A stochastic approach to the modelling of gas transport in bentonite

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ABSTRACT: In the past years, transport of gases in clay buffer materials for Engineered Barrier Systems (EBS) has been deeply investigated by many research programs in the field of nuclear waste disposal. The phenomenological features of gas transport obtained from experimental tests have been used to define modelling approaches to interpret gas-induced effects in bentonite buffer material. The aim of these models is to analyze the performance of clay buffer materials in a long term nuclear waste disposal scenario. The gas transport phenomenon is strongly influenced by the microstructural features of the bentonite. In this paper, an advanced finite element approach is used to simulate the gas migration phenomena in the framework of stochastic finite element analysis. This tool highlights the most relevant factors affecting the accuracy of the modelling process to improve model efficiency and to estimate the simulation uncertainties. The approach highlights the need of an accurate microstructural characterization of bentonite to define the water retention behavior and the hydraulic response of the buffer material.

1 INTRODUCTION

The problem of gas transport in clay buffer materials for Engineered Barrier Systems (EBS) has been deeply investigated in the past years in the framework of the storage of High Level Nuclear Wastes. A multitude of experimental programs devoted to the definition of the basic phenomena involved in the gas-migration have been conducted and many modelling approaches to interpret the laboratory results on bentonite buffer material have been developed. The final objective of these modelling activities is to analyze the performance of clay buffer materials in a long term nuclear waste disposal scenario. At the present state, literature models provide qualitative description of the gas transport phenomena but their predictive capabilities are still limited, indicating that the physical phenomena that rule the gas transport process are still far to be fully understood.

This work is based on a series of gas injection tests performed on saturated granular bentonite by Romero and Gonzalez-Blanco (2017) in the framework of the Engineered Barrier System Task Force. This experimental work includes a series of microstructural investigations that can be used to define and calibrate advanced modelling approaches.

The proposed modelling approach is based on the Finite Element Code Lagamine (Charlier et al., 2001). The code can address multi-physical problems (THMC) adopting a mixtures theory approach.

The modelling is based on a detailed analysis of the microstructure of the analyzed granular bentonite to define the water retention behavior and to calibrate the hydraulic and constitutive models. The deterministic approach based on the best estimate of the material parameters provides good simulations of the gas injection tests.

A series of analysis have been performed with the aim of identifying the uncertainties of the modelling process. The stochastic sensitivity analysis tool has been employed to identify the impact of each modelling assumption on the accuracy of model outputs.

2 MATERIALS AND EXPERIMENTAL DATA

2.1 *Material and sample preparation*

This work is based on the experimental work performed by Romero and Gonzalez-Blanco (2017) on National Standard WP2 Na-bentonite from Wyoming (USA). The material properties of WP2 bentonite are assumed to be similar to Mx-80 Na-bentonite due to the equivalent montmorillonite content. The experimental characterization performed by Romero and Gonzalez-Blanco (2017) has been integrated with MX-80 bentonite data from the relevant literature published elsewhere (Seiphoori et al., 2014a; Seiphoori, 2014b; Villar, 2004; Delage et al., 2006).

The sample preparation process has been described in detail by Romero and Gonzalez-Blanco (2017). The bentonite has been wetted from the hygroscopic water content (7.2%) to a target water content of 20%. The samples have been then statically compacted in oedometric conditions to a target dry density of 1.55 Mg/m^3. In a subsequent phase, the samples have been saturated with deionized water in the oedometric cell at constant vertical stress (the process lasted from 3 to 4 weeks to ensure the full saturation). During the samples preparation process the hydromechanical response has been carefully monitored.

The microstructural features of the obtained samples have been investigated with Mercury Intrusion Porosimetry to compare the pore size distributions of saturated state and as-compacted state.

