Gas generation and potential impact on repository performance

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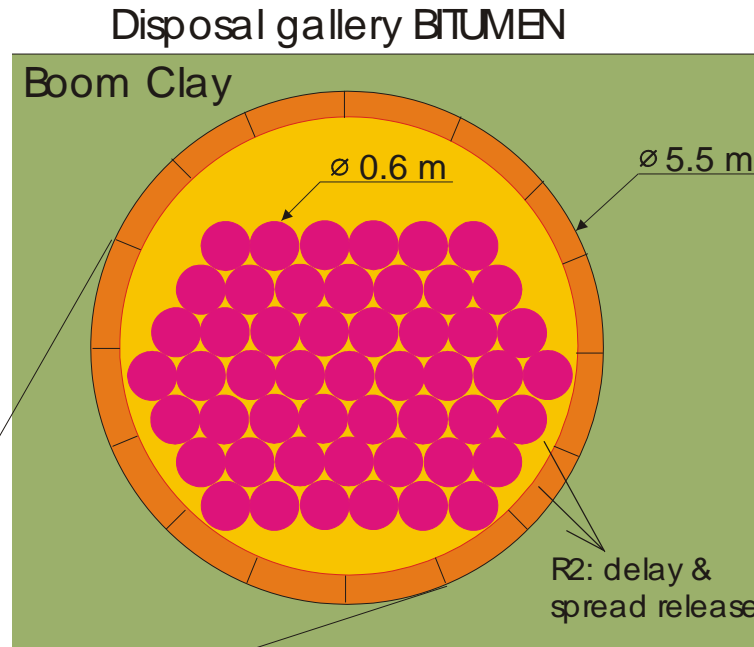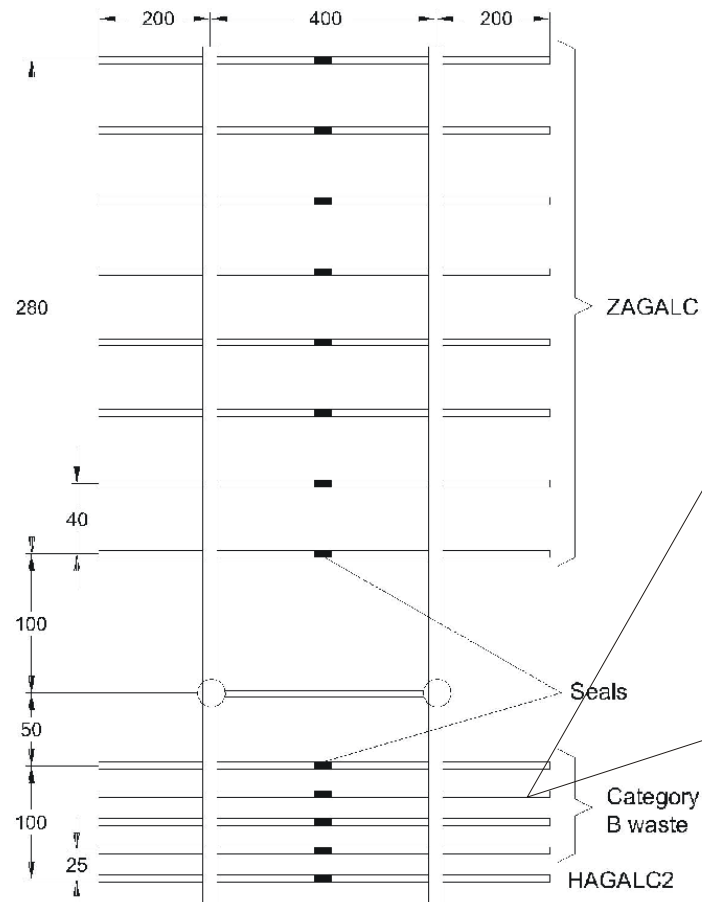# Perturbation of the R2 safety function by gas generation and transport ?

- Large amounts of H_2 gas generated due to anaerobic corrosion of steel in disposal gallery

- More gas generated than diffusive transport via Boom Clay=> excess gas in gallery

- "Growing gas phase"
  - ⇒ Water from gallery is expelled ("pushed") into Boom Clay
  - ⇒ Gas pressure too high (= local total pressure): sudden gas break-through via micro fissures (preferential flow paths) in Boom Clay
  - ⇒ Gas pressure drops after gas has been evacuated

⇒ Perturbation of safety function R2 (delay and spread release)?
  - ⇒ Will free gas phase exist or not?
  - ⇒ Expelled water: contaminated with radionucliden (timing)?
  - ⇒ Is gas breakthrough combined with water (and RN) transport?
  - ⇒ Are micro fissures permanent, or close again after pressure drops below breakthrough pressure?
  - ⇒ Is the safety function "delay and spread release" bypassed?

