Materials and Structures/Matériaux et Constructions, Vol. **32**, January-February 1999, pp 3-8

Gas permeability of concrete in relation to its degree of saturation

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Paper received: December 12, 1997; Paper accepted: April 2, 1998

ABSTRACT

Permeability, along with diffusion and absorption, is used to quantify durability characteristics of a concrete. The measured value of gas permeability of concrete depends strongly on its degree of saturation. Moreover, when the size of pores is of the same order of magnitude as the mean free path of molecules of the percolating gas, there is some molecular flow which violates the assumptions of the Darcy's law. As a result, the coefficient of permeability varies with the applied pressure. In an attempt to take these effects into account, we have tried to quantify different types of flow and we propose a method to calculate the apparent coefficient of permeability and hence the gas flow through concrete having any given degree of saturation and being under a given pressure difference across its extremities. For this purpose, we are characterizing concrete with an intrinsic permeability value. The variation of this intrinsic permeability and that of the contribution of non-viscous flow is studied, for a single concrete mix design in relation to the degree of saturation using a constant head permeametre named as CEMBUREAU and oxygen as the percolating gas.

RÉSUMÉ

La perméabilité, de même que la diffusion et l'absorption est utilisée dans le but de quantifier la durabilité d'un béton. La mesure de la perméabilité au gaz des bétons dépend fortement de leur taux de saturation. De plus la taille des pores pouvant être de l'ordre de grandeur du libre parcours moyen des molécules du gaz percolant, on a alors apparition d'un écoulement de type moléculaire qui ne respecte pas les hypothèses de la loi de Darcy et conséquemment la perméabilité varie en fonction de la pression moyenne de l'essai. Afin de prendre en compte ces problèmes, nous avons essayé de quantifier les différents types d'écoulement en présence et nous proposons une méthode de calcul de la perméabilité apparente puis du débit gazeux, pour n'importe quel taux de saturation du béton et pour n'importe quel gradient de pression. Pour cela il convient de déterminer la perméabilité intrinsèque du matériau qui est indépendante de la pression d'essai.

Pour une composition de béton standard nous étudions les variations de la perméabilité intrinsèque et celle de la part d'écoulement moléculaire en fonction du taux de saturation du matériau. Les perméabilités sont mesurées à l'aide d'un perméamètre à oxygène à charge constante de type CEMBUREAU.

1. INTRODUCTION

The durability of concrete depends upon its ability to prevent the ingress of aggressive chemical species. For more than a century, Darcy's coefficient of permeability has been used as a measure of its leaktightedness and its durability is often derived from this leaktightedness. An accurate and reproducible measurement of this coefficient with water becomes tedious and very difficult to achieve as the quality of concrete improves. This has shifted the focus from water permeability to gas permeability which is easier to measure, requires less time and produces more reproducible results. Also from the point of view of carbonation and ingress of other aggressive gases, it is more suitable to measure the gas permeability.

Measurement of gas permeability of concrete, despite being simple presents some difficulties discussed as follows:

– Firstly, the calculation of the coefficient of permeability is based on Darcy's law. This law assumes a laminar flow regime which is not strictly true for all pores of the porous network in a concrete sample. The dimensions of certain pores imply that the flow is partly of molecular nature which evidently does not follow Darcy's law. This causes the coefficient of permeability to vary with the applied pressure.

– Secondly, gas permeability varies significantly with the distribution and the amount of moisture present in the porous network. This effect is more pronounced when

Editorial note

LMDC, The Laboratoire Matériaux et Durabilité des Constructions, *is a RILEM Titular Member. It is directed by Prof. Jean-Pierre Ollivier, a RILEM Senior Member who is member of the RILEM Coordinating Committee.*

the concrete is nearly dry. There exist various preconditioning methods [2, 5] designed to achieve a standardized state with respect to the amount of moisture inside the pores, having varying success. These procedures have some disadvantages. They try to achieve an almost dry condition and hence a slight unconformity to the standardized procedure may result in significant errors. Also, these methods, even if sufficient for classifying a material, fail to predict the gas flow across a concrete subjected to natural atmospheric humidity conditions, which is obviously not in a standardized state. We shall very frequently use the term 'degree of saturation' as a measure of amount of water in the pores. It is defined as the volume of voids filled with water divided by the total volume of voids. This is expressed as a percentage.

