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EVALUATION OF GAS TRANSPORT PROPERTIES OF BACKFILL MATERIALS FOR WASTE DISPOSAL: H_2 MIGRATION EXPERIMENTS IN COMPACTED Fo-Ca CLAY

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Abstract—Swelling clays may play a major role in the underground disposal of high-level nuclear waste (HLW) in deep geological formations. A multibarrier concept including a waste container, a steel overpack, an engineered barrier consisting of compacted clay (buffer) and the host rock is a potential technique for such HLW disposal. It is anticipated that H_2 will be the main gas generated, mainly by anaerobic metal corrosion during disposal lifetime. After complete resaturation, the clay barrier will have a very low permeability and H_2 may accumulate in the space between the overpack and the clay barrier. This could result in pressures exceeding the resistance of the clay and damage to the entire engineered barrier system (EBS).

The French Atomic Energy Commission (CEA) has performed an experimental program on H_2 gas migration in a French clay referenced Fo-Ca. The tests were conducted with a specific odometer-type cell developed by the Power Reactor and Nuclear Fuel Corporation (PNC) in Japan. Permeability tests on compacted Fo-Ca clay samples of specific dry densities between 1.6 and 1.9 and for water saturation degrees between 70 and 100% provided significant H_2 permeability data (ranging from 10^{-15} to 10^{-21} m^2). Gas migration experiments were also performed to study the behavior of Fo-Ca clay under high gas pressure. Two kinds of gas transport threshold pressures were detected in unsaturated compacted clay. The first one, called "critical pressure", is the pressure over which a gas outflow migrates into the clay. This pressure probably exceeds the capillary pressure of the largest pores within the clay. The second one, called "breakthrough pressure", is detected with increasing gas injection pressure. A sudden rise of gas outflow is subsequently observed. The occurrence of the gas breakthrough is associated with the aperture and propagation of preferential gas transport pathways in the clay. This is apparently linked to the hydromechanical properties and stress state of the clay. When clay samples are not fully water-saturated, the critical pressure is always lower than the breakthrough pressure. However, when the clay is saturated, the 2 pressures appear to be very close. Finally, this study showed that breakthrough pressure—a key parameter for the long-term stability of the clay buffer—varies considerably with the gas injection increments and with the size of the sample. The results of these experiments are consistent with those reported previously by other investigators.

Key Words—Density, Fo-Ca Clay, Gas Critical and Breakthrough Pressures, Gas Preferential Pathways, H_2 Migration, Water Saturation Degree.

INTRODUCTION

Within the framework of management of its HLW, encapsulated vitrified waste and/or spent fuel, and based on current options, France plans to temporarily place its wastes into surface interim storage (for waste cooling) while awaiting possible disposal in a deep geological formation (French Parliament law of December 30, 1991). It should be noted that long-term surface interim storage for HLW is also studied in France by the CEA but that, so far, no decision has been made by the French Agency for Radioactive Waste Management (ANDRA) concerning the final concept. The feasibility and long-term safety of deep disposal depends on the long-term stability of the multibarrier system (waste container, overpack, clay engineered barrier, host rock). It is therefore essential to identify and characterize the phenomena that may jeopardize the long-term performance of the multibarrier disposal option.

It is well known that, after disposal ceases, underground water flow will be the primary transport mechanism for radionuclides through the geosphere and later to the biosphere (Besnus 1991; Besnus and Voinis 1991). The gases generated by mechanisms such as anoxic corrosion of metals (mainly) and possibly by radiolysis and microbial activity will play a less important role in that transport. In the event of significant gas generation, it has been shown that the diffusion transport mechanism is too slow to evacuate the gases that may accumulate in the space between the overpack and the clay engineered barrier fully saturated with water (Harrington and Horseman 1997a). This accumulation of gas could lead to an overpressurization of the clay barrier and the entire EBS, creating microcracks and altering its mechanical (swelling), physical (permeability) and chemical (sorption) performance. This overpressurization could also impact the stability of the near-field by altering water movement and the hydrodynamic regime. Gas generation and gas trans-

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Table 1. Main characteristics of the French Fo-Ca clay.

Mineralogical composition (%)	
Interstratified kaolinite/beidellite	80 ± 2
Kaolinite	4 ± 2
Quartz	6 ± 1
Goethite	6 ± 1
Hematite	0.25
Calcite	1.4 ± 0.2
Gypsum	0.4
Organic matter	0.1
Physical and chemical properties	
<2 μm fraction (%)	90%
Ambient water content (%)	10–12%
Specific density (g/cm³)	2.67
Specific area (m²/g)	430
Liquid limit (%)	112
Plastic limit (%)	50
CEC (meq/100 g)	71
Thermal, hydrological and mechanical properties (1.75 density)	
Swelling pressure (MPa)	13
Hydraulic conductivity (m/s)	$5\ 10^{-14}$
Thermal conductivity (W/m K)	0.95

port in the disposal near-field (a few meters around the waste packages) and the behavior of the compacted clay barrier under high gas pressure are essential issues in assessing the stability of underground disposal of nuclear waste.

OBJECTIVES

Within a deep geological multibarrier disposal system, highly compacted swelling clays are likely to play a major role. Because of their physical, chemical and mechanical characteristics, swelling clays may be used as backfill materials. Because of the very low permeability of the clay, the produced gases may accumulate in the space between the overpack and the clay of the engineered barrier. Production of gases could subsequently greatly increase pressure in the near-field. The results of a previous study (Neretnieks 1985) indicate it could exceed hydrostatic pressure up to 30 MPa. As gas overpressures may impact the stability of the near-field by creating new preferential circulation pathways (microcracks) for deep underground waters and later for radionuclides, it is essential to understand how gases migrate into the clay and how the compacted clay reacts to the resulting pressure. It is well known that hydrogen (H_2) will be the primary gas generated in HLW disposal—mainly through anaerobic corrosion of metals (steels in particular). The H_2 produced will then dissolve in the porewater of the saturated clay barrier. But, since gas diffusion in such a material is very low (about 10^{-11} m² s^{-1}), the gas flow evacuated by this transport mechanism will not be sufficient and a gas phase will from and migrate by advection (Horseman and Volckaert 1995). This is why CEA, in the framework of an R&D program on buffer materials for deep disposal in France, decided to investigate H_2 migration in Fo-Ca clay, a possible candidate for the buffer material.

