Bentonite buffer

Wojciech Sołowski, Aalto University
Importance of the bentonite buffer

Nuclear waste storage safety important for the society
- highest research standards required
- highest ethical standards
- research must be of such high quality that it is undisputable

No currently operating deep geological storages for nuclear waste
- research should be novel and of importance not only for Finland but worldwide
- opportunity to gain world-leading expertise
- if our research is world leading, it should be published in best journals and highly cited
Talk

1. Research overview from the outside review / financing body perspective
   - some introduction about science, research and research quality
   - overview taking into account the special societal importance of the research related to nuclear waste storage solutions

2. Technical overview of the research results related to bentonite buffer funded by KYT2018 programme
Blue Monday

Has anyone heard about Blue Monday (Sininen Maanantai)?

Who thinks there is research behind it?

Who trust the research?
Why TODAY is the most depressing day of the year: Combination of post-Christmas finances, bad weather and long wait for summer combine for 'Blue Monday'

- Researchers have dubbed January 21 as Blue Monday based on surveys
- 87% of the population feel the weather has a direct influence on their mood
- Two-thirds (61%) of people will feel depressed on their way to work today
- Manchester and Leeds topped the list of the places which suffer worst

Tänään on vuoden masentavin päivä

"Sininen maanantai"


THEBES: KYT project investigating bentonite
Blue Monday

- No credible research behind Blue Monday!
- Designed as a PR stunt to sell holidays
- For some reason became successful story

_Guardian_ columnist _Ben Goldacre_ reported that the press release was delivered substantially pre-written to a number of academics by _public relations_ agency _Porter Novelli_, who offered them money to put their names to it.
Research credibility

Many recent hits to research credibility including:
- A Guardian investigation, in collaboration with German broadcaster Norddeutscher Rundfunk, reveals the open-access publishers who accept any article submitted for a fee (2018)
- Three US researchers have pulled off a sophisticated hoax by publishing fake research with ridiculous conclusions in sociology journals to expose what they see as ideological bias and a lack of rigorous vetting at these publications (2018).
- **Beforehand**, scandals related to climate warming research – Climategate at the Climatic Research Unit (CRU) at the University of East Anglia (UEA) (2009)
- 6 Elsevier journals exposed to be established by producers of pharmaceutics, to prop up their products (2005)
Research credibility: positives

6 Elsevier journals exposed to be established by producers of pharmaceutics, to prop up their products (2005)
  • exposed, removed

Climategate at at the Climatic Research Unit (CRU) at the University of East Anglia (UEA) (2009) – cleared of all the charges
  • First announced in December 2009, a British investigation commissioned by the UEA and chaired by Sir Muir Russell, published its final report in July 2010. The commission cleared the scientists and dismissed allegations that they manipulated their data. The "rigour and honesty" of the scientists at the Climatic Research Unit were found not to be in doubt. The panel found that they did not subvert the peer review process to censor criticism as alleged, and that the key data needed to reproduce their findings was freely available to any "competent" researcher.
Research credibility: US experiment

Three US researchers have pulled off a sophisticated hoax by publishing fake research with ridiculous conclusions in sociology / gender studies journals to expose what they see as ideological bias and a lack of rigorous vetting at these publications (2018).

- Managed to get 7 out of 20 papers published, some after many rejections
- The experiment has been cut short due to external inquiries
- Some good journals accepted clearly fake papers – there is a problem with peer review

- Typically top 10% (90+) is good, top 25% is acceptable (75+)
- Finnish JUFO system has its faults, but can be also used (free to check)
- Very low ranked journals (bottom 10% or unlisted) may accept any research with very poor review
Research credibility

Peer review has its faults, but it is still very difficult to get a substandard paper published in a good journal

