Experimental and modeling study of salt binding and release by stabilized MSWI fly ash wastes

Today presentation main contributors

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Sustainable Landfill Founda

Outline

I. Stabilized MSWI fly ash waste

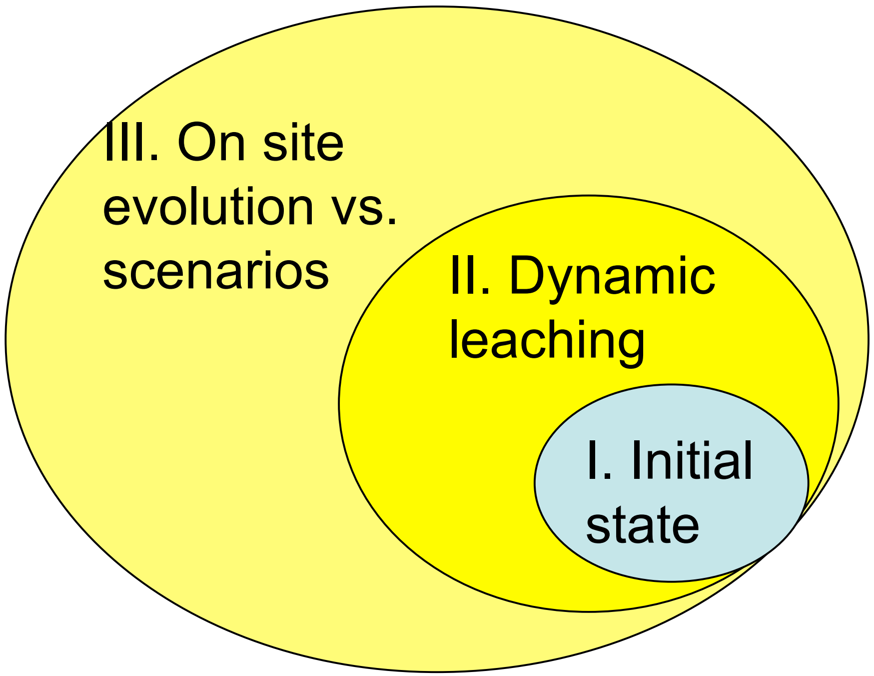
II. Modeling approach

III. Application to dynamic leaching test

IV. An overview of disposal facility modeling

Methodology

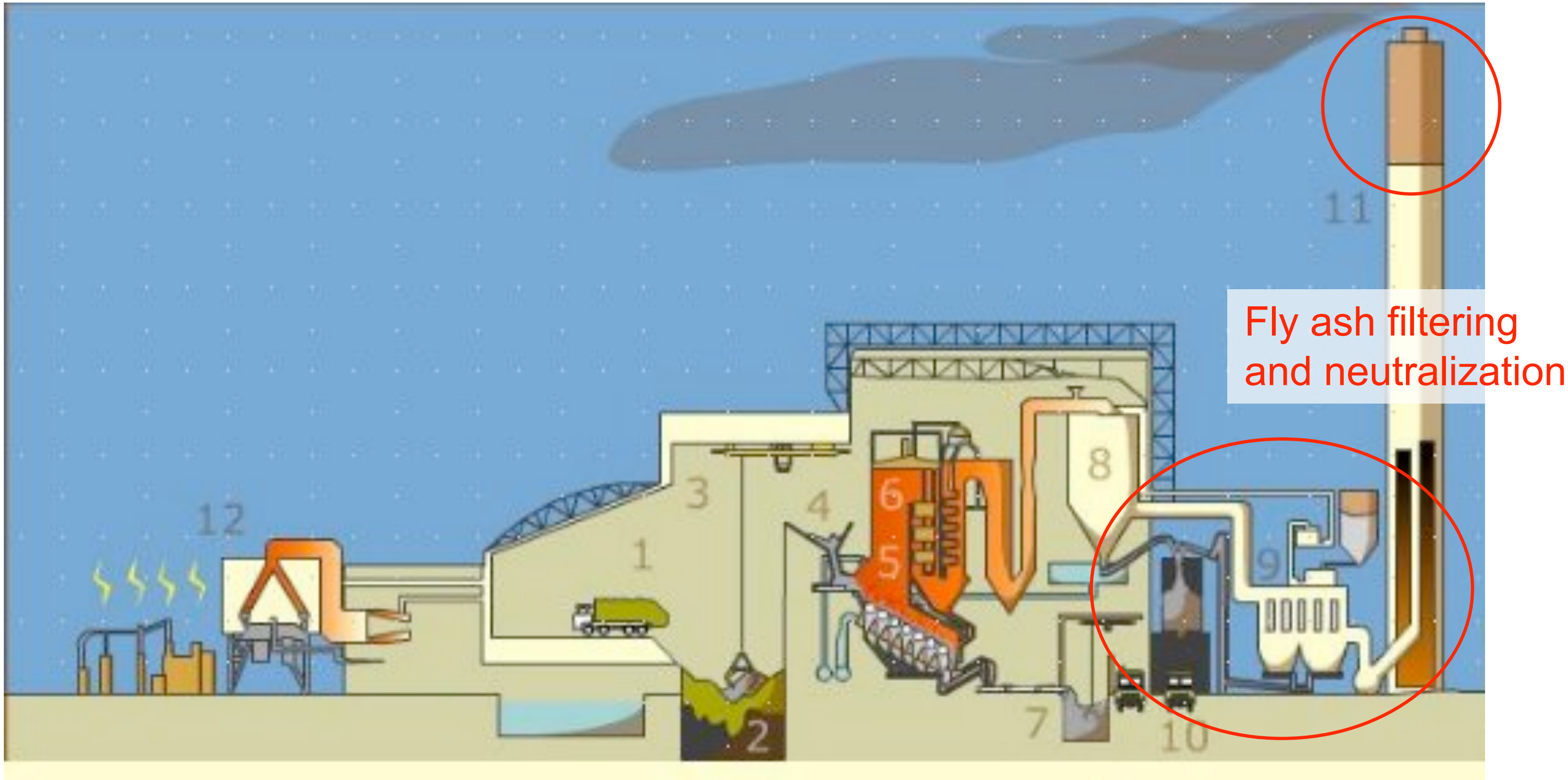
- **Performance AND environmental impact assessment of waste disposal (or recycling scenarios)**
  - Dynamic leaching tests to better characterize the cementitious waste long-term evolution
  - Understanding of leaching mechanisms to link laboratory tests to engineered barrier systems (disposal) or waste/environment interactions (disposal, recycling)



- **Needs for a "common" modeling approach and code applied to different scales, as mechanistic as possible**
  - Reactive transport codes are good candidates

- I + II: *Waste Management (2007)*
- III: *J. Hazardous Mater. (2007)*

Municipal waste incineration plant



$2\,HCl + CaO \rightarrow CaCl_2 + H_2O$ (semi-wet process) ; 50 kg/T of waste

MSWI fly ash

- Leaching test X31-210 → 40% of highly soluble fraction, dominated by chloride and sulphate salts

- Bulk chemistry

Zn	→	6 000 ppm
Pb	→	2 000
Cu	→	400
Cr	→	100
…		

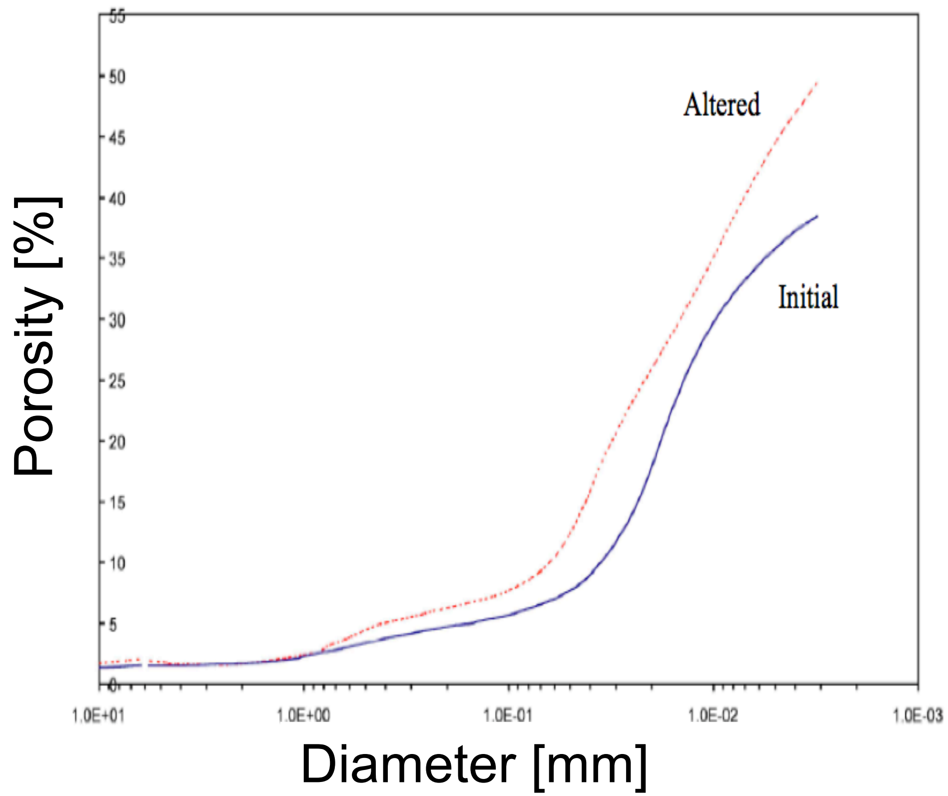
- Required stabilization before disposal, essentially through hydraulic binders

