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MODELLING OF GAS MIGRATION IN CLAY

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RESUMEN

Actualmente se estudia en Bélgica, Francia, España y los Países Bajos la opción de disponer residuos radioactivos en un manto de arcilla profundo. Inmediatamente después del cierre y sellado de un repositorio geológico de residuos radioactivos se generarán distintos tipos de gases. Debido a la baja permeabilidad de los materiales arcillosos estos gases no podrán escapar fácilmente. Esto puede llevar a un aumento de la presión de gas en el repositorio y a la creación de caminos preferenciales de migración de gas.

En este trabajo, como un primer paso hacia el desarrollo de técnicas computacionales para modelar el flujo de gas a través de arcillas naturales y compactadas, y avanzar en la comprensión básica del problema de la migración de gas asociado con la seguridad del repositorio, se han explorado las capacidades de un código general para análisis termo-hidro-mecánico acoplado. Para comparar los resultados del modelo con mediciones realizadas en laboratorio se han seleccionado algunos ensayos de inyección de gas sobre muestras de arcillas naturales.

Los resultados muestran que la contribución de un modelo de tipo continuo constituye una herramienta potente para ganar una comprensión mayor de la migración de gas en arcilla, el cual es capaz de predecir la mayoría de los fenómenos observados en experimentos de flujo de gas. Por lo tanto, podría ser apropiado para describir la migración de gas a través de una barrera de arcilla en un repositorio profundo.

ABSTRACT

The option of the disposal of radioactive waste in a deep clay layer is actually studied in Belgium, France, Spain and Netherlands. Immediately after closure and sealing of a geological radioactive waste repository a range of gases will be generated. Due to the low permeability of the clay materials these gases cannot easily escape. This can lead to an increase in pressure in the repository and to the creation of preferential gas pathways.

In this paper, as a first step towards the development of computational techniques to handle flow of gas through compacted and natural clays, and to advance in the basic understanding of the gas migration issues associated with repository safety, the capabilities of a general code for coupled thermo-hydro-mechanical analysis have been explored. Some laboratory gas injection experiments carried out on natural clay samples have been selected in order to compare model computations with measured specimen of natural clay performance.

The results show that the contribution of a continuum type of model is a powerful tool for gaining a greater understanding of the gas migration in clay, which is capable of predicting most of observed phenomena in gas flow experiments. It may therefore be appropriated for describe gas migration through a clay barrier in a deep repository.

INTRODUCTION

The option of the disposal of radioactive waste in a deep clay layer is actually studied in Belgium, France, Spain and Netherlands. Swelling clays are studied as backfill and sealing materials to be used in clay or granite repositories in Belgium, France, Spain, Sweden, Finland, Switzerland and Canada. Clays as host rock or backfill/sealing material are mainly chosen because of their low permeability and good radionuclide sorption properties.

A range of gases will be produced in a radioactive waste repository after sealing and closure. The most important of these gases are hydrogen, methane, helium, radon and iodine. They will be mainly generated by anaerobic corrosion of metals (canisters, drums, overpacks, scrap metal) and also by decomposition of organic waste and radioactive decay. These gases may give rise to a range of potential hazards. The gases generated in bulk could cause pressurization effects within and around repository due to the low permeability of the clay materials. Pressurization could lead to enhance groundwater movement and to the creation of preferential gas pathways. If the bulk gases, hydrogen and methane, were emitted locally at the surface in sufficiently high concentrations in air, they could be ignited. Similarly, the radioactive and toxic gases could pose radiological and toxicological hazards in the biosphere.

In this work, as a first step towards the development of computational techniques to handle flow of gas through natural clays and for gaining a greater understanding of the gas migration issues associated with repository safety, the capabilities of a fully coupled Thermo-Hydro-Mechanical code (THM) has been explored. The code used is CODE_BRIGHT (Olivella et al.[1,2]). It solves for isothermal problems, in a coupled way, the flow of liquid and gas in unsaturated medium which deforms as total stresses, gas pressure and water pressure change. Advective and diffusive flows are considered through generalized Darcy and Fick formulations. Several constitutive laws are available to define the stress-strain relationship of an unsaturated soil. General laboratory gas injection experiments carried out on natural Boom Clay as reported by Volckaert et al.[3], Ortiz et al.[4] and Harrington and Horseman[5,6] have been selected in order to compare model computations with measured specimen performance. Material properties correspond to natural Boom Clay specimens, as described in Delahaye and Alonso[7], have been selected as the reference common set of parameters. They were defined on the basis of available experimental results. A sensitivity analysis has been carried out varying the most significant soil properties of Boom Clay (water retention curve, water and gas relative permeabilities), and specially its mechanical properties, as an aid to understand the variety of interacting phenomena observed in the experiments.

Once the soil model is defined, CODE_BRIGHT is subsequently used to perform the analysis. Therefore a basic consistent framework for hydro-mechanical interactions is maintained. The results of the different analyses carried out will be compared in order to advance in the basic understanding of gas migration. In a final chapter a discussion of the relevance of soil behaviour, on the basis of the analysis performed, will be presented.

THEORETICAL FRAMEWORK

Modelling the problem outlined requires the performance of coupled Thermo-Hydro-Mechanical (THM) analysis that is formulated using a multi-phase, multi-species approach. The theoretical framework is composed of three main parts: balance equations, equilibrium restrictions and constitutive equations.

The formulation adopted in this work has been described in detail in Olivella et al.[1]. The balance equations are established for the porous medium as a whole. The compositional approach (Panday and Corapcioglu[8]) is adopted to establish the balance equations. The balance equations are: internal energy, water, gas and momentum (equilibrium). According to the compositional approach adopted, phase changes do not appear explicitly in the formulation. The assumption of local equilibrium implies that the species concentration in the various phases can be considered as dependent variables. Equilibrium restrictions are given for the concentration of water vapour in gas (psychrometric law) and dissolved dry gas in water (Henry's law). The constitutive equations are as follow: the conductive heat flow is governed by Fourier's law, liquid and gas flow follow Darcy's law and the molecular diffusion of vapour in gas and of gas in liquid is governed by Fick's law. The variation of intrinsic permeability with porosity is given by Kozeny's relationship and the relative permeabilities are made dependent on effective degree of saturation.

