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Multiscale lattice Boltzmann-finite element modelling of chloride diffusivity in cementitious materials. Part I: Algorithms and implementation

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ABSTRACT

Chloride diffusivity in cementitious materials depends on both the environmental conditions and the evolution of their underlying microstructures over a wide range of length scales. Part I of this two-part investigation presents the algorithms and implementation of a hybrid lattice Boltzmann-finite element method that combines the advantages of lattice Boltzmann method and finite element method to estimate the chloride diffusivity in cementitious materials. Lattice Boltzmann method is used as micro-scale solver to predict the time-dependent chloride diffusivity in cement paste and interfacial transition zone (ITZ), the microstructures of which are generated from the HYMOSTRUC3D model. Finite element method is selected as meso-scale solver for estimating the chloride diffusivity in mortar and concrete, which are modelled as three-phase composites consisting of aggregate, matrix and ITZ, respectively. The upscaling between the micro-scale and meso-scale simulations is accomplished by using the volume averaging technique. The representative elementary volume (REV) of cementitious materials at a lower scale is determined with a numerical-statistical approach. Chloride diffusivity in the REV of cementitious materials at a lower scale is considered as input to predict the chloride diffusivity in cementitious materials at a higher scale. The developed multiscale lattice Boltzmann-finite element modelling scheme enables to acquire a meso-scale solution, i.e. chloride diffusivity, while still capturing the micro-scale information. The simulation results and validation are presented in detail in Part II.

1. Introduction

The durability of cementitious materials has been a major concern due to the premature failure and serviceability issues of many reinforced concrete structures. Durability of cementitious materials is to a large extent dependent on their resistance to the ingress of aggressive species, such as chloride, sulfates and carbon dioxide. For example, chloride-induced corrosion of reinforcing steel is one of the main causes of deterioration of reinforced concrete structures. The time to corrosion initiation of steel is considered dominant and usually defined as the service life, which depends on the ingress of chloride ions into concrete. In such a case, chloride diffusivity is usually regarded as an indicator to evaluate the durability and predict the service life of reinforced concrete structures. In principle, an ideal model for chloride diffusivity should be based on direct measurements of the microstructure, especially the pore structure of cementitious materials (Garboczi and Bentz, 1998). Precise prediction of chloride diffusivity is still a great challenge, since the underlying microstructure of cementitious materials is extremely heterogeneous, complex and generally ranges over a wide length scales.

In order to solve such multiscale problems, it is desirable to develop a multiscale method that can directly capture the fine-scale features. Over the past decades, a number of multiscale methods, such as homogenous multiscale method, heterogeneous multiscale method, variational multiscale method, multiscale finite element method and multiscale finite volume method have been proposed and developed to investigate the mechanical behaviour and transport properties in porous materials (Weinan and Engquist, 2003; Hughes et al., 1998; Hou and Wu, 1997; Jenny et al., 2003). A brief overview of the existing multiscale methods is given in Zhang (2013). Because of its simplicity and easy implementation, homogeneous multiscale methods are widely applied to cementitious materials. For example, Bentz et al. (1998) proposed a simulation

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procedure to predict the chloride diffusivity in saturated concrete based on the virtual concrete microstructure. At micro-scale, the chloride diffusivities in cement paste and interfacial transition zone (ITZ) were estimated using an empirical relationship between chloride diffusivity and porosity. The chloride diffusivity obtained in micro-scale was directly used to calculate the chloride diffusivities in mortar and concrete. Kamali-Bernard et al. (2009) developed a numerical tool to predict the effective diffusivity of tritiated water in saturated mortar. A finite element code (ABAQUS) and the inverse method were applied to determine micro-scale solutions, i.e. the effective diffusivity of tritiated water in cement paste and that in the ITZ, respectively. The micro-scale solutions were directly regarded as input data to predict the effective diffusivity in mortar by using ABAQUS. The interaction between the micro and meso-levels was not considered.

The main purpose of this study is to develop a multiscale modelling scheme, named lattice Boltzmann-finite element method, to estimate the chloride diffusivity in cementitious materials capturing the changing structural information at each scale. Lattice Boltzmann method is used as micro-scale solver to estimate the chloride diffusivities in cement paste and ITZ, the results of which are regarded as input to predict the chloride diffusivities in mortar and concrete by using finite element method. Volume averaging approach is applied to bridge the gap between micro-scale and meso-scale. The representative elementary volume (REV) size is determined with a numerical-statistical approach. The current paper gives a detailed description of the algorithms and implementation of the multiscale modelling scheme.