Stabilized MSWI fly ash

- Bulk chemical composition

CaO	40 % wt Dry Material
SiO_2	18.5 %
Al_2O_3	7 %
Na_2O	1.5 %
K_2O	1.7 %
SO_3	2.5 %
Cl	8.5 %

Stabilized MSWI fly ash



Initial porosity ~ 40 %

- Relatively high porosity and hydrodynamic parameters

(K ~ 10^{-11} m/s, Dp ~ 10^{-10} m^2/s)

Stabilized MSWI fly ash

- An example of the proportion of the main solid phases

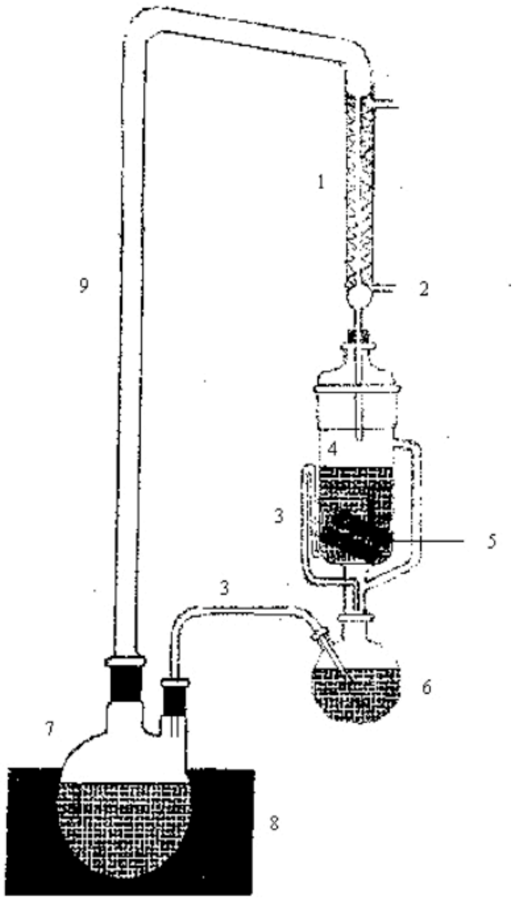
$CaCl_2Ca(OH)_2:H_2O$	5.5 % wt
Calcite	4.5
CSH 1.5	34.5
Ettringite (AFt)	11.5
Friedel's salt (AFm)	22
Halite	2.5
Sylvite	3.0
Portlandite	6
Quartz	4.5

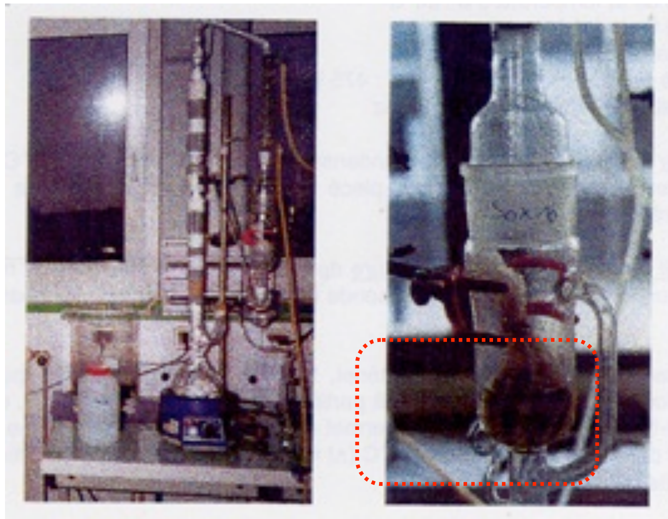
Stabilized MSWI fly ash

- An idea of the initial pore water chemistry (calculation)

pH	12	
Na	35 500 mg/L	1.5 mol/L
K	47 000 mg/L	1.2 mol/L
Ca	16 500 mg/L	0.4 mol/L
SiO_2	1 mg/L	10^{-5} mol/L
Cl	125 000 mg/L	3.6 mol/L
SO_4	> 300 mg/L	> 5 10^{-3} mol/L

Set-up of the dynamic leaching test

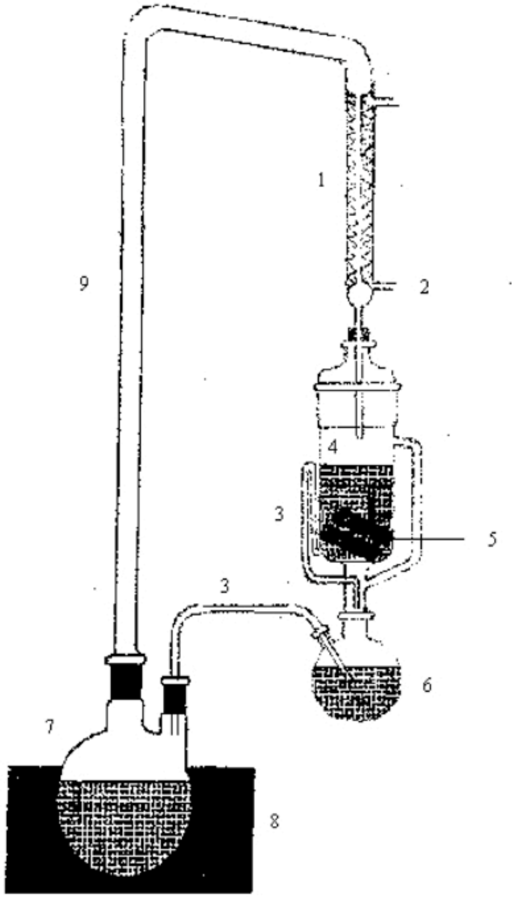


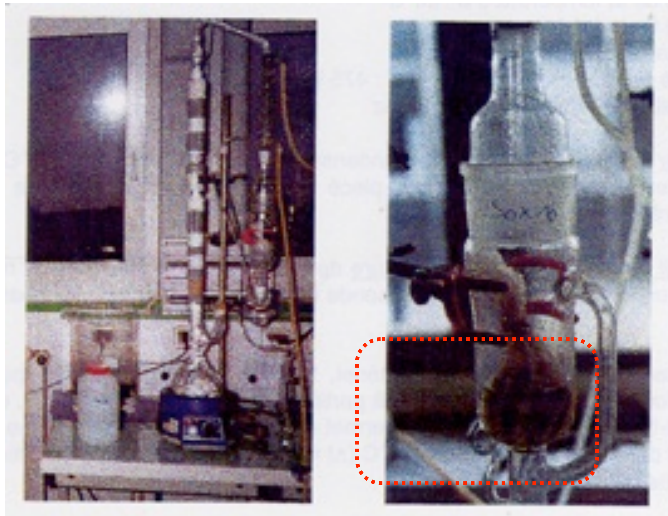


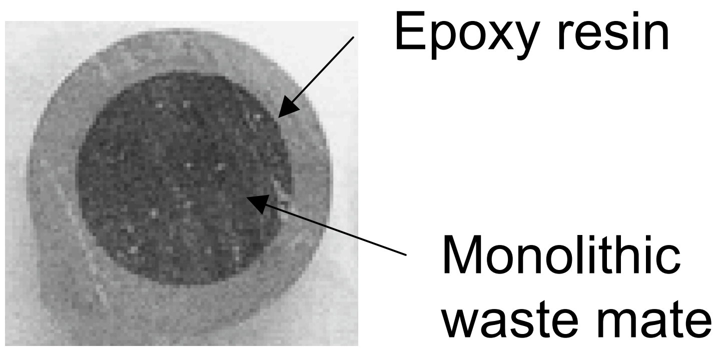
- Renewal at 100 ml/h
- T = 20 C
- Partially open conditions

➤ Soxhlet-like leaching test

Set-up of the dynamic leaching test







Thickness
= 1 cm

Diameter
= 4 cm

- Soxhlet-like leaching test

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Reactive transport code HYTEC

Chemistry

- aqueous chemistry
- dissolution/precipitation of solids
- sorption
- microbiological module

- local thermodynamic equilibrium
- kinetics on redox, sorption and solid reactivity

Hydrodynamics

- 1D, 2D-cylindrical geometry (REV)
- Advective and diffusive transport

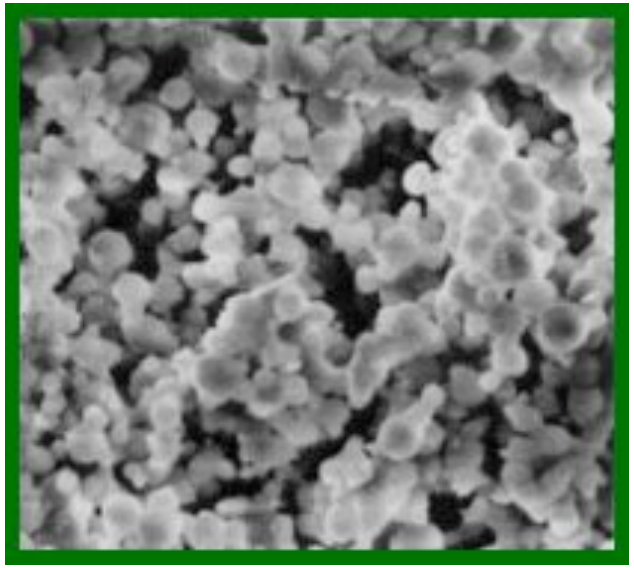
$$\frac{\partial \omega c_i}{\partial t} = \nabla \cdot (D_d \nabla c_i - c_i U) - \frac{\partial \omega \bar{c}_i}{\partial t}$$

