Complex multi-physics multi-layers modelling approaches for iron-environment interactions

upscaling to a cell scale of geological repository radioactive waste

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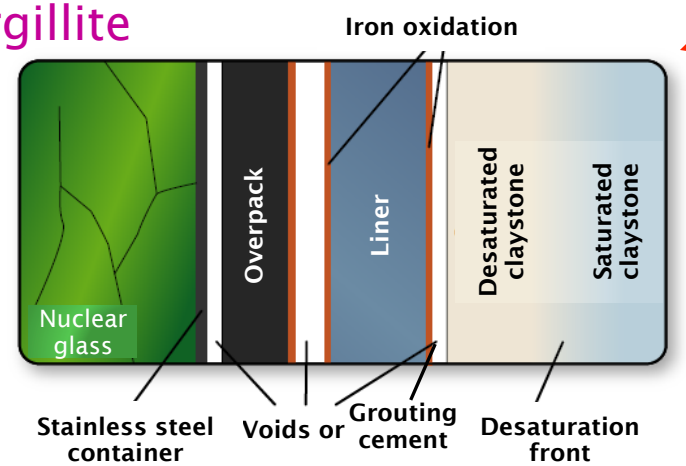
Context

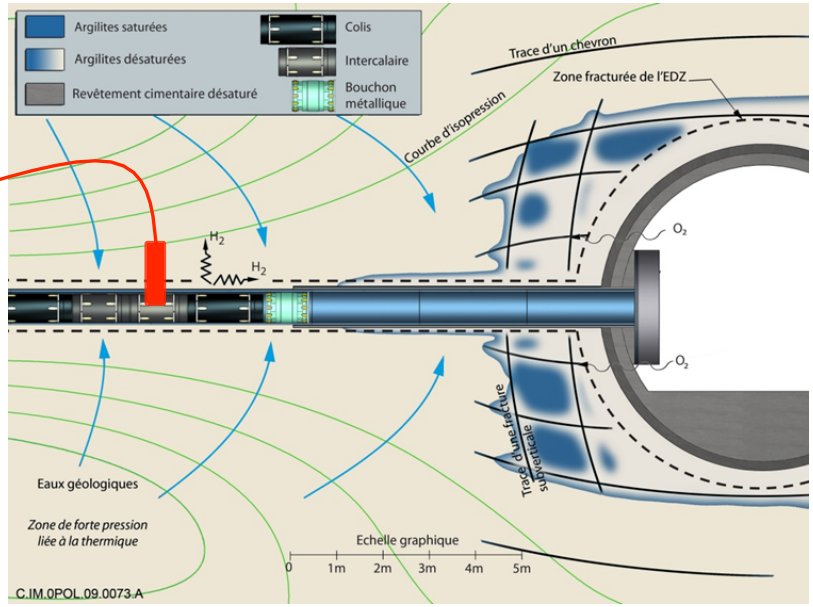
Steel in the geological disposal is present at different levels and in different forms:

- Iron concrete, bolts, liner to assure mechanical stability of the galleries/cells and ILW-LL overpack
- HLW overpack to delay the coming of water

Multi interfaces between steel components and environment

- Voids
- Cement
- Argillite

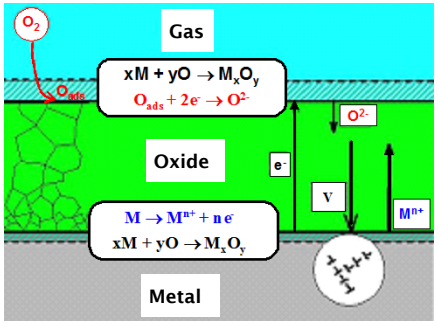


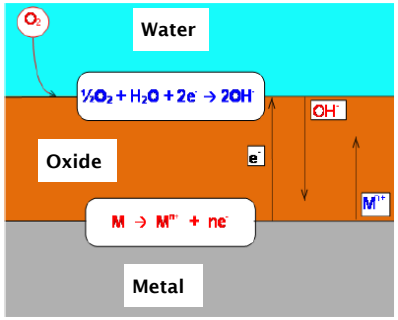


Context

Corrosion processes are function of thermodynamic and fluid composition environment:

- Dry corrosion
- Water corrosion:
  - atmospheric corrosion
  - aqueous corrosion





Corrosion modes are function of interface conditions:

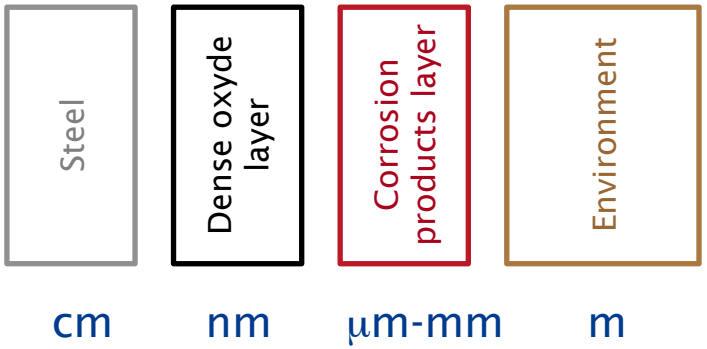
- General corrosion
- Localized corrosion
- …

Three macro-processes:

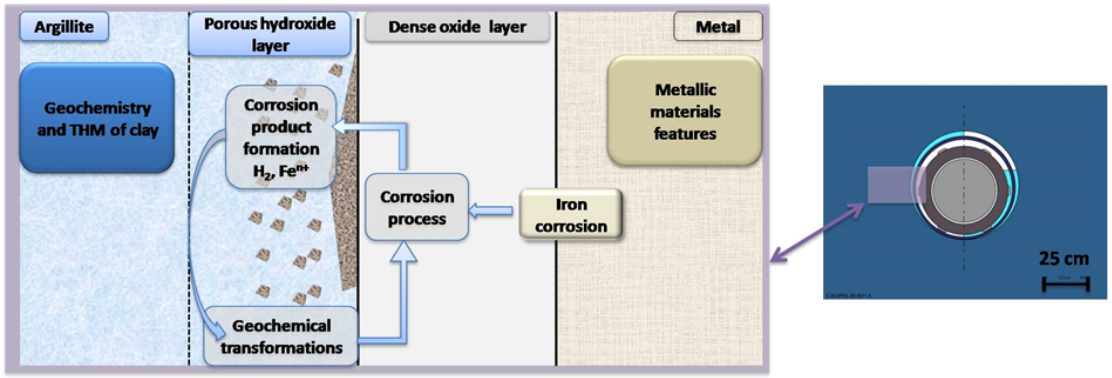
- Cathodic process: input and reduction of oxidizing species
- Anodic process: metal oxidation and metallic cation release
- Formation process and corrosion products growth

Context

- Space scale heterogeneity



- Coupled effects of physical processes
  - Electrochemistry
  - Geochemistry
  - THM (Thermal, Hydraulic and Mechanical processes)



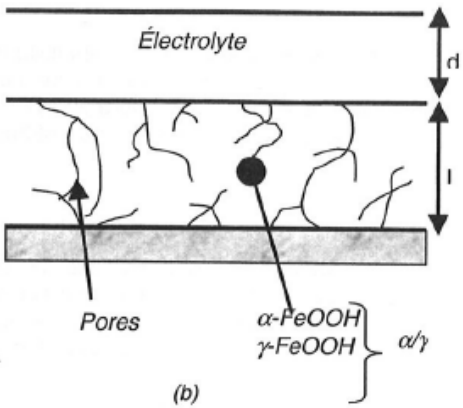
Needs multilevel modelling

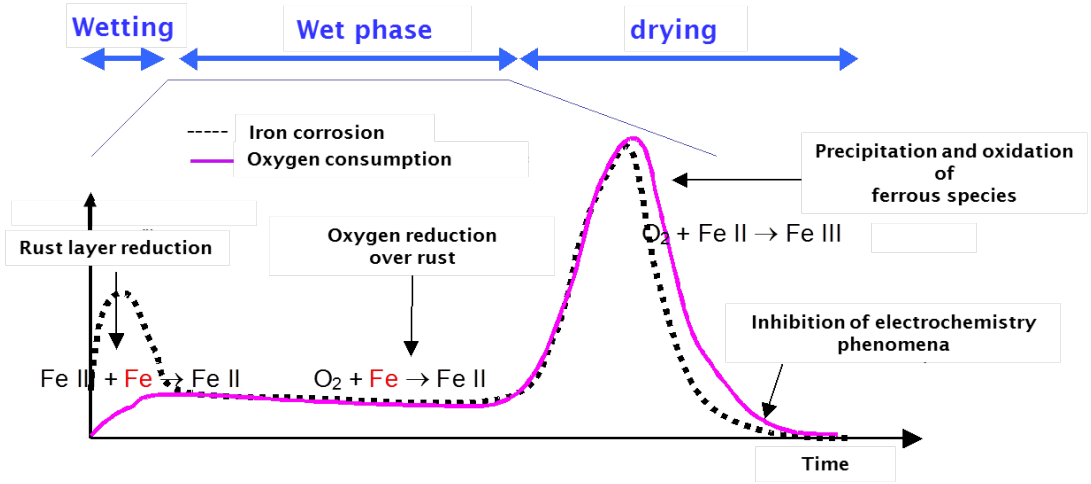
Oxide-hydroxide layer modelling

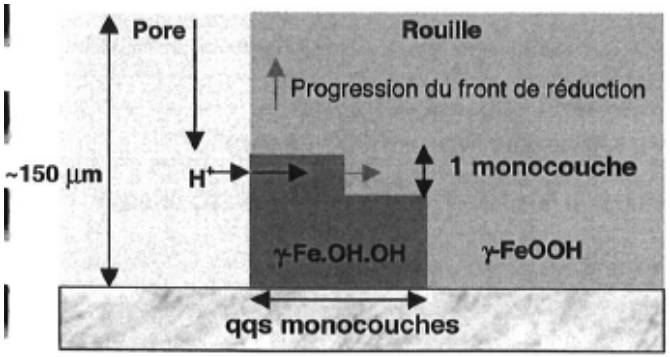
Oxide-hydroxide layer modelling: porous layer

Conceptualisation

- Modelling oxide-hydroxide layer (rust) result of the oxidation by dissolved oxygen in a water film (electrolyte)
- Electrolyte formation depends on air condition
- Consume iron Velocity depends on wetting-drying cycle







Oxide-hydroxide layer modelling: porous layer

Assumption

- A first rust layer exists
- Thickness rust layer is of constant size

Modelling approach:

- Modular approach (CEA / CAST3M)
  - Thermo-hydrodynamic module
    - Predict the characteristics of wetting-drying cycle
    - Assessment of the characteristics electrolyte film
  - Electrochemical module
    - mechanistic description of physical-chemical phenomena
    - Prediction of iron damage

Rust layer actively participates in corrosion mechanisms

- Its properties determine its electrochemical reactivity
- Its properties control the diffusive phenomena

