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THERMOHYDRAULIC TEST ON BENTONITE

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Swelling clays like bentonite are proposed as a buffer in high level nuclear waste repositories. An experimental device useful for thermo-hydraulic analysis of the samples behavior is outlined from room temperature to 373 K. The temperature distribution is established by axial heating of cylindrical samples. The spreading of the water content distribution is also determined from sample sections at the end of the run. An analytical model is build for steady states coherent with the experimental measurements. The transient temperature distribution and the final water content are also outlined via a numerical model. From experimental measurements the thermal conductivity value of the bentonite is $0.47^{1-\mathrm{Sr}}\cdot 1.15^{\mathrm{Sr}}$ W m^{-1} K^{-1}, where Sr is the bentonite degree of saturation (volume of water/volume of pores).

Keywords: *bentonite, nuclear waste, swelling clays, thermal conductivity*

Introduction

The nuclear waste management is a relevant problem in the nuclear industry. Nowadays, it is impossible to eliminate completely their radioactive activity. One of the solutions is to store the nuclear waste in places where would not be able to affect the population and environment. The storage proposed for different nuclear agencies of countries with nuclear power plants consists in vitrifying the nuclear waste in a canister. The nuclear waste produces heat and the vitrified material requires that the temperature increases does not change their properties [1, 2]. These canisters are introduced along the axis of cylindrical excavations in a host rock and surrounded by swelling clay. The groundwater would go into the gallery and the clay would swell, so the repository would be sealed.

Predictive modeling of thermo-hydro-mechanical (THM) behavior of clay barriers in radioactive waste repositories needs to use complex conceptual and numerical models where coupling between different processes that take place in the barrier must be taken into account. For instance, the thermal gradient originated by heat generation process in waste canisters induces flux of water in vapor phase that has influence on the water movement in liquid phase and induces changes in water saturation and soil deformation. Also these changes alter notably water and gas permeability and the thermal conductivity of the barrier.

During last years, numerical models have been developed to analyse the fully coupled THM behaviour of clay barriers [3–8] among others. The number of parameters that is necessary to know in order to carry out the numerical models is large: in a typical thermo-hydro-mechanical analysis the number of parameters needed to characterize each material may be about 15. In fact, it is important any simplification of the problem to evaluate the importance of the different phenomena which occurs in a clay barrier. A standard method is to separate the mass and heat flow from the mechanical part, which is related with the stress and strains, normally more difficult to solve than the heat flow part.

For the swelling clay, a new device has been developed. In the tests, a controlled heat flow is applied on a specimen with no deformation restrictions and with global water content remaining constant. These types of tests were already performed in a preliminary version back in 1949 [9]. More recently [10–12] have developed similar experiments in a clay barrier context. In the tests, temperature evolution at some points of the clay specimen is monitored and final water content distribution along the sample is measured. The aim of the present work is to show the test and its difficulties, because the material testing is a porous media with three phases, and the capabilities and limitations of the different models, analytic and numeric, for simulate the heat and water transport in steady and non steady state.

Experimental

The objective of the test is to apply a controlled axial flux of heat on one of the ends of a cylindrical specimen (38 mm diameter, 76 mm height) and to maintain