Within the framework of the collaboration between CEA and PNC, responsible for nuclear waste disposal research in Japan, an experimental program on the migration of H_2 gas in compacted Fo-Ca clay was performed in 1995. The main objectives of this program were to obtain data on the clay H_2 permeability and to investigate gas transport threshold pressures. These parameters were studied by taking into account the dry density and water content of the compacted clay samples.

New data were obtained on H_2 permeability and on gas migration threshold pressures in a dense clay medium. Many studies carried out on gas transport into compacted clays focused on water-saturated materials. Assuming that, in the disposal, gas could migrate into a not-fully saturated clay barrier, the present work focuses on H_2 migration in partly saturated Fo-Ca clay.

MATERIALS

In France, Fo-Ca clay is a potential material for the deep disposal engineered barrier. Fo-Ca clay is a sedimentary clay from the Vexin region, located west of the Parisian basin. It is a Ca-smectite, primarily consisting of interstratified kaolinite and beidellite (lime-containing smectite). Other important minerals are kaolinite, quartz, goethite and calcite. It contains approximately 11% ambient water and its specific density is 2.67. For a 1.75 dry density, its swelling pressure is approximately 13 MPa. For a uniaxial compaction and same density, the hydraulic conductivity of the Fo-Ca clay is around $5\ 10^{-14}$ m s^{-1} (Coulon 1987; Geneste et al. 1990; Lajudie et al. 1995). The physical and chemical characteristics of the Fo-Ca clay are summarized in Table 1.

EXPERIMENTAL EQUIPMENT

The H_2 migration Fo-Ca clay experiments were performed with the HYDROGEN equipment (small cell) designed and built by PNC (Tanai and Asano 1994). This device was constructed within the framework of the Japanese experimental research program called Engineering Scale Test and Research Facility (ENTRY) and was designed to assess the feasibility and performance of deep geological disposal for nuclear HLW in Japan (Apted et al. 1991; Mano and Ishikawa 1993). The purpose of the HYDROGEN experiment is to study the migration of H_2 gas (H_2 migration mechanisms, gas breakthrough phenomenon, transport model development) in the Japanese bentonite known as Kunigel V1 (Ishikawa et al. 1989). For this program, PNC developed 2 types of cells (Tanai and Asano 1994; Tanai et al. 1997): a cell for large specimens (diameter: 300 mm, height: 1000 mm) designed to study scale effects and to determine the distances over

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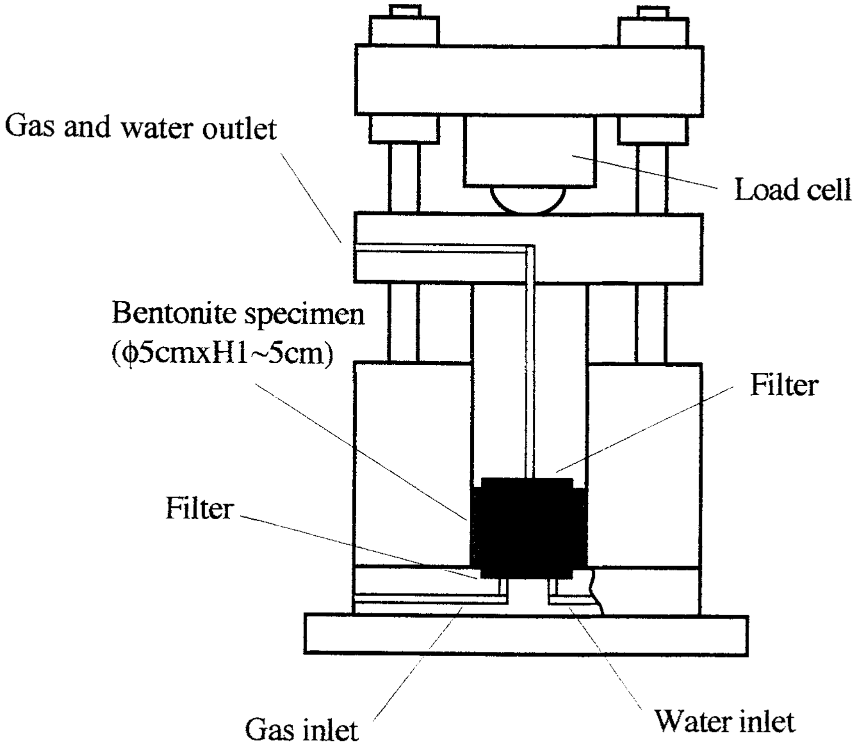

Figure 1. Schematic of the HYDROGEN odometer-type cell.

which gas transport occurs, and a cell for small specimens (diameter: 50 mm, height: 10 to 40 mm) designed for more basic studies (gas permeability, drained water volume, breakthrough pressure, etc.). The small odometer-type cell (Figure 1) is characterized by a maximum inlet gas pressure of 9.3 MPa, a swelling pressure gauge limited to 25 MPa and a water supply pump of 9.8 MPa (maximum).

An H_2 supply system (Figure 2), a control and data acquisition unit (injection pressure, swelling pressure, injection flow rates, etc.) as well as devices measuring downstream gas flow rates (bubble flowmeter and gas collection chamber) and drained water volumes (water collection chamber) are associated with both cells.

