Long Term Diffusion project (LTD)

HYRL, JAEA, NRI/RAWRA, AIST, NAGRA

Helsinki, 9th May 2008

Overview

- 1. LTD Background
- 2. Overview of phase I
- 3. Phase II planning

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The problem

- Matrix diffusion has been assessed on samples disturbed by collection and preparation which causes:

  - *Overestimation of the rock diffusivity (several orders of magnitude)*

  - *Overestimation in porosity (by a factor of two to three)*

-> Leads to an overestimation of matrix diffusion

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Values of geosphere model parameters for PG-85 and K-I

	PG-85	**K-I**
Diffusion depth	**0.5 m**	**0.05 m**
Diffusion volume	**1.3 %**	**0.02 %**

However:

- Connected Porosity (CP) experiment at Grimsel:
  - *Connected porosity extends several metres behind water conducting feature*
  - *Very conservative assumptions from PA*

- LTD NHC-9 also confirms this

Overall LTD aims:

- Study of *in-situ* diffusion -> **monopole and dipole experiments**
- Characterisation of pore space geometry -> ***in-situ* porosity versus laboratory-derived data**
- Coupled with **natural tracer studies** and **diffusion process studies**

LTD phased approach

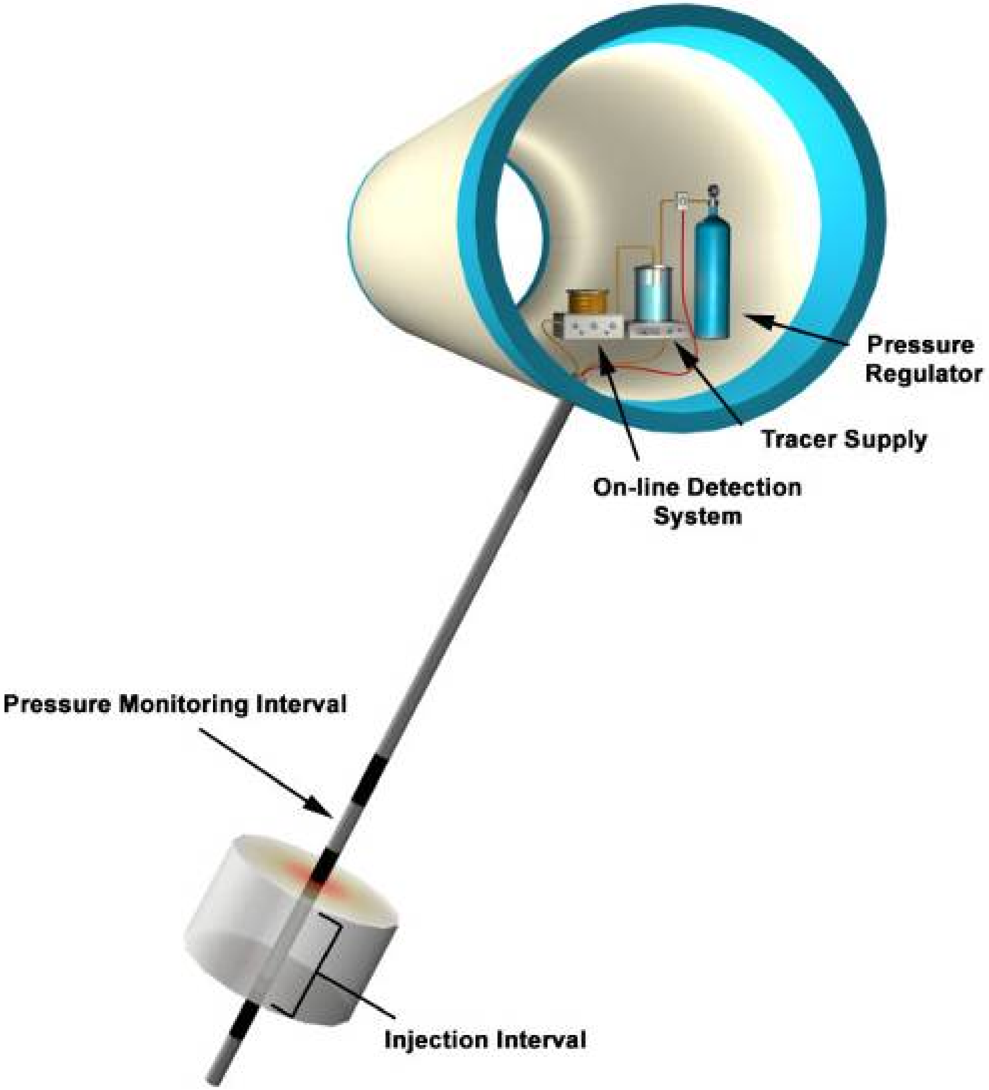
Phase I: 4 year project 2004 to 2008

- **WP 1 The Monopole Phase I 2005 to 2008 and preparation for long term test**
- **WP 2 *In-situ* porosity determination**
- **WP 3 Natural Tracer Studies (NTS)**
- **WP 4 Diffusion Processes Study (DPS)**
- WP 5 Final reporting and synthesis / LTD Phase II planning

Phase II: Starting in 2009

- WP 6 Long term monopole(s)
- WP 7 Mock-up tests (overcore with fracture)
- WP 8 Circulation of tracers through a fracture
- WP 9 Reporting Phase II and overall synthesis Phases I & II

WP-1:IN-SITU DIFFUSION EXPERIMENT



- Demonstration of matrix diffusion in undisturbed rock as a geosphere retardation process
- Develop long term monitoring techniques and strategies

