sck cen
Belgian Nuclear Research Centre

eurad
European Joint Programme on Radioactive Waste Management

Diederik Jacques - 25/10/2024

EURAD WP ACED

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Assessment of the Chemical Evolution at the Disposal Cell Scale

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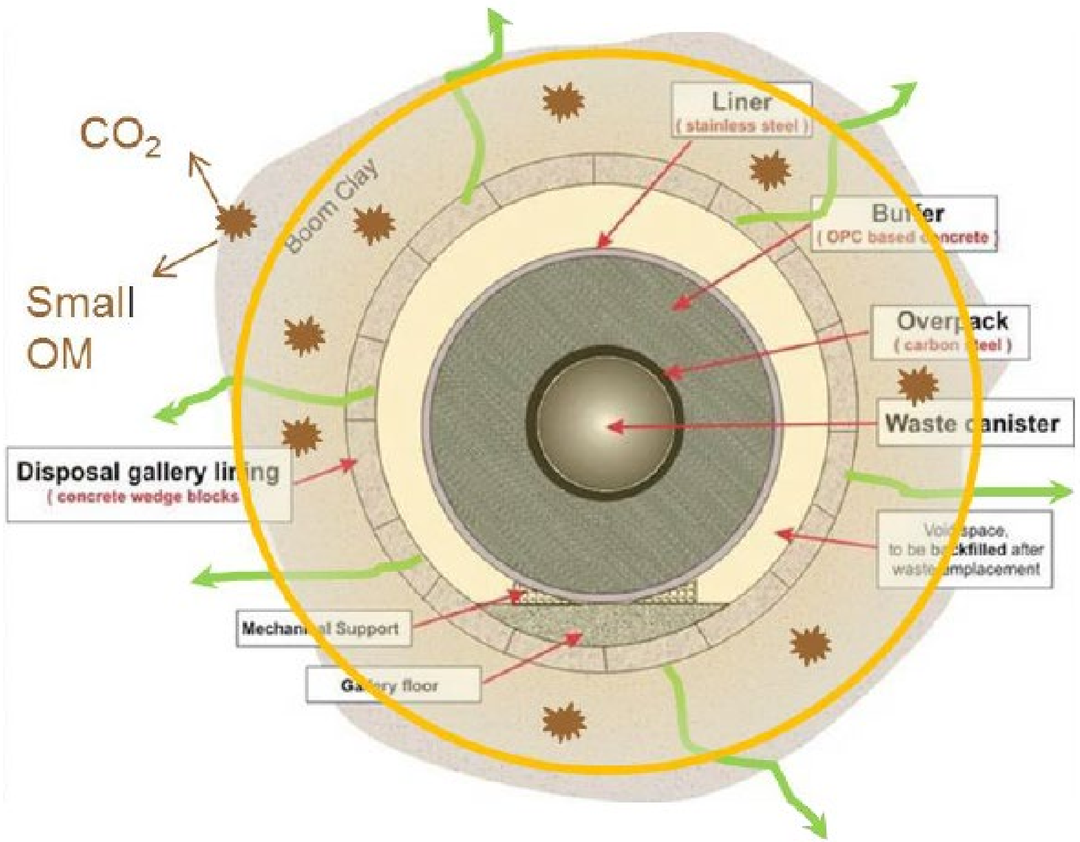
WP in a glance



- **DISPOSAL CELL**.
  - Intermediate level waste package
  - High level waste package
  - Engineered barrier system
  - Immediate surrounding host rock (clay, crystaline)
- **CHEMICAL EVOLUTION**
  - Different materials (glass, steel, different types of cement-based materials, host rocks)
  - Chemical gradients – Transport
  - Disequilibrium – Geochemical reactions
- **ASSESSMENT**
  - Large structures
  - Long times

WP in a glance

Long term chemical evolution forms an important input for the assessment of the evolution of a disposal system and the assessment of safety- and performance-related aspects

AIM – A better conceptual and mathematical representation of the chemical evolution to:

- Improve the assessment and quantification of generic safety functions such as isolation and containment of waste constituents
    - Input to (or coupling with) other processes M - B
- Obtain a better substantiation of conservatism and reduction of uncertainty
- Increase the scientific basis for definition of requirements of materials

Consortium

ANDRA (FR)	MTA-EK (HU)
Bel-V (BE)	PSI (CH)
BRGM (FR)	SCK•CEN (BE)
CEA (FR)	SUBATECH (FR)
CIEMAT (ES)	SURAO (CZ)
CNRS/GeoRess (FR)	UAM (ES)
COVRA (NL)	UDC (ES)
Ecole des Mines (FR)	UFZ (DE)
EDF (FR)	UJV (CZ)
FZJ (DE)	UBern (CH)
IRSN (FR)	VTT (FI)
LEI (LT)	ZAG (SI)