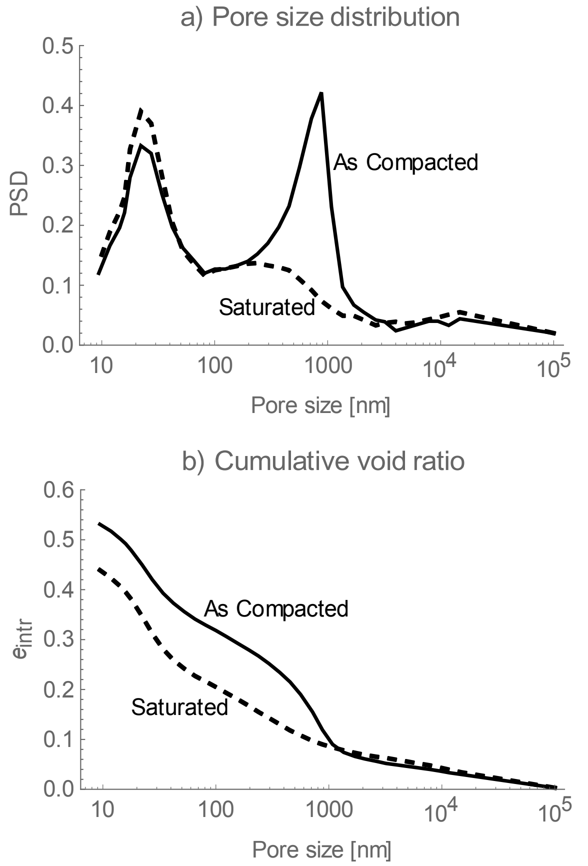

Figure 1. Mercury Intrusion Porosimetry results of Na-bentonites: a) Pore Size Distribution, b) Cumulative void ratio (Romero and Gonzalez-Blanco, 2017).

The results of the MIP tests performed on WP2 bentonite are shown in Figure 1 (Romero and Gonzalez-Blanco, 2017). At the as-compacted state the material exhibits a clear bimodal pore size distribution as observed by Seiphoori et al. (2014a) on MX-80 bentonite. During the saturation process the microstructure of bentonite evolves with a slight increase of the micropores amount and a strong decrease of the higher dimension pores. This behaviour is related to the hydration of the bentonite assemblages that tends to reduce the volume of the macropores. The amount of macroporosity at the saturated state at this relative low value of dry density is still relevant compared to the values measured at a dry density of 1.80 Mg/m^3 (Seiphoori et al., 2014a).

This aspect is of particular interest in the modelling of the gas injection tests because it can significantly affect the water retention behaviour in the high degree of saturation domain.

2.2 Test setup

The gas injection tests have been performed with a high pressure oedometric cell. The vertical stress has been applied with a hydraulic piston and the vertical displacements have been measured with an external LVDT sensor. Three pressure and volume controllers have been used to apply water and air pressures at the boundaries of the sample. In particular, the water pressure has been controlled at the two sides and air pressure has been controlled at the bottom side.

The test protocol involves four phases: a) equalization of the initial saturated state ($\sigma_v \approx$ 5 MPa, $p_w =$ 0.5 MPa); b) replacement of the water in the lower side porous stone with air; c) air injection phase at constant air flow up to the maximum air injection phase (2 to 4.5 MPa); and d) gas dissipation phase performed at constant volume of the injection circuit.

Four tests have been performed with different injection pressures and air injection rates. The laboratory procedures and results are explained in detail by Romero and Gonzalez-Blanco (2017).

3 DEFINITION OF THE MODELLING APPROACH

The modelling approach is based on the finite element code Lagamine (Charlier et al., 2001) that adopts a fully coupled thermo-hydro-mechanical formulation.

The four gas injection tests have been simulated starting from the saturated state neglecting the sample preparation phases.

To model the air dissipation phase it is necessary to properly consider the dead volumes of the gas injection and recovery circuits. One of the possible strategies to take into account the presence of the dead volumes is to introduce two dummy porous reservoirs with the same void volumes of the oedometric apparatus circuits. The hydro-mechanical properties of the dummy reservoirs have been chosen to avoid any modification of specimen hydromechanical response (i.e. extremely low air entry value, high permeability, and rigid mechanical response).

The boundary conditions applied to the model have been defined to mimic the laboratory conditions.

The simulation is based on four phases:

a) initial condition: the bentonite specimen, the upstream and downstream reservoirs are saturated;
b) bottom reservoir desaturation: the water pressure at the bottom side has been slightly reduced to de-

saturate the bottom reservoir. This process doesn't influence the specimen due to the high contrast in the air entry value between the porous reservoir and the bentonite;

c) air injection: the bottom pressure has been increased to reach the maximum injection pressure;

d) air dissipation: the lower boundary becomes impervious (to air and gas) to simulate the closing of the upstream valve.

