

A Lattice Boltzmann approach for advection-diffusion problems in porous media including dissolution

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Micro Abstract

A LB model is presented to simulate advection-diffusion processes in porous media. Chemical reactions occurring on the pore surfaces causing dissolution require continuous surface adaptation. The approach utilizes the Cumulant LB model to model advection, a second LBM for the concentration of the chemical species, and an explicit surface dynamics solver governing the transient evolution of the surface geometry.

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Introduction

Reactive transport processes are dominant in natural and man-made porous material like concrete (or more generally cementitious materials). Chemical processes that occur in such materials include the hydration of cement, alkali-aggregate reactions, the corrosion of steel reinforcement or the degradation of hardened cement pastes due to the attack of chemical species on the solid phases produced by the cement hydration process. The latter will eventually lead to a gradual degradation of the solid phases either by dissolution or by precipitation. In dissolution reactions, chemical species force some of the solid phases to leach (e.g. decalcification and external sulphate attack) [1]. On the other hand, precipitation causes the formation of salt crystals inside the pores of the cementitious materials and due to their increased volume, this may induce internal stresses on the inner surfaces of the pores causing cracking ultimately leading to a loss of material strength (e.g., the reaction of de-icing salts with the monosulphate phase producing Friedels's salt) [3]. The aforementioned dissolution/ precipitation reactions mainly occur at the surface of the pores and they typically alter the position of the interface between the solid and the fluid / gas phases and thus modify the mechanical and transport properties of the material giving rise to a non-linear reaction process in turn. The importance of studying such chemical reactions stems from its relationship to cementitious materials' durability, which is considered a measure of the service-life of the concrete structures. Since, as mentioned above, cementitious materials are vulnerable against chemical attacks, and before any attempt is to be made to optimize the material in a way that reduces its vulnerability i.e., attempts to increase the life-time of the structure, a thorough understanding of the phenomena which occur in the micro pores is required. Although, the problem domain takes place in the macro-scale (continuum scale) from a practical point of view, it is rooted at the smallest pore (micro-scale) and spreads among a hierarchy of length and time scales. Thus, the motivation of this study is the development of a framework for predicting degradation of cementitious materials exposed to chemical attack. Enhancements of existing numerical models are being taken into consideration while building the framework to guarantee correct predictions of transport and reaction in cementitious materials. Moreover, a potential outcome from such a study would be a durability assessment tool that assists engineers in e.g. mitigating ineffective repair solutions for existing structures, or effectively motivate modifications of reinforced concrete design codes requirements for durability.

1 A Brief historical view

For a quite long time, simulations have been in use to model transport phenomena within the concrete materials research community [1]. At the start was the chloride ingress modelling as its transport is related to the steel reinforcement corrosion. A simple Fick-type diffusion was used in [2] shown below, where diffusion was the only mechanism that had been accounted for:

$$\frac{\partial c}{\partial t} - D^* \frac{\partial^2 c}{\partial x^2} = 0 \quad (1)$$

Here $c(x,t)$ is the concentration field and D^* is the diffusion coefficient. Fick's diffusion model was used afterwards for a long time until it has been then realised that the predictions of Fick's model were not in agreement with the measured chloride profiles due to the exposure of the samples to intermittent wetting/ drying cycles. A careful study [6] assessed the validity of Fick's law and concluded that Fick's model is based on assumptions that do not reflect the behaviour of the cementitious materials and hence the model lacks reliability e.g. it does not account for chemical interactions between the solid phases and the pore solution. A more sophisticated advection-diffusion with reaction model is used for simulating reactive transport processes. There is a variety of approaches using conventional numerical methods like [7] or hybrid methods combining e.g. FDM and LBM as in [11]. Recently Lattice Boltzmann models (LBM) have demonstrated their suitability for such problems due to its data locality implying parallel scalability, its computational efficiency and its conservation properties for flows in complex geometries such as porous media as well as its ability to simulate multiphase and multicomponent flows [9]. A twofold homogeneous heterogeneous approach for simulating pore scale reactive flow was proposed in [8] and Ion exchange model coupled with LBM is proposed in the work of [5].