Contents

- Sources of gas generation
- Hydrogen gas generation
  - Principles, amount, rates
- Lab and in-situ gas experiments
  - Are pathways permanent after pressure drop ?
- Modelling gas transport
  - Diffusive transport in Boom Clay (free gas phase?)
  - Two-phase flow in near field (timing and volume of expelled water?)
- Conclusions

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Disposal concept-EUROBITUM

Disposal gallery BITUMEN

Boom Clay

∅ 0.6 m

∅ 5.5 m

R2: delay & spread release

ZAGALC

280

40

100

Seals

50

Category B waste

100

25

HAGALC2

200   400   200

- **Conditioned in 220 L drums**
  - **7590 Stainless steel**
  - **5816 Cr-Carbon steel (90% Fe)**

- **Former disposal concept: 50 drums/m**

  gallery (14 or 34/m in new concept)
  - **~300 m gallery**
  - **grout backfill**

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Sources of gas generation (1)

1. ## Radiolytic gas generation (Valcke et al., 1998)

   ➢ H_2 is the most important radiolytic gas

   ➢ Highest contribution from α-irradiation

   ➢ Contribution of β/γ-irradiation is negligible

   ➢ 0.1-6 m³ (avg=3 m³) H_2 per drum of 216 kg after 100.000 years (0.03 dm³/drum/y)

   ➔ Very small volume of gas generated

Sources of gas generation (2)

2. Microbial gas generation

- Bitumen:
  - ♣ Very difficult to make reliable estimates
  - ♣ Production and consumption of gases (N_2, N_2O, CO_2, CH_4)
  - ♣ 8 dm³/drum 1st year, < 1 dm³/drum/y after 40 y
 - ➔ Small volume of gas generated (anaerobic cond.)
- Nitrate (Ortiz, 2004):
  - ♣ Denitrification (generation of N_2O)
  - ♣ At present only qualitative results (unlikely to be of importance under disposal conditions)
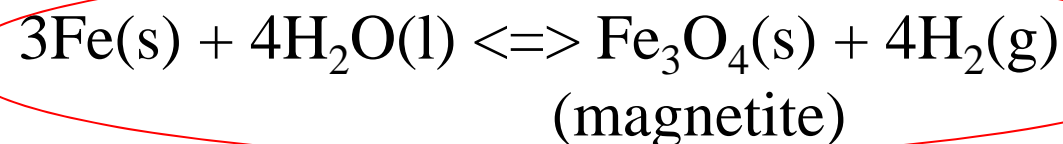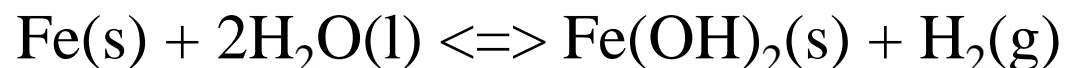
3. Anaerobic corrosion of steel (package!)

- ~**12.2** m³ per drum (carbon steel) (20 dm³/drum/y)
- ➔ Most important (H_2) gas generation process

H$_2$-gas generation: principles & mechanisms

- During aerobic phase of repository (operational phase & first few years after closure): aerobic corrosion

$$4Fe(s) + 3O_2(g) + 6H_2O(l) <=> 4Fe(OH)_3(s)$$
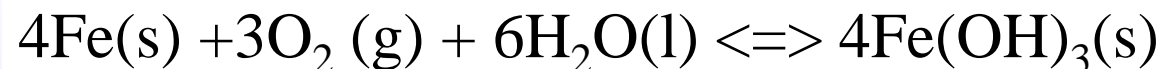
- When repository becomes anaerobic: anaerobic corrosion of iron

$$Fe(s) + 2H_2O(l) <=> Fe(OH)_2(s) + H_2(g)$$

$$3Fe(s) + 4H_2O(l) <=> Fe_3O_4(s) + 4H_2(g)$$
$$(magnetite)$$

➢ 1 mole iron => 4/3 mole H$_2$ (magnetite, pH > 7)

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Gas generation: quantities

- Inventory
  - Fe (C-steel drums) ~ 134 ton
  - Fe (Stainless steel drums) ~ 99 ton

- 1 ton Fe => max 530 m³ STP hydrogen gas
  - C-steel : total ~ 7.09×10^4 m³ STP H_2
  - Stainless steel: total ~ 5.2×10^4 m³ STP H_2

Gas generation: rates (H_2)

- Anaerobic corrosion rate C-steel
  - based on lab and in-situ experiments (Boom Clay; pH = 8.2; Eh=-250 mV; ionic conductivity=1.8 mS/cm):
  - literature (Agg, 1993)
    - pH>8.5 (in cement environment): 0.1 - 1 μm /y;
    - pH<7: max = 1 – 10 μm /y
  - best estimate **1 μm/y** (range 0.2 to 2 μm/y)
  - gas generation rate = 41 mol/m/y (former design)
    (LLW: 50 mol/m/y)
  - gas generation during ~700 years

