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Modelling the diffusivity of mortar and concrete using a three-dimensional mesostructure with several aggregate shapes

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ABSTRACT

This paper presents a numerical investigation into the effect of ITZ and aggregate shape on the diffusivity of mortar and concrete using a three-dimensional model. Concrete is treated as a three-phase composite consisting of aggregate particles, bulk cement paste and aggregate–paste interface, i.e. the 'interfacial transition zone' (ITZ). The model is set up in two stages. First, a three-dimensional representative volume element of the concrete mesostructure is generated. Then, a finite difference method is used to simulate molecular diffusion through the mesostructure. The transport properties of the conductive phases (bulk cement paste and ITZ) are determined based on the water/cement ratio, degree of hydration and porosity gradients away from aggregate particles. The model is validated against available experimental data and compared with analytical relationships for ideal cases. The model is then used to study the effect of aggregate shape on diffusivity, which has not been attempted before in three-dimensions. The model is also applied to assess the effects of water/cement ratio, degree of hydration, aggregate size, volume fraction, shape and orientation, ITZ width and percolation on diffusivity. Some of these effects are impractical to quantify from laboratory experimentation alone. It was found that the shape and orientation of aggregate particles have a significant effect on diffusivity. Diffusivity decreased when spherical aggregate particles are replaced with ellipsoidal particles due to the consequent increase in tortuosity of the cement paste.

1. Introduction

Diffusion is one of the most important transport processes influencing the durability of cement-based materials. Therefore, the ability to estimate the diffusivity of concrete based on mixture proportions and microstructure is attractive, as it would assist in the development of service life prediction models and durability-based design codes.

Lab based transport testing has been carried out for many years to gain a better understanding of different variables influencing the diffusivity of cement-based materials. Since the microstructure of concrete is highly complex and its transport properties are influenced by many interacting parameters, many experiments should be carried out in order to identify the effect of different variables. However, it is often difficult to isolate the effects of specific variables because other influencing parameters inevitably vary. While some of these effects can be reduced or avoided entirely, others are difficult if not impossible to control, and so must be accounted for when analysing results. Moreover, experiments are time-consuming and expensive.

Thus, it would be extremely useful to be able to predict diffusivity based on the microstructure of concrete, either using an analytical or numerical model. However, existing analytical models are restricted to simplified microstructures, e.g. spherical aggregate particles dispersed in a medium [1,2], so they are likely to be inaccurate. The tremendous increase in computational capabilities has strongly favoured the development of numerical simulations based on more precise description of the microstructure. Bentz et al. [3] used random walk simulations to study the diffusivity of concrete. Concrete is assumed to be a composite material consisting of aggregate particles, ITZ and bulk cement paste, and all three phases are treated as uniform continuum materials. Kamali-Bernard et al. [4,5] computed the diffusivity of mortars from a digitized mesostructure by applying Fick's law. The mortar is assumed as a composite, in which the transport properties of the conductive phases (bulk cement paste and ITZ) are constants obtained by fitting experimental results. Zheng et al. [6] proposed a three-phase composite sphere model to calculate the steady-state chloride diffusivity of concrete. However, the simulations were carried out using a composite sphere model and may not be sufficient to capture the effects of ITZ percolation and tortuosity due to multi-sized aggregate particles.

Representing aggregate particles as spheres in a model is probably a gross simplification since the aggregate shape can affect the

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properties of concrete. Most modelling works published to date have been carried out assuming spherical aggregate particles. Recently, some researchers have attempted to model the influence of aggregate shape on the transport properties of cement-based materials. However, the simulations were carried out in two dimensions and the findings appear to be inconclusive [7,8]. Zheng et al. [7] used a two dimensional lattice model to investigate the effect of elliptical aggregate shapes on transport properties. They found that when the aspect ratio of elliptical aggregate particles increases, the chloride diffusivity in concrete decreases for a given chloride diffusivity ratio of ITZ to cement paste. Li et al. [8] used the finite element method to investigate chloride diffusion in concrete. The concrete is treated as a two-dimensional material composed of only cement paste and aggregate particles. They found that the shape of aggregate has a small influence on the diffusion coefficient of chlorides in concrete. To the authors' knowledge, the effect of general aggregate shape on the transport properties of mortar and concrete has not been investigated in three-dimensions.

This paper presents a numerical investigation into factors influencing the diffusivity of mortars and concretes. This approach is particularly useful because a large number of related factors can potentially influence diffusivity. Many laboratory experiments would be required in order to understand their significance and interactions, and even so, may not be able to isolate all of these effects. For example, it is impossible to change the width of the ITZ and examine its effect on transport independent of other variables in a laboratory experiment. Increasing the volume fraction of ITZ (for example by changing the aggregate shape or particle size distribution) causes densification of the bulk cement paste and may change the tortuosity of the cement paste. These have different effects on transport properties, but cannot be isolated in experimental studies.

