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Multi-cycle and multi-scale cellular automata for hydration simulation (of Portland-cement)

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ABSTRACT

CEMHYD3D is a cellular automata (or agent) based computer model for the hydration of cementitious materials, which is able to predict the microstructure and physical properties of such hydrating systems. In this paper, CEMHYD3D is successfully extended for multi-cycle and multi-scale modelling. Multi-cycle means the possibility to zoom in and out on the hydration process with respect to time. Multi-cycle modelling enables the user to study the hydration in more detail in both the early phase (minutes) and on the long term (years). This modelling is needed, for instance to be able to model the hydration of reactants with different reaction rates (e.g. Portland cement and calcium sulphates). The multi-scale modifications enable the use of smaller particles than the standard minimum size of 1 μm, which permits the incorporation of submicron particles in the model. These particles are present in most cementitious binders, and their inclusion will improve the predictions of properties during simulation.

Based on statistical considerations, the dissolution and nucleation probabilities and the number of diffusion steps from the original model have been modified in order to enable multi-scale and multi-cycle modelling. For the multi-scale, two variants to obtain the microstructure at higher resolution have been applied, voxel splitting and rescaling, which differ in the way they deal with the particle shape.

All modifications have been tested for the modifications separately as well as both combined, for a system consisting of an OPC cement, using CEMHYD3D. All simulations showed good agreements between the results at different resolutions (for both scaling methods) and applying different time-steps, confirming the validity of the generally applicable equations presented here.

1. Introduction

Hydration models are used to reduce the number of the real tests needed to optimize a mix design. Examples of these hydration models are CEMHYD3D [1], HYMOSTRUC [2], the Navi and Pignat model [3], μic [4] and HydratiCA [5,6]. These models are able to represent the physical properties and present reaction mechanism during hydration. As pointed out by Chen and Brouwers [7], the physical properties of a hydrating microstructure are limited by the smallest element-size available in the system, the so-called system resolution. Besides the representation of the properties, the system resolution also influences the needed computation power and time needed to run a simulation. In this paper it is shown that resolutions of 0.2 μm nowadays are possible due to increased computations power, while in the original version of CEMHYD3D the used resolution is 1 μm. The use of lower resolutions leads to problem that the amount of hydrated cement per cycle decreases when the system resolution increases [8] and therefore the predictions of the models significantly depend on the system resolution. Garboczi and Bentz [8] discuss the required resolution needed and concluded that 0.2 μm is sufficient. Chen and Brouwers [7] have incorporated the diffusion layer system of Van Breugel [2] into the CEMHYD3D-model.

This paper intends to improve the predictions of the CEMHYD3D model for different system resolution (different scales) by further modification of the dissolution and nucleation probability and the number of diffusion steps within the model. The CEMHYD3D is chosen because of it is believed to be one of the most advanced, well-known and most widely-used computer hydration models [2,4].

Besides the multi-scale modelling, this paper also pays attention to multi-cycle modelling. Multi-cycle modelling enables to zoom in and out into hydration process with regard to time. This is needed for instance when one wishes to study the hydration of calcium sulphate hemihydrates, which have a extreme short hydration time compared to cements, or when one wishes to study

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the long-term reaction behaviour of cementitious materials, when the reaction rate becomes extremely slow.

Both modifications will lead to modifications in the modelling of the hydration process, which consists of dissolution, diffusion and nucleation stage of the process.

2. CEMHYD3D: a 3-D computer-based hydration model

The computer model CEMHYD3D was originally developed by Bentz and Garboczi [9] to represent the hydration process of Portland cement in two-dimensions. Later on, the model was later extended to a 3D-computermodel and other cementitious materials, like fly-ash and silica fume, as reactants were included [10]. Van Eijk [11] has calibrated the model with two Dutch cements CEM I 32.5R and 52.5R, and introduced pore water chemistry. Chen [12] and Chen et al. [13] have introduced slag blended cement into the hydration model including the new phases and their reactions as well as the difference in reactivity between cement and slag material. Furthermore further improvements were done in order to overcome some side effects of changing system resolution. At last mineral shrinkage compensating mixtures were designed by Chen and Brouwers [7] and Chen [12] based on simulations and real tests. Bentz [14] compared CEMHYD3D and real 3-D X-ray microtomography cement paste microstructures using correlation functions. Igarashi et al. [15] compared the simulated microstructures of CEMHYD3D with SEM-BSE image analysis of cement pastes microstructures. Smilauer [16] studied the elastic properties of hydrating cement paste by applying elastic homogenization methods on the microstructures provided by CEMHYD3D. Koster [17] uses CEMHYD3D for the simulation of 3-dimensional moisture transport through and moisture absorption by capillary-porous building materials. Feng et al. [18] uses the initial microstructure provided by CEMHYD3D to simulate the influence of leaching on hydrated cement pastes. Robeyst et al. [19] related the fundamental changes in the microstructure, noticed using ultrasonic measurement during the setting of concrete, with the microstructure development simulated by CEMHYD3D. Newest developments introduced by NIST can be found in the work of Bentz [14,20–22].