- Stainless Steel (AISI 316 L) **< 0.05 μm/y**
 - gas generation during ~10 000 years
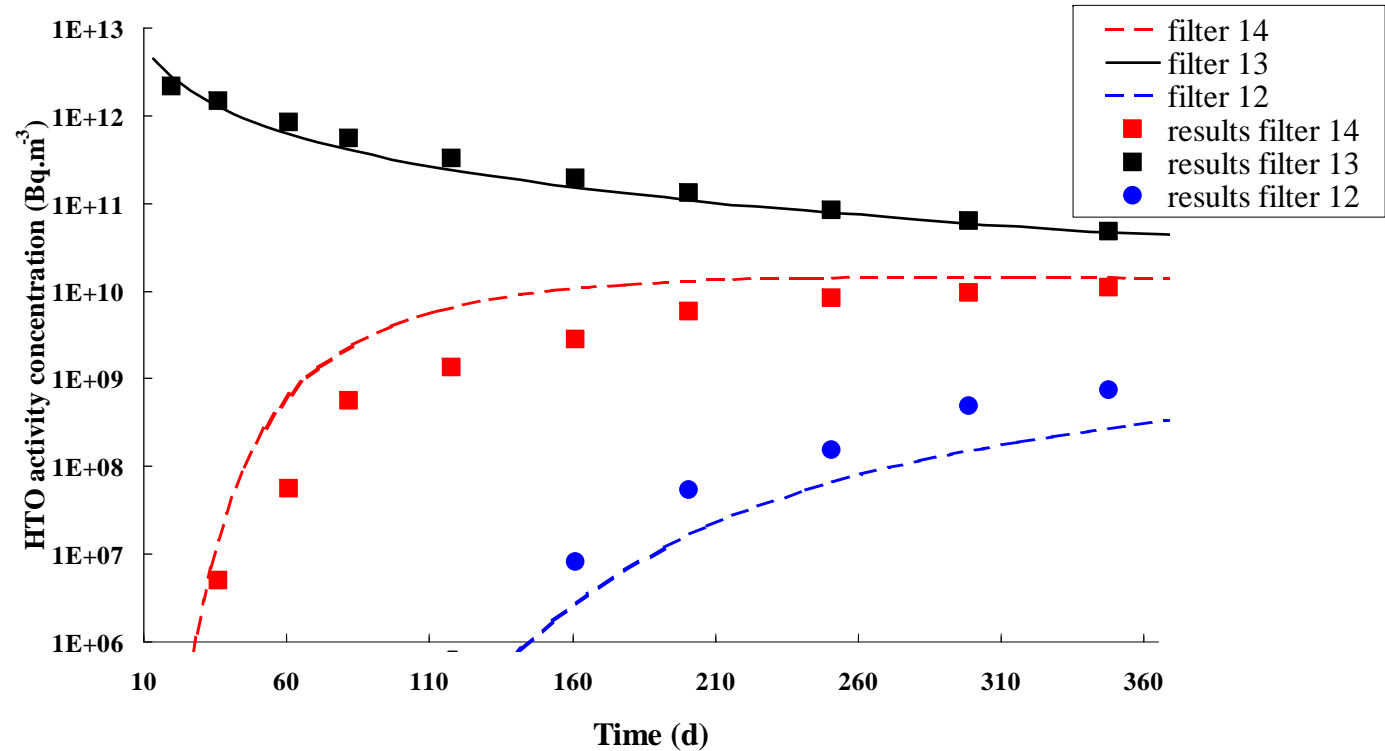
Gas transport: in-situ experiments

- ## In-situ (HADES): MEGAS E5

Observation filter

C

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A

B
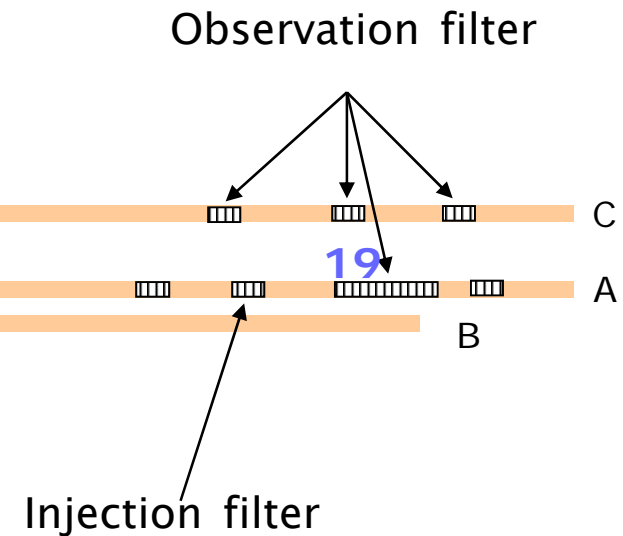
Injection filter

- Gas breakthrough after 1 month ~2.36 MPa (filter 19) (4.4 MPa theor.)
- Continuous gas pathway at breakthrough: $P_{\text{injection}} = P_{\text{filter}}$

Gas transport: in-situ experiments
Tritium injection (MEGAS experiment)

- Injection of water (tritium) after end of gas injection

⇒ **Clay closes complete No preferential flow of water (micro-fissures have closed)**
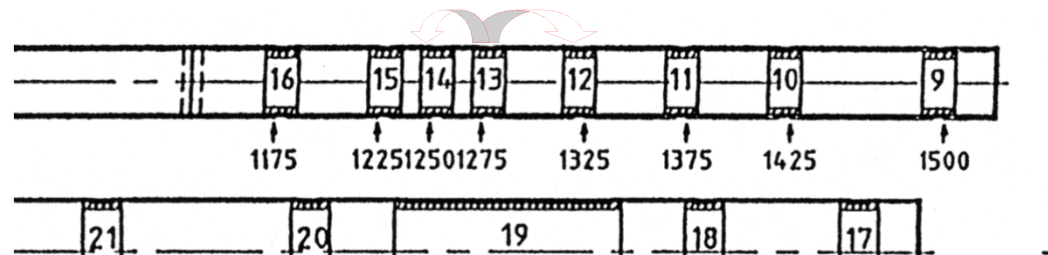
Experimental evidence: gas transport through Boom Clay

- **Advective gas flow: laboratory and in situ experiments**
  - ➤ Breakthrough when gas pressure = total pressure in Boom Clay
  - ➤ Formation of preferential pathway (gas flow)
  - ➤ Breakthrough is geomechanically controlled
  - ➤ Desaturation at breakthrough = few %
 - ➤ Self-healing after stopping gas injection