In order to investigate the effect of aggregate shape, aggregate particles are modelled as ellipsoids with varying aspect ratios, thus providing a dynamic range of shapes from spheres to tri-axial ellipsoidal, prolate and oblate spheroidal to resemble elongated or flaky aggregate shapes. The developed model is regarded as interactive because the transport properties of the ITZ and bulk paste are estimated from their porosities, which depend on the ITZ characteristics (width, porosity gradient), water/cement ratio and degree of hydration. The total porosity of the cement paste (ITZ + bulk paste) is set to be equal to the porosity of a cement paste of the same water to cement (w/c) ratio and hydration degree. A realistic aggregate size distribution is used as an input to the model. The model was validated against available theoretical and experimental results and was then applied to evaluate the effects of aggregate shape, content, size and orientation, water/cement ratio, degree of hydration, ITZ width and percolation on diffusivity.

2. Approach

To model transport phenomena in any material, an input structure coupled with a transport algorithm is required. Therefore, our approach consists of the following steps. First, a three-dimensional mesostructure is generated where concrete is idealised as a composite of aggregate particles, bulk cement paste and ITZ. An algorithm is utilised to convert the mesostructure into a corresponding mesh based on voxel analysis. The model may also include other phases such as air voids and cracks. The transport property of each phase is then defined according to its pore volume fraction. The ITZ is not assumed to be a single shell of uniform property, instead the porosity and diffusivity of the ITZ are allowed to vary with distance from the aggregate surface and with the progress of hydration. After a desired hydration degree is achieved, the capillary porosity is calculated as a function of distance from the nearest aggregate surface and is converted into diffusivity using a previously derived relationship [9]. Then the ITZ diffusivity is averaged over its thickness. A finite difference scheme is applied to the entire mesostructure to simulate bulk diffusion at steady-state conditions. This approach is advantageous because a fairly representative description of the microstructure is used as an input. Since the approach is applied to a digitised mesostructure, it serves as a platform for applying real images of microstructure as inputs in the future, and other transport properties such as pressure-induced water flow and capillary absorption of water can be simulated using the appropriate lattice methods. The finite difference technique requires less computational resource compared to finite element methods, thus investigation at higher discretisation is possible which may lead to improved accuracy.

3. Generation of the model

3.1. Mesostructure

To generate the mesostructure of a mortar or concrete, the aggregate size distribution is first determined either experimentally in a conventional sieve analysis or generated from a theoretical gradation such as Fuller-Thompson [10]. The smallest aggregate size considered was 0.15 mm. Aggregate particles within each sieve class are substituted with identical spheres at the required volume fraction, and placed randomly in the computational cube with the largest particles first with no overlaps. A periodic boundary condition is applied to all sides of the computational cube. In doing so, aggregate particles that extend beyond the cube are completed (wrapped) into the opposite face of the cube. For a cube of 7.5 mm containing 30% volume of aggregate particles ranging from 0.15 to 2 mm (Fig. 1), around eight thousand aggregate particles were required. Voxels representing the cement paste matrix are classified as either ITZ or bulk paste depending on their location relative to the nearest aggregate particle. A large number of voxels are required to realistically represent the mesostructure, but this comes at higher computational cost. A suitable sample size and resolution to obtain representative results will be examined later in this paper.

3.2. Non-spherical aggregate particles

Non-spherical aggregate particles are incorporated into the model to examine the effect of aggregate shape on transport properties. For this study, a range of ellipsoidal particles will be used. An ellipsoid is defined by the following equation:

$$(\boldsymbol{x}-\boldsymbol{v})^t A^{-1}(\boldsymbol{x}-\boldsymbol{v}) = 1 \quad (1)$$

where v is a vector representing the centre of the ellipsoid and A is a positive definite matrix with eigenvectors which are the principal directions of the ellipsoid and eigenvalues which are the squares of the semi-axes: a^2, b^2 and c^2.

The mesostructure is projected into a regular lattice, so collision and overlap detection is a straightforward exercise of finding out whether two particles occupy the same site(s), rather than having to compute and test intersections between any two particles, which is usually very expensive for non-spherical particles [11]. By varying the ratio a:b:c, four distinctive shapes ranging from spheres to elongated and disc shaped particles can be obtained. Table 1 shows the types of ellipsoids that will be used to represent aggregate particles in this study. Note that the ratios a:b:c were selected such that the particle volume (=4/3 πabc) remains constant. Fig. 1 shows examples of randomly generated mesostructure containing 30% volume of spherical and non-spherical aggregate parti-