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Modelling microbiology, gas reaction and chemical evolution of geological disposal facilities

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Outline

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- WG5 expectations
- Specific interests- Biogeochemical Modelling
  - Approach for LLW surface disposal, UK
  - Microbial Gas generation, TVO Finland
  - Mont Terri Bitumen Nitrate experiment
  - Cement pH, UK Bigrad Consortium
- Benefit to the safety case
- Proposal ideas

Expectations on the outcome of WG-5

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- An integrated and balanced proposal to address microbial effects that may be both beneficial or detrimental to the safety case:

- **Detrimental**
  - Corrosion (MIC)
  - Methanogenesis
    - via acetate (LLW/ILW)
    - bulk gas
    - ^{14}C release

- **Beneficial**
  - Mediation of Eh & radionuclide speciation
  - H_2 (& CH_4) consumption
    - oxidation by Fe(III) & SO_4^{2-}
    - Methanogenesis (H_2 only)
  - Fate of organic complexants

"*The safety case must be able to describe the evolution of the repository in a way that may be seen as a reasonable representation of what might happen….*"

(Topic 1, IGD-TP Strategic Research Agenda, 2011)

Biogeochemical Modelling

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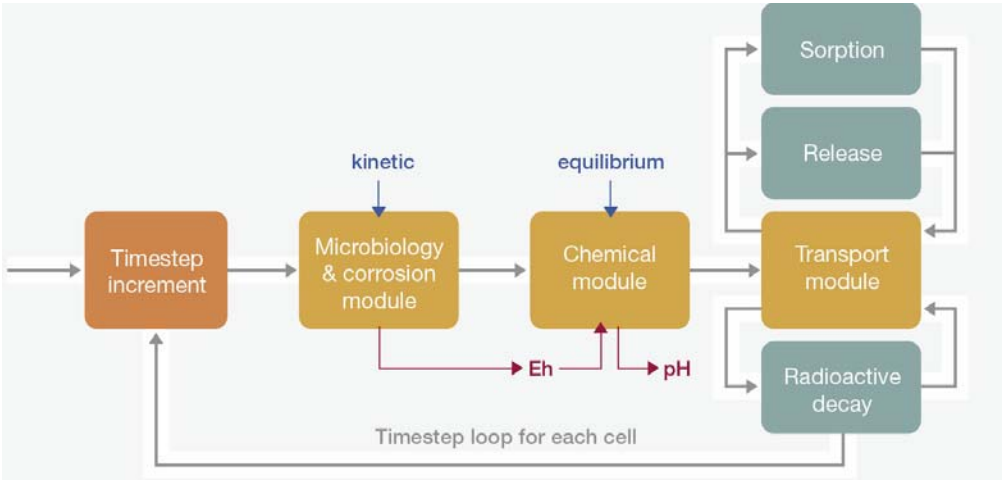
- Concepts and computer model developed in the UK for LLW surface disposal
- Key component of 2002 (BNFL) and 2011 (LLWR Ltd) safety cases
- Underpins near-field conceptual model
  - Eh evolution
  - pH evolution (neutral & cement wasteform)
  - Gas generation (CH_4, H_2)
  - Radionuclide speciation and solubility (U, Tc)
  - ^{14}C partitioning to gas, groundwater, carbonate precipitation



- Generalised Repository Model (GRM)
  - Coupled reactive-transport model
  - Representation of main microbial metabolic processes:
    - Aerobic
    - Denitrification
    - Fe(III) reduction
    - Sulphate reduction
    - Fermentation
    - Methanogenesis
  - Rections between:
    - H_2, organics (cellulose, acetate), reduced minerals
    - O_2, NO_3^-, $Fe(OH)_3$, SO_4^{2-}

