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ASSESSMENT OF HIGH-TEMPERATURE COUPLED EFFECTS IN MODELLING COMPACTED BENTONITES FOR NUCLEAR WASTE DISPOSAL

Gema URRACA[1], Ángel YUSTRES[2], Vicente NAVARRO[3], Laura ASENSIO[4]

ABSTRACT

The aim of this study is to examine thermo-hydro-mechanical and thermo-hydro-chemical effects in modelling bentonite at temperatures exceeding 100°C. For this purpose, two numerical tools using a double porosity approach are used. The inspection case is a bench-scale experiment with Wyoming bentonite. The results of both exercises contribute to enhance the understanding of thermal coupled effects in bentonites at high temperatures under repository conditions, and highlight the advisability of including fully coupled THCM (thermo-hydro-chemo-mechanical) effects in future modelling.

Keywords: nuclear waste disposal, bentonite, double porosity model, multiphysics coupled modelling.

INTRODUCTION

For some countries, nuclear energy is an important component of their strategies to address climate change while assuring access to cost-effective and reliable energy to support economic growth and human development (NEA, 2020). There is a broad consensus within operators and industry, safety authorities, policy makers and research and development institutions in the International Atomic Energy Agency and the OECD states that deep geological disposal is the most suitable end-point for the management of high-level radioactive waste (HLW) generated by nuclear energy (IAEA 2003; NEA, 2008).

Repositories for HLW use a multi-barrier system, including a natural geological barrier and an engineered barrier system, to isolate waste from the biosphere (NEA, 2003). As a part of this system, among other barriers, HLW is typically enclosed in metallic canisters, and buffer materials are to be placed between the canister and the host geological barrier. The use of compacted bentonite as a buffer barrier is being evaluated or has been decided by several regulatory national authorities around the world; for instance, in Finland, Sweden, France, Switzerland, Canada, Japan and Spain (Sellin and Leupin, 2013). As a buffer barrier, bentonite will be subject to complex THCM (thermo-hydro-chemo-mechanical) conditions. These conditions, involving heating, hydration, swelling and chemical interactions, can alter the initial state of the bentonite. In the near field of the canisters, temperatures of over 100°C can be expected. To simulate strong coupled effects over long periods, conceptual and numerical models must represent relevant THCM processes. Temperature effects have been studied, but not extensively above 100°C. The aim of this work is to assess thermo-mechanical and thermo-chemical influences using coupled numerical tools with a double porosity approach. This will lead to a better understanding of thermal effects in bentonites at high temperatures under repository conditions.

MATERIALS AND METHODS

A bench-scale experiment conducted by Chang et al. (2023) was modelled using the XMm model (Navarro et al., 2019) implemented in Comsol Multiphysics (Comsol AB, 2020). The test was carried out in a compacted Wyoming bentonite column, which experienced both heating to 200°C in the center and hydration from a sand-clay interface surrounding the column. The material tested was a sodium Wyoming bentonite (Cetco) with granular particles consisting mainly of montmorillonite (79%). The samples were prepared to a dry density of 1.20 Mg/m^3 and a water content of 18%. The saturating fluid contained mainly Na^+ and SO_4^{2-} ions, with an ionic strength of 0.314 M.

In the experiment, an aluminium vessel was used with cross-sample characterisation methods. The inner diameter was 0.17 m and the length was 0.46 m. A cartridge heater (0.30 m long) was housed in a 0.01 m diameter shaft through the center of the vessel. Thermocouple sensors were placed at various distances from the heater shaft (0 mm, 10 mm, 20 mm, 32 mm, 40 mm, 54 mm, 67 mm and 72 mm) to monitor the temperature distribution. The sand layer was saturated with the fluid at

1 *Universidad de Castilla-La Mancha, Ciudad Real, Spain, mgemma.urraca@uclm.es*
2 *Dr., Universidad de Castilla-La Mancha, Ciudad Real, Spain, angel.yustres@uclm.es*
3 *Prof., Universidad de Castilla-La Mancha, Ciudad Real, Spain, vicente.navarro@uclm.es*
4 *Dr., Universidad de Castilla-La Mancha, Ciudad Real, Spain, laura.asensio@uclm.es*

t=1 day under a pressure of 0.83 MPa. The heater was activated at t=2 days and the temperature of the bentonite on the heater shaft was stabilised at 200°C after 8 days. After 1.5 years, a post-dismantling analysis was conducted.

CONCEPTUAL MODEL

XMm model uses a double porosity approach, based on the framework of the Barcelona Expansive Model (Gens and Alonso, 1992). The macrostructure is associated with the space between bentonite particle aggregates, while the microstructure (assumed to be saturated) is associated with the intra-aggregate voids.

In the approach adopted, each structural level is assigned to a different continuum medium, while both media occupy the same spatial domain. The mass exchange between the two structural levels can be interpreted as a phase change process governed by the difference between chemical potentials (Navarro et al., 2015).

Thermal Problem

The thermal problem is addressed by solving the enthalpy balance. The enthalpy per unit volume of mineral is calculated as the sum of specific enthalpies of the different constituents, each multiplied by their respective masses per mineral volume (Gharbieh, 2021). The global heat capacity is influenced by the mass of water in different phases, and the heat of vaporisation is considered (Asensio et al., 2023). Enthalpy flow relative to the solid skeleton includes both convection and conduction terms, with thermal conductivity computed based on the degree of saturation according to Villar (2002).

