Pergamon

Available online at www.sciencedirect.com

SCIENCE DIRECT®

Cement and Concrete Research 34 (2004) 737–744

CEMENT AND CONCRETE RESEARCH

Prediction of the relative permeability to gas flow of cement-based materials

J.P. Monlouis-Bonnaire*, J. Verdier, B. Perrin

Laboratoire Matériaux et Durabilité des Construction (LMDC), INSA—UPS Génie Civil, 135, Avenue de Rangueil, 31077, Toulouse Cedex 04, France

Received 13 June 2002; accepted 13 February 2003

Abstract

The macroscopic physical modeling of coupled air and water transport in unsaturated porous material requires transport coefficients for the two phases along with the diffusion coefficient of vapor in air.

As their experimental determination is often difficult and time consuming, relations have been proposed for a long time to allow the calculation of all the needed parameters, starting from experimental properties that are easier to obtain, such as water pressure head as a function of the degree of saturation or isothermal sorption curves.

These relations, validated for soils, are also used for cement, mortars or concrete, although they have not been really justified in these cases.

The aim of this study is to determine if soil physics principles can be applied to unsaturated moisture movement in concrete.

We measured the air permeability of concrete specimens from which we obtained the relative permeability. Evidently, the parameters introduced in the relations given by Van Genuchten [Ann. Geophys. 3 (1985) 615] have to be revised. New values are proposed for the parameters for the concrete studied. These values are justified by the good reproduction of the experimental air permeability and airflow evolution observed during experiments.

Keywords: Permeability; Concrete; Modeling

1. Introduction

The modeling of complex fluid movements within unsaturated cement-based materials is always topical because many points remain unclear due to the complexity of the microstructure and all the physicochemical interactions between the different phases.

Moreover, macroscopic models that are used to describe the behaviour of these materials need as input parameters transport properties such as permeability, conductivity or constituent diffusivities. The measurement of these parameters is generally very difficult to carry out and time consuming.

Because of this, researchers have often tried to calculate these properties starting from other, easier to obtain, experimental results, such as the water retention curve. Van Genuchten [2], whose works are based on Mualem's research [3], has proposed very interesting relations that have been validated in the field of soil mechanics.

The purpose of this paper is to determine if these relations are relevant for cement-based materials [4]. Therefore, we measured the air permeability of concrete specimens in different saturation states, which allowed us to obtain the relative permeability. The water retention curve gives the different Van Genuchten's parameters, allowing a direct comparison between calculated and experimental permeability [1].

2. Theoretical approach

The modeling of fluid transport is based on mass balance equations for the liquid and the gas phases.

$$\frac{\partial}{\partial t}(\rho_l\theta_l + \rho_v\theta_g) = -\nabla[(\rho_l u_l)\theta_l + (\rho_v u_g + j_v^{\mathrm{diff}})\theta_g] \quad (1)$$

$$\frac{\partial}{\partial t}(\rho_a\theta_g) = -\nabla[(\rho_a u_g - j_v^{\mathrm{diff}})\theta_g] \quad (2)$$

where θ_l, θ_g [%] are the volumetric fractions for liquid and gas, respectively; u_l, u_g [m/s] are the macroscopic mean

---

* Corresponding author. Tel.: +33-561-559926; fax: +33-561-559949.
E-mail address: montloui@insa-tlse.fr (J.P. Monlouis-Bonnaire).

0008-8846/$ – see front matter © 2004 Published by Elsevier Ltd.
doi:10.1016/S0008-8846(03)00071-1

mass liquid and gas velocities; $j_{\mathrm{v}}^{\mathrm{diff}}$ [kg/m^{2}s] is the vapor density diffusion flux; and ρ_{l}, ρ_{v}, ρ_{a} [kg/m^3] are the liquid, vapor and air mass densities.

The advective parts are expressed with Darcy's law,

$$\rho_{\mathrm{l}} u_{\mathrm{l}} = -K_{\mathrm{l}} \nabla p_{\mathrm{l}} \quad \rho_{\mathrm{g}} u_{\mathrm{g}} = -K_{\mathrm{g}} \nabla p_{\mathrm{g}} \tag{3}$$

and the diffusive parts with Fick's law,

$$j_{\mathrm{v}}^{\mathrm{diff}} = -D_{\mathrm{v}}^{\mathrm{macro}} \nabla \rho_{\mathrm{v}} \tag{4}$$