In saturated porous materials, mechanical behaviour is best understood in terms of effective stress $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - P_l\,\boldsymbol{m}$, where $\boldsymbol{m}^T$ is the auxiliary vector [1,1,1,0,0,0]. For unsaturated materials such as a compacted soil it is necessary to consider two independent stress variables (Gens[9]). Here net stresses $(\boldsymbol{\sigma} - P_g\,\boldsymbol{m})$ and capillary suction, $(s = P_g - P_l)$ have been adopted. P_g and P_l are gas and liquid pressures. Net stress is the excess of total stress over gas pressure. If full saturation is achieved, net stress becomes effective stress. In this work, a thermoplastic constitutive law as described in detail in Alonso et al.[10] and Gens[11] has been adopted.

Computer code

All the analyses have been carried out using the computer code CODE_BRIGHT (Olivella et al.[1,2]). It is a finite element code designed to solve thermo-hydro-mechanical problems in geological media. Although developed originally for saline media, it can be applied to other geological environments by appropriate selection of the relevant terms of the formulation. The main theoretical aspects of the code can be summarised as follows:

- State variables are: solid velocity, $d\boldsymbol{u}/dt$ (one, two or three spatial directions); liquid pressure, P_l ; gas pressure, P_g ; and temperature T .
- Balance of momentum for the medium as a whole is reduced to the equation of stress equilibrium together with a mechanical constitutive model to relate stresses with strains. Strains are defined in terms of displacements.
- Small strains and small strain rates are assumed for solid deformation. Advective terms due to solid displacement are neglected after the formulation is transformed in terms of material derivatives (in fact, material derivatives are approximated as eulerian time derivatives). In this way, volumetric strain is properly considered.
- Dry gas is considered a single species and it is the main component of the gaseous phase. Henry's law is used to express equilibrium of the dissolved dry gas.
- Thermal equilibrium between phases is assumed. This means that the three phases are at the same temperature.
- Concentration of vapour under planar surface (in psychrometric law), surface tension (in retention curve), dynamic viscosity (in Darcy's law), are strongly dependent on temperature.

The main features of the numerical approach are:

- Linear interpolation functions on segments, triangles, quadrilaterals, tetrahedrons and triangular prisms. Analytical integration or numerical integration depending on the element type.
- Coupled THM joint element for the modelling of gaps and interfaces.
- Implicit scheme for time integration. Newton-Raphson method for solution of the non-linear system.
- Automatic discretization of time. Increase or reduction of time increment according to convergence conditions or output requirements.
- LU decomposition and back-substitution (non-symmetric matrix) to solve the system of equations.
- Convergence criteria: in terms of forces/flows and state variables.
- Output options: Time evolution of variables in nodes or elements, contour maps in the solution domain. Maps of stresses or velocities.

ANALYSIS OF THE MEGAS EXPERIMENTAL RESULTS WITH CODE_BRIGHT

Some experiments reported in MEGAS reports (Volckaert et al.[3]; Ortiz et al.[4]) have been selected in order to compare model computations with measured specimen performance. A typical test sequence comprises: (a) A natural Boom clay plug is prepared and immediately transferred to the permeameter cell of the experimental set-up. (b) Water is injected at the bottom side of the plug. In- and outflow are measured until they become equal and all air has been evacuated from the system. From the measurements the hydraulic conductivity is determined. (c) Gas is then injected at the top of the clay plug with a pressure difference below 1 MPa and the outflow of water is followed. The gas injection is continued while the water pressure at the bottom side of the plug is kept constant. If the output flow remains zero the gas injection pressure is raised step by step. After each step the pressure is kept constant for at least two days before the next step is applied. When the gas pressure has been raised to 3 MPa and still no gas flow can be detected, the water counter pressure is stepwise lowered. Once a gas flow is detected the breakthrough pressure has been reached and the experiment is stopped. Depending on the hydraulic conductivity and length of the plug the experiment takes about 1 to 4 months.

Material properties correspond to natural Boom clay. Water retention and relative permeability parameters were defined on the basis of available experimental results. Parameters are listed in Table 1 for the so-called “base case”. Boundary conditions refer to gas and water pressures and/or flow rates specified at the specimen boundaries. Three tests as described in MEGAS reports mentioned above were selected for the simulation: Oedometer tests No. 23B5.5K2, 23B4.5 and 23B10.5K1. One hundred one-dimensional linear elements were used to discretize the specimens.

A sensitivity analysis was carried out changing some parameters (one at a time) defined for the base case. Changes affected the molecular diffusion of gas in water, the intrinsic permeability, the water retention curve and the soil stiffness.

The reference case led to very high breakthrough times and a very small gas outflow rates (if compared with actual measurements). The final, steady state, distribution of degree of saturation along the sample was however close to actual measurements as indicated in Figure 1 for "case a" (reference case) and "case e" ($D' = 10\,D$, D: coefficient of molecular diffusion of helium in water for the reference case). In order to improve the accuracy of computed results, two significant changes had to be introduced into the material properties: an increased gas permeability and a reduced bulk soil stiffness. Bulk stiffness reduction results in increased volumetric deformations as the gas pressure increases. As a result porosity increases and permeability is further increased. In this way computed gas flow rates may approximate in a better way measured results. However the history of gas release through the downstream end could hardly be reproduced with acceptable accuracy (Figure 2). Measured gas flow rates are significantly larger than computed values even if the specimen stiffness was reduced to unacceptable low values.

