EUROPEAN COMMISSION

Community research

Long-term Performance of Engineered Barrier Systems

PEBS

Long-term Performance of Engineered Barrier Systems (PEBS)

Grant Agreement Number 249681

Documents of 3rd Workshop

(DELIVERABLE-N°: D5-13)

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Dissemination Level		
PU	Public	PU
RE	Restricted to a group specified by the partners of the [acronym] project	
CO	Confidential, only for partners of the [acronym] project	

EURATOM

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Workshop objectives

The Second Regulatory Workshop was arranged to present and discuss the latest results of the PEBS project with representatives of European Regulatory Authorities. The Workshop was a follow-up meeting of the First Regulatory Workshop in April 2012 (see Deliverable D5-5).

The focus of this Workshop was on the analysis of potential impacts of the research results on long-term safety functions and implications for repository design. The research work within the PEBS project is providing valuable results that lead to a deeper understanding of the barrier performance. The results presented at the workshop included:

- Results of the PEBS laboratory and in-situ experiments,
- Results of numerical modelling of the thermo-hydro-mechanical and chemical (THM-C) processes in the engineered barrier systems (EBS),
- Identification of the significant processes during the early evolution of the EBS,
- Development of a process-related description of the early post-closure stage of the repository and the residual uncertainties in the evolution,
- Extrapolation of results over the transient phase to the long-term behaviour and associated uncertainties,
- Possible impacts of the extrapolated behaviour and associated uncertainties on the long-term safety functions.

The major areas of uncertainty regarding the EBS performance were captured in four "cases":

1. Water uptake in the bentonite buffer (T<100°C),
2. EBS performance at temperatures exceeding 100°C,
3. HM evolution of the buffer,
4. Impact of the geochemical evolution of bentonite barriers on repository safety functions.

Cases 1, 2, and 4 were presented and discussed at the Second Regulatory Workshop; the corresponding presentations can be found in this deliverable.

Workshop agenda

The Second Regulatory Workshop was held together with the Annual Project Meeting in Zurich, Switzerland on December 3-5, 2013. The agenda of the meeting can be found on pages 3-5.

On December 5, 2013, the day after the workshop a visit to the Grimsel Test Site was arranged by Nagra. The Grimsel Test Site located in the Swiss Alps was established in 1984 as a centre for underground Research and Development supporting a wide range of research projects on the geological disposal of radioactive waste.

Long-term Performance of Engineered Barrier Systems
PEBS

Agenda

Yearly Project Meeting & Regulatory Workshop

December 3-5, 2013 – Zurich, Switzerland

December 3, 2013 (Tuesday)

11:00	Welcome of guests *Overview, background*	BGR / Nagra
	Work Package 2	
11:20	*WP 2 overview*	ENRESA
11:30	*WP 2 Technical Presentation (Task 2.1, 2.2)*	CIEMAT
12:30	Lunch break	
	Work Package 2	
13:30	*WP 2 Technical Presentation (Task 2.2.1)*	CLAY TECH
13:40	*WP 2 Technical Presentation (Task 2.2.2)*	NAGRA, BGR
14:05	*WP 2 Technical Presentation (Task 2.3)*	CIEMAT, UAM
14:30	**Work Package 3**	
	Overview of Work Package 3 organization	GRS
	Status of work and planning (Tasks 3.1, 3.2, 3.3, 3.5)	CIMNE
15:15	Coffee break	
15:45 – 17:00	*Status of work and planning (Tasks 3.1, 3.2, 3.3, 3.5)*	CIMNE
	Status of work and planning (Tasks 3.2, 3.3, 3.5)	Nagra / TK Consult

19:00	Dinner hosted by Nagra	

December 4, 2013 (Wednesday)

09:00	Welcome of guests *(to be confirmed)* *Overview, background*	BGR / Nagra
09:20	**Work Package 3**	
	Status of work and planning (Tasks 3.2, 3.5)	GRS
	Status of work and planning (Task 3.3)	SKB / Clay Technology
10:30	Coffee break	
11:00	**Work Package 3**	
	Status of work and planning (Tasks 3.4, 3.5)	UDC
11:30	**Work Package 4** *- Approach taken in WP4* *- Contents and planning of the WP4 report*	Nagra
12:30	Lunch	
13:30	**Work Package B** – Technical presentations *China mock-up experiment*	BRIUG
14:15	Discussion with representatives of the regulatory authorities	
	Case 1 – Water uptake in bentonite buffer	ENRESA *(to be confirmed)*
	Case 2 – EBS performance at temperatures above 100°C	NAGRA *(to be confirmed)*
15:15	Coffee break	
15:45 – 17:00	Discussion with representatives of the regulatory authorities	
	Case 4 – Impact of the geochemical evolution of bentonite barriers on repository safety functions	GRS *(to be confirmed)*

December 5, 2013 (Thursday)

07:00	Departure from the hotel
	Visit of Grimsel Test Site *The Grimsel Test Site located in the Swiss Alps was established in 1984 as a centre for underground Research and Development supporting a wide range of research projects on the geological disposal of radioactive waste.*
16:00 (approx.)	Return to Zurich

PEBS Long-term Performance of Engineered Barrier Systems

Project Meeting & Regulatory Workshop

December 3-5, 2013 – Zurich, Switzerland

Introduction

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December 03 – 05, 2013 PEBS Project Meeting – Zurich

BGR Bundesanstalt für Geowissenschaften und Rohstoffe GEOZENTRUM HANNOVER

PEBS Long-term Performance of Engineered Barrier Systems

Agenda - Tuesday

11:20	Work Package 2
12:30	Lunch break
13:30	Work Package 2
14:30	Work Package 3
15:15	Coffee break
-17:00	Work Package 3
19:00	Dinner hosted by Nagra

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December 03 – 05, 2013 PEBS Project Meeting – Zurich