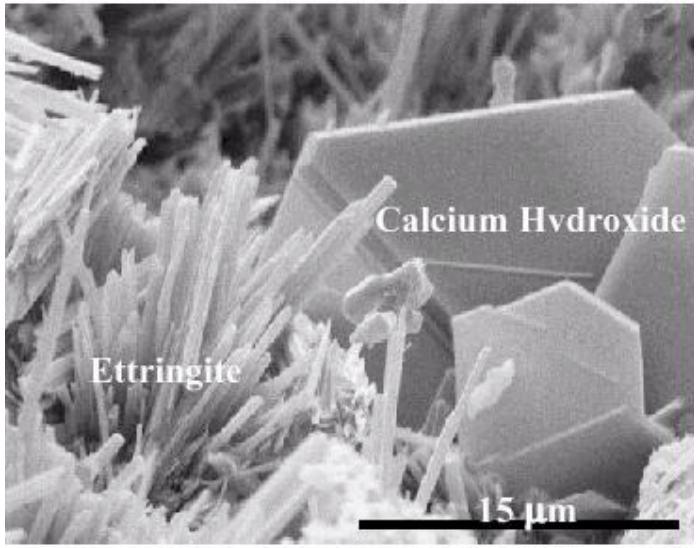
for (un)saturated hydric conditions

- feedback of chemistry on ω and D_e

$$D_e(\omega) = D_e(\omega_0) \left(\frac{\omega - \omega_c}{\omega_0 - \omega_c} \right)^m$$

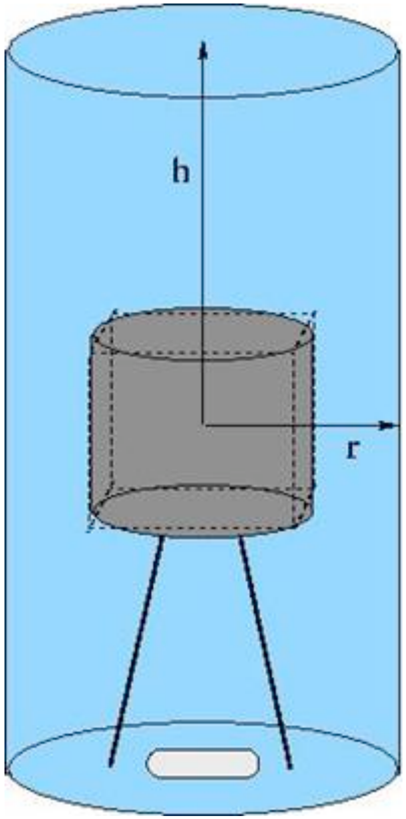
What's the surface of a porous media

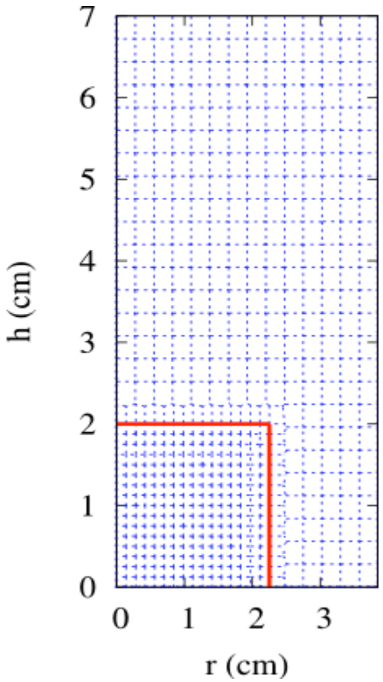




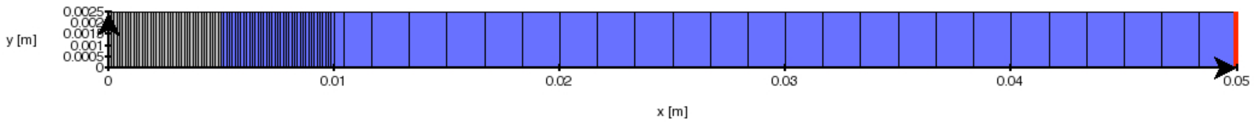
First models used diffuion of salts + global kinetic dissolution of the waste surface

REV modeling





From 1D to 2D geometrical configurations



- Elementary Volume Representation of the interface rather than a geometrical surface
- Equilibrium approach, kinetics is diffusion-controlled (in a first step)

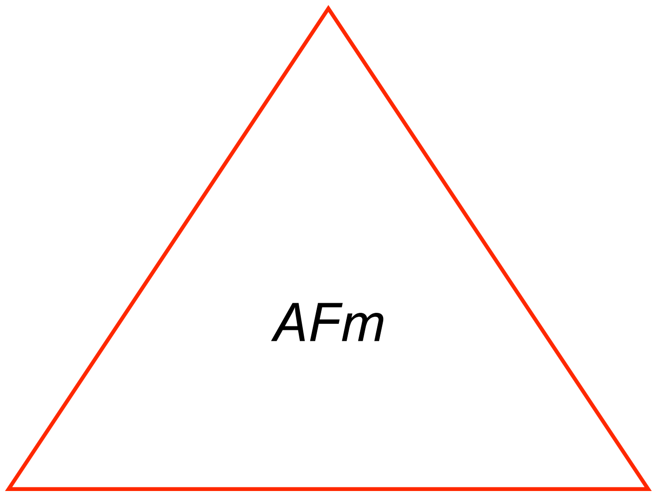
Thermodynamic database

- Database
  - EQ3/6 data base with additional data on cement phases
  - Pure discrete phase approach

    Ex 1 : silica gel - CSH 0.8 - CSH 1.1 - CSH 1.5 - CSH 1.8

    Ex 2 :

$Ca_4Al_2SO_4(OH)_{12}$
Monosulfo



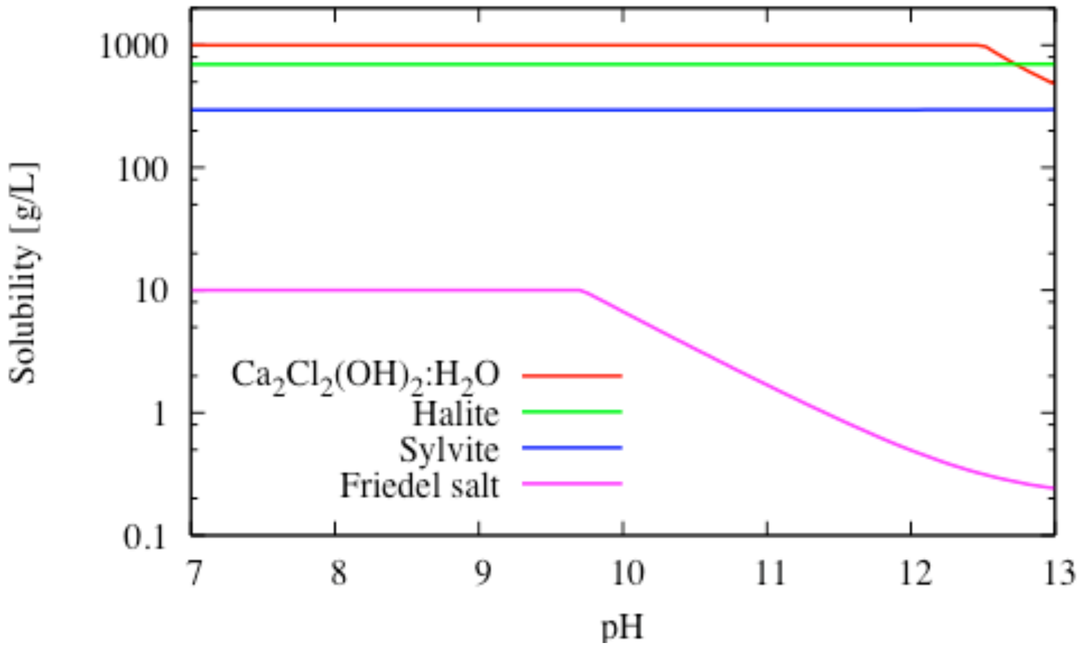
$Ca_4Al_2CO_3(OH)_{12}:6H_2O$
Monocarbo

$Ca_4Al_2Cl_2(OH)_{12}:4H_2O$
Friedel salt

Activity correction model

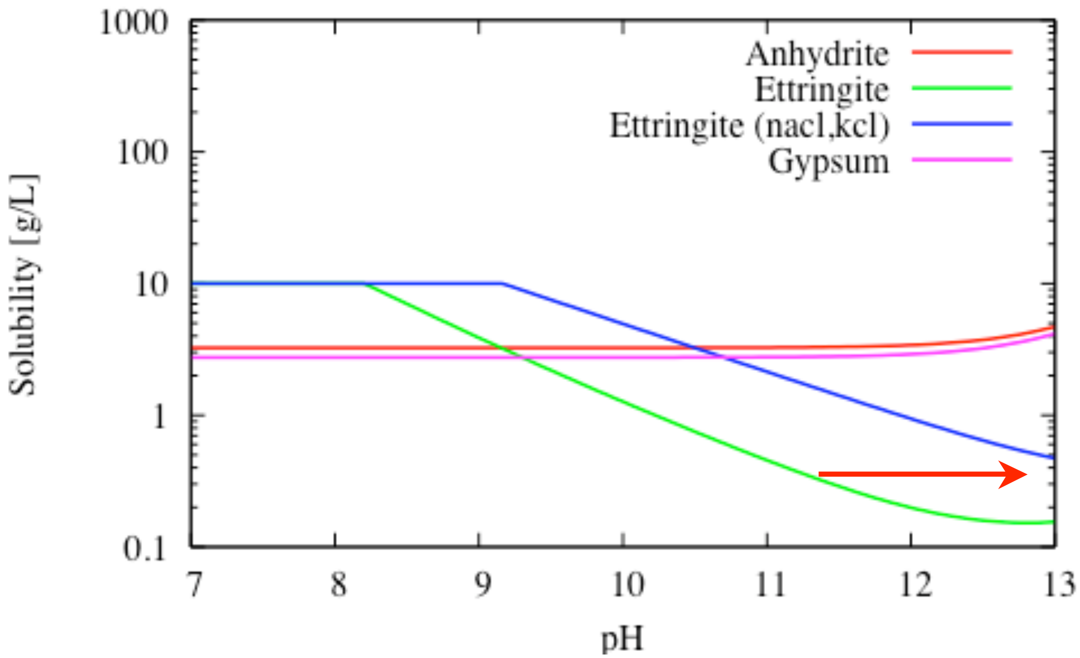
- B-dot model
  - calibrated for NaCl solution
  - for ionic strength ≤ 1 - 2 mol/L
  - applicable on a wide range of temperature
  - gives access to the details of the aqueous speciation
- Helgeson's model
  - for water activity

