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Experimental study of gas and liquid permeability of a mortar

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Abstract

Eight samples cored from the same mortar were used to investigate their respective gas, ethanol and water permeability. Two gas and liquid permeability cells, using special devices for measuring the injected flow under steady conditions, were designed and presented in this paper. The obtained results showed that water permeability was systematically lower (in an order of magnitude from 1 to 2) than gas permeability whereas ethanol permeability was intermediate between these two values. Nevertheless, ethanol and gas permeabilities were found of the same order and, when gas permeability is corrected from the Klinkenberg (or slippage) effect, the results given by these two fluids are virtually identical and can be considered to be the intrinsic permeability value. Thus, the differences observed between water and gas permeability values have to be explained by other phenomena such as rehydration, dissolution and migration of fine elements or water adsorption in the thinnest pores.

Keywords: Transport properties; Mortar; Permeability; Klinkenberg

1. Introduction

This experimental study forms part of a much wider research program, undertaken by our laboratory, on the ageing process of cement matrix materials. It represents more than 2 years of mechanical testing coupled with permeability to gas and liquids. Permeability is one of the most important characteristics of these materials, particularly when used for the design of storage structures. In fact, the increase in permeability and porosity of such materials is currently accepted as providing a reliable indication of their degradation (at least on a qualitative level) whether it be of mechanical or physicochemical origins resulting from the cement matrix being attacked by aggressive products [1,2]. It has also been proven from mechanical tests that high compressive strengths have to be confirmed by low permeability values to indicate a reliable concrete or mortar durability [3], as this property is very sensitive to changes in porosity or microcracking phenomenon [4]. On the other hand, in order to prevent any contamination problems, the determination of permeability becomes essential. In spite of its importance, permeability is not clearly specified in the criteria for designing concrete or mortar even if its characterisation is increasingly recommended by French standardisation organisations working in that field of materials [5,6]. In our view, among the numerous permeability measurement possibilities, there is no agreement on a single test and the fluid (liquid or gas) to be used. According to Darcy's law and assumptions, permeability is an intrinsic property of a material, since it does not have to depend on the injected fluid. This is commonly verified when measuring rock permeability, such as sandstone [7] or granites [8] for example. Measuring cement matrix material permeabilities present some difficulties and phenomena, which are not completely understood at the present time. It is not known why the intrinsic permeability of certain types of concrete is sometimes a thousand times lower when measuring with water than with gas, whereas the values for rocks are virtually identical. Many researchers have attributed that to the self-sealing phenomena due to the hydration of previously unreacted cement. However, specimens with no remaining nonhydrated cement exhibit self-sealing [9]. More commonly, the difference between gas and water permeability values is explained by the theory of gas slippage or the Klinkenberg effect [10,11]. See, for example, the result from Bamforth [12], Perraton et al. [13] and Sullivan [14]. The presence of this effect results in an increase in the apparent

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Table 1
Proportion of the mortar constituents

Constituent	Proportion (kg/m^3)
Water	256
Cement	639
Fine sand (0.16–2 mm)	966
Coarse sand (0.8–3.15 mm)	414

medium permeability. It appears when the mean free path of the gas molecules is sufficiently large compared to the pore diameter and thus depends on the mean gas pressure and the porous network. Another factor, frequently mentioned in the literature [15,16], is the moisture content of the material, which can obviously affect the gas permeability. However, this problem is closer to relative gas permeability measurements under various liquid saturation conditions rather than intrinsic permeability evaluation [7].

The aim of this study is to evaluate to what extent the differences between gas and water permeabilities could be attributed to the Klinkenberg effect or to other reasons (carefully supposed to be physicochemical mechanisms appearing with water). To achieve this objective, the permeability of eight samples of the same mortar, initially perfectly dry, were measured with three different fluids: two liquids (water and ethanol) and a gas (pure argon). As ethanol and argon are chemically neutral compared with water, the measured permeability, by using these fluids, can be assumed to be the intrinsic value as soon as, for gas, the apparent value is corrected by the Klinkenberg effect.