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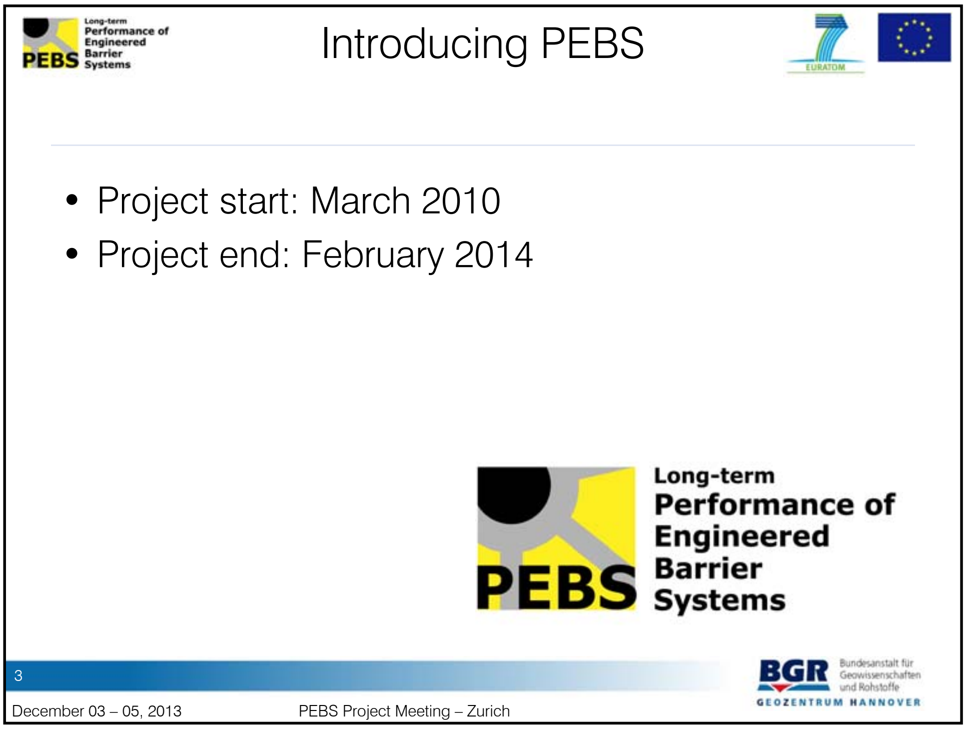
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• Project end: February 2014
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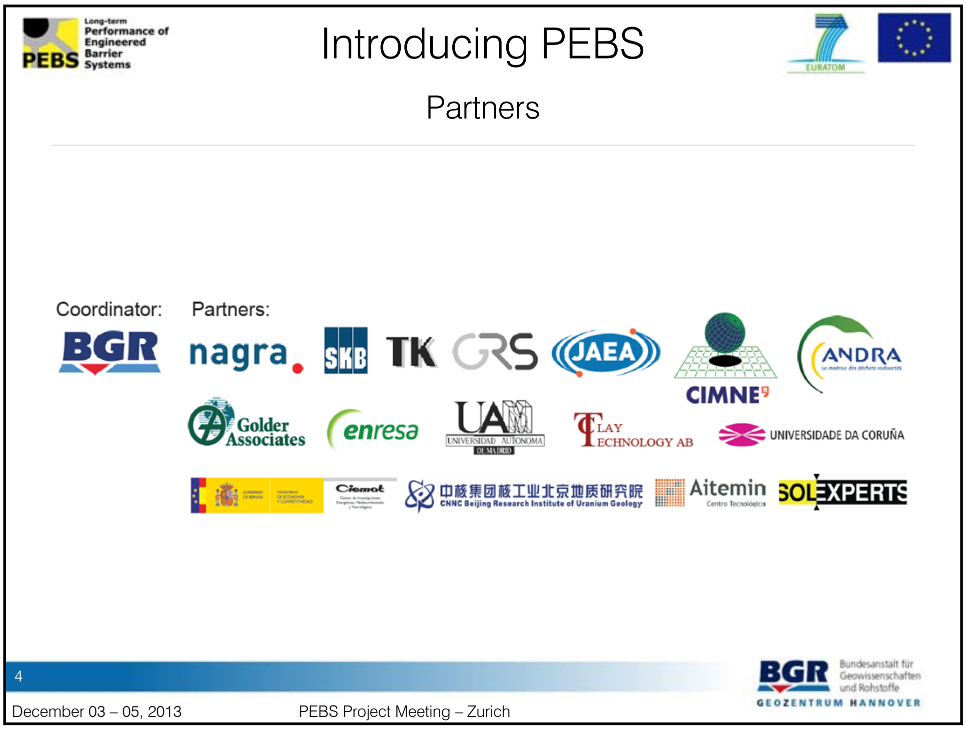
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CLAY TECHNOLOGY AB
UNIVERSIDADE DA CORUÑA
Ciemat
中核集团核工业北京地质研究院
CNNC Beijing Research Institute of Uranium Geology
Aitemin
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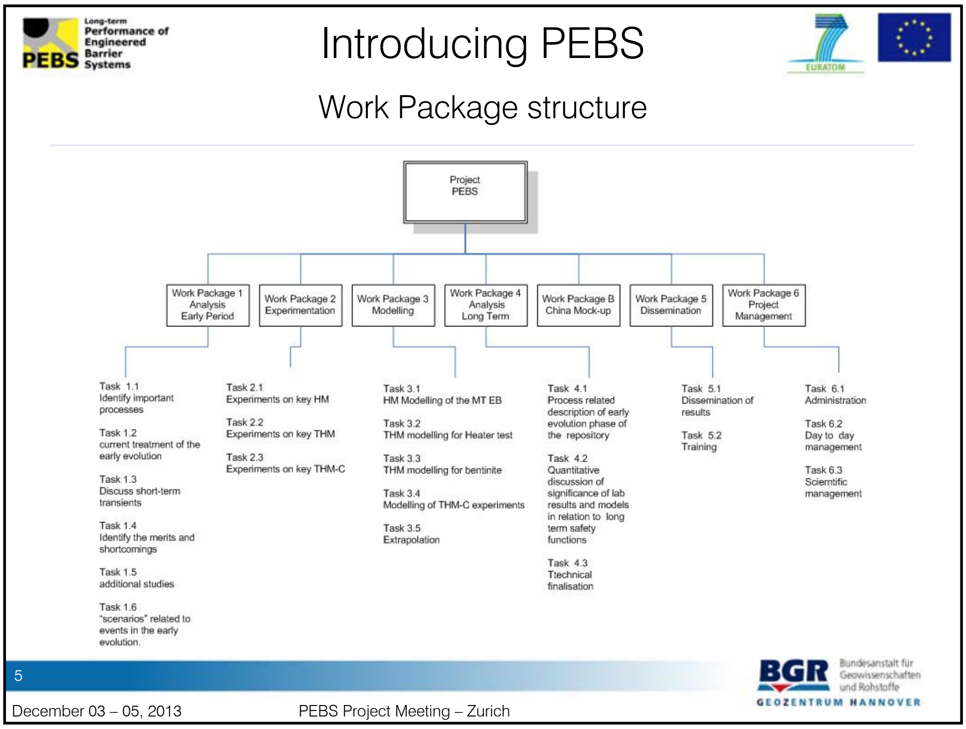
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Work Package 1 Analysis Early Period
Work Package 2 Experimentation
Work Package 3 Modelling
Work Package 4 Analysis Long Term
Work Package B China Mock-up
Work Package 5 Dissemination
Work Package 6 Project Management
Task 1.1 Identify important processes
Task 1.2 current treatment of the early evolution
Task 1.3 Discuss short-term transients
Task 1.4 Identify the merits and shortcomings
Task 1.5 additional studies
Task 1.6 "scenarios" related to events in the early evolution.
Task 2.1 Experiments on key HM
Task 2.2 Experiments on key THM
Task 2.3 Experiments on key THM-C
Task 3.1 HM Modelling of the MT EB
Task 3.2 THM modelling for Heater test
Task 3.3 THM modelling for bentinite
Task 3.4 Modelling of THM-C experiments
Task 3.5 Extrapolation
Task 4.1 Process related description of early evolution phase of the repository
Task 4.2 Quantitative discussion of significance of lab results and models in relation to long term safety functions
Task 4.3 Ttechnical finalisation
Task 5.1 Dissemination of results
Task 5.2 Training
Task 6.1 Administration
Task 6.2 Day to day management
Task 6.3 Scientific management
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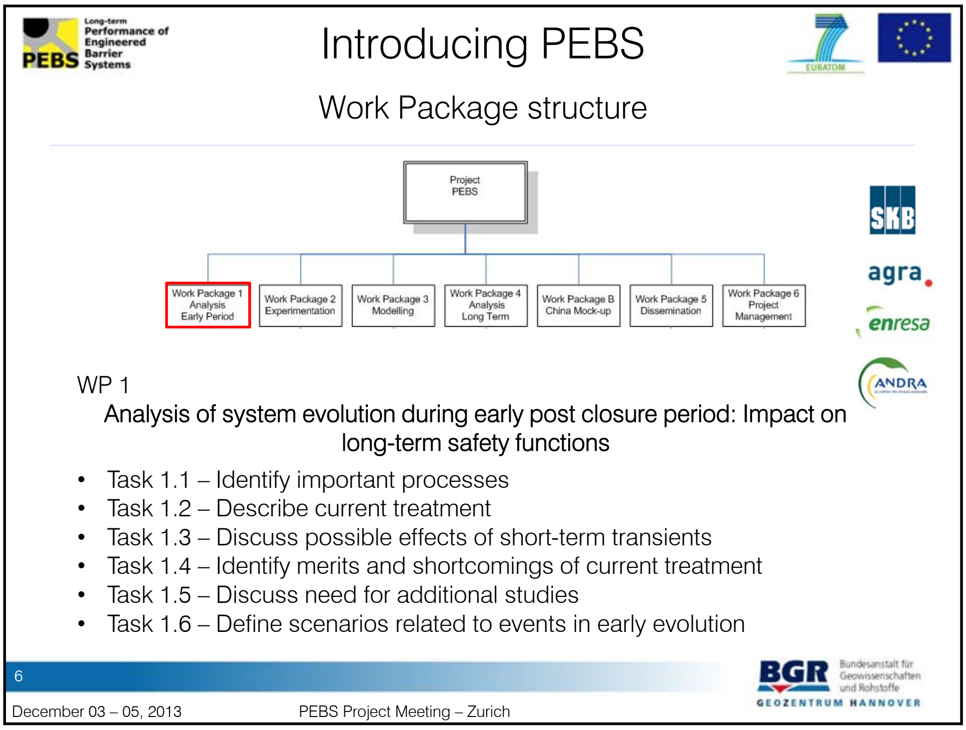
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Work Package 6 Project Management
SKB
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ANDRA
WP 1
Analysis of system evolution during early post closure period: Impact on long-term safety functions
• Task 1.1 – Identify important processes
• Task 1.2 – Describe current treatment
• Task 1.3 – Discuss possible effects of short-term transients
• Task 1.4 – Identify merits and shortcomings of current treatment
• Task 1.5 – Discuss need for additional studies
• Task 1.6 – Define scenarios related to events in early evolution
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Introducing PEBS

Work Package structure

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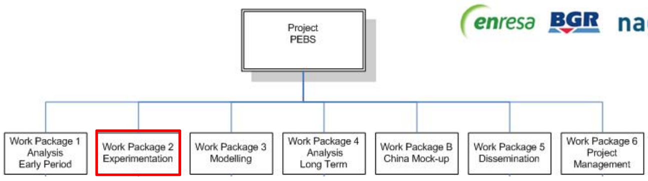

WP 2

Experimentation on key EBS processes and parameters

- Task 2.1 – Experimentation on key HM processes and parameters
- Task 2.2 – Experimentation on key THM processes and parameters
- Task 2.3 – Experimentation on key THM-C processes and parameters

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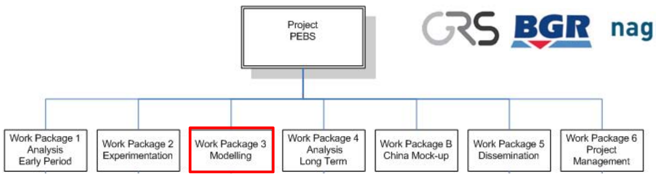

WP 3

Modelling of short-term effects and extrapolation to long-term evolution

- Task 3.1 – HM modelling of the Mont Terri Engineered Barrier (EB) Experiment
- Task 3.2 – THM modelling for the planned heater test HE-E
- Task 3.3 – THM modelling of bentonite buffer
- Task 3.4 – Modelling of THM-C experiments on bentonite buffer
- Task 3.5 – Extrapolation to repository long term evolution