Chloride phases



- Stability of the chloride solid phases vs. pH (HYTEC)

Sulphate phases



➢ Stability of the sulphate solid phases vs. pH (HYTEC)

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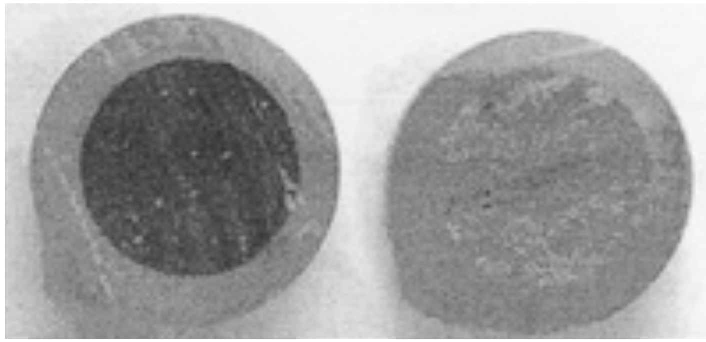
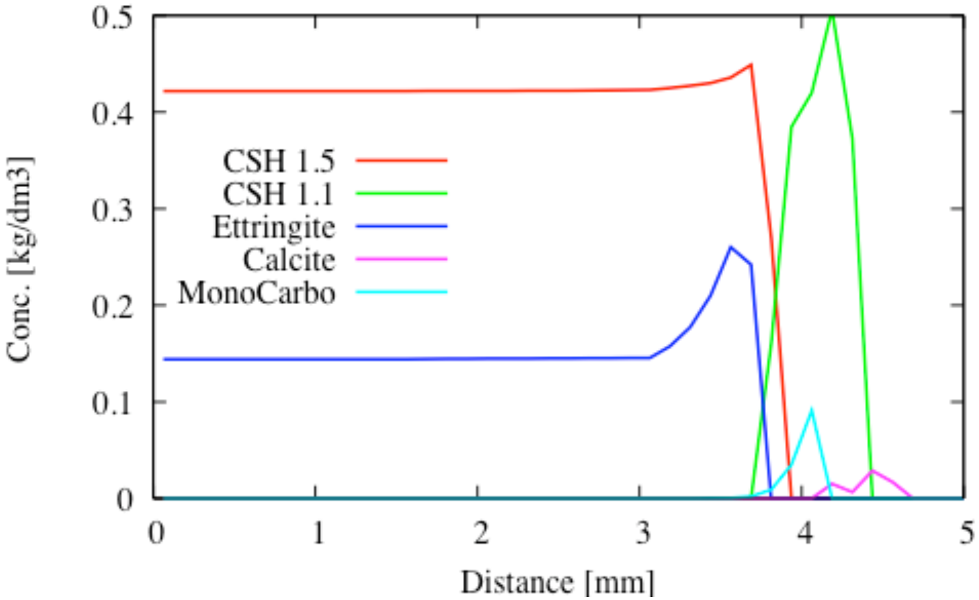
Mineralogy evolution



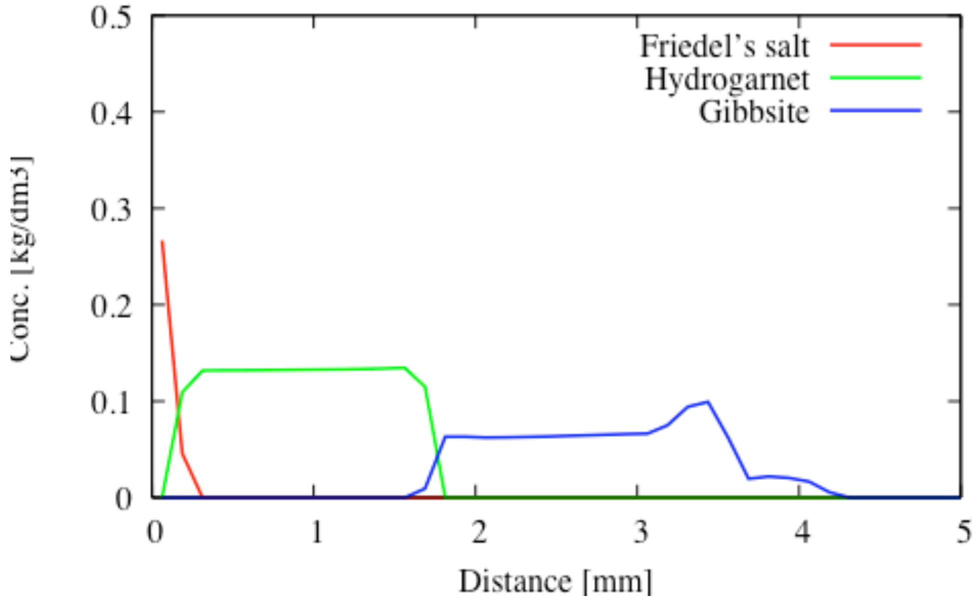
Picture of the sample before and after leaching during 6 months

Mineralogy evolution

6 months

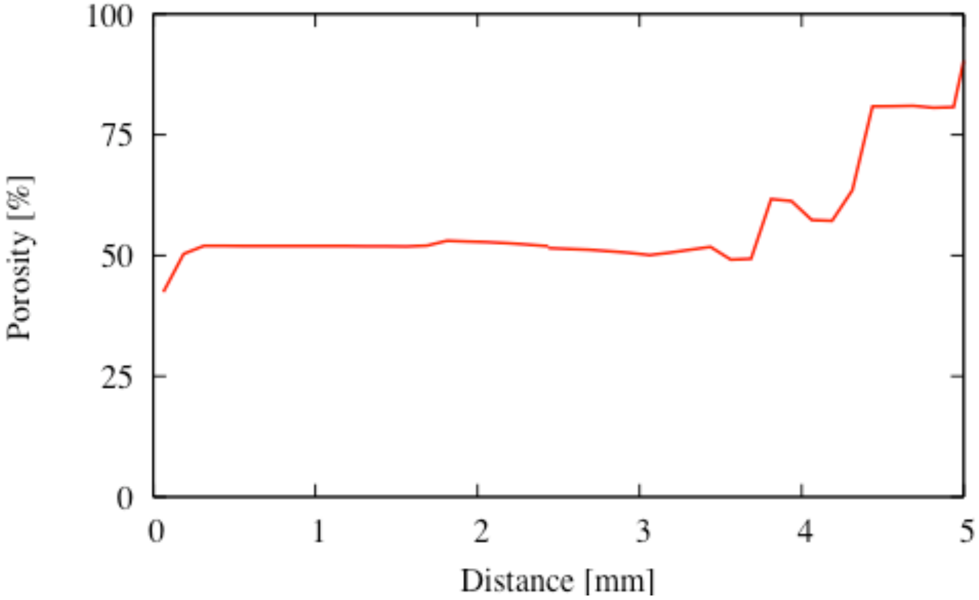


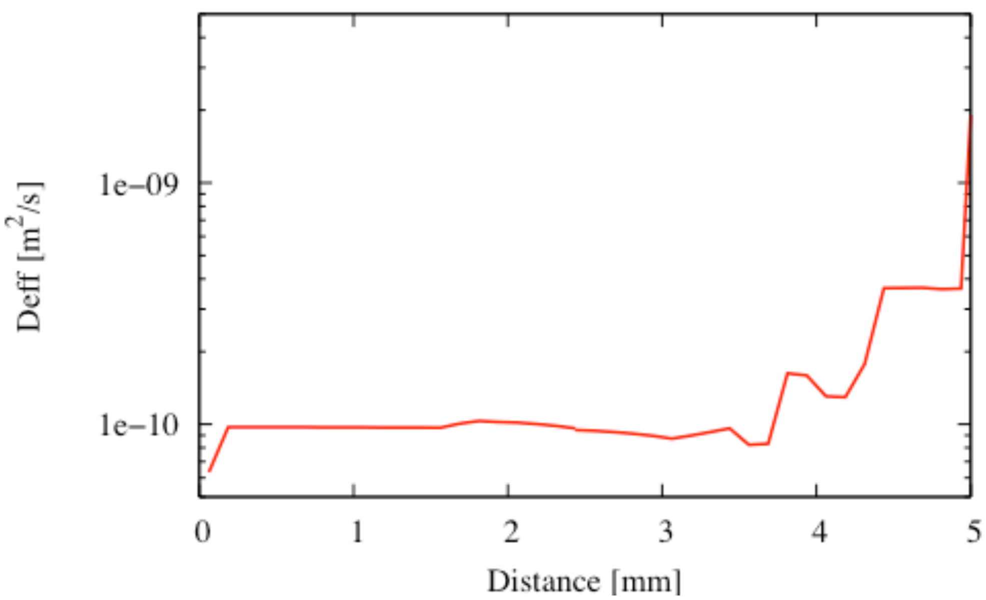
(5 months)



- Calculated position of the mineralogical fronts after leaching (variable porosity and Deff)