1.1 Objective

A study is carried out with the aim to provide a method of predicting gas flow through a concrete having any value of moisture content between the saturated state and perfectly dry[1] and under a given pressure gradient across its extremities. To achieve this objective, first concrete is characterized by its '*intrinsic coefficient of permeability*', which is independent of the applied intensity of pressure and is a characteristic of the porous network alone. Second, the variation of this parameter is studied against the variation of the degree of saturation of the material. So our study is composed of three parts:

– Study of variation of the coefficient of permeability with pressure.

– Study of the variation of intrinsic coefficient of permeability.

– Study of the contribution of viscous and non-viscous flows.

All three studies are in relation with the variation of the degree of saturation.

1.2 Theoretical background

Permeability is defined as the property that governs the rate of flow of a fluid through a porous solid. The coefficient of permeability, K is defined from the well known Darcy's law. In this article for the sake of simplicity 'coefficient of permeability' is written as 'permeability' unless otherwise stated.

The gas permeability of a porous solid is calculated at constant pressure using the Hagen-Poiseuille expression [4] for a compressible fluid as given below:

$$K_{app} = \frac{2QP_{atm}L\mu}{A\left(P^2 - P_{atm}^2\right)}$$

K = Coefficient of permeability of gas (m^2)

Q = gas flow (m^3/sec)

A = cross-sectional Area (m^2)

μ = Coefficient of viscosity of the gas (N.s/m^2)

P = applied absolute Pressure (N/m^2)

P_{atm} = atmospheric Pressure (N/m^2)

This gas permeability value deviates from its permeability to liquids. Also, it varies with the applied mean pressure. This difference in behavior can be explained using the concept first proposed by Knudt et Warburg. They explained this effect taking into consideration non zero ' drift' or 'slip' velocities at the walls of the pores in case of gases. This effect is significant when the mean free path of gas molecules is of comparable magnitude to the pore size percolated by the gas in the material. When the mean free path is much less than the pore size, the slip velocity becomes negligible. The other limiting situation in which the mean free path is much greater than the diameter of the capillary or its length is called 'molecular streaming' or 'Knudsen flow.' Under these circumstances, flow takes place by diffusive, in contrast to viscous mechanisms. Various methods for the calculation of non-viscous flow exist. The most widely used method is the equation proposed by Klinkenberg [3, 6] introducing the concept of an 'intrinsic coefficient of permeability K_{int}'. This is the limiting value of gas permeability when the mean pressure P_m tends towards infinity.

$$K_{app} = K_{\text{int}}\left[1 + \frac{\beta}{P}\right]$$

The value of K_{int} is independent of the mean pressure and hence is a characteristic of the porous network alone whereas β is a constant, characteristic of the porous solid and the percolating gas. The contribution of molecular flow in the total gas flow can easily be calculated with this factor. The method of determination of K_{int} consists in measuring K_{app}, the apparent value of 'K' at different pressures and plotting it against the inverse of the mean pressure. After a straight line fit to the data, we get the value of K_{int} as intercept and $\beta.K_{int}$ as slope. More detail is provided while discussing experimental results (§ 3.1).

Another aspect which requires some explanation here is the calculation of pore sizes percolated by oxygen during our tests at different degrees of saturation. Using Kelvin-Laplace equation as given below [2], we can interrelate the minimum pore sizes (diameter 'd') emptied at a given degree of saturation through atmospheric relative humidity 'ψ' when the moisture inside the concrete porous network is in equilibrium with it.

$$\ln \psi = \frac{-4\sigma\upsilon}{dRT}$$

Here 'σ' and 'ν' are surface tension and molar volume of water respectively, 'R' is the perfect gas constant and 'T' is the absolute temperature.

(1) The perfectly dry condition may be defined as constant mass (of samples) i.e. *less than 0.5% variation of mass, during 24 hours at 105°C in an oven according to AFREM [2].*

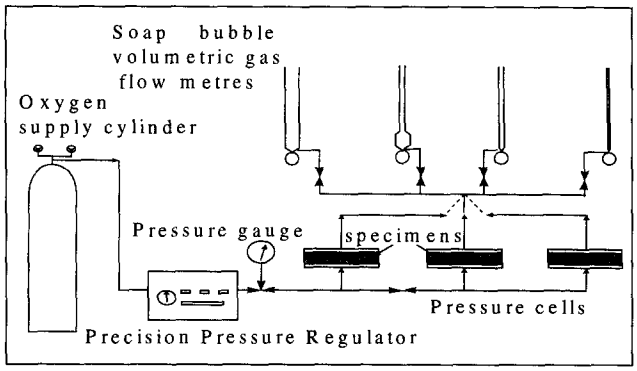

Fig. 1 – Schematic layout of the experimental setup.