2. Gas permeability measurements

2.1. Sample preparation and mortar used

The samples are cylinders of 37 mm in diameter and 70 mm high. They are cored from larger cylinders preserved, over 28 days, in water saturated with lime at a constant temperature of 20 °C and then rectified to obtain a perfect geometry. The mortar used is of a fine grain type, made up of Portland cement CPA CEM I 42.5 from Origny and Hostun sand. The mixture is prepared with a 0.4 W/C ratio. Table 1 gives the proportions of the various constituents.

As the permeability test is based on the measurement of intrinsic permeability by gas injection, the sample must be dried before the test. An ethanol injection was performed and then a moderate oven-drying at a temperature of 60 °C was carried out. Drying was stopped as soon as the sample weight remained constant. A comparison of this method with the more aggressive oven-drying method at 105 °C was made. It appeared very effective and did not exhibit significant thermal microcracking. A hydrostatic compression test, presented in Fig. 1, shows the perfect linearity of the stress–strain relation and thus the absence of microcracking due to drying.

2.2. Experimental device

The general principle of the test is to carry out a continuous gas flow through the sample under steady conditions. The pressure cell was designed to subject the sample to a constant gas pressure P_i at one end and atmospheric pressure P_0 at the other end. Fig. 2 shows an overall diagram of the experimental apparatus used. The test specimen, whose circumferential surface is sealed by a Vitton membrane, is placed in a pressure-confining cell. The confining pressure is maintained by a Gilson type pump. The gas, more than 99% pure argon, is injected via a buffer reservoir whose use will be described later.

2.3. Measuring gas permeability

As the material is supposed to be completely filled with gas, the measured permeability can be considered intrinsic

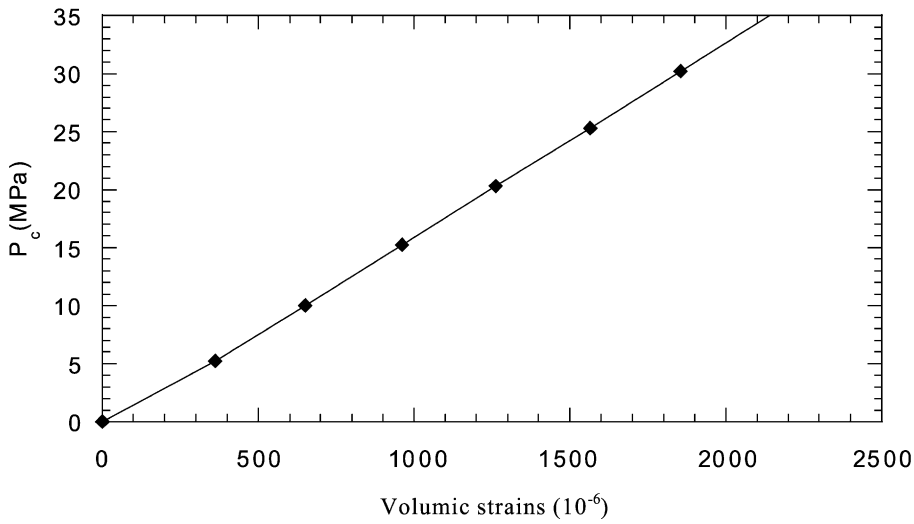

Fig. 1. Results from hydrostatic test after drying.