It was recognised at this initial stage of the investigation the sensitivity of computed results against relatively minor changes in material parameters. The preceding results, which are discussed in more detail in Delahaye and Alonso[7], did not provide a comprehensive information on the effect of some key constitutive parameters in modelling gas migration experiments. In particular, the gas relative permeability was soon identified as a critical property. Further work along the lines summarised here is reported further on.

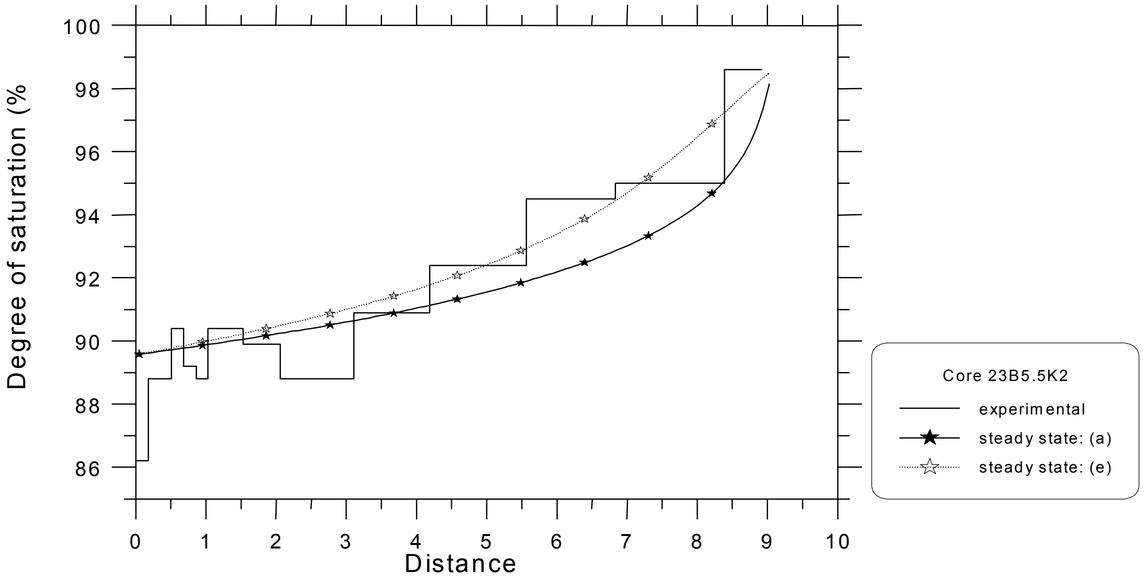

Figure 1: Computed and measured saturation profile of specimen 23B5.5K2.

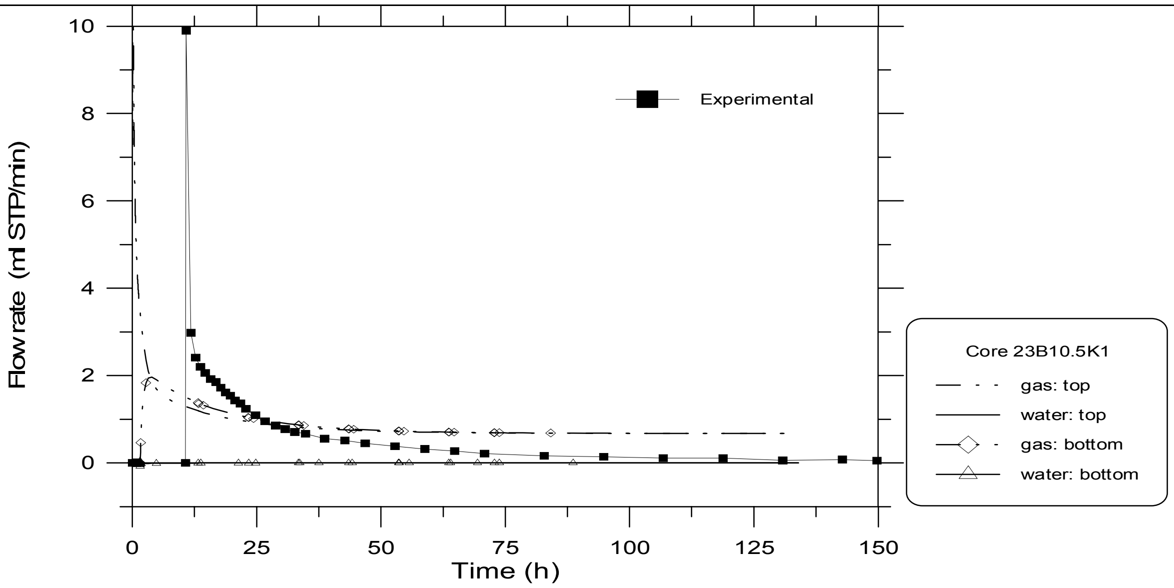

Figure 2: Computed and measured gas flow rate of specimen 23B10.5K1.

TESTING OF CODE_BRIGHT

Test case. The isotropic T4S1 test described in Harrington and Horseman[5,6] was selected for comparison with code calculations. A typical test sequence comprises: (a) resaturation and consolidation at in situ effective stress, (b) measurement of hydraulic conductivity and intrinsic permeability, (c) re-equilibration, (d) gas injection at a number pre-determined pumping rates, and (e) measurement of post-test physical properties. An initial gas pumping rate of 375 μl/hr is used to raise the gas pressure to the point of breakthrough. The injection pressure of the gas at the upstream end of the sample, the water pressure at the downstream end, the confining pressure, in- and outflow rate are measured. Data measured during the gas injection phase is shown in Figure 3. The figure shows the flow rate into the sample, the measured excess gas pressure in the sample (defined as the excess of gas pressure in the upstream end of the sample over the water backpressure, (P_{gi} - P_{wo})) and the gas flow rate out of core. The test shows a breakthrough time close to 0.9×10^6 sec and a marked peak of output gas flow rate when the breakthrough takes place. Once the gas breakthrough has occurred input gas flow rate is decreased in steps and the output gas flow reacts in a parallel way as the excess gas pressure decreases. Once the input gas flow rate is reduced to zero, the excess gas pressure decreases steadily until a final constant value is apparently reached.