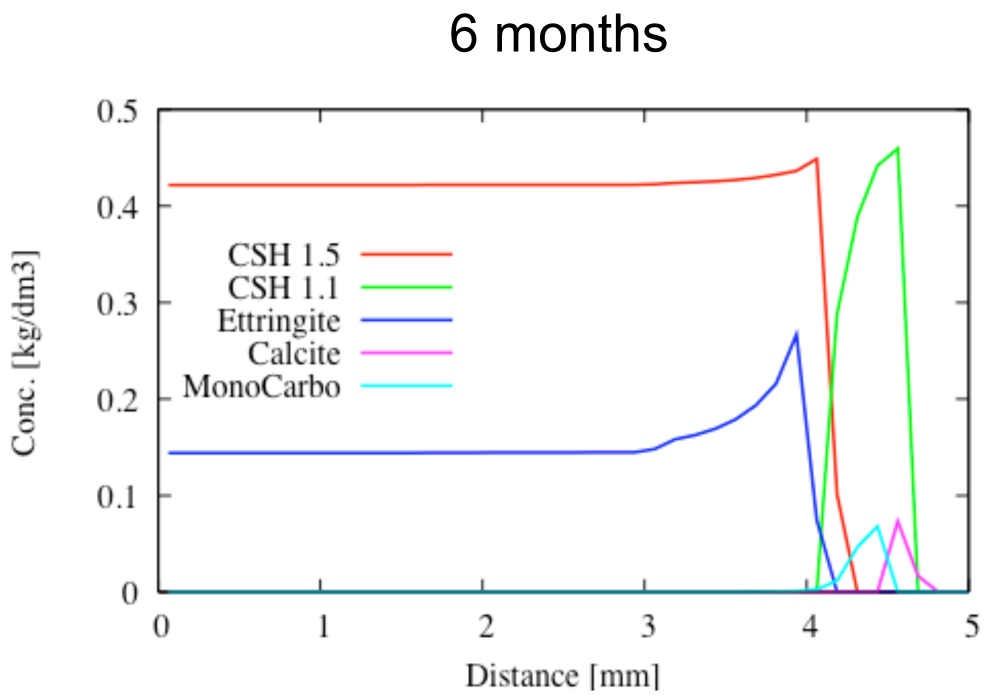
Porosity evolution

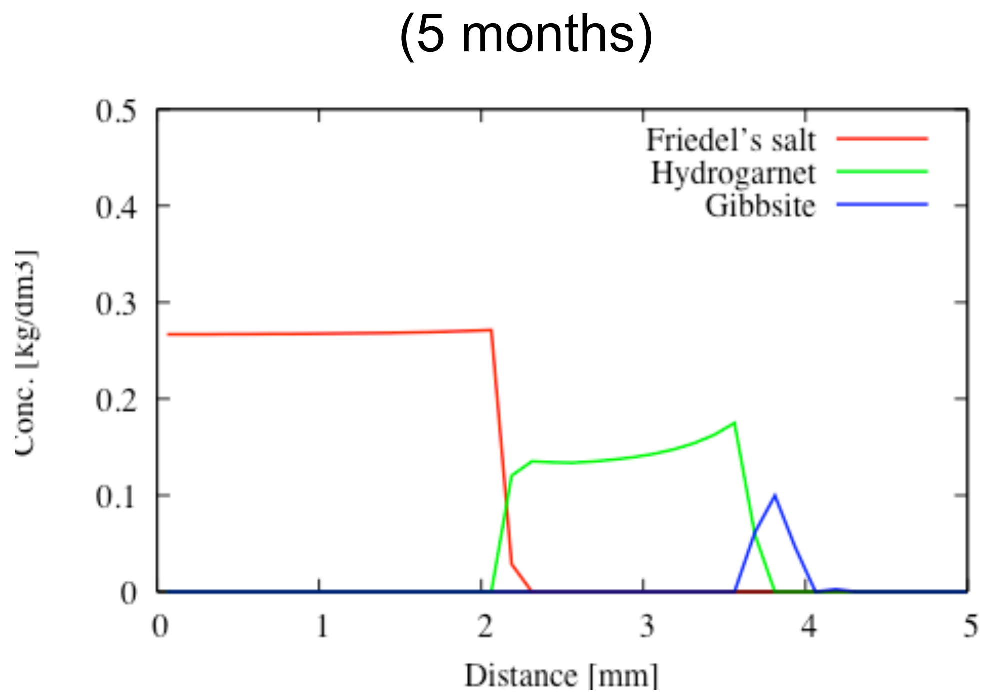




- Calculated evolution of porosity and effective diffusion coefficient after leaching

Mineralogy evolution

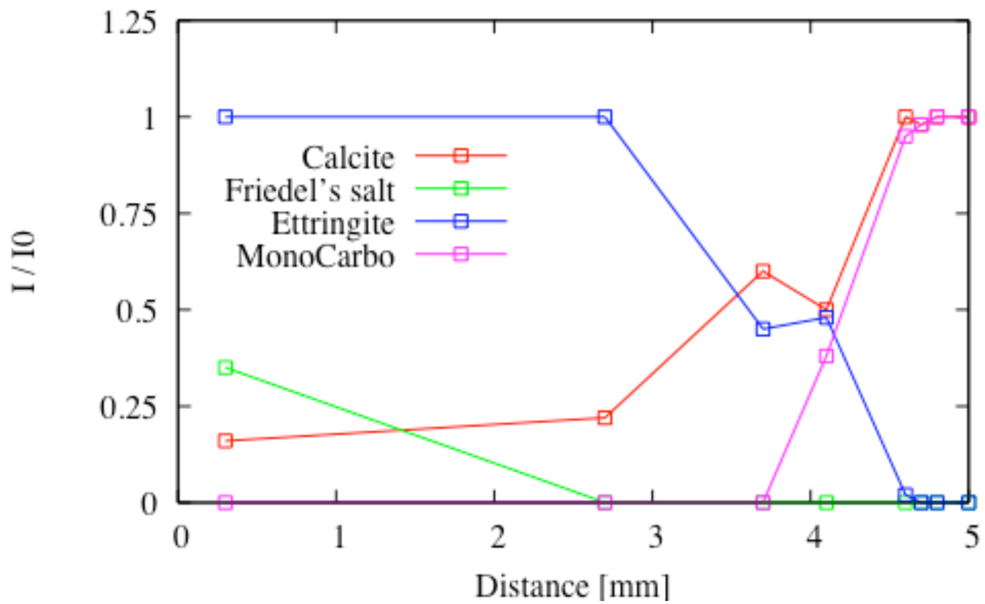




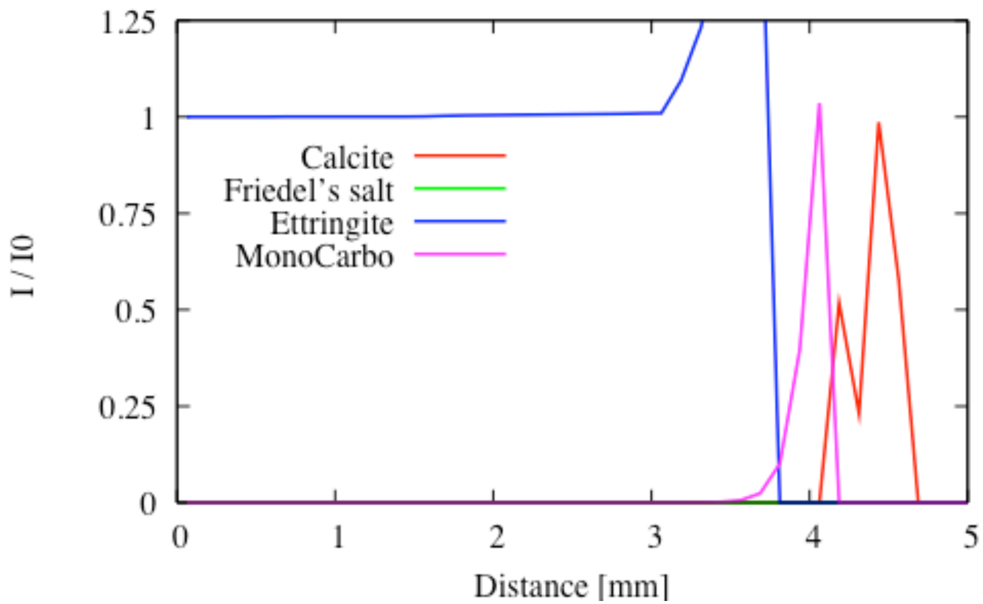
- Calculated position of the mineralogical fronts after leaching
  (fixed porosity and Deff)