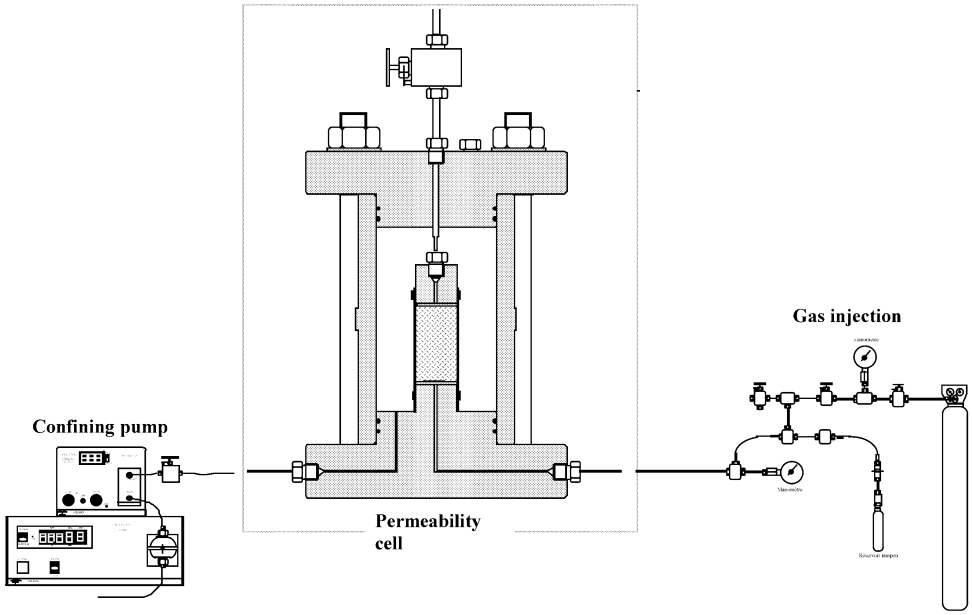

Fig. 2. Experimental device used for gas permeability measurements.

(according to generalised Darcy's law) and not relative. This would have been the case if water (or other liquid) was significantly present in the porous network. The experimental apparatus enables permeability ranging from 10^{-12} to 10^{-21} m^2 to be measured (representing hydraulic conductivity between 10^{-5} and 10^{-14} m/s for water at 20 °C). Measurement is carried out in a steady flow state with a moderate injection pressure of 1.5 MPa. To apply Darcy's law, the medium should be saturated with a single fluid phase and the flow must be laminar. We may assert that a laminar flow takes place if the Reynolds number is lower than 10^3 [17]. This number can be evaluated by $R=\rho V_c l_c/\eta$, where ρ is the specific mass of the gas, η is the dynamic viscosity and V_c is the relative velocity with respect to the solid skeleton. l_c is a characteristic length, which is of the same order as $\sqrt{tr\overline{\overline{K}}}$ where $\overline{\overline{K}}$ is the intrinsic permeability tensor. With an expected permeability value of 10^{-17} m^2, an extremely small value of R ($<10^{-3}$) is obtained. This proves the flow will be laminar. Such precautions allow the study to be placed within the conditions of Darcy's law, which is used for a one-dimensional flow. Argon is also assumed to be an ideal gas and this can be justified within the range of the pressure used [11]. Fig. 3 below presents the boundary conditions to which the sample of length h is subjected. For $x=0$, $P(x)=P_i$ injection pressure and for $x=h$ $P(x)=P_o$ draining pressure, which is maintained at atmospheric level in this case. In this test, longitudinal permeability, noted K_x, will be measured.

In a steady injection state, the pressure variation in the sample is given by the well-known expression:

$$P(x)=\sqrt{P_i^2\left(1-\frac{x}{h}\right)+P_o^2\frac{x}{h}} \quad (1)$$

Darcy's law can be written for a one-dimensional flow:

$$V_x=-\frac{K_x}{\mu}\mathrm{grad}P(x) \text{ or } V_x=-\frac{K_x}{\mu}\frac{dP}{dx} \quad (2)$$

V_x is the gas velocity and μ its viscosity (2.2×10^{-5} Pa s).

The method of measurement is based on an assessment of the mean entry gas flow Q_m in the sample. The injection pressure of 1.5 MPa and the very low value of the flow make the use of commercial flow transducers difficult; as a result, an indirect measurement method is developed, using a buffer reservoir at the entry point of the sample (Fig. 4), in order to measure the volume flow rate entering through the sample. This method is carried out in three steps.

Step 1: The gas is injected via the buffer reservoir, which is supplied with gas by an external source.

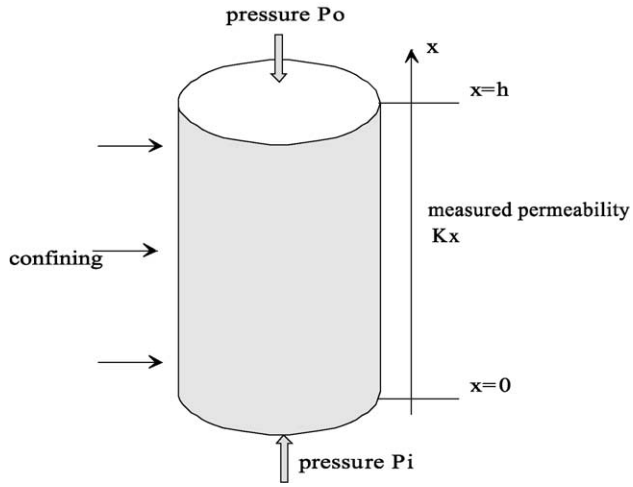

Fig. 3. Boundary conditions.