Figure 4 shows the definition of the test for computational purposes. An upper reservoir simulates actual test conditions. The reservoir volume and the input mass flow rate was derived from the actual measured gas pressure on the upstream reservoir and the known volumetric flow rate input into the system. The test sample and upper reservoir were simulated by 675 plane strain elements (625 elements for the soil sample and 50 elements for the upstream reservoir). A number of hydraulic and hydro-mechanical simulations were carried out in order to understand the effect of some key material parameters. Figure 4 indicates that the sample was confined under an isotropic stress σ = 4.4MPa and was initially saturated under P_{wo} = 2.23MPa.

Material parameters: Basic soil properties of Boom Clay as described in Delahaye and Alonso[7,12] were selected as a reference set of parameters for all the analyses carried out. Table 1, under "base case", provides the reference hydraulic properties. Also shown in the table are the mathematical expressions used for the retention curve and the relative permeabilities. The Van Genuchten equations (Van Genuchten[13]) were used for the water retention characteristic and water relative permeability. A power law was however found more suitable for the gas relative permeability. A second column of parameters ("alternatives" in Table 1) provides values used for water retention and the range of parameters of the relative gas permeability expression to be used subsequently in a sensitivity analysis for the homogeneous approach. For the basic soil the water retention curve is given in Figure 5 for the porosity (ϕ) equal to 0.39. It is the best approximation to actual laboratory data. A plot of the relative permeability functions for water and gas is shown in Figure 6 in terms of the specimen degree of saturation. Some experimental data provided by Volckaert et al.[3] is also shown. Note that the relative gas permeability has two irreducible extreme values (S_{gs} and S_{gr}), which differ from the theoretical values, one and zero respectively. The role of S_{gr} in controlling the gas breakthrough phenomena has been found to be of maximum relevance.

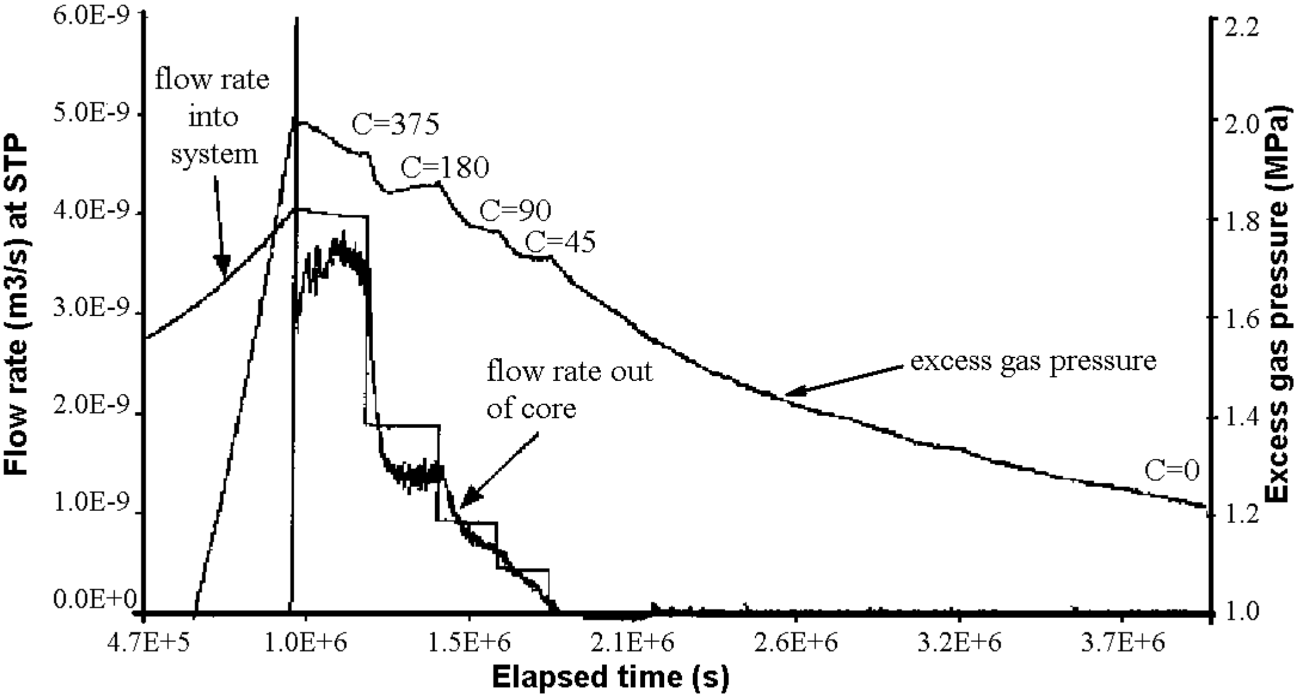

Figure 3: Experimental history T4S1G1. Excess gas pressure ($P_{gi} - P_{wo}$) and volumetric flow rates (STP) into the testing system and out of the specimen plotted against elapsed time (from Harrington and Horseman[5]).