- Peer review is currently the best way to ensure the quality of the research
- Better journals have much better review quality, and thus the research is usually trustworthy
  - Typically top 10% (90+) is very good, top 25% is ‘good enough’ (75+), as listed by Scopus
  - Finnish JUFO system has its faults, but can be also used (free to check)
  - very low ranked journals (bottom 10% or unlisted) may accept any research with very poor review; bottom 50% can be problematic
- Perhaps best – but not available immediately – is to see how the research is followed – the number of citations (discipline / subject adjusted) are still quite reliable metrics
Research credibility: conclusions

**Peer review** has its faults, but it is still very difficult to get a substandard paper published in a good journal

- Single publications in poor journals may (**but does not have to**) indicate relatively poor quality of the work
  - incremental research
  - may be e.g. an invited paper
- Consistent publishing in low ranked journals may indicate possible low quality of research (or the researcher’s lack of care about the publications impact)
- Interaction with the general society (invited talks, seminars etc.) may indicate high quality of research
- Cooperation with best research institutions in the field can indicate high quality of research
KYT2018 bentonite: research overview

Wojciech Sołowski, Aalto University
KYT2018 bentonite research evaluation

Alphabetical order of institutions

Aalto University (THEBES)

Journal papers:


Acknowledgements

The authors would like to gratefully acknowledge that the presented research has been funded by KYT2018 Finnish Research Programme on Nuclear Waste Management via THEBES project.
KYT2018 bentonite research evaluation

Alphabetical order of institutions

Aalto University (THEBES)

Conference papers – if Scopus listed, they are peer reviewed, but they more indicate interaction with society and gathering early feedback for further publishing – high quality research normally published in journal papers (in the end)


3. Abed, A, Sołowski, WT, Romero, E & Gens, A (2018), Inclusion of chemical effect in a fully coupled THM finite element code. in C Ng, A Leung, A Chiu & C Zhou (eds), Unsaturated Soils vol. 2, The Hong Kong University of Science and Technology, Hong Kong, pp. 827-832, International Conference on Unsaturated Soils, Hong Kong, Hong Kong, 03/08/2018. Indexed in Scopus


Seminar talks:

- 2017 Politecnico di Milano
- 2019 Imperial College London

Website maintained: http://solowski.info/thebes/

All acknowledge KYT2018 programme as the only source of financing
KYT2018 bentonite research evaluation
Alphabetical order of institutions
Numerola Oy (THEBES)

Conference papers

Indexed in Scopus
KYT2018 bentonite research evaluation

Alphabetical order of institutions

University of Jyväskylä (THEBES)

Journal paper

Acknowledgements
This work was funded by the Finnish Research Programme on Nuclear Waste Management KYT2014 through the project “Assessment of bentonite characteristics (BOA)”. The authors appreciate the contribution by Pirkko Hölttä (University of Helsinki, Laboratory of Radiochemistry) who provided the synthetic groundwater used in the wetting experiments.

Conference papers


Not indexed in Scopus

© Scopus

Presentations in BelBar project and Beacon seminar (EU project seminars)
KYT2018 bentonite research evaluation

Alphabetical order of institutions

VTT (THEBES)

Journal papers


ACKNOWLEDGMENTS

Andrew Root (Magsol) is acknowledged for running the NMR experiments and fruitful discussions. SAXS experiments were performed by Ville Liljestrom at the X-ray laboratory of Department of Physics at Helsinki University. BOA project (KYT2014) is acknowledged for funding.

MRS Advances indexed in Scopus, but no metrics for MRS Advances is

No KYT programme acknowledged

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THEBES: KYT project investigating bentonite
KYT2018 bentonite research evaluation

Alphabetical order of institutions

VTT (THEBES)

Journal papers

Report


Acknowledgements

The research leading to these results received funding from Posiva Oy (Finland). Andrew Root (MagSol) is acknowledged for carrying out NMR measurements. The authors thank the Alternative Buffer Material (ABM) project and Clay Tech for the opportunity to use the samples for these experiments. Dr A. Muurinen, Dr T. Carlsson and Prof. M. Olin are acknowledged for discussions and ideas on the subject. Dr A.M. Fernández and Dr H. Van Baelen are acknowledged for many constructive comments and suggestions on the manuscript. © 2016 The Mineralogical Society