ENRESA (SP) via LTP

Workplan – Chemical evolution at different scales

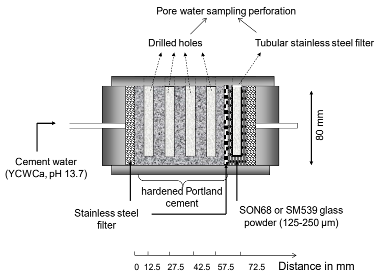
INTERFACE SCALE

WASTE PACKAGE SCALE

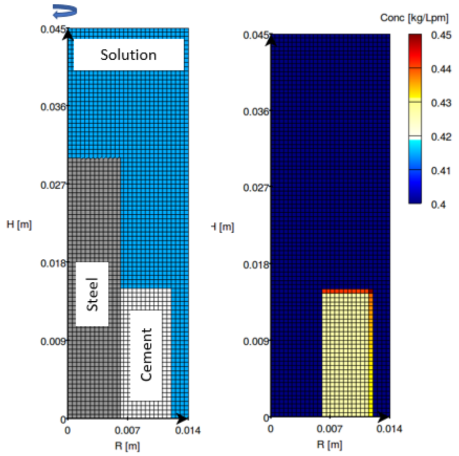
DISPOSAL CELL SCALE

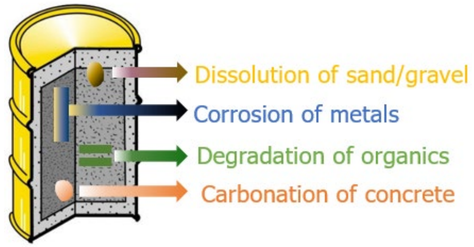
Experimental study

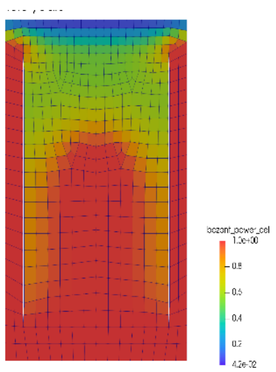
STEEL-CONCRETE and STEEL-CLAY

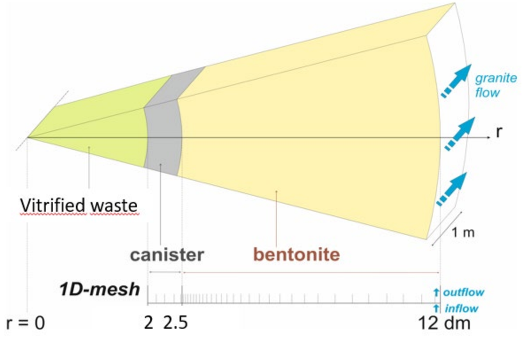


Numerical study – coupled reactive transport models





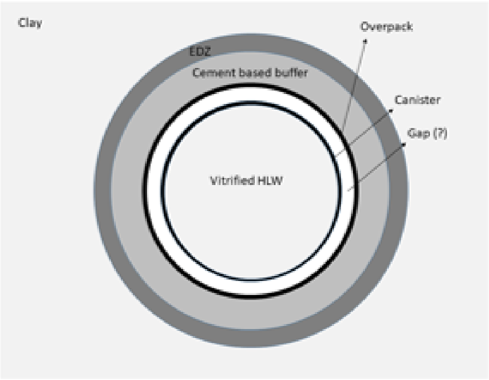


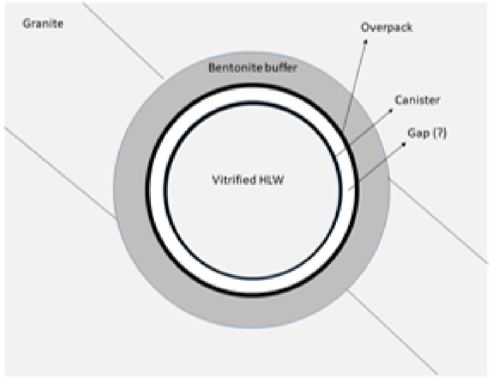


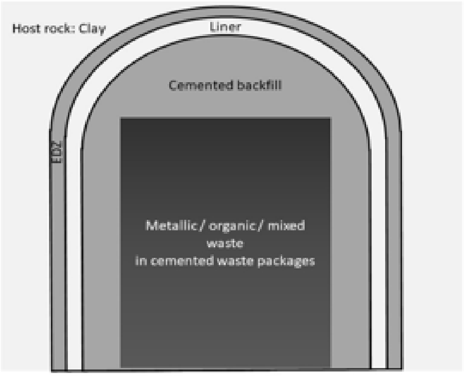
Generic disposal cell

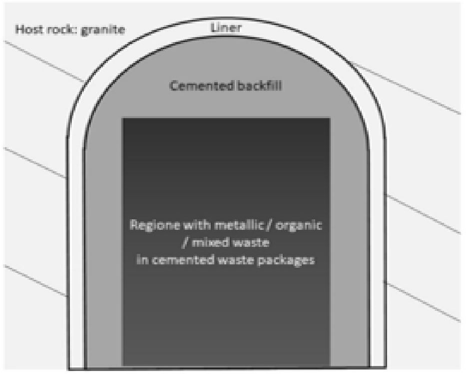
HIGH LEVEL ACTIVITY DISPOSAL CELLS

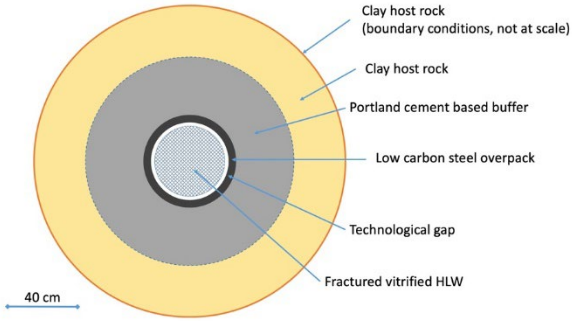
INTERMEDIATE LEVEL ACTIVITY DISPOSAL CELLS

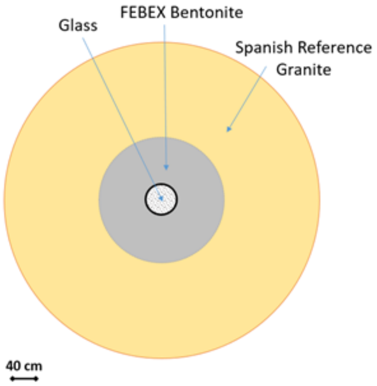


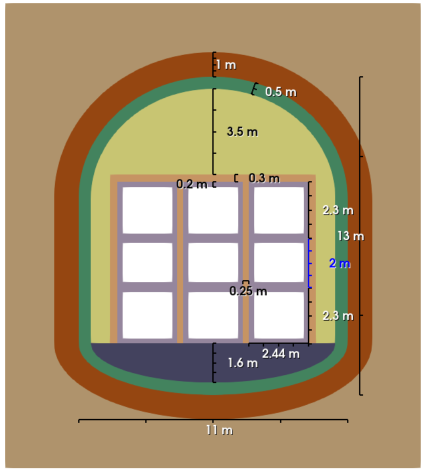


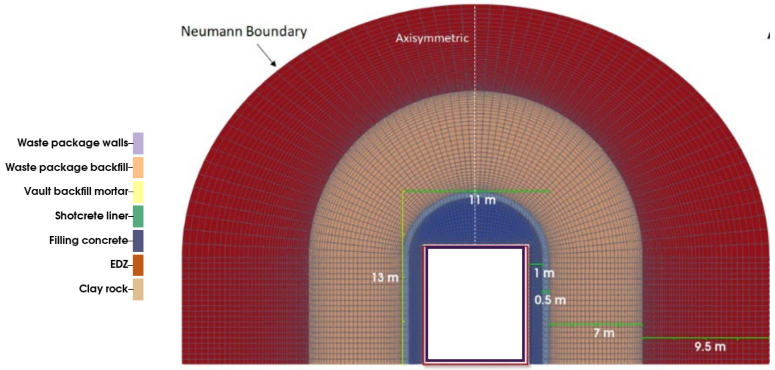








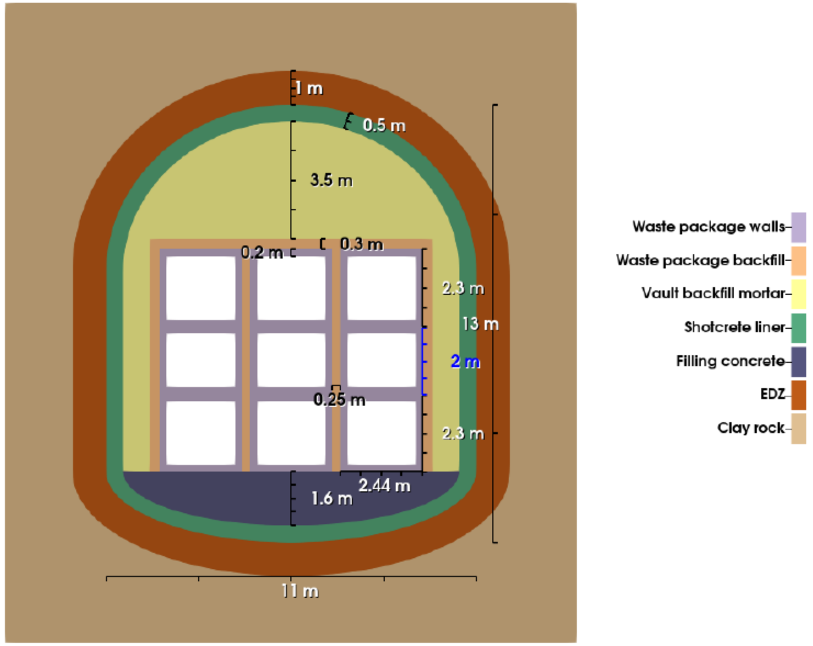


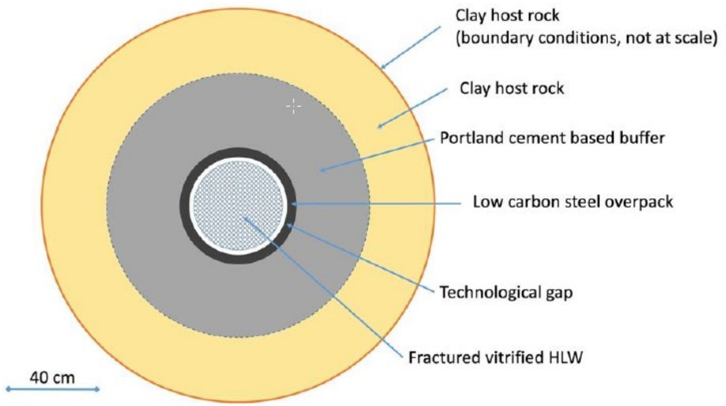


Disposal cell scale models

NARRATIVE, CONCEPTUAL and MATHEMATICAL MODELS

- **Integrated** model describing chemical reactions between engineered barriers at the disposal cell scale
- Models were developed by interaction between partners from different college's and countries
- Contains a lot of models and parameters



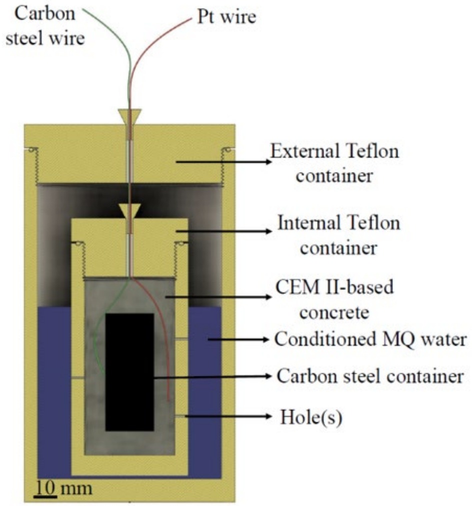


Interfaces

Phenomena at interfaces: experimental Steel/Cement

Corrosion measured

- High Temperature (80°C)
- Different pH (CEM-I/CEM-II – low pH cement-grout)
- In-situ / mock-ups