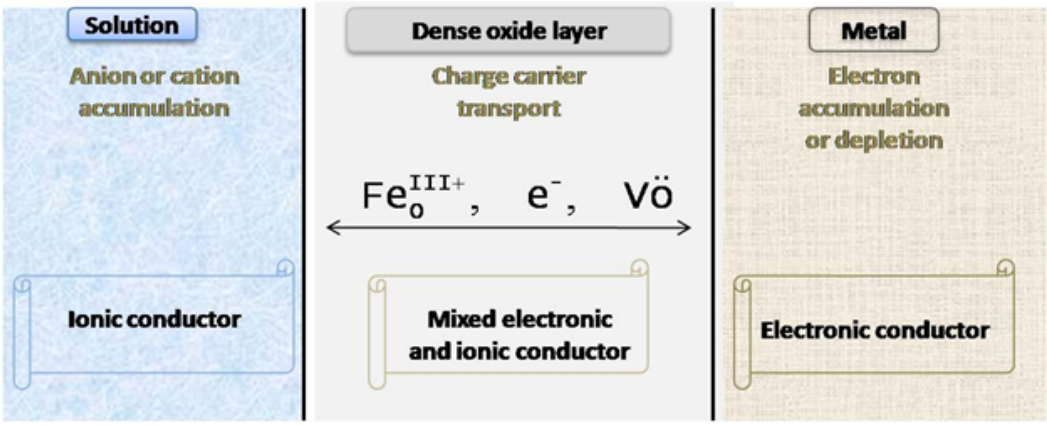
Oxide-hydroxide layer modelling: dense layer

Conceptualisation

- Modelling the oxidation of a metal covered by an oxide layer
- Dedicated software: Calipso (Andra/CEA)

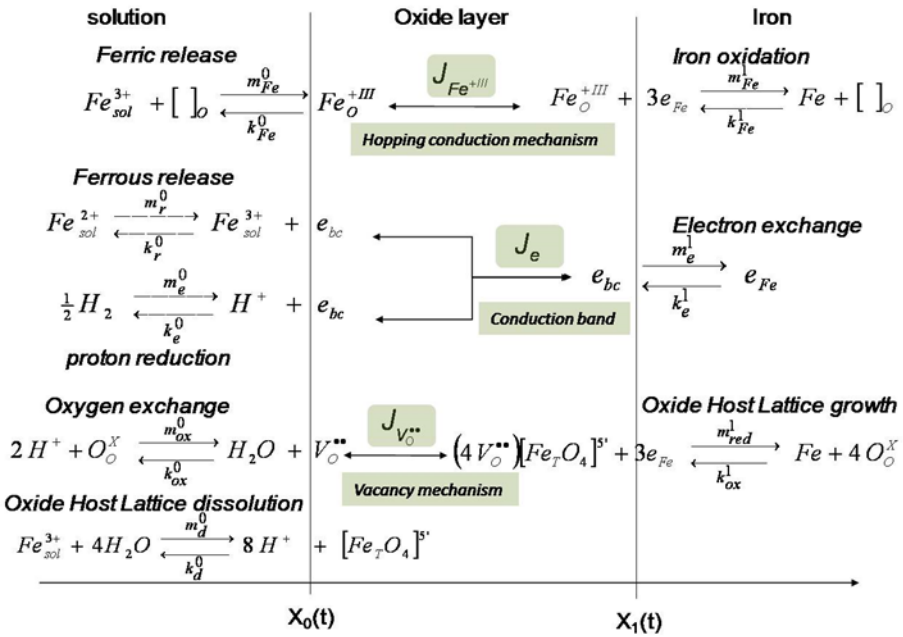
Assumption

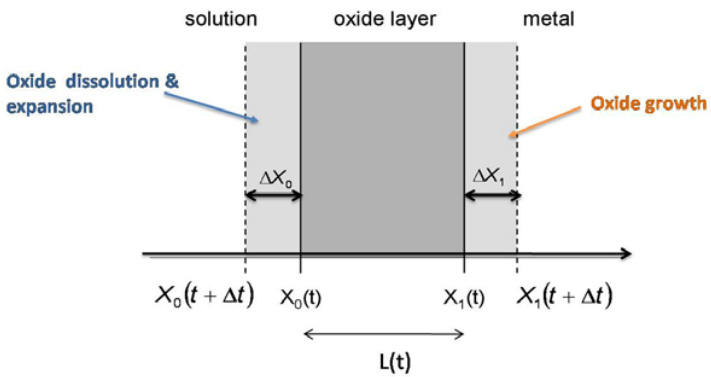
- The metal (iron) is assumed to be covered by a spinel iron oxide layer ($Fe_{(3-\delta)}O_4$)
- The metal is a pure electronic conductor
- The solution is a pure ionic conductor



Modelling approach

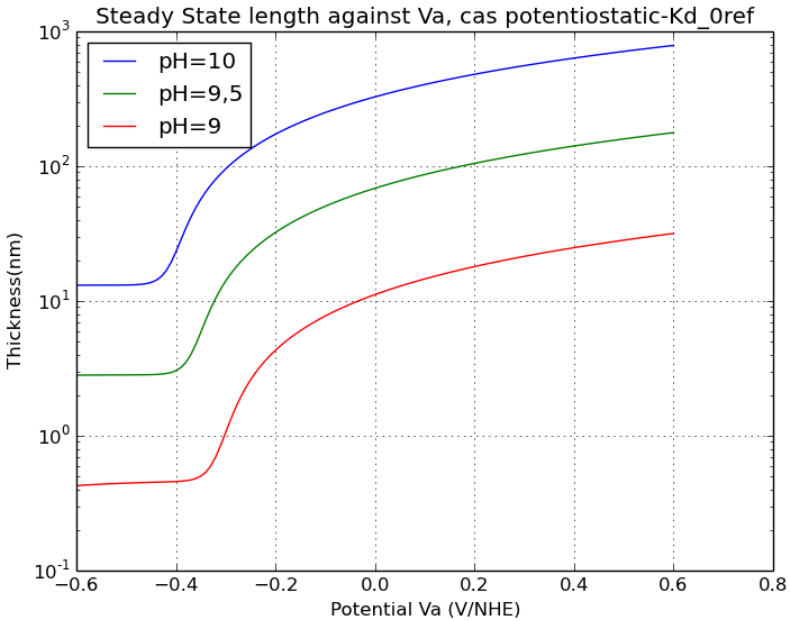
- DPC (Diffusion Poisson Coupled) Model (C. Bataillon – CEA)
  - convection-diffusion equations for the charge carrier concentrations coupled with a Poisson equation for the electric potential
  - discretization of the fluxes by the Scharfetter-Gummel scheme
  - Robin boundary conditions and variable boundaries