Modelling gas transport:
Theoretical background

- ## For initially water-saturated Boom Clay, three types of gas transport may be identified:

  - transport of dissolved gas molecules according to Fickian <u>diffusion principle</u> (no free gas phase present)
  - <u>two-phase flow</u> according to Darcy's law (gasflux is depending on the relative gas permeability) assuming a partial desaturation of the clay (free gas phase present)
  - flow of gas along <u>preferential pathways</u> (non-Darcy flow) created by excess gas pressures (free gas phase present)
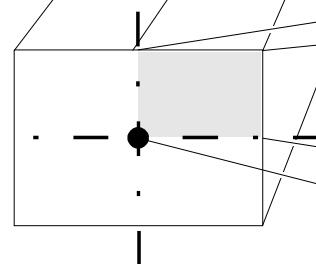
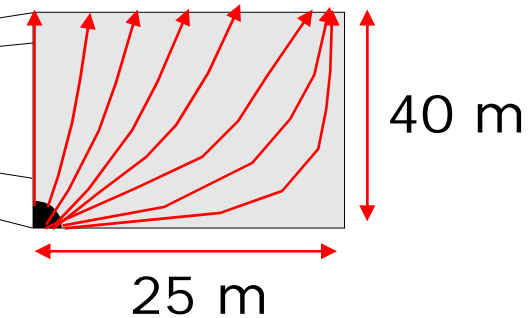# Modelling gas migration in Boom Clay: conceptual model for diffusive transport

a) 3D world

Clay layer

Disposal galleries

b) 2D model

c) 1/4 2D model

40 m

25 m

- Simplified BC model

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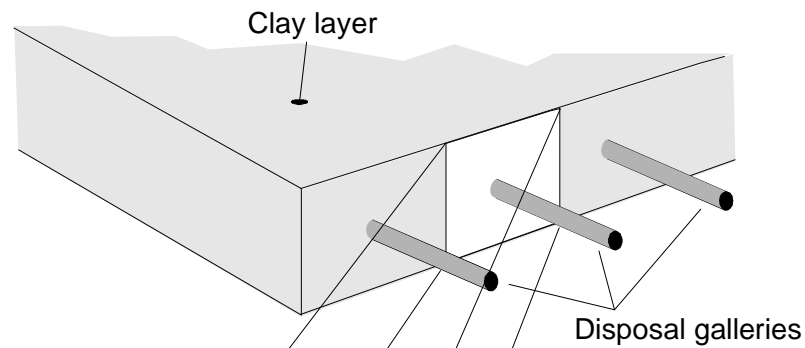
Gas production & diffusive gas transport in Boom Clay

- Microbial conversion:

$$CO_2(aq) + 4H_2 <=> CH_4(aq) + 2H_2O(l)$$

- Former

design (conservative)!

- Gas production >>

gas transport => free gas phase

EUROBITUM (MAGALE)

Y-axis: **Cumulative gas production and transport (mole/m)**

X-axis: **Time (a)**

Legend:
- D = 8E-11 m²/s , P=2.2MPa
- D = 5E-10 m²/s , P=2.2MPa
- D = 8E-11 m²/s , P=4.4MPa
- D = 5E-10 m²/s , P=4.4MPa

Curve labels: H_2 (CS), CH_4 (CS), H_2 (SS), CH_4 (SS)

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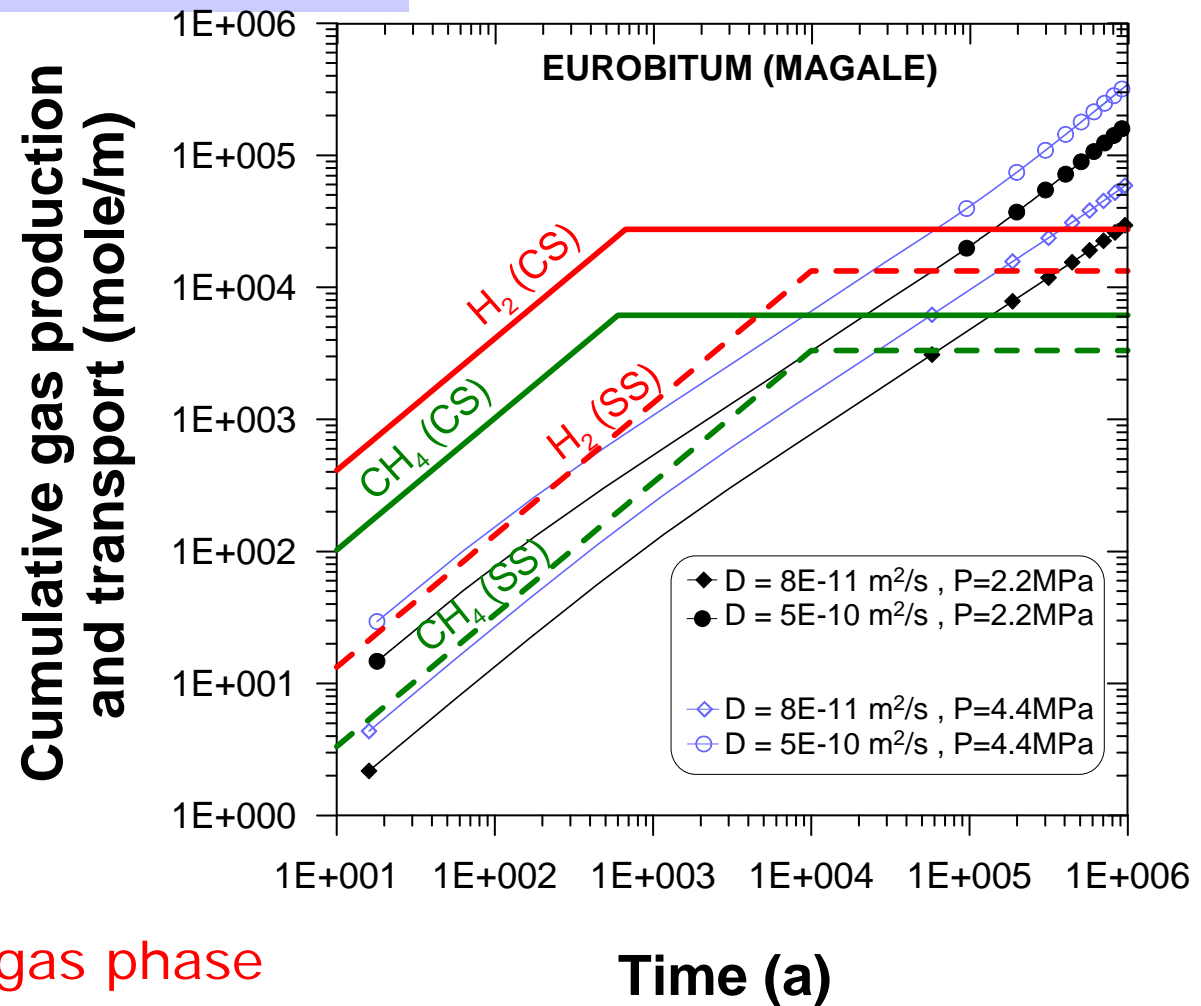
Gas transport modelling: Two-phase flow