2 Theory and numerical model

2.1 The physicochemical model

The physicochemical model incorporates the governing equations for fluid flow, mass transport namely, Navier-Stokes equations, the advection-diffusion equation, and dissolution/ precipitation heterogeneous reaction. The dimensionless form of the model is given by

$$\nabla \cdot \mathbf{u} = 0 \quad (2)$$

$$\frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot (\mathbf{u}\mathbf{u}) = -\nabla p + \frac{1}{Re} \nabla^2 \mathbf{u} + \frac{1}{Fr} \mathbf{g} \quad (3)$$

$$\frac{\partial c^j}{\partial t} + \mathbf{u} \cdot \nabla c^j = \frac{1}{Pe} \nabla^2 c^j + R \quad (4)$$

$$\mathbf{n} \cdot (\nabla c^j)|_{\text{Interface}} = Da \cdot c^j \quad (5)$$

Where \mathbf{u} is the velocity vector field, c^j is the concentration field of the j^{th} species and R represents the sink-source term due to the chemical reactions for the j^{th} species. The non-dimensional parameters used include the Reynolds number, $Re = \frac{UL}{\nu}$, the Froude number, $Fr = \frac{U^2}{gL}$, the Peclet number $Pe = \frac{UL}{D}$, and the Damköhler number $Da = \frac{kL}{D}$. Where U and L is a characteristic fluid flow velocity and characteristic length respectively, ν is the kinematic viscosity, g is the gravitational acceleration, D is the diffusion coefficient and k is the reaction rate. These four independent non-dimensional numbers characterize the flow, solute transport and chemical reaction.

2.2 Lattice Boltzmann Method

Lattice Boltzmann Methods can be described as a family of numerical schemes based on kinetic theory used for solving a general class of advection-diffusion partial differential equations. This is done by solving Boltzmann-type equations with discrete velocity stencils instead of discretising and solving the original transport PDE directly as done in most CFD methods. In LBM, the PDEs' degrees of freedom are represented in terms of a single-particle density distribution function $f_i(\mathbf{r}, t)$, which describes the mass density of particles moving with a velocity \mathbf{e}_i at some position \mathbf{r} and time t . The observable quantities e.g., the hydrodynamic fields in case of solving for flow are the moments of the distribution. The discretised density distribution is transported over a regular spatial lattice. The basic evolution ansatz for LBM reads

$$f_i(\mathbf{r} + \mathbf{e}_i \Delta t, t + \Delta t) = f_i(\mathbf{r}, t) + \Omega_i^{BGK/MRT/CUM}(\mathbf{r}, t) \quad (6)$$

Where \mathbf{r} is the position vector of a given grid node of a Cartesian Lattice and Δt is the time step. The complexity and accuracy of the so-called collision term Ω appearing in the above equation strongly depends on the use of either a single (as in the so-called BGK model) or multiple relaxation times (MRT and the so-called cumulant approach [4]). All the components of the distribution functions or moments are typically relaxed towards an equilibrium distribution and the hydrodynamic (i.e. macroscopic) quantities are obtained as algebraic moments of the spatiotemporal distribution function. For simulating the fluid flow, we are using the Cumulant LBM, which by relaxing cumulants instead of raw or central moments overcomes some problems such as the lack of Galilean invariance [4]. A model with seven microscopic velocities proposed by [10] is used for solving the advection-diffusion equation.

3 Results

The cumulant LBM is benchmarked in [4] and our results for the permeability of a porous medium based on a μ -CT geometry reproduces a value within the range of previous numerical and experimental studies. As an additional preliminary validation study, the MRT-LBM [10] is used to solve what is known as the Taylor-Aris dispersion problem, where the dispersion of a concentration ϕ is considered under the background flow between two parallel plates. In the long time regime, the section-averaged distribution of the concentration is supposed to have an approximate Gaussian distribution irrespective of the initial condition. A coefficient that characterizes the solution called the longitudinal dispersion coefficient D^L measures the temporal growth of the variance of the section-averaged concentration. A set up for the test is conducted, D_n^L is computed and compared to its analytical counterpart D_a^L . A very good agreement found between both for different Pe numbers as observed in Table 1.

