

LABORATORY MEASUREMENTS AND PREDICTIVE EQUATIONS FOR GAS DIFFUSION COEFFICIENT OF UNSATURATED SOILS

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ABSTRACT

Molecular diffusion is an important mechanism for gas transport through soil covers placed over municipal dumps (from which bio-gas must not escape) and above acid generating mine tailings (where oxygen availability must be controlled). Gas flux through cover systems depends on the effective diffusion coefficient D_e of the cover materials. In this paper, the authors describe a laboratory procedure for oxygen diffusion tests, and present results obtained on different types of porous materials at various degrees of saturation. The determination of D_e from experimental measurements is based on analytical and numerical solutions to the diffusion equations. The measured values of D_e are compared to values calculated from two predictive models. A new expression is finally proposed as a simple means of estimating D_e .

RÉSUMÉ

La diffusion moléculaire est un mécanisme important de transport de gaz à travers les couvertures en sols mises en place sur les décharges municipales (d'où le bio-gaz ne doit pas s'échapper) et sur les parc à résidus miniers générateurs d'acide (où la disponibilité de l'oxygène doit être contrôlée). Le flux de gaz à travers les systèmes de recouvrement dépend du coefficient de diffusion effective D_e des matériaux de la couverture. Les auteurs décrivent ici un procédé pour réaliser des essais de diffusion à l'oxygène au laboratoire et présentent des résultats obtenus sur différents types de matériaux poreux à divers degrés de saturation. La détermination de D_e à partir des mesures expérimentales est basée sur des solutions analytiques et numériques aux équations de diffusion. Les valeurs expérimentales mesurées sont comparées aux valeurs calculées à partir de deux modèles de prédiction. On propose finalement une nouvelle expression comme moyen simple d'estimer D_e .

1. INTRODUCTION

There are many natural and man-made systems where transfer of fluids (liquid and gas) in unsaturated materials has to be evaluated. This is the case with soil covers constructed to protect waste disposal sites used for domestic, industrial, mining or other residues, which may represent potential sources of contamination. Such covers generally aim at limiting water infiltration, and also sometimes at reducing migration of gases (methane, CO_2 , radon, etc.) in or out from the wastes. In the mining industry, the gas migration aspect is particularly significant for sulphidic rock wastes and tailings, as an acid leachate can be produced when reactive minerals (like pyrite and pyrrhotite) are in contact with water and oxygen. The ensuing acid mine drainage (AMD) is recognized as a major environmental problem facing the mining industry. In this case, soil cover systems can be used advantageously to reduce oxygen flux from the atmosphere, and thus reduce the rate of acid generation (Nicholson et al. 1989; Collin and Rasmuson 1990; Aubertin and Chapuis 1991; Yanful 1993; Aubertin et al. 1993, 1995).

In recent years, a few layered soil covers have been constructed to control the long term generation of AMD upon mine closure (e.g. MEND 2001). Different materials can be considered for the design of covers, depending on their availability near the site and on the local exposure conditions. In that regard, covers with capillary barrier effects (CCBE) have been evaluated positively, in the

laboratory and in the field (e.g. Aubertin et al. 1995, 1997, 1999; Aachib et al. 1998a, 1998b; Ricard et al. 1999; Dagenais et al. 2001), as an effective method to limit AMD. In a CCBE, the moisture retaining layer, made of a fine textured material (silt or clay) confined between coarse textured soils (sand or gravel), can remain close to saturation for a long time without addition of water (Aachib et al. 1993)

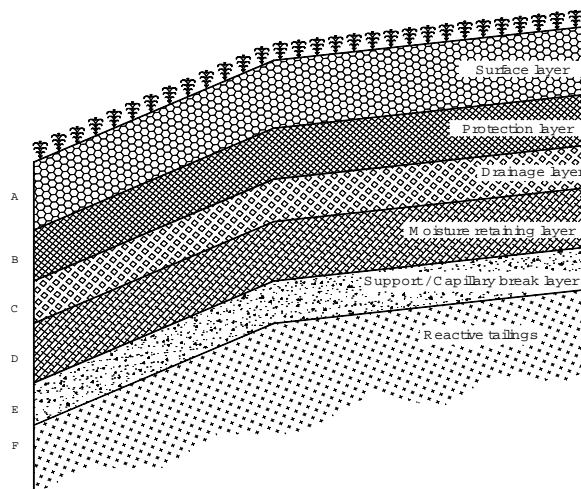


Figure 1. Typical section of a layered CCBE (after Aubertin et Chapuis 1991; Aubertin et al. 1995).

