#### Construction and Building Materials 191 (2018) 193-205

Contents lists available at ScienceDirect

## **Construction and Building Materials**

journal homepage: www.elsevier.com/locate/conbuildmat

## Determination of water permeability for a moisture transport model with minimized batch effect

Zhidong Zhang<sup>\*,1</sup>, George Scherer

Department of Civil and Environmental Engineering, Princeton University, Princeton, NJ 08544, USA

#### HIGHLIGHTS

• Specimens for all methods were prepared from the same cement paste cylinder.

• Permeabilities determined by various methods were used in a moisture transport model.

• Permeability determined by beam bending can provide closer results to measured ones.

• Katz-Thompson and Kozeny-Carman equations largely overestimate permeability.

• The effect of choosing an appropriate method is more important than the batch effect.

#### ARTICLE INFO

Article history: Received 6 December 2017 Received in revised form 23 September 2018 Accepted 27 September 2018

Keywords: Cementitious materials Beam-bending Permeability Microstructural alteration Inverse analysis Solvent exchange

### ABSTRACT

Values of water permeability for cementitious materials reported in the literature show a large scatter. This is partially attributed to the fact that materials used in these studies are different. To eliminate the effects of cements, specimen preparation, curing conditions and other batch effects, this study employs a single cement paste to prepare all specimens for a variety of permeability determination methods, such as beam-bending, sorptivity, Katz–Thompson and Kozeny-Carman equations. Permeabilities determined by these methods are then used in a moisture transport model. Compared with the measured mass loss curves, we found that permeability determined by the beam-bending method is more suitable for the moisture transport model than the other methods. The difference results from the use of a saturated specimen in the beam-bending method, while specimens in the other methods are dried (or rewetted). As already shown in the literature, the microstructure of the dried or rewetted specimens is altered and different to the original microstructure of the water saturated specimens. In addition, we found that drying tests for the inverse analysis method must be done at high RHs (63% in this study) to reduce the effect of vapor diffusion on the determination of water permeability.

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#### 1. Introduction

The moisture transport properties of cementitious materials are essential for evaluating the durability of concrete structures, because the transport of aggressive agents (e.g., chloride ions) and gases ( $CO_2$  and  $O_2$ ) is related to the moisture state [1,2]. Moisture transport in partially saturated porous media, such as cementitious materials, is mainly governed by the transport of three phases: liquid water, water vapor, and dry air. Mainguy et al. showed that dry air makes a very small contribution to the mass of moisture transported and only causes fluctuating air pressure in the material [3]. This conclusion was also drawn by the asymptotic analysis performed by Coussy and Thiéry [4,5]. In addition, considering that the liquid phase remains incompressible and total gas pressure is constant, the mass balance equations of moisture transport can be represented by a single equation, including only liquid water and vapor [6,7]. Mainguy et al. [3] further simplified the model for specific conditions, by considering only liquid water transport and neglecting the vapor diffusion. They found that such a model can give results for simulating drying mass loss curves very similar to the multiphase model. Hence, the transport coefficient - water permeability - that governs the liquid water transport becomes extremely important. For a given material, the ideal situation to perform moisture transport simulations is that water permeability is experimentally determined and then used in the moisture transport model.







<sup>\*</sup> Corresponding author.

E-mail address: zhangzhi@ethz.ch (Z. Zhang).

<sup>&</sup>lt;sup>1</sup> Present address: Institute for Building Materials, ETH Zurich, 8093 Zurich, Switzerland.

In most studies, water permeability  $K_l$  (with a unit of m<sup>2</sup>) is considered as an intrinsic property of the porous material, meaning that  $K_l$  only depends on the microstructure and should be valid for the other fluids. That is why  $K_l$  is called "intrinsic" permeability in some studies (e.g., [3,7,8]). Nevertheless, "intrinsic" permeabilities measured by gases (oxygen and nitrogen [9]) and solvents (methanol [10] and isopropanol [11,12]) are often found to be greater than  $K_l$ . Reasons for this will be discussed later (see the Section 6.4). To avoid confusion, in this paper, the terminology of water permeability instead of intrinsic permeability is used hereafter.

For cementitious materials, the determination of water permeability is an active research field reported in a voluminous literature. As reviewed in the literature [13,8], the permeability can be determined either directly by experiments or indirectly by theoretical models based on other measured data. Conventional methods to measure  $K_1$  are classified as flow-through techniques, as they measure the flux under steady state conditions for fully saturated specimens, with the geometry of either truncated cones [14] or cylinders/disks [15-18,13,19,12]. During measurements, liquid water is pressurized at one face and the outflow at the opposite face is continuously measured until the flow reaches steady state, and then  $K_l$  can be calculated by using Darcy's law. In these methods, it may take a long time to reach steady state flow (e.g., several weeks) for low permeable materials. To reduce the measurement time, it was suggested to increase the applied pressure [19], but this may risk altering the structure of materials and increase the water leak at the interface between the specimen and the pressure cell. Instead of applying continuous constant pressure, pressure relaxation methods involve increasing pressure on one side and observing the decrease of pressure due to liquid being pushed to another side (e.g., [20]). Applying a constant water flow was also used in some studies (e.g., [13]). These methods are rapid but they still need high pressure and thus have the same problems as the other conventional flow-through methods.