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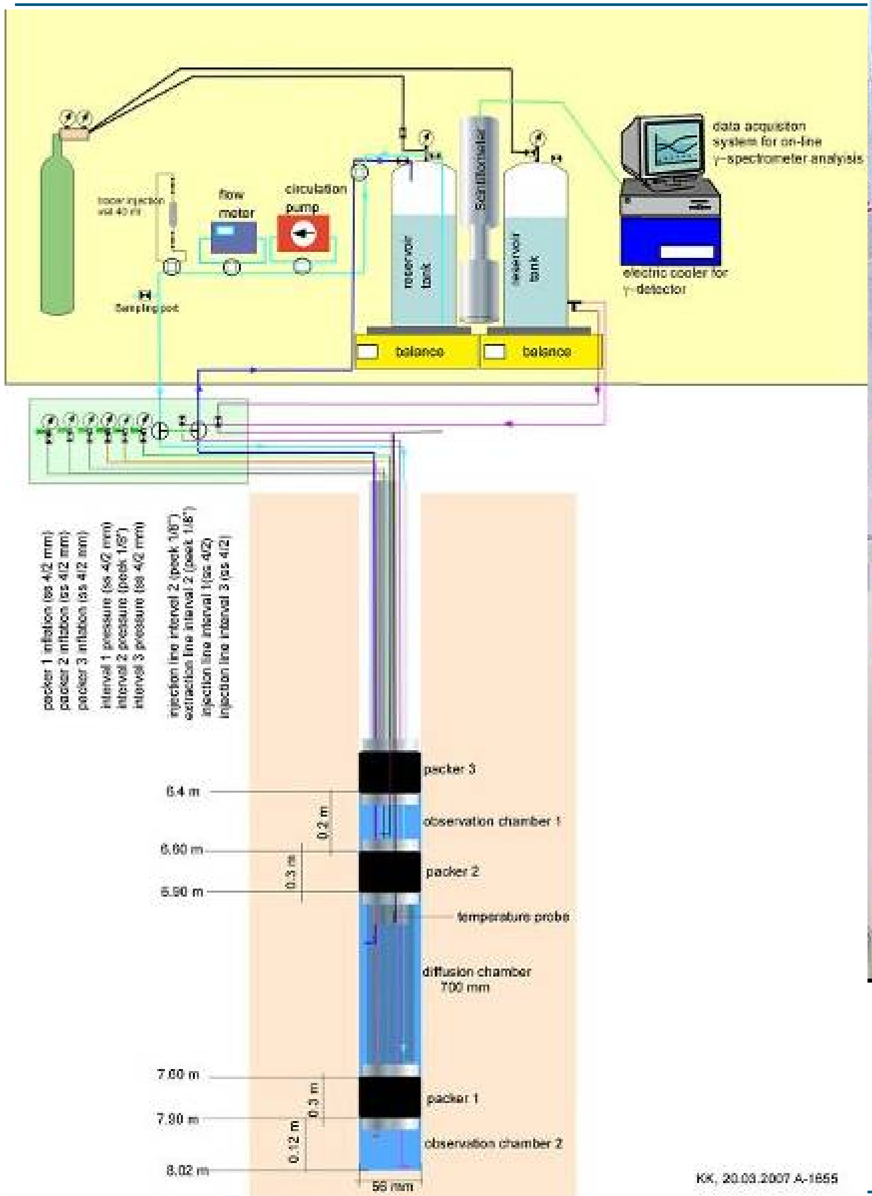
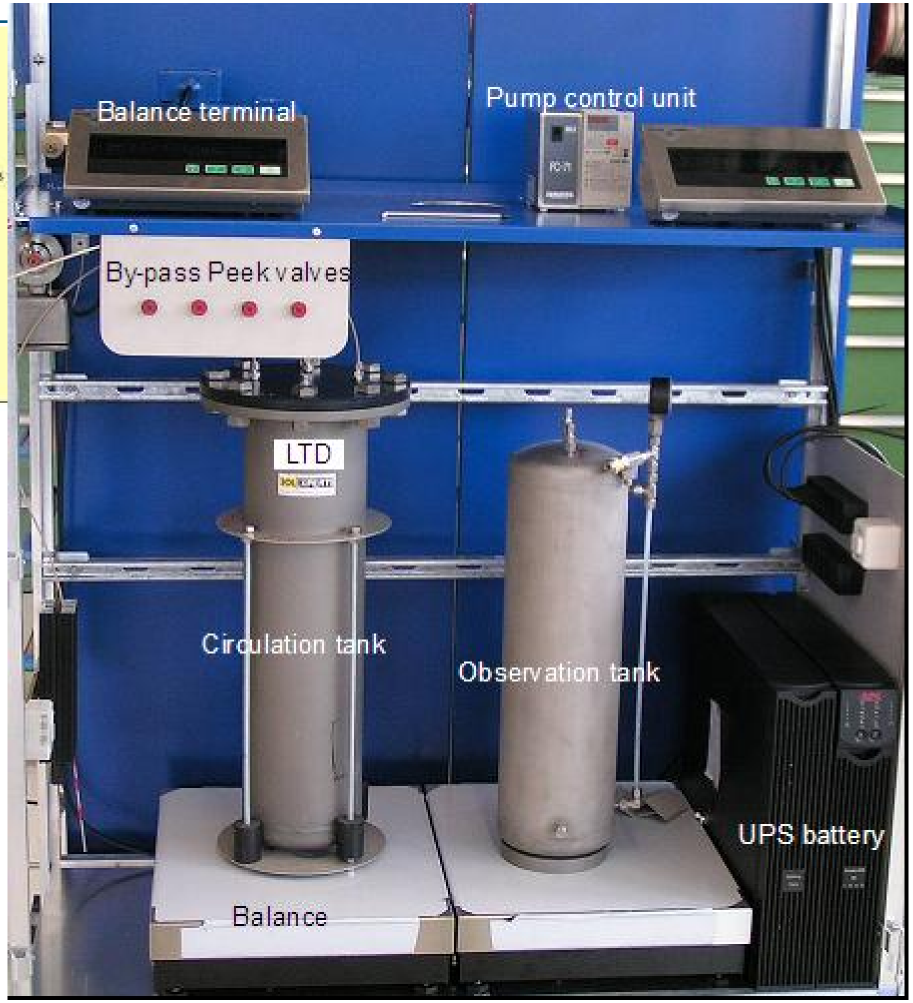
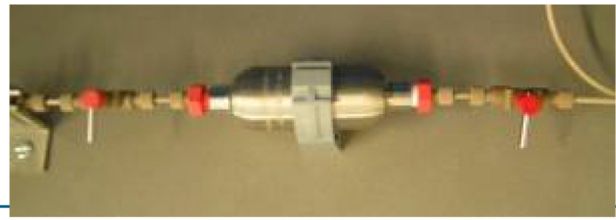
WP-1: MONOPOLE EXPERIMENT







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WP-1: Radionuclide cocktail

RN	Half life	Licensed activity [Bq]	Activity delivered in 40mL vial [Bq]	Justification
^{3}H	12.35 a	1.0E+08	**8,8.0E+07**	Non-sorbing tracer, large diffusion depth, low radiological significance
^{22}Na	2.602 a	5.0E+06	**4,3E+06**	Moderately sorbing tracer, online measurements, optimal t½
^{131}I	8.04 d	1.3E+07	**1.1E+07**	Allows ^{127}I uptake to be monitored for 1st month of experiment. Rapid decay. Easy to measure online
^{134}Cs	2.062 a	5.0E+06	**4,3E+06**	Sorbing tracer for duration of experiment, online measurements, optimal t½

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Predictive modelling

- Modelling teams
  - Institut de Ciències de la Terra “Jaume Almera” (CSIC), Spain
  - Taisei Corporation (supported by JAEA), Japan
  - Nuclear Research Institute Rez, Czech Republik
  - University of Bern, Switzerland

- 4 models general consensus with radionuclide diffusion into rock matrix
- 1-D and 3-D models almost similar results
- H-3 penetrates rock up to 30 cm after 2 years
- Na-22 several centimetres
- Cs-143 first few millimetres (due primarily to sorption)
- I-127 several centimetres

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WP-1: Monitoring

- **Online parameters:**
- Tracer activity in circulation tank
- Pressure in injection and observation intervals
- Mass in surface tanks (balances)

- **Offline sampling**
- Samples collected for PSI analysis (2mL)
- Monthly samples
- Extensive sampling in 1st month (total of 9 samples for 1st month)
- pH, Eh and conductivity

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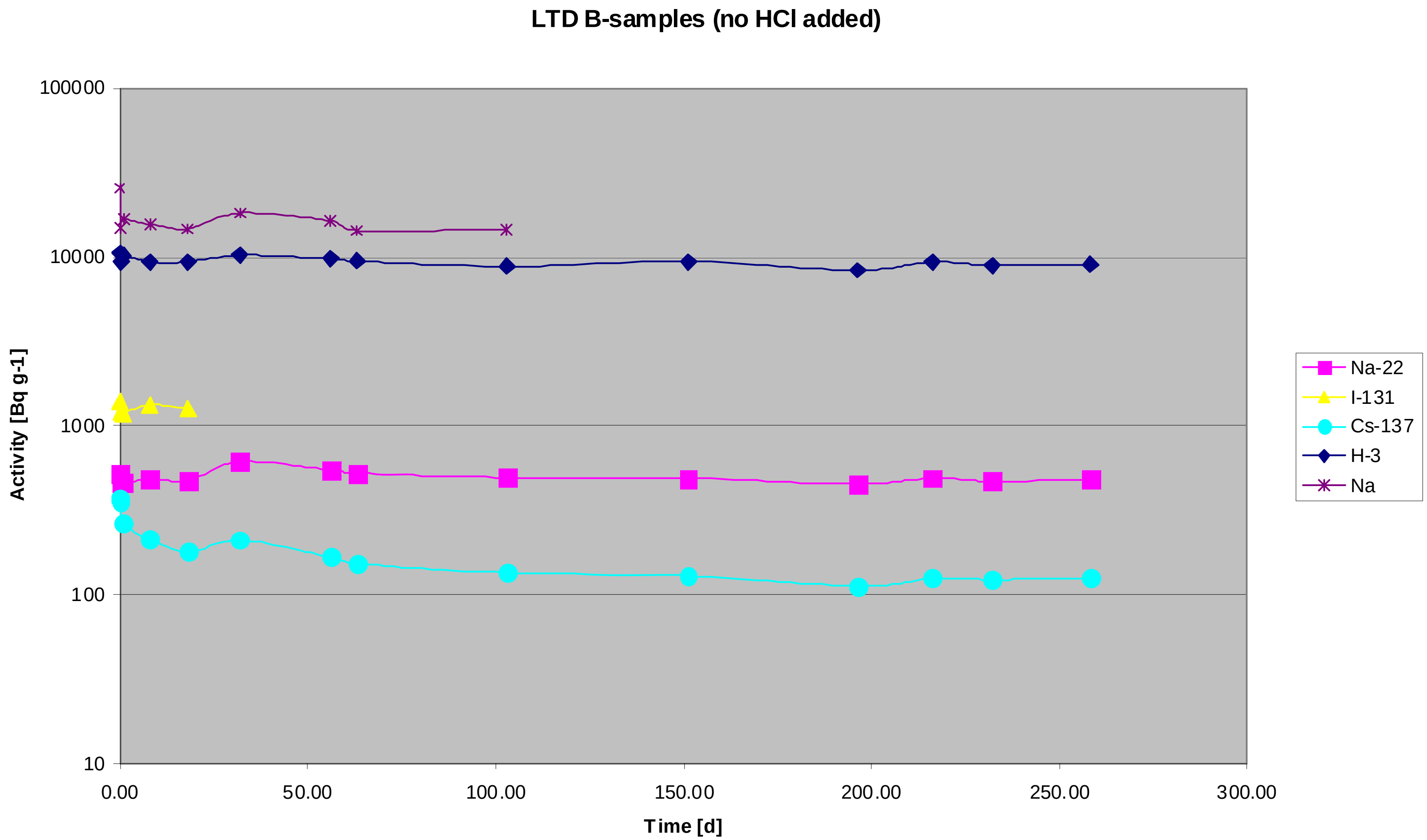
Circulation in borehole started at 8.40am on 7/6/2007