☺ Two-phase system: liquid-gas

  ➢ Unsaturated poreus medium

  ➢ Saturation degree - capillaire pressure relations for each poreus medium (host rock, engineered barriers)

  ➢ Transport of water & gas => saturation degree -relative permeability relations for each poreus medium (for water & gas!)

☹ Gas production coupled to water availability
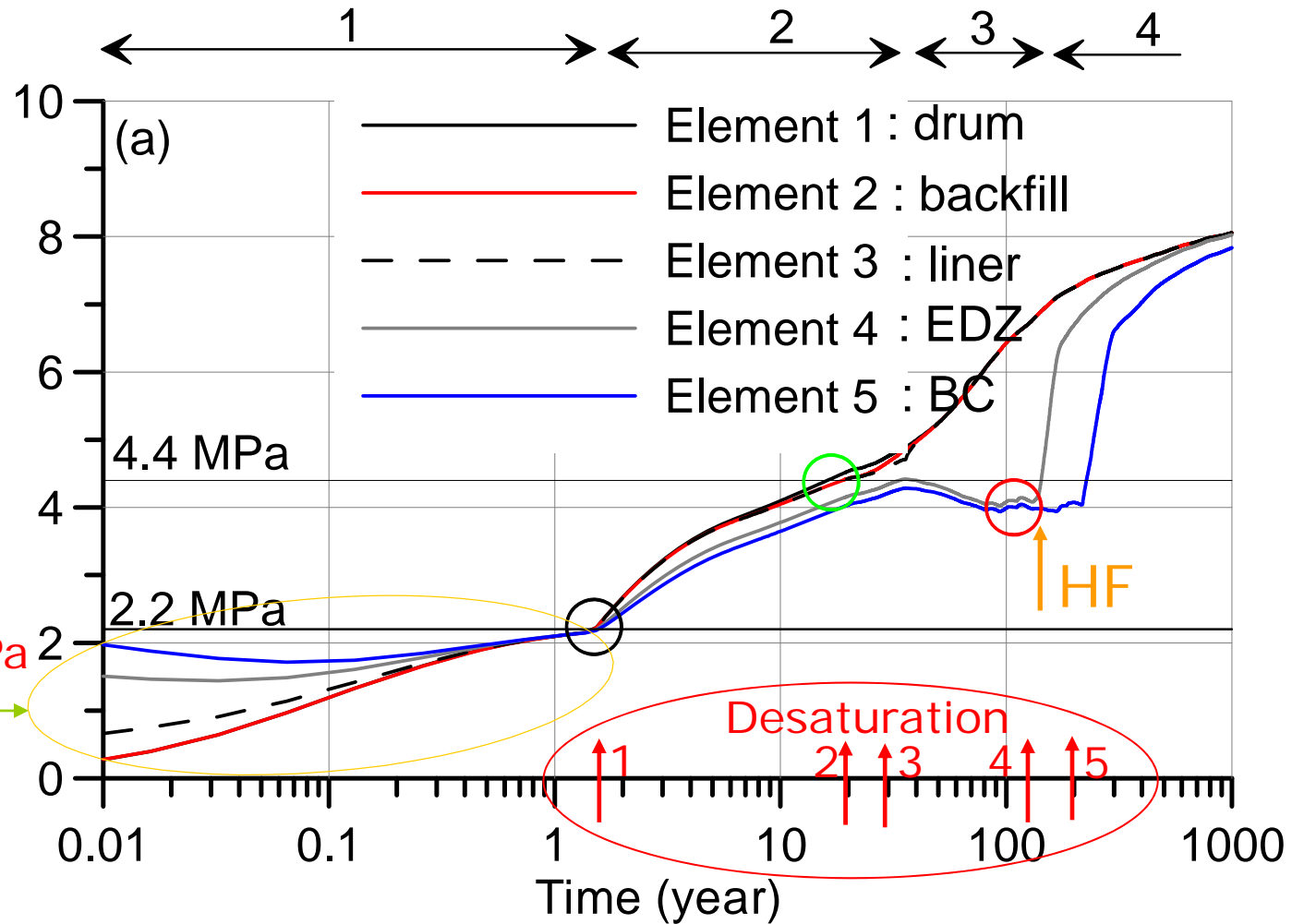 (will update codes in near future)