PROGRAM AND TEST PROCEDURE

Two types of parameters—gas permeability and gas transport threshold pressures—were processed through gas migration tests over a period of 6 mo. The clay dry density and water saturation degree were taken into account. The H_2 migration experiments were performed on compacted clay specimens with dry densities of 1.6, 1.7, 1.8 and 1.9 for water saturation degrees between 70 and 100%. It should be noted that most of the tests were conducted using unsaturated clay samples. Clay specimens were uniaxially compacted in the cell with a 10-ton Shimadzu press. They were prepared with powdered clay that had been stored for several months in sealed containers in which saline solutions had been used to control the relative humidity. When gas injection tests are performed with saturated samples, saturation is achieved by applying a water pressure higher than the atmospheric pressure. Saturation is complete when a water flow is observed in the water collection chamber.

For a typical gas migration test, an incremental H_2 gas pressure was applied (increments between 0.1 and 0.05 MPa on average) to one of the ends of the compacted clay specimen. During the tests, no water backpressure and no mechanical stress were applied to the specimen. Each pressure step was maintained between 12 and 24 h to reach steady state for the system. When a free gas outflow was observed at the downstream end of the clay sample, gas flow rates were measured to determine the H_2 permeability. This operation was repeated for each pressure increment until the gas breakthrough phenomenon was detected. Effective gas permeability (k) was calculated with Darcy's law as modified by the Hagen-Poiseuille formula:

$$k = 2\,\mu\,Q\,L\,P_0/S(P_1^2 - P_2^2) \qquad [1]$$

where k is the gas permeability (m^2), Q the flow rate (m^3 s^{-1}), L the specimen length (m), P_0, the pressure at which the flow rate is measured (Pa), μ the dynamic gas viscosity (Pa s), S the specimen cross-sectional area (m^2), P_1 the absolute injection pressure (Pa), and

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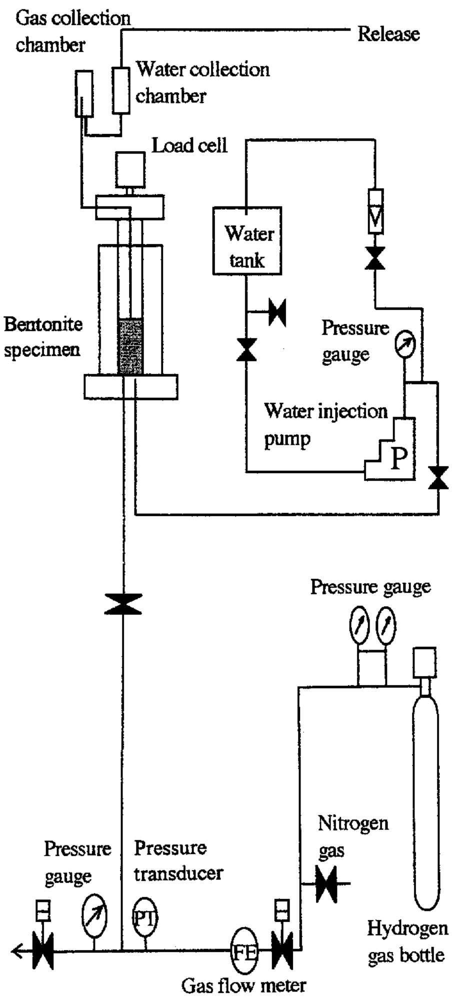

Figure 2. Schematic of the gas injection system related to the HYDROGEN equipment.

P_2 the exit pressure (Pa). Due to the rather large differences of temperature noticed inside the cell during the tests, permeability values were corrected taking into account temperature effects on H_2 viscosity. On the other hand, it should be noted that no evolution of the clay chemical characteristics (pH, Eh, . . .) were monitored during the experiments.

RESULTS AND INTERPRETATION

The main experimental results observed during the 15 H_2 migration tests with compacted Fo-Ca clay samples are summarized in Table 2 (1 MPa = 10^6 Pa = 145 psi).

H_2 Permeability

The H_2 permeability of the compacted Fo-Ca systematically decreases as the water saturation degree increases (Figure 3). When the degree of water saturation increases, more and more capillaries, making up the clay porous network, are filled with water (liquid or vapor). It is therefore increasingly difficult for the gas to migrate. As a result, the material becomes more impermeable to gas. At the same time, the H_2 permeability decreases as the clay density increases (Table 3). This can be explained by the fact that dry density is directly related to porosity. With a specific density of 2.67, the porosities of Fo-Ca clay compacted at dry densities of 1.6, 1.7, 1.8 and 1.9 are 40, 36, 32 and 29%, respectively.

A gas migration experiment with highly compacted and fully water-saturated clays is rather long (between several weeks and several months). As a consequence, only a few tests were carried out using this configuration. However, it was assumed that the variation of H_2 permeability between a 70% and 100% water saturation degree could be regarded as linear and extrapolations were made for total saturation. For dry densities of 1.6, 1.7, 1.8 and 1.9, permeability values should be around 10^{-18}, 10^{-19}, 10^{-20} and 10^{-21} m^2, respectively. These orders of magnitude will have to be confirmed by additional experiments. It should be noted that a gas permeability value of 1.3 10^{-18} m^2 was obtained during an experiment with a 1.6 dry density sample. The Fo-Ca clay H_2 permeability results can be summarized as follows: for a specific dry density and for water saturation degrees ranging from 70 to 100%, the gas permeability globally varies over 3 orders of magnitude. In addition, for a specific water saturation degree, a density variation of 0.1 leads to a gas permeability change of 1 order of magnitude.

Gas Transport Threshold Pressures

In the second part of this work, interesting results were obtained on the hydromechanical behavior of compacted Fo-Ca clay subject to high gas pressures. During controlled H_2 gas injection experiments, 2 kinds of gas transport threshold pressures were observed. The typical results are illustrated by Figures 4a, 4b, 4c and 4d.

