Microbial influence on the immobilization of radionuclides in crystalline rock environments

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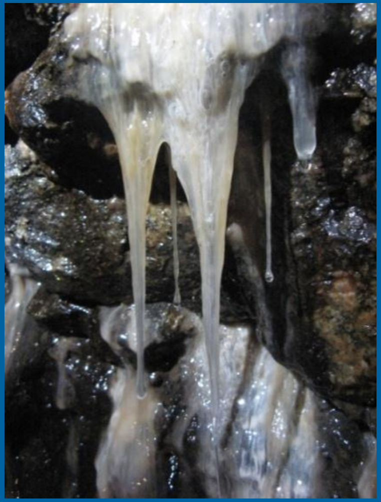


IGD-TP 4th Exchange Forum
WG 5 „Microbial studies“
Prague, Czech Republic
October 29th – 30th, 2013

Microbial influence on radionuclide immobilization

- In nonporous crystalline rock, the dominant transport medium for RN is groundwater flowing through subsurface fractures.
- Fracture surfaces support biological growth.
  - ⇨ Microbial diversity of biofilms depends on microbial consortia and chemical composition of the fracture water.
  - ⇨ The microbial consortia can significantly affect subsurface biogeochemical interactions, leading to adsorption of RN.

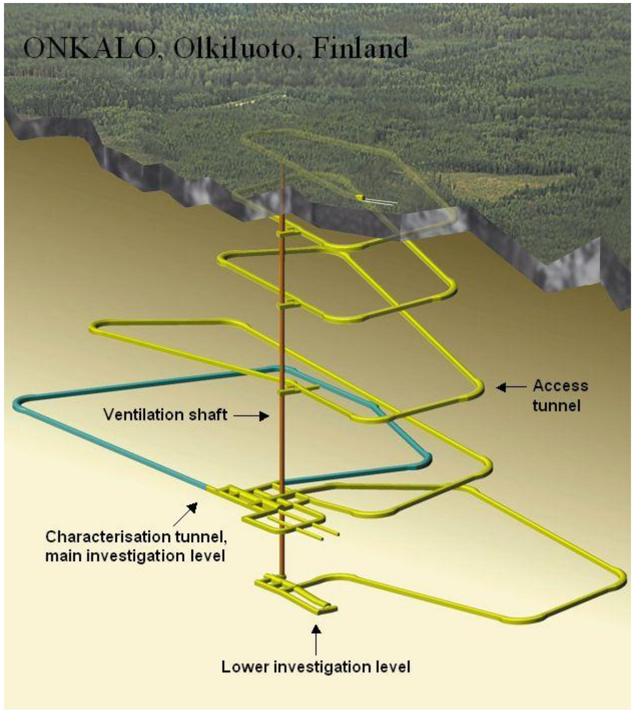
ONKALO: 20 % U uptake by micro-organisms, forming hardly soluble minerals	Äspö: 85 % U and 95 % Np uptake by bacteriogenic iron oxide precipitates (BIOS)

- Subsurface biofilms have a significant effect on the adsorption capacity of host rock formations by forming a barrier between the rock surface and the groundwater.
  - ⇨ Biofilms growing on fracture surfaces decrease the rock adsorption capacity for migrating RN.

➡ **Biofilm coverage can not be ignored**

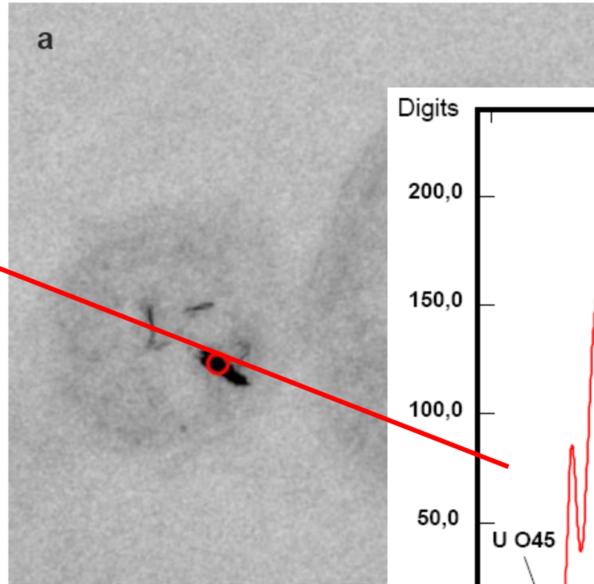
Example 1:

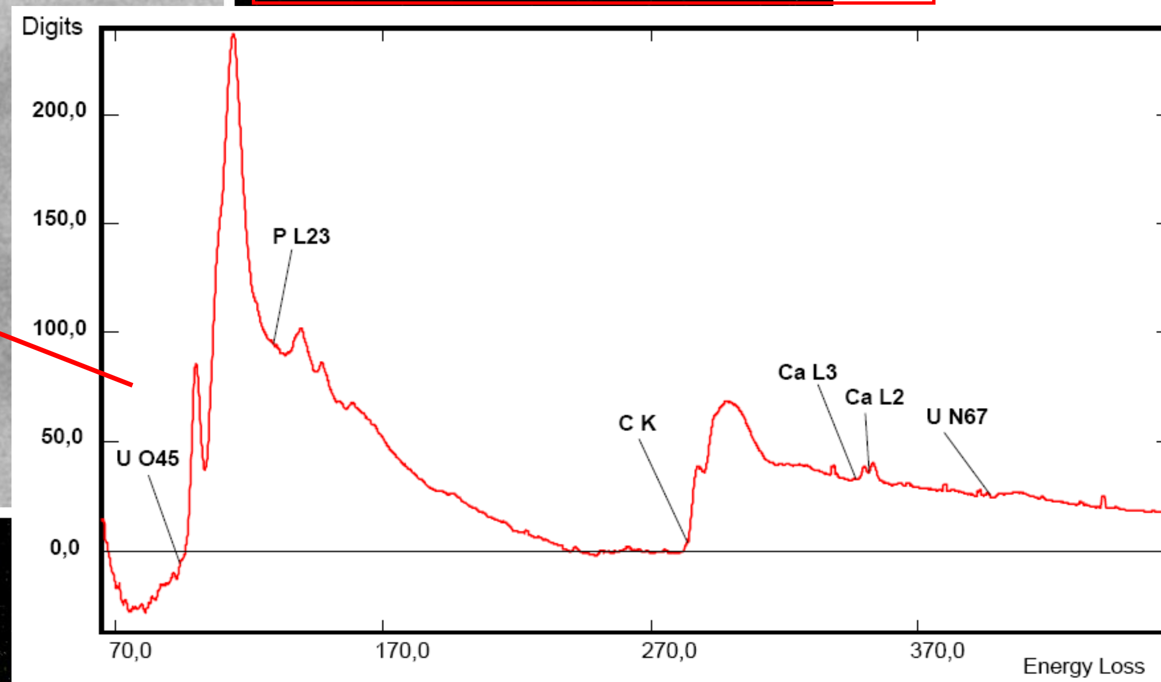
Future permanent nuclear-waste repository ONKALO, Finland