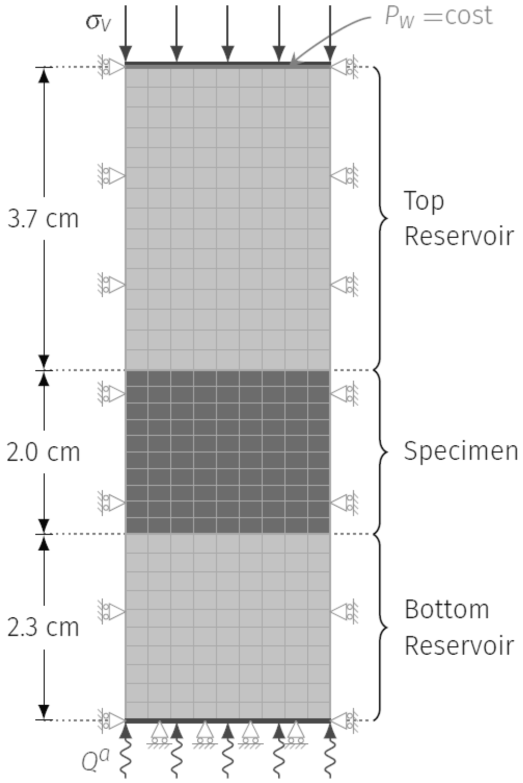

Figure 2. Sketch of the finite element mesh: test idealization, loading and boundary conditions.

A scheme of the finite element model is shown in Figure 2 to highlight the components of the model and the applied boundary conditions. The finite element mesh is based on 8-nodes quadrilateral elements with four integration points.

4 MODEL CALIBRATION

The model calibration has been conducted starting from the material characterization performed on the tested material (Romero and Gonzalez-Blanco, 2017) with some complementary data from other experimental works conducted on MX-80 bentonite.

The response of bentonite to gas injection is strongly influenced by the water retention behavior and the accuracy of model prediction is strongly related to the adopted water retention model.

In this work, an advanced water retention model developed by Diedudonné et al. (2017) for compacted bentonite has been adopted. It is based on the distinction of two retention mechanisms in the compacted bentonite: adsorption in the intra-aggregate pores (macropores), and capillarity in the inter-aggregate pores (micropores). The two mechanisms are combined to take into account the measured distribution of micro and macro porosities. In addition, the evolution of the structure is considered introducing a dependence of the inter-aggregate porosity on the degree of saturation of the material.

The model is formulated in terms of water ratio (e_w: volume of water over solid volume) that is expressed as:

$$e_w = e_{wm} + e_{wM} \tag{1}$$

where e_{wm} accounts for the water stored in the micropores and e_{wM} accounts for the water stored in the macropores. The water retention curve is defined as follows:

$$e_w = e_m \exp\left[-\left(C_{ads} s\right)^{n_{ads}}\right] + \left(e - e_m\right)\left\{1 + \left[\left(e - e_m\right)\frac{s}{A}\right]^n\right\}^{-m} \tag{2}$$

where e_m is the microstructural void ratio, and s is the suction. In addition, the evolution of the microstructural void ratio is directly considered as follows:

$$e_m = e_{m0} + \beta_0 e_w \, . \tag{3}$$

The definition of the water retention model is based on 7 parameters (C_{ads}, n_{ads}, A, n, m, e_{m0}, β_0).