Gas transport through a porous media mainly occurs by molecular diffusion and/or advection through the pores. In fine grained materials such as soils used in covers, gas moves predominantly by molecular diffusion (Collin 1987). The ease with which a gas travels through a cover system under a concentration gradient depends on the effective diffusion coefficient D_e of the fine grained layer. The efficiency against gas migration of a CCBE is due to the fact that the moisture retaining layer remains close to saturation. This limits the flux because the value of D_e for oxygen is much lower in a water filled media than in a rather dry one (with gas filled pores).

Following numerous experimental investigations, many empirical and semi-empirical models have been developed to relate D_e to basic material properties. Nevertheless, for highly saturated media, some work is still needed to establish a simple and efficient measurement technique and a practical equation to predict the oxygen effective gas diffusion coefficient. In this paper, the authors review briefly the theory of gas diffusion in unsaturated soils. Then, an experimental method using a diffusion cell with a decreasing source concentration of oxygen, and the associated analytical and numerical interpretation procedures to evaluate the value of D_e are presented. Experimental results obtained from tests performed at varying water content on various types of materials including soils, non-reactive tailings and a geosynthetic clay liner (GCL) are shown. The authors results, together with data taken from the literature, are used to assess the accuracy of two gas diffusion coefficient predictive models. Finally, a new simple expression for D_e is derived from these two models and from the available data.

2. THEORETICAL APPROACH

Molecular diffusion in relatively fine grained soils can occur both in the interstitial gas and aqueous phase of the media. When the degree of saturation is relatively low, gas transport mostly results from diffusion through the partially gas-filled pores, owing to the much lower diffusion coefficient of O_2 in water (relative to air). The diffusion component in the liquid (water) phase only becomes significant when the degree of saturation S_r is above 85% to 90% approximately. In a CCBE, both components should be considered as S_r is expected to be high in one of the layers.

For one-dimensional gas diffusion, the gas flux is given by Fick's first law (Crank 1975; Hillel 1980) :

$$F = -D_e \frac{\partial C}{\partial z} \quad [1]$$

where F is the diffusive flux of a gas [$ML^{-2}T^{-1}$ or $MolL^{-2}T^{-1}$], D_e the oxygen effective diffusive coefficient [L^2T^{-1}], C the O_2 concentration [ML^{-3} or $MolL^{-3}$] and z the depth or distance [L].

In a transient state, the oxygen distribution over space and time following diffusion through an inert three phase (solid-water-air) media, in which O_2 is neither consumed nor

produced, can be obtained from the following one-dimensional differential equation representing Fick's second law:

$$\theta_{eq} \frac{\partial C}{\partial t} = D_e \frac{\partial^2 C}{\partial z^2} \quad [2]$$

where θ_{eq} is the equivalent diffusion porosity [L^3L^{-3}], and t is the time [T]. The equivalent porosity [L^3L^{-3}] is defined here as (Aubertin et al. 1999, 2000):

$$\theta_{eq} = \theta_a + H\theta_w \quad [3]$$

where θ_a and θ_w are the volumetric air and water contents [L^3L^{-3}] respectively, and H is the dimensionless form of Henry's equilibrium constant. H is equal to the ratio C_w/C_a (C_a is the O_2 concentration in air phase and C_w is the O_2 concentration in dissolved phase). At 20°C, $H \approx 0.03$ for oxygen. In the θ_{eq} expression, θ_a and θ_w are related to the degree of saturation S_r and porosity n in the following manner:

$$\theta_a = n(1 - S_r) \quad [4]$$

$$\theta_w = n S_r \quad [5]$$

The second term of the right hand side of Eq. 3 (i.e. $H\theta_w$) is introduced to take into account gas diffusion in the aqueous phase which, as previously mentioned, is significant when the air phase becomes discontinuous (for $S_r \geq 85$ to 90%).