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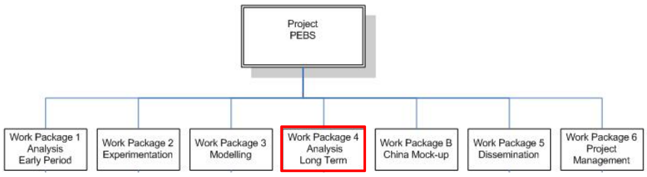

WP 4

Analysis of impact on long-term safety and guidance for repository design and construction

- Task 4.1 – Process-related description of the early evolution phase of the repository
- Task 4.2 – Quantitative discussion of significance of lab experiments and models in relation to long-term safety functions
- Task 4.3 – Technical finalisation

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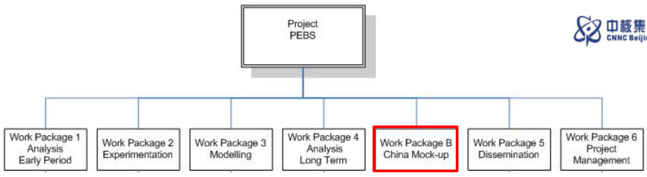

WP B

China-Mock-up Test on Compacted Bentonite-Buffer

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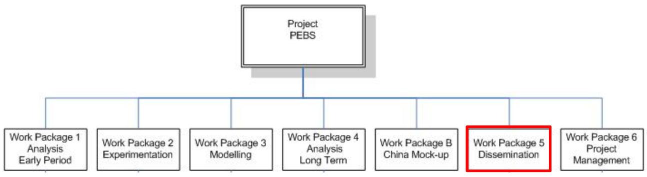


All partners

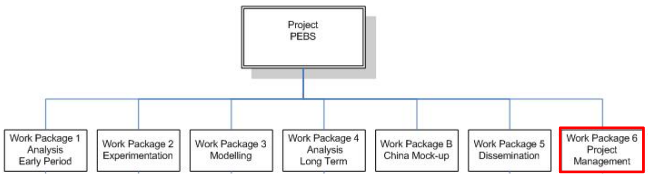
WP 5

Dissemination

- Task 5.1 – Dissemination of results
- Task 5.2 – Training

Introducing PEBS

Work Package structure



WP 6

Project Management

- Task 6.1 – Administration
- Task 6.2 – Day to day management
- Task 6.3 – Scientific management

Long-term Performance of Engineered Barrier Systems PEBS

International Conference on the Performance of Engineered Barriers

February 6-7, 2013 – Hannover

Preliminary programme - overview	
Day 1	Keynote
Day 1	PEBS – Case 1
Day 1	PEBS – Case 2
Day 1	PEBS – Case 3
Day 1	PEBS – Case 4
Day 1 & Day 2	Two parallel sessions & poster session
Day 2	Panel discussion

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Thank you!

The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement n° 249681.

Case 1: Water uptake of the buffer (T<100 ºC)

Juan Carlos Mayor, Enresa

Long-term Performance of Engineered Barrier Systems

4 December 2013 PEBS Workshop 1

Outline

- Introduction
- Process description
- Motivation of Case 1
- Uncertainty analysis
- Potential impact on the long term safety functions
- Conclusions

4 December 2013 PEBS Workshop 2

Long-term Performance of Engineered Barrier Systems PEBS

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Introduction

- Water uptake in clay components in the EBS is one of the key safety-relevant processes that have been extracted in the framework of the PEBS project from the integrated assessment of the processes for four different repository concepts (France, Sweden, Switzerland, Spain)
- These key processes are formulated such that they are overarching and thus safety-relevant for several disposal concepts

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Long-term Performance of Engineered Barrier Systems PEBS

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Long-term safety context

- The safety functions of a bentonite barrier are common to all systems
  - Low hydraulic conductivity and low diffusivity
  - Adequate swelling capacity and swelling pressure
  - Mechanical support
  - Resistance to mineral transformation
  - High radionuclide sorption
  - Minimization of microbial activity
- But the quantitative requirements and importance for long-term safety differs depending on the disposal system (granite or clay rock)

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Process description

- During the early stage of the repository evolution, the emplaced buffer takes up water from the surrounding bedrock and starts swelling.
- The swelling is restricted by the rock wall and a swelling pressure develops. The process depends on the properties of the buffer as well as on the local hydraulic conditions
- After final saturation, the hydraulic conductivity of the buffer will be very low and the swelling pressure will be high
- This process is common for all concepts with a bentonite buffer and is also relevant for bentonite seals. The timescale for the saturation process is however strongly dependent on the boundary conditions.

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Motivation

- Good agreement in THM modelling between models and data for different laboratory, mock up and in situ experiments (with high saturation rate/water supply)
- However, in a number of these experiments the progress of saturation at the later stages of hydration is lower than anticipated by the conventional coupled THM models

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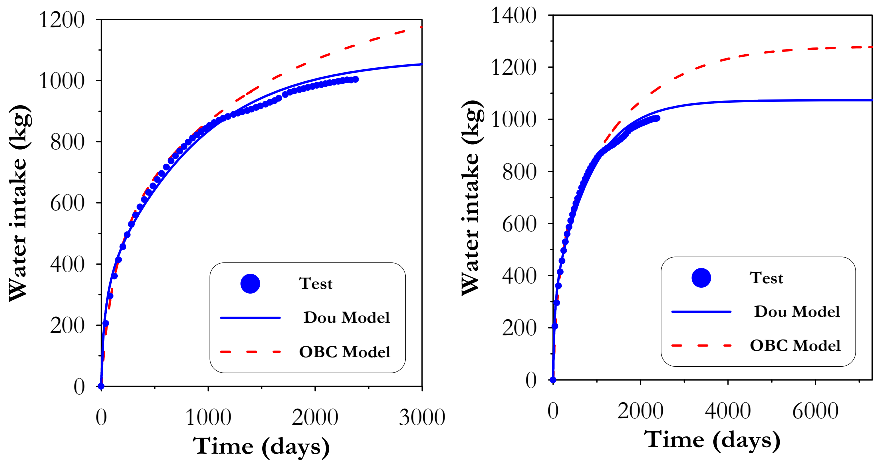
FEBEX mock-up: water inflow



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Uncertainty analysis (I)

- Three hypotheses have been examined to explain this discrepancy: existence of a threshold gradient in Darcy's law, thermo-osmosis phenomena, and evolution of bentonite microstructure during hydration
- Numerical modelling has shown that each of these possibilities are capable of providing results in agreement with observations but, on their own, they are unable to identify with certainty what is the phenomenon (or combination of phenomena) underlying the observed slowing down of hydration

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Uncertainty analysis (II)

- Although PEBS project is not designed to establish experimentally the potential existence and effects of those three phenomena, the performance of THM modelling of the FEBEX mock up test (16y of observations) gives some useful information on the effects likely to be associated with each one of the individual hypotheses
- In addition, performance of long-term THM computations for design repository conditions will provide estimations of the potential significance or otherwise of those phenomena for safety assessment.

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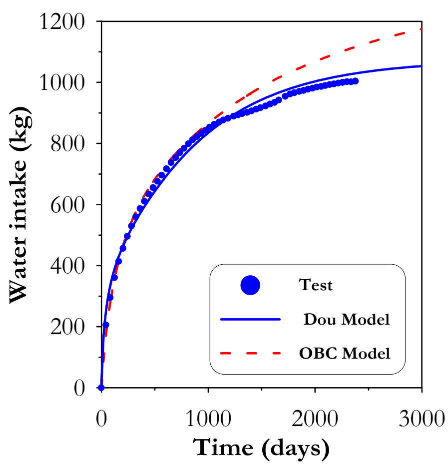
Potential impact on the long term safety functions(I)



The detected uncertainties (or model discrepancies) when present ("evidence is not clear cut") are noticeable at "advanced stages of the saturation process", according to the input (Ciemat, UPC) to the Questionnaire of the Pebs project

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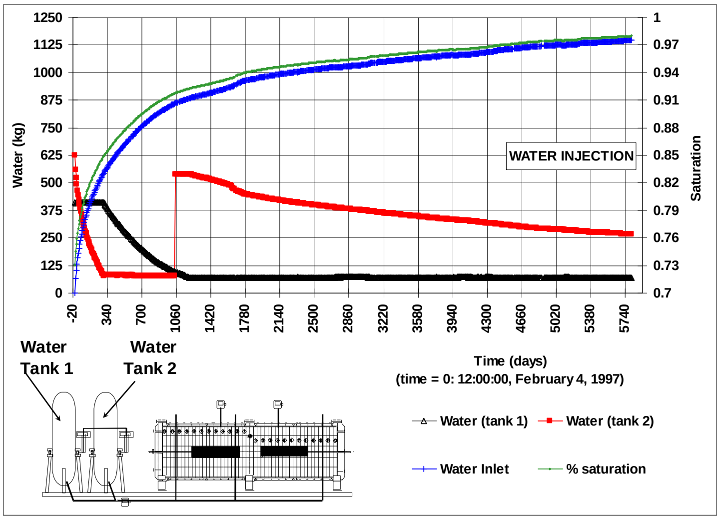
THM behaviour: water inlet



Injected water volume was 1147 L, on 31/12/12.
Saturation is higher than 97%.

