KYT 2018: Bentonite investigations

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Outline

I. Introduction & overview (15 min)

II. Research projects in KYT2018 programme (15 min)
   I. UEFBENT (University of Eastern Finland)
   II. Helsinki University investigation
   III. THEBES (Aalto, VTT, Jyväskylä & Numerola)
Bentonite

Highly compacted bricks, made of dry bentonite, used to construct a barrier preventing possible contamination

during construction

during dismantling

© Villar, 2016
Bentonite

Dry compacted bentonite bricks swell if wetted, hopefully leading to a self-healing and impermeable barrier.

© Villar, 2016
Bentonite - microstructure

Bentonite behaviour is complex and to predict it reliably we need to understand the physical processes which leads to the observable macroscopic behaviour.

© Villar, 2016
Bentonite behaviour is complex and to predict it reliably we need to understand the physical processes which leads to the observable macroscopic behaviour.
Bentonite - microstructure

Forces acting in bentonite:

**Macroscale:** forces from mechanical loading

**Meso:** capillary forces due to water menisci between aggregates

**Micro:** Forces within laminae of clay minerals (mainly montmorillonite) – inter-particle distance exceeds ~30-40 Å van der Waals attraction & electric repulsion on atomic level

inter-particle distance below ~10-20 Å – electric repulsion on atomic level

© Santamarina, 2001
Bentonite swelling

1. Number of Montmorillonite platelets in a pile decrease, spacing between them increase.

Interlaminar porosity increases.

Some swelling occurs.

More water in the smallest pores

© Villar, 2016

THERES: KYT project investigating bentonite
Bentonite - microstructure

MX 80 clay
$\rho = 2 \text{ Mg/m}^3$

SAMPLE AT HIGHER WATER CONTENT AND LOWER SUCTION HAS MORE WATER LOCATED IN VERY SMALL PORES (< 35 nm)

Delage et al., Géot. 2006
Bentonite swelling

2. Size of the aggregates changes. They become also more easy to break. Double porosity structure slowly disappears.

Interlaminar porosity still increases.

More some swelling occurs.

© Delage et al. 2006
Bentonite swelling: montmorillonite water absorption

3. Amount of water between layers of montmorillonite changes

Additionally, the swelling is affected by the mesoscale – size of the aggregates changes and capillary forces make a difference.

© Delage, 2015
© Sayiouri et al. 2000
Bentonite swelling

4. Swelling pressure is affected by the mechanical, thermal, conditions, as well as availability of water and water composition (e.g. salinity).

Similarly, the number of layers / volume change is different for different mechanical conditions: complex thermo-hydro-mechanical-chemical coupling

© Delage, 2015
Water transport

Due to complex microstructure, water transport is complex, mixture of transport via vapour and liquid.

Also highly temperature dependent

Necessary to predict swelling, as swelling depends on amount of water available.

Prediction in time is crucial, as material is affected by stress history

Cui et al., Phys. Chem. Earth 2008
Temperature

Temperature affects forces between particles, both in microscale, as well as mesoscale.

Therefore the material properties are temperature dependent.

Thermal conductivity non-linear and coupled to water & mechanical properties

Prediction in time is crucial, as material is affected by stress history

© Tang et al., 2008
Water salinity affects the hydraulic behaviour of bentonite, as well as mechanical properties. It also affects the swelling of bentonite aggregates and generally lowers the swelling pressure.
Water in liquid phase transport salt, but water vapour not! In non-isothermal condition creation of high saline zones (zone reached wetted by liquid water which evaporates) likely. Effects on material being investigated.
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UEFBENT (University of Eastern Finland)

Investigates swelling of the montmorillonite when saturated with different solutions.