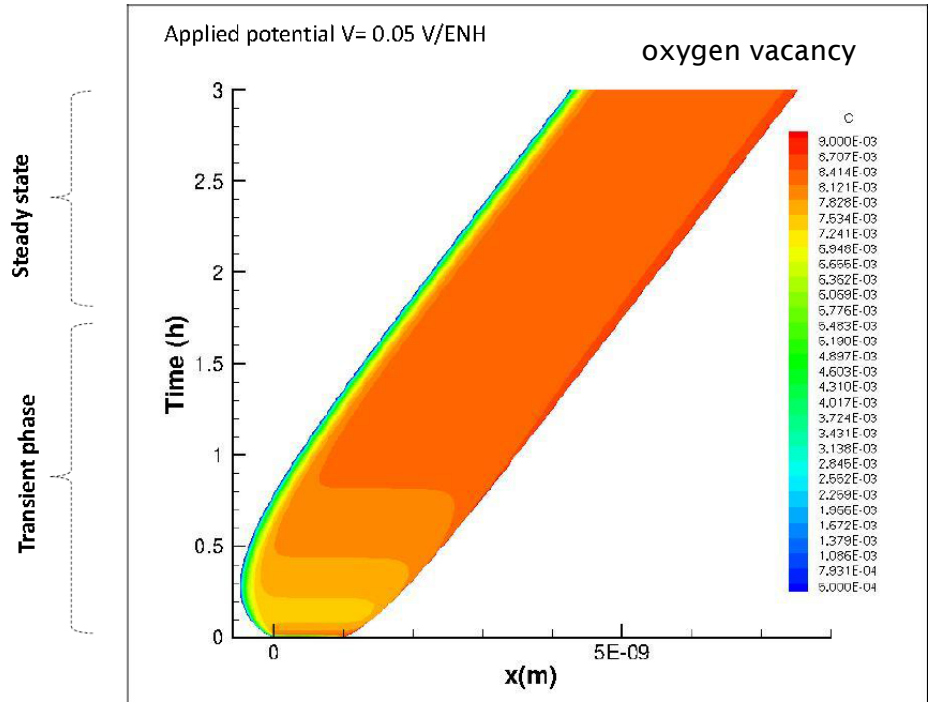


Interface motion

Some results



pH influence on oxide layer thickness



Temporal evolution of oxide layer

corrosion products layer modelling

Corrosion products layer modelling

Conceptualisation

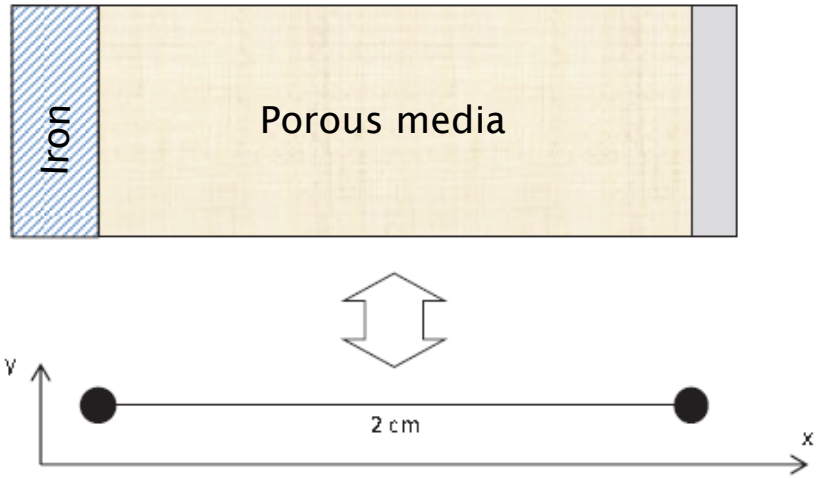
- Transport reactive approach
- Electrochemical laws of Butler-Volmer type (iron flux imposed)

Assumption

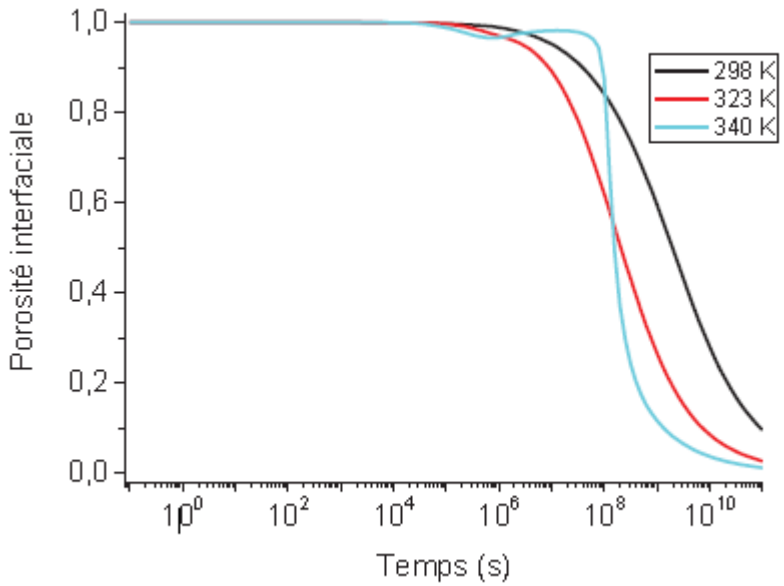
- Electroneutrality
- Chemical system reduced
- porosity changes are not taken into account