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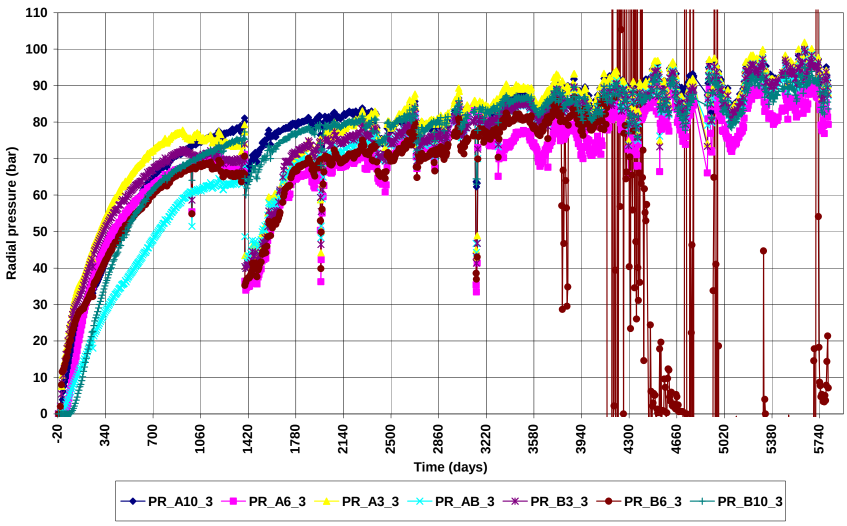
THM behaviour: radial pressures



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Potential impact on the long term safety functions(II)

Two different phases could be distinguished in the saturation process:

Phase 1: from the initial Sr up to about 90%

- Bentonite saturates fast (one thousand days in the Febex mock-up)
- Advective flow from the rock mass
- No model discrepancies

Phase 2: from Sr = 90% to full saturation

- Bentonite saturates more slowly (Sr is 98% after 5740 days in the mock-up)
- Advective flow from the rock mass ceases
- Model discrepancies do exist: the model underpredicts resaturation times

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Potential impact on the long term safety functions (III)

1. At the end of the Phase 1 (Sr=90%) the bentonite buffer will already fulfill its safety functions: K and Swp will fall within the PA acceptable limits
2. Hence the uncertainty identified in Phase 2 (longer saturation times) does not seem to be safety-relevant.
3. This residual uncertainty could be qualified as a “positive uncertainty”: it is likely that the advective flow conditions towards the buffer are kept longer than estimated by the models.
4. However, the evolution of the bentonite parameters (dry density, K, Swp) during a longer saturation period may be important (Case 3)

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CONCLUSIONS

- The context of the existing model uncertainty, from a long-term safety perspective, clearly improved – it can be stated that even though saturation is not yet fully achieved (e.g. after 16 years of hydration of the FEBEX mock-up), the safety functions assigned to the bentonite are achieved because a sufficient swelling pressure and low enough permeability are already reached throughout the barrier at 85-90% average degree of saturation

- The model uncertainty is thus not important from a long-term safety perspective (not safety-relevant)

"The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement n° 249681"

CASE 2: EBS performance at temperatures exceeding 100°C – general findings and application to Nagra's concept

Lawrence Johnson, Irina Gaus, Olivier Leupin
Nagra

Long-term Performance of Engineered Barrier Systems

Outline

- Generic assessment of bentonite barrier evolution vs. a specific case (Nagra's concept) – the importance of host rock and safety assessment context
- Summary of the findings (within PEBS and other projects) on the performance of a bentonite barrier for early evolution exceeding 100°C
- Impact of processes on Nagra safety functions
- Uncertainties and their significance to bentonite performance in the Nagra repository system context

The spectrum of conditions for repositories

- Bentonite buffer considered under a broad range of conditions
  - Pellets and blocks
    - Large range of initial water contents (5-20%), thermal conductivities (0.3-0.9W/mK)
    - Initial host rock temperature
      - Fennoscandian Shield at 400-500 m: 12-15°C
      - Opalinus Clay at 600 m: ~40°C
    - Various bentonites
  - Large range of water inflow rates (resaturation times of 10 to 1000 years)
- A broad envelope of THMC evolution results

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The spectrum of long-term safety requirements

- Common to all systems
  - Low HC and low diffusivity
  - Adequate swelling capacity and swelling pressure
  - Mechanical support
  - Resistance to mineral transformation
  - RN sorption
  - Minimization of MIC
- But…. the relative importance of buffer in safety assessment depends on the host rock
  - How much alteration can be accepted over how long a period and what types of alteration are relevant?

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THC Processes occurring in bentonite after emplacement and potential uncertainties

- Thermally induced drying near the canister and resaturation from the host rock
  - With bentonite pellets, very low moisture content for 10-20 years in Opalinus Clay – above 100°C no corrosion
- Cementation in the porespace due to dissolution/precipitation of minerals
- Transformation of montmorillonite to illite
- Transformation of montmorillonite to beidellite, saponite and/or chlorite
- Redox evolution and Fe-bentonite interaction

Assessment of the processes and impacts is based on a broad range of experimental evidence

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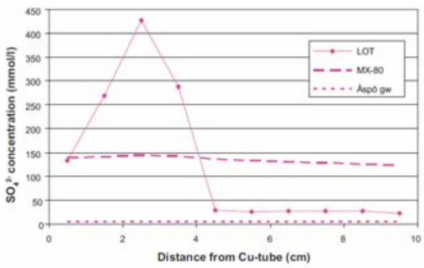
Cementation of the intergranular porespace by precipitation of minerals – evidence from the ABM and LOT experiment

- In the thermal gradient in the partially saturated nearfield it is expected that partly amorphous materials (e.g. SO_4, SiO_2, CO_3…) will precipitate as a function of their temperature dependent solubility. This has been shown to happen in field and lab scale experiments.
- This may locally reduce porosity (increase gas percolation threshold?).



Sulphate concentration in LOT samples as a function of the distance from the copper tube presented per the volume of the porewater (LOT) concentration, which sulphate of MX-80 would cause when evenly dissolved in the porewater (MX-80), and sulphate concentration in Äspö groundwater (Äspö gw). TR-09-29

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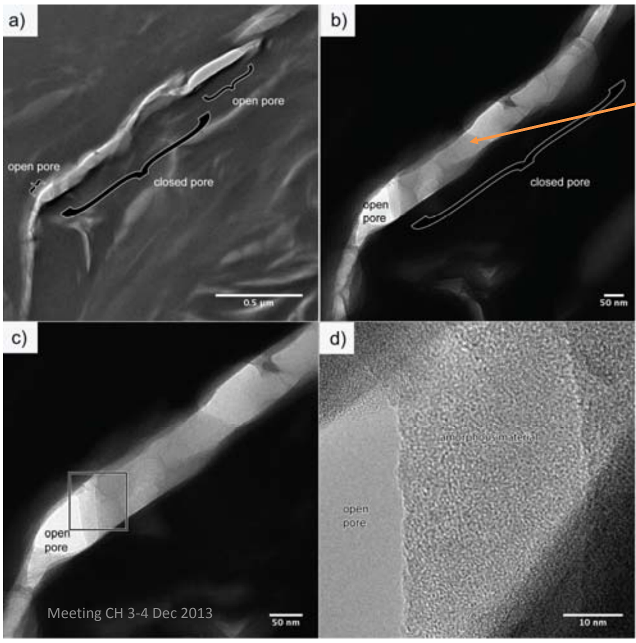
Cementation of the intergranular porespace by precipitation of minerals – evidence from the ABM and LOT experiment



Internal structure

A Redistribution of soluble minerals will be induced by the early phase temperature gradient
The pictures show bentonite MX-80 pellets near to the heater from the ABM experiment.

o *Lukas Keller, 2013; ZHAW*

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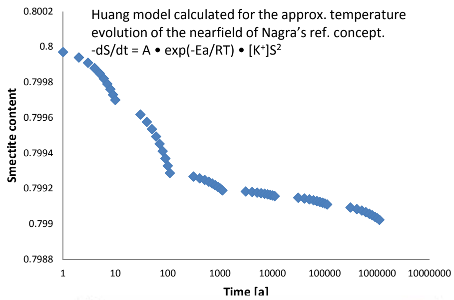
Transformation to illite

- Using models from sedimentary basins (Huang, 1993) transformation of montmorillonite to illite in the repository near field is shown to be a very slow process
- High temperatures and a high K activity are needed for the transformation of montmorillonite:
  Smectite + K^+ + H^+ ↔ Illite + SiO_2 + Mg^{2+} + Na^+ + H_2O
- So far no clear evidence has been found in field or lab scale experiments for any illitization process under repository conditions. However, loss of silica, slightly higher CEC and Mg on the exchanger measured near to the heating sources (LOT and ABM) could potentially be precursor reactions to the illitization.



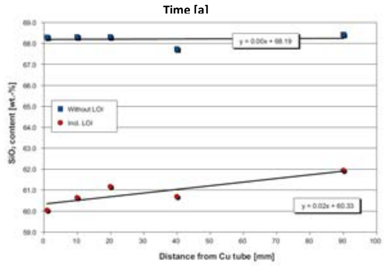


Figure A7-2. Comparison of SiO_2 content before and after correction of XRF data for the loss on ignition (LOI).

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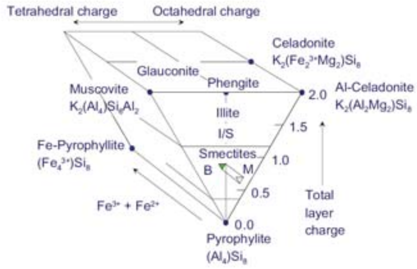
Transformation to beidellite, saponite and chlorite

- Due to initial high temperatures and high Fe-gradients in the nearfield transformation of smectite to other silicate minerals cannot be excluded.
- Potentially resulting sheet silicates might lead to a relatively low hydraulic conductivity but swelling pressure might be partially lost.
- The uncertainties related to the duration of partially saturated conditions and high temperatures prevailing in the near field make it difficult to predict at which rate smectite might be transforming.