Offline analysis results (until March 2007)



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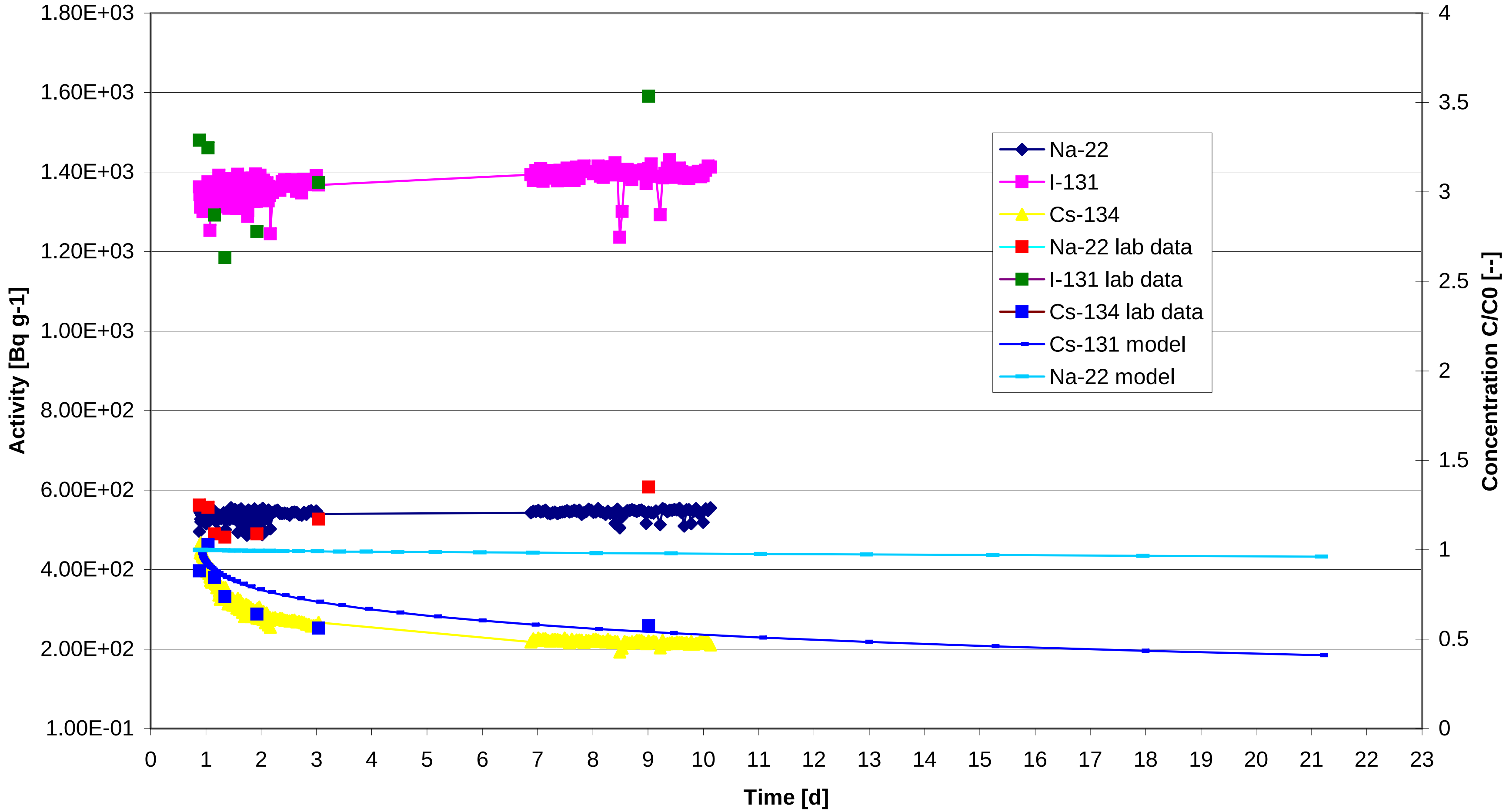
WP-1: Preliminary comparison with model predictions

- Injection into test interval after 0.88 days of tracer circulation



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WP-2: IN-SITU POROSITY (C-14 PMMA and NHC-9)

- Demonstration of an interconnected porosity within the rock matrix and characterisation of the pore tortuosity
- Determination of *in situ* porosity around a water-conducting fracture
- Improve diffusion modelling based on the geometry of matrix pores

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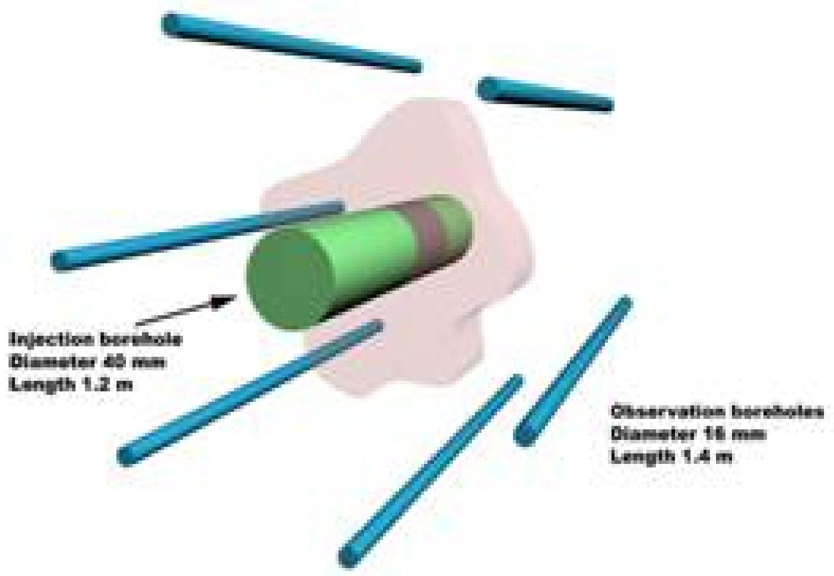
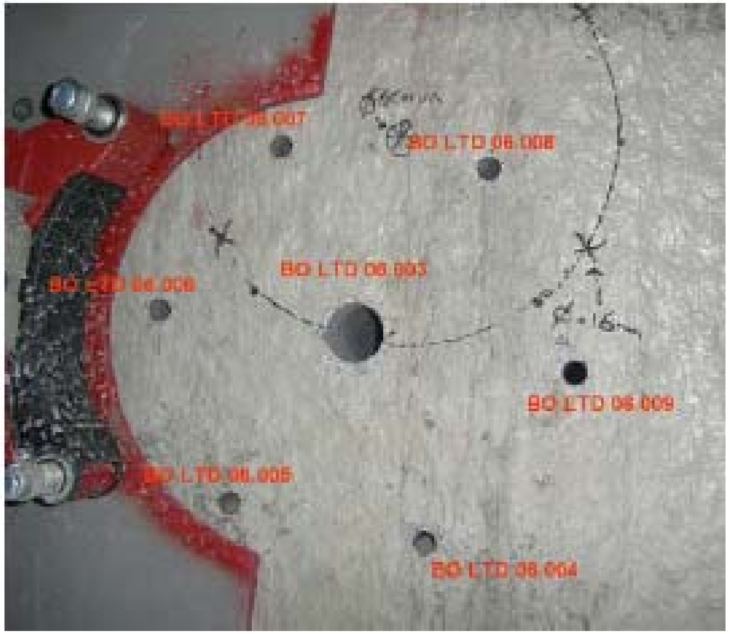
WP-2a: Drilling