Clay Minerals

Earth and Planetary Sciences
Geochemistry and Petrology

#65/113 42nd

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KYT2018 bentonite research evaluation

Alphabetical order of institutions

University of Eastern Finland

Journal papers

Acknowledgements

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Acknowledgements

Financial support provided by Posiva Oy and Finnish Research Programme on Nuclear Waste Management (KYT) is gratefully acknowledged. Timothy Schatz is gratefully acknowledged for helpful discussions. The computations were made possible by use of the Finnish Grid Infrastructure resources.
KYT2018 bentonite research evaluation

University of Eastern Finland

Journal papers

Acknowledgments

Financial support provided by Posiva Oy was gratefully acknowledged. The computations were made possible by use of the Finnish Grid Infrastructure resources.


Acknowledgment

Financial support provided by the Finnish Funding Agency for Technology and Innovation TEKES and the European Union/European Regional Development Fund (ERDF) for the “Sliding Surfaces” project and Posiva Oy are gratefully acknowledged. The computations were made possible by use of the Finnish Grid Infrastructure resources.

2 Conference presentations, but no publications associated
KA2018 bentonite research evaluation

University of Helsinki

Journal paper

Acknowledgements:
The authors would like to thank the staff at the beamline BM20, ESRF, for technical help during the EXAFS measurements. Karsten Heim and Carola Eckardt are thanked for their help with the ATR-FT IR investigations and N₂-BET analyses, respectively. This research was financially supported by the European Atomic Energy community’s Seventh Framework Programme (FP7/2007-2011) under grant agreement n° 295487 and Finnish Research Programme on Nuclear Waste Management (KYT 2018). Additional funding from the German Ministry of Education and Research (BMBF), Project number 02NUK021CX, for travel costs is also acknowledged.

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2 Conference posters, but no publications associated

THEBES: K YT project investigating bentonite
THEBES: KYT project investigating bentonite
Conclusions:

- To maintain trust in your research, publishing in best scientific journals is key
  - consistent publishing in low ranked journals leads to questions
- Once the trust is broken, it is very difficult to regain it
- Any breaks in your research integrity will be used to criticise your research
- The new project funding decisions should be made very careful, as insufficiently transparent decisions, as well as decisions which are not clearly fair, will be criticised

- It is essential to maintain trust in the KYT funded research as fairly and objectively distributed, independently from any external pressures
Technical overview

Wojciech Sołowski, Aalto University
THEBES network

Wojciech Sołowski
THEBES overview

Network activities: meetings, cooperation, dissemination

Yearly seminars with invited speakers (autumn / winter):
- 2015 - Prof. Pierre Delage (Ecole des Ponts ParisTech)
- 2016 - Dr. Dr María Victoria Villar (CIEMAT)
- 2017 - Prof. Olli Ikkala (Aalto University)

Other international cooperation, seminars and visitors:
- 2015 – visit & seminar at Université de Pau et des Pays de l'Adour (UPPA)
- 2015 – visit & seminars of Dr. Klaus-Peter Kröhn
- 2017 – seminar at Politecnico di Milano by Sołowski
- 2015, 2016, 2017 – visits of Abed in UPC BarcelonaTech
- 2018 – visit and seminar of Dr João Mendes (Northumbria University)
- 2019 – seminar at Imperial College London by Sołowski

Yearly management meetings (June):
- 2015 & 2018 – Helsinki
- 2016 & 2017 - Jyväskylä

Long-term website: solowski.info/thebes
Aalto University
(THEBES project)
Wojciech Sołowski
Thermo-hydro-mechanical-chemical 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.  
New model relating mineralogy, microstructure and water retention behaviour