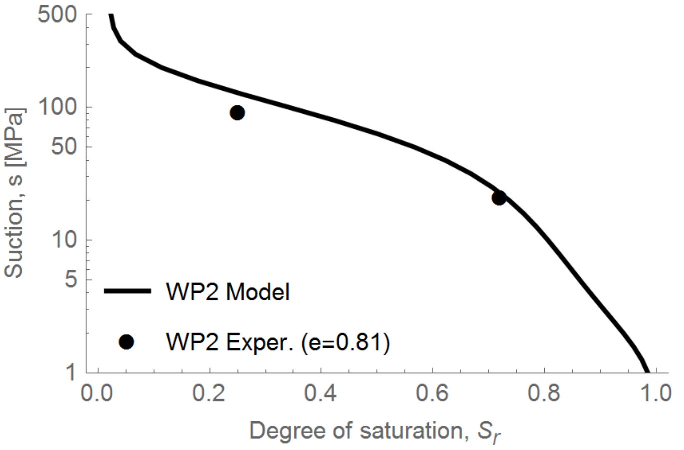

Figure 3. Comparison between the analytical and the measured water retention curve for WP2 bentonite at ρd=1.55 Mg/m3 (e=0.80).

In this work, the model has been calibrated on the basis of a large data base available on MX-80 bentonite (Diedudonné et al., 2017) introducing the results of the MIP investigation of Figure 1. This assumption includes the effects of compaction and saturation processes to the material configuration represented by the ratio between the macrostructural and the microstructural void ratio. From Figure 1 it is possible to define the value of e_{wM} of the studied material. Assuming a threshold value of 200 nm to separate micro-pores and macro-pores, the macrostructural void ratio results 0.17.

Figure 3 shows the obtained water retention curve compared with the experimental points by Romero and Gonzalez-Blanco (2017). The presence of a significant amount of macrostructural pores strongly affects the water retention response for high values of the degrees of saturation.

At this stage the mechanical behaviour of the saturated bentonite has been simulated with a simple linear elastic model. This assumption is admissible since the simulation starts from the saturated state. It allows to analyse the model response to have a qualitative description of the hydromechanical coupling.

Table 1. Summary of sensitivity analysis results.

	Parameter	*Best estimate*
Water retention model Dieudonné et al. (2017)	C_{ads}	0.0075
	n_{ads}	1.5
	A	0.2
	m	3.0
	n	0.15
	e_{m0}	0.37
	β_0	0.34
Intrinsic permeability	k_{intr} [m^2]	1×10^{-20}
Dissolved air diffusion c.	D_l^a [m/s^2]	5×10^{-11}
Henry coefficient	H	0.0234
Porosity	n	0.44
Mechanical elastic model	E [MPa]	2.98
	υ	0.3

The calibration of the hydraulic, diffusive and mechanical models has been conducted on the basis of the results by Romero and Gonzalez-Blanco (2017) (i.e. constant head permeability tests, low pressure gas injection tests, elastic loading). The best estimate of the model calibration is shown in Table 1.

5 ANALYSIS RESULTS

The four gas injection tests at different injection pressures have been simulated with a deterministic approach adopting the best estimate parameter calibration of Table 1.

Figure 4 shows the results of the four analysis in terms of gas pressure at the injection boundary and outflow volume. Some interesting features can be highlighted from the analysis of the results.

The gas migration when a low gas pressure is applied is dominated by a diffusive gas flow (Sample 4 and first injection of Sample 6). The outflow volume during the dissipation stage is very low and the bottom gas pressure remains practically constant. This phenomenon is well captured by the model.

Increasing the injection pressure the advective gas flow becomes more important with the increase of the outflow volumes and the drop of the gas pressure at the injection point. The simulation well predicts the outflow volumes whereas underestimates the pressure drops. Note that the laboratory measured outflow of Sample 6 is not consistent with the other samples at similar injection pressures. This effect is probably related to problems in this experimental test.

Further analysis will be conducted to focus the effect of hydromechanical coupling effects that are neglected with the presented linear elastic mechanical model.

6 STOCHASTIC SENSITIVITY ANALISYS

The use of deterministic approach in numerical analysis leads to a lack of information concerning effects of the model calibration on the analysis outputs. One of the possible approaches to evaluate the model reliability and to identify the most critical model inputs affecting the results is the sensitivity analysis tool.