Mineralogy evolution

DRX



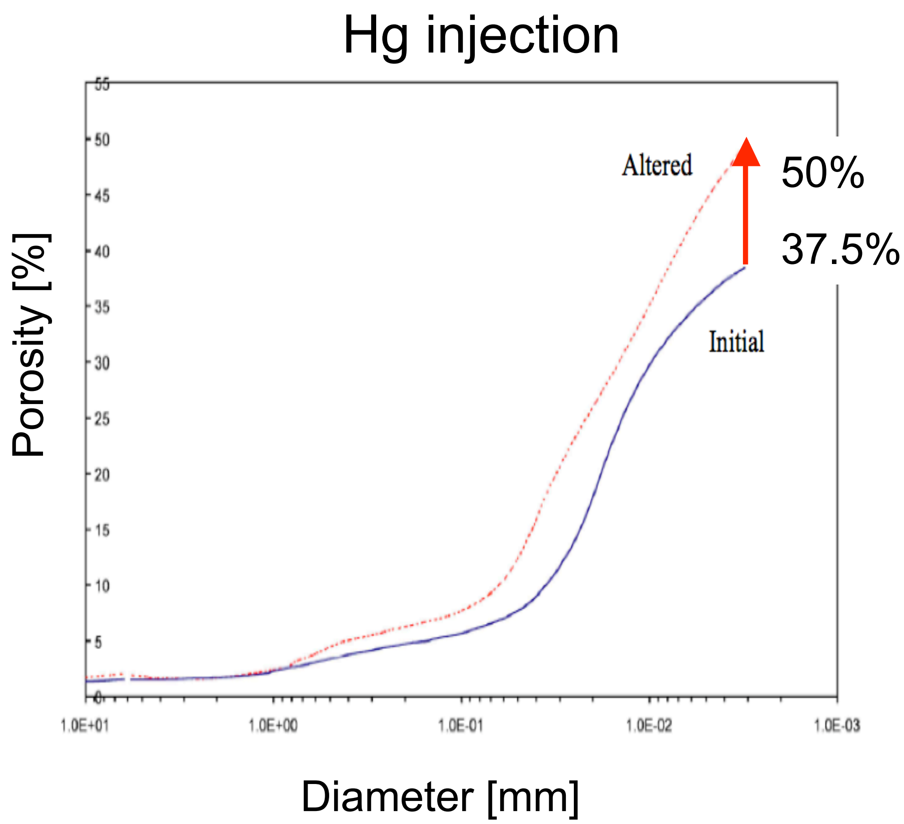
Calculated

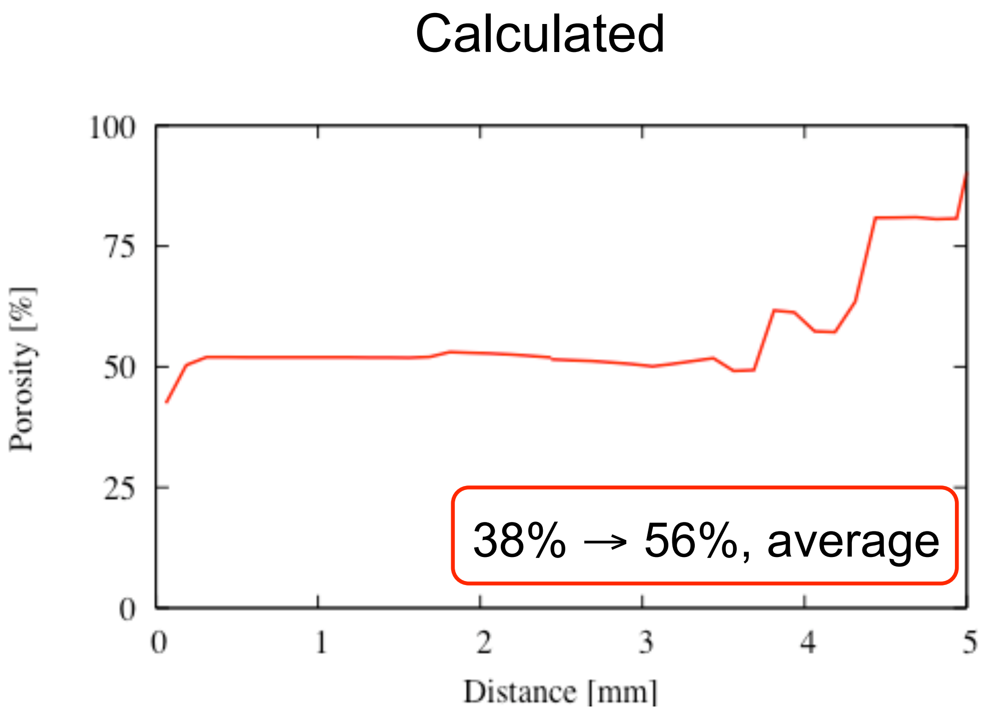


Full depletion of portlandite in both cases

- Comparison between DRX and calculated profiles

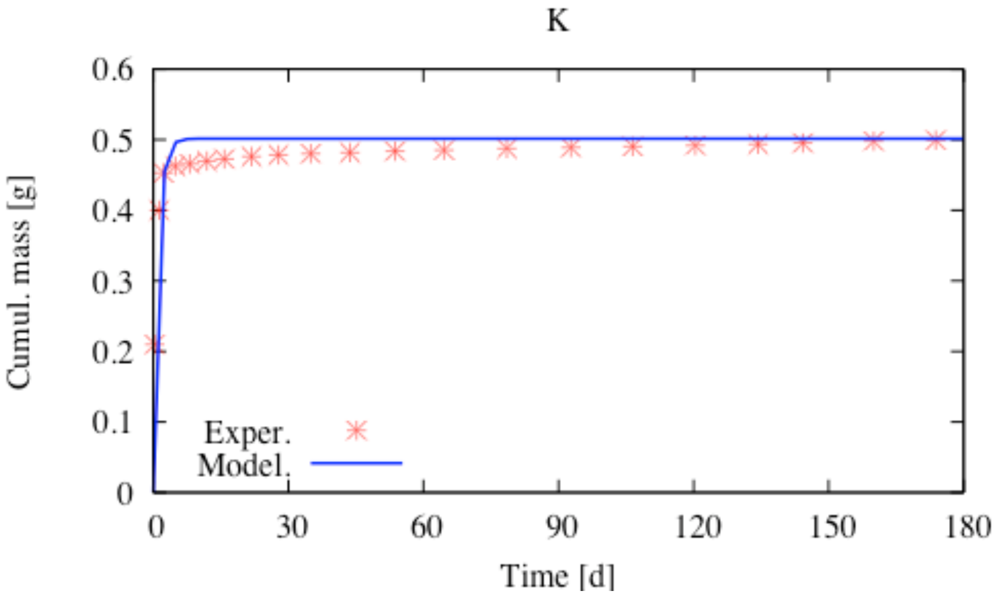
Porosity evolution

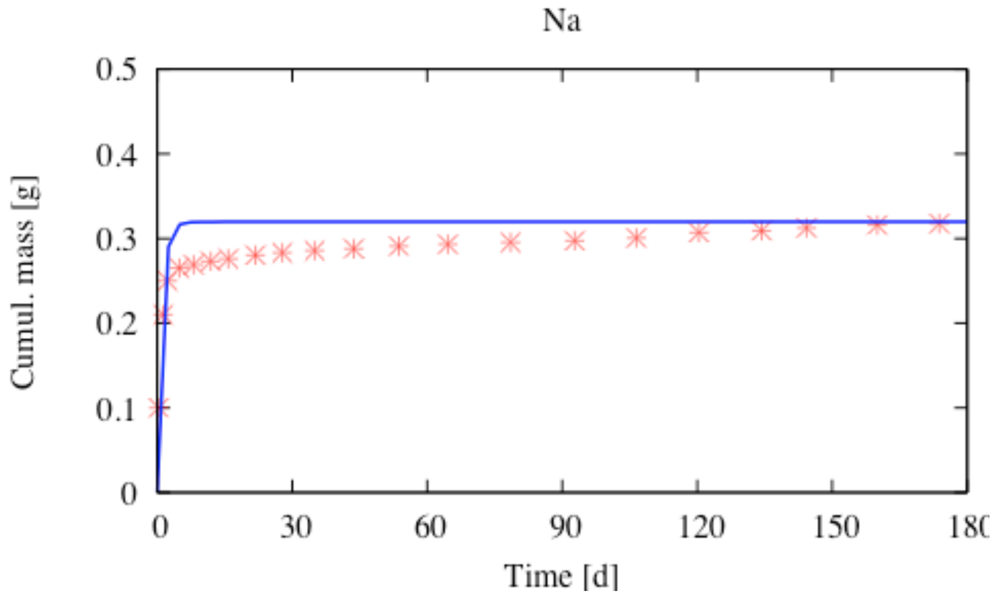




- ➢ Comparison between experimental and calculated porosity profiles

Cumulative release of alkaline elements