☹ Coupling fluid dynamics – mechanics of clay
  (not yet for the near future)

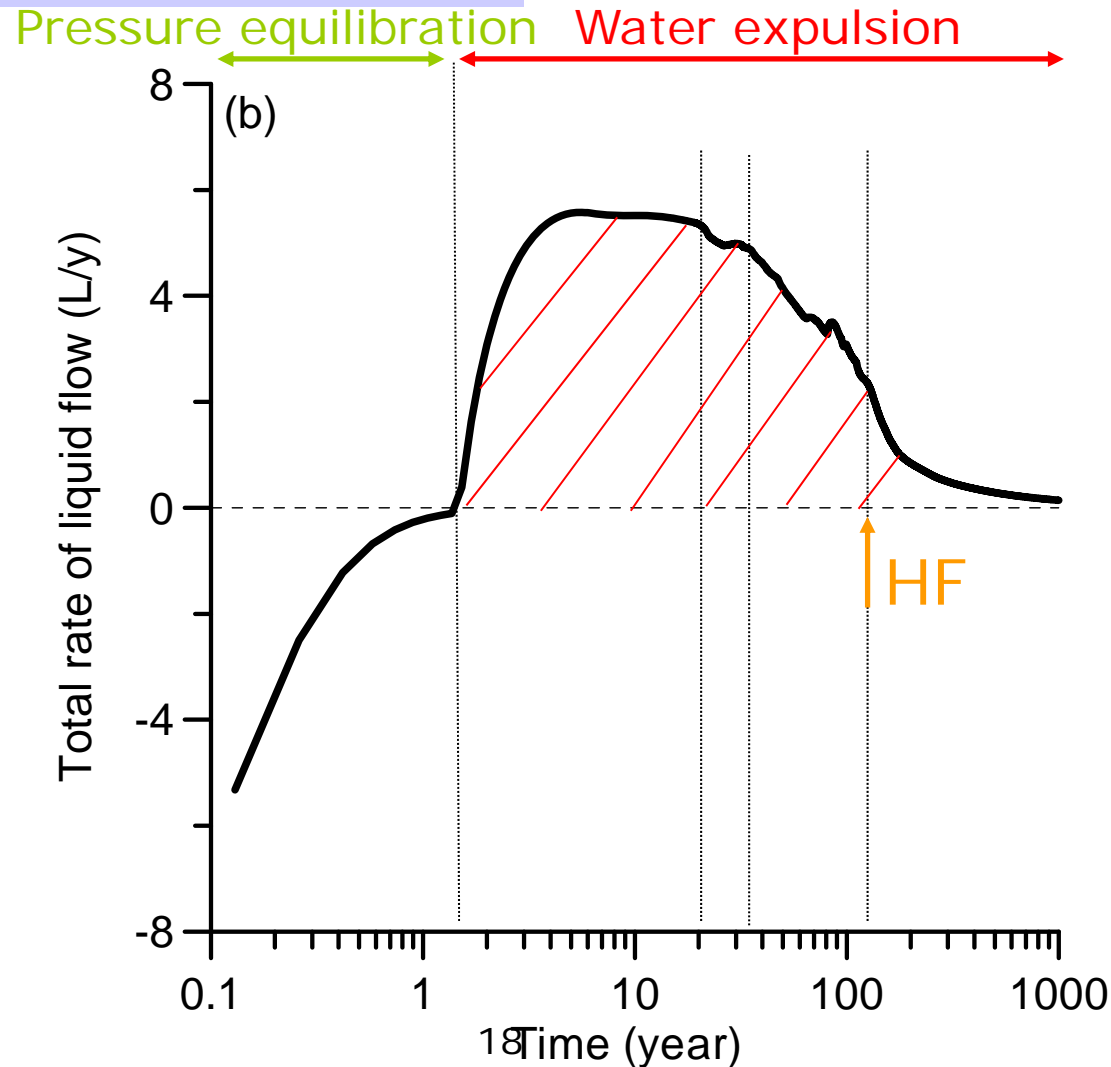16

Base case (LLW)

P vs *t*: pressure (*P*w; *P*g)

Water flow into/out of near field:
Base case (LLW): $S_l = 1$
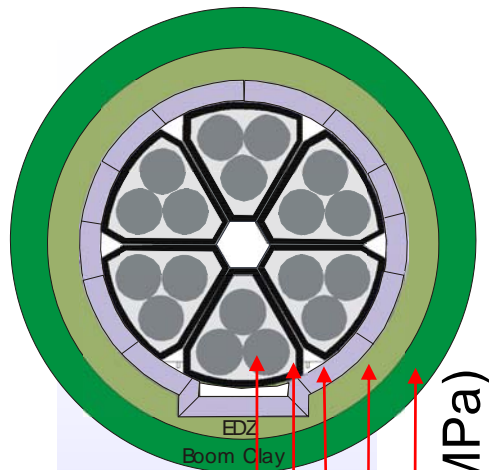
Pressure equilibration   Water expulsion

