Measurement of gas entry pressure

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EURATOM

Fate of repository gases (FORGE)

The multiple barrier concept is the cornerstone of all proposed schemes for underground disposal of radioactive wastes. The concept invokes a series of barriers, both engineered and natural, between the waste and the surface. Achieving this concept is the primary objective of all disposal programmes, from site appraisal and characterisation to repository design and construction. However, the performance of the repository as a whole (waste, buffer, engineering disturbed zone, host rock), and in particular its gas transport properties, are still poorly understood. Issues still to be adequately examined that relate to understanding basic processes include: dilational versus visco-capillary flow mechanisms; long-term integrity of seals, in particular gas flow along contacts; role of the EDZ as a conduit for preferential flow; laboratory to field up-scaling. Understanding gas generation and migration is thus vital in the quantitative assessment of repositories and is the focus of the research in this integrated, multi-disciplinary project. The FORGE project is a pan-European project with links to international radioactive waste management organisations, regulators and academia, specifically designed to tackle the key research issues associated with the generation and movement of repository gasses. Of particular importance are the long-term performance of bentonite buffers, plastic clays, indurated mudrocks and crystalline formations. Further experimental data are required to reduce uncertainty relating to the quantitative treatment of gas in performance assessment. FORGE will address these issues through a series of laboratory and field-scale experiments, including the development of new methods for up-scaling allowing the optimisation of concepts through detailed scenario analysis. The FORGE partners are committed to training and CPD through a broad portfolio of training opportunities and initiatives which form a significant part of the project.

Further details on the FORGE project and its outcomes can be accessed at www.FORGEproject.org.

Intermediate Forge report D3.13

Measurement of gas entry pressure

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1. Introduction

The main goal of the present study, conducted in our laboratory within the Forge European Project, is to evaluate the hydraulic behavior of the interface between argillite and bentonite. The main assumption to validate is that, after water saturation and swelling, there could be a weaker zone as regards gas flow, which is the contact zone between the rock host (argillite) and the bentonite barrier. This means that gas, under pressure (mainly hydrogen), could flow preferentially along this interface. This issue is clearly present in the storage waste project initiated by Andra at Bure. In the laboratory, the question we propose to answer is the following: if gas is injected through a mixed plug (figure 1) constituted of a swollen bentonite (and sand) plug placed within an argillite cylinder, will this gas flow through argillite, bentonite or through the interface? A first answer will be given by gas entry pressure measurements (or gas breakthrough pressure measurements) of saturated argillite and/or bentonite alone, then of a mixed plug (figure 1). As a consequence, the first part of this study is devoted to argillite and bentonite gas entry pressure measurements. It will be followed (in the near future) with gas entry pressure tests on mixed argillite-bentonite plugs.

Figure 1: Example of an argillite-bentonite plug (after dismounting and drying)

In fact, the term “gas breakthrough pressure” appears more appropriate than “gas entry pressure”. Indeed, when gas is injected on one side of a saturated porous medium, its passage is progressive, as shown in figure 2 (from Hildenbrand et al. (2002)). When gas pressure reaches the so-called gas entry pressure, gas begins to pass through the material and pushes a small amount of water through to the downstream sample side. Experimentally, it is difficult to measure accurately gas entry pressure, mainly because its value is so low, that it may be confused with thermally-induced pressure variations (all tests are performed in an insufficiently thermally-regulated experimental room). Rather, we measure gas breakthrough pressure (GBP), defined as the injected gas pressure at which gas is expelled on the sample downstream side.