These relations can produce the three parameters K_{l}, K_{g}, $D_{\mathrm{v}}^{\mathrm{macro}}$, defined as:

$$K_{\mathrm{l}} = \rho_{\mathrm{l}} \frac{k k_{\mathrm{rl}}}{\mu_{\mathrm{l}}} = \rho_{\mathrm{l}} \frac{k_{\mathrm{l}}^{\mathrm{eff}}}{\mu_{\mathrm{l}}} \quad K_{\mathrm{g}} = \rho_{\mathrm{g}} \frac{k k_{\mathrm{rg}}}{\mu_{\mathrm{g}}} = \rho_{\mathrm{g}} \frac{k_{\mathrm{g}}^{\mathrm{eff}}}{\mu_{\mathrm{g}}} \tag{5}$$

where k [m^2] is the absolute permeability, k_{rl} and k_{rg} [$-$] are the relative permeabilities to liquid and gas, μ_{g} and μ_{l} [kg m^{-1} s^{-1}] are the dynamic viscosities, $k_{\mathrm{l}}^{\mathrm{eff}}$ and $k_{\mathrm{g}}^{\mathrm{eff}}$ [m^2] are the effective liquid and gas permeabilities, respectively, and $D_{\mathrm{v}}^{\mathrm{macro}}$ [m^2/s] is a macroscopic diffusive coefficient for vapor.

The macroscopic diffusion coefficient for the vapor in the porous medium depends on the diffusion coefficient of vapor in air, D_{vo}, the tortuosity factor $\tau(\theta_{\mathrm{l}})$, and the part of the porosity occupied by the gaseous phase, $(\varepsilon - \theta_{\mathrm{l}})$, expressed by the difference between the global porosity (ε) and the water content (θ_{l}):

$$D_{\mathrm{v}}^{\mathrm{macro}} = \tau(\theta_{\mathrm{l}})(\varepsilon - \theta_{\mathrm{l}}) D_{\mathrm{vo}} \tag{6}$$

Now we will present the relations between these parameters, which are well known in the field of soil physics, and which have been extracted from many studies, particularly from those of Mualem [3] and Van Genuchten [2].

2.1. Van Genuchten's relations

These relations have the great advantage of being based on the water capillary retention curve and the open porosity of the material, which are easy to determine experimentally. The objective of this work is not to discuss the origin of these relations but only to analyse their capacity to describe the material behaviour.

Two reduced variables characterising the effective degree of saturation of the wetting (water) and nonwetting (air) fluids are represented, respectively, by $\overline{S}$ and $\overline{S}*$. The following relations define them:

$$\overline{S} = \frac{\theta_{\mathrm{w}} - \theta_{\mathrm{wr}}}{\varepsilon - \theta_{\mathrm{wr}}} \quad \overline{S}* = \frac{\varepsilon - \theta_{\mathrm{nwr}} - \theta_{\mathrm{w}}}{\varepsilon - \theta_{\mathrm{nwr}}} \tag{7}$$

where θ_{w}, θ_{wr}, θ_{nwr} and ε are the actual volumetric water content, residual water content (volumetric water content as threshold of incoherently distributed wetting fluid), residual air content (volumetric air content as threshold of incoherently distributed nonwetting fluid) and porosity, respectively. The pressure head, h, is then expressed, according to Van Genuchten, by [5]:

$$\overline{S} = [1 + (ah)^n]^{-m} \tag{8}$$

The inverse relation expressed in term of capillary pressure p_{c} gives:

$$p_{\mathrm{c}} = a\left[\overline{S}^{(-1/m)} - 1\right]^{(1/n)} \tag{9}$$

where a, n and m represent empirical parameters.

According to Mualem [3], an expression for the unsaturated hydraulic conductivity of cement-based materials could be determined such as:

$$k_{\mathrm{rl}} = (\overline{S})^{1}\left[\frac{f(s_{\mathrm{l}})}{f(1)}\right]^2 \text{ with } f(s_{\mathrm{l}}) = \int_0^{s_{\mathrm{l}}} \frac{1}{h(x)} \mathrm{d}x \tag{10}$$

where $s_{\mathrm{l}} = \theta_{\mathrm{w}}/\varepsilon$ is the water degree of saturation. Substituting the expression of capillary pressure (Eq. (9)), the precedent equation leads to an integral expression representing a particular form of the incomplete beta function (Eq. (11)) and, in its most general case, no closed form expression can be derived [2].

$$f(s_{\mathrm{l}}) = m\left[\int_0^{s_{\mathrm{l}}^{(1/m)}} y^{(m-1+1/n)}(1-y)^{(-1/n)} \mathrm{d}x\right] \tag{11}$$

However, analytical solutions do exist for integer values of the exponent $(m - 1 + 1/n)$. To obtain relative simple predictive closed-form analytical expressions for the relative conductivities, Van Genuchten has taken restricted relations between m and n used in Eqs. (7)–(11), with three choices:

$$m = 1 - 1/n \quad m = 1 - 2/n \quad n = \infty,\ m = 1$$

The first equation is the most pertinent for some soils [1]. According to Van Genuchten [2], a new expression for unsaturated hydraulic conductivity could be determined, assuming the first relation $m = 1 - 1/n$ (Eq. (12)):

$$k_{\mathrm{rl}} = (\overline{S})^{l}\left[1 - \left(1 - \overline{S}^{(1/m)}\right)^{m}\right]^2 \tag{12}$$