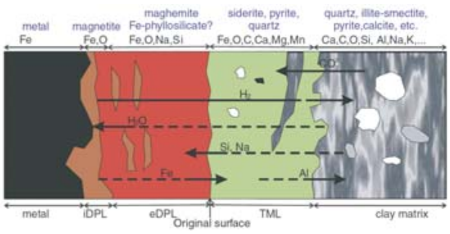
Composition of Na-montmorillonite of the Wyoming bentonite reference material indicated as a triangle in the Beidellite (B) – Montmorillonite (M) range. The basis is O20(OH)4 /Karnland and Birgersson 2006/, modified from /Newman and Brown 1987/. TR-09-29

Reduction of structural Fe and diffusion of Fe^{2+} into the bentonite

The interaction of corrosion-derived iron and bentonite influences corrosion rates and may influence the safety functions of the bentonite buffer. Such adverse effects include:

(1) local cementation of the bentonite via precipitation of Fe(II)/(III) oxides and hydroxides,
(2) destabilisation of the dioctahedral smectite structure (Lantenois et al. 2005),
(3) transformation of the montmorillonite to a non-swelling iron phyllosilicate, such as berthierine (Mosser-Ruck et al. 2010),
(4) dissolution of the montmorillonite via pH increase (Kumpulainen et al. 2010), or even
(5) direct interaction of H_2 with structural Fe in the smectite (Didier et al. 2012). The mechanisms leading to these adverse effects are still poorly understood. Moreover, their relevance for long-term safety is unclear.

ABM Block 7; picture by U.Mäder (NAB 11-19)



- Schlegel, M. L.; et al. Metal corrosion and argillite transformation at the water-saturated, high-temperature iron-clay interface:; Appl. Geochem. 2008, 23 (9), 2619-2633.

THM impacts at T> 100°C and their uncertainties

- THM processes are continuous (no sudden change in behaviour at 100°C).
- However, information on THM parameter characterisation and parameter dependencies at temperatures > 100°C is not extensive.
- At very high temperatures (>200°C), THM behaviour changes significantly (as a consequence of mineralogical changes, loss of swelling capacity) assuming presence of water
- Driving questions for THM laboratory and modelling activities are
  - Impact of hot vapour (steam) (lab) - cementation
  - Impacts on swelling pressure and hydraulic conductivity (lab)
  - Impact on shear strength and maximum strain (lab, URL)
  - Calibration of parameters at the URL scale (URL, modelling)
  - Pellets versus block bulk behaviour (lab, URL, modelling)

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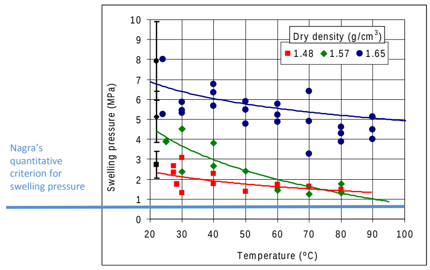
THM impacts at T up to 100°C – properties - lab tests

- Swelling capacity
  - Extensive testing of HM properties <100°C indicate a slight decrease in swelling capacity with T.
- Hydraulic Conductivity
  - Also a slight (?) increase is observed (partly attributed to the decrease in water viscosity).
- Retention curve
  - Modest changes for T<100°C (Romero et al., 2001, 2003; Rizzi et al., 2012)
- Extrapolation points at overall preservation of properties at least up to 100°C (Villar & Gomez-Espina, 2009; Villar et al., (2010))
- Observed differences from various experiments are not comprehensively described



Swelling pressure of FEBEX bentonite for different dry densities and temperatures (updated from {Villar, 2009 #715})

$$K_l(T)/K_l(T_0) = 1 + \beta_T(T - T_0)$$

Material	$\beta_T (K^{-1})$	Reference
Compacted Boom clay	0.010	Romero et al. (2001)
Natural Boom clay	0.030	Lima (2009)
Saturated kaolinite	0.042	Towhata et al., (1993)
Saturated kaolinite	0.010	Kemissa (1998)
Saturated montmorillonite	0.014	Volckaert et al. (1996)
Saturated bentonite	0.010	Cho et al. (1999)

Rate of variation of hydraulic conductivity with temperature. For the variation to be accounted for by viscosity variation only, the parameter b_T should be about 0.03.

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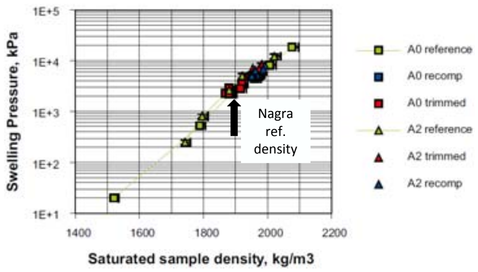
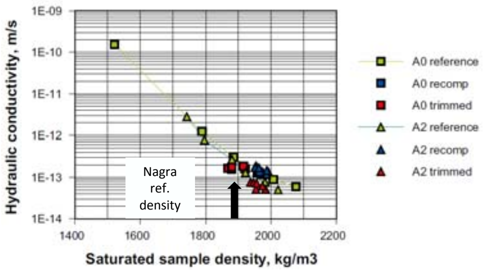
THM Impacts at T>100°C - cementation – decreasing swelling capacity (URL tests)





- Different lab and field-scale experiments have shown that swelling pressure and hydraulic conductivity are not altered by thermal treatment (~130°C) of compacted saturated bentonite

Max. T 130°C

Both figures are from the LOT experiment (TR-09-29)

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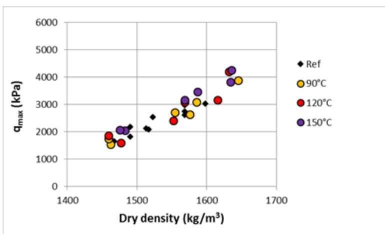
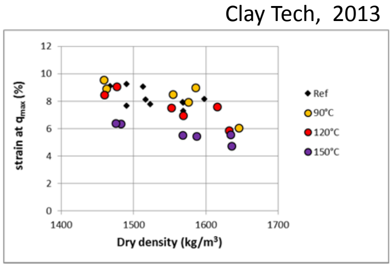
THM impacts at T> 100°C - Impact on shear strength and shear strain (lab)

- Small but significantly lower strain at failure was measured on resaturated material exposed to high temperature (>90°C). A change was seen already after a few hours of exposure
- No clear deviations regarding stress at failure (up to 150°C- PEBS lab tests), some at or above this temperature (field and lab test).

Clay Tech, 2013





Influence of heat and content of CaSO4	MX-80+CaSO4	150	unclear	unclear	G
Influence of heating after saturation	MX-80	90°	no	no	L
Influence of heating after saturation	MX-80	120°	no	-	L
Influence of heating after saturation	MX-80	150°	+	-	L
Deviation when heating before saturation	MX-80		no	unclear	K

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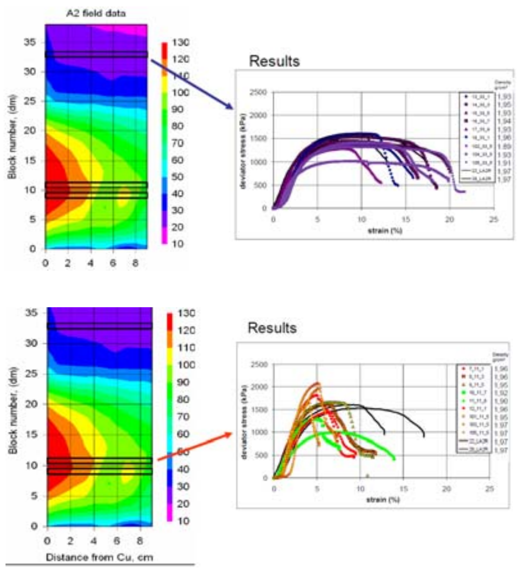
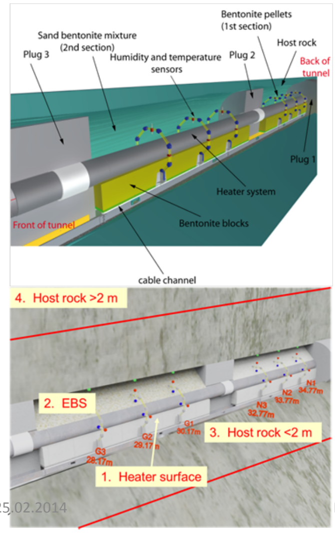
THM Impacts at T>100°C - Cementation - decreasing plasticity (URL)