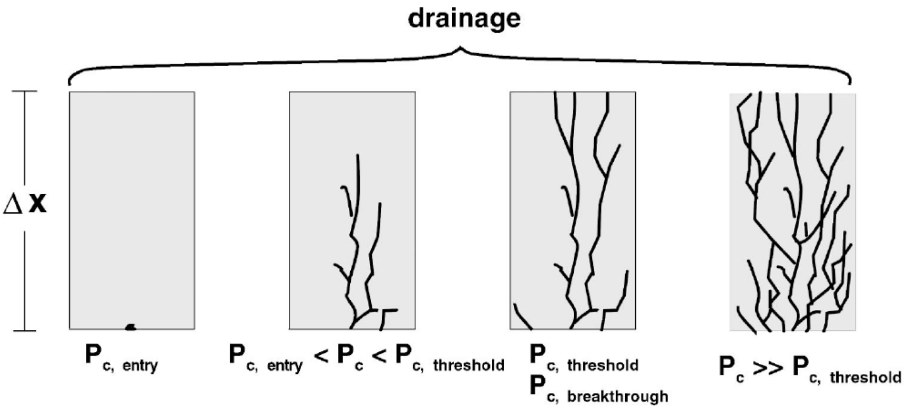

Figure 2: Stages of capillary gas passage through a fine-grained rock (from Hildenbrand et al. (2002))

2. Experimental procedures

a. Case of argillite

Sample preparation

Two series of experiments have been performed on Bure argillite samples, both cored to cylinders of diameter 37mm. The first series was on three samples cut to a height of 10 or 20mm, from core EST25600, see Table 1. The second series of tests involved five samples all of 10mm height, originating from cores EST34394, EST33271 and EST34450, see Table 2.

Experimental method

Fig. 3: Photograph of the triaxial cell and of the downstream chamber system used to record gas passage.

The detection method of GBP is as follows. First, the sample is placed between two sintered, porous, stainless steel disks, in a triaxial cell, and subjected to a hydrostatic stress (confinement) P_c=5MPa or 12MPa (similar to *in situ* stress levels). Sintered disks are placed on each side of the sample from the test start, in order to guarantee proper fluid distribution, and in order to avoid argillite leakage through the fluid pipes. Water is injected at 4MPa pressure, which is the lithostatic water pressure, first, on the upstream side, then (after ca. half an hour), on both sample sides until it is fully saturated. The full saturation state is achieved when water permeability values fall below (10^{-19}-10^{-21}m^2). Exact values are provided for the second test series, see Table 2. This saturation step is reached after several weeks, so that a limited number of samples can be tested on a limited duration.

Following this, all upstream pipes are emptied from water (volume of 5 cl). Sintered disks and downstream pipes are kept in place, without being emptied, in order to avoid sample premature failure. The sample is kept at a constant P_c=5 to 12MPa. The downstream chamber (volume of 2cl) is closed by a dedicated valve, and its pressure is recorded with a pressure transducer, see figure 3. Argon gas pressure is increased very slowly on the upstream side (at a rate of 1 to 10 days between two ΔP_{gas}=0.5 MPa steps), until gas presence is detected in the downstream chamber. For the first series of test, the gas pressure increase rate is provided in Table 1 (see column "Duration" for each upstream gas pressure value). Gas detection is performed using both the downstream pressure transducer (±100 Pa) and a dedicated argon gas detector (±0.1µl/sec). Fluid movement towards the sample downstream side is detected via downstream pressure increase, see Table 1. In such instance, the valve connected to the downstream chamber is carefully opened, so that argon is expelled and recorded by the gas detector, whenever it is actually present (see right column in Tabs. 1 and 2). It should be noted here that downstream gas pressure goes back to zero after each valve opening, between two upstream gas pressure steps. Moreover, this method of gas detection (on the sample downstream side) does not ensure whether gas actually passes through the porous medium by dissolution and diffusion in pore water, or by capillarity. These elements have to be taken into account in further modeling approaches.

b. Case of swollen saturated bentonite

Swollen bentonite is currently tested for GBP by the same method as that carefully designed for argillite, see above. It is in the water saturation phase (swelling is not stabilized yet).

3. Experimental results

a. Case of argillite

For each test, whatever the core series, as soon as gas pressure is applied on the sample upstream side, fluid pressure increases systematically on the downstream side, whatever the upstream gas pressure value (as low as 0.2MPa). This is attributed to water expelled on the downstream side, pushed by gas on the upstream side. Therefore, potentially, gas entry may have begun, yet no device is available in this experiment to check it thoroughly.