From Eq. (8), we can also apply the predictive model of Mualem [3] for a gaseous phase, similar to what was done by Brooks and Corey [6] with Burdine's model [7]:

$$k_{\mathrm{rg}} = (\overline{S}*)^{p}\left[\frac{f(s_{\mathrm{l}})}{f(1)}\right]^2 \text{ with } f(s_{\mathrm{l}}) = \int_{s_{\mathrm{l}}}^{1} \frac{1}{h(x)} \mathrm{d}x \tag{13}$$

Using Eqs. (11) and (13) leads to:

$$\begin{aligned} k_{\mathrm{rg}} &= (\overline{S}*)^{p}\left[1 - (1 - \overline{S}*)^{(1/m)}\right]^{2m} \\ &= (1 - \overline{S})^{p}\left[1 - (\overline{S})^{(1/m)}\right]^{2m} \end{aligned} \tag{14}$$

In these relations (Eqs. (10), (12)–(14)), the exponent coefficients l and p are assumed to be the same. Van Genuchten has determined these coefficients, from comparisons between model and experimental data for 45 soils [3], for a range of values of l [$-l \leq 1 \leq 3$]. The best value for those soils correspond to $l = 0.5$.

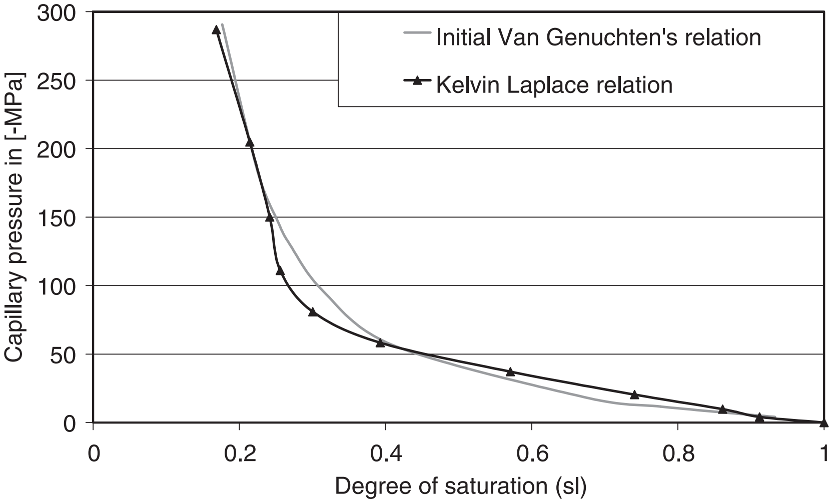

Fig. 1. Capillary pressure versus degree of saturation.

The following expression allows the calculation of the first two terms on the right side of Eq. (6) as a function of s_l [8]:

$$\tau(\theta_l)(\varepsilon - \theta_l) = (\varepsilon s_l)^{4/3}(1 - s_l)^2 \tag{15}$$

3. Materials

To test the relevance of Van Genuchten's relations for cement-based materials, the air permeability of concrete specimens in different saturation states were measured, from which k_g^{eff}, k_{rg} and k can be deduced.

The concrete used had a 0.48 W/C ratio (mass of water/mass of cement used). The constituents were ordinary Portland cement (375 kg m^{-3}) and various limestone aggregates, three sands (0–2.5, 0.63–1.25 and 1.63–2.5 mm) and two gravels (2–6.3 and 10–20 mm). A superplasticizer, added to the water, was used (0.4% of the cement mass). After casting, the concrete, which was covered with a plastic film to avoid water evaporation, was kept in the moulds for 24 hours. Then, each cylindrical sample used (diameter, 15 cm; thickness, 20 cm) was removed from its mould and cured in tap water at a temperature of 20 ± 2 C for 28 days to stabilise the hydration mechanism. After 21 days of curing, samples of 5-cm thickness were sawn from the original specimen and the first 20 mm of both ends were removed in an attempt to avoid skin effects. The thickness and diameter of each sample were precisely measured at eight points. After the curing time, compressive strength and open water porosity tests were performed. Mean values were 62.3 MPa and 13.2 ± 1%, respectively.

