The estimation of effective properties of heterogeneous materials is made through the submission of the specimen to three boundary conditions. In Kinematic Uniform Boundary Conditions (KUBC), the whole external surfaces of the sample are submitted to prescribe displacements. While in

Static Uniform Boundary Conditions (SUBC), the whole external surfaces of the sample are submitted to prescribe load. The third kind is the periodic boundary conditions (PBC) which can be considered as mixed but non uniform boundary conditions [9].

In computational mechanics, any porous volume present in the microstructure is usually discretized into Finite Elements for which near-zero elastic properties are assigned. The estimation of overall mechanical properties is then performed through the classical concept of prescribed uniform displacement and prescribed traction, which are applied at the boundary of a Representative Volume Element (RVE). The application of these three boundary conditions to heterogeneous solids (containing only solid phases) having the RVE should predict similar effective properties [3-9]. But the application of this approach to porous systems often gives a large difference between the three boundary conditions because of the high contrast between the properties of the solid phases and those assigned to the pore space. Hence, by considering the real porosity present in the computational volume (all the structure are meshed unless porosity), the effect induced by the presence of pores is definitively eliminated.

To be able to apply any type of loading on such complex and highly porous microstructure, the specimen was put in solid box with unknown material properties. The latter are determined through an iterative procedure until they reach the apparent properties of the specimen. However an error may be made here with the adding of the external layer due the complex stress and strain fields at the interface between the microstructure and the added solid box.

In the simulation process, the elastic properties were investigated on the microstructures with the edge of 50, 75 and 100 µm. The particles are randomly placed in the numerical volume with w/c=0.4 and for each size three specimens -with different spatial configurations- were generated. The material properties of the three solid phases adopted for the calculation are given in Table1.

| | $C_3S$ | CH | CSH |
|---|---|---|---|
| E (GPa) | 135 | 40 | 31 |
| ν [-] | 0.31 | 0.3 | 0.28 |

Table 1: Mechanical properties of the three solid phases

For each step of hydration, the variation of the concentration of the existing phases is given in Fig.10. Fig.11 shows the evolution of the Young's modulus in terms of hydration degree obtained under three different types of boundary conditions. The effect of boundary conditions is clearly shown. As expected, the upper bound is obtained under kinematic conditions (KUBC) while the lower one is obtained under static ones (SUBC). This means that the size of the computational volume used in the numerical simulation is lower than the RVE. Hence, the calculated properties can not considered as the effective properties but only apparent ones which are size dependent [3]. Moreover, the results obtained under periodic conditions are bounded by those under the pure kinematic and pure static conditions. This means that the periodic calculated E-modulus is closer to the effective.

The effect of RVE size on the elastic properties has been investigated on a microstructures with the degree of hydration α=0.3. Fig.11 shows the Young's modulus calculated under the three types of boundary conditions described above. The results show the quasi-independency of the PBC-calculated E-modulus vis-à-vis of the specimen size. Hence, in the rest of paper, the numerical simulation will be performed only under periodic conditions.

We can see also (Fig.12) that the numerical results mainly in the beginning of hydration are overestimating the real properties. This can be explained by the excess of contact area induced by the regular mesh technique which is unable to capture the real microstructure generated by the hydration model. Nevertheless, it will be interesting to study the correlation between the Young's modulus and the contact area.

