An Innovative Reactive Transport Modeling Approach for the Chemical Evolution of a HLW Cell in the Callovo-Oxfordian Formation

Jorge Molinero (jorge.molinero@amphos21.com), David García, Mireia Grivé, Albert Nardi

Amphos 21 Consulting S.L

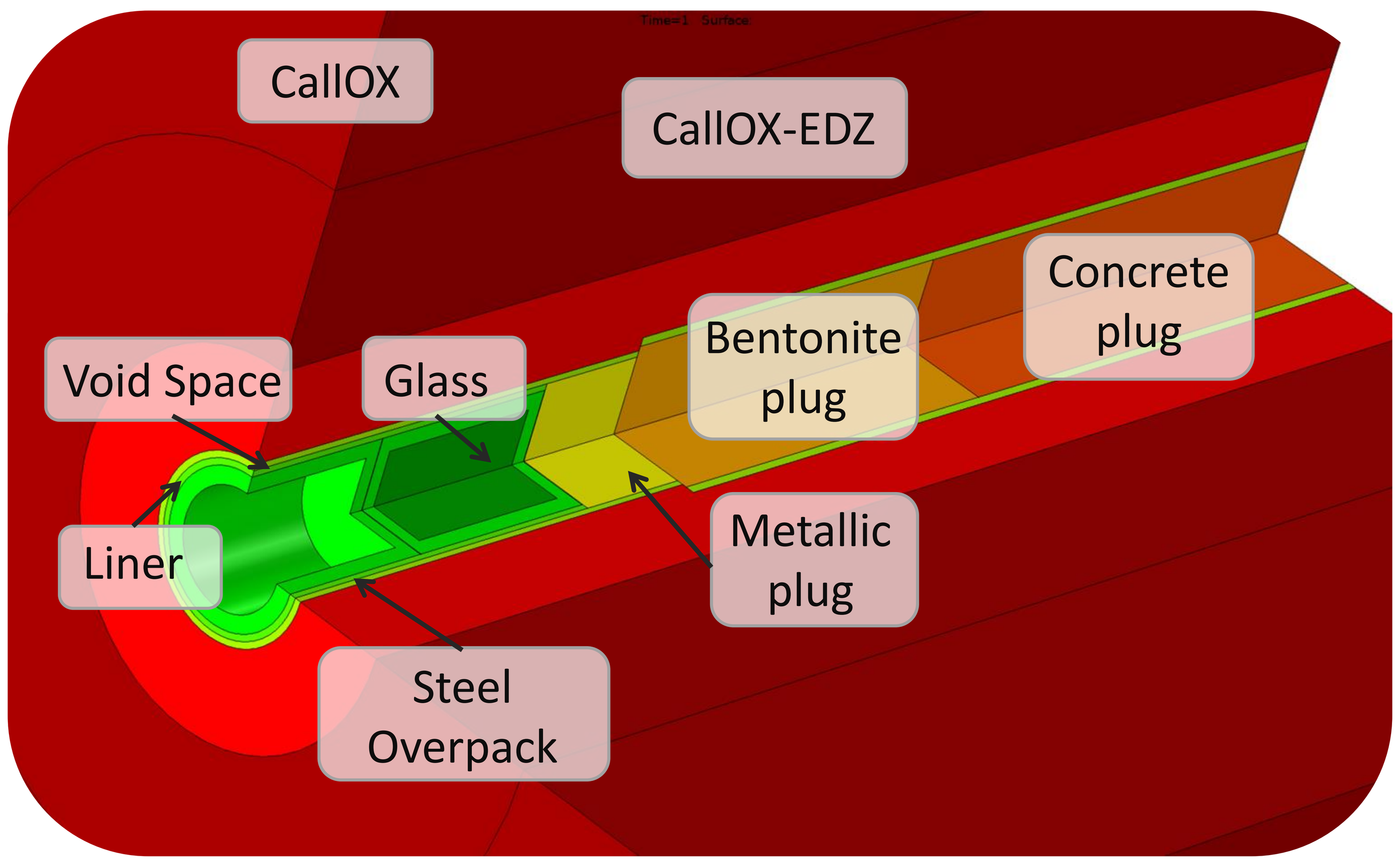
Introduction

Andra (The French National Radioactive Waste Management Agency) envisages the safe disposal of High-Level Waste (HLW) and Intermediate-Level Long-Lived Waste (IL-LLW) in deep geological storage using a multibarrier system. To ensure the containment of radioactivity, the principle of storage is based on a clay formation with low permeability, homogeneity and continuity (i.e Callovo-Oxfordian (CallOX) formation), properties that delay and limit the dispersion of the waste [1].