After 29 days, samples were placed in an oven and dried at 50 C. Thus, by weighing, it was possible to obtain various degrees of saturation inside the sample to get information on the permeability saturation dependence. In

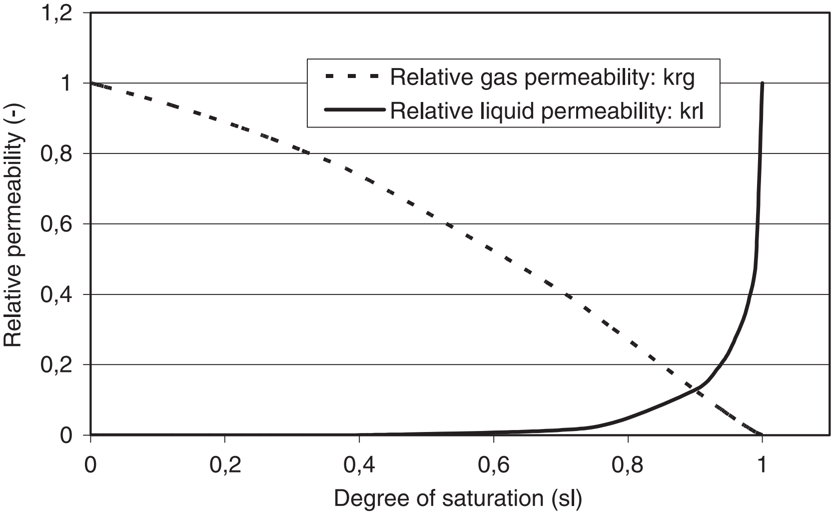

Fig. 2. Relative permeability versus degree of saturation (initial Van Genuchten's relation).

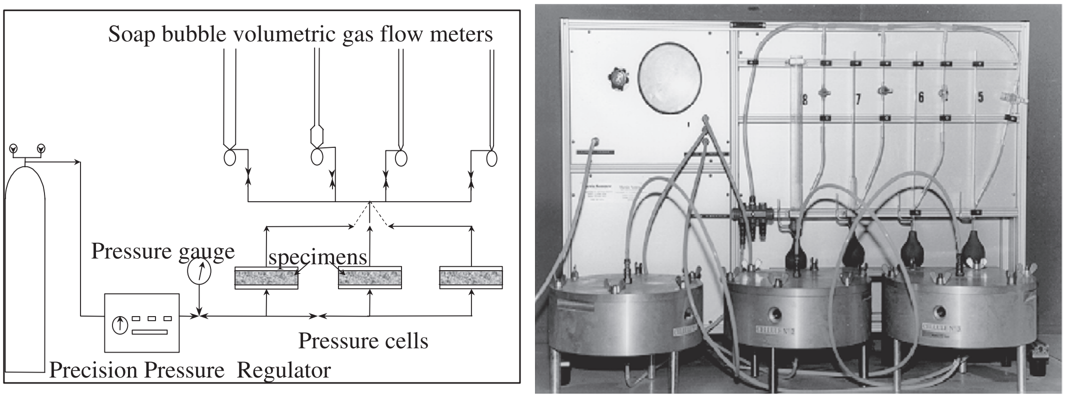

Fig. 3. Schematic layout of the experimental constant head permeameter.

order to have a uniform saturation state the samples were covered with cellophane and aluminum paper and placed in a watertight bag for the same duration as the previous drying time [9]. Accounting for the slow drying process, due to drying step by step, the relatively small specimen thickness and the lack of mass loss during the water redistribution period, we expected good saturation state uniformity.

4. Identification of the parameters allowing the calculation of the relative permeability

First, we identify the Van Genuchten's parameters to determine the relative permeability, which will then be compared to the experimental values.

The experimentally determined isothermal desorption curve allows conversion of the relative humidity, via the Kelvin–Laplace relation, to the capillary pressure p_c. The value of p_c as a function of s_l is presented in Fig. 1. The parameters a and m (Eq. (9)) are then identified by obtaining a calculated curve reproducing as best as possible the experimental one (Fig. 1).

From these parameters, the relative permeabilities k_{rl} and k_{rg}, expressed by Eqs. (12) and (14), are deduced (Fig. 2).

5. Experimental determination of gas permeability

Permeability was measured with a constant head CEMBUREAU permeameter [10]. The apparatus used, illustrated in Fig. 3, allowed recording of the flow.

Upstream and downstream flows were measured with mass flowmeters and the volume flows at normal pressure (1 bar) and temperature (273.15 K) conditions were then