Solving differential Equation 2 for relatively complex boundary and initial conditions often requires a numerical treatment. This equation can nevertheless be solved analytically under well defined, but relatively simple boundary and initial conditions, for steady as well as transient state conditions, generally using Laplace transform or the method of variables separation (e.g. Crank 1975). The calculated concentration profile $C(z,t)$ can in turn provide the flux (with Eq. 1). Such analytical solutions for O_2 diffusion flux in CCBE have been recently proposed by Mbonimpa et al. (2001).

To apply numerical or analytical solutions, the effective diffusion coefficient D_e has to be known. The laboratory testing procedure and the interpretation method developed to obtain D_e are given below.

3. LABORATORY TESTING AND INTERPRETATION PROCEDURES

A few techniques have been proposed to determine the in situ gas diffusion coefficient in the vadose zone (e.g. Jellick and Schnabel 1986; Rolston 1986; Rolston et al. 1991; Hers et al. 2000). However, it is generally easier and more convenient to evaluate the effective diffusion coefficient D_e under well controlled conditions in the laboratory on representative samples. Available techniques have been described by Sallam et al. (1984), Rolston (1986), Glauz and Rolston (1989), Shackelford (1991), Achib et al.

(1993), Yanful (1993), Tremblay (1995), etc.. The experimental set-up developed and used in this project is shown in Figure 2. To measure oxygen diffusion through inert materials (that do not react with oxygen), a closed reservoir consisting of a 8.4 cm diameter plexiglas cylinder with a length of 20 to 30 cm is used. A "Teledyne 320P/D" sensor fixed in the source reservoir measures the oxygen concentration. The analyzer is calibrated in free flowing air and in pure nitrogen before each test.

Laboratory measurements to evaluate D_e with this set up can be done at various degrees of saturation S_r , up to complete saturation. The tested material (including sand, silts, and non reactive tailings) is humidified and densified, to

the desired porosity n and saturation S_r , directly into the cell. Tested samples typically have a thickness ($2L$) of about 4 cm. The heights H_i and H_e of the source and collector reservoirs may vary slightly with each test. The measurement approach assumes that diffusivity, pressure, water content and temperature are uniform and constant throughout the various components within the cell. For tests performed at high S_r values, the fined grained soil layer is placed on a coarser material layer so that capillary effects help to maintain water in the finer material. The water content of the sample is controlled before and after each test. The volume of voids in the bottom sand layer is included in the collector reservoir volume, which is used to calculate the associated height H_e .

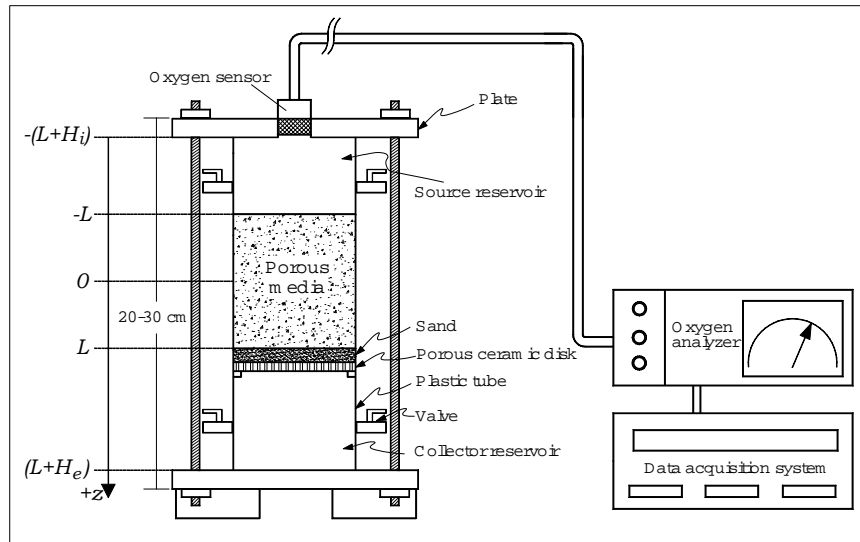


Figure 2. Schematic representation of the diffusion cell used to measure oxygen concentration in the source reservoir (adapted from Aubertin et al. 1995, 1999; Achib 1997).