The objectives of this work have been to develop and settle up a geochemical model for quantifying the thermo-hydro-chemical (THC) processes that could be later applied in reactive transport calculations to precisely evaluate the chemical interaction and migration of different radionuclides.

Geometrical Set-up

The geometrical set-up of this modelling work is based on the scheme of an HLW after closure defined in the Dossier Jalon 2009 [2].



Geochemical System

A complex geochemical system has been considered for each geometrical domain including radionuclides (Cs, Se, Tc and U) solubility control as well as sorption on and exchange within mineral phases.

CallOx / CallOx EDZ
Illite
Smectite
Chlorite
Microcline
Pyrite
Quartz
Calcite
Dolomite
Siderite

CallOx / CallOx EDZexchange
NaCox
KCOx
$CaCox_2$
$MgCox_2$
$FeCox_2$
$SrCox_2$

Steel Overpack, Liner, metallic plug
Steel dissolution rate
Magnetite
Siderite
H2(g)
Calcite
Greenalite
Crondstedtite
Berthierine
Lizardite
Vermiculite

Concrete Plug
CSH1.6
Ettringite
Calcite
CSH0.8
Etringite-Fe
Lizardite
SiO_2(am)
Magnetite
Greenalite
Crondstedtite
Berthierine

Bentonite Plug
Montmorillonite
Quartz
Albite
Microcline
Pyrite
SiO_2(am)
Calcite
Siderite
Magnetite
Greenalite
Crondstedtite
Berthierine
Vermiculite

Bentonite plug exchange
NaX
KX
CaX_2
MgX_2

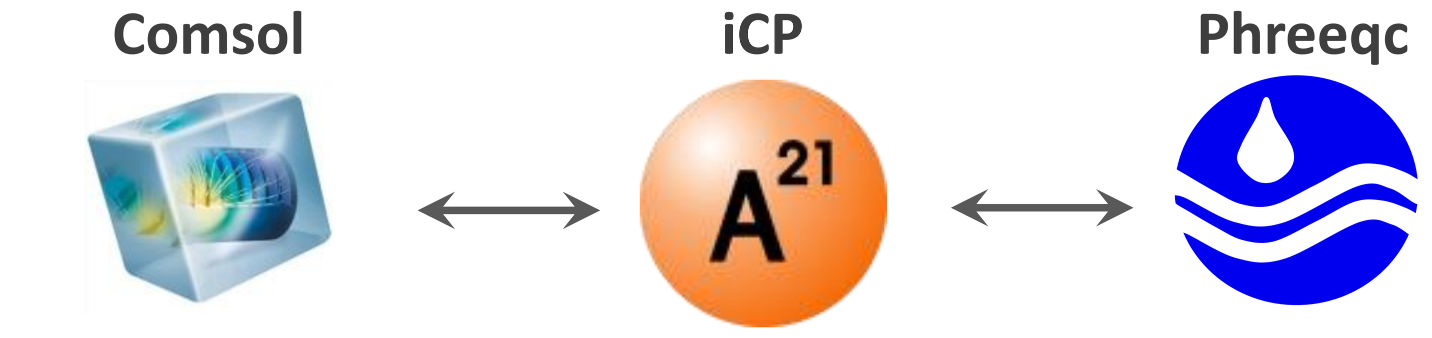
Bentonite plug surface reactions
SsOH
Sw_1OH
Sw_2OH

Glass
Glass dissolution rate
SiO_2(am)
Vermiculite
Calcite
Saponite
Borax
Magnetite
Siderite
Greenalite
Crondstedtite
Berthierine