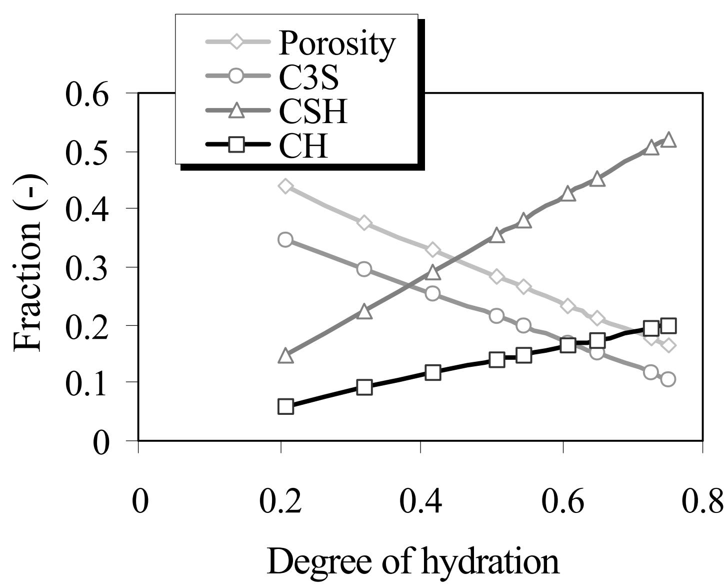


Fig.10: Concentration of different phases during the hydration process

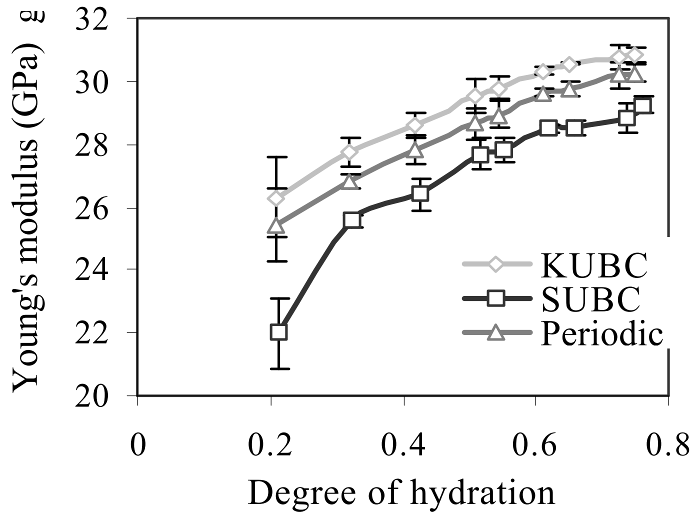


Fig.11: E-modulus of $C_3S$ in terms of hydration degree,
Specimen size = 50 x 50x 50 µm

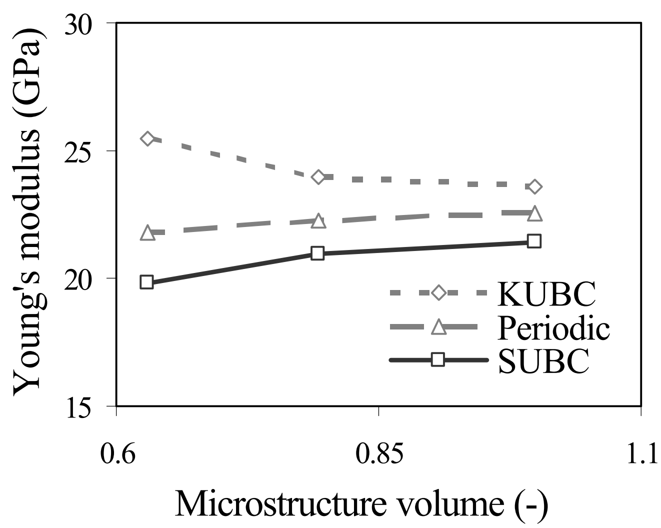


Fig.12: Effect of RVE size on E-modulus, α=0.3

### 4.3 Contact area

The contact area is a good indicator of the development of the mechanical behavior of cement paste such as the strength and the Young's modulus. Although the contact area is not a measurable quantity, it is a suitable tool to quantify the degree of connection between the solid particles in the hydrating microstructure [10]. As mentioned above, the contact area is a microstructure parameter which can be calculated by the hydration model [6-7]. A correlation between Young's modulus and contact area between hydrated grains, similar to the typical relationship between compressive strength and gel space ratio [11] or strength and porosity [12] has been studied (see Fig.13). The result shows a good correlation between Young's modulus under periodic boundary conditions and specific contact area for the computational volume having the size of 100 x 100 x100 µm.

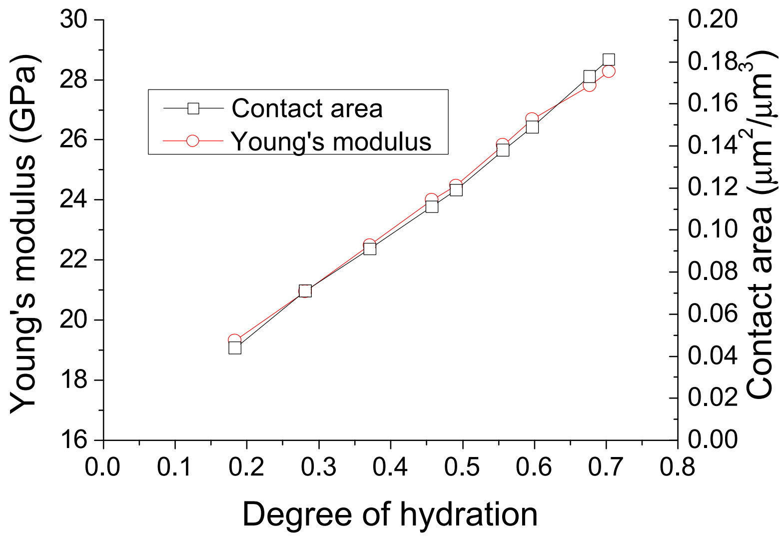


Fig.13: Correlation between young's modulus and contact surfaces, specimen size =100x 100 x 100 µm

5. Conclusion

It has been shown that both the development of the microstructure and the evolution of the mechanical properties of cementitious materials can be described or predicted either by numerical simulation model or by ultrasonic wave characterization method. The latter may a valuable tool to validate the results obtained by numerical modeling. A good correlation has been found between the evolution of Young's modulus predicted by the numerical model and the specific contact area. I turns out that the elastic modulus is not only sensitive to the volume of the connected solid particles as it's widely accepted in the literature, it can also be directly related to the degree of connectivity of the different solid phases present in the macrostructure. However, we should notice that, even if the trend is the same for the methods, the elastic modulus obtained by the numerical model is always overestimating the measured one. This is mainly a mesh generation problem that may solved by a better non-regular mesh technique which now under development.

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