Water expulsion per

m of gallery =

1 m^3 after ~1000 y
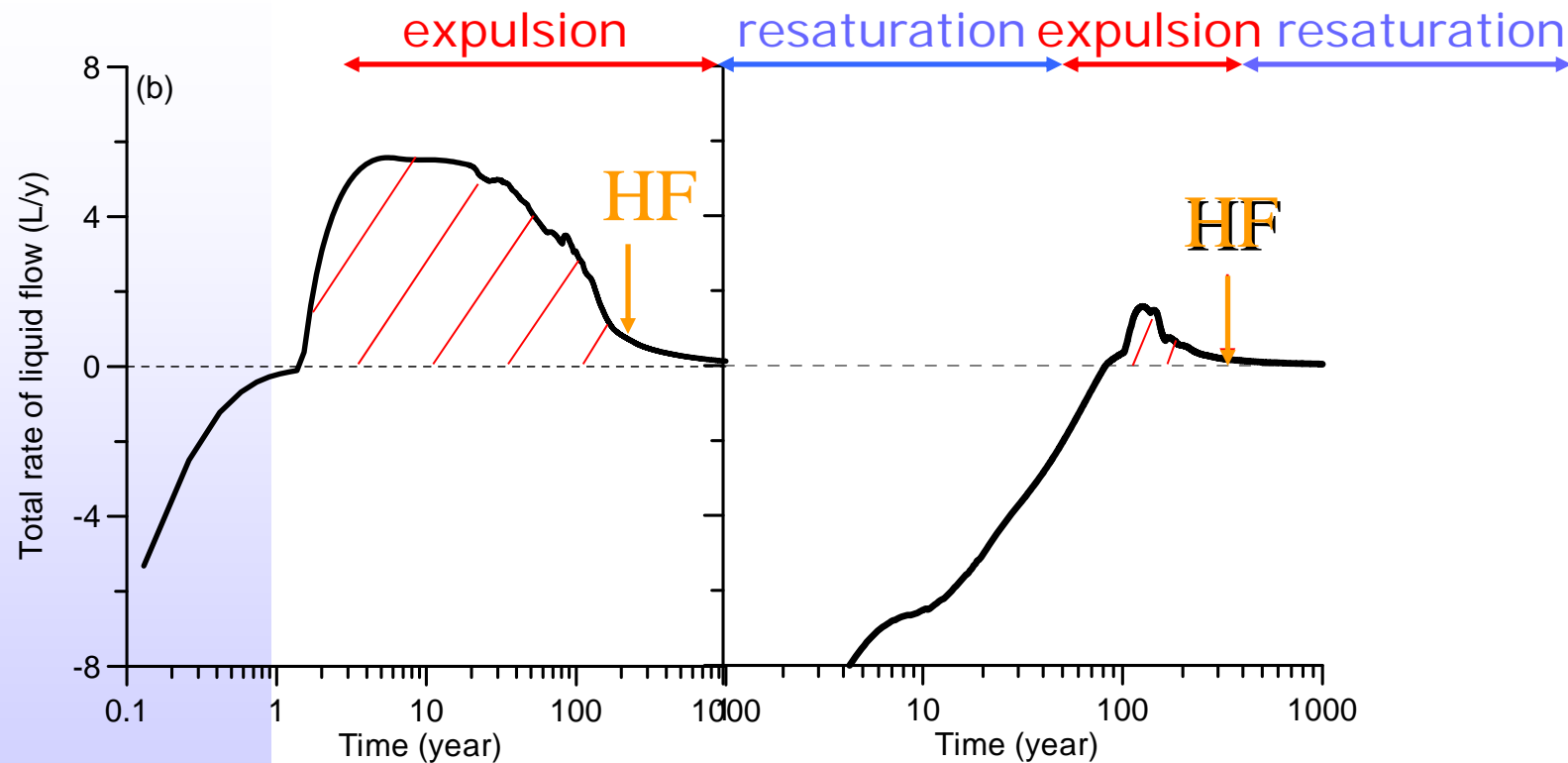
Water flow into/out of near field:
Low initial saturation (LLW): $S_l = 0.5$

Water expulsion per
m of gallery =
0.2 m^3 after ~1000 y

Saturation of near field   Water expulsion

Total rate of liquid flow (L/y)