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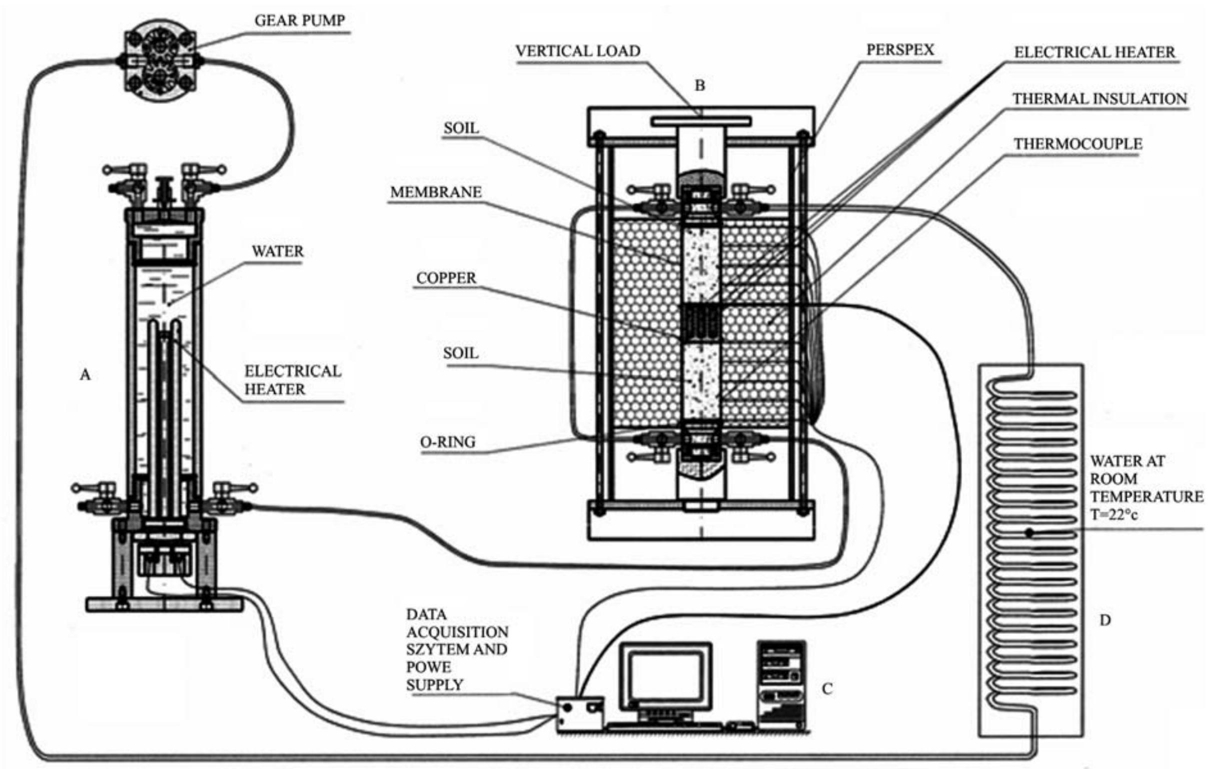

Fig. 1 Scheme of the experimental device. A – Thermostat ensuring the reference temperature B – Test device with heater, the two samples (up and down) and radial thermal protection, C – Controller, D – Cooler using water

the other end at constant temperature. A latex membrane that allows the deformation and keeps constant the overall water content, and a layer (5.5 cm thick) of heat insulating deformable foam surrounds the specimen. All parts are located into a container (a cylinder in perspex) that gives stiffness to the whole equipment. In order to assure the knowledge of heat flux entering in the sample, two specimens symmetrically placed respect to the heater are used in the tests. Figures 1 and 2 show a scheme of the equipment developed.

The heater is a copper cylinder (38 mm diameter, 50 mm height) with five small electrical resistances inside. The resistances are connected to an adjustable DC power supply that allows controlling the input power from 0 to 5 W. In the measurements, a constant power of 2.17 W has been used, reaching steady temperatures in the range of 70–80ºC in the hotter end of the specimen. In the cold end, a constant temperature of 30ºC is maintained by flowing water in a stainless steel canister (A and A' in Fig. 2) in contact with the soil sample. A temperature regulation system keeps constant the temperature of the contact between the head and the soil with variations smaller than 0.5 K.

In order to improve the sealing of the soil and the durability of the latex membrane in the contact with the heater, the membrane is surrounded by liquid silicone that solidifies in few hours. In the whole specimen, the water loss during the test due to diffusion through the membrane is about 0.1 g/day. The specimens are located in vertical position. In order to assure a good contact between the heads and the sample, a low stress (about 0.05 MPa) is applied on the upper zone of the equipment. Thermal conductivity of insulating foam (polyethylene 500) has been obtained from backanalysis of temperature measurements. A value of 0.039 W m^{-1} K^{-1} was obtained and was used in further analyses as a fixed and known value. Figure 3 shows the temperature distribution in the device along the test, computed using the formulation