CEM-I/CEM-II

- Confirmation of low corrosion rate
- Identification of Fe-phases in cement

Low pH cement grout

- Localized corrosion mechanism (S(-2))
- Coalescence of pits seems to lead to a more generalized corrosion mechanism over a period of about 2 y
- Identification of phases led to conceptual model for development of corrosion products

Phenomena at interfaces: experimental Steel/Clay

long-term lab tests

iron/FEBEX compacted bentonite interface

Impact of simultaneous hydration/heating

corrosion processes/products/rates determination

batch and diffusion experiments

Fe^{2+}/montmorillonite interactions

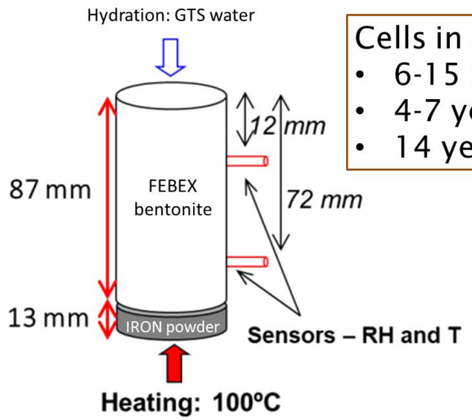
Fe^{2+} transfer in montmorillonite

Redox processes (electron transfers Fe^{2+}_{dis} / Fe^{3+}_{str})

Cell experiments

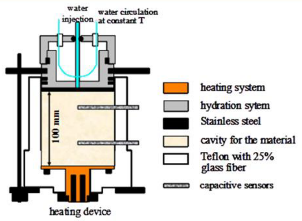
carbon steel/Boda clay interactions

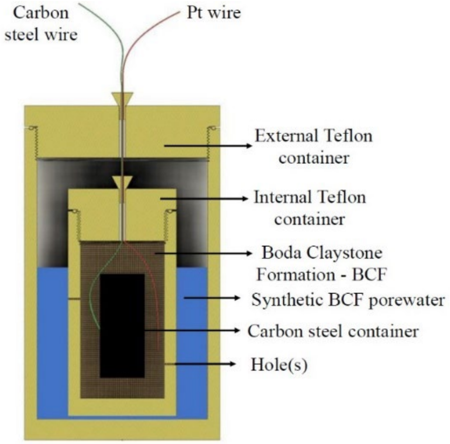
corrosion processes/products/rates determination

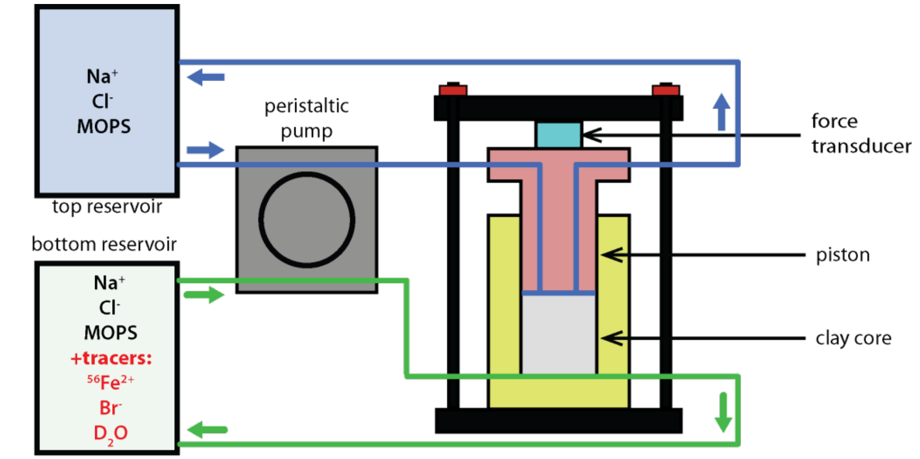


Cells in operation since 2006

- 6-15 months (FB1-2)
- 4-7 years (FB3-4)
- 14 years (FB5)







Phenomena at interfaces: Modelling Steel/Cement

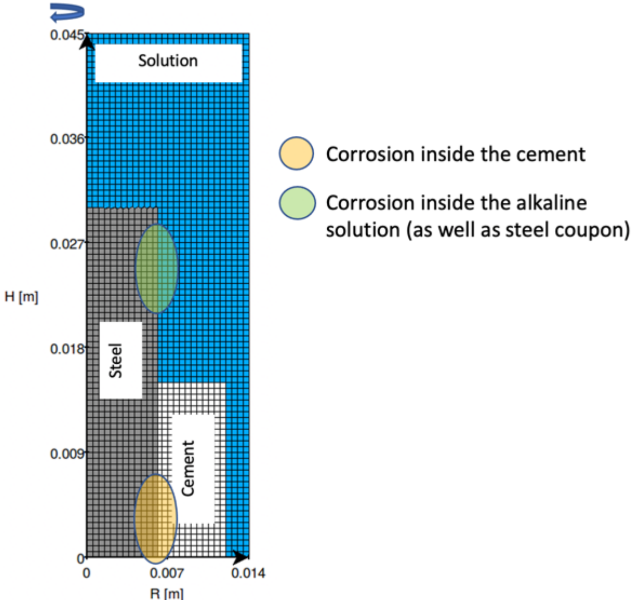
Thermodynamics of Fe-bearing cement hydrates

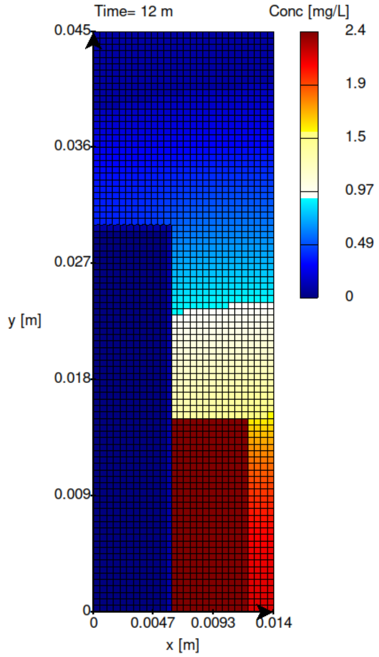
- Thermodynamics – Fe-garnet
- Extended C-A-S-H$^+$ model for Fe(III) & Fe(II)
  - Uptake Fe(III) >> Fe(II)

- Extend the flexibility of using multisite C-A-S-H$^+$ model
  - Script to translate solid solution model to discrete model usable in many more coupled reactive transport models

Modelling Mock-up experiments

- Geometry implemented for steel in contact with water or steel
- Models for material evolution were implemented including effects of temperature and pH





Phenomena at interfaces: Modelling Steel/Clay

long-term lab tests modelling

THCM reactive transport model

water content/temperature/porosity gradient

geochemical (pH, dissolved ions, CEC) distribution

FEBEX in situ experiment modelling

(i) reactive transport code (simplified corrosion approach)

(ii) steel-bentonite interaction model

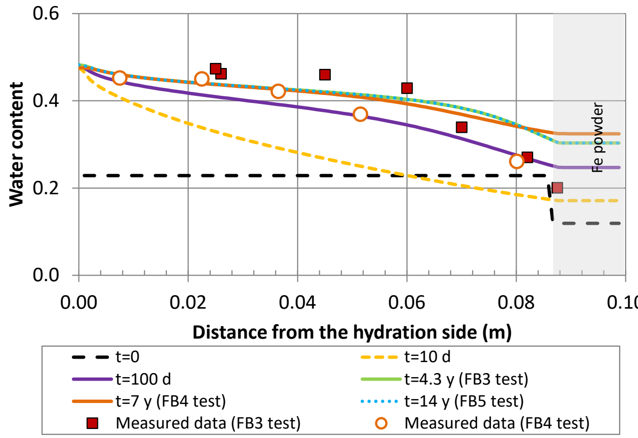
Corrosion rate and corrosion products estimation

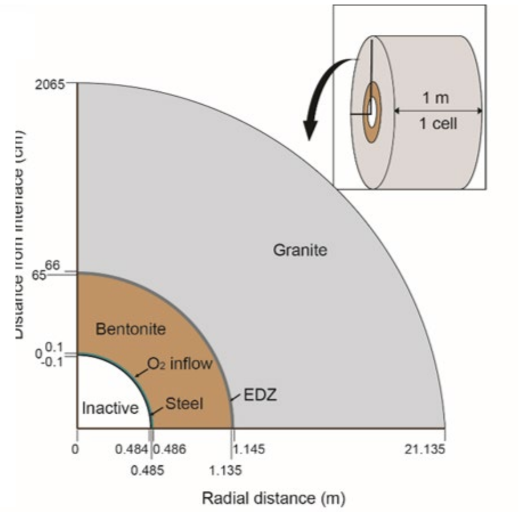
Fe^{2+}/montmorillonite interactions modelling