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

**chemical**: simple model which takes into account salt and salt migration in water.
Thermo-hydro-mechanical-chemical FE simulations
Taking into account well established laws of physics and thermodynamics

Governing Balance Equations
Mass balance (water, dry air, salt and solid component)

Compositional method is employed to derive the balance equations (Panday & Corapcioglu, 1989)

Idealization: three phases (liquid, gas, solid) and three components (water, dry air, solid particles and salt)
Validation: 1D isothermal infiltration

 Isothermal infiltration results

\( h_{w} = 122.32\, \text{m} \)

FEM model – hydraulic and mechanical boundary conditions

Relative humidity over time: Numerical results versus measurements

- Measurements
- CODE_BRIGHT
- Aalto Code

Point A
Point B
Point C
Validation: CIEMAT Mock-Up test
Full experimental details and results are given in Martin et al. (2006)

Simulation of the first 2500 days
Validation: CIEMAT Mock-Up test
Finite Element Model

- Axisymmetric conditions
- Geometry and Finite Element Mesh
- Mechanical boundary conditions
- Hydraulic boundary conditions
- Thermal boundary conditions

THEBES: KYT project investigating bentonite
Validation: CIEMAT Mock-Up test
Measurements versus numerical results

Relative humidity

Temperature

THEBES: KYT project investigating bentonite
Validation: CIEMAT Mock-Up test
Measurements versus numerical results

Water intake

Swelling pressure
Validation: CIEMAT Mock-Up test
Some numerical results over time

- Temperature
- Relative humidity
Validation: CIEMAT Mock-Up test
Some numerical results over time

Porosity

Displacement
Simple coupling of chemical effect
Governing mass balance

\[
\begin{align*}
\frac{\partial (\phi^i \rho^i \omega^i_k)}{\partial t} + \nabla \cdot \left( \phi^i \rho^i \omega^i_k \mathbf{v}^i \right) + \nabla \cdot \mathbf{j}^i_k &= Q^i_k \\
\frac{\partial ((1-n)\rho^s C_s)}{\partial t} + \frac{\partial (nS^l C)}{\partial t} + [nS^l C + (1-n)\rho^s C_s] \frac{\partial \mathbf{v}}{\partial t} + \nabla \cdot (q^l C) - \nabla \cdot (nS^l D_h \nabla C) &= 0
\end{align*}
\]

\[C_s = K_d C; \quad D_h = \tau D_o + \alpha \left| \frac{q^l}{nS^l} \right|\]
Simple coupling of chemical effect
Validation against experimental results salt transport (Warrick et al. 1971)

n = 0.38
$D_0 = 6.94 \times 10^{-10} \text{ m}^2/\text{s}$
$\alpha_L = 0.01 \text{ m}$
$\alpha_T = 0.0 \text{ m}$
$k_w = 4.4558 \times 10^{-6} \text{ m/s}$
$K_d = 0.0$

- The test starts by water infiltration and salt injection at the top boundary with concentration of 209 meq/l.
- At $t = 168.0 \text{ min}$, the salt concentration dropped to zero.
- Simulation time: 10 hours.
Verification: Density dependent solute transport (Elder 1967)
Flow field due to varying salt concentration
Water retention curve including microstructure effect:
Considered structural levels

- Clay aggregates
- inter-aggregates pore (macro-pore)
- Clay layer
- inter-layers pore
- inter-layers water molecules (0.4Å-12Å)
- empty intra-aggregate pore (micro-pore 12Å-200nm)
- empty macro-pore (> 200nm)
- water-filled micro-pore
- water filled macro-pore

THEBES: KYT project investigating bentonite
Water retention curve including microstructure effect:
Evolution of micro-void ratio with saturation

Under review (2 round) in Geotechnique (leading journal in the field), other paper submitted to International Journal of Numerical Methods in Geomechanics (outcome uncertain)
Developments
Block–pellet Homogenization

**Figure 2.** Schematic of the laboratory test.
Thebes code numerical results
Some graphical output

Horizontal stress
Vertical stress
Total water flux

Dry density
Degree of saturation
Total displacements x 5
Numerola Oy

THEBES project

Mika Laitinen
Numerola Oy – Models and software for simulating bentonite

• Numerola Oy has developed software for simulating thermal, hydrological and mechanical evolution of bentonite.