The experimental procedure used to determine the effective diffusion coefficient D_e is based on decreasing source concentration with time (Shackelford 1991; Yanful 1993; Tremblay 1995; Aubertin et al. 1995). Before beginning the experiment, the cell is purged with humidified nitrogen (to prevent humidity loss in the sample) until the oxygen concentration of the entire cell stabilizes to zero (almost no entrapped oxygen remains in the small sample). The source reservoir is then briefly opened to reach atmospheric conditions (about 21% oxygen). Once the cell is closed, oxygen migrates by diffusion from the source to the receptor reservoir (initially void of O_2) due to the concentration gradient. The closed system is then allowed to approach equilibrium (or steady state). The temporal evolution of oxygen concentration in the source reservoir is monitored with the oxygen analyzer. The interpretation procedure adopted to calculate D_e consists in comparing the experimentally measured concentration to the numerically or analytically calculated values.

If H_i , H_e , and L represent the heights of the source reservoir, of the collector reservoir, and of the sample respectively (see Figure 2), the initial concentration conditions $C(z, t=0)$

can be described by the following top boundary conditions ($z=\pm L$):

$$C(z,0) = C_0 \quad \text{for } -(L+H_i) \leq z \leq -L \quad [6]$$

$$C(z,0) = 0 \quad \text{for } -L < z \leq L+H_e \quad [7]$$

where C_0 is the atmospheric oxygen concentration. For these conditions, the temporal evolution of the oxygen concentration C_i and C_e in the source and collector reservoirs are given by:

$$C_i(t) = C_0 - \frac{1}{H_i} \int_0^t (F)_{z=-L} dt \quad [8]$$

$$C_e(t) = \frac{1}{H_e} \int_0^t (F)_{z=L} dt \quad [9]$$

The concentration C and the gas flux F are considered continuous at the interfaces ($z = \pm L$). The flux (F) in Eq. [8] and [9] is given by Eq. [1]. To facilitate the calculations,

Equations [2], [8] and [9] are made dimensionless by setting $c = \frac{C}{C_0}$, $\xi = \frac{D_e}{\theta_{eq} L^2} t$, $\eta = \frac{z}{L}$, $\beta = \frac{H_i}{\theta_{eq} L}$ and $\gamma = \frac{H_i}{H_e}$. One then obtains:

$$\frac{\partial c}{\partial \xi} = \frac{\partial^2 c}{\partial \eta^2} \quad [10]$$

and

$$c_i = 1 + \frac{1}{\beta} \int_0^\xi \left(\frac{\partial c}{\partial \eta} \right)_{\eta=-1} d\xi \quad [11]$$

$$c_e = -\frac{\gamma}{\beta} \int_0^\xi \left(\frac{\partial c}{\partial \eta} \right)_{\eta=1} d\xi \quad [12]$$

where c is a normalized concentration, c_i and c_e are the normalized concentrations in the source and collector reservoirs respectively.

An analytical solution to Eqs. [10], [11], and [12] can be obtained using Laplace transform (Shair and Cohen 1969; Glauz and Rolston 1989). The solution corresponding to the upper soil boundary ($\eta=-1$) in contact with the source reservoir is expressed as follow:

$$c_i = \left(\frac{C}{C_0} \right)_{\eta=-1} = \frac{1}{1 + \frac{1}{\gamma} + \frac{2}{\beta}} + \sum_{j=1}^{\infty} \frac{A(\alpha_j)}{B(\alpha_j)} \exp(-\alpha_j^2 \xi) \quad [13]$$

where

$$A = \frac{\alpha_j^2}{\gamma} + \frac{\gamma}{\beta^2} \quad [14]$$

$$B = \alpha_j^4 \frac{\beta}{\gamma} + \alpha_j^2 \left(\frac{1}{\gamma\beta} + \frac{\gamma}{\beta} + \frac{1}{2\gamma} + \frac{1}{2} \right) + \frac{\gamma}{\beta^3} + \frac{\gamma}{2\beta^2} + \frac{1}{2\beta^2} \quad [15]$$

and α_j are the roots of:

$$\tan(2\alpha_j) = \frac{\alpha_j \beta (1+\gamma)}{\alpha_j^2 \beta^2 - \gamma} \quad [16]$$