Void
Calcite
Siderite
Magnetite
Lizardite
Vermiculite
Greenalite
Crondstedtite
Berthierine

interface Comsol-Phreeqc (iCP)

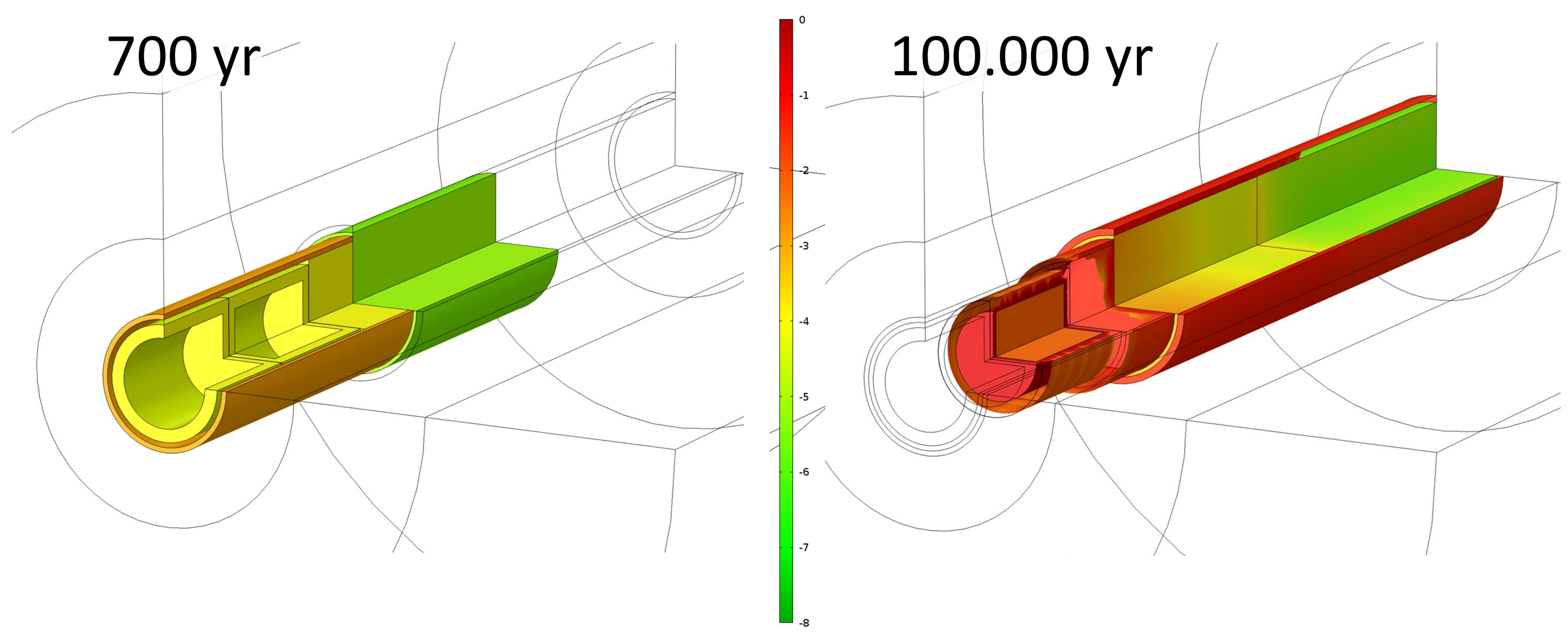
All the geochemical processes considered in the conceptual model have been implemented in a powerful high performance computing framework, based on the coupling of COMSOL and PHREEQC [3] simulators [4].