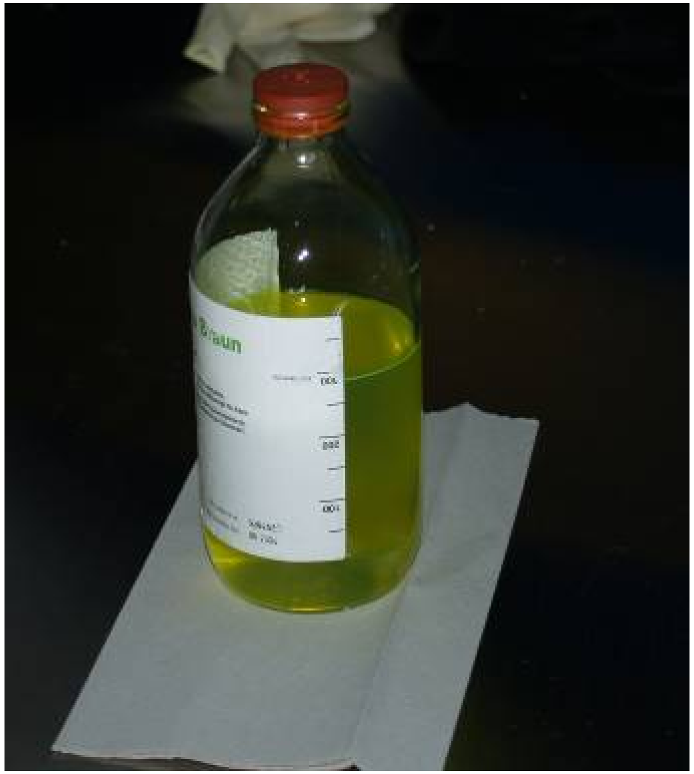
- **7 boreholes drilled:**

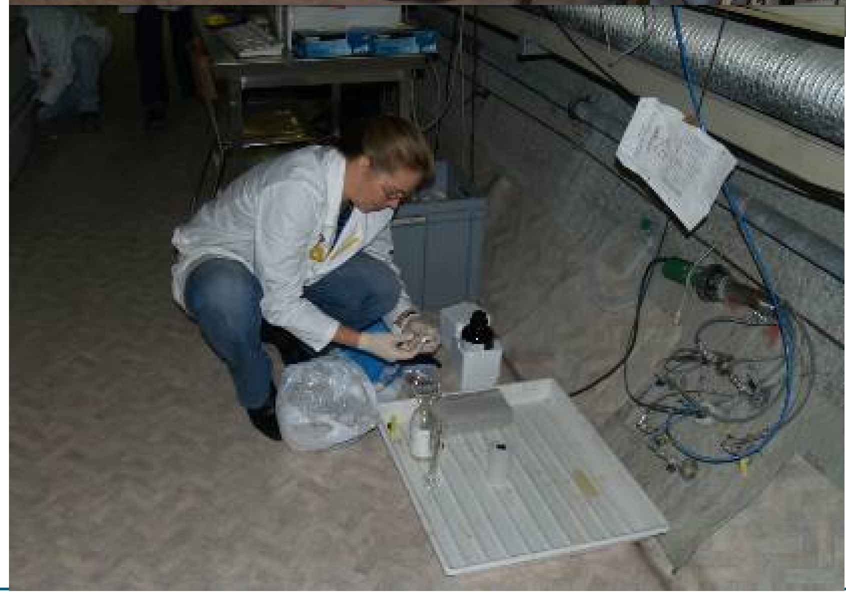
  - Central injection borehole (41 mm)
  - Six observation boreholes (16 mm)





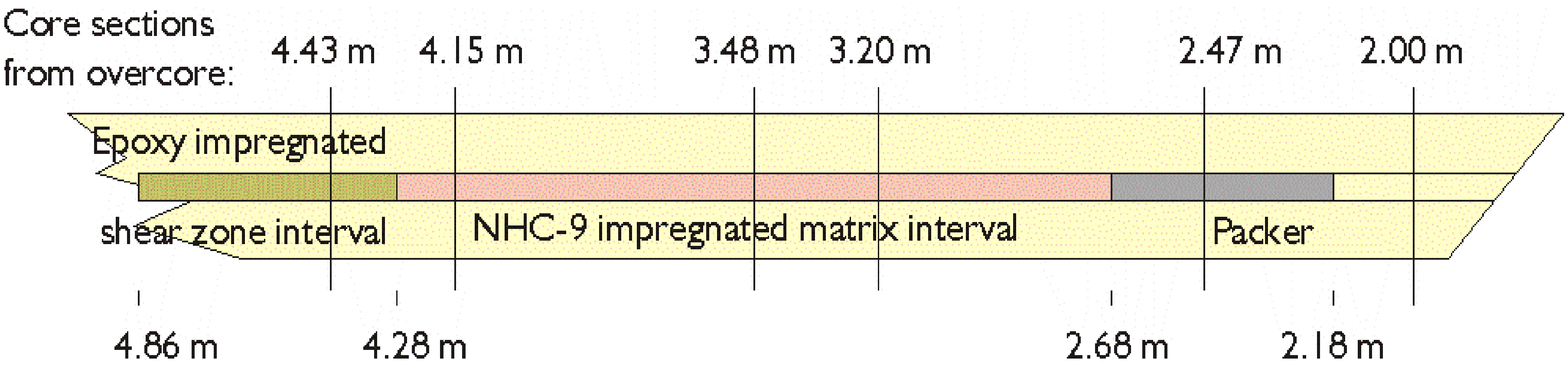
WP-2a: Injection of C14 PMMA resin 14 Nov. 2006





WP-2a: Overcoring

WP 2b: Experimental layout

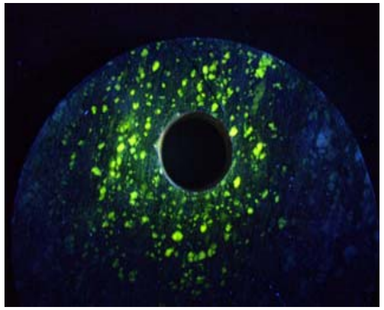


1. NHC-9 resin was injected *in situ* into the rock matrix close to a shear zone
2. Resin polymerisation was achieved by heating
3. Overcoring and sub-sampling took place at the GTS (sawing into slabs)
4. Investigation of slabs under UV light and under the UV microscope
5. Laboratory impregnations of rock samples with NHC-9

WP 2b Concept and aims of chemical porosity determination

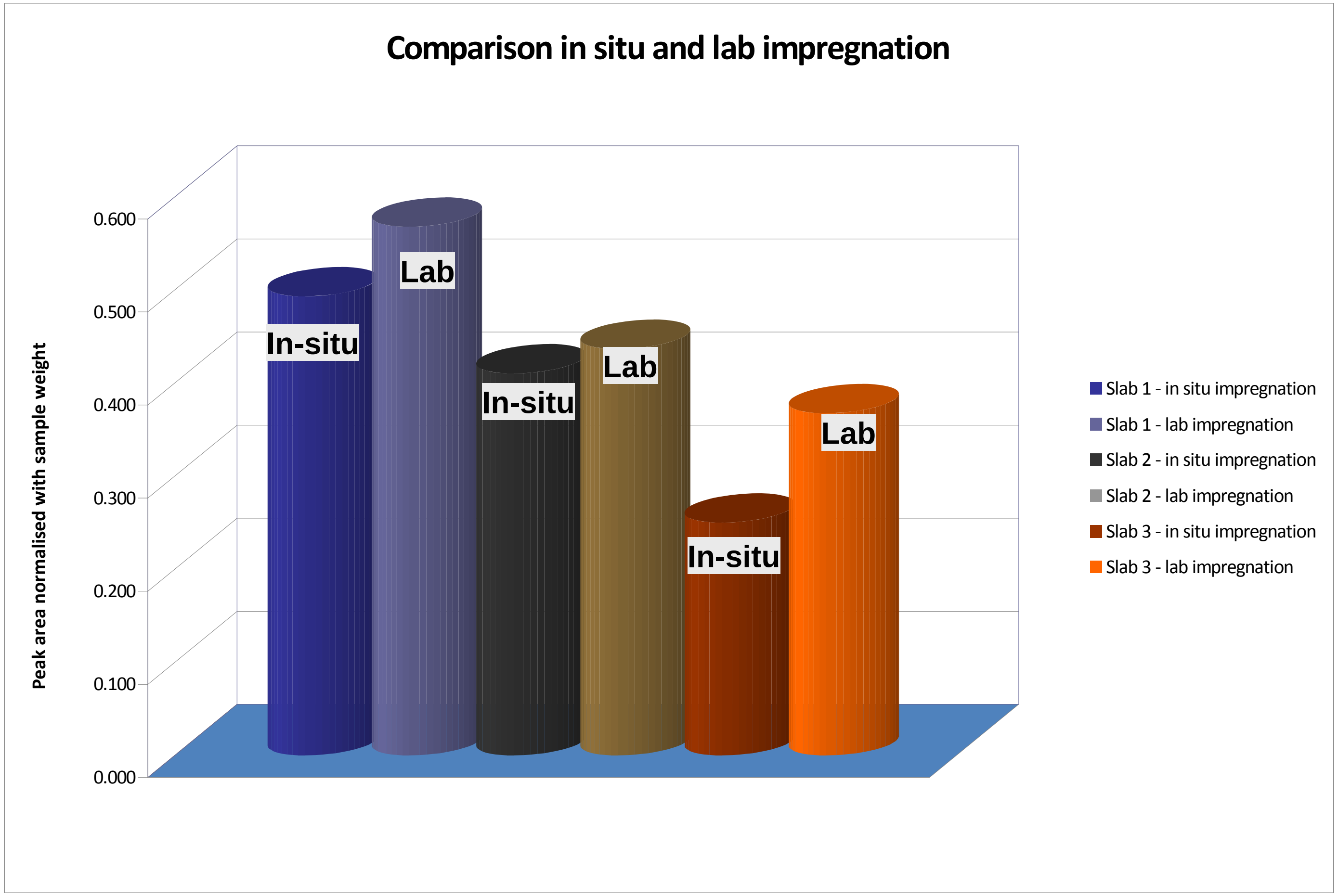
- NHC-9 resin consists of 42 weight% of C
- The Grimsel matrix is almost free of C (confirmed by measurements)
- There is a direct correlation between C content (resin content) and porosity