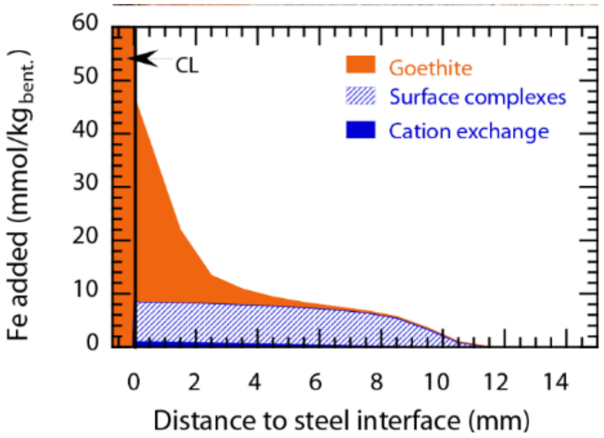
Sorption (batch) and reactive transport (diff + sorption) models

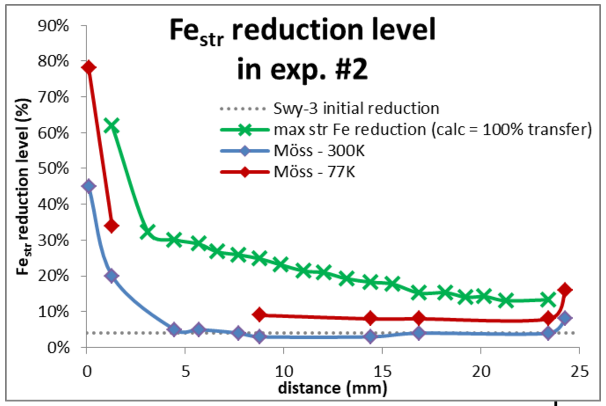
Solute diffusion and Fe sorption (complexe, cation exch, redox sorpt)

water content, pH, dissolved ions spatial distribution







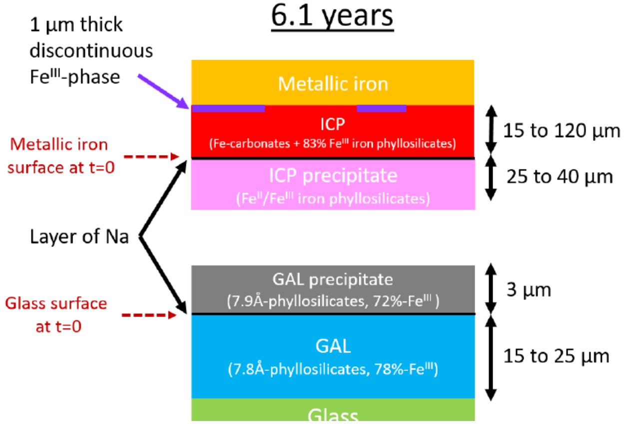


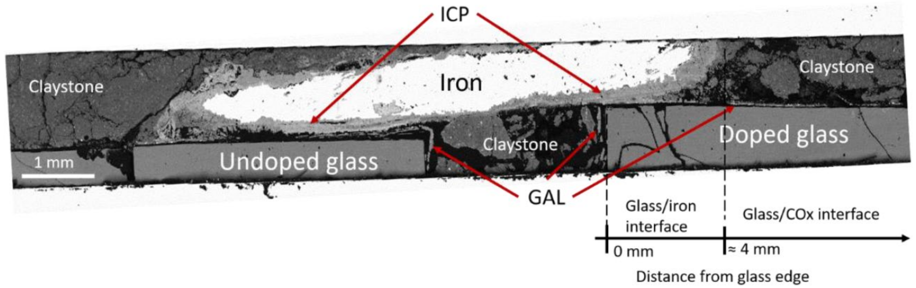
Waste packages

Phenomena at multiple interfaces – Experiments HLW

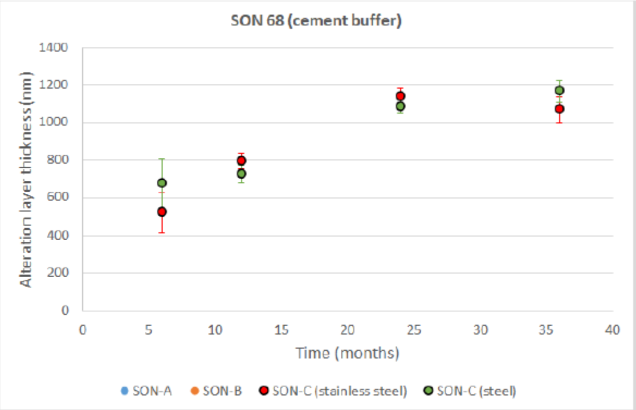
Experiments

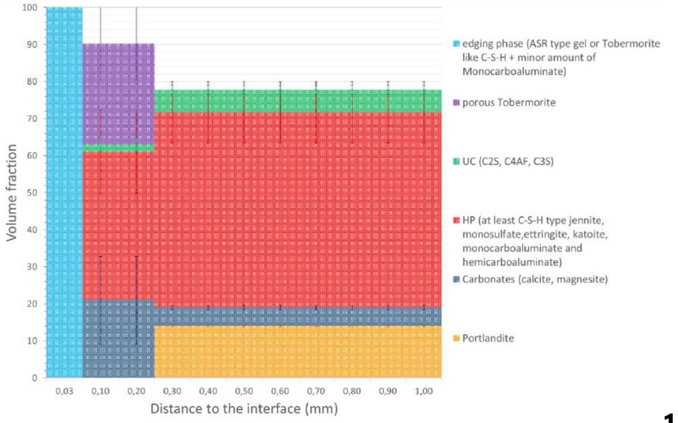
Glass/steel/clay — Glass/steel/cement buffer/clay — Glass/steel/cement





- Identification of neoformed phases (although difficult because small amounts & poorly crystallized) has been done with multiscale analytical approaches
- Pore water sampling for representative disposal conditions (high solid/liquid ratio) was successful in 1 set-up
- Insight in development of alteration zones
- Estimates of alteration/corrosion rates





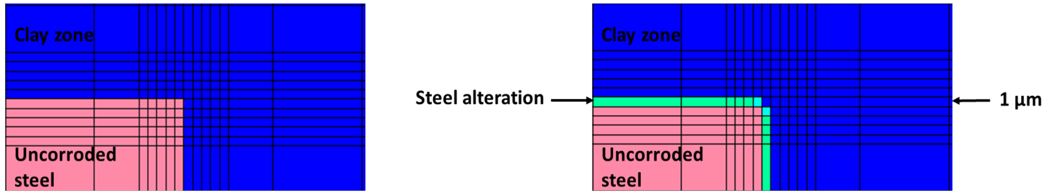
Phenomena at multiple interfaces – Modelling HLW

Modelling

Glass dissolution models for

- “Low” pH – GRAAL model implemented in PHREEQC-based reactive transport model
- “High” pH model for interaction with cement

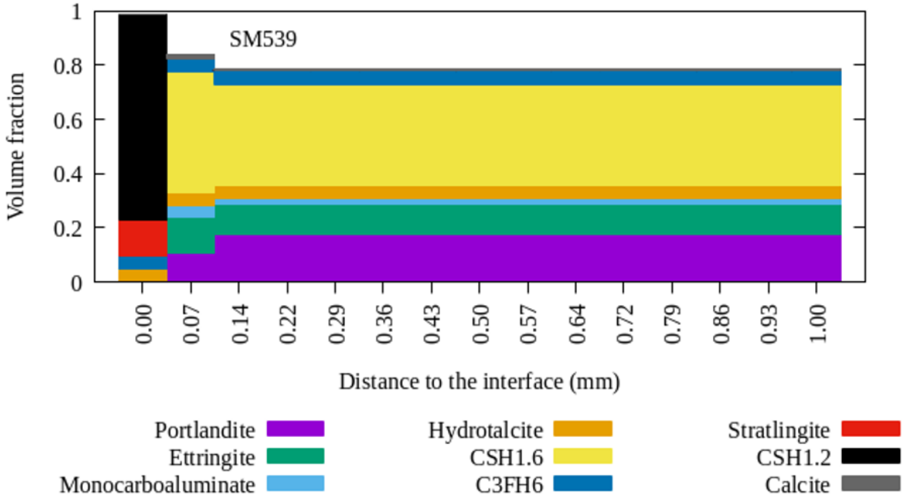
Implementation of layer by layer corrosion model of steel