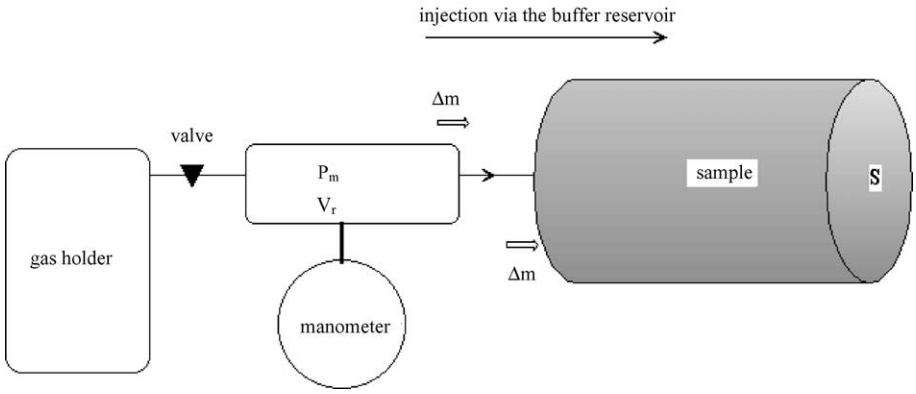

Fig. 4. Simplified diagram: mean flow rate measurement.

Step 2: The reservoir is no longer supplied by the external source but gas, initially at pressure P_i, is continuously flowing into the sample. This involves a drop in pressure in the reservoir, which is limited to a low value ΔP_i (0.1 MPa) during a measured injection time Δt. Thereafter, it is assumed that every thing occurs as if a steady-state flow was taking place at a constant mean injection pressure P_m given by $P_m=P_i-(\Delta P_i/2)$.

Step 3: The mean volume flow rate Q_m is calculated by $Q_m=\Delta\rho V_r/\rho\Delta t$ where V_r is the volume of the buffer reservoir and the piping, $\Delta\rho$ is the variation of the gas specific mass due to ΔP_i. As mentioned before, the gas is assumed to be ideal and the test lasts sufficiently long for it to be under isothermal conditions. Consequently, it can be shown that the mean volume flow is written [6]:

$$Q_m = \frac{V_r \Delta P_i}{\Delta t P_m} \tag{3}$$

This method has been checked on many occasions when it was possible to directly measure the flow. A similar