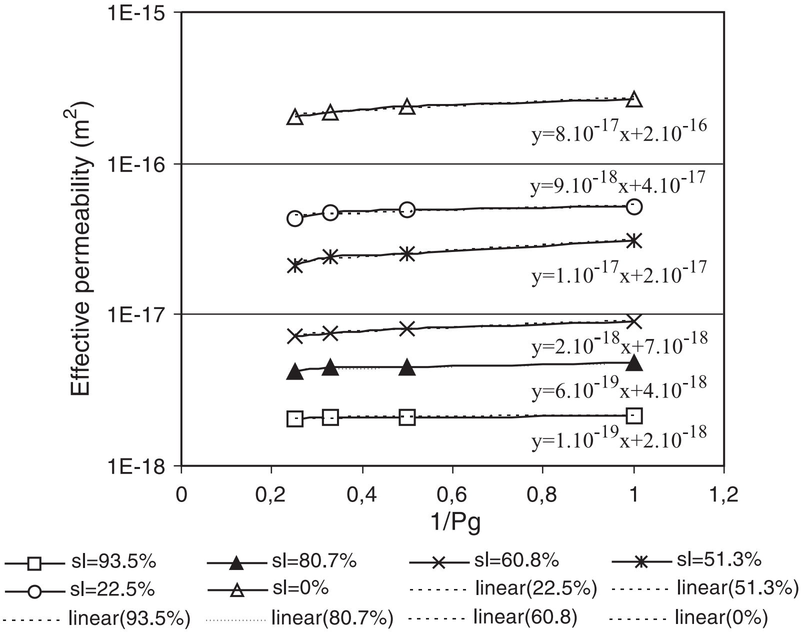

Fig. 4. Effective permeability for different degrees of saturation and gas pressures.

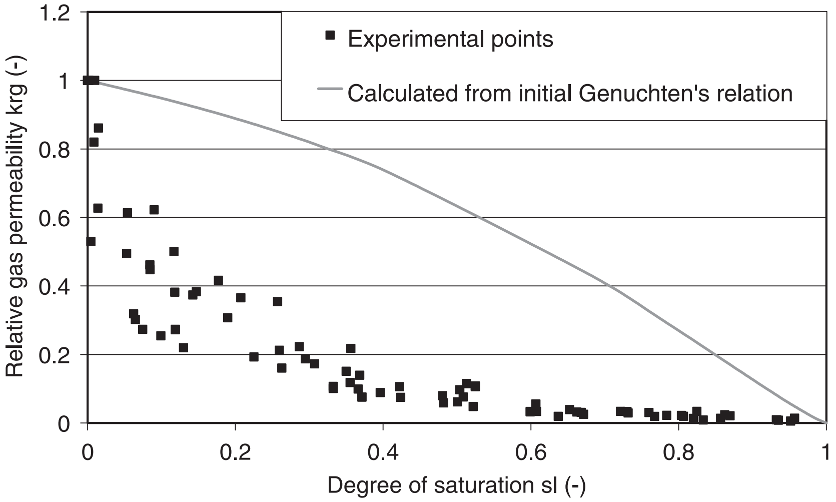

Fig. 5. Experimental gas permeability compared to calculation obtained with the initial Van Genuchten's relation.

calculated (expressed in normal liters per minute, nl/min). A data-recording system (HP 34970 multimeter and PC) gave the variation of the volume flow with time. In order to obtain values representative of a material, the test was conducted on a minimum of three samples [11]. A 1-MPa lateral confinement was chosen to ensure radial and orthoradial stresses in the sample.

The following relation gives the effective permeability of the material tested when the flow is assumed to be laminar and unidirectional [10]:

$$k_g^{\mathrm{eff}} = \frac{2\mu_g L P_s Q_s}{A(P_e^2 - P_s^2)} \tag{16}$$

Where L [m] is the sample thickness, A [m^2] is the section subjected to flow, P_e [Pa] is the upstream absolute pressure and P_s is the downstream absolute pressure.

The effective permeability k_g^{eff} depends on the pressure gradient, temperature, water saturation and material microstructure. The permeability given by Eq. (16) assumes a laminar flow but in fact a nonviscous contribution due to the pore fineness in concrete is frequently observed (molecular effect and slip flow, Knudsen's effect). These contributions and pressure dependence are well known since Klinkenberg [12] proposed a correction, parameterized by the factor β, depending on the mean free path of the gas molecules. The mean free path is a function of pressure and the degree of saturation. The effective permeability can then be written:

$$k_g^{\mathrm{eff}} = k k_{rg} \beta(p_g, s_l) \tag{17}$$

For a given degree of saturation, the effective permeability is generally a linear function of $(1/p_g)$, where p_g is the total