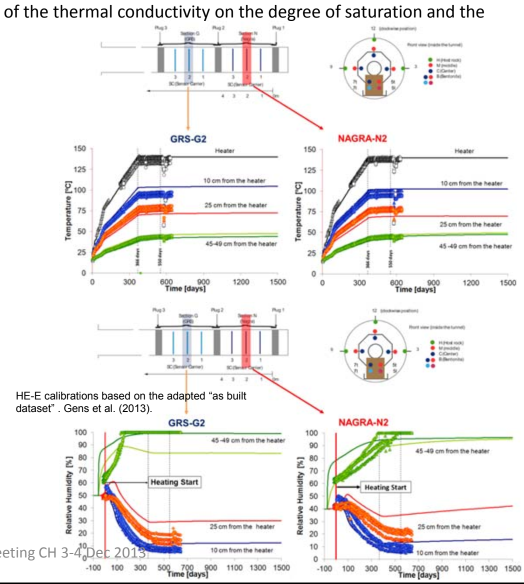


- The strain at failure is less (increased brittleness) for all the drilled specimens taken from the warm section of the field exposed material compared to the reference material taking the density into account.
- The specimens from the warm section of the field exposed material show a qualitatively different shear behaviour involving a more pronounced failure. In addition a nearly vertical failure surface is seen on some of these specimens after failure but not seen on specimens exposed to short time heating in the laboratory.
- The increased brittleness and pronounced failure seen on these drilled specimens from the warm section of the field exposed material is destroyed when the specimens are dried, milled and re-compacted.
- o *Ann Dueck, 2008; clay technology*

THM Impacts at T>100°C - large scale HE-E

- With the current TH(M) conceptualisation (TOUGH; Codebright) the very early dry phase can be reproduced in terms of temperature evolution
- Some uncertainty regarding the dependence of the thermal conductivity on the degree of saturation and the retention curve for the pellets remains





Insights from modelling

- Results from HE-E modelling regarding temperature, degree of saturation of bentonite, pore pressures in rock etc. should provide valuable insights regarding correct representation of the system
- Extrapolation over time should establish the bounds for system behaviour (envelope of T, resaturation vs. time)
- But input on time-dependent bentonite alteration will have to come from expert judgement based on results from laboratory programs

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Effects of partially saturated conditions at 100-150°C

- Couture (1985) reported substantial reduction in expandability upon exposure of bentonite powder to steam at 150 - 250°C
- Oscarson & Dixon (1990) showed that compacted bentonite heated at 90-150°C under unsaturated conditions did not show any significant change in mineralogy, P_s or k
- Pusch (2000) saw only slight changes in P_s and k of compacted bentonite that was allowed to expand on exposure to steam for T of 90 - 110°C
- Pusch et al. (2003) found dense bentonite pellet samples had some reduction of P_s at 125°C and a significant reduction at 150°C upon exposure to water vapour
- Valter and Plötze (2013) found at $T \geq 120$ °C with a degree of saturation $S_r < 0.25$ a distinctive influence on different properties of MX-80 sodium bentonite:
  - water uptake capacity under free swelling conditions decreased with increasing temperature over time
  - water vapor adsorption ability showed a significant drop at T=120 °C. The ability to recover by mechanical remolding was explained with cementing processes during the thermal treatment.
  - no significant mineralogical changes

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Overall findings and uncertainties (1)

- Results from several long-term heater-buffer experiments show minor thermal transient effects:
  - Decrease in plasticity of bentonite
  - Slight decrease in hydraulic conductivity
  - Slight decrease in swelling pressure
- Thermally-induced mineralogical transformation of bentonite likely to be very limited even over very long times
- Iron-bentonite impacts remain uncertain
  - Potential to locally reduce swelling and cause cementation, but this is a very slow process

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PEBS Long-term Performance of Engineered Barrier Systems

Overall findings and uncertainties (2)

- Some impact of vapor on bentonite properties under repository relevant conditions (120-150°C).
- A comprehensive view on the impact of T up to 150°C on swelling pressure and hydraulic conductivity (although not dramatic) is not yet in place
- The impact of chemistry induced by reactive gases in partially saturated bentonite under suction has not been been constrained (potentially relevant for long unsaturated time periods at elevated temperatures).
- The relation between thermal conductivity and degree of saturation needs further data support and upscaling, including its correlation with retention curves

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What is the significance of the uncertainties for the Nagra disposal system?

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Nagra's EBS[1] design principles

- Multi-barrier system
- Initial complete containment (> 1000 years)
- Attenuation of radionuclide release
- Simple materials with stability, longevity and good predictability
- Practical, reliable waste and buffer emplacement
- Robustness (insensitivity to disturbances and residual uncertainties)
- Redundancy
- Compartmentalisation concept

[1]Refers to complete NF engineered system

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Requirements for the buffer

Safety-relevant attributes	Favours/contributes to ...	Criteria Buffer / preferred values
Low hydraulic conductivity	Attenuation safety function of buffer, by ensuring diffusive transport	$K < 10^{-11}$ m s^{-1} for buffer around canister
Chemical retention of radionuclides	Attenuation safety function of buffer, by retarding transportfrom the buffer	No quantitative criterion , strong sorption is favored
Sufficient density	Attenuation safety function of buffer, by preventing colloid transport	$\rho_s > 1.650$ Mg m^{-3}
Sufficient swelling pressure	Attenuation safety function of rock, by providing mechanical stabilization of rooms, and hence avoiding significant extension of EDZ	0.2 MPa $< P_s <$ minimum principal stress
Mechanical support	Safety function of canister, by ensuring it is surrounded by a protective layer of buffer (stress buffering)	Buffer must be sufficiently viscous to avoid canister sinking
Sufficient gas transport capacity	Attenuation safety function of buffer, by ensuring gas can migrate without compromising hydraulic barrier	No quantitative criterion ; less than the minimum principal stress
Minimize microbial corrosion	Safety function of canister, by ensuring conditions favorable to slow corrosion	A dry density of > 1.45 Mg m^{-3} preferred to limit microbial activity
Resistance to mineral transformation	Longevity of other safety relevant attributes of buffer	No quantitative criterion
Stress buffering	Safety function of canister, by providing stress buffering	Beneficial, but not a required property
Suitable heat conduction	Safety function of canister, by ensuring favorable maximum temperature conditions	$0.4 < Tc < 2$ W m^{-1} K^{-1} (for a specific thermal heat load of 1500 W)

A dry density value that satisfies all criteria is 1.45 Mg m^{-3} ($k = 10^{-13}$ m s^{-1} and P_s = 3-4 MPa)

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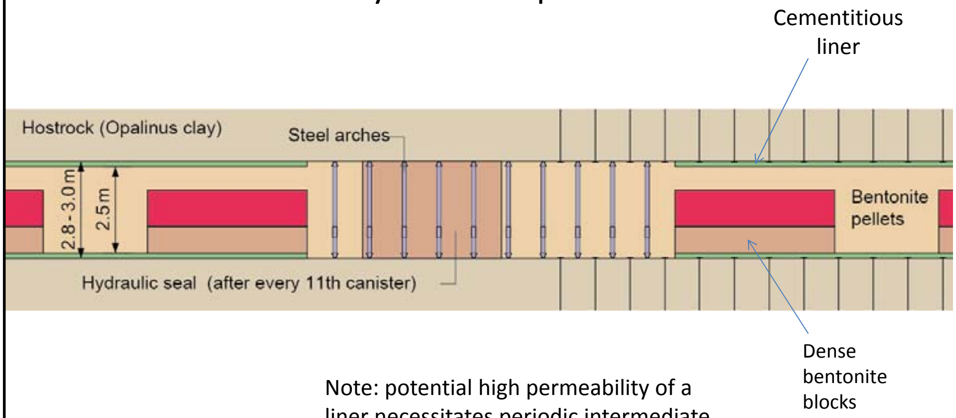
Nagra's waste emplacement tunnel layout concept



Note: potential high permeability of a liner necessitates periodic intermediate bentonite seals (but seals are relevant only if there are transmissive features in Opalinus Clay)

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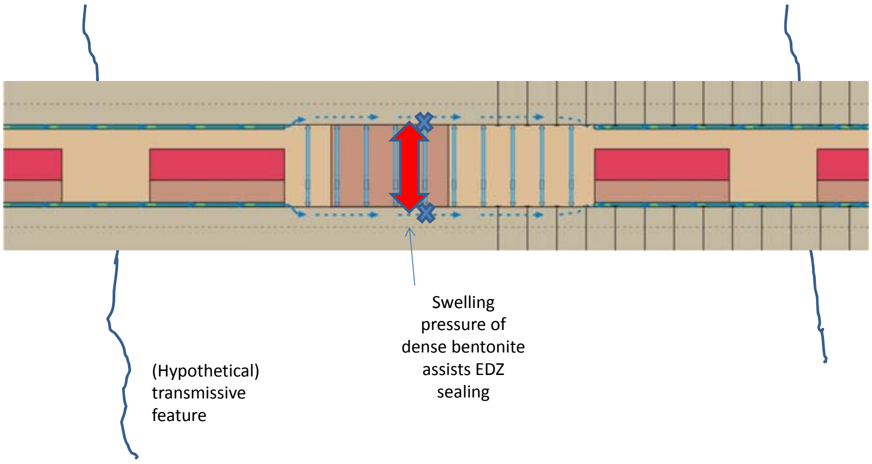
Function of intermediate seal



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Buffer performance in Nagra assessment cases

- Reference case – diffusion in rock
  - Radiological perspective: Requirements on buffer from mass transport perspective are minimal – if there is no near field retention and canisters have a short lifetime, the radiological consequences are unchanged relative to buffer and canister performing «ideally»
- Alternative assessment cases
  - Hydraulically transmissive features in Opalinus Clay intersecting emplacement tunnels (what-if? scenario)
    - Doses are well below dose limit but robustness can be enhanced with an intermediate seal
  - Borehole intrusion (human intrusion scenario)
    - Low hydraulic conductivity of buffer is beneficial

