NUMERICAL SIMULATION FOR NUCLEAR WASTE MANAGEMENT

M. Caron-Charles, G. Bernard-Michel, M. Filippi, P. Montarnal
CEA Saclay
91 191 Gif /Yvette FRANCE

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ABSTRACT

CEA and ANDRA have jointly developed the simulation platform Alliances, dedicated to radionuclide transport computing in porous geological site. It is designed for achieving three objectives: performance and safety studies, modeling and numerical research, capitalization of knowledge. Modeling results are presented, dealing with extended transport and reactive transport. Then, thermal conductivity of clay is estimated using inverse method.

CONTEXT

One of the challenges of the nuclear energy industry is management of waste, especially high level radioactive waste.

To assess a solution for waste repository, radionuclide release and transport must be estimated for different situations, taking into account coupling physical phenomena occurring for different distance and different time scales. The final objective is predictive simulation of the transport in the whole of the geological site, meaning to large scale (> 1 km) and for large time (> 100 millions of years).

Among the solutions studied by the French agency (ANDRA) and CEA, high radioactive waste might be stored in underground formation of porous clay.

The underground storage concept proceeds with successive barriers around the waste canister: engineered barrier then geological barrier selected for their transport properties, in order to minimize transfer to the geological surrounding.

The storage concept is very modular, and divided into compartments. So that phenomena are analysed in each compartments: distinction is made between near field and far field. In the same way, elementary phenomena take place at different time, allowing therefore simplification for the numerical coupling. Two axes may then be derived for the whole problem resolution: length scale and time scale.

A simplified representation of an underground repository might be as follows. Nuclear wastes are confined in container, either concrete for middle radioactivity or glass matrix for high level. The containers are then set up into modules and insulated from the host rock using an engineered

clay barrier, that absorbs water and fully expands to act as a sealing material. Galleries are provided for access in the repository site.

As the design is modular, it allows the study of physical phenomena occurring after the storage unit closure, chained or coupled: thermal hydraulics, mechanics, chemical reactions. Radionuclide transport must be then evaluated for different situations, fully depending on the time and space scales [CNE].

Container degradation provides the source term for radionuclide release. Degradation may be controlled by chemical kinetics, such as corrosion for canister, or glass / gel transition for glass matrix.

For the radionuclide (RN) transfer in the surrounding medium, distinctive RN comportment are to be taken into account, depending on their ionic state and their specific chemical retention with the mineral phase, including also the isotopic decay, radioactive specie generation process…..

NUMERICAL SIMULATION

As experimental measurement can only provide part of the information, owing to the complexity of the phenomenology, and most overall because of the time scale involved for performance assessment, numerical simulation is the best fit solution for extrapolation then prediction.

Many difficulties are encountered in the numerical simulation of RN transfer in porous media. First, the global problem to be solved involves many physical phenomena, at different space and time scale. Local phenomena must be appropriately described, such as the RN release source term, but flux or RN concentration must be evaluated at every point or frontier of the geological site, over than 20 km. Then boundary conditions may differ depending of the site frontier, and may vary as a function of time too. As for the RN concentration values, they range from high value in the near field to very low value in the far field (10^{-13} m/l).

The usual difficulties encountered are bounded to meshing deformation (non regular meshing), coarse gird (till 500 000 meshes), and non stable numerical solution leading to concentration oscillation or negative concentration, particularly for low values.

Numerical Patform

CEA and ANDRA have jointly developed the simulation platform ALLIANCES dedicated to radionuclide transport computing in porous media. It is designed for achieving three objectives: performance and safety studies, modelling and numerical research, capitalisation of knowledge.

The simulation platform proceeds by integration of specific codes for simulation of radionuclide transport in porous media, using the same data model (Python script). It is important to notice that these codes are selected as the best ones for specific modelling, before their integration in the platform [Bengaouer A. Tallandier J et al 2003].

The numerical platform is designed with three successive layers:

- The basic layer manages the whole computing sequence: supervisor, meshing, displaying tools, physical data inputs….
- Integration schemes, coupling algorithms and parameter uncertainties evaluation are included in the second layer
- Then specific modules such as container degradation models, or extended transport, or chemical models, or sensitive analysis models defined the basis modules.

Since the numerical platform is designed for performance assessment computing, leading to more than 1 000 calculations, industrial characteristics are necessary: all geometry meshed for multi –dimensional calculations, human machine interface, results display, computing time as short as possible, memory size as small as possible……

Currently, the physical models implemented are related to RN transport in saturated conditions.

The use of specific codes for each physical phenomena, provides the best estimation for the key process simulation. Then, since a single data model is used for all the codes, this easily allows specific couplings or chains.