A first gas threshold pressure was generally detected in the low-pressure field. When this pressure was reached, a gas outflow (around 0.5 cm^3 s^{-1} minimum) was measured downstream of the clay sample. In samples characterized by 1.6, 1.7, 1.8 and 1.9 dry densities with respective water saturation degrees of 96, 82, 91 and 74%, gas outflows were 1, 0.9, 0.6 and 1.5 cm^3 s^{-1}, respectively (Figures 4a, 4b, 4c and 4d). This gas threshold pressure that we called critical pressure (P_C) is the pressure below which the H_2 gas cannot migrate into the compacted clay medium. In unsaturated clay, all the capillaries do not have the same saturation state, depending on their average access radius (Kelvin-Laplace's law). For a given gas injection pressure, capillaries are therefore likely to be subjected to stress in different ways. Gas transport is facilitated accordingly. When the injection pressure is higher than the capil-

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Table 2. Characteristics and results of H_2 migration tests with compacted Fo-Ca clay.

Test reference	Specimen reference	DD	S_R (%)	⌀, H (cm)	P_C (MPa)	P_B (MPa)	k (m²)	P_I (MPa)
MIG1	FoCa1H16SA	1.6	100	⌀ 5, H 1	4.1	4.3	$1.3\ 10^{-18}$	4.1
MIG2	FoCa1H1669	1.6	69	⌀ 5, H 1	—	—	$1\ 10^{-15}$	0.13
MIG3	FoCa4H1891	1.8	91	⌀ 5, H 4	1.4	3.0	$1.6\ 10^{-19}$	1.6
							$1.6\ 10^{-19}$	2.9
MIG4	FoCa1H1891	1.8	91	⌀ 5, H 1	—	—	—	—
MIG5	FoCa4H1675	1.6	75	⌀ 5, H 4	0.15	0.3	$1.2\ 10^{-16}$	0.2
							$1.6\ 10^{-16}$	0.3
MIG6	FoCa4H1982	1.9	82	⌀ 5, H 4	1.6	>9.3	$3.6\ 10^{-20}$	1.8
							$2\ 10^{-20}$	5.1
MIG7	FoCa4H16SA	1.6	100	⌀ 5, H 4	—	>9.3	—	—
MIG8	FoCa4H1883	1.8	83	⌀ 5, H 4	≈0.6	2.1	$2.2\ 10^{-18}$	0.6
							$3.5\ 10^{-18}$	1.6
MIG9	FoCa4H1898	1.8	98	⌀ 5, H 4	≈1.6	2.1	$5.1\ 10^{-20}$	1.6
MIG10	FoCa4H1696	1.6	96	⌀ 5, H 4	≈0.2	≈0.55	$3.1\ 10^{-18}$	0.35
							$2.7\ 10^{-18}$	0.5
MIG11	FoCa4H1782	1.7	82	⌀ 5, H 4	≈0.13	≈0.8	$1.2\ 10^{-17}$	0.35
							$1.1\ 10^{-17}$	0.5
MIG12	FoCa4H1776	1.7	76	⌀ 5, H 4	<0.15	—	$3.9\ 10^{-17}$	0.2
							$2.7\ 10^{-17}$	0.6
MIG13	FoCa4H1974	1.9	74	⌀ 5, H 4	<5.1	>9.3	$3.1\ 10^{-19}$	0.6
							$2.7\ 10^{-19}$	9.1
MIG14	FoCa4H1873	1.8	73	⌀ 5, H 4	0.15	1.2	$8.2\ 10^{-18}$	0.2
							$4.3\ 10^{-18}$	1.1
MIG15	FoCa4H1789	1.7	89	⌀ 5, H 4	<0.19	0.6	$1.1\ 10^{-18}$	0.2
							$2.3\ 10^{-18}$	0.6

Key: Dry density (DD), water saturation degree (S_R), specimen diameter (⌀), specimen height (H), absolute critical pressure (P_C), absolute breakthrough pressure (P_B), average gas permeability (k) and absolute injection pressure (P_I).

lary pressure of the largest clay pores, gas transport by advection begins. From this moment on, if the injection pressure is maintained, gas can migrate through the sample and a continuous gas outflow is observed. As a consequence, when the gas injection pressure goes up, an increasing flow rate is recorded downstream (exiting the specimen). On the other hand, gas migration experiments showed that gas outflow also depends on gas humidity. During gas injection, the circulation of dry H_2 gas modified the internal hydric conditions of the material. This phenomenon, which has to be taken into account for test interpretation, leads to a significant desaturation of clay samples. For example, after a 6-day test, the degree of water satu-

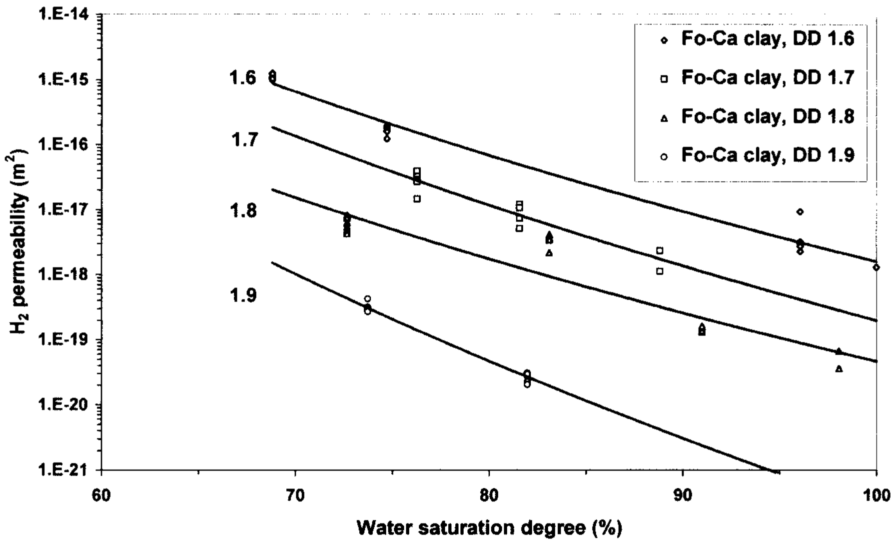

Figure 3. Relationship between H_2 permeability, dry density (DD) and water saturation degree for Fo-Ca clay.

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Table 3. H_2 permeability (m^2), ranges of water saturation degrees (S_R) and dry density (DD) for compacted Fo-Ca clay.

