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Computational investigation of pore permeability and connectivity from transmission X-ray microscope images of a cement paste specimen

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HIGHLIGHTS

- Generate 3D digital samples with nanoscale TXM images from a microscale cement paste sample.
- Explain the governing equations and numerical scheme for permeability calculation.
- Apply permeability solver to compute pore permeability and connectivity with digital samples.
- Compare the calculated permeability with analytical values from Kat–Thompson equation.

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ABSTRACT

This study applied the Transmission X-ray Microscope (TXM) characterization techniques and permeability-solver computational program to investigate the transport properties of a microscale cement paste specimen. The TXM techniques allow fast-image acquisition of pore microstructure at a resolution of 30 nm. The microscale cement paste specimen with 0.45 water/cement ratio was specially prepared with a capillary tube. The pore microstructure of the microscale paste specimen was characterized by using the Advanced Photon Source at the Argonne National Lab. The image processing technique was conducted to identify the pore distribution in the captured images. The digital samples with different porosities were generated to compute the transport properties. The burning algorithm was employed to estimate the pore connectivity. The finite difference method with artificial compressibility relaxation algorithm was applied to simulate water transport in capillary pores with Stokes equation. The pore permeability was computed with the calculated average flow velocity using Darcy's Law. The computed permeability results of digital samples were also compared with the predicted values from the Katz–Thompson equation to demonstrate the computational accuracy.

1. Introduction

The multiscale transport properties of cement paste affect the concrete durability since the penetrated solution will reduce the resistance of chemical attacks. Based on former research [1–3], the permeability of cementitious materials relies on the microstructure of the network, including total porosity, pore size distribution and connectivity. Thus, it is essential to understand the relationship between pore transport properties and microstructure characteristics.

The microstructure of cement paste is heterogeneous and consists of multi-phases. Unhydrated cement particles, capillary pores and various hydration products including Calcium silicate hydrate (C–S–H), Calcium hydroxide (C–H) and Calcium sulfoaluminates (Etringite and Monosulfoaluminate) are randomly distributed. Three types of voids are presented in the hydrated cement paste: interlayer space (gel pores), capillary pores and air voids. Interlayer space that is between C–S–H layers is usually less than 10 nm, thus its impact on permeability is negligible. Capillary pores (typically ranging from 10 nm to 50 μm), as illustrated in one scanning electron microscope image of cement mortar in Fig. 1, affect the permeability of cement paste since they form interconnected networks through which bulk water flow occurs. Mehta and Monteiro [4] show that capillary porosity is directly proportional to the water–cement ratio and inversely proportional to the degree of hydration. An exponential drop in the permeability was observed when the volume fraction of capillary pores decreased from 0.4 to 0.3 [5]. Air voids are typically ranging from 50 μm to

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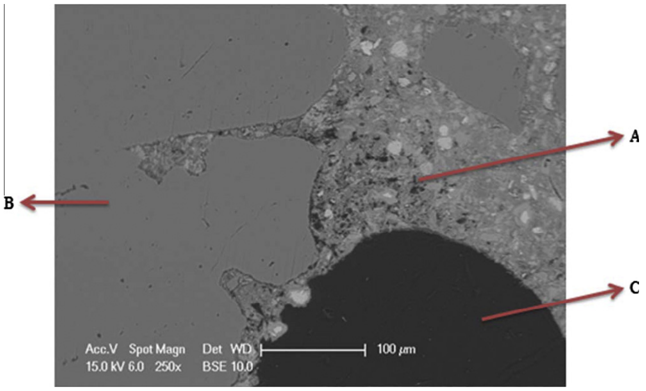

Fig. 1. Demonstration of (A) capillary pore, (B) fine aggregates and (C) air void in one Scanning electron microscope (SEM) image of a cement paste sample.

3 mm as shown in Fig. 1. The trapped air voids can be controlled by using air-entraining admixture for reducing freeze–thaw damage [6].