For the first test series, Table 1 shows that argon is not detected from the test start, yet at an upstream pressure ranging from 1.26MPa up to 3MPa, depending on the sample considered and on its height. As for confining pressure P_c, no significant trend is recorded: for Sample 1, GBP is of 1.65MPa at P_c=5MPa and it increases up 1.8MPa at P_c=12MPa, whereas, for Sample 2, it is of 3MPa at P_c=5MPa and it decreases down to 2.5MPa at P_c=12MPa. On the opposite, sample height h is influential: GBP ranges from 1.26 up to 1.8MPa for h=10mm, whereas it is of 2.5-3MPa for h=20mm. There is also some scatter from one sample to another, with GBP values ranging between 1.26 and 1.8MPa at h=10mm. The second test series has been performed to check these first observations, using different sample origins (different cores).

Table 1: Results for the first series of GBP experiments on COx argillite samples from core n. EST25600 (horizontal). Upstream gas pressures at gas breakthrough are indicated in bold.

Sample n.	h (mm)	P_c (MPa)	P_{gas} (MPa) Up-stream	Duration (h)	P_{gas} (MPa) Down-stream	Gas?
1	10	5	0.2	48	0.0015	No
			0.7		0.007	
			0.98		0.009	
			1.35	24	0.0065	Doubt
			1.4	48	0.0144	
			1.65	**1**	**0.01**	**Yes**
		12	1.5	48	0.005	No
			1.8	**1**	**>0.01**	**Yes**
2	20	5	1.45	72	0.0011	No
			2	72	0.002	Doubt
			2.5	96	0.003	
			3	**1**	**0.004**	**Yes**
		12	2	96	0.0042	No
			2.5		**0.0038**	Yes
			3		0.0033	
			3.5	72	0.0071	
			4		0.0145	
3	10	5	0.5	72	0.0032	No
			0.88		0.0045	

			1.26		**0.012**	**Yes**

For the second test series, as for the first series, no clear relationship is observed between GBP and confinement, see Table 2: for Sample EST34394-6, GBP=0.3MPa at P_c=6MPa, which is identical to GBP for Sample EST34450, obtained at P_c=11 to 12MPa. On the opposite, Sample EST34394-7 has a GBP ranging from 0.2MPa at P_c=6MPa to 1.3MPa at P_c=12MPa. This is very close to the variation range from one sample to another. A significant variation in GBP is therefore observed, depending on the sample considered (rather than on P_c).

Finally, while GBP values range from 1.26MPa and up to 3MPa for the first test series, it is almost an order of magnitude lower for the second test series, with values ranging from 0.2 to 1.3MPa. This is attributed to sample initial micro or macro-cracking state, which is all the more so great as argillite is stored longer before test.

Table 2: Results for the second series of GBP experiments on COx argillite samples (height h=10mm). K is water permeability before the GBP experiment.

Core and sample n.	P_c (MPa□	K (m^2)	P_{gas} (MPa) Upstream	Gas passage?
EST34394-6 (horizontal)	6	$7e^{-20}$	0.3	Yes
EST34394-7 (horizontal, sept 2009)	6	$1e^{-19}$	0.2	Yes
	12	$1e^{-19}$	1.3	Yes
EST34394-4 (horizontal)	12	$1.5e^{-20}$	0.5	Yes
EST33271 (inclined)	11	$4.2e^{-21}$	1.3	Yes
EST34450, (horizontal, sept 2009)	11-12	$9e^{-21}$	0.3	Yes

b. Case of swollen bentonite

Ongoing work- see next report

4. Partial conclusion

Although results on argillite only are available, a first important conclusion can be drawn, due to the fact that the GBP of argillite is quite low: measured values are within the range 0.3 to 2MPa depending on the *in situ* core location. For a few samples; the GBP value is higher than 2MPa but it never excesses 3MPa. Also, argillite is a transverse anisotropic material: it is a sedimentary indurated clay, composed of diagenetic bedding planes. Whenever GBP tests are performed along the bedding planes, as during both test series presented here, lower values may be expected rather than when performing GBP experiments perpendicularly to the bedding planes. This aspect will also be investigated in further work.

First results on swollen bentonite, which will be presented in our next report, display GBP values significantly higher than those measured on argillite. This means that, in a first approach, gas would rather flow through argillite than through saturated bentonite. This has to be confirmed.