Insight of evolution of alteration zones through modelling

Qualitative(/quantitative) agreement with experiments

However, careful selection of “minerals” for alteration products is required

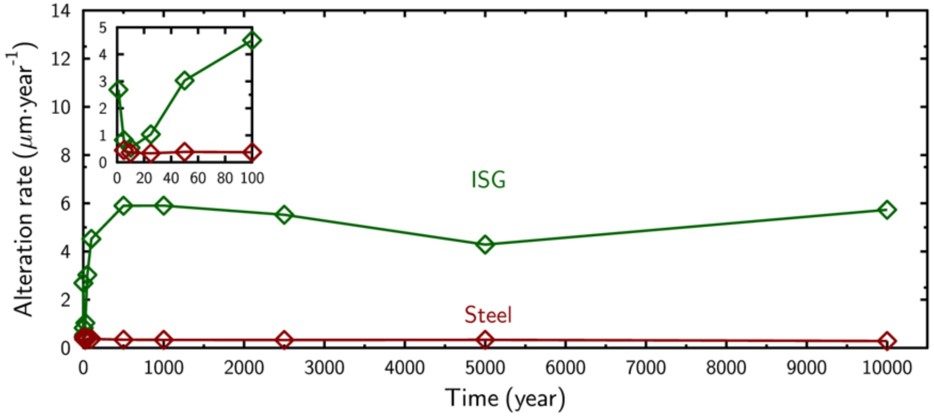


Upscaling to waste package and disposal cell – HLW

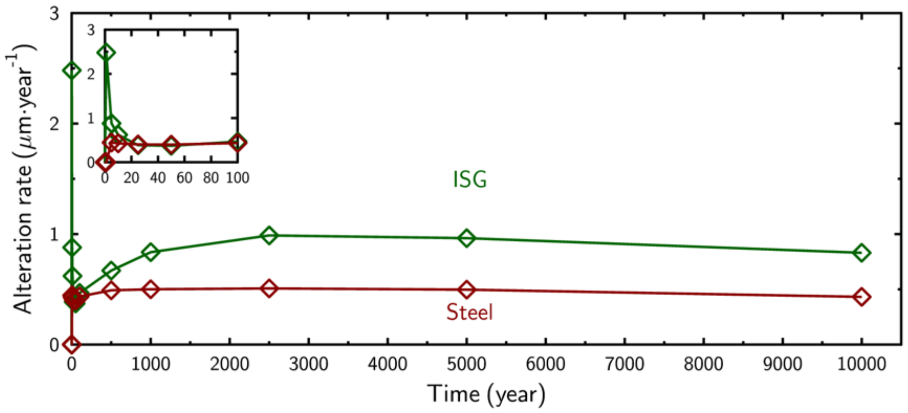
Waste Package

Upscaling to larger time and spatial scales (by considering coarser discretization)

Upscaled models allowed assessment of time-variable alteration zones as a function of geochemical conditions

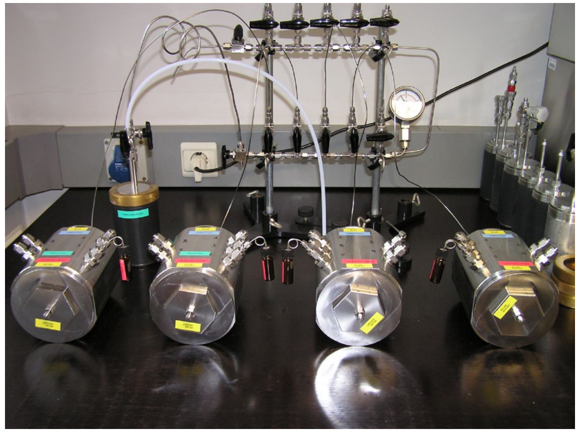


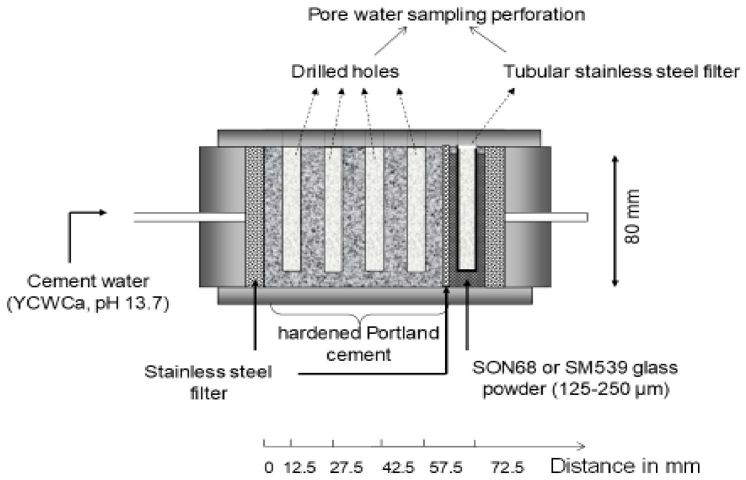
Alteration rates calculated at the waste package scale with the BRGM model at the glass – steel interface with a young concrete for a cement buffer of 100 cm thickness at 25°C.



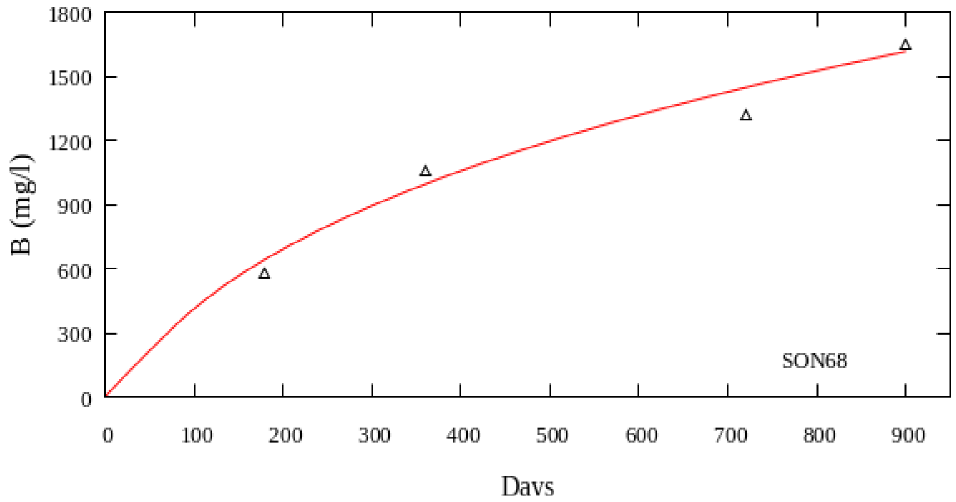
Alteration rates at the glass – steel interface, calculated with the BRGM model for an aged concrete at the waste package scale at 25°C.

