Experimental and numerical investigation on water exchange of Opalinus Clay samples

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Abstract. Opalinus Clay is a fine-grained sedimentary geomaterial with a mineral composition consisting mainly of silicates, carbonates and quartz. Due to its favourable barrier properties, this geomaterial has been selected as the host rock for the underground disposal of radioactive waste in Switzerland. In this context, the hydro-mechanical state of Opalinus Clay will be affected by subsequent drying and wetting phenomena. Design procedures require then an appropriate understanding of the processes related to water exchange.

To analyse the behaviour of Opalinus Clay upon suction changes and to define its water retention properties, the vapour equilibrium technique is combined with an accurate assessment of the deformations in the two orthogonal directions using strain gauges. Based on those results, a 3D Thermo-Hydro-Mechanical Finite Element model is implemented and validated. Preliminary analysis shows overall a good agreement between experimental data and modelling results in terms of change in water content, degree of saturation and equilibrium time. The numerical model will be used to define an accurate law for the tortuosity and permeability evolution of Opalinus Clay when subjected to suction variations, that is required to simulate the behaviour of a nuclear waste disposal system.

Keywords Opalinus Clay, hydro-mechanical behaviour, vapour equilibrium technique, FE-model

1 Introduction

The current or past use of nuclear energy still poses serious environmental concerns related to the disposal of the generated nuclear waste. From a geomechanical perspective, the design of a geological disposal system requires a deep understanding of the thermo-hydro-mechanical behaviour of the involved materials (i.e. the host rock and the engineered barriers). Opalinus Clay has been selected to serve as the host formation for the Swiss underground disposal of radioactive waste thanks to its favourable properties (e.g., low permeability and porosity, high retention and self-sealing properties). Over the lifespan of the repository, the hydro-mechanical state of Opalinus Clay will be affected by the tunnel excavation, ventilation and re-saturation phases, leading to subsequent drying and wetting phenomena [1]. These processes are related to the heat and water fluxes that take place between the geomaterial and the surrounding environment. Such evaluation requires taking into account the couplings between the balance of water mass and energy, together with the relevant boundary conditions. By numerical modelling, the role, the magnitude and the effect of each transport mechanism can be identified. Interestingly, the quantitative evaluation of these fluxes is required in many other relevant applications for civil and environmental engineering, like climate change effects induced by global warming, natural or artificial slope stability and transport of contaminants in the groundwater.

In this context, experimental results on Opalinus Clay have shown high water retention properties and the dependency of the air entry value on the void ratio [2], and have demonstrated an irreversible volumetric response to suction variations [3, 4]. Consistent observation from numerical investigation of the water exchange on Opalinus Clay due to suction changes, suggests that the development of micro-cracks may enhance the flow rate of water vapour [5].

In this paper, a 3D numerical model capable of reproducing the vapour equilibrium technique (VET) is implemented in the commercial Finite Element code Comsol Multiphysics® and validated on tests carried out on four specimens of Opalinus Clay [4]. This numerical model will be used to study in depth the role of advective and diffusive fluxes during the drying and wetting processes to which the Opalinus Clay will be subjected during its lifespan, as well as the effect that the environmental conditions (temperature T, relative humidity h_r, total suction ψ) and the material properties (porosity ϕ, mineralogical composition and fabric) have on the hydro-mechanical behaviour of this material.

2 Material and methods

Opalinus Clay is a fine-grained sedimentary geomaterial with a mineral composition consisting mainly of silicates, carbonates and quartz. In 2015, a borehole was drilled close to the village of Lausen (north-western Switzerland), where Opalinus Clay was encountered at a shallow depth from 6 to 71 m [6]. The samples considered in this study (namely L3, L8, L11 and L13) were retrieved from this borehole and the initial geotechnical characteristics of the tested specimens are discussed in [4].