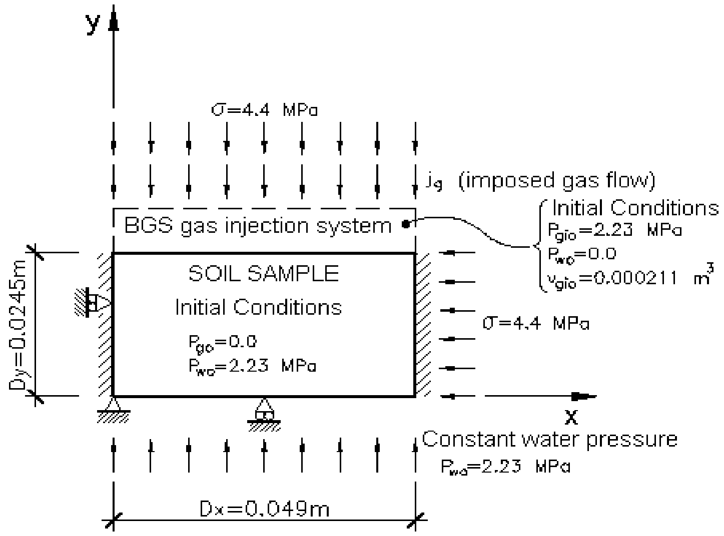

Figure 4: Definition of solved case.

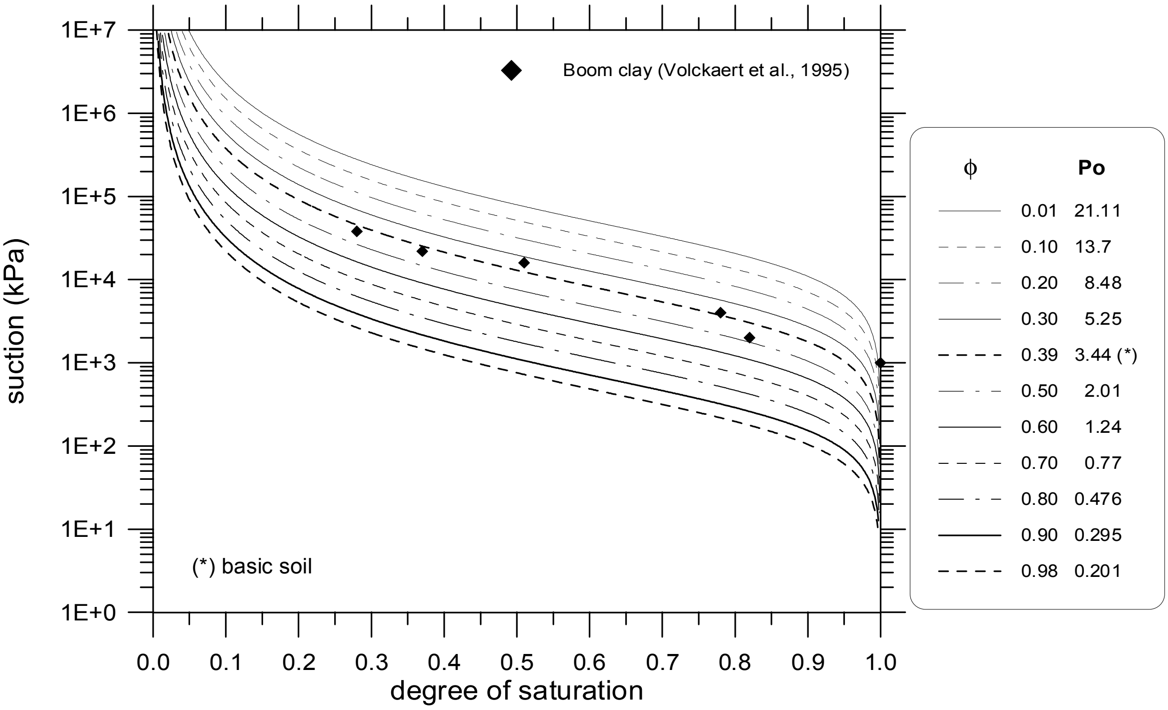

Figure 5: Water retention curve data of Boom clay specimens adopted.

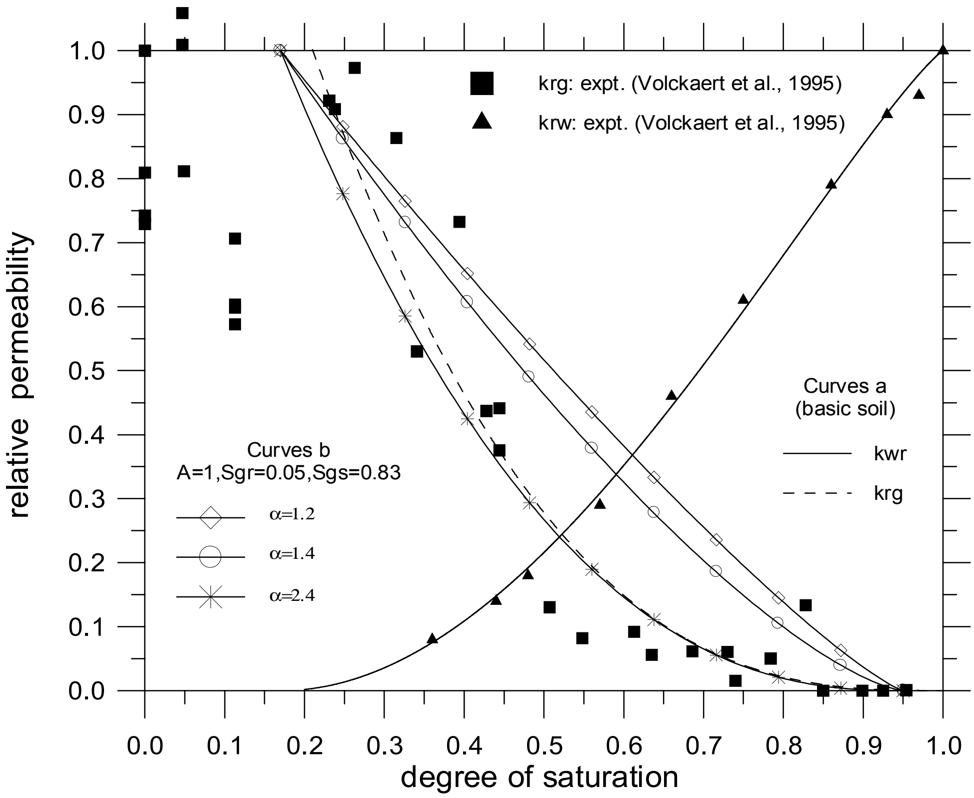

Figure 6: Relative permeability data of Boom clay specimens and relationships adopted.