These Equations have been implemented into MATLAB to obtain the concentration variation over time. More details are given in Aachib et al. (2002). Equations [10] to [12] can also be solved numerically using a technique proposed by Talbot (1979). The computer program POLLUTE (Rowe et al. 1994) integrates such technique. POLLUTE was initially developed for modeling solute transport through porous media. It has been adapted here for solving gas diffusion by using the equivalent porosity θ_{eq} ; the effective diffusion coefficient D_e is then given by $\theta_{eq} D^*$, where D^* is the bulk gas diffusion coefficient in a porous medium (D^* is the value introduced into POLLUTE). More details on the experimental procedure and interpretation can be found in Mbonimpa et al. (2001) and Aachib et al. (2002).

The authors have conducted many experiments on various materials at different S_r , up to full saturation (e.g. Aubertin et al. 1995, 1999, 2000). For each test, the effective diffusion coefficient (D_e) is determined by fitting the analytical and/or numerical concentration curve to the experimental data (as shown in Fig. 3). As can be seen, results obtained from both calculation methods (analytical and/or numerical) are practically identical for these tests.

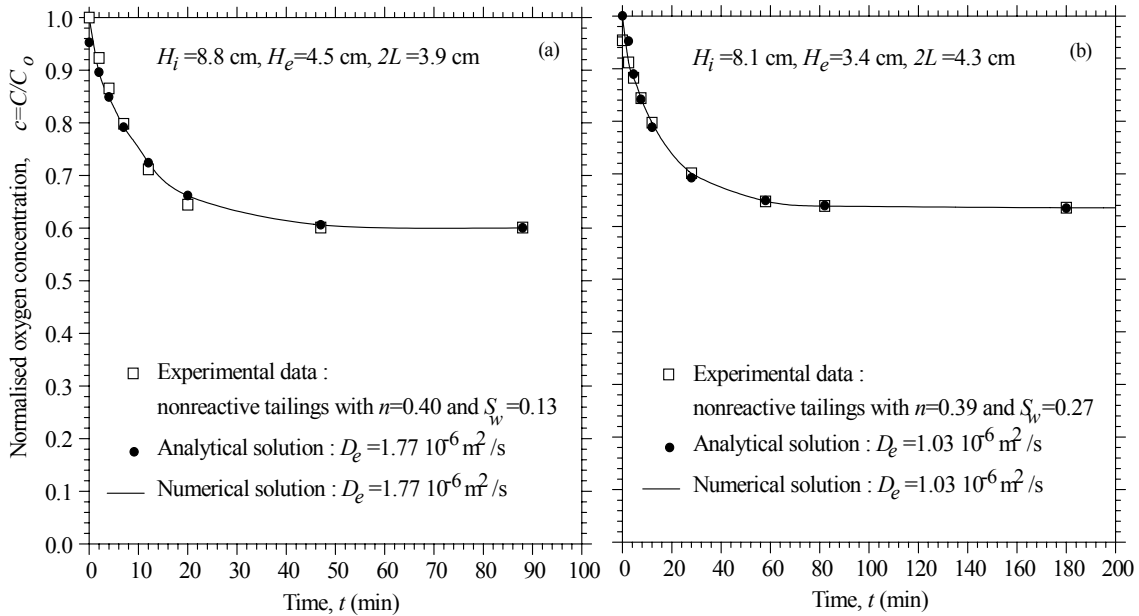


Figure 3. Concentration profiles measured and calculated in the source reservoirs (taken from Aachib, 1997, and Aachib et al. 2002); in this figure, S_w represents the degree of saturation S_r .