4 Conclusion

The objective of this work is to develop a state-of-the-art model for multicomponent reactive flow in cementitious materials. The next step is the implementation of a mass-conserving scheme to take into account the surface dynamics due to chemical reaction. Differently from previous approaches dealing with surface reaction, we are aiming at explicitly capturing a sharp interface while at the same time maintaining local mass conservation. To the best of our knowledge

Pe	$\frac{D_n^L}{D}$	$\frac{D_a^L}{D}$	Error(%)
25	4.018	3.976	1.053
50	13.154	12.905	1.929

Table 1. Taylor-Aris dispersion

previous schemes are either pixel/voxel-based implying first order accuracy or lacking mass conservation.

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References

- [1] M. Alexander, A. Bertron, and N. De Belie. *Performance of Cement-Based Materials in Aggressive Aqueous Environments: State-of-the-Art Report, RILEM TC 211 - PAE*. RILEM State-of-the-Art Reports. Springer Netherlands, 2012.
- [2] M. Collepardi, A. Marcialis, and R. Turriziani. Penetration of Chloride Ions into Cement Pastes and Concretes. *Journal of the American Ceramic Society*, 55(10):534–535, 1972.
- [3] Y. Farnam, S. Dick, A. Wiese, J. Davis, D. Bentz, and J. Weiss. The influence of calcium chloride deicing salt on phase changes and damage development in cementitious materials. *Cement and Concrete Composites*, 64:1–15, 2015.
- [4] M. Geier, M. Schönherr, A. Pasquali, and M. Krafczyk. The cumulant lattice Boltzmann equation in three dimensions: Theory and validation. *Computers & Mathematics with Applications*, 70(4):507–547, 2015.
- [5] A. Hiorth, E. Jetttestuen, L. M. Cathles, and M. V. Madland. Precipitation, dissolution, and ion exchange processes coupled with a lattice Boltzmann advection diffusion solver. *Geochimica et Cosmochimica Acta*, 104:99–110, 2013.
- [6] J. Marchand and E. Samson. Predicting the service-life of concrete structures – Limitations of simplified models. *Cement and Concrete Composites*, 31(8):515–521, 2009.
- [7] S. Molins, D. Trebotich, C. I. Steefel, and C. Shen. An investigation of the effect of pore scale flow on average geochemical reaction rates using direct numerical simulation. *Water Resources Research*, 48(3), 2012.
- [8] R. A. Patel, J. Perko, D. Jacques, G. D. Schutter, K. V. Breugel, and G. Ye. A versatile pore-scale multicomponent reactive transport approach based on lattice Boltzmann method: Application to portlandite dissolution. *Physics and Chemistry of the Earth, Parts A/B/C*, 70:127–137, 2014.
- [9] X. Yang, Y. Mehmani, W. A. Perkins, A. Pasquali, M. Schönherr, K. Kim, M. Perego, M. L. Parks, N. Trask, M. T. Balhoff, M. C. Richmond, M. Geier, M. Krafczyk, L.-S. Luo, A. M. Tartakovsky, and T. D. Scheibe. Intercomparison of 3D pore-scale flow and solute transport simulation methods. *Adv. Water Resour.*, 95:176–189, 2016.
- [10] H. Yoshida and M. Nagaoka. Multiple-relaxation-time lattice Boltzmann model for the convection and anisotropic diffusion equation. *Journal of Computational Physics*, 229(20):7774–7795, 2010.
- [11] D. Yu and A. J. C. Ladd. A numerical simulation method for dissolution in porous and fractured media. *Journal of Computational Physics*, 229(18):6450–6465, 2010.