The full description of the mechanical behaviour of clays is very complex. The constitutive models adopted here (Alonso et al.[10], Gens[11]) require a high number of parameters to be obtained, necessarily, from a limited set of data. In addition, for most parameters there is not a direct manner to be determined but, because of the coupling between different parts of the model, they have to be estimated from back-analysis of laboratory test results. The set of parameters adopted for the Boom clay is indicated in Table 2. They were defined on the basis of experimental results (Alonso et al.[14]; Gens et al.[15]; Volckaert et al.[16]).

Sensitivity analysis: The effect of gas relative permeability has been examined through a sensitivity analysis, varying the *A* and S_{gr} parameters controlling k_{rg} (see Table 1 "Alternatives" column). The remaining flow and mechanical parameters were kept as indicated for the base case in Table 1 and 2. A volumetric gas pumping rate C = 375µl/hr was imposed in all cases in order to raise the gas pressure to the point of breakthrough. The main conclusion of this analysis, which cannot be reported here in detail, may be summarised as follows:

a) Gas breakthrough times are highly dependent on the relative gas permeability function in the sense that breakthrough time increases the higher S_{gr} and the higher the α coefficient.

b) A gas flow rate peak at the time of breakthrough is predicted for nonzero values of S_{gr} and relatively low values of the coefficient α. A continuous increase in gas flow rate until and beyond the gas breakthrough point is predicted when $S_{gr} = 0$ and α is high. The flow peak response of the specimen is enhanced the higher S_{gr} and the lower α.

This pattern of behaviour was found for rigid soil and also for the mechanical parameters given in Table 2. Actually the mechanical parameters given in Table 2 did not modify the purely hydraulic analysis. Most likely this conclusion cannot be extended to other soil stiffnesses. Note in particular that the high apparent saturated preconsolidation stress, p_o^* in Table 2, is high (12MPa) and this implies an elastic behaviour of the specimen throughout the test.

Table 1: Water retention and relative permeability parameters. Experiment T4S1

Relationship	Parameter	Base Case	Alternatives
Retention curve $S_e = \left(1 + \left(\frac{P_g - P_l}{P_o}\right)^{(1-\lambda)^{-1}}\right)^{-\lambda}$ $S_e = (S_l - S_{lr})/(S_{ls} - S_{lr})$	P_o (MPa)	3.443	$P_o(\phi)$
	σ_o (N m^{-1})	0.072	0.072
	λ	0.329	0.329
	S_{lr}	0.	0.
	S_{ls}	1.	1.
Water relative permeability $k_{rl} = \sqrt{S_e}\left(1 - \left(1 - S_e^{1/\lambda}\right)^{\lambda}\right)^2$	λ	1.3	1.3
	S_{lr}	0.17	0.17
	S_{ls}	1.	1.
Gas relative permeability $k_{rg} = A\ S_{eg}^{\alpha}$ $S_{eg} = (S_g - S_{gr})/(S_{gs} - S_{gr})$	A	1.15	1
	α	2.8	2.4, 1.4, 1.2
	S_{gr}	0.	0, 0.02, 0.05
	S_{gs}	0.83	0.83

Table 2: Mechanical parameters

Elastic		Plastic	
k_{io}	2.65 E-02	$\lambda\ (o)$	0.26
k_{so}	3.22 e-03	r	0.564
ν	0.333	β	54.4 MPa^{-1}
α_{ss}	0.00 MPa^{-1}	k	0.00732
α_{is}	0.00 MPa^{-1}	P^c	0.06 MPa
α_{sp}	0.00	M	1
P_r	0.01	α	0.395
		p_o^*	12MPa

Modelling T4S1 Test: A full thermo-hydro-mechanics analysis was run. The best fit was found for the following parameters controlling the gas relative permeability: A = 0.4, α = 1.2, Sgr = 0.038, Sgs = 0.83. The remaining hydraulic and mechanical parameters were kept unchanged. They are given in Table 1 (base case) and Table 2. The full history of gas injection was imposed to the sample. The initial volumetric flow rate (375μl/hr) was kept constant until breakthrough. Later, the flow rate was reduced in steps (180, 90, 45, 0 μl/hr) following the actual test conditions.

Figure 7 shows a comparison between measured excess gas pressure ($\left(P_{gi} - P_{wo}\right)$ and computed values. Also indicated in the figure are the computed upstream and downstream gas pressures.