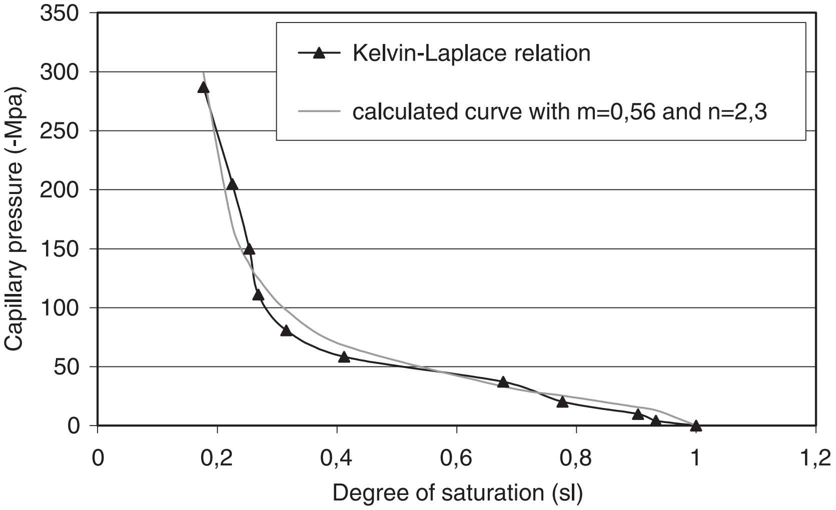

Fig. 6. Adjustment of parameters in order to reach a calculated curve reproducing the experimental capillary pressure.

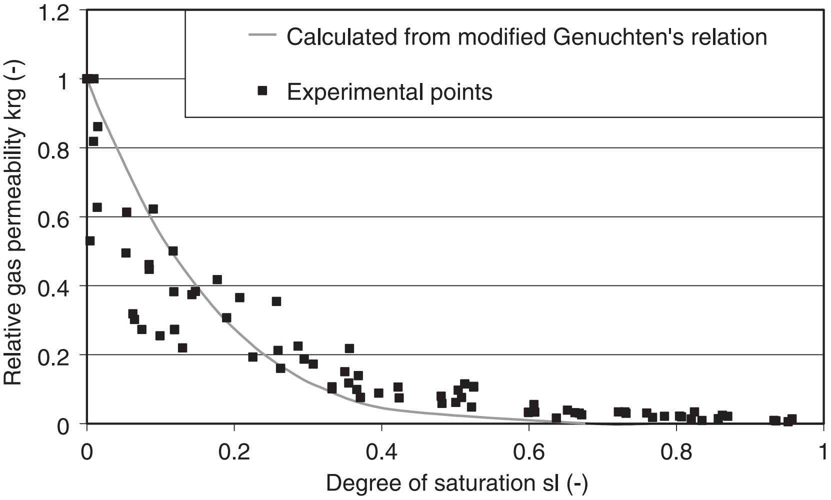

Fig. 7. Adjustment of parameters in order to reach a calculated curve reproducing the experimental relative permeability.

pressure of the gaseous phase. Consequently, we can admit a relation of the following form:

$$\beta = \left(1 + \frac{a}{p_g}\right) \tag{18}$$

The way to obtain k consists, for a dry material, in measuring k_g^{eff} for different levels of air pressure, and defining k as the limit of k_g^{eff} as $1/p_g$ tends towards zero. The relative permeability is calculated for different degrees of saturation, accounting for the previously obtained k value and as $1/p_g$ tends towards zero.

The effective permeability, k_g^{eff}, has been calculated for different degrees of saturation and gas pressure gradients (Fig. 4). The results confirm the very good linearity between k_g^{eff} and $1/p_g$.

The relative gas permeability for all the samples of the same material were calculated and the values are shown in Fig. 5. The relative gas permeability deduced from the initial Van Genuchten's relation is also represented in this figure. Obviously the calculated permeability does not describe the experimental behaviour.

Both of the curves do not have the same asymptote for $s_l = 0$. For a small variation of water saturation in the 0–0.2 range, the experimental gas relative permeability decreases strongly, contrary to the Van Genuchten relation (Fig. 5).

Without changing the form of the relations Eqs. (9), (12) and (14), the parameters seem to be unsuitable for our cement materials and should be revised. We proceeded then as follows: The m value, the relation between m and n being conserved, is adjusted until the calculated capillary pressure curve describes the experimental curve (Fig. 6). Then the value of p is also adjusted to reproduce as well as possible the experimental relative air permeability. The result is presented in Fig. 7. The relative agreement between calculated and experimental values is due to the fact that Eq. (14), giving the relative permeability, is imposed. The values of all the parameters that have been determined on the studied concrete, starting from capillary and relative permeability curves, are given in Table 1.

Table 1
New values for the coefficients of the Van Genuchten's relations

a	p	m	n
11.087	5.5	0.56	$m = 1 - 1/n$

An important difference concerning p is noticed between our results ($p = 5.5$) and the value given by Van Genuchten for soils, which was equal to 0.5. This could be explained by the important difference between the microstructures of concrete and soils, characterised by global porosity, distribution of pore size diameters, tortuosity, connectivity and fineness of pores. The microstructure of the material indeed has a great influence on the p value.

Another set of parameters could have been eventually determined if the relation between m and n had been modified. This procedure would, in this case, change the closed-form relations giving the relative permeability (Eqs. (10)–(12) and (14)).