As a first example, chaining a container degradation model and an extended transport model including radioactive chains, gives an estimation of RN flux in the surrounding geological media, from which a sensitive analysis may be derived.

Another example is the so called reactive transport, solved by the coupling between a transport model and a chemical model. It ensures the best estimation for the modelling of the chemical interaction between the flowing aqueous phase and the mineral phase.

Another important characteristic is the disposal of different codes and different numerical methods for solving a same physical model. For example, two codes may be chosen for extended transport simulation: Porflow (ACRI / USA) and Castem (CEA/ France).This enables result comparison then ensures a best confidence for the prediction. Likewise transport, reactive transport is solved through two different couplings (two transport codes and two chemical codes): MT3D / Phreeqc and Castem/Chess.

Special attention is paid to the qualification and validation program [Mügler C. Miguez R et al 2003]. It proceeds encased tests, with a gradual complexity, first comparing calculation to analytical solution, then defining benchmark exercise, between different modules implemented in the platform or external ones. Then, all the tests achieved are stored as reference test, available for the users.

Extended Tansport

Extended transport solved the Darcy equation for water flow in porous media, and a convection / diffusion equation for RN transport. Castem and Porflow components of Alliances include also radioactive chains, linear or non linear ones, radioactive decay, dissolution and precipitation, and

then chemical retention modelled through a delay factor. These are the major phenomena occurring all over the RN migration in the geological site.

Castem component is developed in CEA [Bernard-Michel G. Le Potier C. 2003]. For the spatial discretization, two numerical schemes are available: Mixed Hybride Finite Element (MHFE), and Finite Volume (FV). In fact, MHFE and FV methods have complementary properties with respect to stability and monotony, particularly for solving the diffusive part of the transport. Every problem may then be solved with the two algorithms, just setting up an option in Alliances.

Darcy flow:

$$\vec{U} = K\,gra\vec{d}\,h$$
$$\frac{\partial h}{\partial t} = div\vec{U} \qquad \text{(Eq. 1)}$$

with:

- K: permeability ; parameter or tensor for anisotropic or heterogeneous properties
- U: flow velocity
- h: hydraulic charge

Transport is solved using a generic equation :

$$A_1 \frac{\partial C}{\partial t} + A_2 div(A_3 gra\vec{d}\,C) + A_4 div(\vec{U}C) + A_5 C + A_6 = 0 \qquad \text{(Eq. 2)}$$

with A_1 A_2 A_3 A_4 A_5 A_6 the fields of transport properties including:

- porosity
- chemical retention or delay factor
- diffusion / dispersion: tensor for anisotropic or heterogeneous properties
- radioactive decay
- precipitation/dissolution kinetics constant
- source term

This generic equation is solved at each step: either a time step, or an iterative step of the linearization phase. This very modular design allows to solve transport for a chemical specie or for each elementary isotopic form of the chemical specie. It will also be applied to unsaturated transport resolution, since the A_i fields have no predefined form. The linear sparse system, which is not symmetric due to the convection operator, is solved either with a BCGStab or a GMRES algorithm, after the matrix preconditioning with ILU algorithm. Different boundary conditions may be taken into account: defined concentration, defined flux or mixed concentration and flux conditions, constant or time variable.

The ANDRA Couplex 1 Validation Test in Alliances

This case is specially defined for codes validation. Iodine 129, a very mobile radionuclide, is released from a storage unit through a virtual geological site, composed with four successive layers with different transport properties. The dimensions: h = 625 m and L = 25 km, leading to very flat meshing cells, as well as the boundary conditions and the simulated physical time duration: 10^7 years, arise strong difficulties for numerical simulation.