Gas generation and microbial processes in repositories

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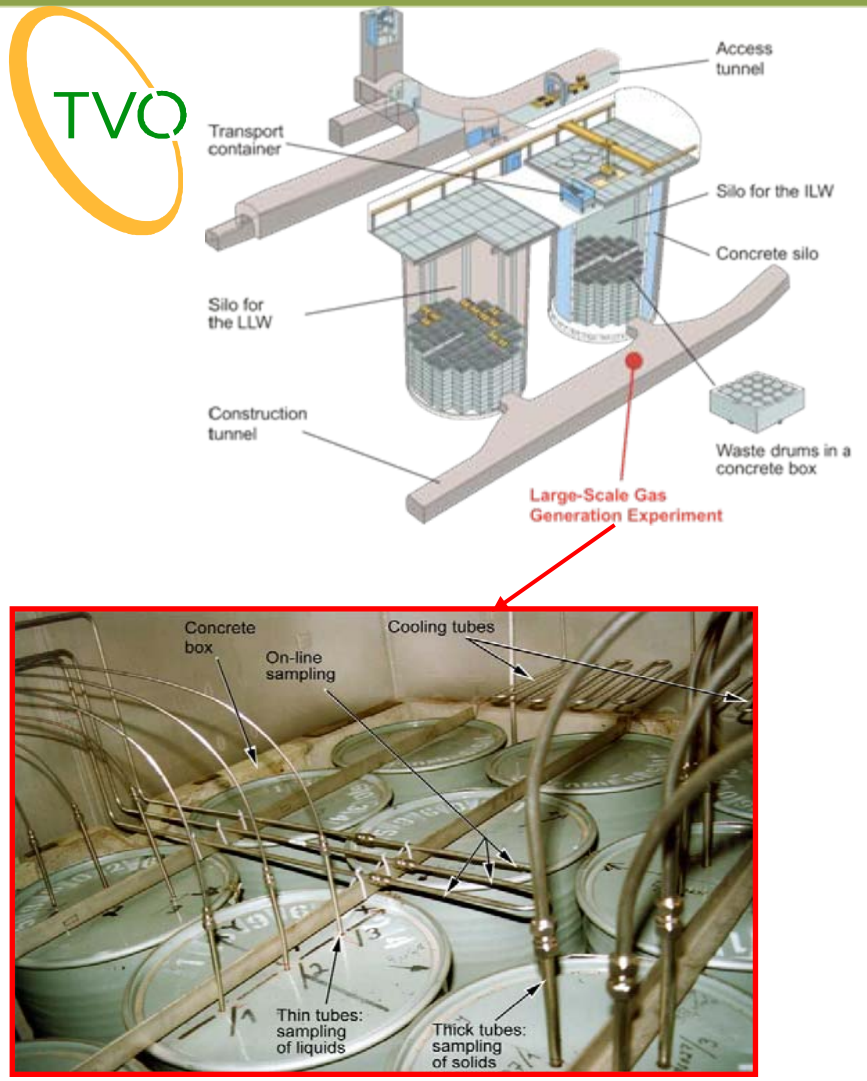
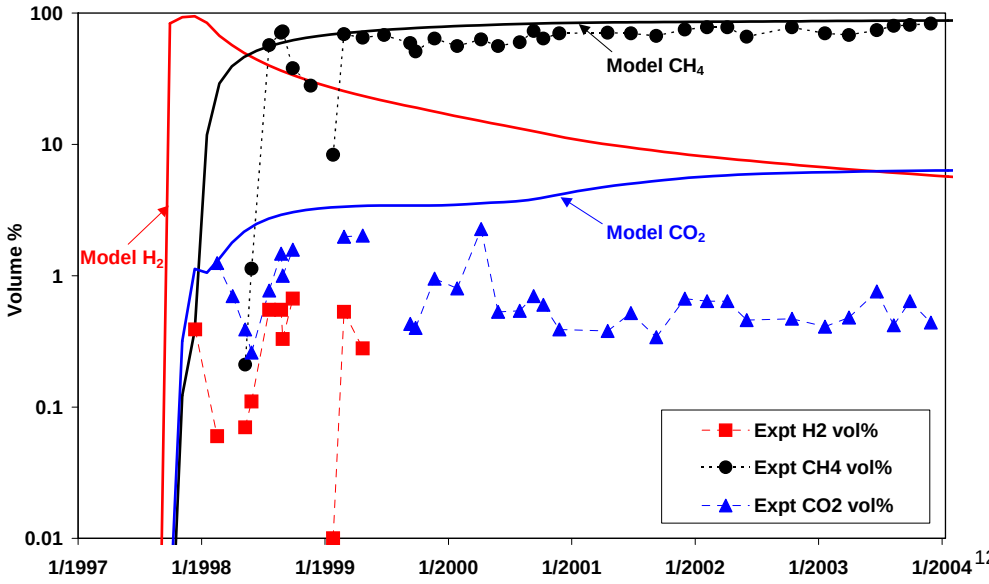
- VLJ Repository, Olkiluoto, Finland (operated by TVO)
- Large Scale Gas Generation expt. Examines CH_4, CO_2, H_2 generation from LLW/ILW
- Initially concrete buffered pH
- High water content