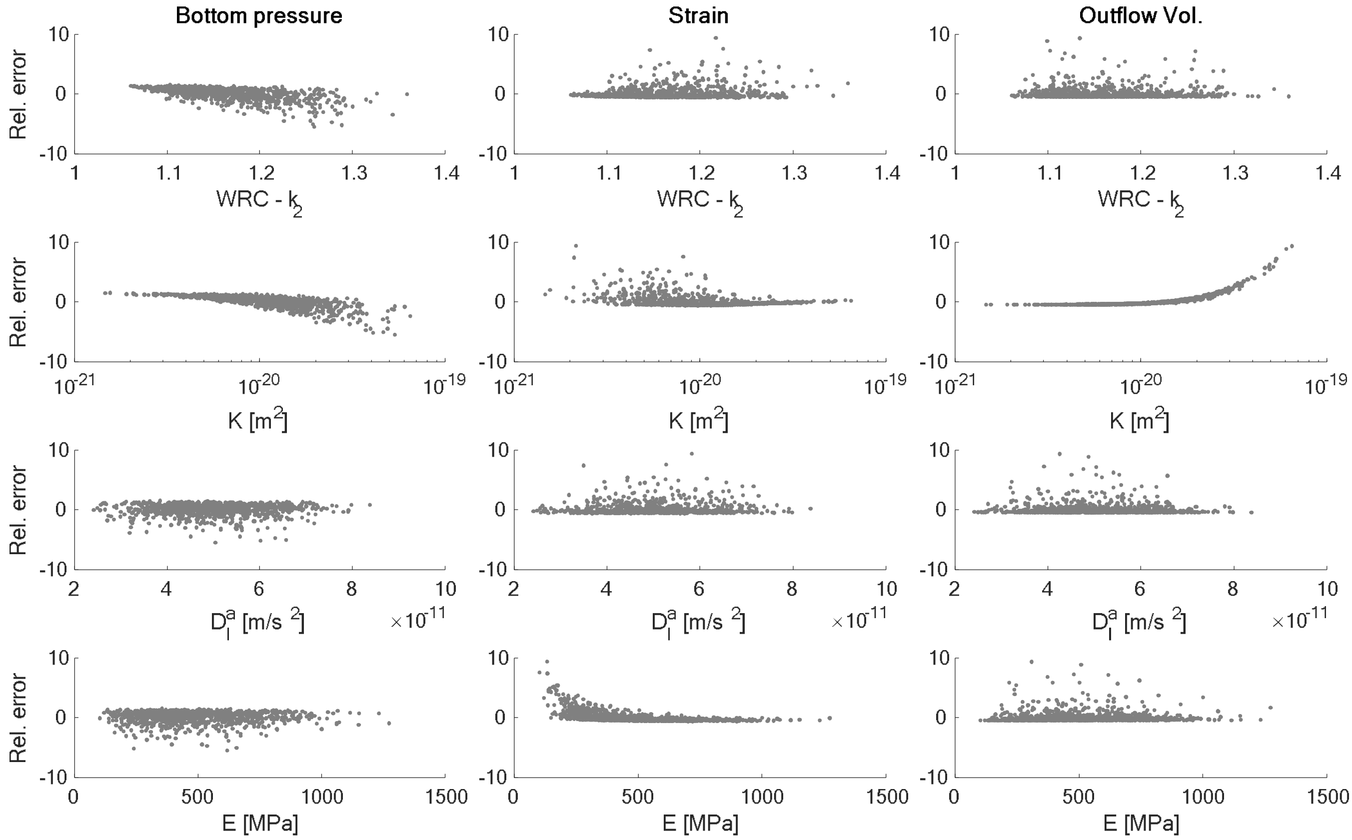

Figure 5. Results of the sensitivity analysis of gas injection tests on saturated bentonite (Sample S7).

In this work we performed a stochastic sensitivity analysis following the approach proposed by Saltelli et al. (2008). The Monte Carlo method with linear regression has been adopted. It consists in the definition of a set of input parameters and of a set of output target quantities. The first step of the sensitivity analysis is the definition of a statistical set of the input parameters sampled from suitable statistical distributions. The model outputs, computed from each input set, are elaborated to obtain a measure of the errors of the simulation with respect to the measured values. The correlation between the distribution of each output error and each input parameter gives the measure of the sensitivity of the numerical model.

The sensitivity of the model outputs to the input parameters can be efficiently analyzed drawing the scatter plots of the errors with respect to each model parameter. The more grouped are the scatter points, the highest is the sensitivity.