2. MATERIALS AND METHODOLOGY

2.1 Experimental setup

An apparatus known as the CEMBUREAU permeameter was used for the determination of permeability. This is a constant head permeameter and oxygen is used as the permeating medium. The general layout of the apparatus is as shown in the Fig. 1:

A pressure difference up to 5 bars (5×10^5 N/m^2) can be applied to the specimens in the pressure cells which are sealed by a tightly fitting polyurethane rubber pressing under high pressure against the curved surface. The volume flow rate through the specimens is measured by means of a soap bubble flow meter.

After initiating the percolation of oxygen through a specimen at a given applied pressure, sufficient time (varying from 40 minutes to several hours) is provided for the establishment of steady state flow before an actual measurement is taken. This condition is verified by taking two measurements separated by a 15 minute time interval. If the two values differ by less than 3%, a steady state flow condition is assumed to be achieved.

The coefficient of permeability K is calculated from the Hagen-Poiseuille equation for laminar flow of a compressible fluid through a porous body under steady state conditions [3].

$$K = \frac{2QP_{atm}L\mu}{A\left(P^2 - P_{atm}^2\right)}$$

2.2 Composition of the concrete studied

A single concrete mix was used in manufacturing the specimens for all aspects of our study and the composition is given in Table 1.

Cement: An ordinary Portland cement was used, with 55 MPa crushing strength and a fineness of 3250 cm^2/g. Its chemical constituents are given in Table 2:

Aggregates: Riverbed rolled gravel from Garonne River has been used. The grain size classification for sand is 0/6, and for gravel it is 6/10 and 10/20.

The resulting concrete had the following characteristics:

Slump	4.5 cm
28 day compressive strength 'f_{c28}'	33 MPa
Entrapped Air	3.1%
Density after 28 days	
– saturated	2.46 g/cm^3
– dry	2.33 g/cm^3
Open porosity (measured by water saturation)	12.6 %

Table 1 – Concrete composition	
Component	Content kg/m^3
Cement CEM I 42,5R	270
Water	180
Plasticizer	4,5
Sand 0/6 mm	860
Gravel 6/10 mm	330
Gravel 10/20 mm	775

Table 2 – Cement constituents (in % age)									
CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	K_2O	Na_2O	SO_3	MnO	TiO_2
64.12	20.23	5.38	2.08	1.96	0.77	0.12	3.2	0.07	0.12

2.3 Casting, curing and preconditioning

The specimens for permeametry were cast in PVC cylindrical molds, in the form of discs compatible with the CEMBUREAU apparatus being 5 cm thick and 15 cm in diameter. They were cured, in the curing room for one day, demolded and then were kept in a temperature controlled room at 20°C wrapped in plastic sheeting and aluminum foil for 27 days.

To carry out a series of K_{int} measurements with concrete specimens having different amounts of moisture, we adopted the following procedure. First, we cast six specimens. We conducted a single K measurement for each of them and the two specimens having a value of K closest to the mean value were chosen. These two specimens were used repeatedly and the degree of saturation was varied by oven drying. All this was done in an effort to minimize the dispersion of results usually present among different samples of the same concrete and to obtain a variation of K_{int} due solely to changes in the degree of saturation. This degree of saturation was varied from almost saturated to totally dry. Between two successive measurements, the specimens were oven dried at 50°C to get a pre-assigned loss of water content. Afterwards, they were wrapped in adhesive aluminum foil and thin sticking plastic sheeting for moisture conservation and were kept at 50°C for 72 hours for its homogenization inside the porous network. Next they were cooled for 24 hours at 20°C before the next measurement was made. The results obtained show a very small dispersion and hence justify the procedure followed.

Drying the specimens during successive measurements gives rise to the question of micro-cracking which may influence permeability. Yssorche *et al.* [8] report that the micro-cracking that arises from drying proce-

dure at 50°C is very superficial and that the whole body of the concrete is identical before and after this procedure. As a precautionary measure the specimens were thoroughly checked under an optical microscope (100 ×) for the occurrence of cracks. There were practically none detected under this magnification even at the end of this series of tests. The possibility of development of cracks is minimized due to the fact that no strong moisture gradient existed at any time during this series of tests due to the homogenization process after every change in saturation value which is the principal factor provoking crack development.

The specimens for the determination of compressive strength were cast in cylindrical molds of 11.5 cm diameter and 23 cm height. They were cured for 28 days in the curing room before being tested.