To analyse the behaviour of Opalinus Clay upon suction variations and define its water retention behaviour, the VET is combined with an accurate assessment of deformations in two orthogonal directions using strain gauges [4] as shown in Fig. 1. The VET allows the application of a known relative humidity inside a sealed container with saturated saline solutions. Through the so-called ‘psychrometric law’, relative humidity can be then converted to total suction [7]. Based on the type of salt, different total suction values can be imposed and allow to perform wetting/drying cycles. All tests were conducted at a controlled temperature of 25°C (T_{env}) and using cylindrical specimens with an initial diameter and height of 25 mm and 20 mm, respectively, with bedding perpendicular to the axis of the cylinder [4]. For each core, two specimens were used for the water retention measurements: one specimen equipped with strain gauges to evaluate volume strains; the second specimen used to monitor the mass variation to assess the water content changes. At each suction step, the achievement of the equilibrium condition is assessed by the stabilization of both strain and mass of the specimen.

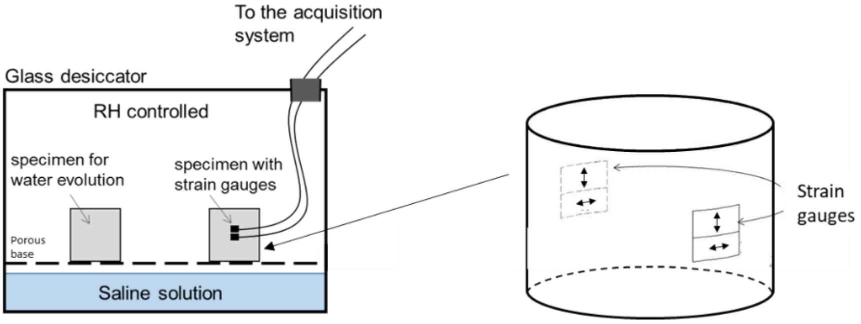

Fig. 1. Experimental setup: sealed container for total suction application (left); strain gauge configuration on the tested specimen (right) (modified after [7])

3 THM model for vapour equilibrium technique

In order to study the water exchange related to the various water transport mechanisms in the geomaterial, a Thermo-Hydro-Mechanical (THM) model is implemented. It integrates the coupled water mass and energy balance equations. Volume changes upon drying and wetting are evaluated by introducing a suitable dependency of porosity on suction [8]. The mass balance of air is neglected under the common assumption of hydrostatic conditions for the air phase [9]. As a consequence, the air pressure is equal to the atmospheric one and is constant in time and space. The diffusive flux of air dissolved in liquid water is also considered as negligible. The evaporative flow from/to the porous medium is modelled as water and heat fluxes at the soil-atmosphere interface. In the following sections, the nomenclatures ‘water’ and ‘air’ indicate the chemical species, while ‘gas’ or ‘liquid’ indicate the phases. The water in the gas phase is indicated as ‘vapour’ and the flow of dry air in the soil is neglected.

Water mass balance equation. The mass balance equation can be expressed in terms of chemical species and, under the above hypotheses, it assumes the form:

$$\frac{\partial}{\partial t}\left[\phi\left(S_l\rho_l + S_g c_g^w\right)\right] + \nabla\cdot\left[-\frac{\mathbf{K}_{sat}k_r}{g}\nabla\left(u_l + \rho_l g z\right)\right] + \nabla\cdot\left(-\phi S_g D_v \boldsymbol{\tau}\nabla c_g^w\right) = 0 \qquad (1)$$

where ϕ is the porosity; S_l and S_g are the liquid and gas degree of saturation ($S_l = 1 - S_g$); ρ_l is the mass density of the liquid phase; c_g^w is the vapour mass concentration; $\mathbf{K}_{\mathbf{sat}}$ is the saturated hydraulic conductivity tensor; k_r is the relative permeability coefficient; u_l is the water pressure in liquid phase; g is the gravitational acceleration; z is the elevation head; D_v is the diffusion coefficient of vapour in free air; and $\boldsymbol{\tau}$ is the tortuosity tensor**.**

The hydraulic storage properties of the soil are introduced by setting the dependence of porosity and liquid degree of saturation on suction. In this work, van Genuchten's expression is used, providing the link between effective degree of saturation S_{eff} and total suction:

$$S_{eff} = \left[1+\left(\alpha\psi\right)^{n}\right]^{-(1-1/n)} \tag{2}$$

where $S_{eff} = (S_l - S_{res}) / (1 - S_{res})$, S_{res} is the liquid residual degree of saturation, and α and n are model parameters.