- GRM Biogeochemical modelling
  - Constrains rates of gas generation
  - Builds confidence in the VLJ safety case
  - Validates microbial modelling approach for gas generation and chemical evolution
  - GRM and expt. data now used internationally
    - e.g. as a validation test case
    - UK, France, Canada, Switzerland



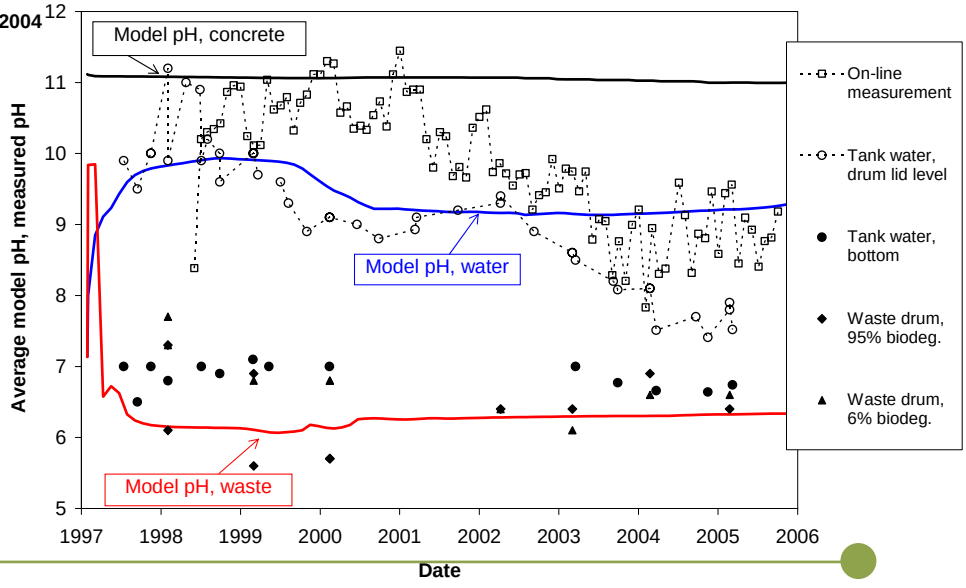
Small, J. **Nykyri, M**., Helin, M., Hovi, U., Sarlin, T., **Itävaara, M**. (2008). Experimental and modelling investigations of the biogeochemistry of gas production from low and intermediate level radioactive waste. *Applied Geochemistry* **23**. 1383-1418

TVO gas generation expt.



