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Gas breaktrough behavior of the Spanish reference bentonite

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The Spanish concept for the geological disposal of radioactive waste implies the use of pre-compacted bentonite, as a main component of the engineering barrier system. The reference bentonite, called FEBEX bentonite, is composed mainly of montmorillonite, has a maximum grain size of 5 mm and a hygroscopic water content of ~14.5%. The aim of this work was to determine the gas breakthrough (BT) pressure on saturated samples of this buffer material.

Samples (20 mm length, 38/50 mm diameter) were uniaxially compacted at 1.5 and 1.6 $g \cdot cm^{-3}$ dry densities (DD), directly in the test cell. Their water content were from hygroscopic to around 26% (full saturation at 1.6 DD), most of them on the wet side of optimum. Compaction pressures ranged between 7 and 1.4 MPa, depending on target DD, water content and granulometry of the bentonite. To check the effect of the as-compacted macrostructure, we tested three grain size distributions (full <5 mm, 1.18<x<2.0 mm, and 2.83<x<4.7 mm) with similar water content (hygroscopic); and the full grain size distribution with different water contents (around 18, 22 and 26%). Mercury intrusion porosimetry (MIP) on small compacted samples (for different DD and granulometric fractions) showed that the main differences between them were macrostructural, with minor effect on mesostructure.

A series of long-term gas injection tests were carried out to determine, by constant pressure steps, the BT pressure. Some improvements were introduced in the experimental gas breaktrough (BT) setup and the testing protocol described in [1], including different configurations of gas up and downstream volumes (~150, 75 and 50 cm^3), where actual pressure (inlet or outlet) is measured (Fig. 1a). A measurement of gas permeability (on the as-compacted sample) and two injection phases (on the fully saturated sample) configure the BT protocol. Each injection phase consists of a water permeability measurement and a long-term gas injection at increasing/decreasing pressures. The gas injection is intended to elicit at least two BT events (Fig 1b). The cells kept the samples in isochoric conditions during the test: after the saturation phase, the swelling pressure is expected to prevent preferential gas flows between the sample and the cell body. Once the gas injection starts no more water was supplied.

Gas permeability was measured on as-compacted samples with the high-pressure steady-state setup described in [2, 3]. The measurements of gas outflow and other variables, including RH/Temperature, permit to calculate the gas permeability and to estimate the sample drying induced by gas flow during the test. In these samples, there was a linear relationship of gas flow with the difference of square pressures for all dry densities tested. From these measurements, intrinsic permeability ranged from 10^{-14} m^2 to 10^{-17} m^2 (for 1.5 DD and 1.6 DD).

The water permeability measurement before the first gas injection phase allows to obtain a base-line value; the one after gas injection allows to determine if the sealing capacity was compromised by the previous BT events. Average values were in the range $(0.7 – 2.0) \cdot 10^{-18}$ m^2.

After [1], the air entry pressure for the FEBEX compacted bentonite is around 25 MPa (for DD 1.6). BT pressures determined in this work were much lower than this value and close to the upper range of the expected swelling pressure, that

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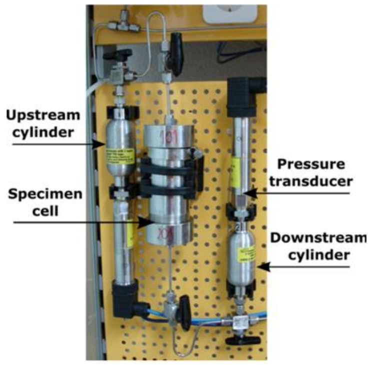
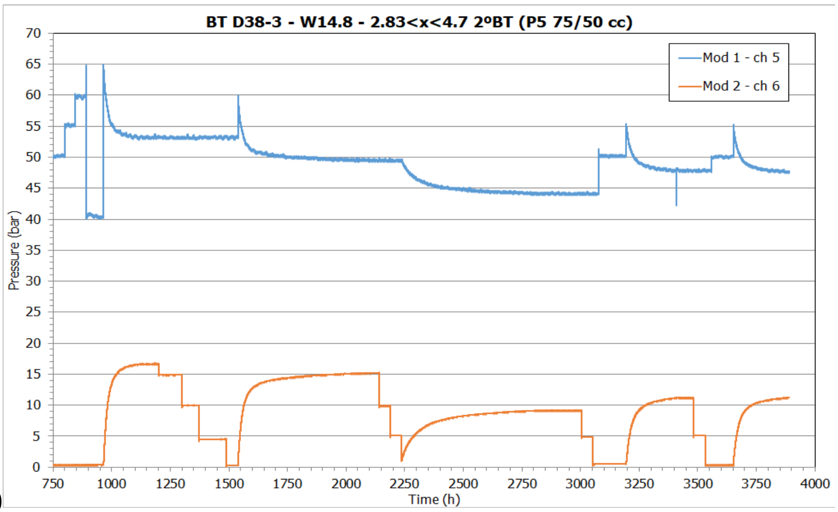
controls the gas entry and the breakthrough events. Local variations in the swelling pressure due to several factors (material heterogeneity, change in suction, temperature, stress) can direct the high-pressure gas through preferential local paths.



a)



b)

Figure 1: BT test: a) Cell set-up with pressure transducer; b) example of BT events on *P-t* curve.

The BT events on pressure-time curves showed a consistent and systematic repetition of BT values (Fig 1b); they had different shapes depending on whether the injection pressure was greater than, close to or just less than the actual BT pressure; and changes in the slope of the curves could indicate different types of flow and the underlying physical concept (from microfracturing, in the case of the instantaneous episodes, to microscopic pathway dilation for the gradual ones).

BT pressures increased with increasing dry density and water content (initial saturation degree) at compaction (considered as initial microstructural state); and the geometry of the sample (decreasing L/D ratio). The BT behavior is considered as a characteristic of a given liquid-solid system (sample): related to the initial state after compaction and the preservation of their structure during the test, at a specified temperature. Changes affecting the structure will affect the BT pressure.

Gas migration in saturated FEBEX bentonite at DD<1.6 was interpreted to occur by the formation and propagation of dilatant pathways.

Contributor statement

Guillermo García-Herrera: data acquisition and treatment; visualization. **Natalia Brea:** MIP analisys**. José-Miguel Barcala:** Test design; Electronics; Programming; Data acquisition. **Pedro-Luis Martín:** Conceptualization, Test design; Methodology, Formal analysis, Writing – Review & Editing; Project lead.

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