Element	Atomic mass (g/mol)	Total mass [g]	Weight%
C	12.011	423.822	42.38
H	1.008	85.122	8.51
O	15.999	487.533	48.75
N	14.007	1.881	0.19
S	32.066	0.322	0.03
Br	79.904	1.155	0.12
Na	22.990	0.166	0.02



- Investigate the pore space connectivity in the rock matrix at different distances from a shear zone
- Compare in situ impregnated rock samples with laboratory impregnated rock samples

WP-2b In situ and laboratory impregnated rock samples



- Systematic increase in C (resin) content in samples that were impregnated in the laboratory

WP-3 (Natural tracer study) Core samples

LCS 07.001

Legend

Lithology / ductile deformation of granodiorite / veins > 2 cm:

- Weakly schistous
- Strongly schistous
- Weakly mylonitic
- Mylonite
- Ultramylonite
- Lamprophyre
- Quarz vein > 2 cm
- Cavity

- Fractured zone, core disturbed
- Bad ETIBS quality

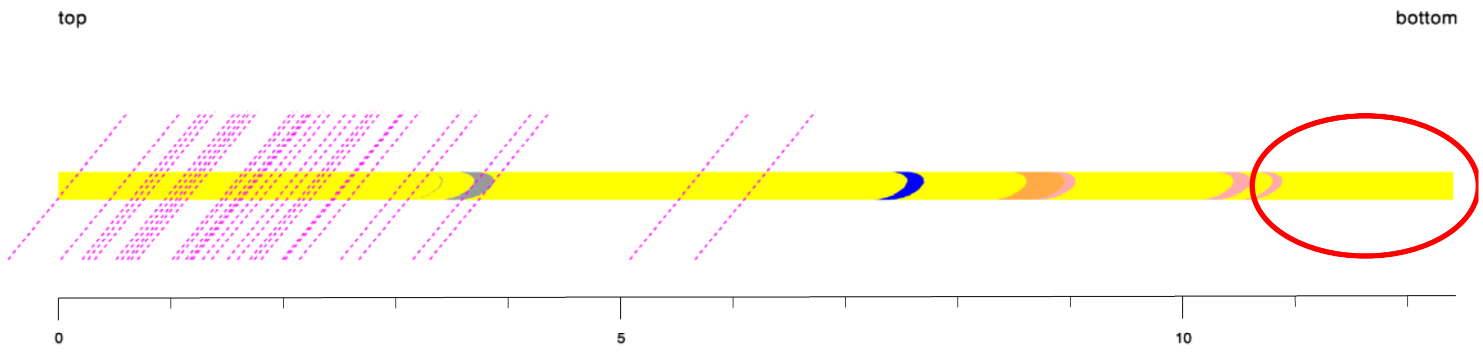
Packer

Fracture width:

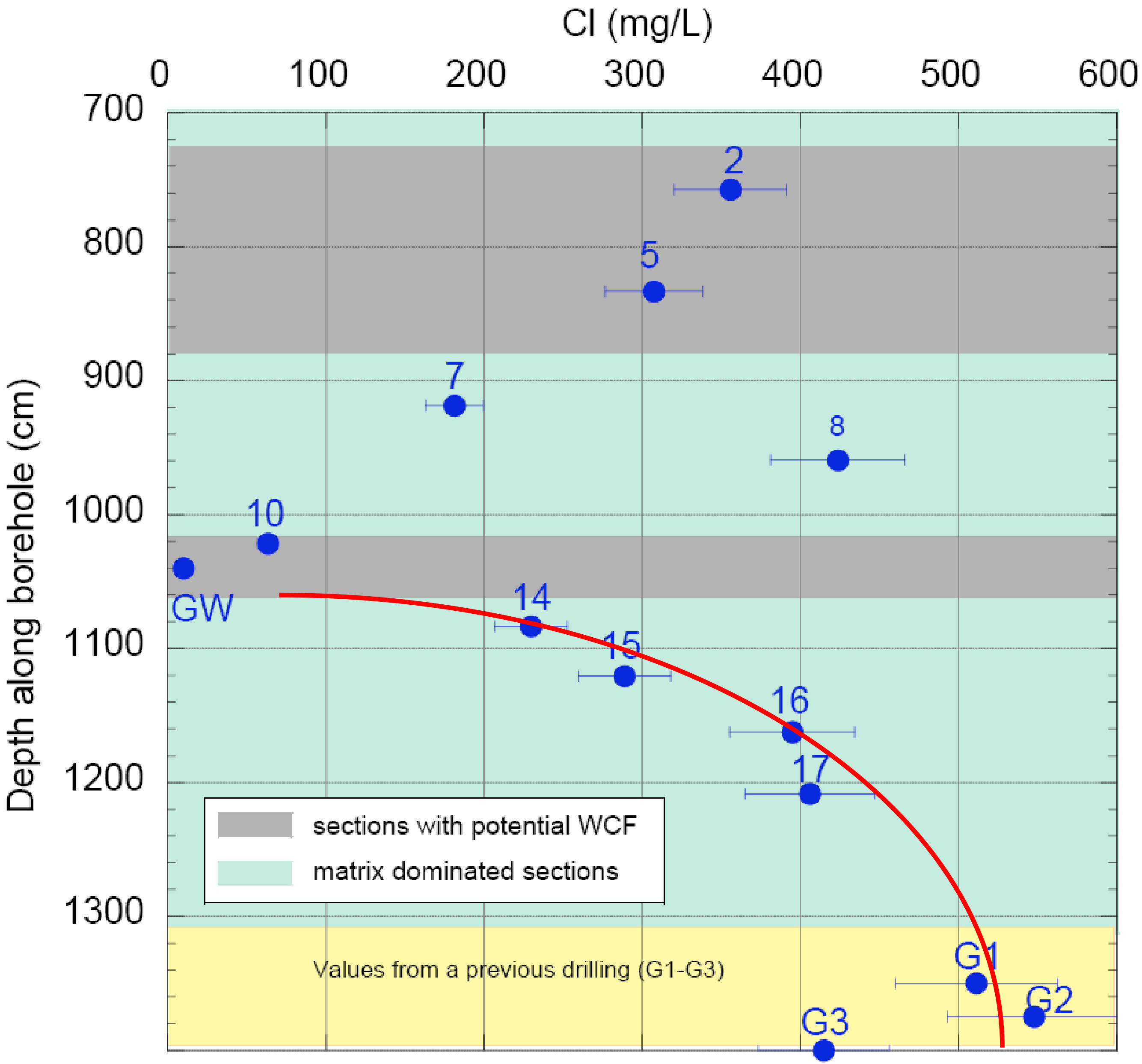
- < 0.1 cm
- 0.25 cm
- 0.75 cm
- > 1.0 cm

Fracture filling:

- no filling observed
- fault gouge
- veins < 2 cm



WP-3 Results: Diffusion profiles



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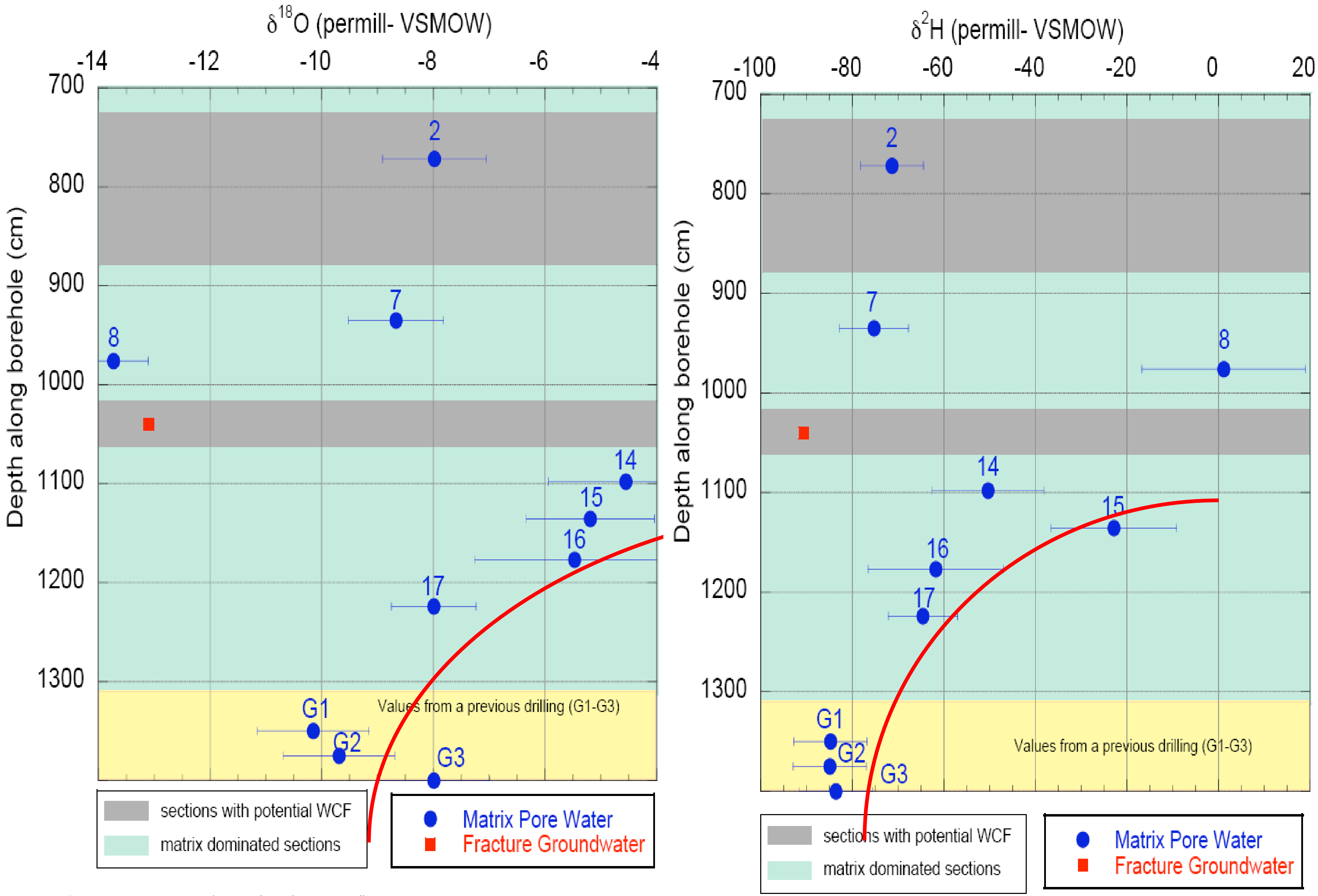
δ18O (permill- VSMOW)
δ2H (permill- VSMOW)
Depth along borehole (cm)
Values from a previous drilling (G1-G3)
sections with potential WCF
matrix dominated sections
Matrix Pore Water
Fracture Groundwater

WP-4: Diffusion Process Study

- **Laser Ablation - Inductively Coupled - Mass Spectrometry (LA-ICP-MS) on solid samples is used in earth sciences to get small scale information of the element distribution in a rock sample**

- **Shows *in situ* matrix diffusion and gives an indication of the retardation sites of radionuclides along the flow path and in the rock matrix**

- **A first study was carried out at LLNL in USA by Max Hu (measurements and lab experiments) and Andreas Möri (sample preparation and interpretation of data)**
  Ref: Radionuclide transport in fractured granite interface zones by Q.H. Hu & A. Möri. Physics and Chemistry of the Earth, 2008.

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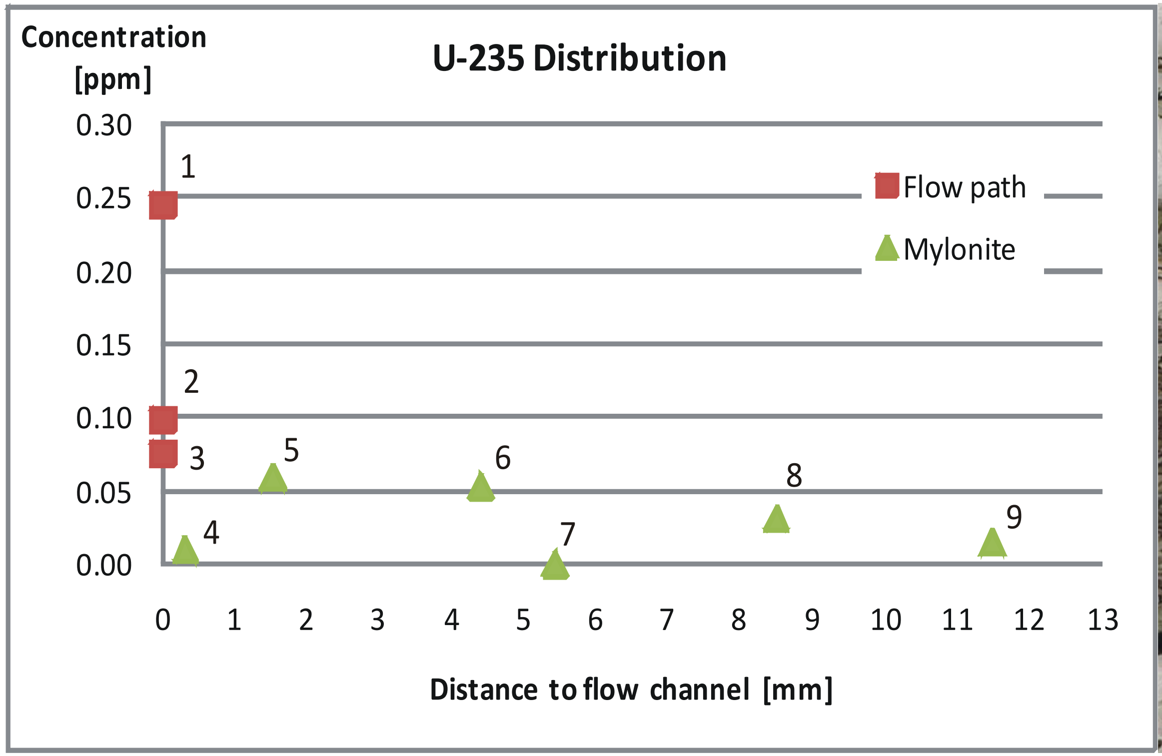
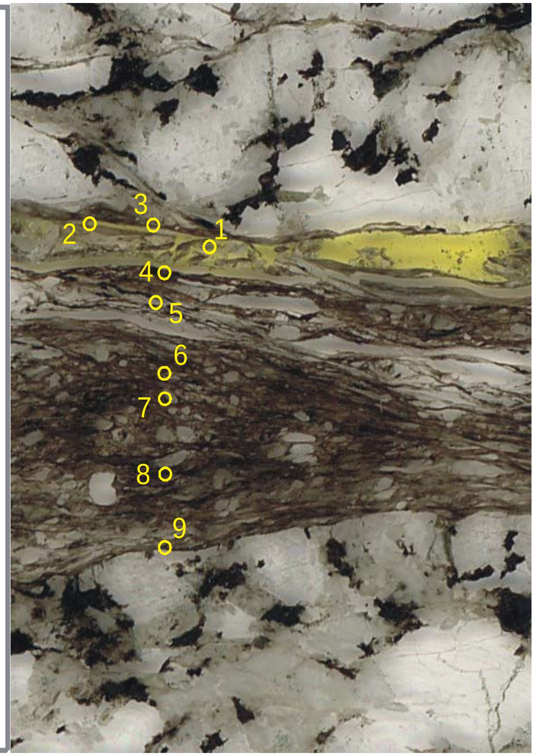
^{235}U profile in mylonite





- **High activity in flow channels (hot spots; pts. 1, 2 & 3)**
- **Low activities in measurements within biotite bands (pts. 4 and 7)**
- **Flat decrease in ^{235}U concentration within mylonite**