- Rate of CH_4 production modelled accurately in “blind test”
- Low levels of H_2 measured in the experiment
- H_2 from corrosion consumed locally in waste by SRBs

- Heterogeneity in pH allows microbial processes to develop at neutral pH in waste drums
- Eventually, concrete buffered alkaline water is neutralised



Effect of nitrate in radioactive waste disposal

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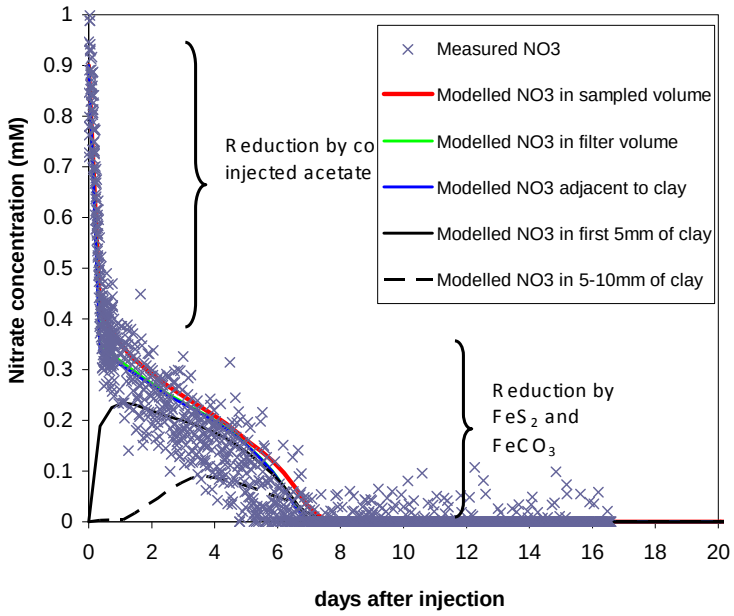
- Nitrate is a strong oxidant (electron acceptor)
- Potential to affect the speciation and transport behaviour of redox sensitive radionuclides *e.g.* Se, Tc, U, Np, Pu
- Large amounts of nitrate salts are present in bituminised waste, produced in France and Belgium
- The Mont Terri Bitumen-Nitrate experiment examines the biogeochemistry of nitrate interaction with a clay formation.

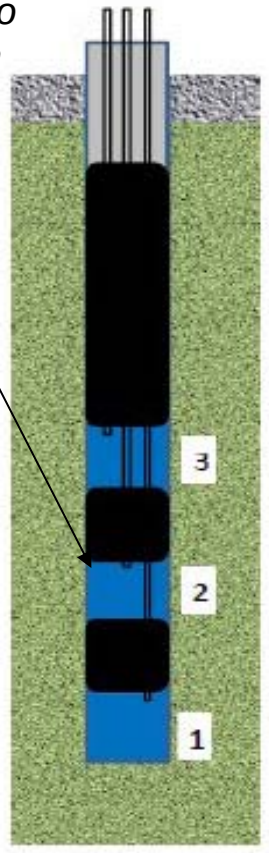
- GRM Model aids the interpretation of expt data
- Has been used to forward model the effect on redox potential (Eh) and U and Tc radionuclide speciation

- Migration 2013, Brighton; Small, J, Abrahamsen L, Albrecht A, Bleyen N, Valcke E



NO_3^- & organics injected into borehole in Opalinus Clay

Online monitoring



Mont Terri Project
Underground Rock Laboratory
Laboratoire souterrain

SCK·CEN ANDRA nagra IRSN

Microbial Biomass:

1. Modelled Biomass of borehole and EDZ
2. MPN cultivation/ATP/SEM analysis of sampled fluid

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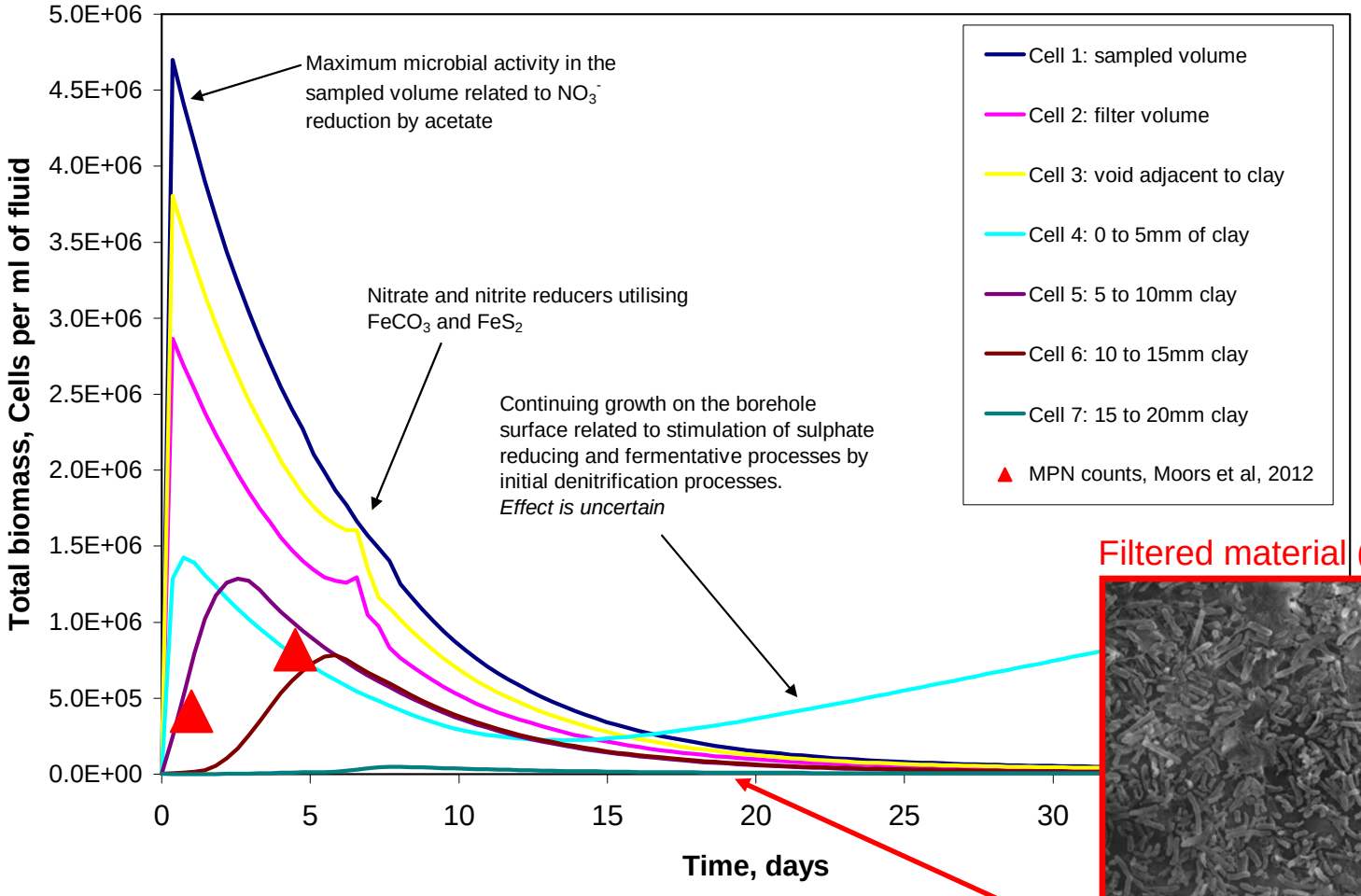
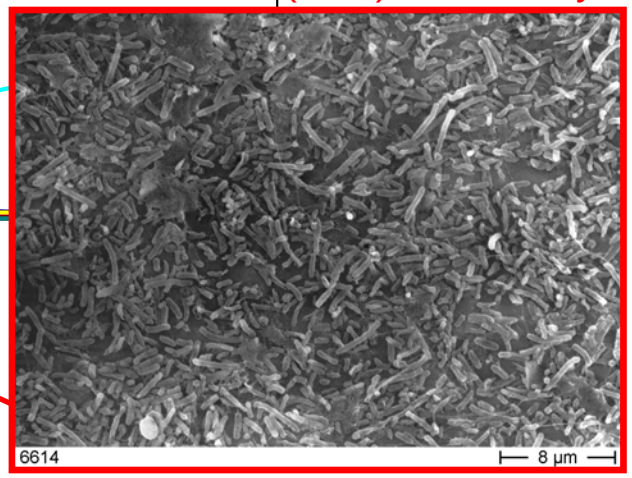