Plots of measured D_e values versus the degree of saturation S_r will be shown below (in Figure 6). The results from this investigation were obtained on sand, silty clay, till and tailings, and some data were also taken from the literature on mine tailings (Reardon and Moddle 1985; Aachib 1997), sand (Aachib 1997), and geosynthetic clay liners (Aubertin et al. 2000).

4. SEMI-EMPIRICAL MODELS TO PREDICT D_e

Experimental tests to evaluate D_e can be difficult to conduct, time consuming, and costly. Furthermore, the obtained results are often erratic or inconclusive. For the preliminary phases of a project and to help validate some questionable data, empirical and semi-empirical prediction models can be used to estimate D_e (e.g. Penman 1940; Marshall 1959; Millington 1959; Currie 1961; Troeh et al. 1982; Sallam et al. 1984; Reardon and Moddle 1985; Elberling et al. 1994; Jin and Jury 1996). Two physically-based (semi-empirical) models have been retained for this investigation. The first model considered was proposed by Millington and Quirk (1961) (M-Q model):

$$D_e = \frac{\theta_a^{10/3}}{n^2} D_a^0 \quad [17]$$

where D_a^0 is the free (undisturbed) diffusion coefficient in air; $D_a^0=1.8 \times 10^{-5} \text{ m}^2/\text{s}$ for oxygen at 22°C (e.g. Fredlund and Rahardjo 1993). Equation 17 can also be rewritten as:

$$D_e = D_a = \theta_a T_a D_a^0 \quad [18]$$

with

$$T_a = \frac{\theta_a^{7/3}}{n^2} = n^{1/3} (1 - S_r)^{7/3} \quad [19]$$

where T_a is the gas-phase tortuosity. The Millington-Quirk model doesn't take into account gas diffusion through the water phase, so that for $S_r=1$ (saturated medium), it leads to an effective diffusion coefficient of zero ($D_e=0$), which is an unrealistic value.

The other model used here does consider diffusion of O_2 through the water phase in a partially saturated porous medium. It has been developed by Collin (1987), who modified the Millington and Shearer (1971) (M-S model) (see also Collin and Rasmuson 1988). Using this approach, one can express D_e as follows:

$$D_e = D_a + H D_w \quad [20]$$

where D_a and D_w are the effective diffusion coefficient in the gas and water phases respectively. D_a is given by equation 18 while D_w can be defined as:

$$D_w = \theta_w T_w D_w^0 \quad [21]$$

where D_w^0 is the free diffusion coefficient in water ($D_w^0 = 2.2 \times 10^{-9} \text{ m}^2/\text{s}$ for oxygen at room temperature; e.g. Fredlund and Rahardjo 1993) and T_w is the gas tortuosity in the water phase. With this model, the tortuosity parameters T_a and T_w depend on porosity, connectivity and shape of the pore canals. Using the equations proposed by Collin and Rasmuson (1988), the following expressions are obtained:

$$T_a = (1 - S_r)^2 [n(1 - S_r)]^{2x-1} = \frac{\theta_a^{2x+1}}{n^2} \quad [22]$$

and

$$T_w = S_r^2 (n S_r)^{2y-1} = \frac{\theta_w^{2y+1}}{n^2} \quad [23]$$

with

$$[n(1 - S_r)]^{2x} + [1 - n(1 - S_r)]^x = \theta_a^{2x} + (1 - \theta_a)^x = 1 \quad [24]$$

$$(n S_r)^{2y} + (1 - n S_r)^y = \theta_w^{2y} + (1 - \theta_w)^y = 1 \quad [25]$$

When Equations [24] and [25] are used to evaluate the values of exponents x and y , it is observed that: x increases while y decreases when the degree of saturation S_r increases; x and y increase with n ; $x = y$ for $S_r = 0.5$; and x for $S_r = 0$ equals y for $S_r = 1$ (Aachib et al. 2002).

The oxygen effective diffusion coefficients obtained with the above models are plotted as a function of the degree of saturation in Figure 4, for porosity values $n=0.40$ and $n=0.80$. As one can see on this figure, the effect of introducing D_w in the model becomes noticeable when $S_r \geq 85\%$ approximately.