S_R (%)	DD 1.6	DD 1.7	DD 1.8	DD 1.9
73–76	$1.7\ 10^{-16}$	$2.8\ 10^{-17}$	$4.3\ 10^{-18}$	$3.1\ 10^{-19}$
81–83	—	$9.2\ 10^{-18}$	$3.4\ 10^{-18}$	$2.7\ 10^{-20}$
89–91	—	$2.3\ 10^{-18}$	$1.3\ 10^{-19}$	—
96–98	$3.9\ 10^{-18}$	—	$5.1\ 10^{-20}$	—

ration of the 1.8 dry density sample was 83%, compared to an initial saturation of 91%.

When the gas injection pressure was increased stepwise, a second gas threshold pressure was observed. This pressure, called "breakthrough pressure" (P_B), was detected when upstream and downstream gas flow rates sharply increased. Breakthrough gas pressure was higher than critical pressure, which provided information about the overall resistance of the material to gas penetration. The updated interpretations of this phenomenon in fully saturated clays relate breakthrough pressure to the formation or development of preferential migration pathways, which may enlarge and propagate in the clay (Volkaert et al. 1995; Horseman et al. 1996; Ortiz et al. 1997; Harrington and Horseman 1997b; Horseman and Harrington 1997). The results obtained with Fo-Ca clay apparently show that a similar phenomenon occurs for a lower water saturation state.

It should be noted that, for a specific clay density, the higher the degree of saturation, the higher the critical and breakthrough pressures (Table 4). Results also showed that a ratio exists between critical and breakthrough pressures. A cross-interpretation of the data obtained for both pressures apparently shows that, when the water saturation degree nears 100%, the critical pressure (also called "gas entry pressure" in that case) ratio over breakthrough pressure nears 1. This result can be illustrated by the gas injection test performed with a fully water-saturated Fo-Ca clay sample with a 1.6 dry density. For this sample, critical gas and gas breakthrough pressures were estimated at 4.1 MPa and 4.3 MPa, respectively, and this is why critical gas pressure was difficult to detect. We also tried to correlate breakthrough pressure and the H_2 permeability. This correlation—interpolated with power law—is shown in Figure 5. Three ranges of water saturation (Sr) were selected: 70–80%, 80–90% and 90–100%. The relationship between permeability and water saturation is similar regardless of the saturation range.

Finally, it is important to note that determination of gas critical and breakthrough pressures in a clay material—saturated or not—is probably sensitive to a double scale effect. This was revealed by 2 tests with saturated compacted Fo-Ca clay samples with a dry density of 1.6 (Figure 6). During a preliminary test performed with a 1-cm-thick specimen (sample 1), a gas entry pressure of 4.1 MPa and a gas breakthrough pressure of 4.3 MPa were recorded after a 35-d test. A second experiment with a 4-cm-thick specimen (sample 2), over a period of 9 d and with a final injection pressure of 9.3 MPa, did not allow breakthrough pressure to be reached. These experiments clearly suggest a scale effect related to the size of the specimens, the duration of the experiment and especially to the duration of each pressure increment used to obtain steady flow. The determination of gas transport threshold pressures is therefore greatly influenced by this purely experimental factor.

DISCUSSION

Gas transport in highly compacted clay media have been studied by many authors (Pusch et al. 1983, 1985; Neretnieks 1985; Lineham 1989; Wikramaratha et al. 1993; Wikramaratha and Goodfield 1994; Volckaert et al. 1995; Horseman et al. 1996; Gray et al. 1996; Tanai et al. 1997; Horseman and Harrington 1997). Most of these investigations studied the impact of gas pressure on water-saturated clays. It is generally considered that the gas phase will form during the anaerobic corrosion of metals when the disposal is completely resaturated (over a few decades and several hundred years). However, given that aerobic conditions in the disposal near-field will not exceed 100 y (Gras 1996), gas generation could start earlier. Gas flows could therefore develop in the engineered clay barrier prior to its being fully saturated with water.

Gas migration in compacted clays has been studied for more than a decade. With regard to gas breakthrough phenomenon, Neretnieks (1985) studied the critical breakthrough pressure at which gas transport by advection starts in saturated bentonite. This pressure is associated with the clay swelling pressure. This breakthrough overpressure would represent between 20 and 90% of the swelling pressure. Other studies tried to show that these breakthrough pressures can provoke the formation of high-conductivity passages through the material. These passages must overcome capillary forces and can lead to the displacement of clay aggregates (Pusch and Forsberg 1983; Pusch et al. 1985). This notion of breakthrough pressure can also be found in Lineham's studies (1989) describing the transfer of N_2 in London clay. Here again, the author mentioned a critical gas pressure higher than the capillary pressure of the largest clay pores as the minimum pressure for initiating gas transport. In the framework of the European project, Modeling and Experiments on Gas Migration in Argillaceous Host Rocks (MEGAS), Volckaert et al. (1992) studied the phenomena likely to occur following gas production in an argillaceous rock (Boom clay). One of the most important findings of this work is that gas overpressures in clay-rich media may be associated with the formation of gas preferential pathways (microcracks). Wikramaratha et al. (1993) and Wikramaratha and