In all cases, good buffer properties are desirable (low hydraulic conductivity, high swelling pressure…..), but compartmentalisation concept with intermediate seal provides «back-up»

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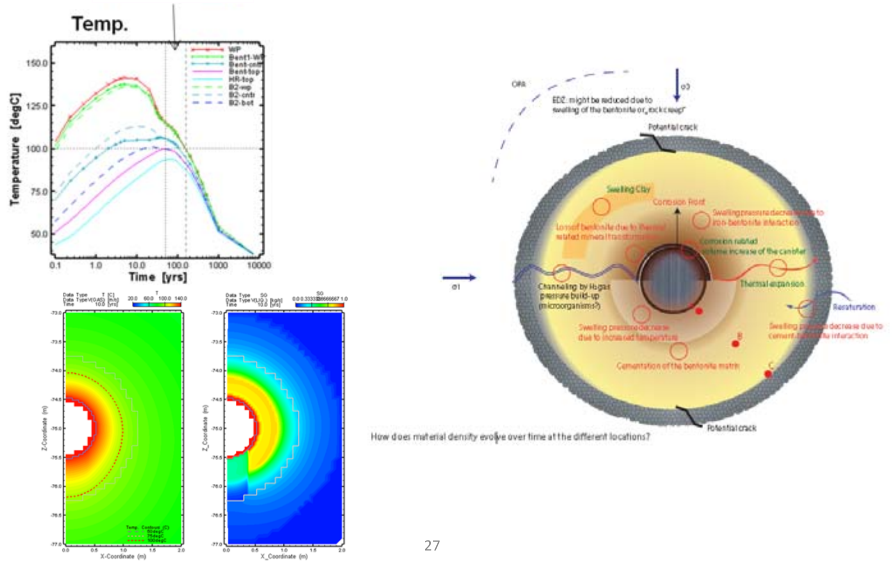
Long-term Performance of Engineered Barrier Systems PEBS

EURATOM

The impact of the uncertainties needs to be placed in the correct spatial and temporal context to assess the consequences



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Long-term Performance of Engineered Barrier Systems PEBS

EURATOM

Uncertainties and their significance

- The effect of high temperatures on the hydraulic properties and swelling pressure of the buffer
  - Context – reduction in swelling as a result of cementation localized to inner half of buffer (very little illitization)
  - Significance – low from RN transport perspective, because much of the buffer is unaffected and diffusion/sorption of radionuclides is almost unaffected; increase in strength of buffer due to local cementation
- The evolution of swelling pressure with time and the interaction with the convergence of the host rock
  - Context – rock stress exceeds swelling pressure of buffer, eventual convergence of rock and compaction if rock creep occurs – time uncertain
  - Significance – low from RN transport perspective

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Uncertainties and their significance

- Chemical interaction between bentonite and Fe (in case of steel canister)
  - Context – cm to tens of cm of bentonite altered to some degree (very slow process)
  - Significance – some loss of swelling capacity, increase in k
- Chemical interaction between bentonite and low pH concrete liner
  - Context – few cm of altered bentonite, plugging of pores, reduced k at interface
  - Significance – extended duration of resaturation

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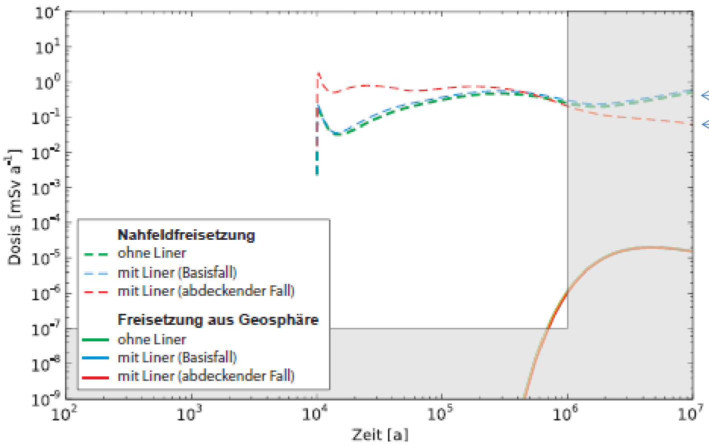
Effect of bentonite degradation on RN release (Nagra concept)



Release from near field

← Reference case

← Degraded bentonite

← Release from geosphere

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Long-term Performance of Engineered Barrier Systems

PEBS

EURATOM

"The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement n° 249681"

PEBS 5th Annual Project Meeting

Impact of the Geochemical Evolution of Bentonite Barriers on Repository Safety Functions - PEBS Case 4

J. Cuevas, J. Samper, M.J. Turrero, K. Wieczorek

Zurich, December 3-5, 2013

Long-term Performance of Engineered Barrier Systems

Fig. 1

Zurich, December 3-5, 2013 Case 4

Presentation Outline

- Introduction
- Case 4 topics
- Example of a question catalogue
- Discussion of the individual Case 4 topics
- Conclusions

Fig. 2

Zurich, December 3-5, 2013 Case 4

Case 4 Introduction

For the case investigations a formal approach with a questionnaire was chosen to facilitate integration of experimental and modelling results from PEBS as well as from earlier projects.

A set of topics was defined for each case, and specific questions were formulated for each topic. Answers were collected from all PEBS partners which could contribute.

Case 4 addresses the impact of the geochemical evolution of bentonite barriers on repository safety functions.

The most important safety function that could be impaired by the chemical effects is the required **limitation of advective flow**: Chemical effects should not inhibit the low permeability and sufficient swelling pressure developing and being maintained in the buffer in the long term.

Fig. 3

Zurich, December 3-5, 2013 Case 4

Case 4 Topics

- Chemical interaction of **corrosion products** with bentonite and the impact on bentonite properties
- Reaction products at the **canister/bentonite** and **cement/bentonite interfaces** and their implications about the geochemical reaction pathways
- **Bentonite alteration** at temperatures **above 100°C**
- **Thickness of the altered layers** at the canister/bentonite and concrete/bentonite interfaces and their properties
- Geochemical **modelling of reactions at the interfaces** in the EBS: Uncertainty in chemical parameters
- Uncertainty in the **bentonite hydration rate**
- The **time-scale of certain geochemical reactions** is too long for experimental verification – what is the impact if such reactions are neglected or simplified?

Fig. 4

Zurich, December 3-5, 2013 Case 4

Example: Question Catalogue for Topic 2

2. Understanding of the geochemical processes that might affect the EBS performance is essential. Reaction products arising at the
   (1) canister/bentonite interface and the
   (2) cement/bentonite interface
may contain information about the geochemical reaction pathways.

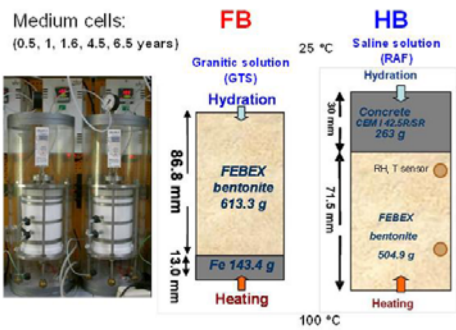
a) *Did you identify such reaction products in your experiments?*
b) *If yes, do the observed reaction products allow identification of the reaction pathways?*
c) *Can the reaction pathways be reproduced by adequate models?*
d) *Do you think that the observed reaction products are stable and the reaction pathways are valid for the medium term (decades to centuries) or the long term (thousands of years)? Or is there any evidence that the chemistry will further evolve and the observed products are only intermediate phases?*

Fig. 5

Zurich, December 3-5, 2013 Case 4

1. Corrosion Products

- Several lab tests were started by CIEMAT in NF-PRO and completed and dismantled in PEBS
- Reaction pathway for iron corrosion under unsaturated and saturated conditions
  → next slide



- No newly-formed iron-rich clay phases and no mineralogical alteration of bentonite
- Pitting occurred only under aerobic conditions