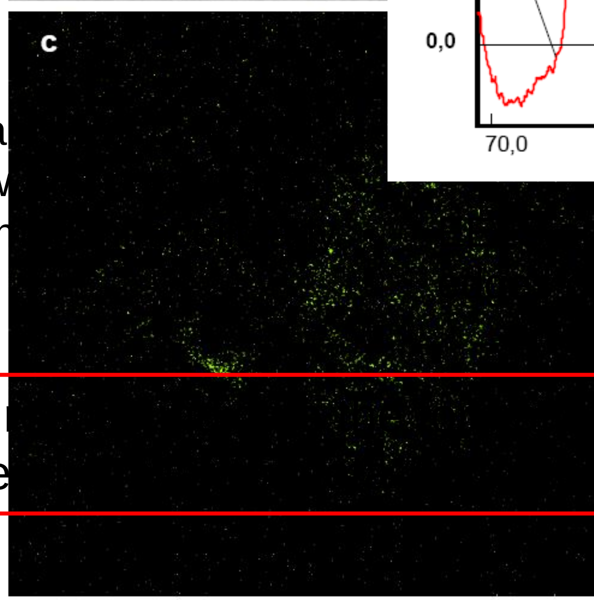


Intracellular formation of U-phophate mineral

Spectroscopic evidence for the
eral







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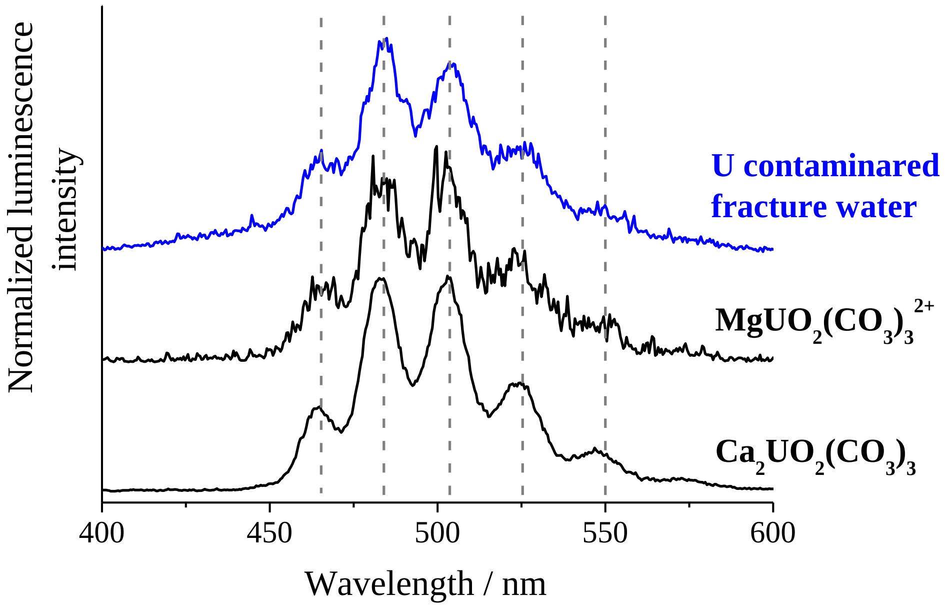
Aqueous uranyl carbonate species in the groundwater

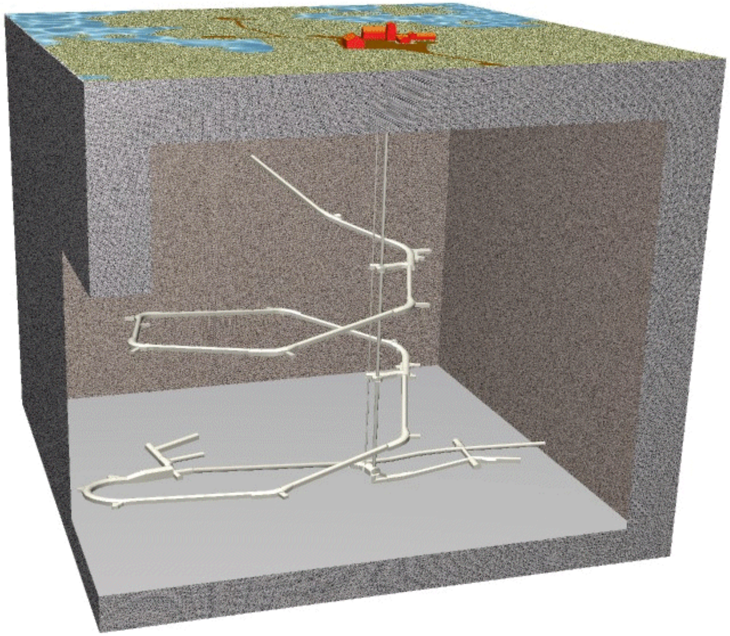


Samples	Luminescence emission bands / nm					Ref.
Fracture water	465.2	483.9	503.6	525.1	549.8	This work
$MgUO_2(CO)_3^{2+}$	466.4	485.0	504.5	526.9	551.2	Geipel et al. (2008)
$Ca_2UO_2(CO_3)_3$	466.0	484.6	504.1	525.4	549.1	Bernhard et al. (2001)

Formation of aqueous uranyl carbonate species in the U contaminated groundwater, most likely $Ca_2UO_2(CO_3)_3$ and $MgUO_2(CO)_3^{2+}$.