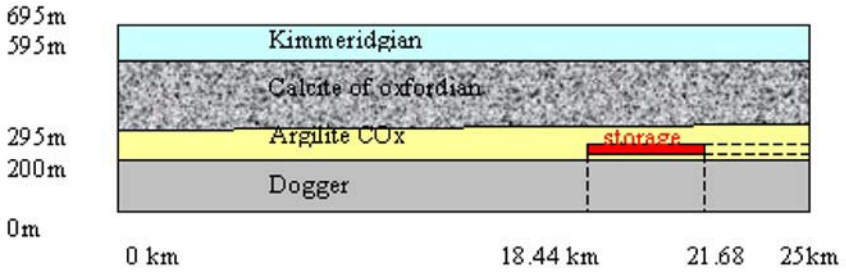


Fig. 1 Geometric description of couplex 1 test

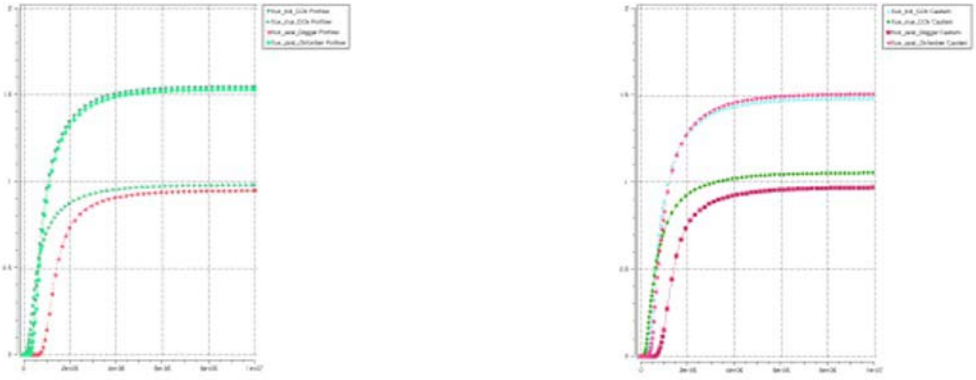
This case is successfully solved in Alliances with the two components Castem (MHFE and FV) and Porflow (FV).

The transport equation is derived from Eq. 2:

$$R\omega(\frac{\partial C}{\partial t}+\lambda C)-div(D\vec{\nabla}C+\vec{U}C)=s \qquad \text{(Eq. 3)}$$

with:
R: latency retardation factor
ω: porosity
λ: radioactive decay
s: source term
D: diffusion/dispersion tensor

The results comparison confirms the codes predicting capacity for Iodine transport in representative coarse scenario and assesses their validation. In the example, mass conservation is clearly stated since instantaneous flux curves are similar for the inner and outer frontier. Then total cumulative mass fluxes simulate Iodine release at the geological site frontiers with a very good accuracy for the two codes.



Porflow component Castem component

Fig. 2 Castem / Porflow Comparisons With Alliances; Cumulative I^{129} Flux Calculated on the Different Frontiers of the sSimulated Domain for Couplex1 test for $t< 10^7$ Years

Reactive Transport

The dissolved radioactive species flowing in the liquid phase, according to a onvection/diffusion process, may chemically react with the mineral solid phase. This is the chemical retention. But chemistry time scale is far much smaller than the transport one, so that one may assume an equilibrium chemistry between the solved and the sorbed concentration in the two phases. For low concentration level, chemical retention is well described with a thermodynamical function Kd, either linear, or Langmuir or Freundlich isotherm function. But this approach, used in the extended transport, is a global approach, not convenient for complex situations, nor specific RN such as Cesium [Jaquier J. et Ly J. 2003].

In Alliances numerical platform, reactive transport may be modelled through a coupling between a specific chemical code and a specific transport code [Montarnal P. 2003]. The chemical codes (Chess or Phreeqc) compute the chemical form of each specie according to the main involved processes: redox reactions, adsorption or interfacial interaction (surface complexation, ion exchange…..), precipitation / dissolution, stating a thermodynamical equilibrium. This ensures a complete description of the geochemical system, very convenient for reactive transport in the near field.

In fact, near field means to the first barrier around the waste container. Then all the RN may flow there and concentration level may be high enough for avoiding a Kd like global approach.

The following example proposes the modelling of Portlandite (concrete form) dissolution by clayed water in a convection/dispersion (1D) process.

The transport equation is derived from Eq. 2:

$$\omega\frac{\partial C}{\partial t}+\omega\frac{\partial F}{\partial t}-div(D\vec{\nabla}C+\vec{U}C)=0 \qquad \text{(Eq. 4)}$$

with:

C: concentration in the mobile phase
F: solid fixed concentration

The distribution between C and F is provided by the chemical code.

The chemical system comprises 5 component species, 18 secondary species in the liquid phase, 4 mineral species and 3 gaseous species. The results clearly display the dissolution interface progression in the concrete liberating calcium. As calcium is dissolved in the clay water, it precipitates as displayed by the second interface. The pH evolution is calculated too, ranging from pH = 7.8 in the clayed water to pH = 12.2 inside the Portlandite.

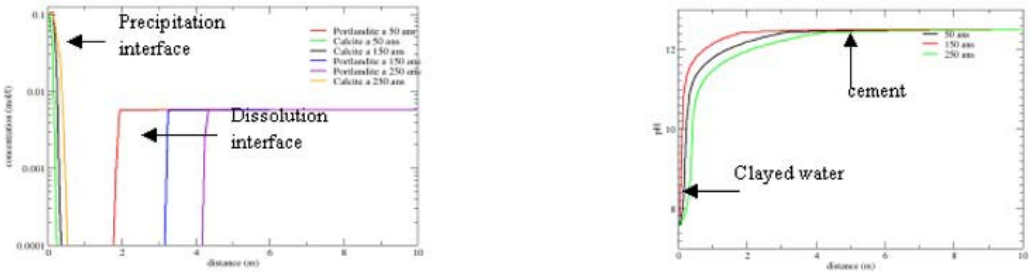


Fig. 3. Cement Dissolution by Clayed Water Calculated With Alliances Coupling Algorithm Castem/Chess; Fixed Concentration for the Calcium in the Geological Medium, Calcium Dissolution in the Cement Phase, Then Calcium Precipitation in the Water Phase; Ph Evolution.