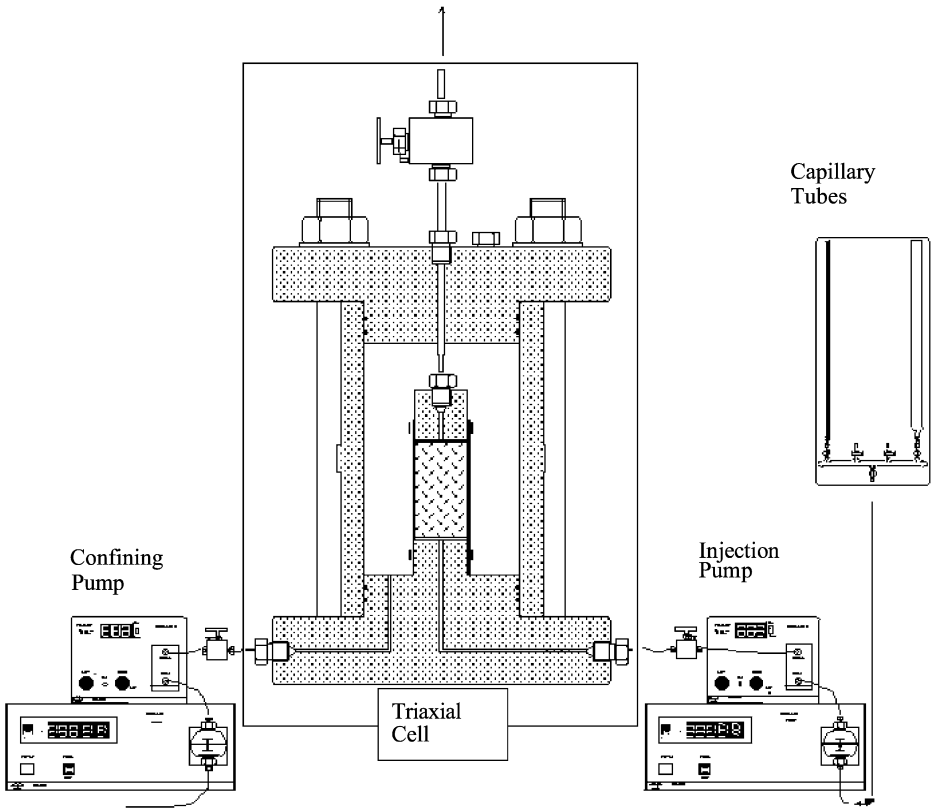

Fig. 5. Liquid permeability device.

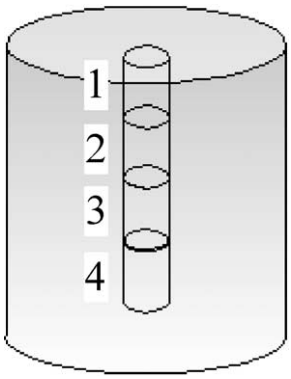

Fig. 6. Location of the different samples.

Table 2
Results obtained with gas (mean gas pressure 0.8 MPa), ethanol and water (K_g, K_e and K_w are respectively the values obtained with gas, ethanol and water)

Sample	K_g (10^{-17} m^2)	K_e (10^{-17} m^2)	K_w (10^{-17} m^2)	K_g/K_e	K_g/K_w	K_e/K_w
1A	1.96	0.96	0.014	2.04	140	69
2A	1.52	0.85	0.054	1.79	28	16
3A	0.96	0.49	0.03	1.96	32	16
4A	1.01	0.53	0.041	1.90	25	13
1B	2.49	0.95	0.11	2.62	23	9
2B	2.56	1.02	0.035	2.51	73	29
3B	2.18	0.75	0.078	2.90	28	10
4B	2.60	1.14	0.28	2.28	9.3	4

method for measuring gas permeability can be found in Ref. [3] under more complex conditions. The use of Darcy's law and of Eq. (3) from which P_i=P_m was taken, gives the following:

$$K_x = \frac{\mu Q_m}{S}\frac{2hP_m}{(P_m^2 - P_o^2)} \quad (4)$$

where S is the sample cross-section.

2.4. Measuring liquid permeability

As mentioned before, as soon as the gas permeability measurement is completed, the sample is subjected to a liquid flow, either ethanol or water. The liquid permeability cell is essentially similar to the gas permeability cell, except that it has a different fluid inlet device (Fig. 5). Liquid is injected, at a constant pressure P_i, by a high-pressure pump (Gilson type) from calibrated capillary tubes (3-mm diameter). Such a device has the advantage that the injected flow can be measured accurately and, as soon as a steady-state flow is reached, it can be easily recorded. Furthermore, it is a direct measurement of the permeability, under steady conditions, which is carried out and based on the entry flow rate Q measurement. As shown above Darcy's law can be applied and leads to:

$$K_x = \frac{\mu Q}{S}\frac{h}{(P_i - P_o)} \quad (5)$$

Q is the volume rate of liquid flow and μ the liquid viscosity. P_i is maintained at 1.5 MPa during the whole test and P_o is the atmospheric pressure. Such pressure conditions are identical to those used for gas permeability measurements.

In order to make sure of ethanol saturation, tests were carried out by high-gradient pressure injection to expel gas from the sample. Complete saturation is supposed to be achieved as soon as the injected flow rate is constant; the permeability test is then proceeded. Several flow rate values were measured (during 2 days) in order to verify whether it remained constant. In reference to the characteristic of ethanol (solubility, surface tension, viscosity, etc.) [22] and the results obtained, we can thus conclude that the chemical activity of ethanol on the mortar used is very low

