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An inter lab comparison of gas transport testing procedures: Oxygen permeability and oxygen diffusivity

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ABSTRACT

The transport properties of concrete are useful measurements as they can provide a potential indication of durability and can be used in the prediction of the service life of concrete elements. Gas permeability and diffusivity test methods are attractive due to the ease of testing, their non-destructive nature and their potential to correlate to in-field carbonation of reinforced concrete structures. Unfortunately, limited information is available on the variability and inter-laboratory variability of the test methods that assess gas transport. This paper presents results of a recent investigation on the repeatability/variability of an oxygen permeameter and of an oxygen diffusivity instrument in two testing laboratories. Generally, a lower variability was seen for the oxygen diffusivity measurements compared to oxygen permeability. Moreover, the results confirm the importance of several parameters such as the flow and pressure used in the test and the samples conditioning method (degree of saturation and percentage of mass variation). In order to obtain repeatable results from gas transport testing between labs, the same boundary conditions (pressure and flow) and the same sample conditioning procedures should be used.

1. Introduction

Transport properties can be used to assess the potential durability of concrete mixtures or to provide critical data that can be used in the prediction of service life [1]. It has been widely accepted that many of the deterioration processes in concrete structures are influenced by the ability of the material to limit the ingress of deleterious aqueous solutions (e.g., chlorides, sulfates, water, etc.) or gases (e.g., carbon dioxide) [1].

The measurement of a permeability coefficient is considered by many to have the potential to serve as a performance criterion that can be related to concrete durability. However, the low permeability of concrete complicates permeability testing. First, samples can require a long conditioning period, making the test lengthy. Second the structure of cementitious materials can change during the test. Moreover, for water permeability the testing phase itself can be challenging, since it requires high pressure or specific sample geometry [2]. Additionally, water might interact with the concrete causing self-healing mechanisms or further hydration for early-age samples [3].

Consequently, due to the ease of testing and the quasi non-destructive nature of gas permeability and gas diffusivity tests, many researchers have favored them for the evaluation of transport properties of cementitious materials [4]. Substantial efforts have been invested in the development of testing procedures for gas permeability [5–8] and gas diffusivity [9–11]. Both properties are determined based on the rate of gas flow through the sample. In the gas permeability test, the flow is generated by a pressure gradient. In the gas diffusivity case the flow is generated by a concentration gradient. While the basic concepts of each test are relatively straightforward, several questions arise that are related to the gas-flow models adopted for evaluating the results of each test method. First, the assumption of laminar flow conditions during gas permeability tests can be questioned [12]. In addition, complexity arises since three main diffusivity mechanisms (ordinary, Knudsen and surface diffusion) may all occur simultaneously in concrete [9,13].

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It is also generally known that transport properties are influenced by the degree of saturation (or alternatively moisture content) and the distribution of the moisture present in the porous network [14,15]. For gas transport, the relative humidity (or alternatively the degree of saturation) at which the samples are conditioned can be expected to influence both the prevalent gas transport mechanisms acting in the samples and the rate of gas transport [46].

At relative humidities greater than 80% the large pores (e.g., capillary pores, pores with radius larger than about 5 nm) contain vapor, except for the adsorbed water layers on the pore walls, while the smaller pores (gel pores, pores with radius smaller than about 5 nm [16]) are fluid-filled [12,17]. At these high relative humidities the gas paths appear to be disconnected. Since in these conditions the mean free path of the diffusing molecules is much shorter than the pore radius and a total gas pressure gradient is present, viscous flow is the primary transport mechanism [18]. Consequently, at high relative humidities (i.e., humidities greater than 80%) the concrete appears almost impermeable to gas transport [12].

At lower relative humidity (50–80% relative humidity range [16,19]) the gel pores begin to empty and the thickness of the layers of water molecules that cover the pores walls decreases [20–23] causing the mean free path of gas molecules to increase [13]. This substantially increases the pore connectivity. The contribution of non-viscous flow (ordinary and Knudsen diffusion) increases, since Knudsen diffusion occurs when the mean free path is relatively long compared to the characteristic length of the pores the molecules collide more frequently with the pore wall than with each other [15]. For concrete materials, Knudsen diffusion contributes primarily for pores smaller than 50 nm [13]. Ordinary diffusion occurs when the mean free path of gas molecules is relatively short compared to the pores size and it is typically shown in large pores [13,15].

As a result, at even lower relative humidity (RH less than 50%), concrete appears highly permeable to gas transport mainly due to the increase of the pore space available to gases and due to the increase of connectivity and to the reduced tortuosity of the gas path.