A power law is used to fit the experimental relationship between porosity and total suction ($\phi = a \cdot \psi^{b}$, where a and b are fitting parameters) and between saturated hydraulic conductivity and void ratio ($K_{sat} = c \cdot e^{d}$ where c and d are fitting parameters).

The vapour mass concentration c_g^w is defined as the product between the saturated concentration of vapour $c_{g,sat}^{w}$ and the relative humidity h_{r} that depends on the temperature T and on total suction ψ via the psychrometric law:

$$c_g^w = c_{g,sat}^w \cdot h_r = c_{g,sat}^w \cdot \exp^{-\psi M^w / \rho_l R T} \tag{3}$$

where M^w is the molar mass of the water and R is the constant of perfect gases.

The vapour diffusivity coefficient in free air D_v and the saturated concentration of vapour $c_{g,sat}^{w}$ depend on temperature T [10], while the tortuosity is assumed equal to: $\tau = (\phi S_g)^{(2/3)}$ [11].

Thermal energy balance equation. The thermal energy balance equation can be expressed as shown in Equation (4), accounting for heat flux due to conductive and convective heat transfer [12]:

$$\frac{\partial}{\partial t}\left[E_{\mathrm{s}}\rho_{\mathrm{s}}\left(1-\phi\right)+E_{\mathrm{l}}\rho_{\mathrm{l}}S_{\mathrm{l}}\phi+E_{\mathrm{g}}\rho_{\mathrm{g}}S_{\mathrm{g}}\phi\right]+\nabla\cdot\left(\mathbf{i}_{\mathrm{c}}+\mathbf{j}_{\mathrm{E_l}}+\mathbf{j}_{\mathrm{E_v}}\right)=0 \tag{4}$$

where $E_s = c_{p,s}T$, $E_l = c_{p,l}T$, $E_g = c_{p,g}T + L_w$ represent the energy stored in the solid, in the liquid and in the gas phase, respectively. Such terms depend on the corresponding specific heat capacities ($c_{p,s}$, $c_{p,l}$ and $c_{p,v}$) and on the latent heat of vaporization $L_w(T)$. The conduction of sensible heat is accounted for by the Fourier law, *i.e.* $i_c = -\lambda \nabla T$, where λ is the isotropic thermal conductivity of the medium and is a function of the thermal conductivities of solid particles, liquid water, and vapour respectively weighted by the corresponding volume fractions. Convective energy fluxes related to the movement of the liquid water and vapour are evaluated as follows:

$$\mathbf{j}_{\mathrm{E_l}} = E_\mathrm{l}\left[-\frac{\mathbf{K}_{sat}k_r}{g}\ \nabla\left(u_l + \rho_l g z\right)\right] \tag{5}$$

$$\mathbf{j}_{\mathrm{E_v}} = E_\mathrm{g}\left(-D_v \boldsymbol{\tau} \nabla c_g^w\right) \tag{6}$$

Boundary conditions at the soil surface. The boundary conditions for the water mass balance are given by the product between the liquid water mass density and the actual evaporation rate at the soil surface, *AE*, as in [13]:

$$\boldsymbol{q}_{\mathbf{evap}} = \rho_\mathrm{l}\boldsymbol{AE}, \quad \boldsymbol{AE} = \boldsymbol{PE}\,\frac{p_{\mathrm{g,soil}} - p_{\mathrm{g,env}}}{p_{\mathrm{g,sat}} - p_{\mathrm{g,env}}} \tag{7}$$