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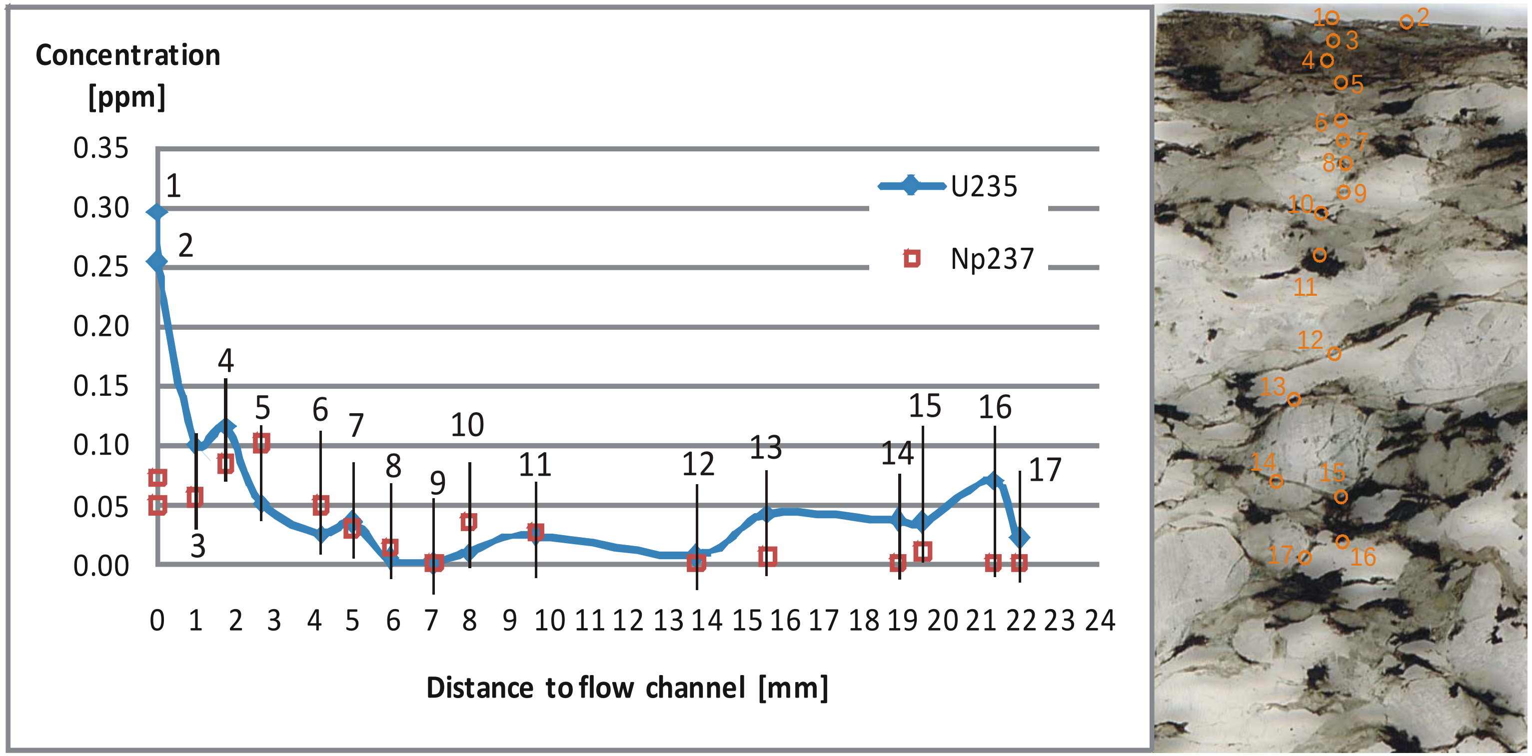
^{235}U and ^{237}Np diffusion profile in matrix



- ^{235}U: a strong decrease in concentration can be observed in the first 7mm. Below this decrease there is still some uranium in the rock
- ^{237}Np: Starting concentration at the flow channel is smaller than in case of ^{235}U. A peak can be observed after about 5 mm; the tailing is much lower that in case of ^{235}U

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Phase I reporting and synthesis

- Will start soon

PHASE II Experiments

- **WP 6a** Extension of existing monopole (total circulation for at least 2 years)

- **WP 6b** New monopole circulation with other heavier / safety relevant radionuclides / coupled with a natural tracer study

- **WP 7** Mock-up tests consisting of overcore with a shear zone

- **WP 8** Diffusion from a shear zone into the rock matrix (non-active and active tracers)

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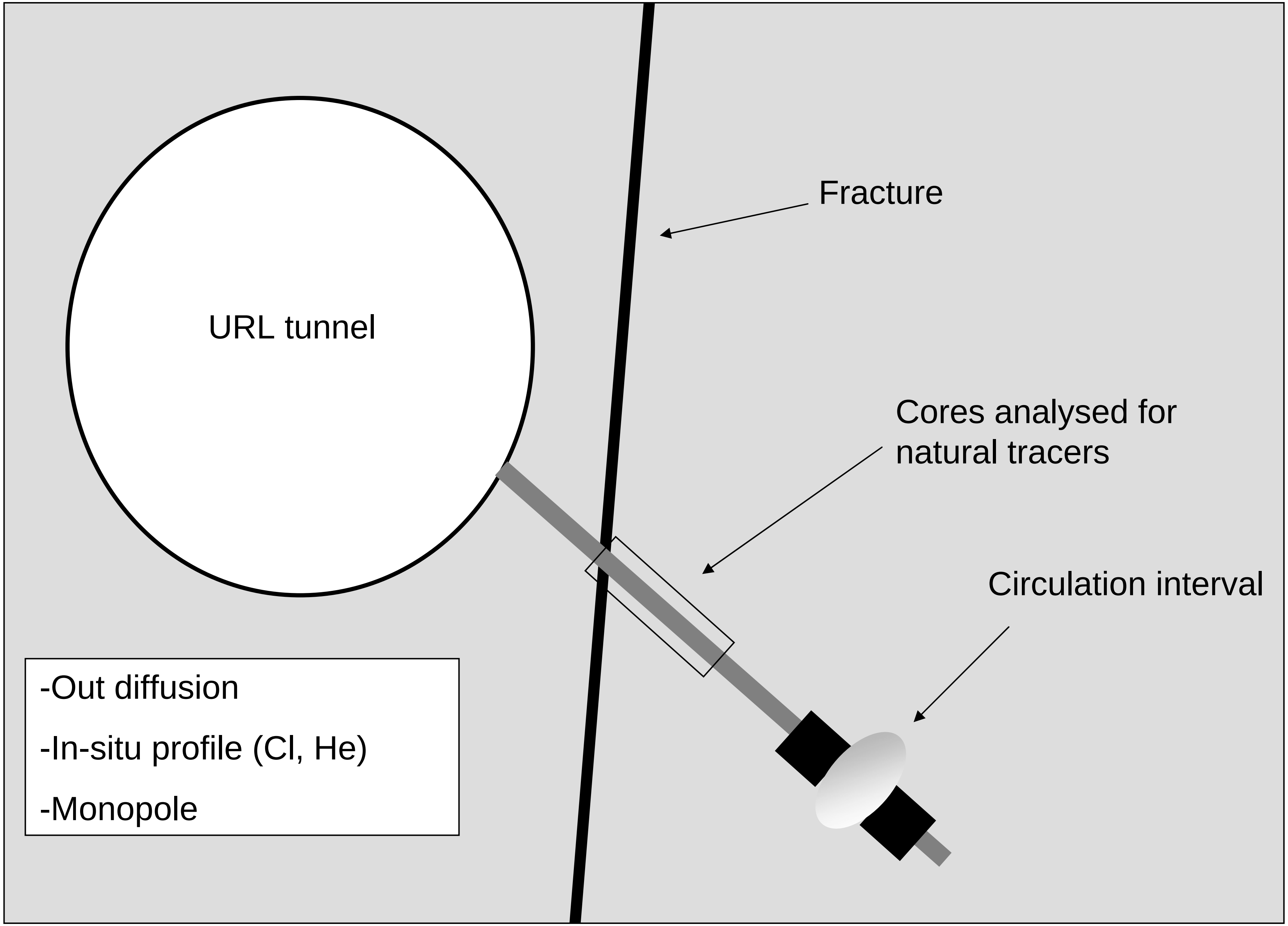

-Out diffusion

-In-situ profile (Cl, He)