Parameter Identification Using Inverse Method

One of the problems arising in radionuclide transport simulation is strongly bound to the uncertainty of the transport properties. In fact, experimental measurements are not always available for geological site, or local heterogeneity may affect the average values. Numerical methods, such as inverse methods, are then very useful for parameter identification. They are applied to in situ experiments, proceeding by minimization of the deviation function between calculated variables and experimental measurements.

The following example deals with the thermal conductivity identification of Opalinus clay. A heating source, temperature controlled, is located in a central borehole. Two other boreholes (index C2 and C3) drilled 50 cm away from the central borehole, and each fitted with 5 thermocouples, provide continuous temperature measurements till reaching the thermal steady state.

Three parameters are numerically estimated:

- The longitudinal and transverse thermal conductivities: λ_L and λ_T that have different values because of the geological anisotropy
- The thermal power loss of the heating source: α, since only part of supplied thermal power is delivered.

The optimisation code KALIF [Martinez JM] defines an experiment plan, proposing different set of values for the three parameters λ_L ; λ_T and α, that are then successively included in the direct calculations. The best set of parameters means to the minimization of the cost function:

$$J = \frac{1}{\sum_i a_i} \cdot \sum_i a_i (T\exp_i - Tcal_i)^2 \qquad \text{(Eq. 5)}$$

For the current problem, 500 000 direct calculations have been performed, using neural network method, in order to short the CPU time.

The three parameters are estimated with a rather good confidence as displayed on Fig. 4. In all the cases, the validation of the simplified solution provided by neural network, is checked through direct calculation with Castem code.
For the thermal loss coefficient: α = 0.77 +/- 3%.

Then, parameter identification clearly notices a dissymmetry for the two boreholes C2 and C3, with respect to the transverse thermal conductivity:

- Borehole C2: λ_L = 1.84 +/- 6% W/K.m and λ_T = 0.55 +/- 9% W/K.m
- Borehole C3: λ_L = 1.90 +/- 6% W/K.m and λ_T = 1.07 +/- 9% W/K.m

First interpretations conclude to a probable effect of the geological surrounding heterogeneity.

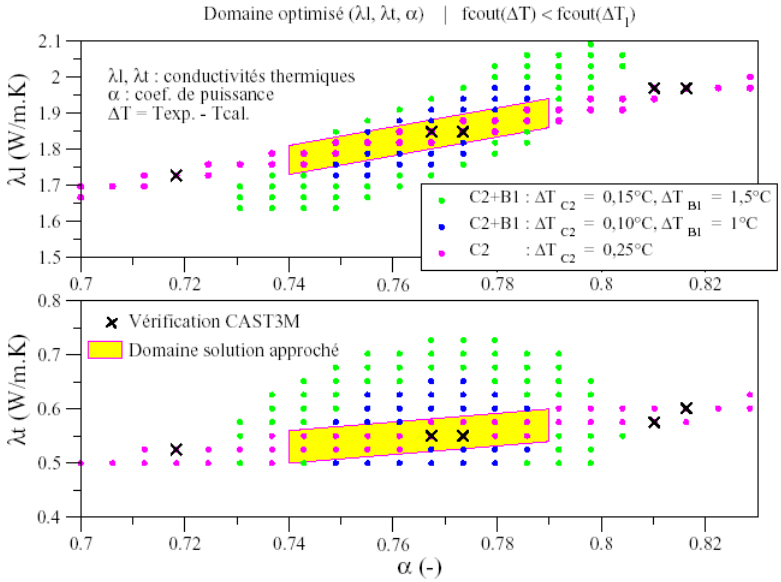


Fig. 4 Optimisation domain for the parameters α λ_l λ_t from temperature measurements; neural network solutions and validation with castem direct calculations

CONCLUSION

Alliances numerical platform is operative and available on a CD form, for PC / LINUX computer system.

The current implemented modules for transport modelling in porous media deal with extended transport and reactive transport in saturated conditions. The source term for RN release, may be derived from container degradation models. Then, a sensitivity analysis module is implemented.

New modules will be integrated in the numerical platform, to fulfil a complete description of the successive phases and coupled phenomena occurring in a waste repository: unsaturated reactive transport, thermo-hydro-mechanics……..

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