Recent studies used hollow cylinders [21] which measured radial flow of water under applied pressure. The main advantage of this kind of method is that the total area through which fluids flow is much larger than the disc specimens; therefore, the measurements showed higher accuracy and repeatability [22,21]. On the other hand, the large area increases the risk of heterogeneity, which means that any cracks or areas having greater water flow can significantly change the results.

Indirect methods, requiring the other data to calculate  $K_{l}$ , are called poromechanical (dynamic pressurization) techniques which monitor the time-dependent deformation of a specimen induced by externally applied stress or temperature change. The dynamic pressurization (DP) is one of such methods, which keeps the specimen in a sealed vessel and suddenly increasing or decreasing the pressure [23,24]. The hollow cylinders were also used in the DP method to improve the measurement accuracy [22,25]. By alternatively pressurizing and depressurizing, the effect of air voids in the unsaturated specimen can be gradually removed [24]. The beambending (BB) method is another example [26–29]. The measured relaxation curve that is obtained when a certain strain is applied to a long and slender specimen includes both hydrodynamic and viscoelastic effects. Therefore,  $K_l$  can be determined by fitting the relaxation curve with a theoretical model. The deformation of a specimen can also be introduced by the thermal expansion [30– 33], in which the rate of thermal strain relaxation is used to determine  $K_l$  based on the fact that the liquid shows a greater expansion than the solid phase.

Measurements of  $K_l$  are very sensitive to saturation conditions since the fully saturated condition is not easy to achieve. The presence of air voids or entrapped air in non-fully saturated materials may have a great influence on the measured results of the poromechanical methods and cause long delays in reaching equilibrium in conventional methods [34]. To ensure the fully saturated condition, various approaches were used in the literature, such as curing the specimen in water/limewater [21,12], vacuum saturation [35] and pressurizing saturation [28,29]. The time needed to fully saturate a porous body increases with the square of its smallest dimension. For the direct methods, the thickness of a disc specimen ranges from 25 to 70 mm (see the review in [19]) depending the size of aggregates as El-Dieb and Hooton [16] suggested that the specimen thickness should be 3 times as large as the aggregate size and a recent study [36] even reported that the specimen needs to be about 10 times as thick as the aggregates; therefore, a specimen may be extremely difficult to saturate. By contrast, the specimen in BB measurements is much easier to saturate since the method is limited to paste and mortar, so the diameter of the cylinder can be smaller than the concrete specimens used in the conventional methods.

In addition to experimental measurements, the microstructural information (i.e., pore size distribution (PSD), porosity, tortuosity, and connectivity) is often used to calculate water permeability. A relationship that was first proposed by Kozeny in 1927 [37] and later modified by Carman [38,39] is commonly known as the Kozeny–Carman (KC) equation. This equation was developed after considering a porous material as an assembly of capillary tubes for which the Navier–Stokes equation can be applied. It yielded  $K_l$  as a function of the porosity, the specific surface, and the shape and tortuosity of channels. It has been found that the KC equation is approximately valid for sands but not for clays [40]. Wong et al. [41] adopted a modified KC equation incorporating tortuosity and constrictivity to predict the oxygen permeability for concrete and they concluded that this equation overestimated the permeability by about one order of magnitude.

The Katz-Thompson model [42] (KTI model) was initially developed based on the measured percolation radius and resistivity and has shown a good estimation of permeability for sedimentary rocks. In another version of this model, the resistivity factor was assessed by mercury intrusion data (KTII model) [43]. This can largely simplify the use of the original version of Katz-Thompson model. For cementitious materials (concretes and mortars), both KTI and KTII models were found to overestimate the permeability [44,45], which was believed due to damage induced by the mercury intrusion. A recent study by Zhou et al. [9] showed that KTII can provide similar results to the measured nitrogen gas permeability with the Klinkenberg correction, but it is about 2–4 orders of magnitude higher than water permeability. The overestimation of KC and KTII equations will be discussed in the Section 6.3.

Other indirect methods based on theoretical models are also reported in the literature. For instance, a practical method is to use measured sorptivity to assess  $K_{l}$  [12], because sorptivity measurements are much easier to perform than the above-mentioned permeability measurements. The first author of this paper introduced two methods to indirectly determine water permeability [8]. One is called "inverse analysis" that utilizes a numerical moisture transport model to back-calculate  $K_l$  based on the measured drying mass loss curve. The other one employs the measured diffusivity curve to fit  $K_l$  by a general expression including both liquid transport and vapor diffusion. One factor that may affect the results of theoretical models is the diffusion of water vapor, as this phase is neglected in some models, but our studies show that the neglect can underestimate mass transport at low RHs [8]. The influence of vapor diffusion on the determination of K<sub>1</sub> will be discussed in Section 6.2.

As stated above, various methods using different specimen geometries or theoretical models in the literature to determine  $K_l$  show that the reported permeability values have great dispersion, ranging from  $10^{-22}$  to  $10^{-17}$  m<sup>2</sup> for materials with the same water-to-cement ratio (see review in [8]). Even though researchers are

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