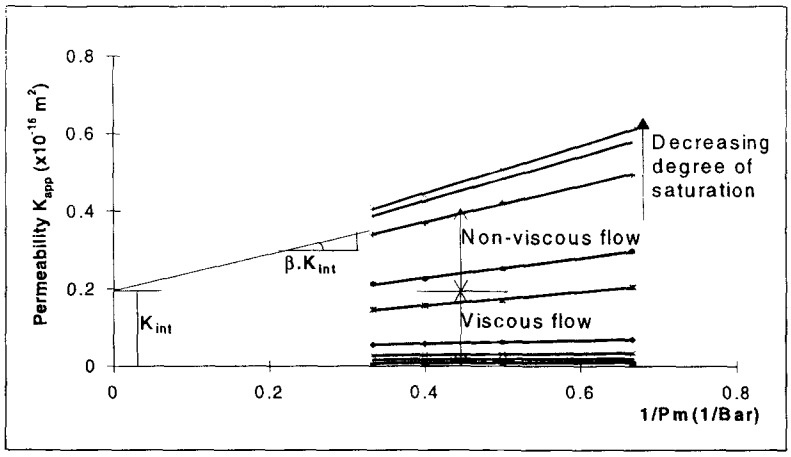

Fig. 2 – Relationship between mean pressure and apparent coefficient of permeability.

3. RESULTS AND ANALYSIS

The results of the three parts of our study are presented below.

3.1 Variation of K_{app} with pressure

First of all, the results which relate the measured value of coefficient of permeability to the mean pressure applied across the specimen will be presented. We shall call this coefficient as apparent coefficient of permeability 'K_{app}'. This nomenclature is used firstly to be able to distinguish it clearly from the intrinsic value K_{int} to be calculated next. Secondly, it is not uniquely dependent upon the porous network, but varies with the applied pressure. The procedure adopted is in line with the Klinkenberg concept. The values of K_{app} are plotted against the mean of the pressures at the two faces of the specimen (one of them always being atmospheric pressure) for each degree of saturation. These measurements cover a range of 2 to 5 bars (2×10^5 to 5×10^5 N/m^2) absolute pressure applied at one face. Through a straight line fit, this plot provides us with two parameters: K_{int} and β. Using the equation:

$$K_{app} = K_{int}\left[1 + \frac{\beta}{P_m}\right]$$

these two parameters enable us to calculate K_{app} for any given mean pressure. For all the straight line fits, we get a regression coefficient of around 0.98-0.99 which shows a near perfect fit. Fig. 2 shows an example of the curves plotted. This method can be better visualized graphically as shown with two parts of 'K' representing two types of flow in relation to each other, the part of viscous flow eventually becoming 100% as P_m tends towards infinity.

3.2 Variation of K_{int} in relation to the degree of saturation

Next we shall present the results concerning the variation of K_{int} in relation to the degree of saturation. Its value, as expected, rises continuously with decreasing moisture in the specimens. The overall change from the least value (0.0025×10^{-16} m^2) to the maximum value (0.23×10^{-16} m^2) is around 100 times, *i.e.* two orders of magnitude. This shows its very strong dependence upon the degree of saturation.

The value of K_{int} shows the same trend throughout the range of measurements. For this, two factors upon which it depends play their part. The rise in K_{app} is very slow when the specimens are almost saturated and becomes notably more rapid when they are nearing perfectly dry condition. This can be explained keeping in mind the inter-connectivity of pores inside the porous network. With more moisture still inside which is homogenized, drying up of

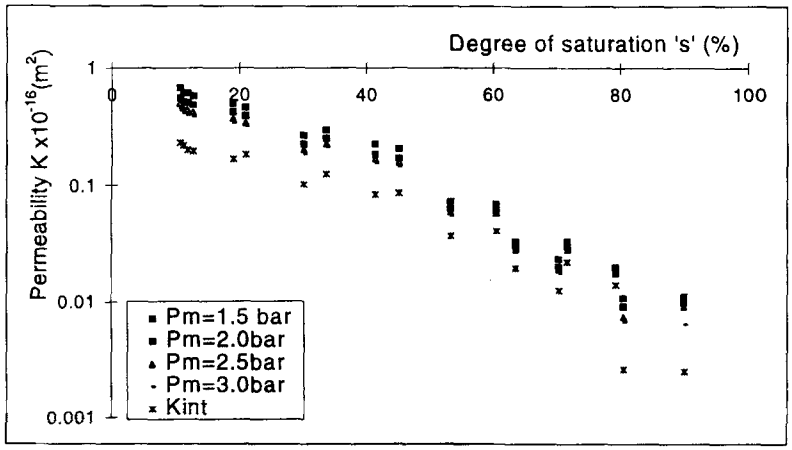

Fig. 3 – Variation of coefficient of permeability in relation to the degree of saturation.