The CEMHYD3D-model usually represent the microstructure of hydrating cement by using a $100 \cdot 100 \cdot 100$ box with 10^6 voxels of size $1 \cdot 1 \cdot 1$. Other resolutions are possible to use, although larger system sizes and smaller voxel sizes represent the microstructure more accurately, the computing time needed for these system and voxel sizes is remarkably higher. According to Smilauer and Bittnar [23], the reasonable microstructure size lies in the range of 20–50 μm and a microstructure edge size above 100 μm has been found to bring no significant accuracy in the hydration model predictions. Garboczi and Bentz [8] point out that 0.2 μm resolution is accurate enough for simulation of cement hydration. Therefore in this paper a microstructure of $100 \cdot 100 \cdot 100$ μm^3 is used while a system resolution up to 0.2 μm is tested.

The particles within the initial microstructure are created by placing voxels in an approximately spherical shape called "digitized particle" within the box. A 1-μm particle in CEMHYD3D is represented by 1 voxel at a system resolution of 1 μm, while at a system resolution of 0.33 μm it is represented by 19 voxels with size of 0.33 μm. An example for the digitized particle of size 1, 3, 5, 7, 11 and 21 voxels is shown in Fig. 1, which contain 1, 19, 81, 179, 739 and 4945 voxels, respectively.

In the original version of CEMHYD3D only phase-boundary-reaction (chemical reaction controlled) was considered. According to Chen [12], the diffusion layer is needed in order to correct for the effect of the system resolution. He incorporated the diffusion layer into the CEMHYD3D-model, based on the work done by Van Breugel [24]. Van Breugel [24] showed that the reaction rate is constant for phase-boundary reaction and decreasing for diffusion controlled circumstances. The transition point between both mechanism is defined by layer thickness δ_{tr}. This is shown in Fig. 3. Both mechanism and the transition layer have been incorporated by Chen [12] and Chen and Brouwers [7]. The dissolution probability is an complicated function depending on temperature, reaction degree, sulphate concentration and other parameters. For readability here for the description of dissolution probability function, $P_{D,1}$ is used. $P_{D,1}$ includes the earlier mentioned parameters. The dissolution probability for the phase-boundary reaction reads;

$$P_{D,1} = P_{D,0} \quad \text{for } \delta \leqslant \delta_{tr} \tag{1}$$

and the dissolution probability for the diffusion controlled part, based on Chen [12] and Chen and Brouwers [7], reads;

$$P_{D,1} = P_{D,0} \cdot \frac{\delta_{tr}}{\delta} \quad \text{for } \delta > \delta_{tr} \tag{2}$$

with $P_{D,0}$ the basic dissolution probability function (here presented as an constant value, but in fact an complicated function), δ the layer thickness and δ_{tr} the theoretical transition layer thickness. According to van Breugel [24], the transition layer thickness for low heat cement equals 2–4 μm and for fast hydrating cements 3–6 μm. In CEMHYD3D a δ_{tr} of 2 μm is chosen [7,13]. A full description of CEMHYD3D can be found in [7,10,25].

3. Multi-cycle modelling

In this section, the option of multi-cycle modelling is introduced. The idea behind multi-cycle is the variation of the length of the time steps in the model. Fig. 2 shows the principle of multi-cycle modelling. In this section the term reference cycle will be used to describe a cycle within the original model. In order to describe this variation, the multi-cycle factor k is introduced. A higher multi-cycle factor results in k smaller time steps during hydration and therefore resulting in k time more cycles to achieve a same degree of hydration. Hence, for a reference cycle holds that $k = 1$.

This is necessary, because the hydration of some reactants, e.g. hemihydrate, may be much faster compared with the hydration of the cement clinkers. The complete hydration of hemihydrate takes place within half an hour, while cement takes a few days. Multi-cycle modelling introduces two major adaptations to the model: the modification of the reaction kinetics and a correction of the calculation of the reaction time.

Within CEMHYD3D the kinetics are mainly regulated by the dissolution and nucleation probabilities and the number of diffusion steps. Due to smaller time step at a larger k-factor, the number of particles that dissolve, diffuse and nucleate is smaller. Therefore these probabilities need to be smaller. In this section, the modification of the reaction kinetics is described. In this paper the reference system refers to the system with system resolution of 1 μm and without any multi-scale (nor multi-cycle) modifications.

3.1. Dissolution

The expectation of a voxel-face to dissolve in the reference system during a cycle equals the dissolution probability ($P_{D,1}$). In general, for k cycles the expectation reads

$$\sum_{i=0}^{k} i \left[\binom{k}{i} P_{D,k}^{k-i} (1 - P_{D,k})^k \right] = k P_{D,k} \tag{3}$$

With $P_{D,k}$ is the dissolution probability when a cycle is divided into k new cycles. This probability is equal to the expectation of dissolution in the reference system, $P_{D,1}$, when