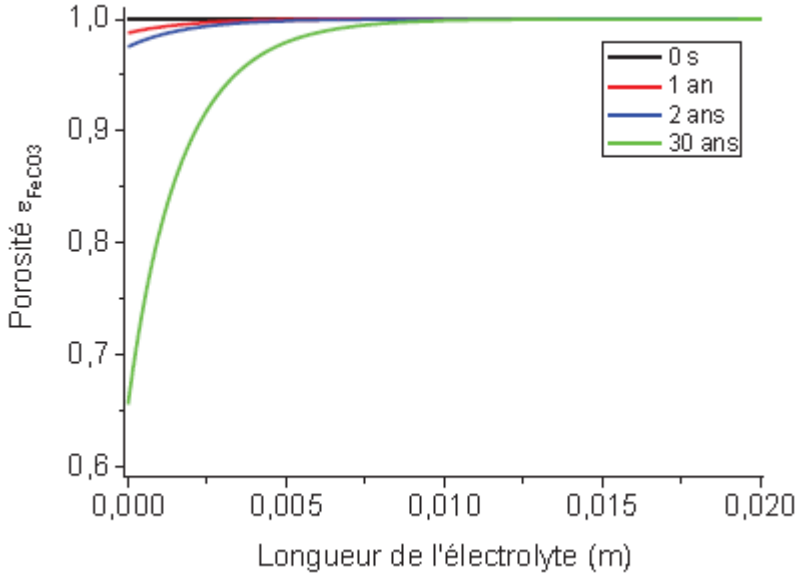
Modelling approach

- COMSOL
- Nernst-Planck equations



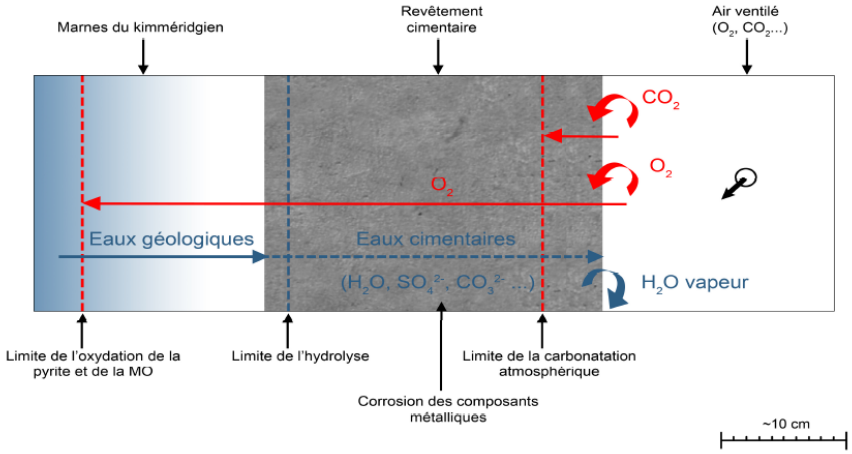
Some results: effects on the porosity

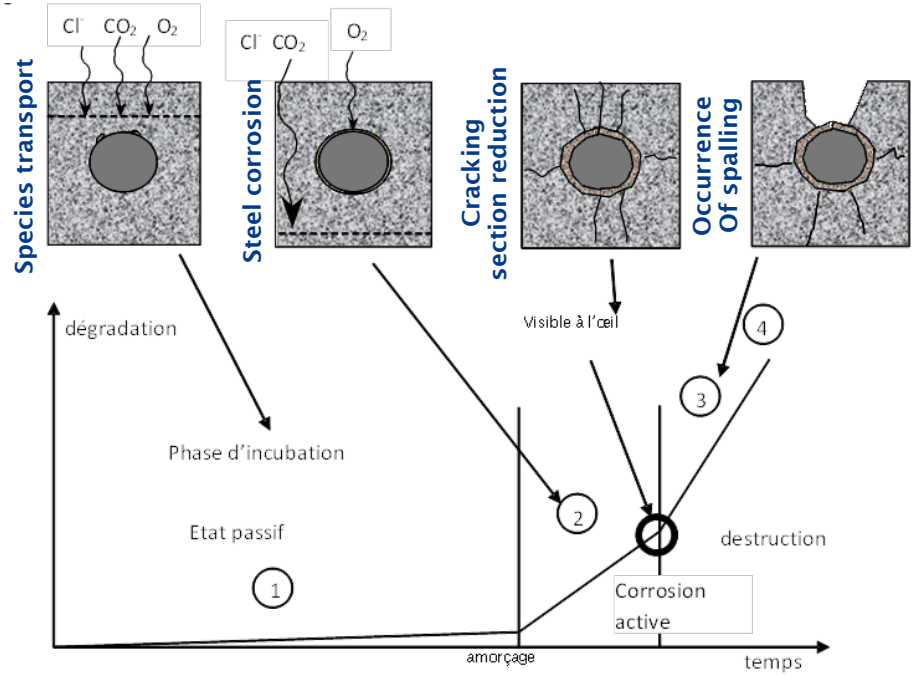




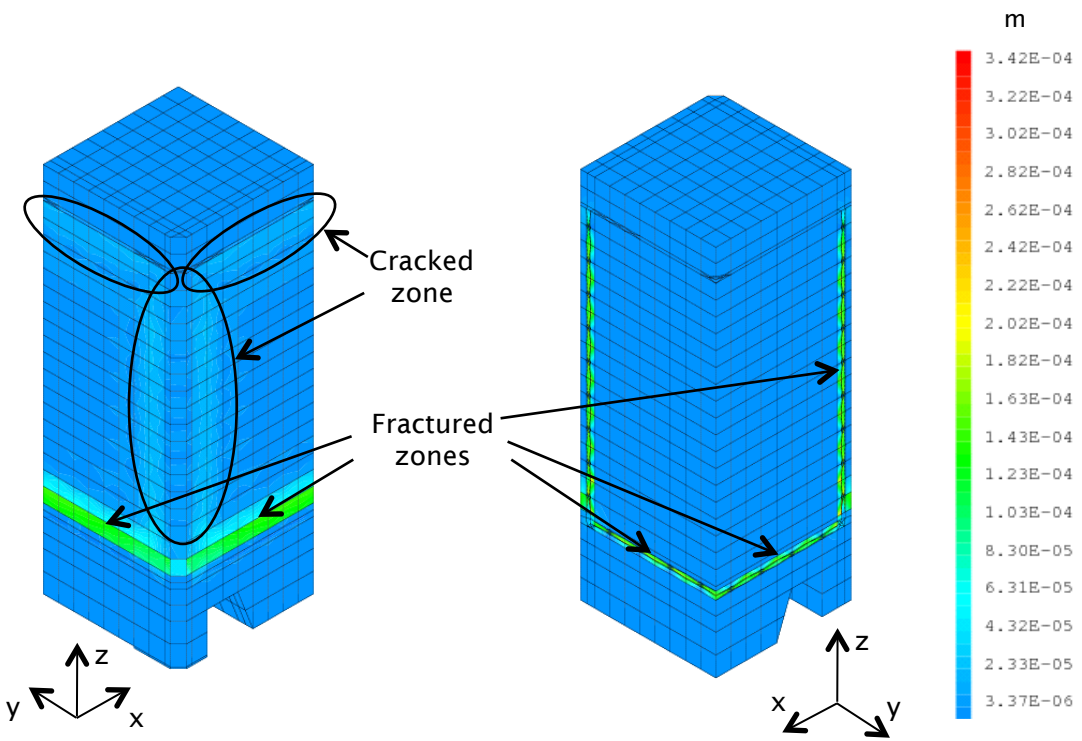
Conceptualisation

- Evaluation of the velocity of corrosion products layer growth are given by empirical formulation
- Relation between velocity and layer thickness
- Modelling mechanical constraints evolution
- Not numerical corrosion/reactive transport model





Example: ILW-LL overpack damage at 10 000 years



Electro-geochemistry coupling

2 levels

Electro-geochemistry coupling: 2 levels