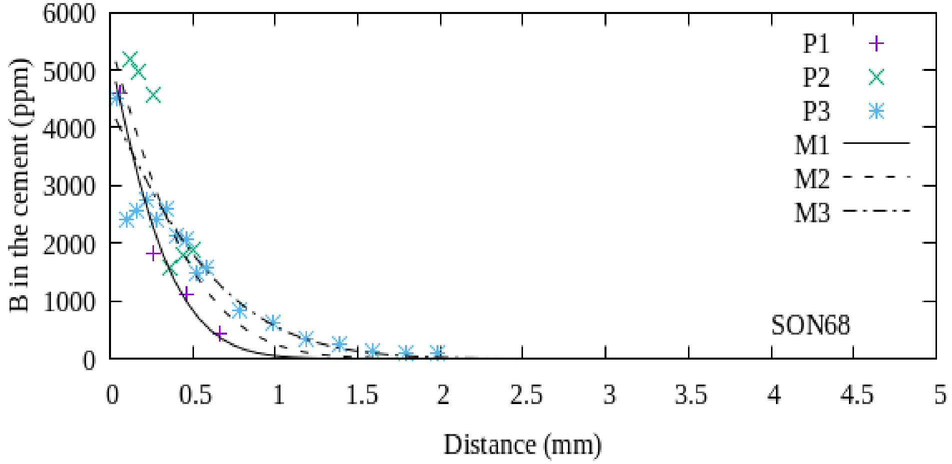
Glass – Steel - Cement





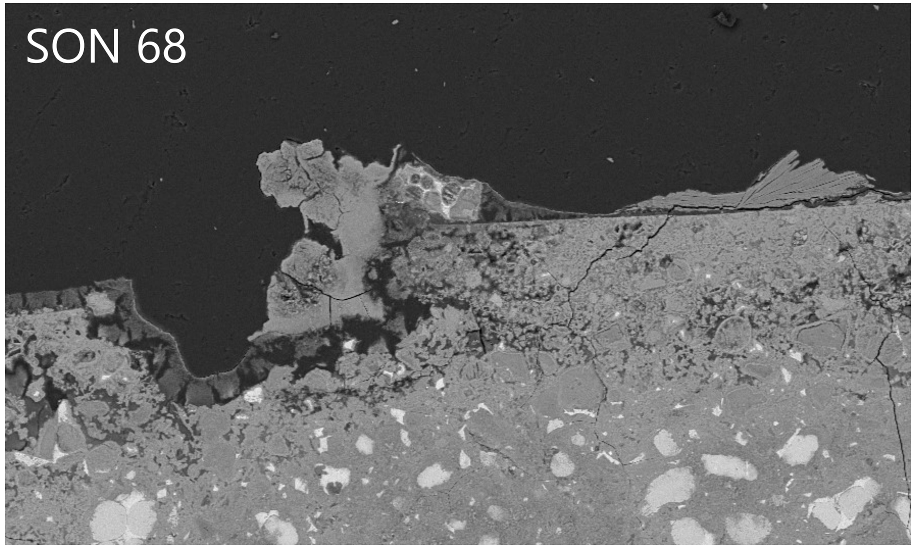
Aqueous chemistry: Model for glass dissolution & diffusion in cement



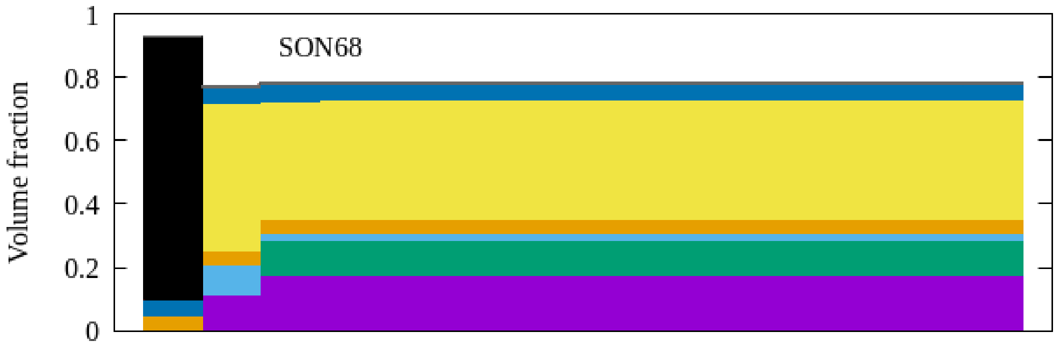


Glass – Steel - Cement

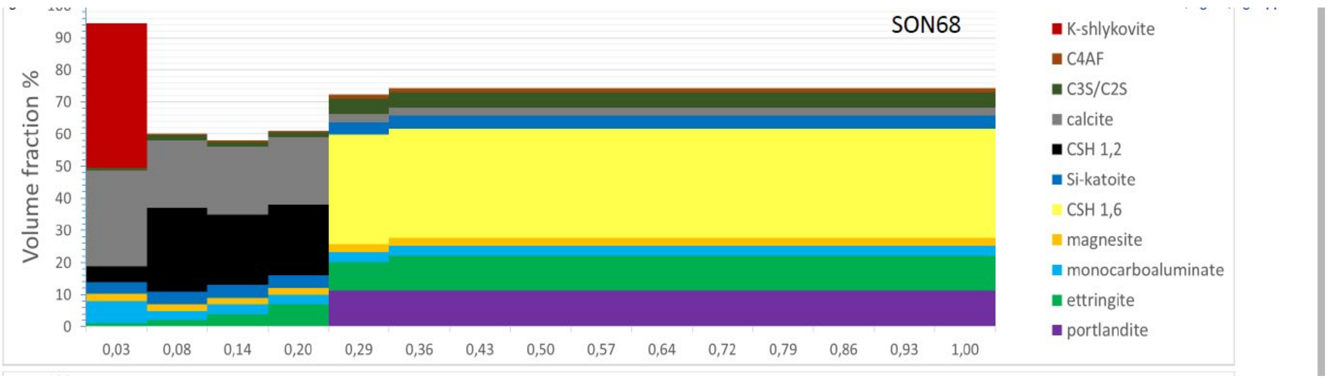
Solid phase characterization and modelling



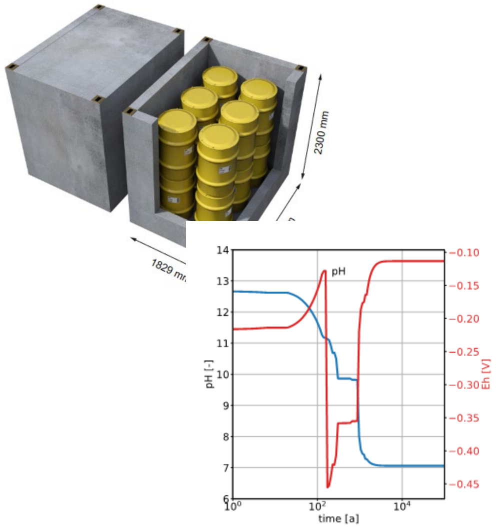
Model



Solid phase characterisation

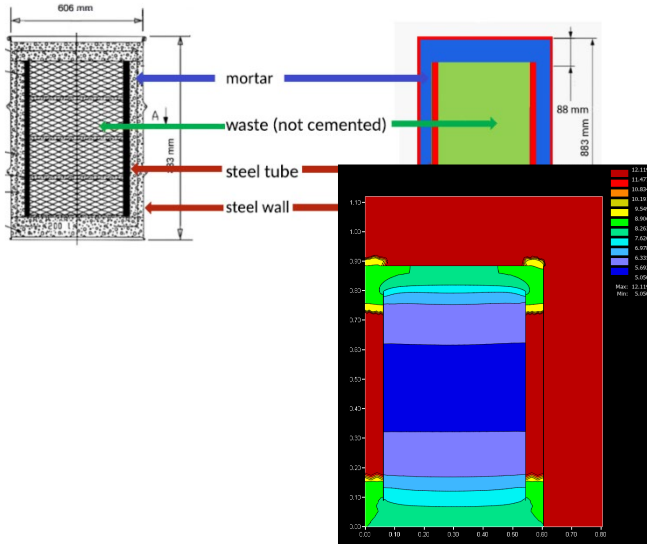


Waste package scale – ILW



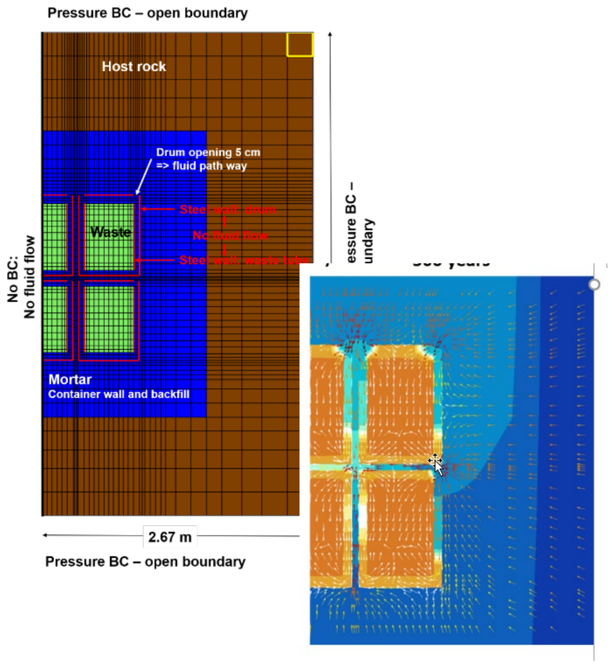
Mixing tank approach

Modelling time-dependent evolution without spatial dimension



Coupled reactive transport

Modelling temporal-spatial evolution – neglecting multiphase phenomena



Multiphase flow & transport

Modelling time-spatial evolution including multiphase phenomena but with abstracted chemistry