2. Chloride diffusion in cementitious materials

Diffusion of ions is generally defined as their movement due to the concentration gradient, or more strictly speaking, chemical potential, from regions of higher concentration to regions of lower concentration of the diffusing substance. The movement of ions in the pore solution (in one dimension) can be described as (Tang, 1996):

$$J = -BC\frac{\partial \mu}{\partial x} = -BRT\left(1+\frac{\partial \ln \gamma}{\partial \ln C}\right)\frac{\partial C}{\partial x} = -D_0\left(1+\frac{\partial \ln \gamma}{\partial \ln C}\right)\frac{\partial C}{\partial x} = -D\frac{\partial C}{\partial x} \quad (1)$$

where B is the proportionality factor and assumed to be constant (Tang, 1996), C is the concentration of species in solution, μ is chemical potential, R is the universal gas constant (J mol^{-1} k^{-1}), T is the absolute temperature, the term $D_0 = BRT$ is a constant for a given temperature, γ is the activity coefficient that is observed to be a complex function of the ionic concentrations in solution (Bockris et al., 2002; Zhang and Gjørv, 2005), and D stands for the diffusion coefficient. If diffusion solely occurs in a pore solution, the diffusion coefficient D only depends on the type of diffusing species and their concentrations. Under this condition, the diffusion coefficient is generally termed the self-diffusion coefficient (D_p). However, when passing through a porous medium, like concrete, the diffusion process is not only affected by the pore structure of concrete but also influenced by the presence of other types of species in the pore solution. If the diffusing ions do not interact physically or chemically with hydration products, the resulting diffusion coefficient is usually referred to as the effective diffusion coefficient D_e, which is a material characteristic describing the ability of transfer for a given molecule or ion driven by a concentration gradient. The variation of concentration within a unit volume can be described by Fick's second law:

$$\frac{\partial C}{\partial t} = -\nabla \cdot J \quad (2)$$

With respect to chloride ions, they are found in cementitious materials in two forms. One is bound chloride and the other is free chloride. Bound chlorides are either the result of chemical binding, i.e. chloride ions are chemically bound by reacting with the cement hydrates or physical binding, i.e. chloride ions are physically adsorbed to the cement gel. The total chloride content in cementitious material is a sum of free and bound chlorides. It is only the free chlorides that can dissolve in the pore solution and diffuse in cementitious materials through the pore solution, reach the surface of steel reinforcement and induce the corrosion process. Therefore, the corrosion of steel is related to the free chlorides. It is better to describe the chloride diffusion in cementitious materials only in terms of free chloride concentration. The total chloride concentration C_t is the summation of free chloride concentration C_f and bound chloride concentration C_b as $C_t = C_f + C_b$. The chloride ions transfer equation (in one dimension) can be written as follows:

$$\frac{\partial C_t}{\partial t} = \frac{\partial C_f}{\partial t} + \frac{\partial C_b}{\partial t} = \left(1+\frac{\partial C_b}{\partial C_f}\right)\frac{\partial C_f}{\partial t} \quad (3)$$

By incorporating Eqs. (1) and (3) into Eq. (2), the governing equation of transient diffusion of chloride ions into cementitious materials can be rewritten as follows:

$$\frac{\partial C_f}{\partial t} = \frac{D_e}{(1+(\partial C_b/\partial C_f))}\frac{\partial^2 C_f}{\partial x^2} = D_a\frac{\partial^2 C_f}{\partial x^2} \quad (4)$$

where D_a denotes the apparent chloride diffusion coefficient that is a function of the effective chloride diffusion coefficient D_e and chloride binding capacity $\partial C_b/\partial C_f$. $\partial C_b/\partial C_f$ represents the influence of chloride binding on the chloride diffusion coefficient and varies greatly with the free chloride concentration C_f. Therefore, the apparent chloride diffusion coefficient D_a should not be constant but relates to the effective chloride diffusion coefficient D_e and the free chloride concentration C_f, both of which depend upon the microstructure of cementitious materials affected by factors including w/c ratio and age, and upon the environmental conditions including temperature and degree of water saturation.

3. Definition of scales

The heterogeneity of cementitious materials manifests itself at different length scales. In order to develop an appropriate multiscale modelling scheme for fluid transport in cementitious materials, it is essential to consider the spatial scales involved and how the physical processes and characteristics of the system associate with the corresponding scale. A careful definition of relevant length scales can clarify any investigation of scale considerations, although such definitions are a matter of choice and modelling approach. In general, the length scale of interest in cementitious system may vary from a nano-level in the order of 10^{-9} m to a macro-level on the order of 10^{0} m for some regional applications. A scale hierarchy related to fluid transport in cementitious materials is defined, which consists of five elementary levels, i.e. nano-scale, sub-micro-scale, micro-scale (pore-scale), meso-scale and macro-scale.

The level of a characteristic length scale of 10^{-9} m is the smallest scale of a physical representation of the complex microstructure of cementitious materials (Ulm et al., 2004). It is the scale of gel pores in the calcium silicate hydrate (C-S-H) solid gel formed at the early ages by the hydration of two calcium silicates (C_3S and C_2S) in cement. This scale is referred to as nano-scale. Jennings (2000) provided a qualitative and quantitative evidence of the microstructure