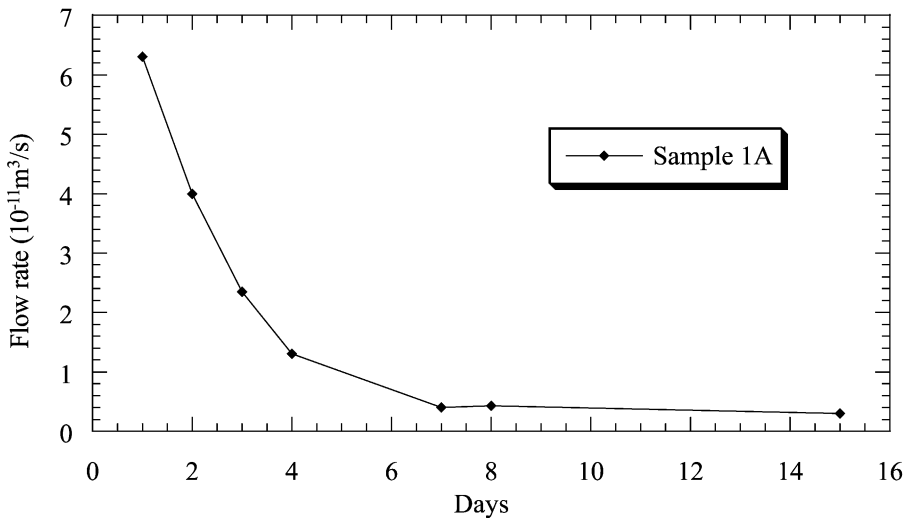

Fig. 7. Evolution of the flow rate with time (Sample 1A).

Table 3
Gas permeability test under higher gas pressures

Mean pressure (MPa)	0.8	1	2	3	4
K_g (10^{-17} m^2)	1.96	1.7	1.34	1.1	1
K_g/K_e	2.04	1.77	1.40	1.15	1.04

or negligible compared to water activity. See, for example, the analogy with sorptivity tests [18].

3. Experimental results

Two series of four samples were respectively cored from two cylinders, A and B. They were located along the height as shown in Fig. 6. After drying, permeability tests were carried out using first the gas and then ethanol and water. As soon as the flow rate remains constant, we proceed to the permeability measurement except for water as there is a continuous decrease in the injected flow rate (i.e., the permeability) with respect to time until the value stabilises (Fig. 7). This phenomenon is quite instantaneous when ethanol is replaced by water as injection liquid; this cannot be explained by their viscosity differences, which are very similar to each other. Moreover, these two liquids are miscible.

Table 2 gives all the results obtained for the eight samples. When measured with gas, the permeability values vary from 10^{-17} to 2.6×10^{-17} m^2 for both Cylinders A and B. For such measurements, these differences are not very significant. The examination of the results does not show either a systematic effect of the sample position in the cylinder even if the mortar from Cylinder A seems less homogeneous than that from Cylinder B. What is more instructive is the observed differences (of the permeability values) according to the various fluids used for injection.

3.1. Comparison of gas–ethanol

Even if systematically lower K_e is of the same order of magnitude as K_g and the ratio K_g/K_e is almost the same with a mean value of 1.92 for Mortar A and 2.6 for Mortar B. This observation leads us to think that these differences are due to the Klinkenberg effect. In order to verify this point, one complementary test was carried out on Sample 1A by measuring K_g with four mean gas pressures (1, 2, 3 and 4 MPa). Table 3 and Fig. 8 give the results of this test. As expected from a possible Klinkenberg effect, the apparent gas permeability clearly decreases when being measured with a higher mean level of pressures. It was not possible to proceed with pressure over 4 MPa, nevertheless, the Klinkenberg effect can be assumed to vanish at this pressure level as the apparent permeability value reaches the obtained value by using ethanol. Thus, permeabilities measured with this liquid can be considered to be intrinsic permeabilities. However, as soon as values obtained from gas injection are corrected by taking into account the Klinkenberg effect, the information providing both types of measurements is virtually the same.