Although it is complex to precisely measure the porosity of cement paste, three methods were used to estimate the value [7]. They are gas adsorption, mercury intrusion porosimetry (MIP) and direct observation techniques including optical microscope and scanning electron microscope (SEM). Gas adsorption measures the amount of condensing vapor adsorbed by accessible pores within the specimen. However, it can only provide accurate measurements of gel pores (which has little impact on transport phenomenon), not capillary pores. MIP forces mercury into specimen with increasing pressure. It has been demonstrated to be very helpful to measure the pore size distribution of the general size range that believed to control permeability [8,9]. The direct observation techniques of porosity on polished thin sections are commonly used. With the development of new imaging technology, X-ray computed tomography was used to study the pore structure in concrete [10].

Another essential aspect of transport study is to measure the permeability of cement paste. A number of techniques has been developed to experimentally measure the permeability of harden concrete, including direct and indirect methods, which were summarized by former researchers [7,11]. Two direct standard methods were recommended by two institutions to measure the permeability [12,13]. The theoretical foundation of these methods is Darcy's Law of flow. To calculate the intrinsic permeability, a pressure gradient can be established over a specimen and the amount of water through which is measured. For the indirect method, the rapid chloride permeability test [14] measures the electrical conductance of concrete sample. It is based on the principle that the electrical current is conducted by charged ions, which are chloride ions in this case, penetrating through the sample. Thus higher current is associated with higher permeability. Recent research shows that electrical impedance spectroscopy can yield an accurate measurement of the true conductivity of concrete specimen [15]. The electrical impedance method was applied to pervious concrete for studying the relationship between pore permeability and aggregate sizes [16–19].

Besides the experimental measurement, the computational analysis of permeability was also attempted by researchers. The basic idea is to solve Stokes equation for the average velocity, and then use the average velocity to calculate permeability based on Darcy's Law. Martys et al. [20] used this method to simulate random packing of spheres and obtained universal curves for permeability. The permeability solver code [21,22] developed at NIST was applied in this research. In this program, a hydraulic pressure was applied as the initial input and the periodic boundary conditions were used. The flow condition in the pore system was considered to be incompressible and steady-state based on computational fluid mechanics. The finite-difference method was applied on image pixels with the artificial compressibility relaxation algorithm to solve Stokes equation. Once the solutions of the average velocity at four different depths converge to a close value, the permeability can be obtained from Darcy equation based on that velocity. By making a comparison to the existing direct measurement of permeability made by other researchers [17,23,24], the accuracy of the code was validated by giving an error of less than 2%. Later on, 3D virtual pervious concrete microstructures generated by the correlation filter reconstruction algorithm were developed. And their permeability and connectivity were computed by the permeability solver code to demonstrate the validation of the reconstruction algorithm [25,26].

Some theoretical models were proposed for the relationship between porosity and permeability. Empirical models including the Darcy–Poiseuille model, the Archie model and the Carman–Kozeny models were first put forward [7,27]. Although these models attempt to connect permeability to some indicators like total porosity, they failed to consider the network of pore structure, thus their predictions were considered to be inadequate [7]. To overcome this, the percolation theory and model were introduced to provide more information about the physical properties. The percolation theory [28] quantifies the pore connectivity in a porous medium by considering the porous medium as a lattice and assuming that each site in the lattice is occupied randomly with a particular probability. The occupation of each site is totally independent. The "site-bond percolation" was considered to be most close to the nature of flow through porous media [28]. The function between phase percolation and cement hydration was studied using a 3D digital image-based simulation. And a universal curve of percolation vs. porosity was plotted for different water–cement ratios and degree of hydration [29,30].

2. Objective and scope

This study is aim to numerically calculate transport properties of a 3D digital cement paste sample generated from the TXM image and investigate the relationship between the permeability, pore connectivity and the porosity in cement pastes. This paper includes three connected work:

1. Prepare a microscale cement paste specimen and conduct 3D TXM image acquisition.
2. Conduct image processing to identify capillary pore distribution and generate digital samples for transport simulation.
3. Apply burning algorithm and permeability solver to compute the pore connectivity and permeability of 3D digital samples and compare the computed permeability with predicted results from the Katz–Thompson (K–T) equation.

This study investigated the transport properties (connectivity and permeability) of a cement paste specimen. The relationship between pore porosity, connectivity and permeability was also demonstrated through computational analysis.

3. Cement paste specimen preparation and TXM image acquisition

3.1. Specimen preparation

The specimens were prepared with w/c ratios of 0.45 to generate more capillary pores, by following the ASTM C305 standard. The mixing process was performed manually since only a small