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Background and the targets of the study

- The aim of the modelling is to rationalize experimental swelling pressure observations and data to be used in the selection of the buffer material.
- We have developed a swelling pressure model based on an atomic-level computational approach.
- To predict swelling pressure trends of montmorillonite-beidellite smectites with respect to variations of layer charges, charge locations, and interlayer cations.
- To investigate the swelling pressure in salt solutions and at different temperatures.
Measurement and simulation of clay swelling pressure

Swelling pressure measurement

Swelling pressure model

Swelling pressure simulation
Simulated swelling pressure of sodium smectites. Expansions of 50%, 100% and 150% correspond to dry densities of 1650kgm$^{-3}$, 1240kgm$^{-3}$ and 990 kgm$^{-3}$ respectively.

- 50%, 100% and 150% expansions have been chosen to represent different swelling states.
- High and low layer charge reduces swelling pressure.
- Charge located in the tetrahedral sheets reduces swelling.
Conclusions and Acknowledgements

- High or low layer charge and high charge fraction on tetrahedral sheets reduces the swelling pressure

- Interlayer cation species strongly influence the swelling characteristics (Na⁺ / Ca²⁺)

- Increased salinity of surrounding water reduces swelling pressure

- New MD model for swelling pressure simulation and clay material selection has been demonstrated
BENTO (University of Helsinki)

Bentonite erosion and radionuclide interaction processes
Bentonite erosion and radionuclide interaction processes (BENTO)
Pirkko Hölttä, Outi Elo and Valtteri Suorsa
University of Helsinki, Department of Chemistry, Radiochemistry

Introduction
Bentonite erosion resulting in the formation of stable and mobile colloids may increase radionuclide transport in the geosphere and have a direct impact on the overall performance of the bentonite buffer.

Experimental
Bentonite erosion, colloid formation and stability
- Batch type experiments (MX-80, Nanocor PGN)
- Photon correlation spectroscopy (PCS): particle size, concentration and zeta potential
- The effect of salinity, pH and time
- Bentonite erosion kinetics
Radionuclide sorption/desorption on bentonite/montmorillonite suspension and colloids
Colloid mobility
Colloid-mediated radionuclide transport

3 Poster presentations
$^{237}$Np sorption and the influence of colloids on Np(V) transport

- MX-80 bentonite colloids, Kuru grey granite
- Batch sorption/desorption (pH, isotherms, solid/liquid, kinetics)
- In-situ ATR FT-IR and EXAFS in HZDR - chemical nature of the complex between Np and Na-montmorillonite or colloid
- Drill core and crushed rock column experiments

**Measured breakthrough curves of Np-237 through drill core column (a) and crushed granite column (b) flow rates of 0.8 ml/h in 10 mM NaClO$_4$ (pH8) in the absence and presence of colloids. Despite the low uptake of Np(V) by the colloids, the Np(V) breakthrough is enhanced in the presence of colloids.**
Radionuclide, bentonite colloid and granite interaction

Block-scale experiments provides an intermediate scale study between conventional laboratory (cm) and in-situ experiments (m)
- Kuru Grey Granite Block with water conducting natural fracture (90 x 94 x 70 cm)
- Flow conditions were determined using $^3$H, $^{36}$Cl and Amino G
- A good reproducibility and similar breakthrough curves obtained with several tests confirm the block set-up is suitable for the further experiments.
- Colloid/radionuclide interaction experiments using $^{152}$Eu as an analogue for trivalent actinides

Supportive experiments to study the colloid stability in the ternary system granite – montmorillonite colloids – radionuclide
Colloid analysis by Laser Induced Breakdown Detection (LIBD) (KIT/ENE)

The breakthrough curves with different conservative tracers.