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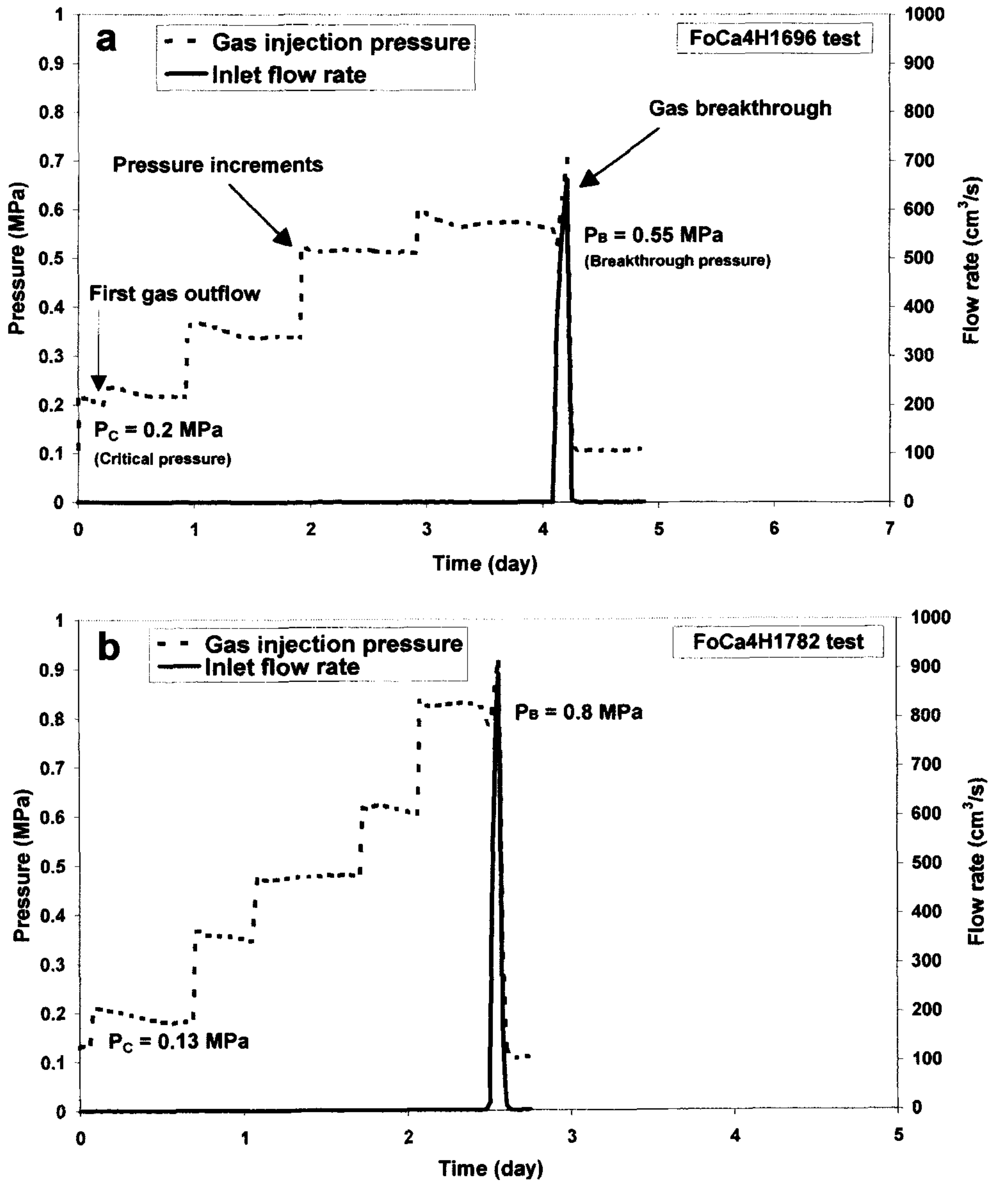

Figure 4. Typical gas breakthrough test history obtained for compacted Fo-Ca clay samples with different dry densities: 1.6 (a), 1.7 (b), 1.8 (c), 1.9 (d).

Goodfield (1994) also evoked the concept of critical pressure, a pressure below which the gas cannot migrate by advection (diffusive regime only in porewater). Opinions are apparently divided regarding the relationship of breakthrough pressure to the material swelling pressure. But there is surely a close connection between these 2 pressures which, overall, would be of the same order of magnitude (Pusch et al. 1983; Neretnieks 1985). Gas migration tests performed by PNC with the Japanese Kunigel V1 bentonite-saturated samples showed that the bentonite breakthrough pressure would be of the same order of magnitude as its swelling pressure (Tanai et al. 1997). Similar observations were made by Gray et al. (1996) with argon gas injection experiments in compacted illite clay and bentonite.

The understanding of gas migration in highly compacted clays is still being actively researched. Recent studies have provided new information (in both the experimental and modeling fields) for understanding the gas breakthrough phenomenon in fully water-saturated clay media (Volckaert et al. 1995; Ortiz et al. 1996; Ortiz et al. 1997; Harrington and Horseman 1997b; Horseman and Harrington 1997; Wood et al.

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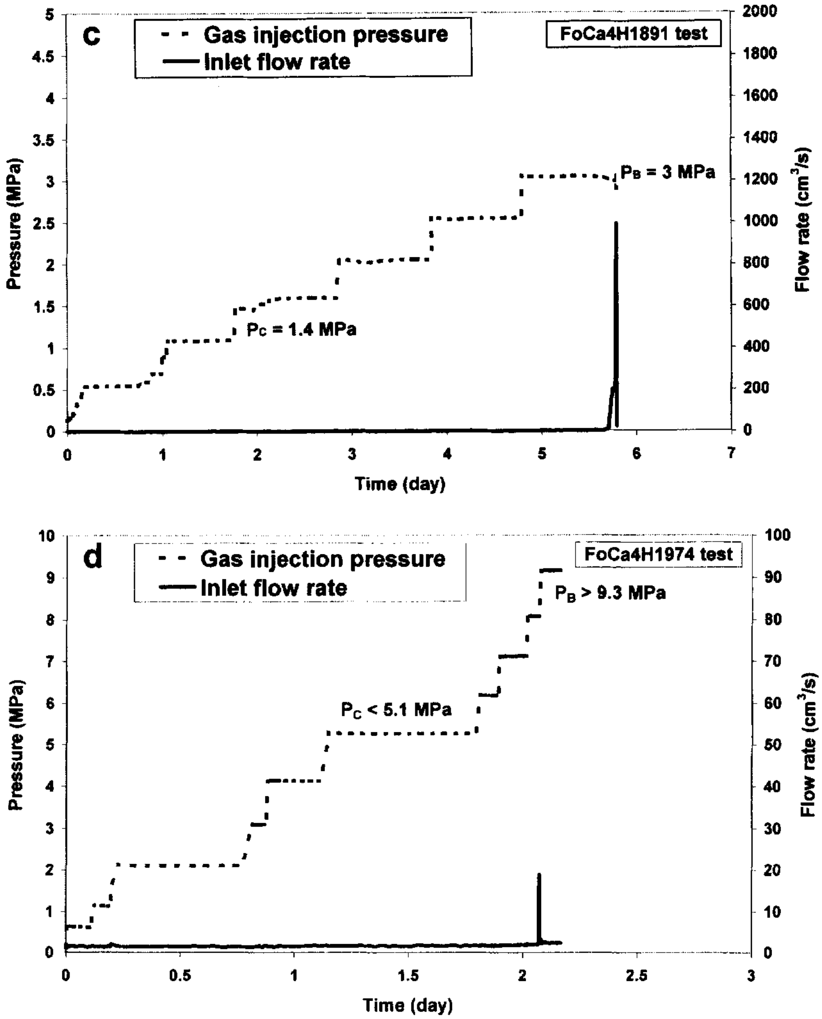