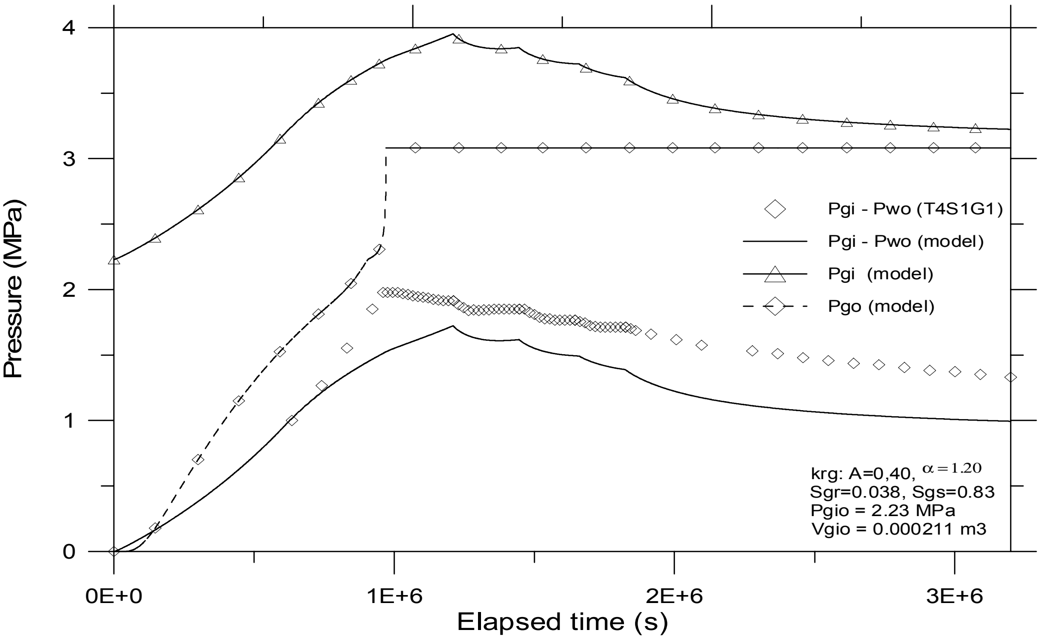

Figure 7: Evolution of gas pressure just inside the sample at the upstream end (P_{gi}), gas pressure just inside the sample at downstream end (P_{go}) and excess gas pressure (P_{gi}-P_{wo}). Some experimental points from T4S1G1 test history have been also indicated.

Measured and predicted gas flow rates are given in Figure 8. The peak behaviour of gas output rate is well reproduced. However, the evolution of the decaying output gas rates beyond peak is smoother than and overpredicts the actual measurements. Output water flow rates are given in Figure 9. The analysis correctly predicts a peak discharge at breakthrough. Low water flow rate at the downstream end is calculated thereafter. Net mean stresses and suction histories at three points along the sample area shown in Figure 10. Suction increases during the first part of the experiment. A maximum value is reached at the breakthrough point and a constant residual value close to 0. 85MPa is computed at steady state. A similar suction value was indicated by Harrington and Horseman[5]. This residual suction value is consistently obtained in the water retention curve for a degree of saturation $S_{ls} = 1 - S_{gr} = 0.962$. A small water flow rate from the downstream end is now wetting the specimen.

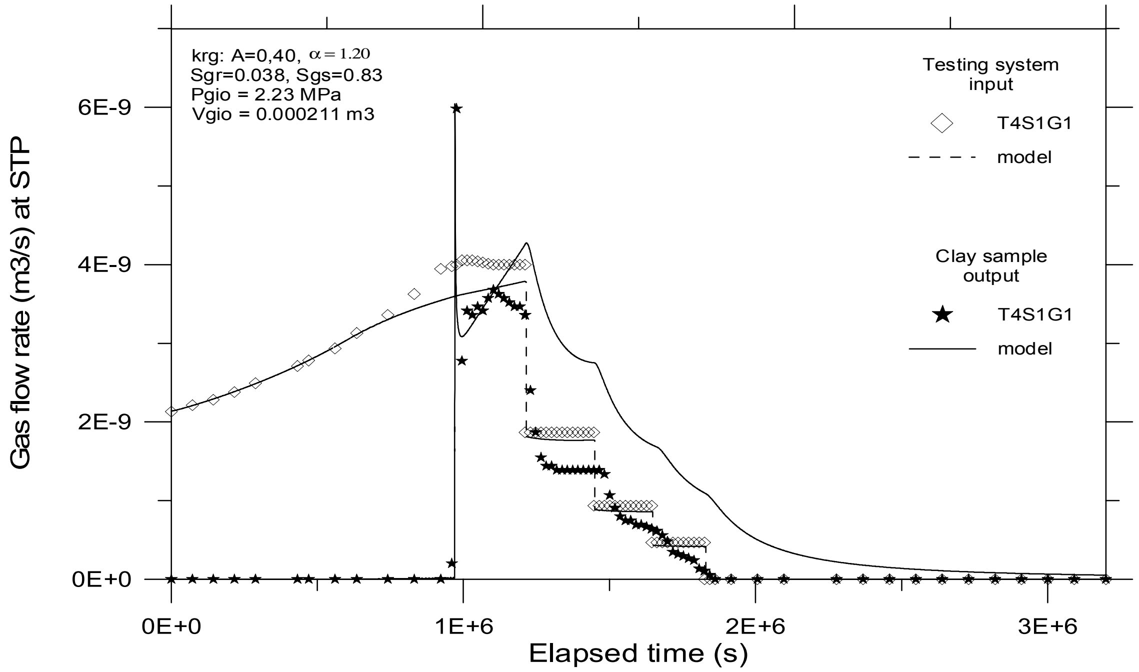

Figure 8: Evolution of volumetric gas flow rates at STP into the testing system and out of the clay specimen. Some experimental points from T4S1G1 test history are also indicated.