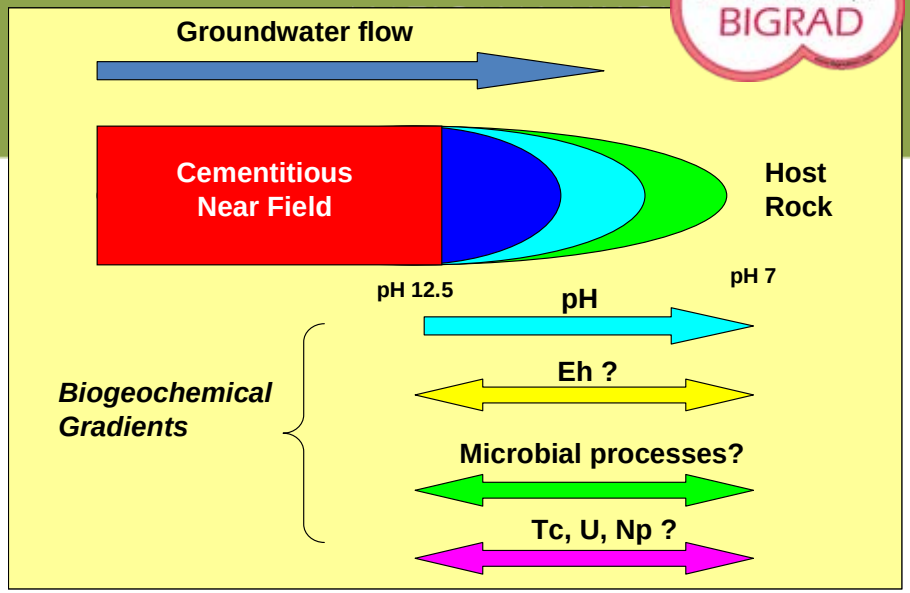
Filtered material (2mls) after 19 days

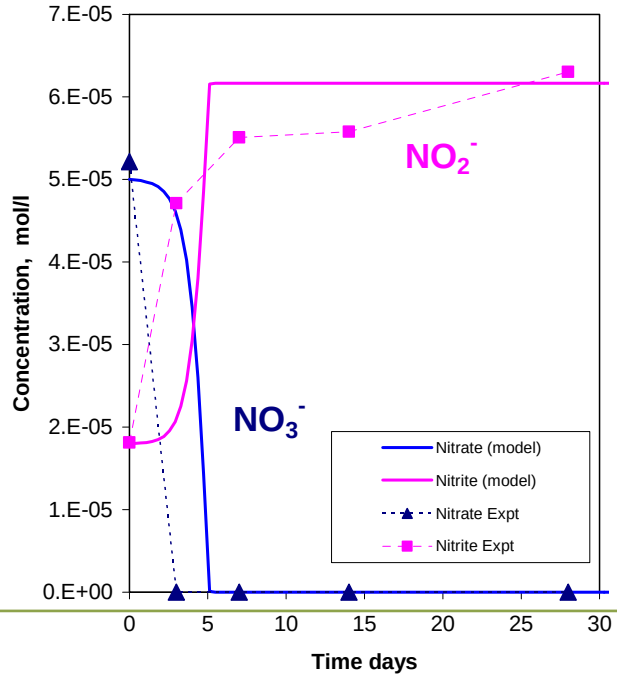


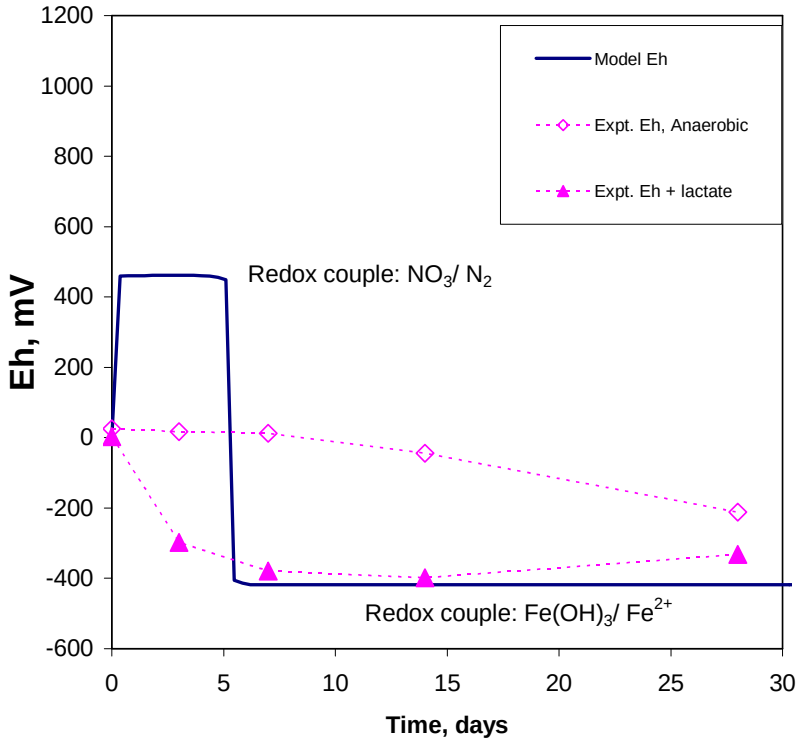
Moors, H., Geissler, A., Boven, P., Selenska-Pobell, S and Leys, N. *BN Experiment: Intermediate results of the microbiological analyses.* Mont Terri Project Technical Note 2011-39, 2012.

Microbial activity under cementitious conditions (pH 9-13)

- See Jon Lloyd presentation
- GRM models of denitrification, Fe reduction, and Eh at pH 10
- SO_4^{2-} reduction and CH_4 formation energetically less favourable at high pH
- Modelled Tc speciation consistent with observed microbial reduction
- U speciation models (based on NEA database) predict U(VI) whereas microbial U reduction observed







The safety assessment perspective: how can we contribute to a competent safety case?

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- Asses the viability of microbial processes under the range of physical and chemical conditions of ILW, HLW/SF wasteforms, engineered barriers and geosphere interface
- Realistic consideration of the fate of gases (H_2), organics (CDPs) and electron acceptors (NO_3^-) released from the waste and engineered barriers
- Development of microbiological process models (conceptual and mathematical) for repository evolution and radionuclide behaviour in the multibarrier system

Remaining issues and a proposal for future research

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- Investigate the viability of specific microbial processes that may be detrimental to the safety case:
  - Sulphate reduction and MIC in bentonite
  - Methanogenesis under cementitious conditions
- Develop a microbiological process model that reasonably describes the fate of repository derived energy sources:
  - H_2 and CH_4 gases,
  - organics, including cellulose degradation products,
  - nitrates and other electron acceptors present, and
  - which takes account of physical and chemical conditions heterogeneities and interfaces which may inhibit or promote microbial activity.
  - To test hypotheses for the Eh evolution and speciation of redox sensitive radionuclides.
- Utilise and develop current biogeochemical modelling approaches as an interpretive tool and to represent the above in the safety case