The visual analysis of output sensitivity can be refined by performing a multilinear regression on the output data to define the correlation coefficients β_{Zj}^2 between the output and the relative input.

The stochastic sensitivity analysis tool is a powerful way to analyze the impact of the calibration of the numerical model and represents the first phase of an efficient uncertainties estimation process.

In this study, the tool has been applied to the analysis to investigate the effects of four input parameters on the three meaningful measured quantities. The analyzed input parameters are: the air entry value of the water retention curve, the water intrinsic permeability, the air diffusion coefficient, and the Young's modulus of bentonite. The analyzed output quantities are: the gas pressure at the injection point, the axial strain, and the outflow volume at the recovery point.

The errors of the simulation have been defined as the ratio between the integrals of the simulated and the measured time histories of each output variable.

The statistical set of input variables has been defined starting from the expected variability of each parameter. It consists of one thousands of input sets sampled from normal or beta distributions (depending on the presence of physical limits of the input parameter).

The results of the sensitivity analysis in terms of scatter plots for Sample S7 are shown in Figure 5. It is possible to highlight the strong sensitivity of the three outputs on the intrinsic permeability (denoted by the grouping of the scatter points). In addition, strong sensitivity can be highlighted also for strain on Young's modulus and for bottom gas pressure on the water retention curve.

A more refined way to analyze output sensitivity is the computation of the correlation coefficients. The results of the multilinear correlation procedure of the four tests are shown in Table 2. These scalar coefficients directly express the degree of sensitivity of each output on the relative input parameter.

From the analysis of the correlation coefficients, it is possible to highlight some interesting features of the proposed approach:

a. The permeability strongly impacts on the three output quantities for all the four tests.
b. The gas pressure at the injection point is mainly related to the water retention behavior.
c. The vertical strain is mainly related to the Young modulus.
d. The diffusion coefficient has an impact on the bottom gas pressure only for Sample 4 (the only specimen subjected to low injection pressure for the entire test).

Table 2. Summary of sensitivity analysis results.

SAMPLE 4			
Parameter	P_g^b	ε_{ax}	V_{out}
WRC	0.23	0.02	0.00
K	0.54	0.28	0.79
D_l^a	0.07	0.00	0.00
E	0.00	0.40	0.00

SAMPLE 5			
Parameter	P_g^b	ε_{ax}	V_{out}
WRC	0.30	0.09	0.00
K	0.54	0.13	0.82
D_l^a	0.00	0.00	0.00
E	0.00	0.45	0.00

SAMPLE 6			
Parameter	P_g^b	ε_{ax}	V_{out}
WRC	0.06	0.02	0.00
K	0.15	0.01	0.91
D_l^a	0.00	0.00	0.00
E	0.00	0.13	0.02

SAMPLE 7			
Parameter	P_g^b	ε_{ax}	V_{out}
WRC	0.33	0.04	0.00
K	0.53	0.05	0.87
D_l^a	0.00	0.00	0.00
E	0.00	0.26	0.00

The sensitivity analysis results will be used to improve the model performances in terms of accuracy of the calibration. In addition, it can be used to orient the laboratory characterization towards the more sensitive aspects of the analyzed phenomena.

7 CONCLUSIONS

The integrity of Engineered Barrier Systems for nuclear waste storage can be deeply affected by the migration of the gas produced by the corrosion of the canister. In the past years, many efforts have been devoted to the characterization and simulation of the gas migration phenomena in the EBS.

In the present work, a series of gas injection tests performed on saturated bentonite have been simulated to assess the reliability of the simulation process and the uncertainties associated to the model predictions.

The fully coupled finite element environment Lagamine has been used as modelling framework and an advanced water retention model for bentonite has been adopted. The deterministic analysis results show a general agreement with the experimental measurements. The subsequent phase of the modelling work will focus the hydromechanical couplings by adopting more advanced mechanical models.

The results of the modelling approach have been analyzed with the sensitivity analysis tool to highlight the impact of model input assumptions on the outputs obtained from the model. This process highlighted the extreme sensitivity of the model results on the intrinsic permeability of the bentonite and other interesting features of the approach that can be employed to improve the modelling framework and to define the uncertainties of the modelling process.

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