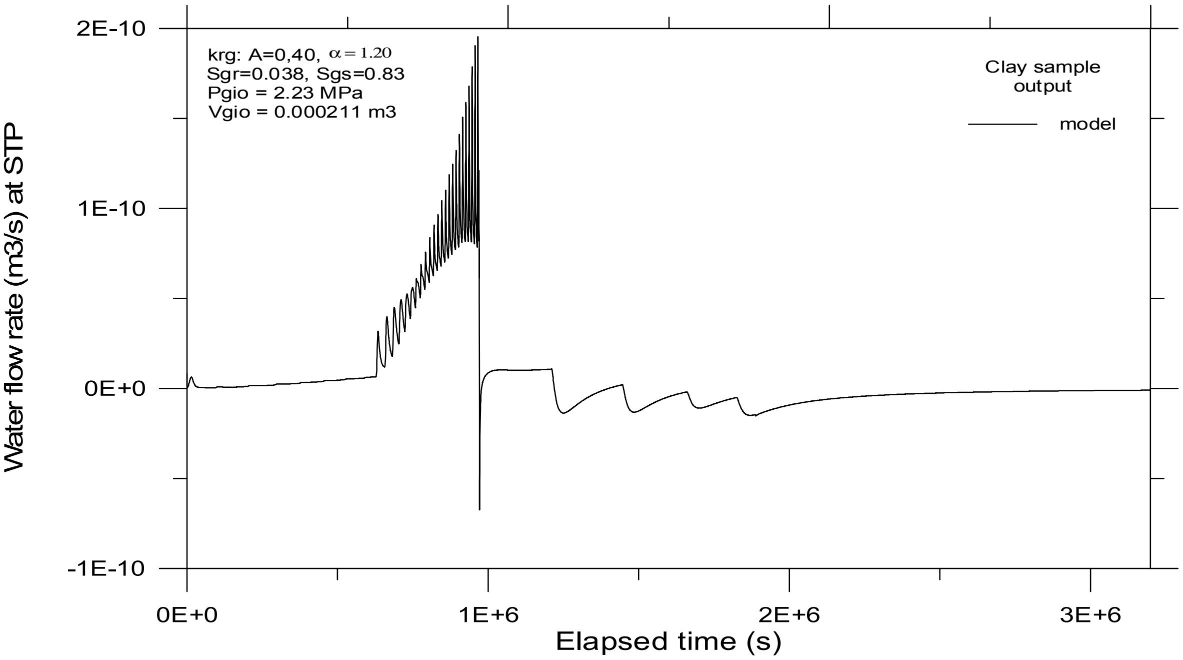

Figure 9: Evolution of volumetric water flow rates out of the clay sample.

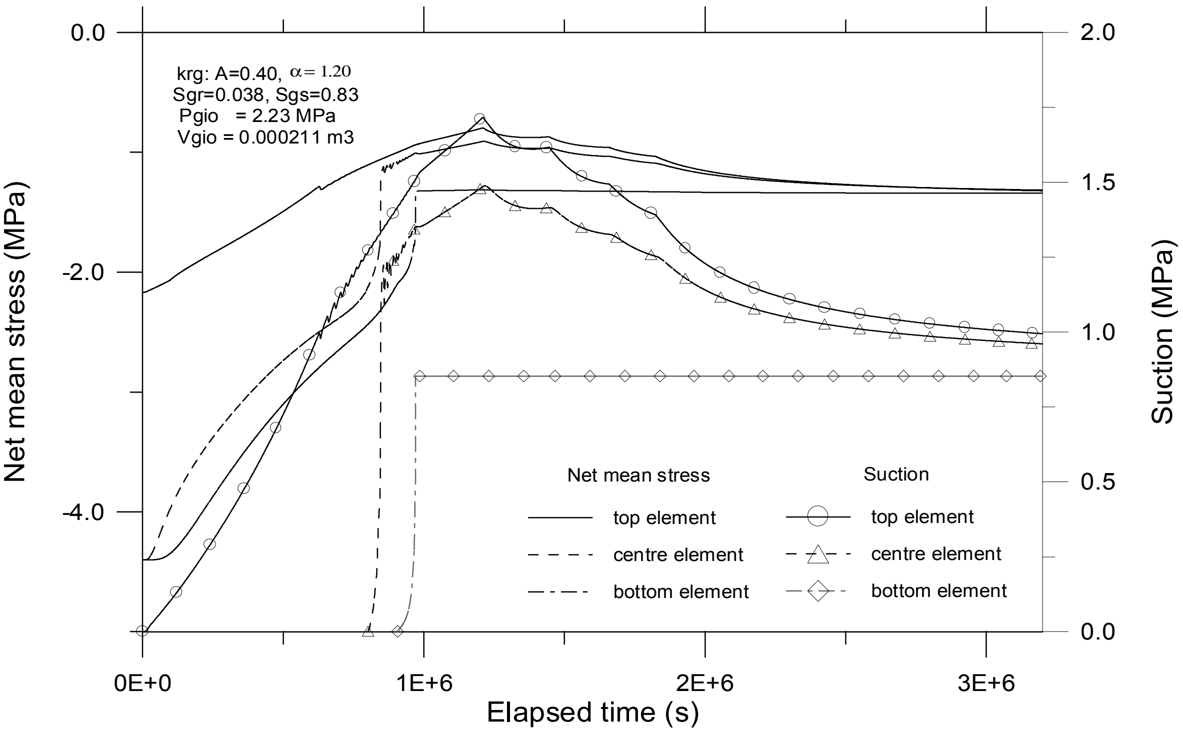

Figure 10: Evolution of net mean stress and suction at three elements (top, centre and bottom) of clay sample.

CONCLUSIONES

A homogeneous analysis of gas migration experiments through CODE_BRIGHT may reproduce in a fairly satisfactory way actual observations. The key variable controlling the results is the gas relative permeability, k_{rg}. If a power law of effective degree of saturation is selected as a mathematical formulation for k_{rg}, the breakthrough time and the peak behaviour of input flow rate are controlled by the power law exponent (α) and the irreducible S_{gr} value. A satisfactory reproduction of the T4S1 test has essentially been achieved by selecting appropriately these two parameters. The remaining hydraulic and mechanical parameters have been directly taken from laboratory experiments on samples of compacted and natural Boom clay. The experimental determination of permeability to gas of an unsaturated specimen is a particularly difficult task. It has been found that the classical formulation in terms of a common intrinsic permeability for water and gas flow is in general not valid. This adds difficulties to the correct interpretation of the gas relative permeability parameters and in particular to the term “A” of the power law. Despite these difficulties it is believed that the continuous approach has an interesting potential to describe gas migration through a saturated barrier. It should be stressed that it is a fully consistent approach, which integrates in a proper way flow and mechanical phenomena. It can therefore be conveniently linked to the evolution of the barrier state as a result of other phenomena of thermal, hydraulic, mechanical or geochemical nature.

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