6. Numerical simulation of the experimental behaviour

The physical modeling as described in Section 2 has been implemented in the numerical code, Delphin [13]. This is a bidimensionnal macroscopic model of coupled heat and mass transfer in porous materials based on a phenomenological description of advective and diffusive transport processes. For more details, see Refs. [14,15].

Experiments described in Section 5 have been numerically simulated to compare the experimental air flow through the specimens to the calculated flow, accounting for the same pressure conditions.

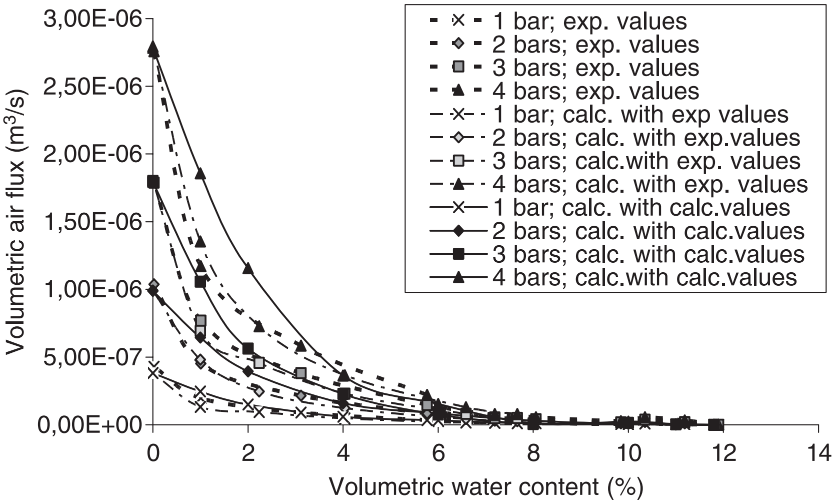

Fig. 8. Numerical simulation and experimental values of airflows obtained during experimentation.

In order to launch the simulations, the following two sets of values have been attributed to the different parameters such as relative permeability and vapour diffusivity.

For the first set, the relative gas permeability was determined from the experimental values, with all the other parameters calculated. The purpose of this work was to validate the numerical calculation and the values attributed to the other parameters.

Then all parameters, including, k_g^{eff}, were calculated with the relations (Eqs. (5), (9), (12) and (14)) accounting for the values of the coefficients given in Table 1.

Fig. 8 presents different kinds of volumetric air flux curves. Experimental results, obtained for three pressure levels (1 to 4 bar), are shown by the dash lines. Calculated values obtained with experimental gas permeabilities are represented by dot dash lines. Finally, calculated values starting from calculated gas permeabilities are represented by solid lines.

Experimental and calculated values agree very well if the simulations are conducted starting from measured values. This correlation is a little less representative if it is started from entirely calculated values. The difference increases with pressure level. Nevertheless, despite these differences, the results are, in our opinion, satisfying and in any case better than the results we obtained with unrevised values of Van Genuchten's parameters.

7. Proof of the revised values for other materials

In order to check if the value obtained for the exponent parameter p is not only valuable for the concrete realised for this study but is useful for other materials, results from

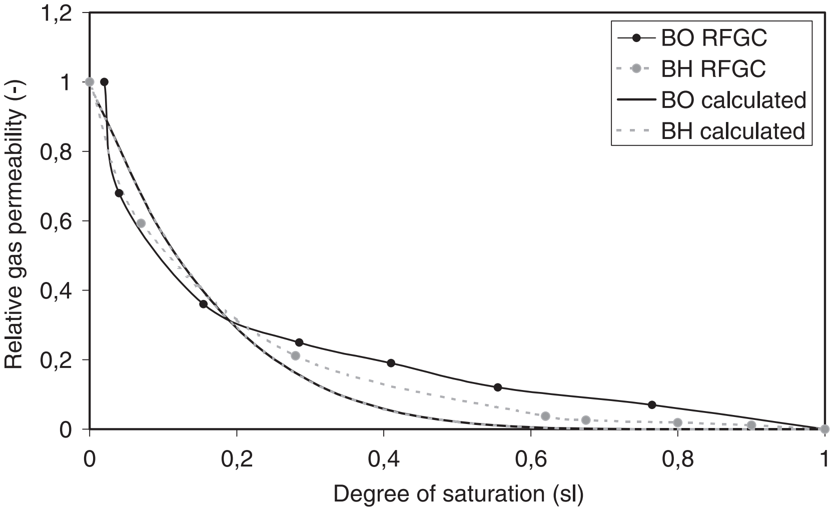

Fig. 9. Experimental and calculated relative permeability for two concretes, BO and BH.

the literature have been analyzed [16] and are presented in Fig. 9.

With p equal to 5.5 (the same value found for the studied concrete), the calculated values give a satisfying representation of the experimental data for the two concretes, BO (ordinary concrete with a compressive strength of 50 MPa after 28 days of curing in tap water) and BH (very high performance concrete compressive strength of 115 MPa after 28 days of curing in tap water), justifying in this way this new value.