Hydraulic Problem

The double porosity model balances water masses in macrostructure and microstructure separately. The macrostructural water mass includes both liquid water and vapour. Suction and degree of saturation at the macrostructural level are related by a double porosity water retention curve that accounts for changes in void ratio (De la Morena et al., 2021). Macrostructural water flow is determined using an extended Darcy law where the intrinsic permeability of bentonite is a function of macrostructural porosity (Gens et al., 2011). The diffusion of water vapour in the gas phase is calculated using Fick's law. Water properties are a function of temperature (Asensio et al., 2023).

Mechanical Problem

The mechanical problem is solved using the equilibrium equation. The increase in constitutive stress is related to the increase in elastic strain through the elastic matrix. The strain caused by the microstructure is computed from the variation of the microstructural void ratio, which, in this approach, is a function of suction and stress. The strain caused by the macrostructure is calculated using the Barcelona Basic Model (Alonso et al., 1990), which includes elastic and plastic terms due to changes in stress, suction and yield. Besides, a linear elastic term accounts for thermal strains (Ma and Hueckel, 1992).

Chemical Problem

Both macrostructural and microstructural water have dissolved chemical species. The THC couplings included in the model are based on Cabrera et al. (2024). It is a reactive transport model including precipitation and dissolution.

Bentonite is composed of inert (montmorillonite) and reactive minerals. Initial porosity distribution is determined using the structural suction of the microstructure (De la Morena et al., 2018 and Navarro et al., 2019). In the model used, the chemical problem is coupled to both suction and temperature.

RESULTS AND DISCUSSION

First, the problem was modelled with a THM model. Temperature evolution over time in the different sensors used in the experimental test is reproduced with high quality, as shown in Figure 1. A radial temperature distribution was created from 200°C (condition imposed on heater) to 95°C at the edge of the water inlet zone.

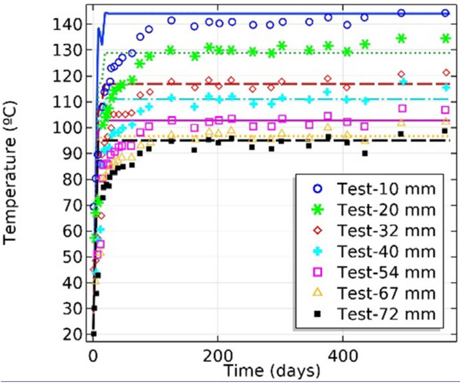

Figure 1 *Temperature evolution over time for different sensors (Lines-Model, Markers-Test)*

Density was analysed to investigate the mechanical problem. Experimental data available from both measurements made after test dismantling and results obtained from X-ray computed tomography (CT) scans performed by Chang et al. (2023) and Yoon et al. (2024) have been used. The fit between the measured bulk density data and the numerical model is satisfactory (Figure 2). Some differences are observed especially in the area close to the heater (about 10-25 mm), where the density in the model is slightly higher than in the experimental data. The density obtained experimentally suggests that the sample has expanded during the test.

Therefore, reduce the differences observed between test and model results, it would be advisable to include deformability at the boundaries of the sample. This could improve the density fit between the experimental results and the model data in the area near the canister.

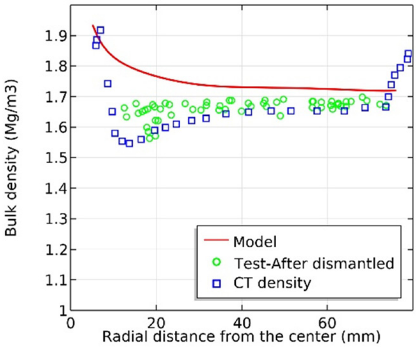

Figure 2 *Post-mortem density profile*

Second, a THC model was used to simulate the problem. Anhydrite, whose initial content was 0%, is observed to precipitate in the model, as expected under high temperatures and presence of calcium and sulphate ions. These results are qualitatively consistent with the experimental results. However, the 100% of anhydrite observed experimentally at the contact with the heater is not reproduced. This would require displacing all other minerals including montmorillonite, which could be achieved introducing deformability in the model.

In addition to the THM and THC couplings, full THCM coupling would be advised in future modelling. It is expected that, by introducing further couplings in a single model, the mechanical deformability will improve the fit of the density profile to the experimental one and contribute to refine the results of the chemical model.

CONCLUSIONS

A bench-scale experiment from the literature was selected to contrast the numerical model since it provides insights into the behaviour of bentonite under heating, hydration, swelling and mineral precipitation, aiding in the modelling of coupled processes in HLW. Coupled THM and THC formulations, implemented in finite element software, were used to model the experiment.

The results obtained show a high quality fit in the temperature distribution profile with time. In the case of the post-mortem profile of density, the fit between the experimental data and the model is satisfactory, with slightly higher density values in the model. About mineral precipitation, the modelling results are qualitatively consistent with the experimental data, but a coupling with the mechanical problem would be needed to refine the results quantitatively.

A numerical tool to solve the fully coupled THCM problem at high temperatures will be developed in further studies. The deformability of the cell suggested by experimental results will be introduced to improve the density results and to allow space for a 100% anhydrite precipitation at the contact with the heater.

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