



Impact of the microstructure model on coupled simulation of drying and accelerated carbonation

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ABSTRACT

During carbonation, the microstructure of a cement paste is strongly modified, which induces a dynamic feedback loop impacting the global advancement of the reaction. In this study, we investigate the effect of the microstructure model on simulations of accelerated carbonation of cement pastes. A variable-porosity reactive transport model with a new microstructure model is used. To match the chemical layers created by the carbonation front, we use a layered microstructure model. We demonstrate that qualitative prediction for a large range of conditions and formulations can only be obtained if the macroscopic parameters of the carbonated layer can be modelled accurately. In particular, the saturation dependent parameters, such as the capillary pressure or the relative diffusion coefficients, control the rate of advancement of the reaction front since a high saturation inhibits the carbon dioxide diffusion.

1. Introduction

Models and simulations are very promising methods to study the durability of cement paste and concrete for many formulations, and under many conditions. However, simulating long-term durability issues such as carbonation is still an open problem. The main difficulty is that most reactions include precipitation and dissolution of solid phases [1]. The changes of the solid phase assemblage modify the microstructure, and thus the macroscopic transport properties. Capturing accurately the feedback of the transport properties on the reactions, and the feedback of the reactions on the transport properties, is necessary to obtain quantitative predictions [2,3].

Carbonation is the reaction of the cement hydrate phases with atmospheric CO₂ [1]. As CO₂ ingresses, it decreases the pH of the pore solution, and phases such as Portlandite and C-S-H dissolve, and finally carbonated phases, mainly calcium carbonate, precipitate. The reaction progresses toward the core of the sample in the form of a sharp reaction front separating the carbonated from the non-carbonated layer.

The CO₂ diffusion mainly occurs through the carbonated layer, from the surface to the reaction front. If the transport and chemical properties of non-carbonated cement paste have been well characterized through the years, the carbonated layer remains mostly unexplored. Notable exceptions include the experimental studies by M. Auroy et al. [3] and A. Morandeu et al. [4]. A common observation is that the

microstructure is strongly modified (see e.g. [3, Fig. 12]). The porosity is not a sufficient descriptor as the pore size distribution is changed [3,4]. In addition, cracks may also occur, e.g. due to carbonation shrinkage [5] and counteract the porosity clogging due to the calcium carbonate [3].

A common method to model the microstructure changes is to modify the macroscopic transport properties as a power law of the porosity [6,7]. Strong theoretical background exists for the power laws [8,9]. However, the assumptions of this theory do not apply to the pore size distributions of typical cement pastes. Their application to cement paste is mainly based on empirical and practical reasons [6,10–12]. In addition, the reference state is often the non-carbonated state [6,7]. Leemann and Moro [13] found a poor correlation between the reaction front propagation and the gas diffusion coefficient of uncarbonated concrete. The main implication is that the gas diffusion coefficient of the non-carbonated layer is not a good descriptor of the microstructure of the carbonated layer. The modification to the microstructure by the carbonation reaction is dependent on the formulation [3,4]. Therefore, the relation between carbonated and non-carbonated properties can not be captured by a single parameter. To model a wide range of formulations, the model must be able to capture the impact of the chemistry on the microstructure, and the impact of the microstructure on the carbonation reactions. Simpler model which can not capture these feedbacks need to be calibrated for each formulation and set of

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conditions. This is an obstacle when extrapolating from accelerated to natural conditions, or when extrapolating to a different formulation.

Coupled simulations of carbonation and drying require the knowledge of many parameters, such as the liquid and gas diffusion coefficients, the liquid permeability, the water sorption... [7,13-18]. Therefore, building an accurate model is challenging. The long-term objective of this study is to develop a numerical model of carbonation providing quantitative predictions. The first step, and the focus of this study, is to investigate the requirements for such a model from the experimental, modelling and numerical point of view.

This article is based on the hypothesis that to obtain quantitative predictions we need to modify how the model is built and tested. The engineering variable is the carbonation depth. As such many models have been validated with respect to the evolution of the carbonation depth with time. Our main argument against this approach is that the carbonation depth follows a square root law as a function of time, typical of diffusion law [14]. Therefore, if both a diffusion law is used, and the coefficient of diffusion is fitted on the same experiments, the evolution of the carbonation depth is not a good observable to decide of the correctness of the model. This is because the fitting exercise only test if the mathematical model can represent the experiments. The diffusion behavior is already well-established in models since the work of Papadakis et al. [14]. The relationship between the evolution of the microstructure due to the carbonation, and the macroscopic parameters should be calibrated on independent experiments. This separation is particularly important if we want to understand the differences between natural and accelerated carbonation. In this article, our objectives are to prove this hypothesis, and to explore new observables to obtain more rigorous method to build and to test carbonation models for quantitative predictions. A main obstacle to this goal is the lack of complete sets of data containing for the same samples: chemistry analysis, microstructure information, macroscopic properties and macroscopic experiments of carbonation. As a consequence, choices must be made. In this paper, we focus on sensitivity analysis rather than strict experiment reproduction because (1) the simulations results have the correct order of magnitudes and (2) many unknown parameters can be modified to obtain results inside the experimental range of uncertainties. These points will also be demonstrated in the paper.