This result allows thinking that this new p value could be generalized for all materials cast with cement. Nevertheless, it will be necessary to obtain and to compare more experimental results for mortars and concretes to be able to eventually generalize this result.

8. Conclusions

The aim of this study was to determine if the relations given by Van Genuchten and based on the work of Mualem, relations that have been largely validated for soils, can be used in predicting unsaturated moisture movement in cement-based materials.

In the first part, we mentioned the different relations that give the relative gas and liquid permeability. We also gave an expression and some values of the coefficients that appear in these relations and are valid for soils.

In the second part, we directly measured the effective air permeability of concrete specimens for different water contents and different air pressure levels. Absolute and relative air permeability can be deduced from these experiments.

Comparison between experimental values and calculated values using Van Genuchten's parameters obviously shows that the values of some parameters have to be revised.

The form of the curve that gives the relative permeability k_{rg} versus the degree of saturation s_l shows that for concrete k_{rg} decreases very rapidly with s_l. The first pores that are filled with water seem to have a great influence upon air transport through the material for cement-based materials; this is not exactly the case for soils.

Our approach was to conserve the relation between m and n, which allowed a closed form relation to be obtained for k_{rg}, and to change only the exponent coefficient p in its expression. The value $p = 0.5$ is then replaced by $p = 5.5$.

This new value allows the airflow evolution obtained during the experimental measurement to be accurately calculated.

The exploitation of experimental results from the literature concerning different concretes that account for the same value of p, lets think that this new value of p could be used to calculate the air relative permeability for all cement-based materials.

Acknowledgements

Thanks to John Grunewald (Dipl-Ing, Technical University of DRESDEN) who allowed us to use and develop his numerical code.

References

[1] M.Th. Van Genuchten, D.R. Nielsen, On describing and predicting the hydraulic properties of unsaturated soils, Ann. Geophys. 3 (5) (1985) 615–628.

[2] M.Th. Van Genuchten, A closed-form equation for predicting the hydraulic conductivity of unsaturated soils, Soil. Sci. Soc. Am. J. 44 (5) (1980) 892–898.

[3] Y. Mualem, A new model for predicting the hydraulic conductivity of unsaturated porous media, Water Resour. Res. 12 (3) (1976) 513–522.

[4] B.M. Savage, D.J. Janssen, Soils physics principles validated for use in predicting unsaturated moisture movement in Portland cement concrete, ACI Mater. J., Jan.–Feb. (1997) 63–70.

[5] L. Luckner, M.Th. Van Genuchten, D.R. Nielsen, A consistent set of parametric models for the two-phase flow of immiscible fluids in the subsurface gaseous phase, Water Resour. Res. 25 (10) (1989) 2187–2193.

[6] R.H. Brooks, A.T. Corey, Properties of porous media affecting fluid flow, J. Irrig. Drain. Eng. 92 (IR2) (1968) 61–66.

[7] N.T. Burdine, Relative permeability calculations from pore-size distribution data, Trans. Am. Inst. Min. Metall. Pet. Eng. Inc. 198 (1953) 71–78.

[8] M. Mainguy, Modèles de diffusion non linéaires en milieu poreux: Application à la dissolution et au séchage des matériaux cimentaires, PhD thesis, ENPC Paris, France, 1999, 262 pp.

[9] M. Carcasses, A. Abbas, J.P. Ollivier, J. Verdier, An optimised preconditioning procedure for gas permeability measurement, Mater. Struct. 35 (2002) 22–27.

[10] J.J. Kollek, The determination of the permeability of concrete to oxygen by the CEMBUREAU method—a recommendation, Mater. Struct. 22 (1989) 225–230.

[11] D. Perraton, La perméabilité au gaz des bétons hydrauliques, PhD thesis, INSA de Toulouse, France, 1992, 225 pp.

[12] L.G. Klinkenberg, The permeability of porous media to liquids and gases, Drill. Prod. Pract. API (1941) 200–213.

[13] J. Grunewald, Documentation of the Numerical Simulation Program DIM 3.1, Theoretical Fundamentals, Institute of Building Climatology, University of Technology, Dresden, Germany, 1999.

[14] P. Haüpl, J. Grunewald, H. Fechner, Coupled heat and moisture transfer in building structures, Int. J. Heat Mass Transfer 40 (7) (1997) 1633–1642.

[15] J. Grunewald, Konvektiver und diffusiver Stoff-und Energietransport in kapillarporösen Baustoffen, PhD thesis, University of Dresden, Germany, 1997, 158 pp.

[16] RFGC Transferts 2000, Transferts dans les bétons et durabilité, Revue Française de Génie Civil, 2001, 396 pp.