Figure 4. Continued.

Table 4. Critical pressure (P_C), breakthrough pressure (P_B) and water saturation degree (S_R) for compacted Fo-Ca clay (1.6, 1.8 dry densities).

S_R (%)	P_C (MPa)	P_B (MPa)	P_C/P_B ratio
1.6 dry density			
75	0.15	0.3	0.5
96	≈0.2	≈0.55	0.4
100	4.1	4.3	0.9
1.8 dry density			
73	0.15	1.3	0.1
83	0.6	2.1	0.3
91	1.4	2.9	0.5
98	1.6	2.1†	0.8

† Breakthrough pressure low value due to a possible lack of adhesion between clay specimen and vessel wall.

1997). These studies showed that gas breakthrough is linked to the formation and propagation of preferential pathways in the clay. Under constant stress boundary conditions, these pathways are relatively unstable, leading to intermittent gas outflow. This suggests that pathways open under high gas pressure and close when the gas injection pressure falls. It is also possible to explain intermittent flow (burst flow type) in terms of gas pathway propagation and collapse (Horseman et al. 1997). It has been found that gas breakthrough leads to a minor desaturation (Ortiz et al. 1996) of the clay material (less than 2%). Thanks to these results, modelers (Ortiz et al. 1995; Volckaert et al. 1995; Harrington and Horseman 1997b; Wood et al. 1997) could develop conceptual models for gas migration in such a material, considering a pathway-type flow concept instead of a classic 2-phase flow concept. On the other hand, a major role is apparently played by geomechanical stress on gas migration pathways (Ortiz et al.

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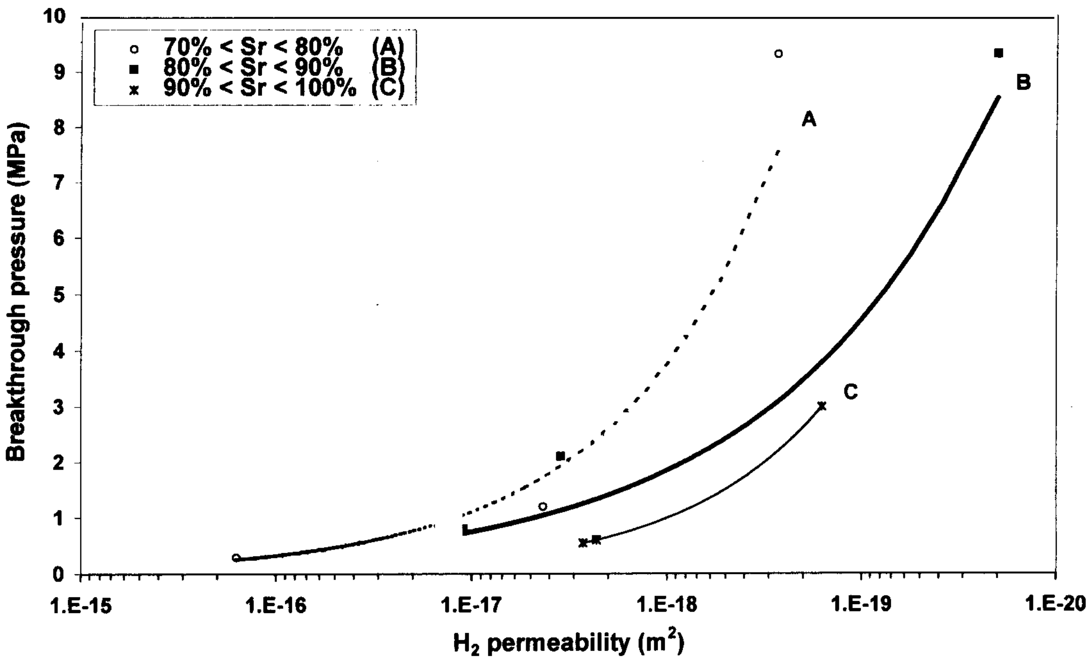

Figure 5. Correlation between breakthrough pressure and H_2 permeability for compacted Fo-Ca clay samples (4-cm-thick) for different water saturation ranges.