-Monopole

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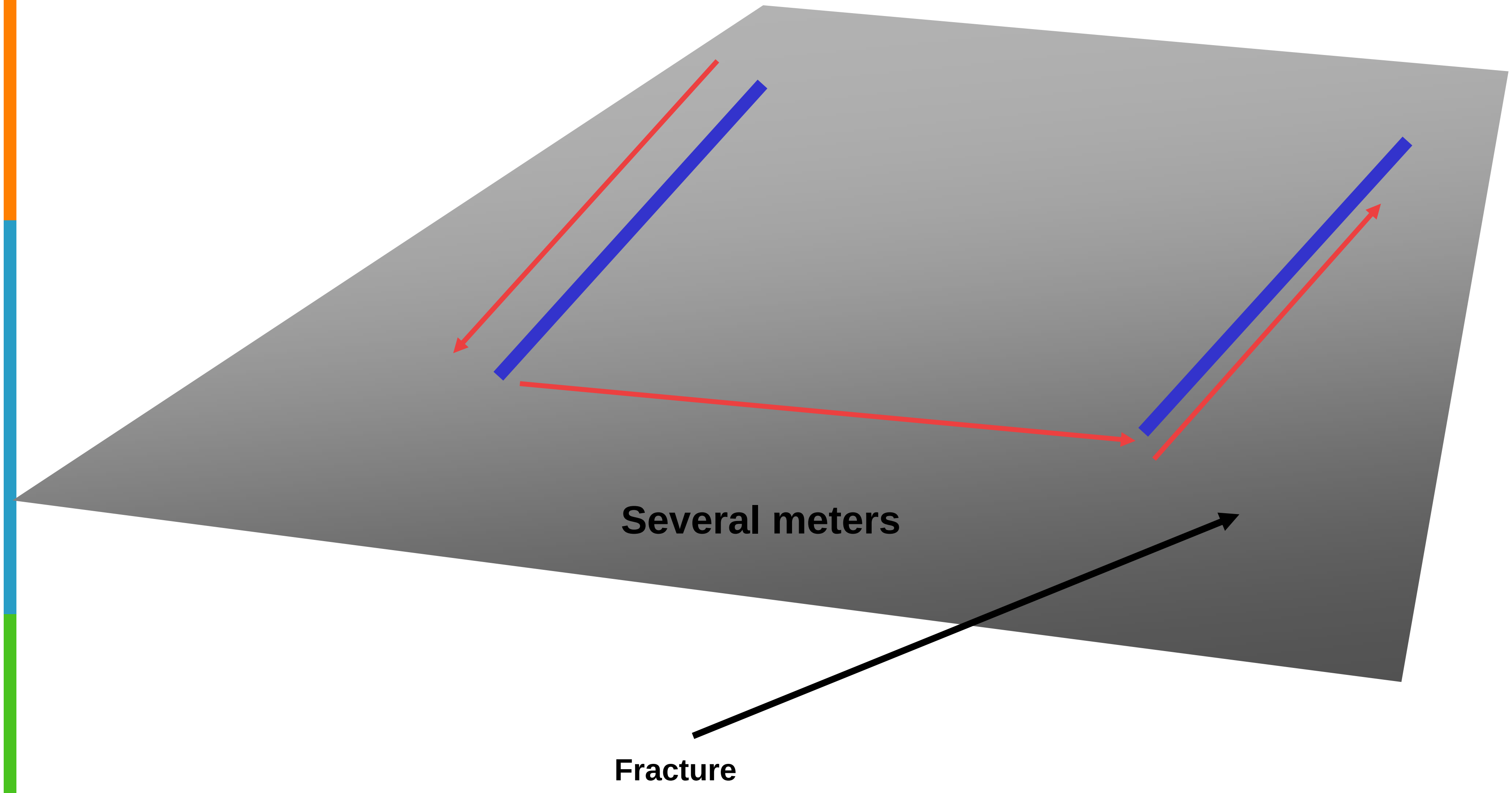
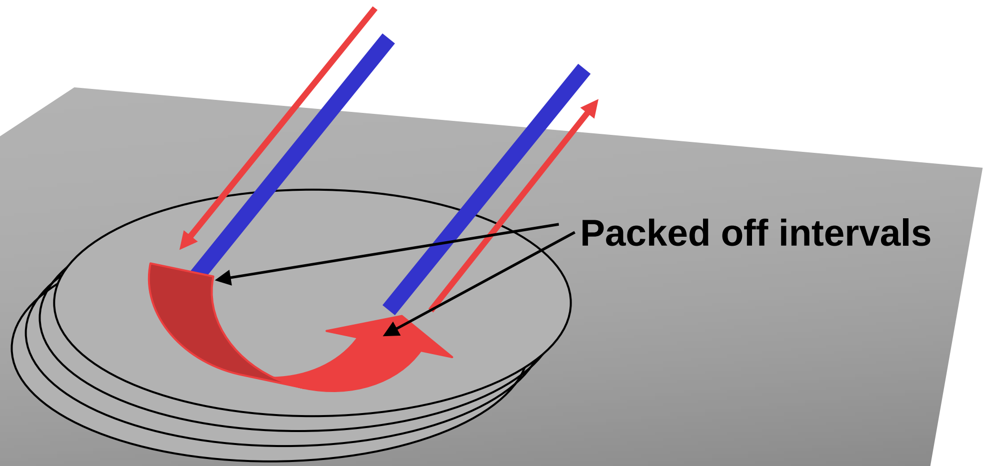
Mock-up test

Use of non-active tracers in open fracture



Long term in-situ diffusion experiment in a fracture

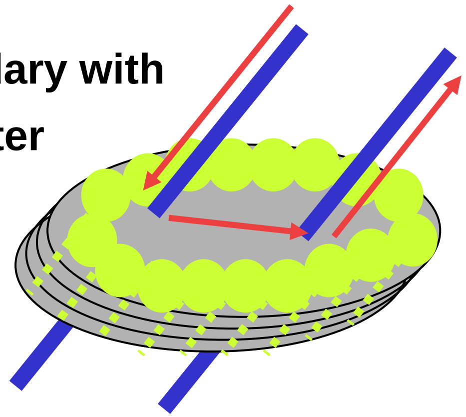
- **Advection** along preferential pathways in fracture
- **Dispersion/diffusion** in the fracture
- **Absoption** in the fracture
- **Diffusion** into the rock matrix (across the interface)



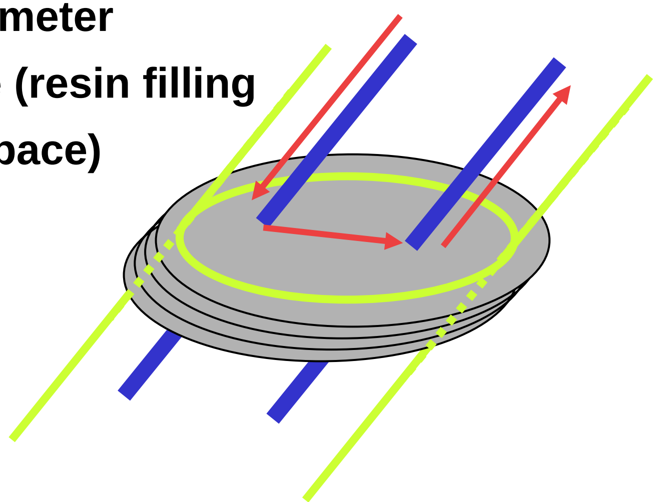
Tracer circulation within a fracture

Options for controlling flow and preventing loss of radionuclides in a fracture

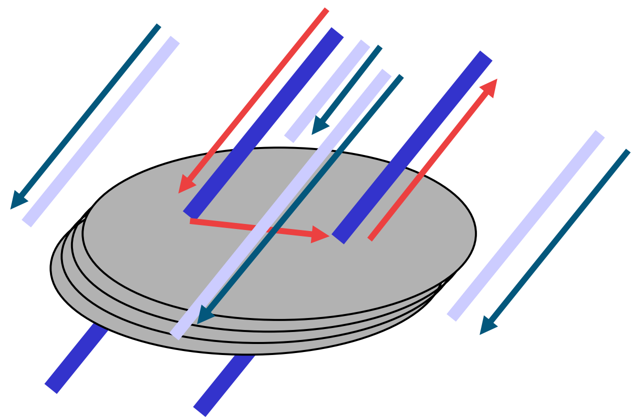
a) Resin boundary with small diameter boreholes



b) Resin boundary with large diameter borehole (resin filling of ring space)



c) Tracer loss prevented by applying external hydraulic pressure via 4 or more boreholes



Summary Phase I

- **WP-1** Predictive modelling complete, monopole circulation and water sampling ongoing -> extension to at least 2 years
- **WP-2** All field activities completed, predictive modelling of tracer injection complete, laboratory analyses of overcores ongoing at Universities of Helsinki and Bern
- **WP-3** Field activities and analyses completed -> reporting ongoing
- **WP-4** All activities completed -> 2 papers published
- WP-5 Synthesis report of all data in Phase I

Phase II planning

- WP-6a Extension of monopole for at least 2 years
- WP-6b New monopole to circulate heavier radionuclides (U,…)
- WP-7 Laboratory mock-up tests
- WP-8 In-situ experiment involving a fracture (feasibility study)
- WP-9 Final synthesis report (Phases I and II)