Moreover, at low relative humidity the contribution of surface diffusion also becomes more considerable. Surface diffusion occurs when gas molecules can diffuse along the pore walls. The adsorbed molecules move from one site to another through a desorption–absorption mechanism [15]. The mechanisms involved in surface diffusion are not fully understood and its contribution seems to become negligible in very large pores.

A schematic of the principal mechanisms involved in the gas transport for different water content is pictured in Fig. 1. It becomes clear from the previous considerations that the sample moisture conditioning process strongly influences gas permeability and diffusivity measurements [12,24–27].

2. Research objectives

This study is part of a large investigation on performance-based transport test methods for permeability evaluation. In this context, this paper presents results on the repeatability/variability of an oxygen permeameter and of an oxygen diffusivity instrument in two testing laboratories. The sensitivity of each instrument to different testing parameters is presented: including the applied pressure, the gas flow rate and the sample moisture content.

3. Testing procedure

3.1. Oxygen permeability test

The oxygen permeability test procedure describes a measurement made using a falling head permeameter [28–30]. Made popular by researchers from South Africa, this instrument has since been used by several researchers around the world. Fig. 2a shows a picture of the device with four test cells, while the details of the test cell are presented in Fig. 2b. The test involves the measurement of the steady unidirectional flow of gas through the specimen due to the presence of a pressure gradient between the upper (P_{atm}) and the lower side (P) of the sample. The lower surface of the sample is in contact with the oxygen contained in the vessel at a known pressure, while the upper surface is at atmospheric pressure. The gas flow is quantified by monitoring the pressure drop with time.

To perform the test, the sample is inserted in a rubber collar, placed in the upper cell and then compressed in order to ensure a good sealing of the sides. The inlet valves are opened and the gas is allowed to flow through the cylinder for a short period of time (i.e., seconds); the outlet valve is then closed and the pressure in the cylinder is allowed to reach approximately 100 kPa (relative pressure). Four samples are tested simultaneously.

The procedure followed in Lab 1 (Purdue University laboratory) used an automatic data logger that monitors the decay in pressure over 6 h (or until the pressure has dropped to 50 kPa) recording the pressure values every 30 s [29]. The procedure followed at Lab 2 (EMPA laboratory) monitored the pressure decay for 8 h and recorded the pressure every 5 min [28].

The oxygen permeability is evaluated using D'Arcy's law for the case of flow of compressible gas. The gas flow per cross sectional area is expressed as function of the pressure decay ($\partial P/\partial z$), of the coefficient of permeability (k_{SA}), and of the gravity acceleration (g) according to Eq. (1):

$$\frac{\partial m}{\partial t} = -\frac{k_{SA}}{g}\frac{\partial P}{\partial z} \tag{1}$$

where z is the spatial coordinate indicating the position across the sample in the direction of the gas flow. Eq. (1) can be combined with the ideal gas law to provide an expression for the evaluation of oxygen permeability (Eq. (2)) [30]:

$$k_{SA} = \frac{\omega V g d}{RAT}\frac{1}{t}\ln\left(\frac{P_0}{P}\right) \tag{2}$$

where ω is the molecular mass of oxygen, 0.032 kg/mol, V is the volume of oxygen under pressure (m^3), R is the universal gas constant, 8.314 (N m)/(K mol), A is the cross sectional area of the sample (m^2), d is the sample thickness (m), T is the absolute temperature, 293.15 K (20 ± 1 °C in Lab 2 and 23 ± 1 °C in Lab 1). The term $\ln(P_0/P)/t$ has been evaluated according to Alexander [29] considering a linear regression forced through the origin. The ordinary least square of the estimator ($\hat{m}$) of the slope of the linear regression will be as in Eq. (3):

$$\frac{1}{t}\ln\left(\frac{P_0}{P}\right) \cong \hat{m} = \frac{\sum_{i=1}^{n}\left[\ln\frac{P_0}{P_i}\right]^2}{\sum_{i=1}^{n}\left[\ln\frac{P_0}{P_i}\right]t_i} \tag{3}$$

where P_0 is the initial pressure and P_i the pressure at time t_i and n is the number of data points (i.e., pressure) being recorded.

Alexander et al. [28] proposed an oxygen permeability index (OPI), which is the logarithm of the average of the permeability values obtained from the four samples tested (Eq. (4)):

$$\mathrm{OPI} = -\log_{10}\left(\frac{\sum_{i=1}^{4}k_{SA\,i}}{4}\right) \tag{4}$$

where $k_{SA\,i}$ is the oxygen permeability of each sample evaluated through Eq. (2).

It is worth mentioning that the coefficient of permeability k_{SA} used for this study (Eq. (2)) is expressed in m/s while typical gas