Example 2:

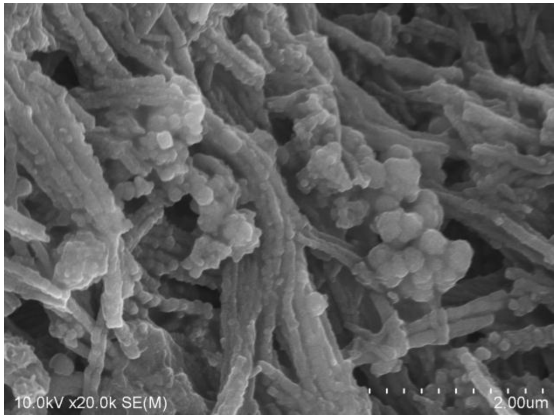
Hard Rock Laboratory Äspö, Sweden



U and Np uptake in BIOS biofilms

	BIOS biofilm dry mass after the experiment with U(VI) [µg/g]
Na	1,17E+04
Mg	4,10E+04
Al	9,45E+01
Si	1,01E+03
P	4,01E+03
K	7,71E+02
Ca	2,06E+04
Fe	3,76E+05
Co	2,29E-01
Ni	3,54E+00
Cu	2,34E+00
As	<5
Br	1,55E+02
Sr	6,19E+02
Sn	4,07E-01
Ba	5,74E+02
Pb	4,24E-01
U	5,00E+02

- Precipitation of ferrihydrite onto the extracellular stalks of *Gallionella ferruginea* depends on:
  - total supply of dissolved ferric iron
  - availability of reactive sorption sites (i.e. carboxyl or phosphoryl groups) in individual cells

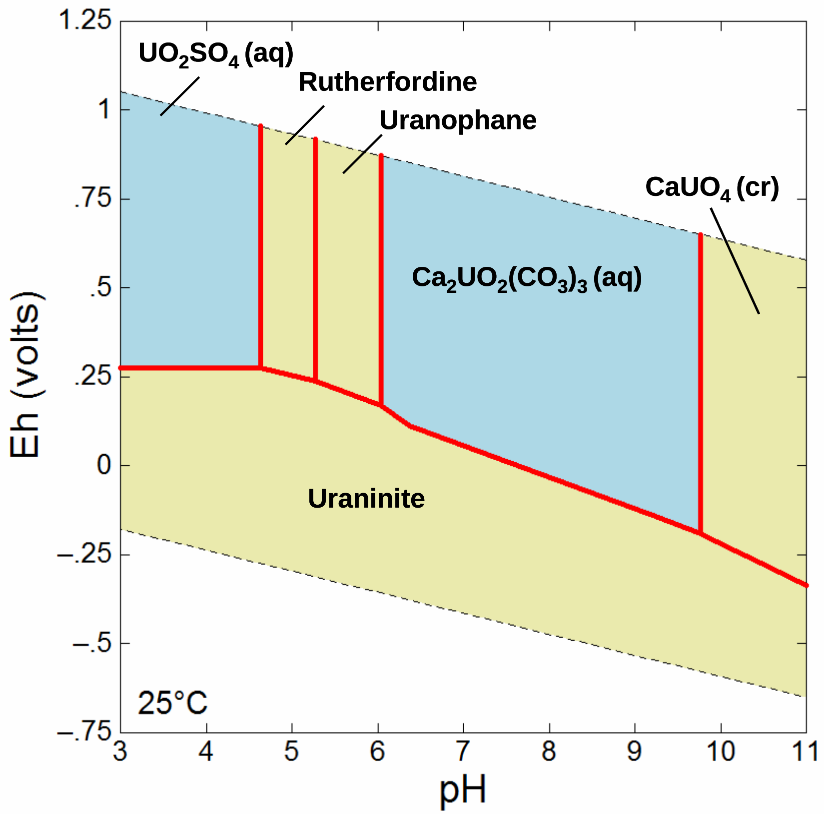


- Substantial uptake of U and Np in BIOS biofilms depends on:
  - high amount of precipitated ferrihydrite (70 % in BIOS biofilm)
  - neutral pH

Uptake of Uranium **(85%)** and Neptunium (**95%)**

by BIOS biofilms in neutral pH range.

Mobile calcium uranyl carbonate species in the groundwater



Calculation of predominance fields of U species * using analytical data of the contaminated ground water and Geochemist´s Workbench model package.

Under the ambient conditions the theoretical predominance fields can be defined:

- U(VI) solids: - Rutherfordine, Uranophane (4.7 < pH < 6.0)
  - $CaUO_4$(cr) (pH > 9.8)
- U(IV) solid: Uraninite (E_h < 260 mV)
- aqueous species: - UO_2SO_4 (pH ≤ 4.7)
  - calcium uranyl carbonate (6.0 ≤ pH ≤ 9.8)

Sensor measurements:

	pH	Eh [mV]
after the experiment	7,52	355

pH ± 0,2; Eh ± 30 mV

The highly mobile calcium uranyl carbonate species in the U contaminated ground water were calculated.