5. PERFORMANCE OF THE PREDICTIVE MODELS

Figure 5 compares experimental results with values of the predicted effective diffusion coefficient. The predicted values obtained with these two models are usually in good agreement with measured values, at least for $S_r \leq 90\%$ (or when $D_e \geq 10^{-9} \text{ m}^2/\text{s}$). Based on these results and on those two models, the authors propose a somewhat simplified expression:

$$D_e = \frac{1}{n^2} [D_a^0 \theta_a^p + H D_w^0 \theta_w^p] \quad [26]$$

where p represents a material constant. Figure 6 shows how D_e varies according to p for a given porosity value $n=0.4$. In this figure, the experimental values (at various n) are also presented. In most cases, it has been found that a good estimate of D_e is obtained with $p=3.5$, while values of p from 2.7 and 4.0 usually provide the maximum and minimum values of D_e respectively.

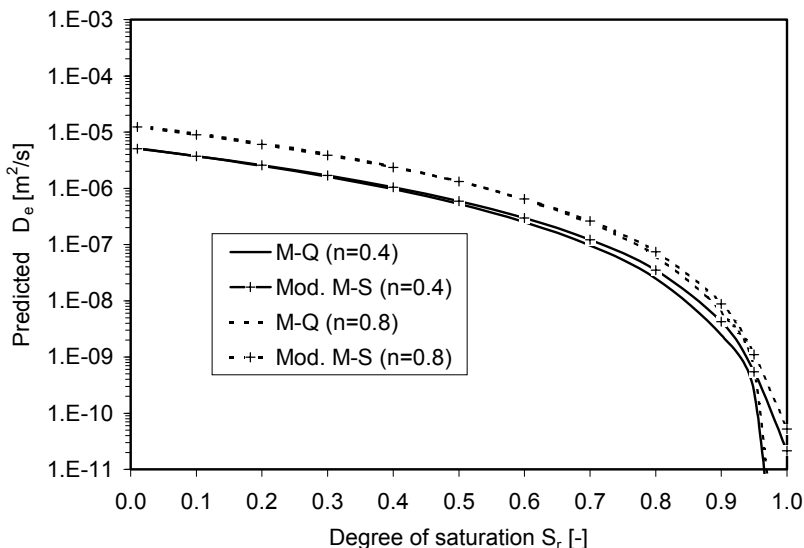


Figure 4. Plotting of D_e as a function of the degree of saturation S_r (for $n=0.40$ and $n=0.80$).

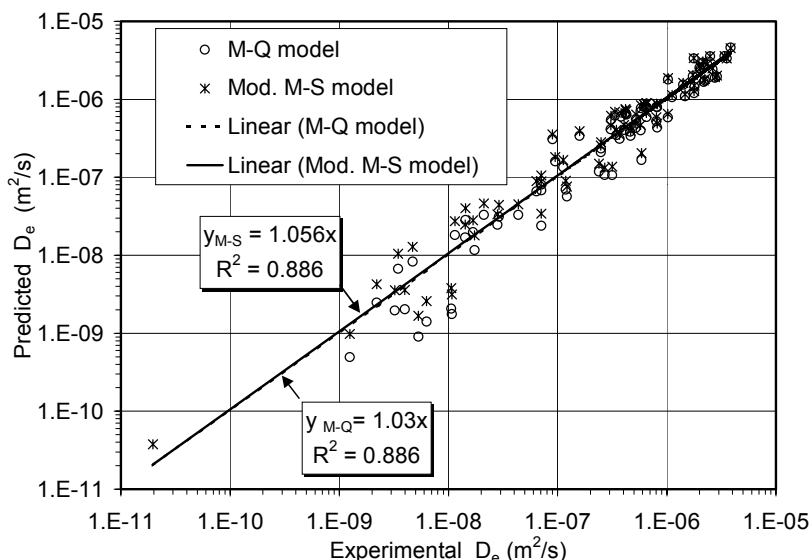


Figure 5. Comparison of experimental and predicted D_e using the two selected models for partially saturated materials.