3.2. Comparison of ethanol–water

Examination of the water permeability results shows a larger scatter than for those obtained with ethanol or gas. K_w is systematically lower than K_e (or K_g) with a ratio K_e/K_w that can vary from 4 to 69. This scatter may be due to a major difficulty with this kind of measurement, which is a continuous decrease in the injected flow rate into the sample. When argon or ethanol are used, the injected flow rate reaches a stable value within a few hours. This is one essential advantage of ethanol or gas as percolating fluids as compared to water, which is the rapidity of measurements. When used over a long period under a high-pressure gradient, water may bring about dissolution and migration of fine elements within

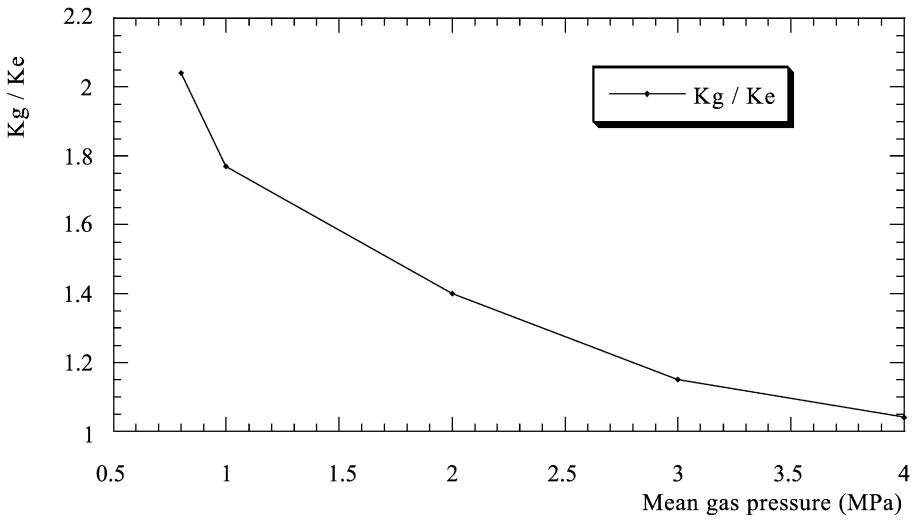

Fig. 8. Variation of the K_g/K_e ratio when the mean gas injection pressure is increased.

the material. This problem that also occurs during tests on rock materials such as granite [8] is a probable disturbing factor of the permeability measurements, which as a result may lead to a lower permeability but over a long testing period of time only. Thus, the instantaneous drop followed by a continuous decrease of the flow rate when substituting water for ethanol and the high K_e/K_w ratio is typically a problem, which occurs in hydrated cement systems. When pure water is injected into the porous space, the chemical equilibrium of the various phases present in the material is no longer verified and leads to dissolution/precipitation phenomena. The rate of these is intrinsically much faster than the rate of transport [19] and can justify the observed instantaneous drop of the flow rate. The continuous decrease of the latter, which follows the first step has also been shown for capillary absorption of water, and is attributed to the swelling of the cement gel [18–20]. As a consequence, the effective diameter of the pores is progressively reduced with water penetration in the sample, which results in a decrease of water permeability. This phenomenon is quite similar to the healing of cracks under water penetration [21].

4. Conclusion

Three permeability tests, using gas and two liquids, were performed on eight samples of the same mortar previously dried at a moderate temperature. These tests have shown that water permeability is systematically lower (from 1 to 2 order of magnitude) than gas permeability. Theses differences are often attributed to the Klinkenberg (or slippage) effect and, in order to verify or otherwise this point, we proceeded to permeability measurements with ethanol, which is a neutral liquid compared to water. Even if propan-2-ol would have certainly been a better choice as it does not alter the mortar microstructure [18], ethanol did not exhibit any measurable effect on the injection flow rate using a high gradient pressure. Thus, it was assumed to be a neutral liquid compared to water. Ethanol permeability values were always intermediate between gas and water values and, when corrected with the Klinkenberg effect, were virtually the same as gas permeability results. Thus, water and gas (or ethanol) permeability differences have to be justified by other causes such as rehydration of nonreacted cement, dissolution and migration of fine elements and water adsorption in the smallest pores of the cement matrix. Thus, water and gas permeability differences have to be justified by two main causes.

- The Klinkenberg effect depending on the pore size distribution and the mean gas pressure level. This effect is reduced when pressure increases; thus, it can be easily corrected. It is clear, in this study, that the Klinkenberg effect is only the smallest part of the observed differences between gas and water permeability.
- The chemical activity of pure water, which may lead to rehydration of nonreacted cement, dissolution/precipitation, migration of fine elements and water adsorption in the smallest pores of the cement matrix. One or a mix of these phenomena is the main cause of the differences mentioned before.

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