Disposal Cell

Upscaling disposal cell – HLW

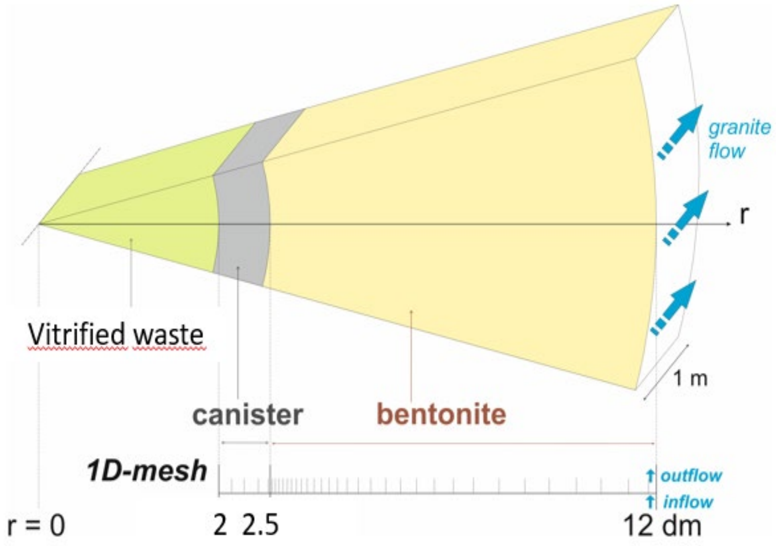
Disposal cell

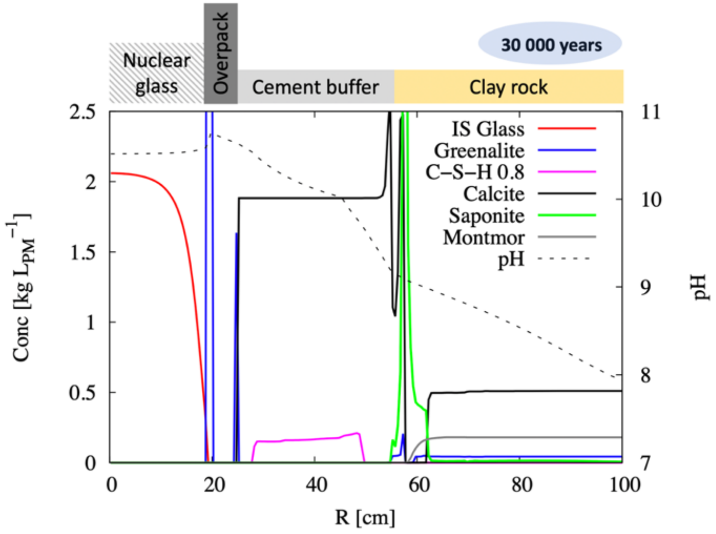
Successful implementation of models starting from detailed mathematical model (D2.16)

Two time frames are implemented (before and after breaching of canister)

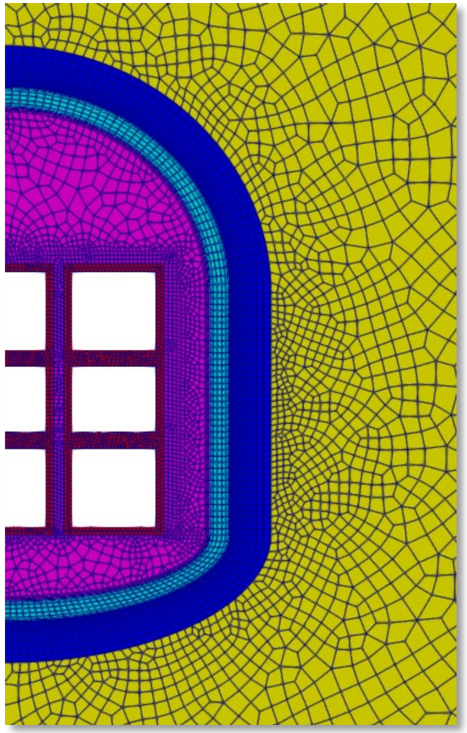
Insight in geochemical evolution caused by interactions of different barriers **(complete system)** over long time frames

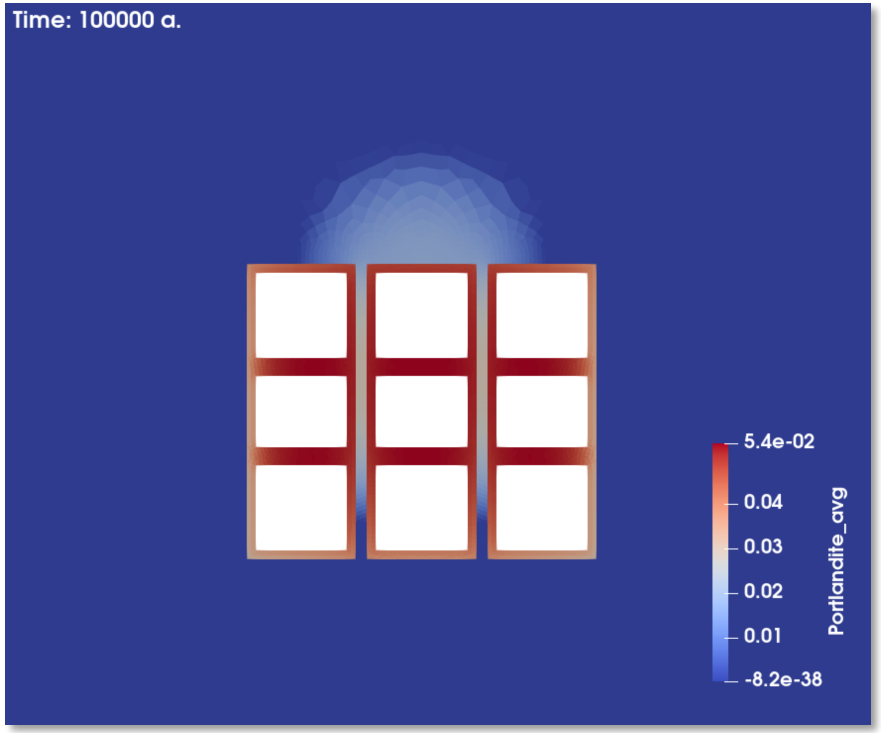
Transients (hydric, temperature) during first 1000 y in system with cement buffer could be neglected for rather thick buffers (>30cm) but accounted for in thin buffers (5 cm)