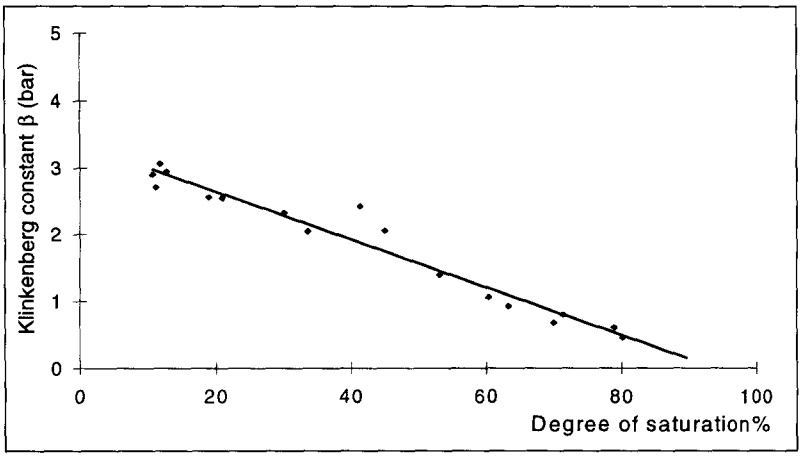

Fig. 4 – Evolution of Klinkenberg constant.

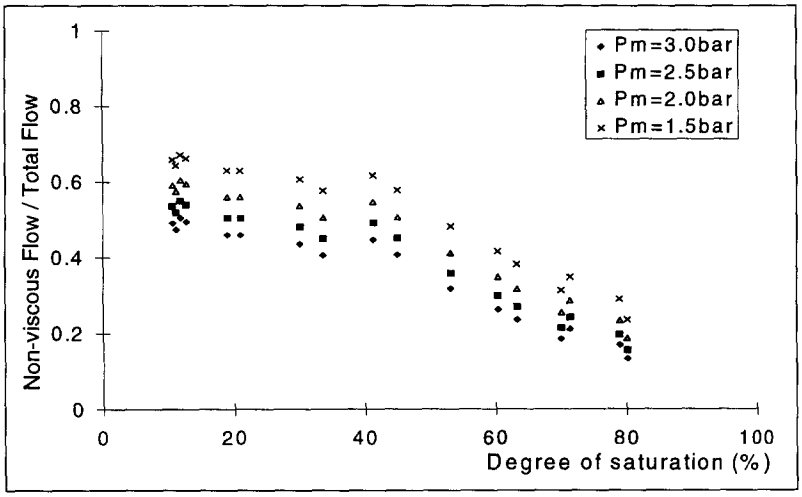

Fig. 5 – Evolution of contribution of non-viscous flow.

some of the pores does not provide many thorough passages for the percolation of oxygen. Variation becomes more significant when the dried up and thus empty pores or air voids as we may call them, start becoming interconnected. At very near to perfectly dry conditions, almost every single dried up pore contributes to oxygen flow. For K_{int} a second factor which keeps the trend of this variation, the same throughout, is the increasing value of β (to be presented next) along with increasing K_{app}. More details are given in the following section.

(See Fig. 3).

A logarithmic curve with only two correlation constants provides a good fit to the K_{int} values (Regression coefficient = 0.97) and it is given below ('s' represents the degree of saturation in percentage):

$$K_{int} = \ln[1.62 / s^{0.108}].10^{-16}m^2$$

which can be expressed in a general form as

$$K_{int} = \ln[a / s^{b}].10^{-16}m^2$$

and where

a = 1.62

and b = 0.108

for this concrete.

We have emphasized many times that K_{int} is a characteristic of the porous network alone. A question arises here that when it is a characteristic solely of porous network then why it is changing? In the partially saturated porous medium, water and solid both behave more or less in the same manner in blocking the flow of a percolating gas. We can note here that the effective porous network through which the gas is percolating is evolving with the changing degree of saturation. Hence K_{int} is indeed every time representing uniquely the porous network, but only the dried up portion which is changing.