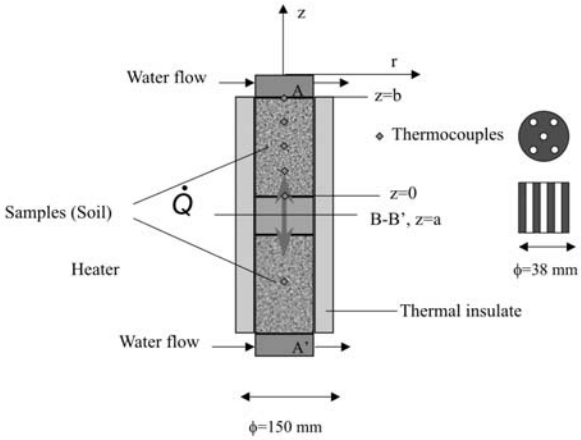

Fig. 2 Left – Outline of the heater (in B–B'), samples, reference temperature (in A–A') and position of the thermocouples. Right – Cu-cylinder, distribution of the resistances in the heater. The arrows indicate the flow direction

Table 1 Thermocouple position and temperature values in the axis device (r=0) (experimental and using different λ_s values)

z/mm	$T_{measured}$/°C	$T_{numerical}$ λ_s=0.879 W m^{-1} K^{-1}	$T_{analytical}$ λ_s=0.879 W m^{-1} K^{-1}	$T_{analytical}$ λ_s=0.972 W m^{-1} K^{-1}
0.0	75.6	75.8	78.7	75.8
20.0	61.5	61.7	63.4	61.6
38.0	50.2	50.4	50.7	49.7
60.0	39.4	38.8	39.8	39.4
78.0	30.0	30.0	30.0	30.0

presented below. Analyses of the heat transfer for cylindrical systems performed with Code Bright, the finite element code with the numerical analysis has been done [3, 13, 14], allowed evaluating the effect of lateral loss of heat, and it has been estimated as a close to 60% of the total heater power. That indicates the importance of performing a 3-D axisymmetrical analysis of the experiment.

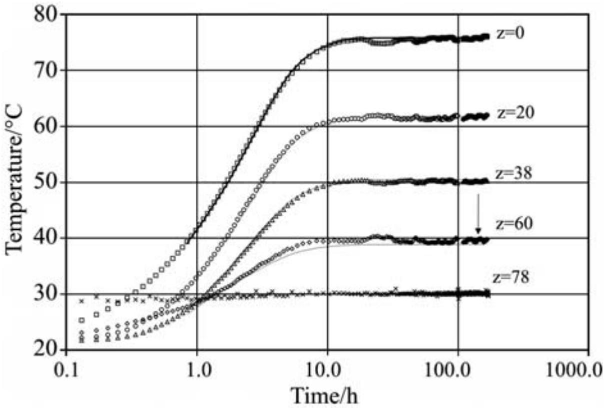

Fig. 3 Evolution of temperature in some points of the sample. The solid lines are the numerical simulation results in steady state conditions. The arrow indicates the daily temperature changes

During the measurements, temperatures in both ends and in three internal points of the specimen, located at regular intervals, are monitored by means of a data acquisition system controlled by a PC. The system is also used to impose a constant temperature (30ºC) at the cold end of the sample. Temperature measurements are concentrated in one of the two specimens of each test, whereas in the other specimen the temperature is only measured at the central point, just to check the symmetry of heat flux. At the end of the tests, diameter change is measured in some points of the specimen with a resolution of 0.01 mm. Finally, the soil samples are cut in six small cylinders and the water content of each one is determined.

An excessive dissipated power leads an excessive increase of the temperature (over 80ºC) and the latex membrane melts down, so there is uncontrolled water losses. This test cannot be maintained long time because there is a vapor leakage even with the latex membrane without any damage. This vapor leakage is quantified in 0.1 g/day, but these leakages depend on the sample initial degree of saturation and on the heater dissipated power. Minor effects can be related to daily temperature changes in the laboratory (24 h peak to peak with a temperature difference of 0.6ºC approximately). See, for instance the temperature ripple indicated by an arrow in Fig. 3.