where $\boldsymbol{PE}$ is the potential rate of evaporation, normal to the soil surface, $p_{g,soil} = p_{g,sat}(T)h_r(T)$ is the current vapour pressure of the soil at the soil-atmosphere interface, $p_{g,env} = p_{g,sat}(T_{env})h_r(T_{env})$ is the vapour pressure in the surrounding environment. $\boldsymbol{PE}$ depends on the mixing characteristics of the air above the evaporating surface. The actual rate of evaporation $\boldsymbol{AE}$ is equal to $\boldsymbol{PE}$ as long as the soil relative humidity is of 100% *i.e.* $p_{g,soil} = p_{g,sat}$. As $p_{g,soil}$ reduces, the rate of actual evaporation progressively decreases, limiting the flux of water vapour across the soil-atmosphere boundary.

In the thermal energy balance equation, the evaporation process acts as a heat sink, causing a reduction in temperature. Furthermore, the temperature difference between the soil and the environment generates a heat flux, which aims at re-establishing equilibrium. The thermal flux, that is imposed in the portion of the boundary subjected to evaporation, is thus (see [14]):

$$\boldsymbol{q}_{\mathbf{heat}} = L_\mathrm{w}\ \boldsymbol{q}_{\mathbf{evap}} - \varepsilon_\mathrm{s}\sigma\left(T_{\mathrm{env}}^4 - T^4\right)\boldsymbol{n} \tag{8}$$

where ε_s is the soil emissivity, σ is Stefan-Boltzmann constant and $\boldsymbol{n}$ is the unit vector normal to the soil surface.

4 Model validation

To investigate the water exchange in Opalinus Clay subjected to suction variations, the THM model described in Section 3 was implemented in the FE code Comsol Multiphysics® and validated against the results of the VET tests on the specimens reported above. [4] provides the durations and suctions of the VET tests, as well as the initial conditions, the water content and the volumetric strains following the drying and wetting processes. The relationship between the liquid degree of saturation and total suction is established under the assumption of homogeneous distribution of water content and strain on the entire tested specimens. The hydraulic conductivities related to the void ratio are estimated from the one-dimensional consolidations presented in [15] and are assumed isotropic. Table 1 summarizes the initial conditions and the samples' parameters used in the numerical simulations.

Table 1. Initial conditions and parameters used in the numerical simulations (D: main drying path and W: main wetting path)

Core sample	L3		L8		L11		L13	
Hydraulic path	D	W	D	W	D	W	D	W
α (MPa^{-1})	0.064	0.260	0.086	0.208	0.262	0.573	0.043	0.126
n	1.51	1.42	1.52	1.46	1.45	1.41	1.69	1.56
a	0.1808	0.1839	0.1513	0.1532	0.1712	0.1725	0.1911	0.1928
b	-0.01	-0.013	-0.007	-0.008	-0.006	-0.007	-0.015	-0.015
c (m/s)	0.0754		1188.1		1.13e7		0.0041	
d	17.3		19.5		27.3		16	
ψ_0 (MPa)	4		4		4		10	
T_0 (°C)	25		25		25		25	

Fig. 2 shows the imposed total suction for all tested specimens as a function of time, as well as the experimental data compared to the results of the numerical simulations, in terms of water content and degree of saturation. These results show that the FE model is able to successfully reproduce the VET tests along the hydraulic drying and wetting paths to which the specimens are subjected. Additionally, the equilibrium time is overall well captured by the model.

Some minor discrepancies can be observed at very high or very low suction levels, mainly due to the non-perfect fitting of van Genuchten's law for the water retention behaviour of the tested specimens. The model can be improved through the back analysis of the transient behaviour considering anisotropic hydraulic conductivity of Opalinus Clay.

This numerical model will be used to study in depth the role of advective and diffusive fluxes during the drying and wetting processes to which the Opalinus Clay will be subjected during its lifespan, as well as the effect that the environmental conditions (T, h_r, ψ) and the material properties (porosity, mineralogical composition and fabric) have on the hydro-mechanical behaviour of this material.