Conceptualisation

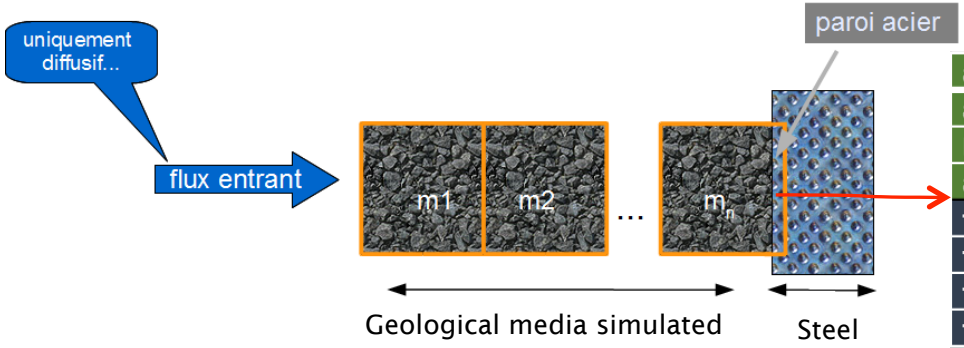
- Electrochemical / reactive transport coupling

Assumption

- Corrosion product layer are not simulated

Modelling approach

- Coupling Kirmat (1D reactive transport model – LHyGeS) / Calipso (corrosion model – Andra/CEA)



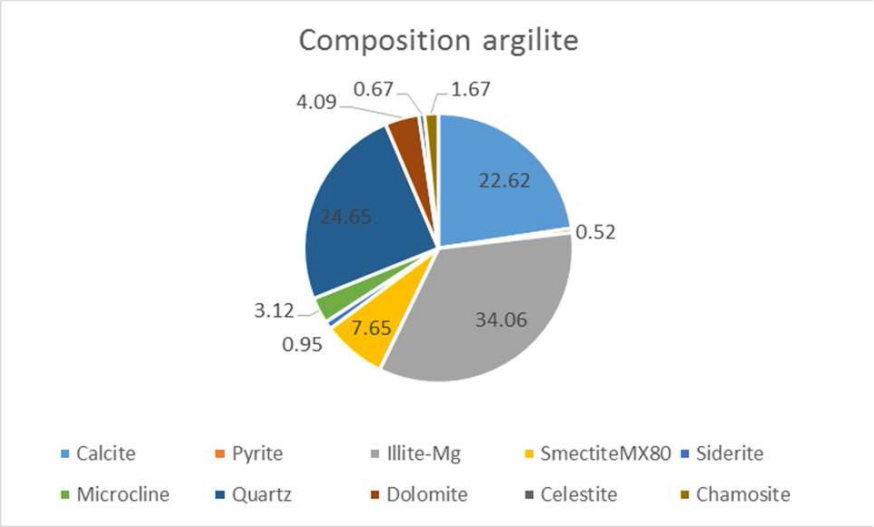
aFe2_input	Ion activity for Fe^{2+}
aFe3_input	Ion activity forFe^{3+}
Eh_input	Eh aqueous solution
aH_input	Ion activity for H^+
fFe2_retour	Flux Fe^{2+} from Calipso (mol/an/kg H_2O)
fFe3_retour	Flux Fe^{3+} from Calipso (mol/an/kg H_2O)
fH2O_retour	Flux H_2O from Calipso (mol/an/kg H_2O)
fH2_retour	Flux H_2 from Calipso (mol/an/kg H_2O)