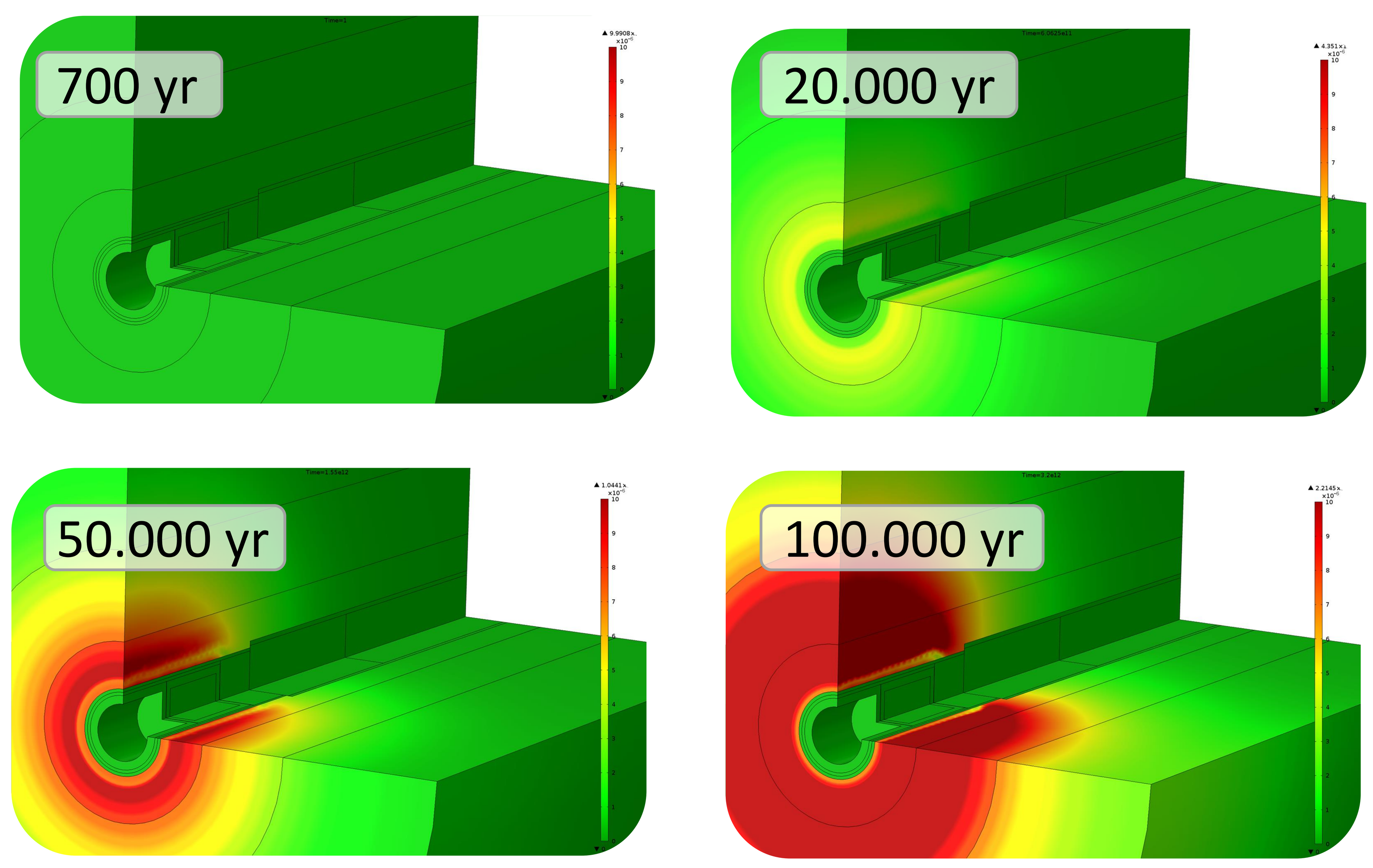
Results

Mineral, sorbed and aqueous specie concentrations evolution up to 100.000 years have been calculated.

Logarithm of greenalite molality



Retention of Cs as function of time within CallOx exchange positions.



Conclusions

- Long-term simulations, up to 100,000 years, have been performed successfully.
- Results obtained are key for understanding the system evolution.

References

[1] Andra (2013) Projet CIGÉO. Andra 504-DCOM/13-0028.
[2] Andra (2009) JALON 2009. C.NT.AHVL.09.0005.B
[3] Parkhurst, David L., and C. A. J. Appelo. "User's guide to PHREEQC (Version 2): A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations." (1999): 312.
[4] Nardi, A., Trinchero, P., de Vries, L., Idiart, A., & Molinero, J. (2012). Coupling multiphysics with geochemistry: The COMSOL-PHREEQC interface. In COMSOL Conference, October.