*** The database used was the thermo.dat accompanying the code, supplemented by the most recent NEA database for aqueous uranium species and mineral uranium phases (Guillaumont et al. 2003 *Chemical Thermodynamics* 5) and Bernhard et al. et al. (2001) *Radiochem. Acta* 89, 511-518.**

Expectation on the outcome of WG 5

- **Exceptance of the community, that the influence of microbial processes cannot be ignored and have to be implemented in the safety assessment.**

- Cooperation with members of WG 5 to expand the knowledge on microbial influence on RN migration behaviour.
  - Meetings in WGs for exchange of knowledge
  - Establishing on-site experiments (e.g. in crystalline rock environments – HRL Äspö) under aerobic and anaerobic conditions using long-lived RN

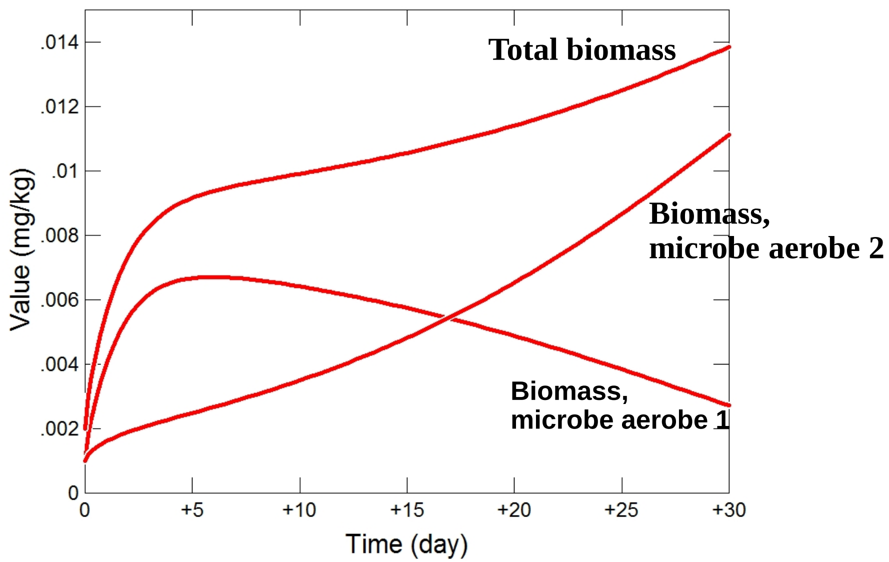
- Bringing forward new research projects

Relevance of microbial processes in a repository environment?

- Kinetics of biofilm growth?

Modelling:

- Trace the effect of microbial metabolism and growth in a geochemical system, using a generalized kinetic rate law.
- Estimating biofilm growth for process description, including different scales and different types of radionuclides.



Geochemist´s Workbench example calculation

Quantification of microbial processes

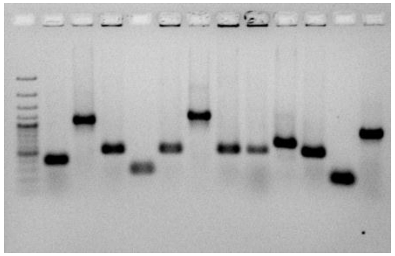
Assessment of microbial mediated retention of radionuclides

Remaining issues

- What will be the future microbial diversity in a repository in the case of:
  - changing geochemical conditions (E_h, pH, T, p)
  - opened and closed deposit
  - aerobic and anaerobic conditions?
- Which will be the sources for electron donors and electron acceptors for microbial activities?
  - in the far-field
  - in the near-field
- How do microbes influence the corrosion of container materials?
- What is the influence of microbial metabolic products?
- Which parameter are needed for modeling the kinetic of microbial growth?

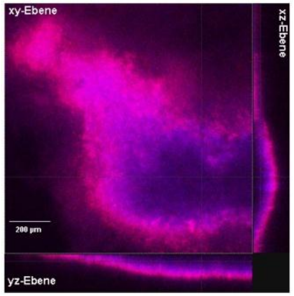
How can HZDR contribute to a competent safety case?