To analyze the importance of carbonated and non-carbonated parameters independently, we used a layered microstructure model to match the chemical layering [6]. With this model, we are able to assess the relative effect of each parameters in each layer, both on the global advancement of the reaction, and individual observables such as the pH or the saturation. Our main contribution is to analyze in detail the feedback cycle between the ingress of CO₂ and drying. In particular, we demonstrate that the mass conservation of water needs to be solved rigorously. This includes the ability to predict the parameters such as the capillary pressure, the relative diffusion and the permeability. To focus only on a few issues, this study contains only simulations for accelerated carbonation in isothermal conditions.

The paper is organized as follows. Section 2 presents the model used in this study, the numerical framework and the simulations we executed. Section 3 presents the results while Section 4 discusses them in the light of our long-term objective of quantitative predictions.

2. Carbonation model and simulations

2.1. Numerical framework : ReactMiCP

The simulations presented in this article were solved using ReactMiCP [19], a reactive transport framework developed by the authors [20]. ReactMiCP aims to be a generic reactive transport simulator, but it was developed to be able to solve the layered carbonation model. A layered microstructure model has strong numerical requirements. A main feature of ReactMiCP is the strong separation between the transport equations and the microstructure model. In addition, to the

transport and chemistry stagger commonly used in operator splitting algorithm, ReactMiCP introduces the upscaling stagger in charge of computing the microstructure model and the macroscopic parameters. This stronger separation requires a robust and flexible solver. A new sequential iterative algorithm was developed to meet these requirements. The convergence criteria used is the residuals of the transport equations (see Eq. (1) and [20]). Other advantages of this method are that variables are always consistent and that the solution is always correct independently of any non-linearity from the macroscopic parameters or the chemical reactions. As an iterative algorithm, it also reduces the dependence of the simulation on the mesh size and the timestep [20]. The transport equations are solved with a custom vertex-centered finite volume solver. It uses an element-by-element finite difference algorithm to build the Jacobian. As a consequence, any nonlinearities from the a-priori unknown macroscopic parameters are taken into account. Implementation details were inspired by Dynaflow, a nonlinear transient finite element analysis program [21]. The chemistry stagger also uses a custom speciation solver, SpecMiCP [22]. This solver uses a rigorous method, based on a complementarity problem, to find the equilibrium phase assemblage instead of the trial and error approach [22]. In addition, it was developed as a reactive transport module hence the coupling is easy and efficient.

The model presented in this paper can be directly implemented in ReactMiCP. To our knowledge, it is the only software with this set of features. As detailed in this article, it opens new investigations of cementitious materials.

2.2. Governing equations

2.2.1. Components

Solving the transport equations for every species is too computationally expensive. For reactive transport, the species, and their corresponding chemical reactions are expressed as a function of a subset of primary species, called the basis [23,24]. The basis is formed of N_c components. Due to the use of complementarity conditions in SpecMiCP [22] only one basis is used during the simulations. The basis used is: H₂O, Ca²⁺, CO₂, Al(OH)₄⁻, SO₄²⁻, Si(OH)₄, Na⁺, K⁺.

The general form of a component mass balance is written as Eq.(1):

$$\frac{\partial \mathcal{C}_i^\alpha}{\partial t} = -\nabla F_i^\alpha + \sum_{\beta \neq \alpha} R_i^{\beta \rightarrow \alpha} \quad i = 1 \dots N_c \quad \alpha, \beta = l, g, s \quad (1)$$

where \mathcal{C}_i^α is the total concentration of component i in phase α per unit volume of material, F_i^α is the flux of component i in phase α per cross-section of material, and $R_i^{\beta \rightarrow \alpha}$ is the rate of transfer of material from phase β to phase α per unit volume of material.

The total concentration of a component is the weighted sum of the concentrations of the species containing this component

$$\mathcal{C}_i^\alpha = c_i^\alpha + \sum_j \nu_{ji} c_j^\alpha \quad (2)$$

where c_k^α is the concentration of species k in phase α , and ν_{ji} is the stoichiometric coefficient in the chemical reaction :

$$A_j = \sum_i \nu_{ji} A_i \quad (3)$$

The physical meaning of a total concentration is explained in [24].

There is no transport in the solid phases so, for all solid phases m it can be simplified as :

$$\frac{\partial \mathcal{C}_i^m}{\partial t} = R_i^{l \rightarrow m} \quad (4)$$

The system is in isothermal conditions (25 °C). Temperature will have an effect on the chemical reactions [25] and the transport properties [26]. However, since most accelerated carbonation experiments are run under isothermal conditions, and the parameters uncertainties are already high, these effects are neglected.

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