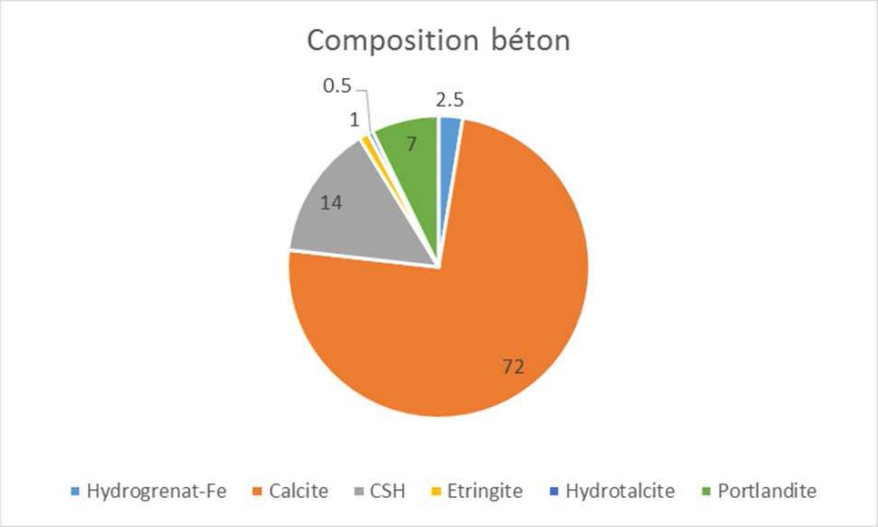
Some result: Test argillite/concrete/iron at 25°C

1 argillite zone:
Length: 1m (20 cells 0.05 m)
Section: 4E-4m2
Porosity : 0.18
Permeability : 1.62E-20 m2
Diffusion : 3.15E-3 m2/an
pH : 7.2
Eh: -171 mV

1 concrete zone:
Length : 0.04 m (4 cells 0.01 m)
Section : 4E-4m2
Porosity : 0.13
Permeability : 5.0E-21m2
Diffusion : 3.15E-3 m2/an
pH : 12.4
Eh: -50.5 mV

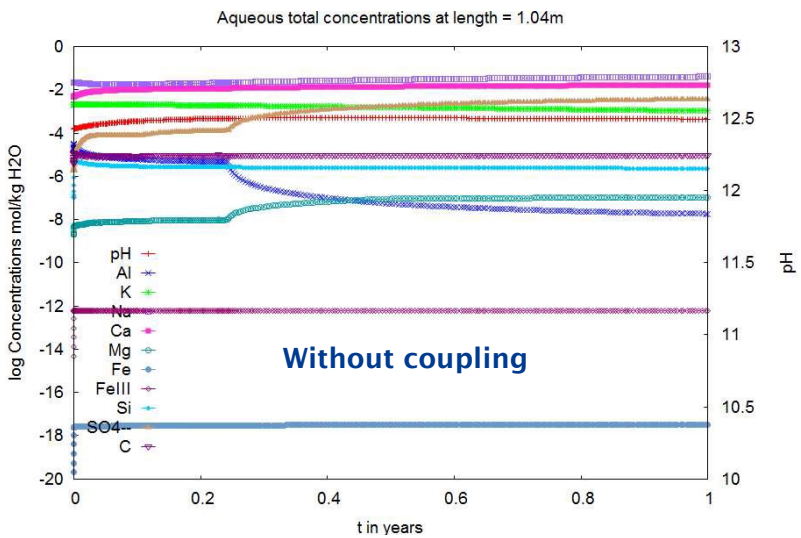
1 Iron zone:
Length : 0.01 m (1 cell)
Section : 4E-4m2
Porosity : 0.075
Permeability : 1.E-30m2
Diffusion : 3.15E-3 m2/an
pH : 12.4
Eh: -50.5 mV