Fig. 6

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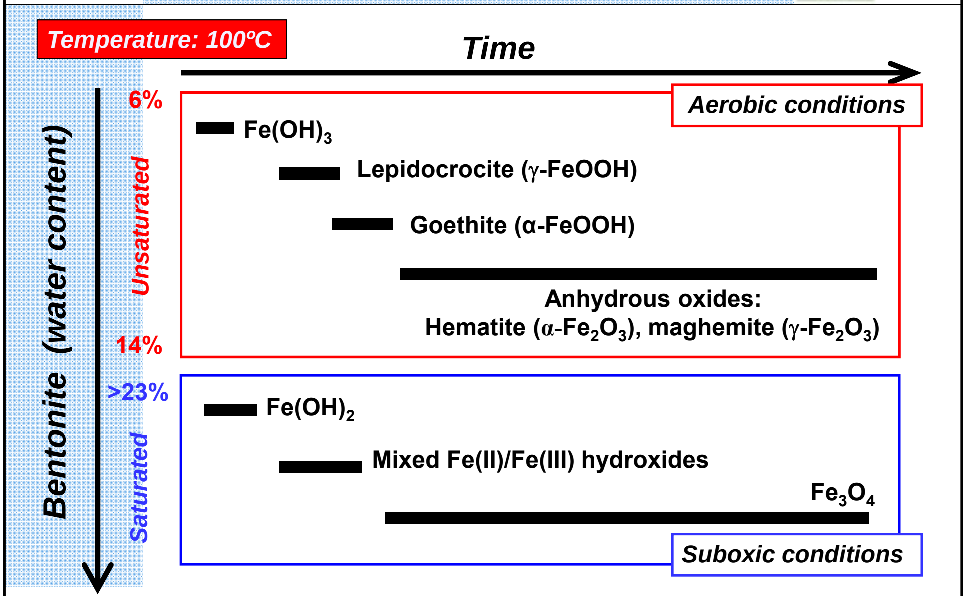
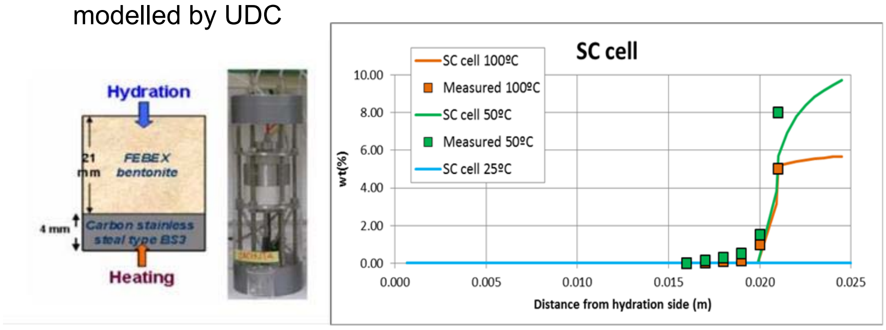
Observed reaction pathways



PEBS Workshop, Barcelona 26/09/13

2. Canister/Bentonite and Cement/Bentonite Interfaces (1)

- For the canister/bentonite interface, the reaction pathway can be identified from the reaction products
- For suboxic conditions which can be expected in the long term after bentonite saturation, respective lab experiments of CIEMAT were modelled by UDC



Modelling results for two corrosion tests in small cells
(Fe weight percent (wt%) versus distance from the hydration port)

Fig. 8

Zurich, December 3-5, 2013 Case 4

2. Canister/Bentonite and Cement/Bentonite Interfaces (2)

- For the bentonite/concrete interface, the reaction pathway is less precisely established (minerals form complex assemblages, very low crystal size) → next slide
- Hypotheses exist in how timescale and the progressive decrease in the high pH conditions will favour the formation of different minerals

  low Si/Al zeolites (i.e., Analcite, phillipsite)
  → CSH of decreasing Ca/Si ratios
  → stable calcite, feldspar, saponite mineral associations

- UDC models were used to simulate the lab experiments performed by CIEMAT and reproduced for the most part the observed mineralogical changes at the cement/bentonite interface

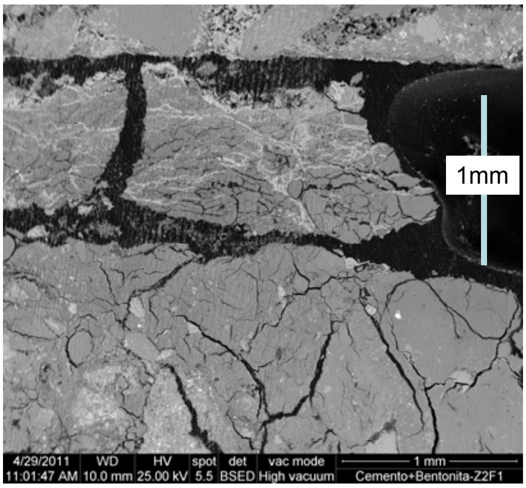
Fig. 9

Zurich, December 3-5, 2013 Case 4

Concrete/bentonite

Design
Corrosion
Concr./bent.
Water
Aqueous
React. products
Thickness/poros.
Chem. comp.
Exchang. Cat
Pathways
Uncertainties
Long-term
Models

Bentonite: calcium rich veinlets



Concrete

reaction products at the interface

Bentonite

PEBS Workshop, Barcelona 26/09/13

3. Bentonite Alteration, T > 100°C

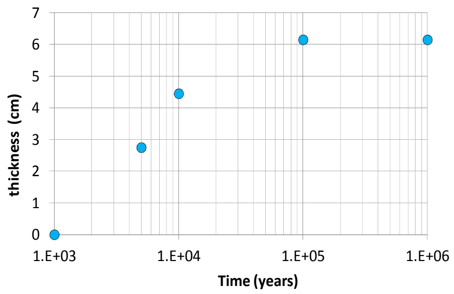
- At higher temperatures higher reaction rates and higher ordered crystals can be expected
- Major alteration processes, like cementation by SiO_2 precipitation and illitization, have been found to occur only at 150 °C and beyond (Wersin et al., 2007)

Fig. 11

Zurich, December 3-5, 2013 Case 4

4. Thickness of the Altered Layers (1)

- Regarding the canister/bentonite interface, corrosion products were found to penetrate a few millimetres into the bentonite in NF-PRO
- In the experiments performed in PEBS the affected thickness was even below 1 mm
- In UDC's model calculations for a repository in granite, the precipitation of corrosion products close to the canister led to a significant decrease of bentonite porosity - the affected bentonite thickness increased with time, reaching a thickness of 6 cm after 1 million years



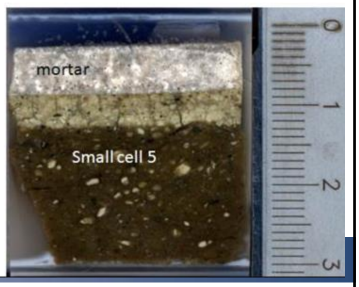
Predictions of the thickness of bentonite in which the decrease of the porosity is larger than 10% of its initial value (0.407)

Fig. 12

Zurich, December 3-5, 2013 Case 4

4. Thickness of the Altered Layers (2)

- At the bentonite/concrete interface, precipitation of carbonates, C(A)SH, MSH and other phases can result in pore clogging, which could affect the hydraulic parameters and the local stress state
- In controlled lime-mortar/bentonite interface experiments an altered layer of bentonite several millimetres thick (<5 mm) is formed (CASH)
- A second layer of several centimetres of thickness may be also affected by changes in the concentrations of exchanged cations, with an enrichment in exchanged calcium and a depletion of magnesium
- Coupled THCm numerical models capture the main trends in mineral dissolution-precipitation



Cemented rim (pale green colour) with low porosity/ low specific external surface of <5 mm thickness at lime mortar (grey-white colour)/bentonite interface. CIEMAT-UAM small cell PEBS experiments

Fig. 13

Zurich, December 3-5, 2013 Case 4

5. Uncertainty in chemical parameters

- Model results are mostly consistent with experimental observations
- Uncertainties with respect to long-term predictions / slow reactions which cannot be directly checked by experiments
- Major uncertainties include the nature of secondary minerals, like low-crystal-size C(A)SH and MSH at the bentonite/concrete interface (important for clogged low-porosity layers which may affect hydration rates / hydraulic behaviour) – and the related porosity / permeability
- Uncertainties regarding equilibrium constants, kinetic parameters (rate constants, reactive surface areas)
- Coupling to mechanics (stress influence), gas generation & transport

Fig. 14

Zurich, December 3-5, 2013 Case 4

6. Uncertainty in the Hydration Rate

- Geochemical effects influencing the hydration in the long term are not really expected (setting aside the influence of thin clogged layers at bentonite/concrete interfaces)
- In the other direction, the hydration rate is important for the geo-chemical evolution of the EBS, because partial saturation involves a gaseous phase with altered corrosion mechanisms - this is the more pronounced the longer the partial saturation phase is kept up

Fig. 15

Zurich, December 3-5, 2013 Case 4

7. Time-Scale of Certain Reactions

- The time-scale of certain geochemical reactions (e.g., smectite dissolution) is too long for experimental verification
- For short to medium term geochemical models these reactions are sometimes excluded
- At the pH conditions of a repository in granite (6.5<pH<9) the smectite dissolution is deemed to be not relevant even for the long term
- In a repository in clay this process may be more relevant for the bentonite subjected to a high-pH plume in contact with concrete

Fig. 16

Zurich, December 3-5, 2013 Case 4

Conclusions (1)

- Thermally-induced mineralogical changes such as the cementation by silica precipitation and the illitization will be relevant mostly above 150 °C
- Interactions of corrosion products and bentonite:
  - The main properties of the bentonite remain unaltered
  - There is a sequence of corrosion products, $Fe(OH)_2(s)$ and magnetite being the end members
  - Iron is sorbed by surface complexation while iron exchange is less relevant than iron sorption
  - Corrosion products penetrate a few mm into the bentonite
  - The coupled THC numerical models mostly reproduce the experimental data

Fig. 17

Zurich, December 3-5, 2013 Case 4

Conclusions (2)

- Interactions of bentonite and concrete:
  - An altered layer of bentonite several millimetres thick (<5 mm) is cemented by the precipitation of new minerals in the pore space
  - A second layer of several centimetres of thickness may be also affected by changes in the concentrations of exchanged cations
  - Coupled THCm numerical models capture the main trends in mineral dissolution-precipitation
  - While there are still open questions regarding the conceptual geochemical model, the pore clogging processes, and the final parameters and properties of the altered zone, current models indicate that the thickness of the altered bentonite will be bounded

Fig. 18

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PEBS Long-term Performance of Engineered Barrier Systems

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Fig. 19

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Fig. 20

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