HF

Uncontaminated
water from
host formation

Time (year)
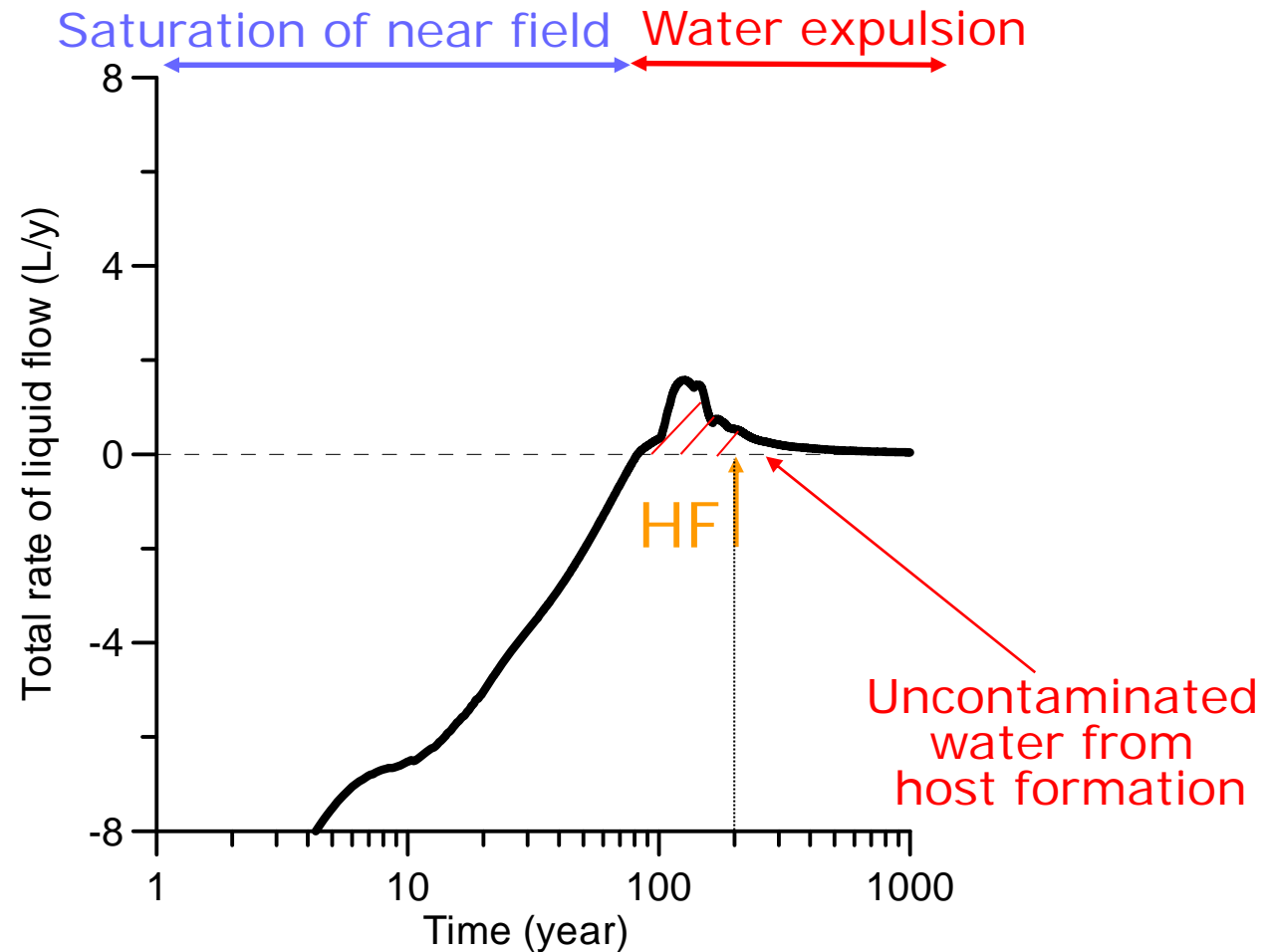
Effect of cyclic water expulsion

1st water expulsion   20 2nd water expulsion

Effect of hydrofracturing

- ## Experiments
  - ➤ Gas transport via hydrofracture does not involve water flow via fracture => no accelerated transport and contamination
    - ♣ desaturation only few %
    - ♣ total pressure in fracture > hydrostatic pressure surrounding the fracture => no water flow possible)
  - ➤ After pressure drop fractures close again, clay obtains its original properties
- ## Modelling
  - ➤ At time of first hydrofracturing, most water expelled
  - ➤ Expelled water not yet contaminated (early time process)
  - ➤ Cyclic pattern of expelling and resaturation mainly involves Boom Clay porewater; near field porewater not expelled

Conclusions (1)

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- EUROBITUM: H_2 gas most important

- Based on former design, H_2-gas production rates EUROBITUM (41 mol/m/y) similar to LLW (50 mol/m/y) = upper limit (conservative estimate)

- <u>Experimental evidence in Boom Clay shows</u>:
  - ➤ gas generation produces hydrofracturing of Boom Clay (lab & in-situ)
  - ➤ does not create accelerated water flow (lab)
  - ➤ fractures are not permanent (self healing of Boom Clay)(lab & in-situ)

- Hydrofractures preferentially form in direction of highest hydraulic conductivity (EDZ and horizontally in Boom Clay due to anisotropy in hydraulic conductivity)

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Conclusions (2)

- ## For LLW, two-phase flow modelling shows:
  - ➤ Water will be expelled first time after a period of ~100 y (not yet contaminated), followed by hydrofracturing and (partial) resaturation of near field
  - ➤ Further cycles of resaturation and water expelled involve small quantities of uncontaminated Boom Clay porewater

- ## For EUROBITUM:
  - ➤ Likely to be even more favourable because less drums/m and/or lower reactive surface compared to LLW
  - ➤ Details about gas pressure built-up, volume of water expelled and timing of processes still need to be evaluated

- ## No permanent preferential pathways (only temporary and very localised mechanical disturbance)

- ## No accelerated release of radionuclides
  - =>Performance of repository is not significantly affected (safety function of near field and Boom Clay still intact)

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