The proportional $^{152}$Eu distribution in GGW solution (blue), on Ni-montmorillonite colloids (green) and granite grains (red) as the function of time.
THEBES

THMC Behaviour of the Swelling Clay Barriers
THEBES goals

Experimental research → Data for modelling → Creation & validation of numerical models → Implementation into numerical software

feedback loop

Soft clay barriers: thermo – hydro – mechanical – chemical coupling

Case studies
THEBES research: VTT

Experimental research → Data for modelling → Creation & validation of numerical models → Implementation into numerical software

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Soft clay barriers: thermo – hydro – mechanical – chemical coupling

Case studies
THEBES research: VTT

- Hydromechanical experiments
- Microstructural studies
- Chemical experiments
- Modelling

Matusewicz 2013
Hydromechanical experiments
- objective: determining the mechanical behaviour of unsaturated bentonite at high confining pressures
- high pressure triaxial cell – 32 MPa
- various water contents, densities, chemical conditions

Microstructural studies
- objective: characterization of bentonite structure and water distribution
- x-ray methods (SAXS), microscopy
- new method: atomic force microscopy (AFM)
  - geometrical characterization of montmorillonite layers
Chemical experiments
• objective: determine porewater chemical composition and evolution by different methods
• in situ ion selective electrodes (ISE)
  • activities (e.g. Cl\textsuperscript{-}) measured directly in compacted bentonite
• squeezing water from bentonite
  • ion diffusion and cation exchange during squeezing

Modelling objective:
develop a large deformation model combining water transport, mechanical behaviour and chemical effects for bentonite
THEBES research: Jyväskylä

Experimental research → Data for modelling → Creation & validation of numerical models → Implementation into numerical software → Case studies

Soft clay barriers: thermo – hydro – mechanical – chemical coupling

feedback loop
THEBES research: Jyväskylä

- X-ray tomographic method was previously developed and used for measuring 4D (3D + time) water content and deformation in MX80 bentonite samples during constant volume wetting → data was used in THEBES project

Measured water content and deformation (vertical distributions)
THEBES research: Jyväskylä

- **X-ray imaging method** was developed and used for measuring water content and deformation in MX80 bentonite samples during axial 1D wetting and swelling

- Phase 1, Free swelling: An extensive set of experiments completed

- Phase 2, Constant volume wetting: Experiments in progress

1. Axial 1D wetting and free swelling

2. Axial 1D wetting in constant volume. Result from the first experiment
THEBES research: Numerola

- Experimental research
- Data for modelling
- Creation & validation of numerical models
- Implementation into numerical software
- Case studies

Soft clay barriers: thermo – hydro – mechanical – chemical coupling
THEBES research: Numerola

- Mechanical model for bentonite (supporting Jyväskylä research)
- Hydraulic transport (Kröhn 2010)
- Thermal transport (Jussila, 2007)
- Numerrin solver development and support (used in the whole THEBES network)

1. Axial 1D wetting and free swelling

2. Axial 1D wetting in constant volume. Result from the first experiment
THEBES research: Aalto

Experimental research → Data for modelling → Creation & validation of numerical models → Implementation into numerical software

feedback loop

Soft clay barriers: thermo – hydro – mechanical – chemical coupling

Case studies
Thermo-hydro-mechanical FE simulations

Taking into account well established laws of physics and thermodynamics

**mechanical:** amended BBM, models by della Vecchia et al. (2013, 2014, 2015), new models which take into account micro and macro structure of bentonite are in development

**hydraulic:** number of models for water retention, Philip & De Vries model for vapour transport, extended Darcy law for liquid water transport, Henry’s law for solubility of water, phase changes and heat effects are taken into account

**thermal:** heat flux in solid, water and gas phases, full energy balance / coupling

Some experiments on water retention behaviour of MX80 bentonite and its microstructure in saline solutions leading to inclusion of those into modelling (chemical coupling)
Validation: CIEMAT Mock-Up test

Example: Simulation of CIEMAT Mock-Up test for 2500 days (Martin et al. 2006)
The hydro-thermal coupling is based on the theory proposed by Philip & De Vries (1957).

Axisymmetric conditions

Temperature

Relative humidity

Porosity

THEBES: KYT project investigating bentonite
Validation: CIEMAT Mock-Up test

Calculated relative humidity versus measurements

![Graph showing calculated relative humidity versus measurements](image)
Long-term safety of nuclear waste management

Safety case

- Canister performance
- Microbiological effects
- Buffer and backfill performance
- Other coordinated projects?
- Other safety studies

Bentonite!
Thank you