1996). Finally, it has been demonstrated that, under constant stress boundary conditions (confining pressure) and taking into account the application of a specific water backpressure which could simulate the underground hydrostatic pressure of the disposal site, the passage of a gas phase into an initially water-saturated buffer clay is only possible if the gas pressure exceeds the sum of the material swelling pressure and water backpressure (Horseman and Harrington 1997). The main stages of a controlled flow rate gas injection experiment until gas breakthrough could be summarized as follows: gas pressure buildup, gas entry pressure and preferential pathways propagation and gas breakthrough pressure. These results were confirmed by our test performed with a 1.6 dry density Fo-Ca clay fully water-saturated sample (1-cm-thick) with a water

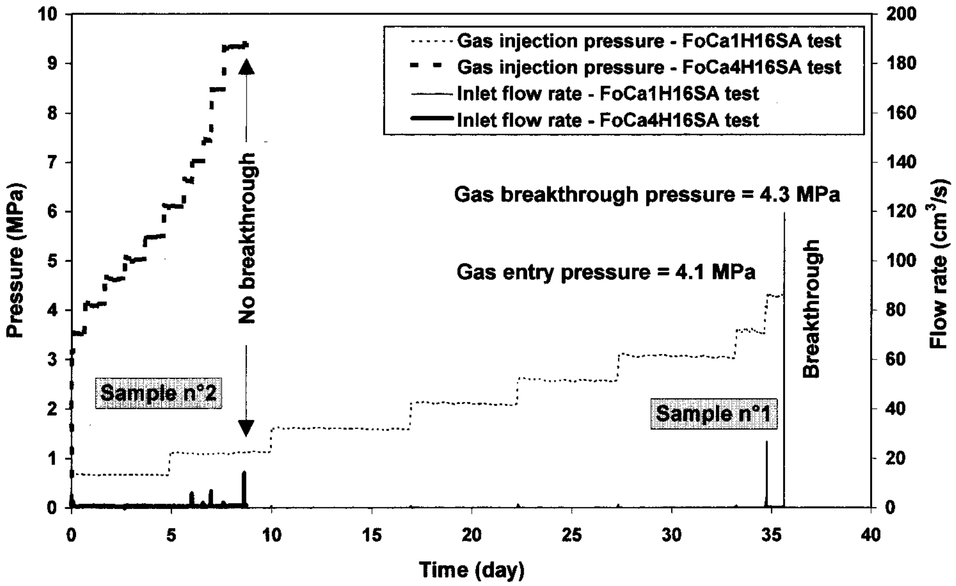

Figure 6. Scale effect on gas entry and breakthrough pressures for compacted and saturated Fo-Ca clay samples.

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backpressure equal to zero. A 4.1-MPa gas entry pressure and a 4.3-MPa breakthrough pressure were obtained for this sample. In addition, swelling pressure tests carried out on 1.6 dry density Fo-Ca clay samples produced an average swelling pressure of around 4 MPa (2.75 MPa with the 1-cm-thick and 4.95 MPa with the 4-cm-thick). This result showed that gas breakthrough pressure strongly depends on clay hydromechanical properties. Additionally, this test provided interesting information about the propagation time of gas preferential pathways. During the gas injection experiment, the elapsed time between the moment when the gas entry pressure was detected and the moment when the breakthrough phenomenon was observed was approximately 20 h. During this period, we believe that gas pathways probably enlarged and propagated into the clay. At the same time, the gas inlet flow rate increased from 3 to 120 $cm^3\ s^{-1}$.

CONCLUSION

As far as the performance and safety assessments of radioactive waste disposal in deep geological formations are concerned, much attention should be paid to the assumption that gases (mostly H_2) may form in significant quantities at the interface between the waste container (or overpack) and the engineered barrier. As a consequence, the near-field barriers and the clay buffer material in particular, could be destabilized. It is therefore essential to determine the impact of gas pressures on the buffer material. This paper is a contribution to the knowledge of gas migration mechanisms and gas breakthrough phenomena in highly compacted clays. In France, the candidate material for the buffer could be the Fo-Ca clay. This material, used to backfill the space between each container (overpack) and the host rock, should delay water contact with the container and radioactivity migration in the far-field (retention of radionuclides by sorption). Gas pressures are likely to locally and globally modify the fundamental properties of the buffer material and impact its effectiveness as a physical and chemical barrier. The potential impacts of gas pressures are illustrated by the H_2 migration experiments in Fo-Ca clay. The data on Fo-Ca clay H_2 permeability were obtained by considering 2 main parameters: the material dry density and the water saturation. For a water saturation degree of around 75%, the H_2 permeability of compacted Fo-Ca clay at densities of 1.6, 1.7, 1.8 and 1.9 are $2\ 10^{-16}$, $3\ 10^{-17}$, $4\ 10^{-18}$ and $3\ 10^{-19}$ m^2, respectively. For specimens of identical densities but with a water saturation degree of around 97%, H_2 permeability values are $4\ 10^{-18}$, 10^{-19} (estimated value), $5\ 10^{-20}$ and 10^{-21} (estimated value) m^2, respectively. It can therefore be considered that, for this range of saturation degrees, an average increase of around 20% leads to a 10^{-2} reduction of the H_2 permeability.

The second important aspect of this study concerns the migration of gas in high-density clay. The experiments performed with unsaturated highly compacted Fo-Ca clay revealed a critical pressure over which gas migration starts in the clay. In that context, gas transport is possible because the largest clay pores have very low capillary pressures that are easily exceeded by gas pressure. A second gas transport threshold pressure, called "breakthrough pressure", at which gas outflows are much larger, is then detected. The breakthrough phenomenon is probably caused by the aperture and propagation of high-conductivity discrete pathways in the clay. For a water saturation degree of around 75%, gas critical and breakthrough pressures for a 1.8 density Fo-Ca clay are around 0.15 and 1.3 MPa, respectively. For a water saturation degree around 90%, these pressures are around 1.4 and 3 MPa, respectively. The evaluation of critical and breakthrough pressures is essential in assessing the pressure fields for which gases can freely migrate into unsaturated clay without excessive constraints (no permanent damages for the clay) and the pressure fields for which the pressure buildup induces stress and leads to microcracks formation and rupture. On the other hand, this study showed that, in case of fully water-saturated clay, the gas entry pressure and breakthrough pressure are very close and that scale effect on gas breakthrough has to be carefully estimated. Finally, the results obtained on gas breakthrough phenomena are consistent with the results of recent studies showing that migration of gas flows into compacted clay is linked to the formation, aperture and propagation of gas preferential pathways. Gas breakthrough pressure is strongly related to clay material hydromechanical properties (swelling pressure) and to stress and hydraulic conditions (water backpressure). Data provided by experiments on water-saturated Fo-Ca confirmed that, globally, gas breakthrough pressure slightly exceeds the sum of swelling pressure and external water backpressure.

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