• Thermal model based on heat conduction depending on water content.

• Hydrological model accounts for diffusion of water and water vapour.

• Hydromechanical model based on elastoplastic model of Markku Kataja and experiments of Jyväskylä University. Mechanical parameters depend on water content.

• Implemented with Numerrin modelling language and package of numerical methods (in-house house software of Numerola Oy).
Numerola Oy – The models and software are being validated by simulating several experiments of JYU and CIEMAT

Simulating bentonite wetting and swelling experiment of JYU. Comparing simulated and measured deformations.
Numerola Oy – The hydromechanical model parameters are fitted to free swelling and constant volume experiments of JYU

Simulating constant volume experiment of JYU. Comparing simulated and measured displacements and mechanical stress.
Univeristy of Jyväskylä
(THEBES project)
Markku Kataja
Jyväskylä research – Free swelling experiments

- X-ray imaging method was developed and used to monitor free swelling of MX80 bentonite samples (Fig. 1).
- A set of samples (Table 1) was measured and the results were collected into a databank (temporary link: users.jyu.fi/~hpatana/bentonite_databank).

![X-ray microtomographic scanner and sample holder used in free swelling experiments.](image)

**Figure 1:**

![Example of results: bentonite and water content at three instants of time.](image)

**Figure 2:**

<table>
<thead>
<tr>
<th>Dry density [g/cm³]</th>
<th>Water content [%]</th>
<th>Water content [%]</th>
<th>Water content [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\rho_{b0} = 1.40$</td>
<td>$w_0 = 12$</td>
<td>$w_0 = 17$</td>
<td>$w_0 = 24$</td>
</tr>
<tr>
<td>$\rho_{b0} = 1.65$</td>
<td>$2/2$</td>
<td>$2/2$</td>
<td>$2/2$</td>
</tr>
<tr>
<td>$\rho_{b0} = 1.90$</td>
<td>$2/2$</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

**Table 2:** Number of experiments (completed/planned) for each case in constant volume experiments (duration of experiment: 4 days).
Jyväskylä research – Constant volume experiments

Table 2: Number of experiments (completed/planned) for each case in constant volume experiments (duration of experiment: 32 days).

<table>
<thead>
<tr>
<th>Dry density [g/cm³]</th>
<th>Water content [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>w₀ = 12</td>
<td>w₀ = 17</td>
</tr>
<tr>
<td>ρb₀ = 1.40</td>
<td>2/2</td>
</tr>
<tr>
<td>ρb₀ = 1.65</td>
<td>2/2</td>
</tr>
<tr>
<td>ρb₀ = 1.90</td>
<td>1/2</td>
</tr>
</tbody>
</table>

- X-ray imaging method was also used to measure water transport and deformation in MX80 bentonite samples kept in a constant volume (Fig. 3)
- At the present, 10 samples out of the planned 12 (Table 2) has been measured. The results are collected into the databank (temporary link: users.jyu.fi/~hpatana/bentonite.databank)
Jyväskylä research – Conclusions

• Non-invasive imaging methods, X-ray tomography and X-ray imaging appear to be useful tools for measuring hydro-mechanical properties of the bentonite buffer.

• Free swelling experiments are completed.

• Constant volume experiments will be completed within 3/2019.

• The results obtained in this project are expected to be useful in developing more reliable numerical models for hydro-mechanical behavior of bentonite.