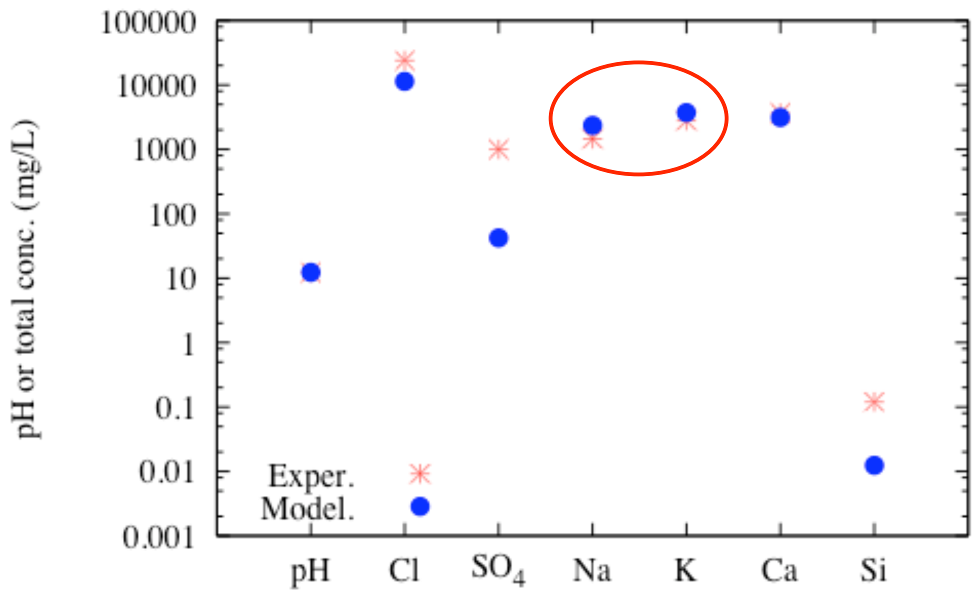




Measured released mass
K = 99.5%, Na = 98.5%

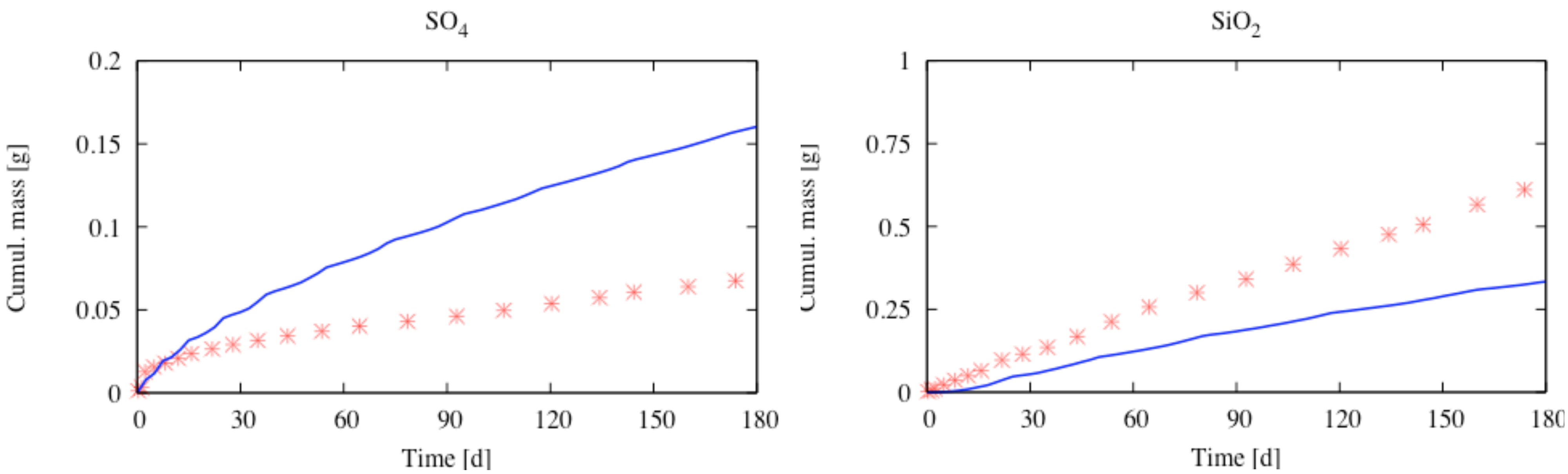
➢ Diffusion-controlled Release (poral source)

Cumulative release of alkaline elements



- Batch test (L/S = 5)

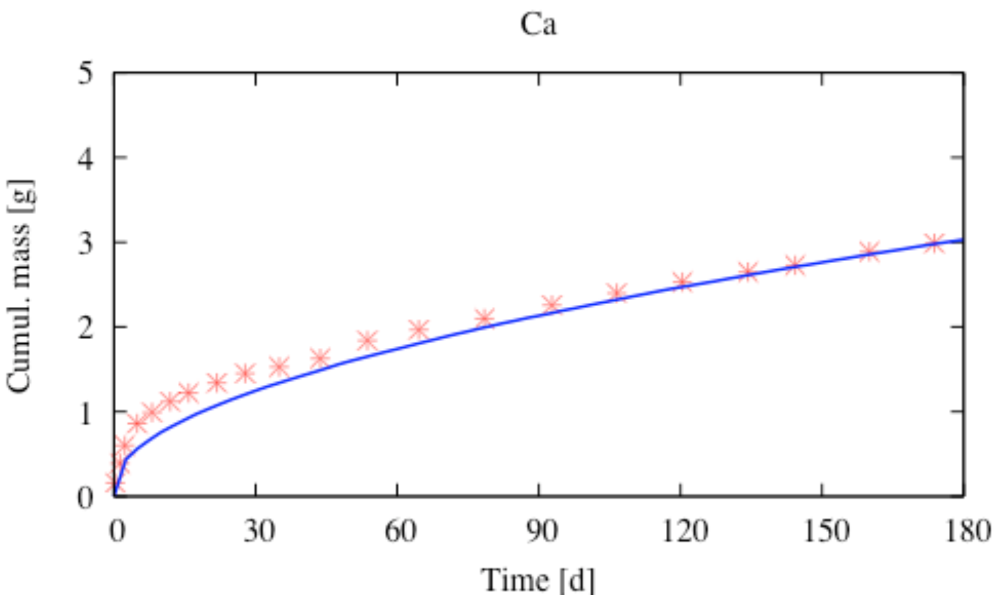
Cumulative release of sulphate and silica

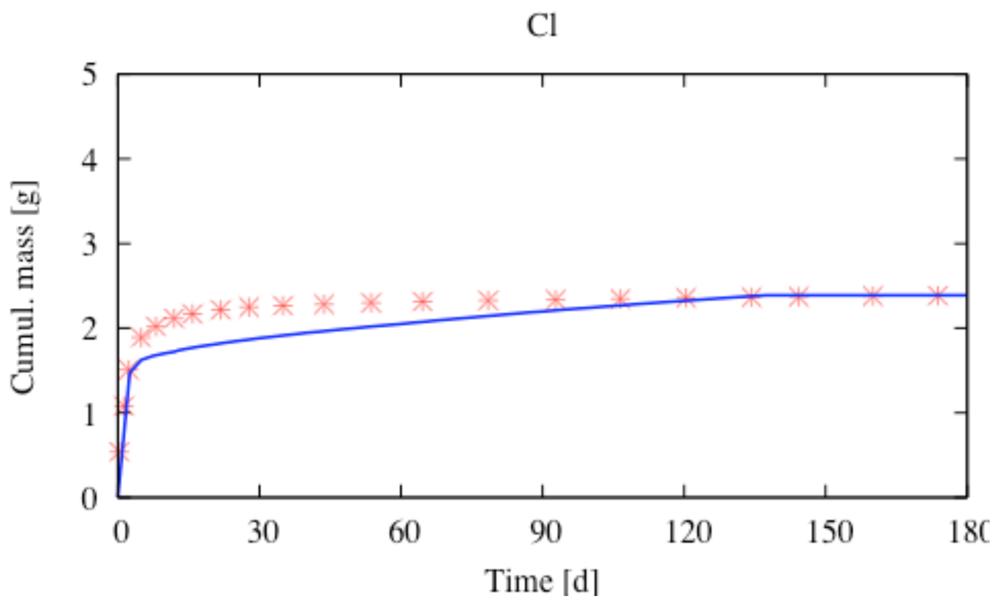


Measured released mass
SO_4 = 5%, Si = 9%

➢ Solubility-controlled release (solid phase source)