- Molecular analyses of the microbial diversity (PCR, cloning, sequencing, pyro-sequencing, FISH techniques)



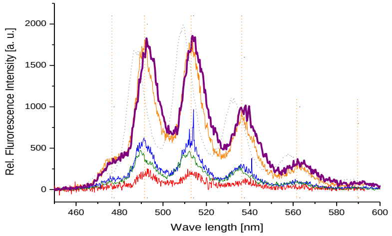
- Identification of the metabolic activity (e.g. CARD-FISH and RNA analyses)

- Culturing of representative microbial model systems in the laboratory

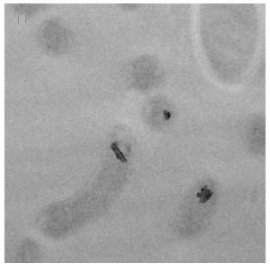


- Confocal laser scanning microscopy combined with Laser induced fluorescence spectroscopy (CLSM / LIFS)
  ⇨ RN interactions in situ, speciation in micrometer scale

- Time-resolved Laser-induced Fluorescence Spectroscopy (TRLFS) ⇨ RN speciation



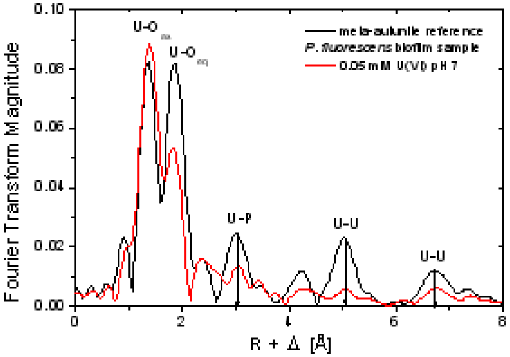
How can HZDR contribute to a competent safety case?



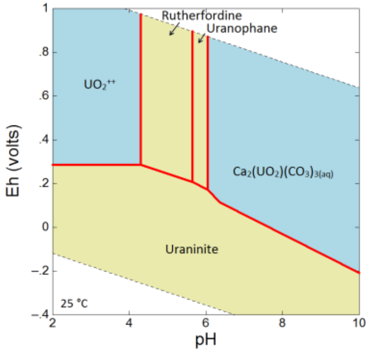
- Electron microscopy methods (SEM)/EDS, TEM/EELS)
  ⇨ ultra structure and elemental composition

- Microelectrode studies for redox determination
  ⇨ gradients of physico-chemical parameters

- Extended X-ray absorption fine structure spectroscopy (EXAFS)
  ⇨ structural information on RN



- Thermodynamic modelling ⇨ calculation of the predominance fields of RN species



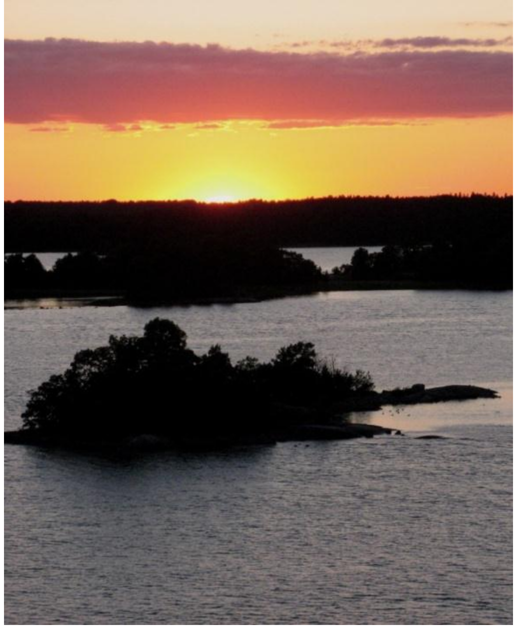
Concluding remarks for the round table

- For a comprehensive long-term safety analyses it is essential to include microbiological aspects.
- It is necessary to develop a framework to address the microbiological influence relevant for repositories.
- Collaborations and exchange of knowledge as well as establishing of on-site experiments are needed.

Open questions related to repositories and microorganisms:

- **How do MO influence the transport of radionuclides?**
- **How can we define the kinetic of microbial growth?**
- **How do MO influence the oxidation/corrosion of container materials?**
- **How do MO influence the stability of reactive barrier material, e.g. bentonite?**

Thanks to you for listening patiently!



Henry Moll´s presentation this afternoon according to microbes in clay and salt host rocks.

The research leading to these results has received funding from Euratom Seventh Framework Programme FP7/2007-2011 under grant agreement n° 212287 (ReCosy project) and under grant agreement n° 269658 (CROCK project).