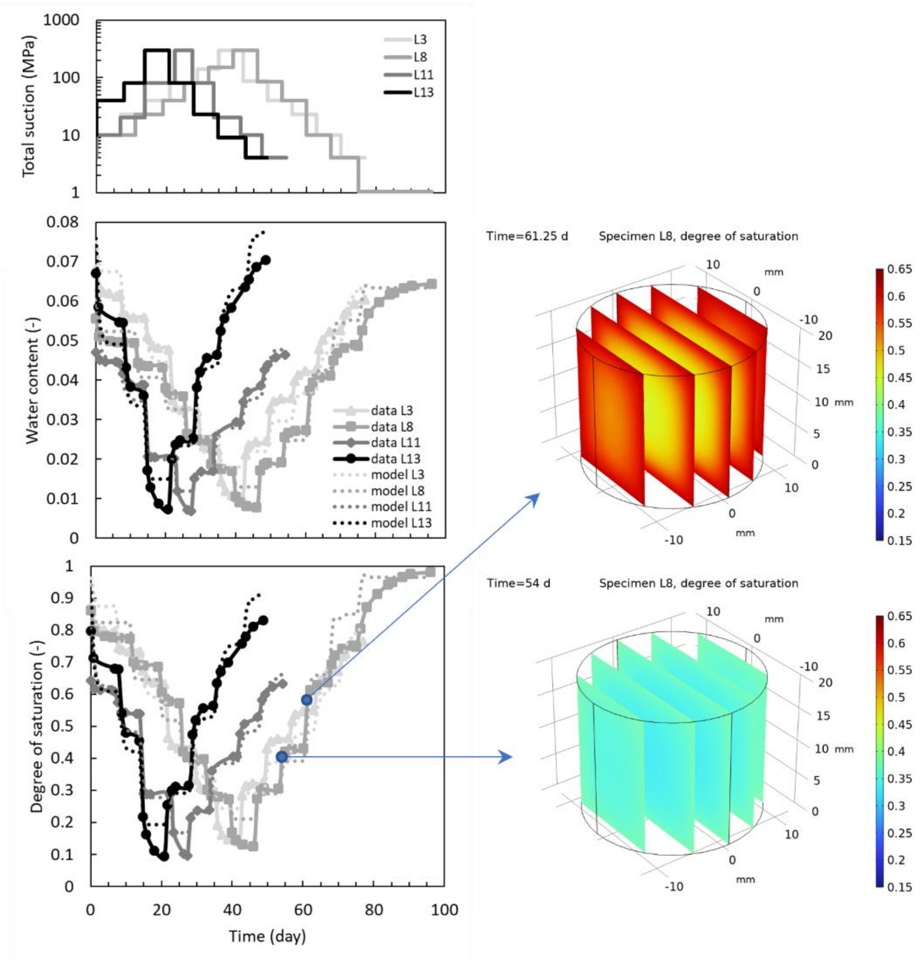

Fig. 2. Comparison between experimental and numerical results of VET tests.

5 Conclusion

In this study, the water exchange that takes place in Opalinus Clay from suction variation is investigated. In this regard, results of VET tests of four samples from Lausen site are used in order to determine the fitting parameters for the water retention properties and the evolution of void ratio and hydraulic conductivity.

The implemented numerical model is composed by the coupled system of water mass and energy balance equations. Appropriate boundary conditions are considered in order to simulate the water and heat flux from/to the geomaterial and its surrounding environment.

Preliminary analysis shows an overall good agreement between experimental data and numerical results in terms of change in water content, degree of saturation and equilibrium time. Perspectives for the improvement of the model are identified, such as the consideration of anisotropic hydraulic conductivity of Opalinus Clay, and better evolution laws for tortuosity and permeability. In this regard, additional experimental data of VET tests on Opalinus Clay from various sites will be considered in order to assess the role of the material heterogeneity on the water exchange mechanisms.

The numerical model will be used to define an accurate law for the tortuosity and permeability evolution of Opalinus Clay when subjected to suction variations, that is required to simulate the conditions of nuclear waste storage.

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