Cumulative release of calcium and chloride





Measured released mass
Ca = 25.5%, Cl = 99.9%

- Mixed release process

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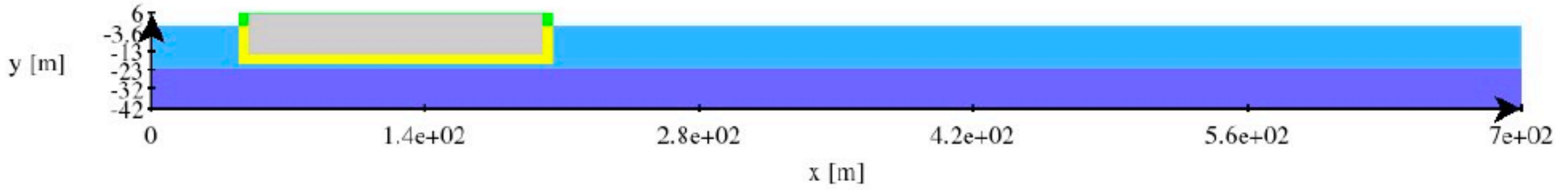
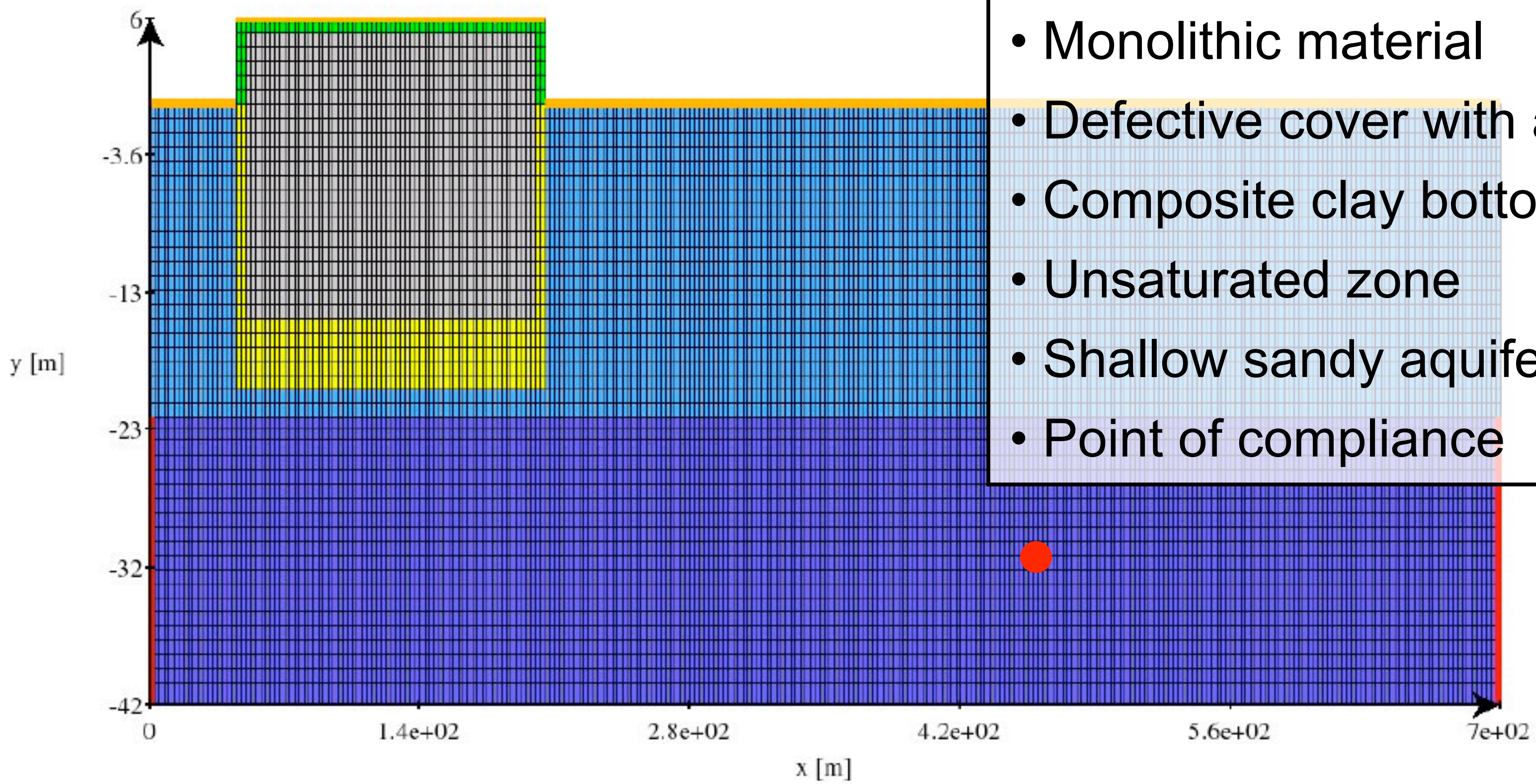
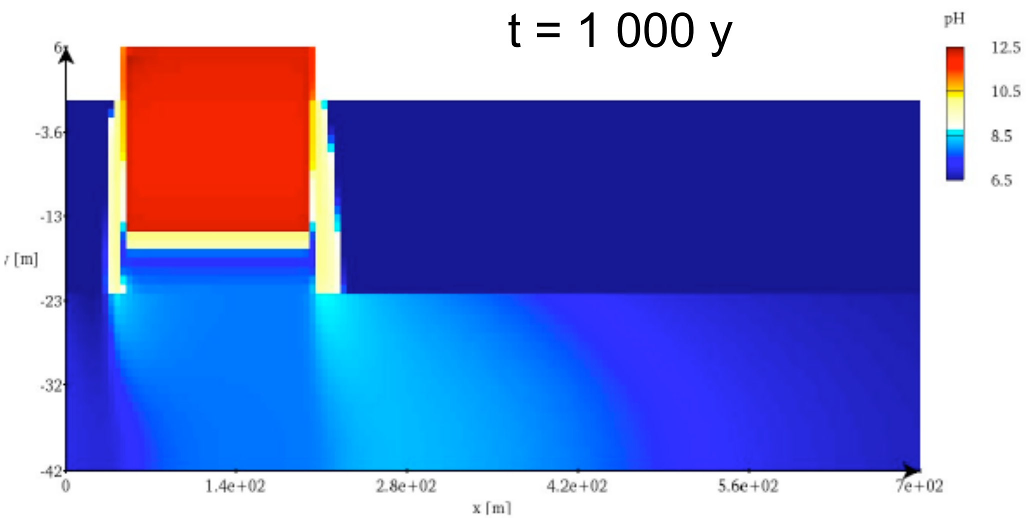
Scheme of the disposal facility

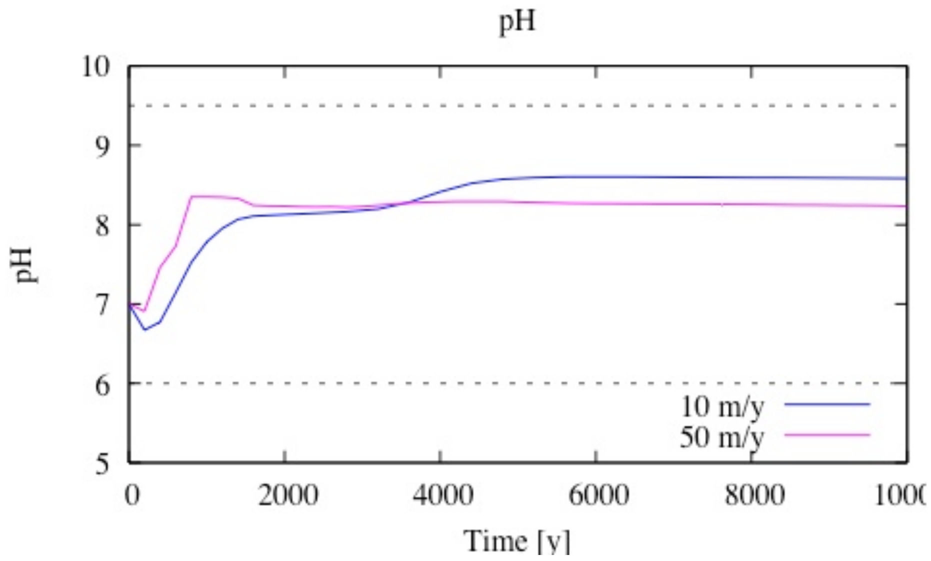


- Waste volume: 150 x 150 x 20 m
- Monolithic material
- Defective cover with an upper clay liner
- Composite clay bottom liner
- Unsaturated zone
- Shallow sandy aquifer (10 m/y)
- Point of compliance



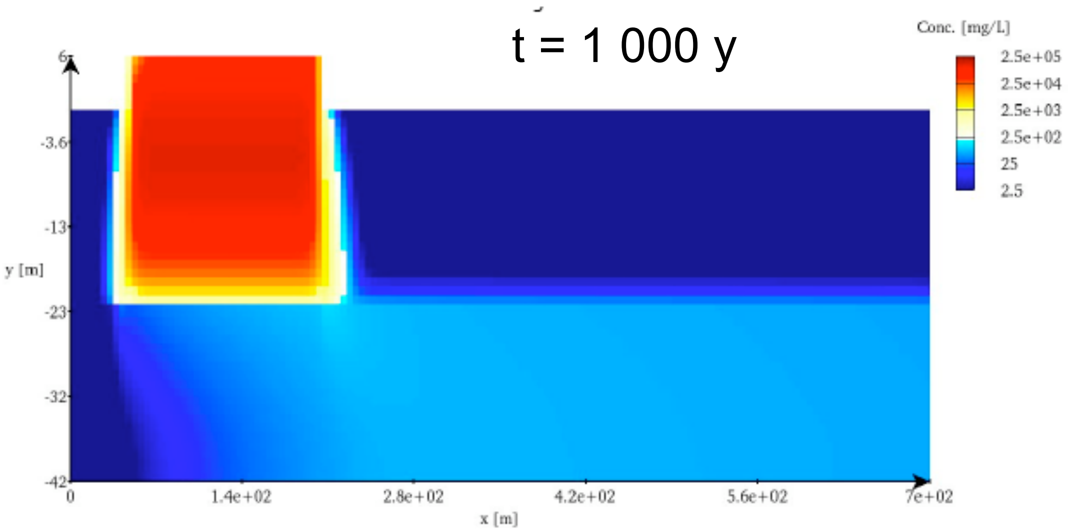
Alkaline plume migration

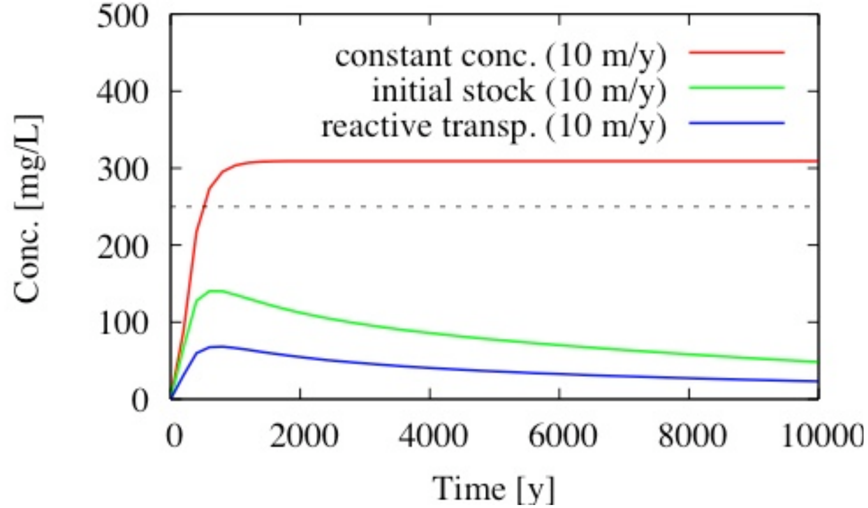




pH: 2D profile and evolution with time at the point of compliance

Alkaline plume migration





Chloride conc.: 2D profile and evolution with time at the point of compliance

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Conclusion (methodology)

- The "long-term" evolution of the stabilized MSWI FA waste was not fully addressed, the present calculations are still in progress!
- However, the agreement between model and experimental data is far to be bad both for the release of major element and the mineralogy evolution
- Capability of reactive transport codes to mechanistically link the laboratory tests to site scenarios, and therefore to support performance and environmental impact assessments in a more consistent way

Conclusion (science)

- The MSWI FA salts are clearly stabilized in the waste form, particularly sulphate but, in a smaller extend, chloride too
- Sensibility analysis on the AFm thermodynamics, especially the destabilization of the Friedel's salt vs. monocarbonate under partially desaturated conditions
- More detailed insights in the laws for porosity evolution and its relationship with Deff
- Confrontation of modeling with core samples collected in 10-year disposal (PASSIFY Project)