Results an discussion

The material tested is a bentonite from the South-East of Spain (FEBEX bentonite), furnished and studied in the frame of FEBEX project [15]. Its geomechanical and geochemical properties are described extensively in [16, 17]. Bentonite has been compacted at dry density of 1.63 g cm^{-3} and with a water content of 15.33% (degree of saturation of 63%). Initial temperature of specimen was 22ºC. During 7 days a power of 2.17 W is dissipated in the heater. Also, the cold end of the specimen was maintained a temperature of 30ºC.

Temperatures measured during the heating period are shown in Fig. 3. Temperature reaches a quasi-steady state regime at 10 h after starting the test. Figure 4 shows water content measured at the end of the test in two samples. The evolution of water content is an evidence of the magnitude of water flows in both liquid and vapour phases. The differences observed (lower than 1%) between both samples can be related by the destructive method used to measure the final water content. Moreover, average final water content

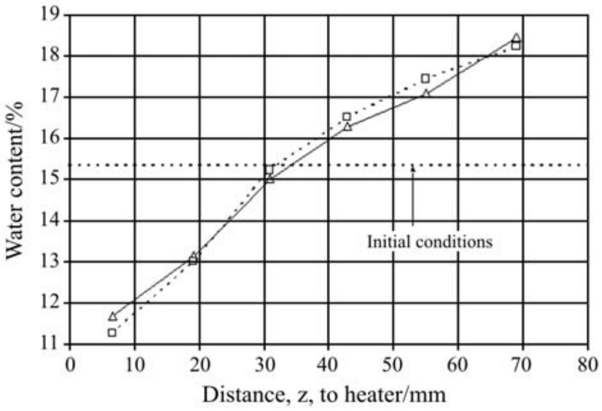

Fig. 4 Water content measured at the end of the test (7 days of heating) for the two samples

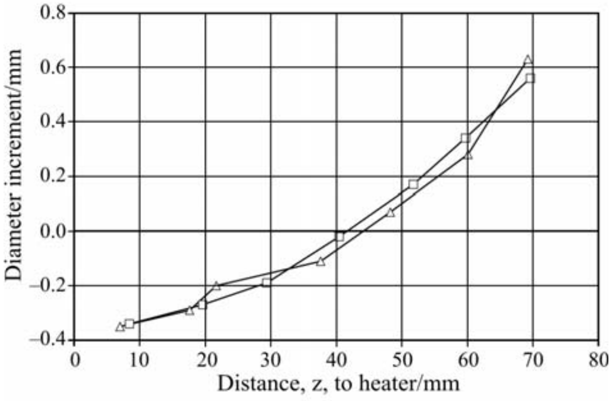

Fig. 5 Change of specimen diameter after 7 days of heating for the two samples

is slightly lower than the initial. The difference seems produced by minor water transport through the elastic membrane.

The change of diameter measured in the soil sample after the end of test is shown in Fig. 5. In the most part of the specimen the shrinkage due to the drying process originated by flow of water to the cold end of the specimen prevails over the dilation originated by temperature increment. Near the zone where the temperature increment has been lower, a significant diameter increment can be observed due to swelling of bentonite.

Analytic model

The heat transport problem and the Fourier's equation solution is a classic problem of the mathematical physics [18, 19] and provide the main tool for steady and for transitory thermal effects. In calorimetric approach the appropriate modeling (general Fourier equation in 1 or 3-D or the RC-analogy) permits an evaluation of the particular behavior of flat detectors when the dissipation is only partially detected by the set of thermocouples [20–22]. In [23] the Fourier equation is solved to apply the solution for the parameters identification. In our case, the analytical solution associated to the experimental device in steady state let to do fast analysis and allows furnishing an approach to experimental parameters. For instance, the cell dimensions, the materials characteristics, and so on. Below, the Fourier equation has been solved for steady state furnishing an approach to the thermal conductivity of the clay.