3.3 Analysis of the contribution of non-viscous flow and variation of Klinkenberg's constant

The results concerning the contribution of molecular flow in the total oxygen discharge are presented next. The parameter used here for this purpose is Klinkenberg's coefficient β obtained in a manner discussed in § 3.1. The part of non-viscous flow (molecular and Knudsen type) is calculated as:

$$Q_{molecular} = \left[\frac{K_{int}\beta}{P_m}\right]\frac{A\left(P_a^2 - P_{atm}^2\right)}{2\mu P_{atm} L}$$

and the fraction

$$\frac{Q_{molecular}}{Q_{total}} = \frac{\beta/P_m}{1+\beta/P_m} = \frac{\beta}{P_m+\beta}$$

The value of β shows a continuous increase with the decrease in the degree of saturation. A straight line fit shows a good regression coefficient of 0.96.

(See Fig. 4).

During the homogenization of moisture inside the porous network, there is a tendency for water being a wetting fluid to occupy pores of relatively smaller diameter due to capillary drift. For air, being a non-wetting fluid, the inverse is the case. That means that drying first empties the largest pores and the smallest ones being the last to be emptied. Quantitatively, these sizes can be calculated using the Kelvin-Laplace equation given in § 1.2.

Using this equation, we can calculate the minimum size of the pores dried up at a given relative humidity. This phenomenon allows us to explain the increasing contribution of molecular flow with the decreasing quantity of moisture inside the porous network.

When there is a lot of moisture inside the porous network, almost all of the pores having sizes comparable to the mean free path of gas are blocked with water. For our case, the mean free path λ is around 800A° for P_m = 1.5 bar and 400A° for P_m = 3.0 bar. These and other calculations of pore sizes corresponding to different degrees of saturation

are based on sorption isotherms at 20°C carried out in the same lab for the same concrete. At degrees of saturation around 86% and 80%, pores of size 800A°and 400A° respectively start to become empty. As concrete with a degree of saturation of more than 80% is almost impermeable making reliable permeability measurements nearly impossible, measurements are invariably made with 's' less than 80%.

(See Fig. 5).

This implies that some part of flow is non viscous (13% to 23%) in the very first measurement at any of the four mean pressure values. At 's' = 55%, pores of 100A° begin to become empty of water and Knudsen flow begins to contribute to the total oxygen discharge. Very close to perfectly dry conditions, the total contribution of non-viscous flow may be as high as 65%. We must not forget that oxygen may start to percolate some larger pores (d > 800A°) very late when they become connected through a very small pore (*e.g.* d ~ 100A°) emptied at a later stage. In this manner, both types of flow evolve at the same time during the drying up process.

4. METHOD FOR CALCULATING GAS FLOW

A problem of calculating gas flow through concrete can be stated in the following manner: We consider a concrete wall of thickness 'L' and area 'A' separating two gaseous media of pressure 'P_1' and 'P_2' and the degree of saturation 's' at ambient temperature.

As is obvious to the reader, this calculation requires some steps interrelating quantities which are needed in the end to calculate the gas flow. The method is described step by step below using the correlations developed for the specific case under our study.

Step 1

Using the degree of saturation 's', we calculate the value of intrinsic coefficient of permeability Kint using the logarithmic correlation:

$$K_{int}\,(m^2) = \ln[1.62 / s^{0.108}]\ 10^{-16}$$

and the Klinkenberg constant β using the straight line fit:

$$\beta(\text{bar}) = 3.37 - 0.036s.$$

Step 2

We use K_{int} and β calculated in step 1, and P_m the mean of the two given pressures across concrete to calculate K_{app}:

$$K_{app}\,(m^2) = K_{int}\,[1+b/Pm].$$

Step 3

With K_{app} and the concrete dimensions, we calculate the gas flow using the Hagen-Poiseuille equation.

5. CONCLUDING REMARKS

Summarizing the details given above, we can say that we are able to formulate a method by which we can relate gas flow through concrete at any given moisture condition and the applied pressure gradient through different steps. The correlation used in these steps use very few constants and the coefficient of regression is every time very close to 1. So a small number of measurements, taken carefully, can be sufficient to obtain the same type of relationship for any other concrete.

The intrinsic coefficient of permeability can be a better parameter for characterization of a concrete for durability compared to the traditional Darcy's coefficient of permeability, as it is independent of the fluid properties and the applied pressure gradient. It is hence, a characteristic of the porous medium alone. It may allow us make a better comparison of values obtained under different experimentation conditions.

This method is still not verified for a large number of concrete compositions and hence it still requires validation for other cementitious materials specially high performance concretes, for its practicality and effectiveness. Also, it remains to be seen whether it works well or not with the concrete compositions containing additives like air entraining agents.

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