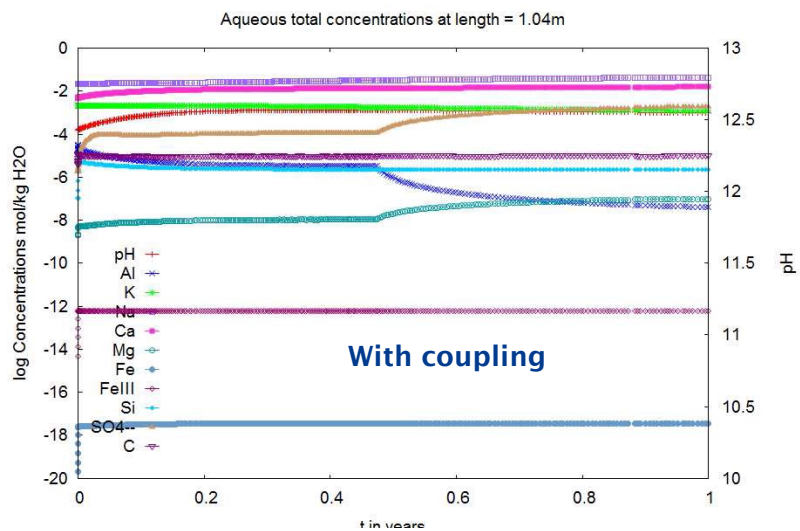


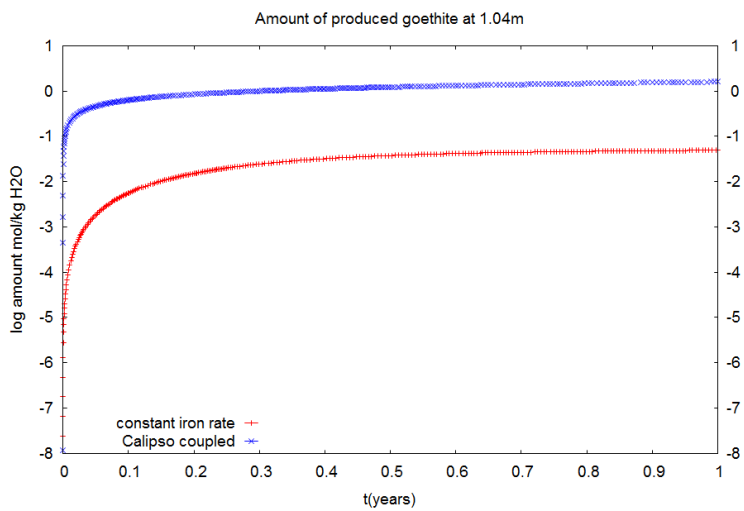


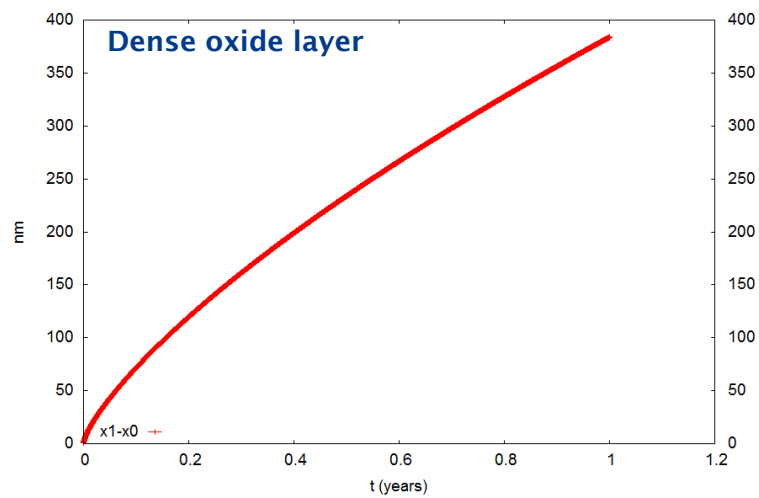
Electro-geochemistry coupling: 2 levels

Result after one year of reaction









Electro-geochemistry coupling

3 levels

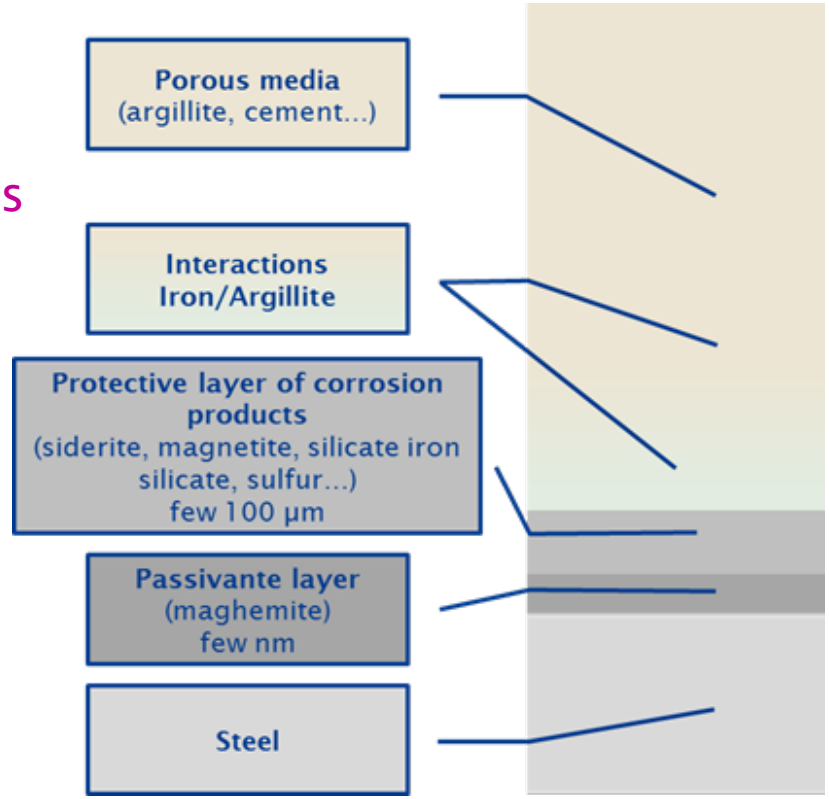
Electro-geochemistry coupling: 3 levels

Conceptualisation

- Electrochemical / reactive transport coupling
- Passivante layer modelling
- Protective layer of corrosion products modelling
- Geochemistry interaction modelling

Work in progress

- University of Bourgogne (ICB) thesis
- Support collaboration Andra/CEA/ LHyGeS



That’s all folks !