• Results are available in databank (temporary link: users.jyu.fi/~hpatana/bentonite_databank)
VTT

(THEBES project)

Veli-Matti Pulkkanen
Hydromechanical experiments

Functioning new laboratory equipment and techniques to study bentonite mechanical behaviour

- Stress path control

Loadframe (250 kN) + high pressure triaxial cell (32 Mpa)

- Disposal conditions can be reached
- Silicon oil: high temperature is possible
- Hydraulic equipment: fast loading is possible
THEBES research: VTT

- Hydromechanical experiments
- Microstructural studies
- Chemical experiments
- Modelling
THEBES: KYT project investigating bentonite

Hydromechanical experiments

• Functioning new laboratory equipment and techniques to study bentonite mechanical behaviour
  • Stress path control

• Loadframe (250 kN) + high pressure triaxial cell (32 Mpa)
  • Disposal conditions can be reached
  • Silicon oil: high temperature is possible
  • Hydraulic equipment: fast loading is possible
Hydromechanical experiments

• Work will continue
  • KYT2022 project Broctio
    • Fast loading
    • Effect of chemistry
  • H2020 HITEC
    • Effect of temperature

Example results from uniaxial confined compression
Microstructural studies

• Focus on Atomic Force Microscope (AFM) studies of bentonite platelets
  • Size and shape distributions
  • Cooperation with KYT2018 Geobiokierto project
• PhD Thesis by M Matusewicz
  • ”The microstructure of bentonite clay”
  • Dissertation held in Dec 2018
Chemical experiments

• In situ ion selective electrode (ISE) development
  • Modification of commercial ISEs
  • Preparation of in-house made ISEs
  • pH, Cl, Na, Ca sensitive electrodes

• Experiments
  • Online ISE measurements
  • Standard chemical analysis of external solutions and post mortem samples

Example results: pH at different depths in bentonite and in the external solution of the diffusion cell sample. The solution chemistry was changed at 271 days from low-pH to high-pH.
Modelling

• D.Sc. (Tech.) thesis by V-M Pulkkanen:
  • “A large deformation model for chemoelastic porous media – bentonite clay in spent nuclear disposal”
  • Pre-examined

Example results from simulations: Water content profiles of bentonite wetting by water vapour (from left)

• solid lines – model in the thesis
• dashed lines – non-coupled model
• dots – experimental results by K-P Kröh
University of Eastern Finland

Tapani Pakkanen
Water retention curve including microstructure effect:
Considered structural levels
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.
- The method predicts swelling pressure trends of montmorillonite-beidellite smectites with respect to variations of layer charges, charge locations, and interlayer cations.
- Swelling pressures have been investigated 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.
Conclusions and Acknowledgements

- High or low layer charge and high charge fraction on tetrahedral sheets reduces the swelling pressure
- Iron substitution decreases clay swelling pressures
- Interlayer cation species strongly influence the swelling characteristics (Na\(^+\) / Ca\(^{2+}\))
- Elevated temperature increases clay swelling pressures
- Increased salinity of surrounding water reduces swelling pressure
- New MD model for swelling pressure simulation and clay material selection has been demonstrated
University of Helsinki

Pirkko Hölttä
Bentonite erosion and radionuclide interaction processes (BENTO)

Pirkko Hölttä, O. Elo, V. Suorsa, E. Honkanämi, N. Pakkanen, E. Puhakka
University of Helsinki, Department of Chemistry, Radiochemistry

Bentonite erosion resulting in the mass loss by 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.

- MX-80 bentonite (76 % montmorillonite),
  Nanocor PGN Montmorillonite (98 % montmorillonite)
- Bentonite erosion, colloid formation and stability
- Radionuclide sorption on bentonite/montmorillonite suspension and colloids
- Colloid-mediated radionuclide transport
Bentonite erosion and colloid stability

- Batch type experiments → Particle size distribution, concentration and zeta potential
- Methods: Photon correlation spectroscopy (PCS), ICP-MS, XRD and SAXS
- The bentonite erosion and colloid stability depended strongly on the ionic strength and the valence of the cations. In dilute solutions (1–10 mM), particle diameter was under 500 nm and ZP lower than -30 mV, indicating stable colloids.
- High pH (cement water) increased bentonite erosion and colloid formation.
- The montmorillonite content affected erosion, Nanocor PGN montmorillonite (98% mmt) eroded more than MX 80 bentonite (76% mmt).

