Engineering Geology 149–150 (2012) 57–69

Contents lists available at SciVerse ScienceDirect

Engineering Geology

journal homepage: www.elsevier.com/locate/enggeo

Long-term experimental evidences of saturation of compacted bentonite under repository conditions

M.V. Villar [a],*, P.L. Martín [a], I. Bárcena [b], J.L. García-Siñeriz [b], R. Gómez-Espina [a], A. Lloret [c]

[a] CIEMAT, Madrid, Spain
[b] AITEMIN, Madrid, Spain
[c] UPC, Barcelona, Spain

ARTICLE INFO

Article history:
Received 30 April 2012
Received in revised form 7 August 2012
Accepted 9 August 2012
Available online 29 August 2012

Keywords:
Bentonite
Hydration
Engineered barrier
Radioactive waste repository
Temperature

ABSTRACT

This paper summarises the information gathered in the last 15 years on the saturation of compacted bentonite obtained from different laboratory-scale tests, a large-scale mock-up test, and a real-scale *in situ* test, that were performed to simulate the conditions of the bentonite barrier in a high-level radioactive waste repository and to better understand the hydration/heating processes. In all the tests the bentonite used was the Spanish FEBEX bentonite, the maximum temperature in the system was 100 °C and the water used was of the granitic type, with low salinity. Some of the tests were running for more than thirteen years.

The migration of water vapour in areas affected by the high temperature induced by the radioactive waste decay is very rapid, its extent depending on the actual temperature and bentonite porosity. The water vapour condensates in cooler areas and this causes water content increases in internal zones of the barrier where the liquid water coming from the host rock has not yet arrived. The hydration kinetics is initially quicker when the temperature is high, provided no vapour phase is formed. Nevertheless, the major effect of the thermal gradient on saturation is a delaying of it in the inner parts of the barrier, which can be very persistent and depends on the actual thermal gradient and consequently, on the barrier thickness and boundary conditions. During the transient period in which the barrier is saturating, important changes in the water content and dry density of the bentonite are generated, which induce bentonite density and water content gradients along its thickness. These gradients could eventually disappear once the barrier is fully saturated, depending on the irreversibility of the deformations.

The average density of the water in the saturated barrier will be higher than 1 g/cm^3, due to the predominance of high-density, interlayer water in the compacted bentonite, and consequently, more water than expected, according to calculations made considering the density of free water, would fit in the bentonite pores.

The rate of hydration of the barrier depends on the bentonite and surrounding media hydraulic properties (that is, water availability), waste temperature and buffer thickness and geometry.

1. Introduction

A current design for engineered barriers in high-level radioactive waste (HLW) repositories includes bentonite compacted blocks initially unsaturated. The heat released by the waste will induce a thermal gradient through the bentonite barrier while groundwater will tend to flow into it. Most models predict that full saturation of the barrier will be reached before the dissipation of the thermal gradient, which will take place between 100 and 1000 years after deposition, depending on the particular characteristics of the repository. However, experimental and modelling results have shown that the barrier saturation can be an extremely slow process greatly affected by the bentonite microstructural modifications that take place upon hydration under constant volume conditions and by other processes such as thermo-osmosis and the effect of low hydraulic gradients on hydraulic conductivity (Sánchez et al., 2007, 2012). Moreover, it still remains unclear whether the high temperatures around the canister would hinder the full saturation of the inner part of the barrier or just delay it.

This paper summarises the information collected in the last 15 years on the saturation of compacted FEBEX bentonite obtained from different laboratory-scale tests, a large-scale mock-up test, and a real-scale *in situ* test, that were performed to simulate the conditions of the clay barrier in the repository under controlled boundary conditions. The results obtained help understand the hydration/heating processes and their consequences on the long-term bentonite performance, what would allow the validation and verification of the near-field thermo-hydro-mechanical (THM) models. The large-scale

---

* Corresponding author at: Avd. Complutense 40, 28040 Madrid, Spain. Tel.: +34 913466391; fax: +34 913466542.
E-mail addresses: mv.villar@ciemat.es (M.V. Villar), pedro.lmartin@ciemat.es (P.L. Martín), ignacio.barcena@aitemin.es (I. Bárcena), jlg.sineriz@aitemin.es (J.L. García-Siñeriz), roberto.gomez@ciemat.es (R. Gómez-Espina), antonio.lloret@upc.edu (A. Lloret).

0013-7952/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.enggeo.2012.08.004

tests were proposed as a complementary step in the task of demonstrating the feasibility of installing a clay engineered barrier surrounding a simulated canister in a gallery excavated in granite, keeping with the ENRESA "Deep Geological Disposal for Granite" reference concept (ENRESA, 1995). In this concept the waste canisters are emplaced horizontally in drifts, surrounded by a barrier of high density compacted bentonite blocks.

In the two large-scale tests the thermal effect of the waste is simulated by means of heaters; hydration is natural in the *in situ* test – which is being performed in a gallery excavated in granite – and controlled in the case of the mock-up. Both in the large-scale and in the laboratory tests, the temperature at the heater surface was fixed at 100 °C, which is the maximum temperature expected on the surface of the waste container in the Spanish concept. Also, in the mock-up and in most of the laboratory tests, the hydration water used was a low salinity granitic type.

The material used in all these tests is the Spanish FEBEX bentonite, selected by ENRESA as suitable material for the backfilling and sealing of HLW repositories. To obtain the swelling pressure and hydraulic conductivity necessary for a good barrier performance it must be compacted to dry densities of about 1.6 g/cm^3 (Villar et al., 2006).