It is possible to study the temperature distribution without take in account the water flow if the soil is supposed as a homogeneous material. This approximation could be good while the water content change does not produce important variations in the heat conductivity. In samples with a large saturation degree, the temperature increases with time in the sample section in contact with heater because the sample dries and the heat conductivity falls. The thermo-hydraulic model used in this paper can reproduce the water content changes and its influence in the temperature distribution. The heat transfer for the steady state in a system with three materials requires some simplified hypothesis:

- The system is symmetric respect the heater horizontal media plane (B–B', Fig. 2) and axial axis (in *z* axis, Fig. 2).
- The dissipated power is homogeneous in the copper cylinder
- The heat flow in the sample and heater is axial while in the insulation is radial, so the flow vector inside the sample has the same direction that the axial axis. This roughly approximation would be only exact when the lateral losses are zero (the simplifications give good results).

Formalisms

For the thermal insulation (Fig. 2), the differential equation is:

$$\frac{\mathrm{d}^2T_\mathrm{a}}{\mathrm{d}r^2}+\frac{1}{r}\frac{\mathrm{d}T_\mathrm{a}}{\mathrm{d}r}=0 \qquad (1)$$

The solution of this differential equation gives the heat flow in function of the sample temperature or copper cylinder, depending which material is in contact with the thermal insulation. The boundary conditions are:

$T_\mathrm{a}(R)=T_\mathrm{c}(R)$ if $a\leq z<0$, $T_\mathrm{a}(R)=T_\mathrm{s}(R)$ if $0\leq z<b$ and $T_\mathrm{a}(R_\mathrm{ext})=T_2$, where T_c and T_s are the copper and soil temperature, R is the sample radius, R_ext includes R and the insulate thickness (Fig. 2) and T_2 the imposed temperature in the external boundary.

For the copper and soil cylinders (Fig. 2), a heat balance in a section has to be done:

- In the copper cylinder:

$$\frac{\mathrm{d}^2T_\mathrm{c}}{\mathrm{d}z^2}-\frac{2k}{\lambda_\mathrm{c}R}T_\mathrm{c}+\frac{Rq_0+2kT_2}{\lambda_\mathrm{c}R}=0 \qquad (2)$$

- In the soil cylinder:

$$\frac{\mathrm{d}^2T_\mathrm{s}}{\mathrm{d}z^2}-\frac{2k}{\lambda_\mathrm{s}R}(T_\mathrm{s}-T_2)=0 \qquad (3)$$

where $$k=\frac{-\lambda_\mathrm{a}}{R\ln(R/R_\mathrm{ext})} \qquad (4)$$

In the equations, z is the vertical coordinate with origin in the contact of soil with the heater (Fig. 2), q_0 is the power supply by volume unit, λ_c the copper heat conductivity, λ_s the soil heat conductivity and λ_a the insulate heat conductivity.

Temperature distribution results:

For the heater temperature along z axis:

$$T_{\mathrm{c}}(z) = A_1 \exp\left[\sqrt{\frac{2k}{\lambda_{\mathrm{c}} R}}\, z\right] + B_1 \exp\left[-\sqrt{\frac{2k}{\lambda_{\mathrm{c}} R}}\, z\right] + \frac{Rq_0 + 2kT_2}{2k} \quad (5)$$

For the soil sample temperature along z axis:

$$T_{\mathrm{s}}(z) = A_2 \exp\left[\sqrt{\frac{2k}{\lambda_{\mathrm{s}} R}}\, z\right] + B_2 \exp\left[-\sqrt{\frac{2k}{\lambda_{\mathrm{s}} R}}\, z\right] + T_2 \quad (6)$$

For the temperature in the isolator:

$$T_{\mathrm{a}}(r) = \frac{T - T_2}{\ln R / R_{\mathrm{ext}}} \ln \frac{r}{R_{\mathrm{ext}}} + T_2 \quad (7)$$

where T is T_{c} or T_{s} depending the material in contact with the isolator.