According to a recent analysis of data from different sources, it has been found that $p=10/3$ (as in Equation 17) provides the best statistical prediction (Aachib et al. 2002).

6. DISCUSSION

The results presented above illustrate how the effective oxygen diffusion coefficient D_e of inert materials (that do not react with O_2) can be obtained experimentally, or estimated using predictive models. The same approaches can be used for other gases. Gas diffusion tests can also be performed in situ using a decreasing source concentration approach (see Mbonimpa et al, this conference), or other techniques (e.g., Jellick and Schnabel 1986, Rolston 1986; Rolston et al. 1991; Hers et al. 2000).

For reactive materials however, such as organic matter or sulphidic tailings, a reaction rate parameter K_r is necessary (beside D_e) to analyze experimental results with Fick's laws. For that purpose, the laboratory procedure has been modified by the authors so that the two parameters can be determined simultaneously from laboratory tests where O_2 concentration is monitored in both reservoirs (source and collector) of the cell presented in Figure 2. Details on this procedure and associated interpretation can be found in Mbonimpa et al. (2001). A procedure used to evaluate D_e and K_r simultaneously in the field is presented in this conference by Mbonimpa et al. with some applications on CCBE presented by Bussi re et al. (this conference).

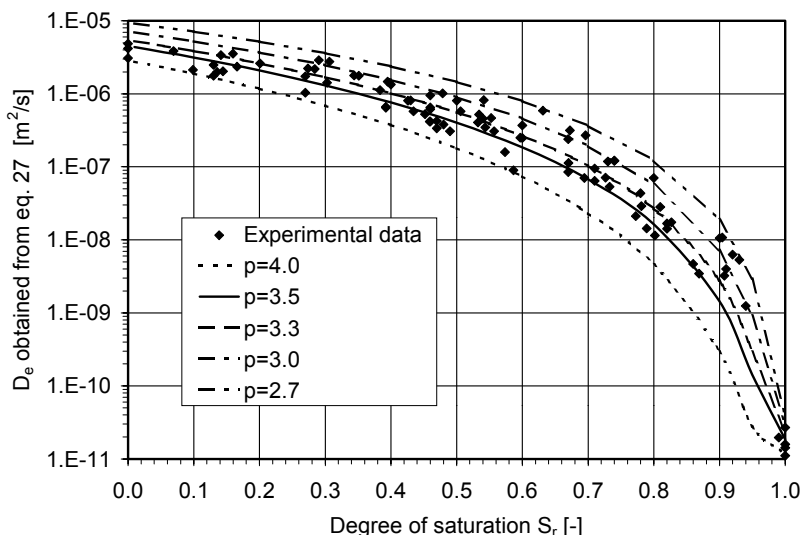


Figure 6. Influence of exponent p in Equation 26 on the predicted values for the effective diffusion coefficient D_e , for a porosity n of 0.40; experimental values are also included (for different n values).

7. CONCLUSION

In this paper, the authors briefly recall an experimental procedure to measure D_e from the temporal evolution of oxygen concentration in the source reservoir of the diffusion cell presented in Figure 2. The effective diffusion coefficient D_e is obtained by iteratively matching the measured oxygen concentration and the concentration obtained numerically or analytically, by solving Fick's laws for the initially and boundary conditions corresponding to the diffusion cell. Diffusion tests have been performed on variably saturated porous materials. The obtained results are compared to values calculated from two predictive models that relate the value of D_e to basic material properties (i.e porosity n and degree of saturation S_r). Predicted and measured values are generally in good agreement. Based on these two models and on available data, a simplified expression to estimate D_e is proposed.

8. ACKNOWLEDGEMENTS

The post-doctoral grant provided to the last author (Mamert Mbonimpa) by the Institut de Recherche en Santé et Sécurité du Travail du Québec (IRSST) is thankfully acknowledged. The authors also received financial support from NSERC and from the participants to the NSERC Polytechnique-UQAT Industrial Chair on Environment and Mine Wastes Management (www.polymtl.ca/enviro-geremi).

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