Nanocor PGN erosion in different ionic strength reference ground water and electrolyte. From left: Allard (4.2 mM), Olso (0.52 M), Olso (5 mM), Olso (1 mM), NaCl (1 mM) and CaCl₂ (1 mM).

MX-80 bentonite and Nanocor PGN montmorillonite erosion experiment in cement and Allard water
Radionuclide sorption onto bentonite

- pH has a great influence on the chemical form of the radionuclides and thus on the sorption.
- Ionic strength decreased the sorption due to the increased specific surface area of the aggregated particles.
- Molecular modelling clarified the sorption mechanism of Cs\(^+\) and Sr\(^{2+}\) onto basal sites and Eu\(^{3+}\) on the edge sites of montmorillonite.
- Nanocor PGN montmorillonite adsorbed more strongly radionuclides than MX-80 bentonite.

Cs-134 sorption on MX-80 bentonite (76 % mmt) and Nanocor PGN montmorillonite (98 % mmt) suspension and colloids in Allard water.

Eu-152 sorption on Nanocor PGN montmorillonite (98 % mmt) colloids in Allard water.

Left: Cs\(^+\) and Sr\(^{2+}\) ion sorption onto the basal sites of the montmorillonite tetrahedral sheet.
Right: Eu(H\(_2\)O)_9\(^{3+}\) onto the edge site of the montmorillonite octahedral sheet.
Block-scale experiment on bentonite colloid – radionuclide interaction

- Kuru grey granite block: natural fracture: 0.9 x 0.9 m
- Intermediate between conventional laboratory and in-situ experiments (Grimsel Colloid formation and migration, CFM)
- $^3$H, $^{36}$Cl and Amino-G, $^{152}$Eu, $^{85}$Sr with and without colloids
- Colloid analysis by Laser Induced Breakdown Detection (KIT/ENE, Germany)
- Supportive batch sorption on colloids and crushed granite

- A good reproducibility in breakthrough curves obtained with several tests confirm the block set-up is suitable for the further experiment
- No breakthrough of $^{152}$Eu with colloids during the experiment

Valtteri Suorsa: Master thesis 2017
Influence of colloids on Np(V) transport

- MX-80 bentonite colloids, Kuru grey granite
- Batch sorption/desorption experiments (pH, isotherms, solid/liquid, kinetics)
- In-situ ATR FT-IR and EXAFS in HZDR, Germany
  - chemical nature of the complex between Np and montmorillonite or colloid
  - confirmation the inner-sphere complex and possible existence of the outer-sphere complex
- Drill core and crushed rock column experiments showed the enhanced effect of colloids on the transport of Np.

Breakthrough curves for Np(V) in the absence (red) and presence of colloids (blue). Transport was modeled using the analytical solution of advection–matrix diffusion equation Voutilainen and Kekäläinen).
Conclusions

- New experimental data on bentonite erosion and sorption of radionuclides.
- Different experimental arrangements and advanced analytical methods were applied.
- Salinity of water is the most important factor for the bentonite erosion and the stability of colloids.
- Spectroscopic methods and molecular modelling provided important information on the sorption mechanisms of radionuclides.
- Novel data on Np-237 (NpO$_{2}^{+}$), which will be a major dose contributor after 100 000 years in the SNF repository.
- Column experiments demonstrated the enhanced effect of colloids on the transport of radionuclides.


O. Elo, M. Voutilainen, N. Huittinen and P. Hölttä, Np(V) transport in granitic rock: The influence of bentonite colloids in the laboratory scale. Resubmitted to Applied Geochemistry.
Thank you!

Questions?