2. Material

The FEBEX bentonite was extracted from the Cortijo de Archidona deposit (Almería, Spain) and the processing at the factory consisted on disaggregation and gently grinding, drying at 60 °C and sieving by 5 mm. The physico-chemical properties of the FEBEX bentonite, as well as its most relevant thermo-hydro-mechanical and geochemical characteristics obtained during the projects FEBEX I and II are summarised in the final reports of the project (ENRESA, 2000, 2006a), and a comprehensive study related to its hydro-mechanical and microstructural properties is given in Lloret et al. (2003). A summary of the results obtained is given below.

The montmorillonite content of the FEBEX bentonite is above 90 wt.% (92 ± 3%). The smectitic phases are actually made up of a smectite–illite mixed layer, with 10–15 wt.% of illite layers. Besides, the bentonite contains variable quantities of quartz (2 ± 1 wt.%), plagioclase (3 ± 1 wt.%), K-feldspar (traces), calcite (1 ± 0.5 wt.%), and cristobalite–trydimite (2 ± 1 wt.%).

The cation exchange capacity of the smectite is 102 ± 4 meq/100 g, the main exchangeable cations being calcium (35 ± 2 meq/100 g), magnesium (31 ± 3 meq/100 g) and sodium (27 ± 1 meq/100 g). The predominant soluble ions are chloride, sulphate, bicarbonate and sodium.

The liquid limit of the bentonite is 102 ± 4%, the plastic limit 53 ± 3%, the density of the solid particles 2.70 ± 0.04 g/cm^3, and 67 ± 3% of particles are smaller than 2 μm. The hygroscopic water content in equilibrium with the laboratory atmosphere (relative humidity 50 ± 10%, temperature 21 ± 3 °C, total suction about 100 MPa) is 13.7 ± 1.3%. The external specific surface area is 32 ± 3 m^2/g and the total specific surface area is about 725 m^2/g.

The saturated hydraulic conductivity of compacted bentonite samples is exponentially related to their dry density. For a dry density of 1.6 g/cm^3 the saturated permeability of the bentonite is about $5\cdot10^{-14}$ m/s at room temperature, either with granitic or deionised water used as percolating fluid. The temperature increase results in an increase in permeability.

The swelling pressure of compacted samples is also exponentially related to the bentonite dry density, and when the bentonite at dry density of 1.6 g/cm^3 is saturated with deionised water at room temperature, the swelling pressure has a value of about 6 MPa. Saturation with granitic water gives similar values, whereas temperature causes a decrease of them.

The retention curve of the bentonite was determined in samples compacted to different dry densities at different temperatures (Lloret et al., 2004; Villar and Lloret, 2004; Villar and Gómez-Espina, 2009). The volume of the samples remained constant during the determinations, since they were confined in constant volume cells. Following an approach similar to that presented by Sánchez (2004) to fit the data from these laboratory determinations, the empirical Eq. (1) can be obtained:

$$w=\left(\left(b\cdot n^{c}\cdot e^{-\alpha(T-T_0)}\right)\cdot\left[1+\left(\frac{s}{P_0\cdot e^{-\eta(n-n_0)}\cdot e^{-\alpha(T-T_0)}}\right)^{\frac{1}{1-\lambda_1}}\right]^{-\lambda_1}\right)\cdot\left(1-\left(\frac{s}{P_{\mathrm{sec}}}\right)^{\lambda_2}\right)\cdot(S_r-S_{lr}) \tag{1}$$

where w is the water content in percentage, n and n_0 the porosity and reference porosity, s the suction in MPa, T and T_0 the temperature and reference temperature in °C, S_r and S_{lr} the liquid degree of saturation and liquid residual degree of saturation, P_0, P_{sec}, λ_1 and λ_2 parameters to define the retention curve at reference temperature and porosity, and b, c, α and η fitting parameters to take into account the influence of temperature and porosity. The values of parameters are indicated in Table 1. The differences between measured values and the estimated values using Eq. (1) are smaller than 2% in terms of water content.

The thermal conductivity (λ, W/m·K) of the compacted bentonite at laboratory temperature is related to the degree of saturation (S_r) through the following expression:

$$\lambda=\frac{0.57-1.28}{1+\exp^{(S_r-0.65)}/_{0.100}}+1.28 \tag{2}$$

Some isothermal infiltration tests and heat flow tests at constant overall water content were performed during the FEBEX I project (ENRESA, 2000, 2006a) and they were back analysed using CODEBRIGHT. The experimental data were fitted using a cubic law for the relative permeability and a value of 0.8 for the tortuosity factor.

In all the tests described below, the bentonite was used compacted with its hygroscopic water content (14%) at dry densities between 1.6 and 1.7 g/cm^3, which is the range expected in the repository.

3. Description of tests

3.1. In situ test

The FEBEX *in situ* test is performed under natural conditions and at full scale in a gallery excavated in the underground laboratory managed by NAGRA at Grimsel, Switzerland (ENRESA, 2000, 2006a). The basic components of the test (Figure 1) were: the gallery, measuring 70 m in length and 2.3 m in diameter, excavated through the Aare granite; the heating system, made up of two heaters placed inside a liner installed concentrically with the gallery and separated one from the other by a distance of 1.0 m, with dimensions and weights analogous to those of the real canisters; the clay barrier, formed by blocks of compacted bentonite; the instrumentation and the monitoring and control system for data acquisition and supervision and control of the test both autonomously and remotely from Madrid. Up to 632 sensors of very diverse types were initially installed to monitor the different thermo-hydro-mechanical processes that occurred in both the clay barrier and the surrounding rock throughout the entire life of the test. The gallery was closed by a concrete plug.

Table 1
Values of parameters in Eq. (1).

b	c	P_0 (MPa)	λ_1	λ_2	η	n_0	α (1/°C)	T_0 (°C)	P_{sec} (MPa)	S_r	S_{lr}
145	1.9	25	0.2	1.1	20	0.4	0.0015	20	1000	1.0	0.01