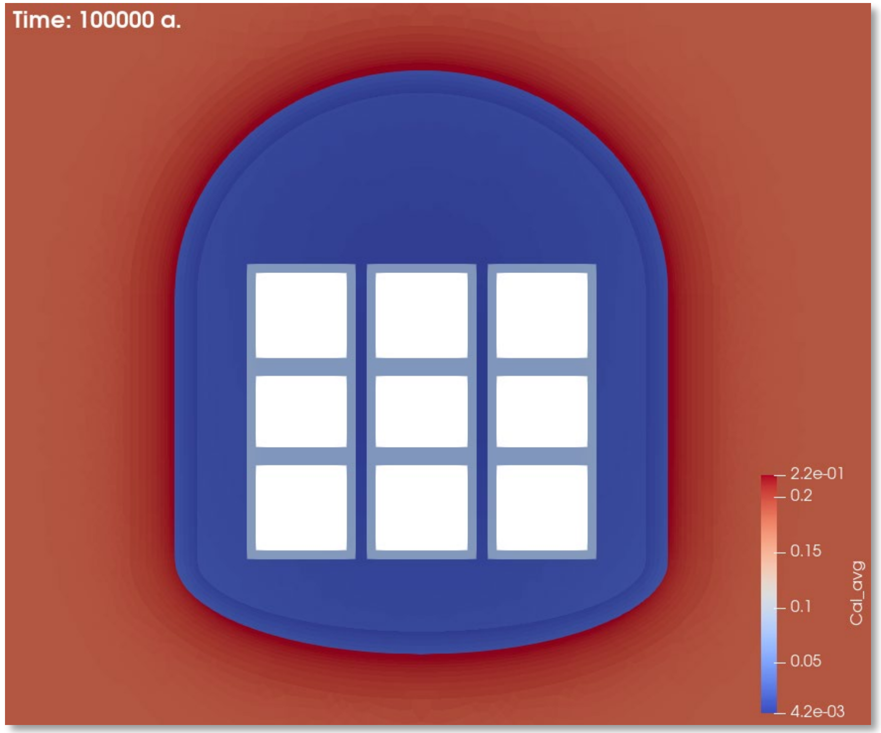




Upscaling to disposal cell – ILW

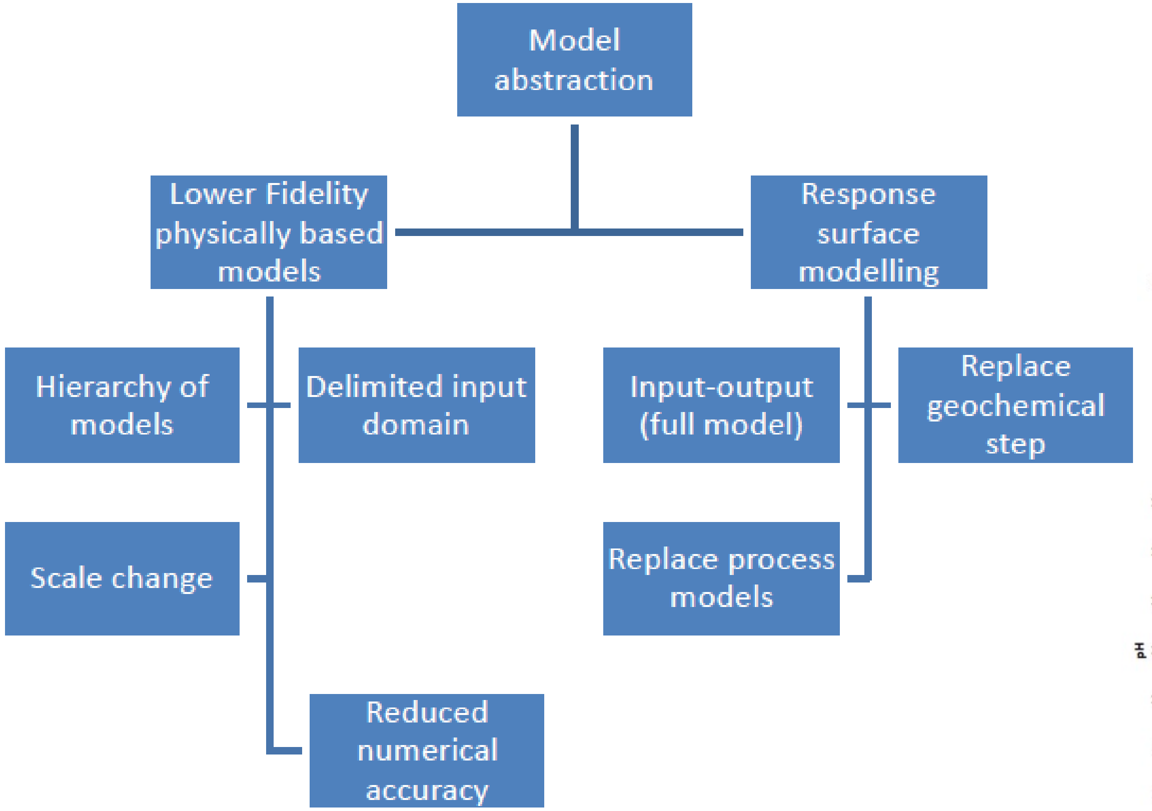


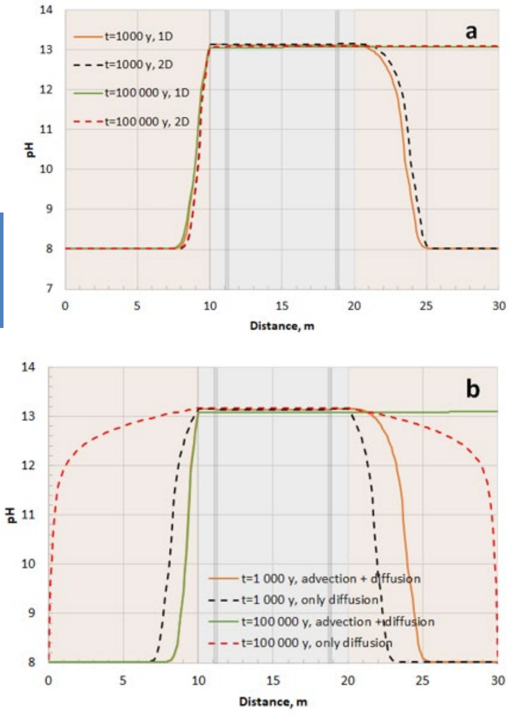


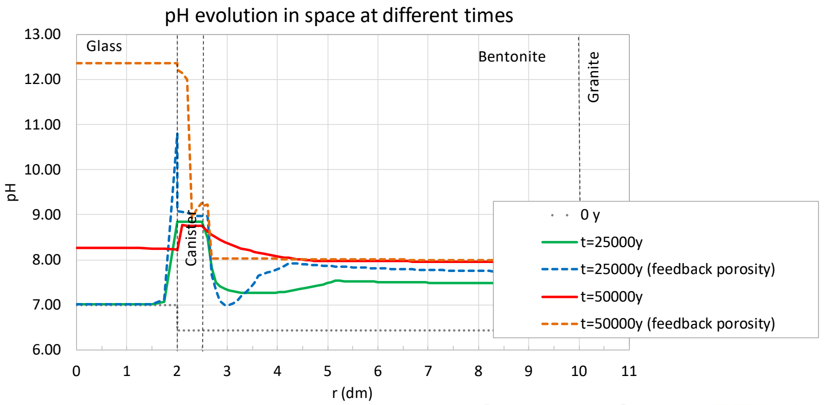


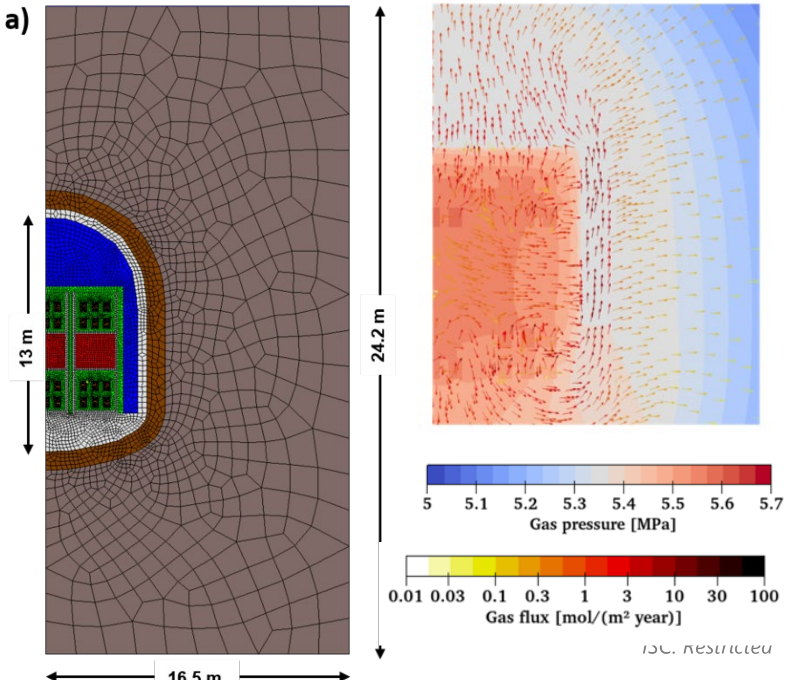
Abstraction - Sensitivity

Classification of abstraction methodologies and recent examples









Conclusions

Key Achievements

Experimental work

Advanced experimental set-ups

Advanced multi-scale characterizations

Training of less-advanced partners

Modelling work

Advancement in reactive transport models

Applications at different scales

Training of less-advanced partners

Insight in chemical processes at studied interfaces

New materials, different conditions

Input toward modelling (e.g. secondary phases)

Numerous examples of "full" and "abstracted" coupled reactive (transport) models

Integrated conceptual models

Approaches to modelling the processes

Lot of parameters

Final statement on « The modelling results in ACED »

Demonstrate the power of coupled reactive transport calculates

- For realist3ic engineering configurations specific for the ILW/HLW in DGR with all its complexities based on the good (mature) but still evolving scientific understanding
- Many first-time implementations of conceptual model where evolution is driven by processes
  - Geochemical conditions – and transient effects – determine degradation of waste, waste packages and barriers degradation with multiple interacting degradation pathways
- However – not the aim to predict the exact geochemical evolution, but rather insight in role of processes from interface to full disposal cell scale
  - Dissolution/degradation rates at time scales far beyond experimental duration

Therefore, a significant step forward has been made to include geochemical evolution calculations in national programs as an input to several aspects of scientific understanding, safety and optimization.

Belgian program

- Characterization of experiments that were running at SCK CEN
  - Additional interpretation and model development

- Disposal cell HLW in clay
  - Many relevant materials and processes for supercontainer
  - Variant available with thicker cement buffer

- General knowledge, models, parameters,… for e.g. Fe speciation and migration in concrete

- Demonstration of models coupling migration and geochemistry at different scales and for integrated systems (different barriers together)

SCK CEN
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Foundation of Public Utility

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