A_1, B_1, A_2 and B_2 parameters are calculated solving the following four equations which appear with the boundary conditions application:

- $z=a$: $\frac{\mathrm{d}T_{\mathrm{c}}}{\mathrm{d}z} = 0$, heater symmetry plane (B–B' axis, Fig. 2),
- $z=b$: $T_{\mathrm{s}}=T_0$, sample in the opposite side respect the heater (A and A', Fig. 2), T_0 is the temperature in this side
- $z=0$: $T_{\mathrm{s}}=T_{\mathrm{c}}$, heater and sample contact, temperature continuity and $\lambda_{\mathrm{s}}(\mathrm{d}T_{\mathrm{s}}/\mathrm{d}z)–\lambda_{\mathrm{c}}(\mathrm{d}T_{\mathrm{c}}/\mathrm{d}z)$ for continuity heat flow.

It is possible to see the influence on the temperature distribution of the different parameters with the equation solution. A generic case has been chosen and the parameters are changed to study their influence in the test. The geometric parameters are the same that the sample tested: (R=19 mm and R_{ext}=75 mm). The temperature in the external boundary of the isolation is fixed in 298 K and in the sample base in 303 K. From the heater voltage and intensity, the power dissipated in the heater by volume unit q_0 is 38216 W m^{-3}. The thermal conductivity for soil and for the isolation are 0.879 and 0.03755 W m^{-1} K^{-1} respectively, obtained with the numerical method calibration which uses the statistical method of maximum likelihood [24]. The copper thermal conductivity is 384 W mK^{-1} [25]. With the trial and error method, for the analytical model, the soil thermal conductivity is 0.972 W mK^{-1}, slightly greater than the thermal conductivity measured with numerical model.

The results from the analytical model in the steady state are compared with the results obtained from the numerical methods (3D with axisymmetry), which simulates the real problem (time dependent) with more approximation than the analytical analysis and it will be briefly described later. When the dissipated power in the heater increases also increases the heat losses in the base. The previous solution has more imprecision when the insulate thickness increases because the heat flow is more bi-dimensional than radial (related with the classical problem in cylindrical heat transfer). For a R_{ext}=250 mm, the maximum difference with the numerical model is 13ºC, and for the cell geometry test (R_{ext}=75 mm), the temperature difference is less than 3ºC.

Numerical modelling of transitory behavior

Finite element code 'Code Bright' [3, 13, 14] has been used to model thermo-hydraulic behavior of clay. Although the code allows studying in a coupled form the mechanical response of soils, only the thermal and water flow capacities of the code have been considered. Initially, the code was developed for non-isothermal multiphase flow of brine and gas through porous deformable saline media [3]. Only a brief description of relevant parts of these equations is included here.

In Code Bright, equations for mass balance were established following the compositional approach. That is, mass balance is performed for water, air and salt species instead of using solid, liquid and gas phases. Equation for balance of energy is established for the medium as a whole.

The final objective is to find the unknowns (temperature T and liquid pressure, P_{l}) from water mass and energy balance equations. Therefore, the dependent variables will have to be related to the unknowns by means of equilibrium restrictions and constitutive equations. The equilibrium restrictions are the psychrometric law, which relates the liquid pressure with the vapor content in the gas, and the Henry's law, which gives the relation between the gas pressure and the air content in the liquid. The constitutive laws are the Darcy's law for the liquid advective flow, Fick's law for the non advective vapor flow, the Fourier's law for the heat conductive transport and the retention curve for the relation between the degree of saturation and the liquid pressure. Gas pressure will be assumed constant in this analysis. All these laws are explained in [3, 13, 14].

In Fig. 3 the temperature measures and the simulation with the optimized results for thermal and hydraulic parameters are presented. It can be seen a very good agreement between measurements and numerical results. The results also show that the heat transport and the water flow works in two different time scales because conductivity heat flow is faster than mass transport.

Conclusions

A device for thermal tests in soils has been developed. It allows studying the heat and water flows in a swelling clay sample. Later, the measurements have been compared with the results from an analytical model and numerical model with satisfactory agreement. The analytical model permits to study the influence of the cell dimensions, boundary conditions and furnishes an approach to material thermal properties.

The numerical model allows an analysis of the measurements in steady and in non steady states with and without hydraulic coupling, so it is possible to separate the heat flow by conductivity from the heat flow by mass transport. It is possible to see the influence of changes in water content in temperature distribution because the water content influences in heat conductivity (increase with the water content increase). This phenomenon is observed in the measurements.

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