

## ThermoChimie project

# Development of a thermodynamic database for performance assessment calculations in relation to geological disposal of radioactive waste

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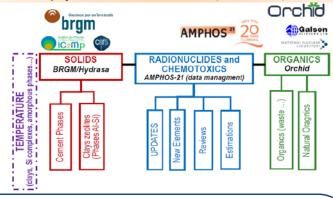
### **Consortium Andra-RWM**

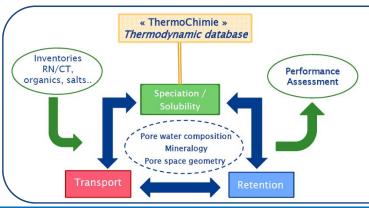
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ThermoChimie (TC) [1], is a thermodynamic database initially created and developed by Andra, since 1996. In October 2014, RWM and Andra formed a ThermoChimie project consortium to further develop the thermodynamic database.





#### **INTRODUCTION - OBJECTIVES**

Thermodynamic or geochemical modelling plays a substantial role in the approaches taken in **Performance Assessment of a Geological Disposal Facility (GDF) for radioactive waste.** Thermodynamic modelling is mainly used in assessing the geochemical evolution of the GDF in terms of the performance of the engineered barriers and host rock systems as well as the migration/retention behaviour of radionuclides. Such calculations require adequate conceptual and numerical models fed by reliable thermodynamic and kinetic data. These are the main building blocks of geochemical modelling and the **robustness of the thermodynamic data** used is mandatory for the accuracy of the geochemical simulation results.

The objective of the work is to identify, within the frame of the French underground repository concept and the UK generic GDF concept, the most significant geochemical environments from a Performance Assessment perspective. This constrains the range of conditions under which ThermoChimie must be applicable (i.e.: 5<pH<14, 15°C<T<100°C, SIT, water domain ...) [1] [2] [3] [4].

#### ThermoChimie database strengths

- ☐ Consistency: thermodynamic functions for a chemical systems
- □ Exhaustivity: ThermoChimie data on various systems ([1][2][3][4]), as:
  - > speciation and solubility limits for radionuclides and chemo-toxics,
  - rigeochemical processes under near-field and far-field conditions (with consideration of thermal, saline and organics perturbations),
  - > assessment of the processes of cement degradation,
  - > assessment of the processes of canister corrosion and stability of corrosion products (from iron steel)
- Traceability: each selected data is associated with an original data source
- □ **Usability**: compatible with numerical tools (PhreeqC, Crunch, Chess, Toughreact ...).

#### Priorities studies: program on 5 years (2014 - 2019).

- ☐ Temperature Corrections (a minima up to 90°C)
  - Clay Phases: smectite/illite transition (thermo-kinetic); fibrous clay; hydration/anhydrous models for clay minerals
  - Transition between amorphous phases and crystallizated phases (oxihydroxides; carbonates...)
  - Correction van't Hoff / HKF => challenge on ΔHr (RN, TC etc...)
  - « U(VI) Ca-CO<sub>3</sub> » aqueous complexes (up to 80°C)
- Stability of organics complexes with RN-TC in hyperalkaline conditions
  - U, Pu, Tc, Ni, Np, Pb, Ca, Fe, Sn, Al, Mg
  - with organics complexants: ISA, TBP/DBP, EDTA, Phtalic, Oxalic
- □ Ionic strength corrections (S.I.T. approach)
- ☐ Extension to thermodynamic data on chemical elements Be, Cu, Zn, La, Ac, Bi.

ThermoChimie covers the data necessary for various applications, including data on major elements, on radioelements, such as actinides and lanthanides, and chemotoxic metals

Aqueous species, solds, gases

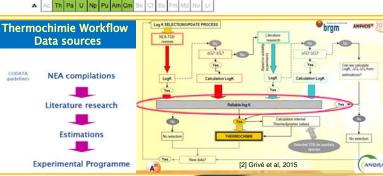
Actinides

X Actinides

X Actinides

X Short-lived radionuclides

X Radionuclides with half-lives longer than 1000 year of the properties of the pro



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[1] Giffaut, E. et al. (2014). "Andra thermodynamic database for performance assessment: ThermoChimie". Applied Geochemistry, 49, 225–236.

[2] Grivé, M., et al. (2015). "Thermodynamic data selection applied to radionuclides and chemotoxic elements: An overview of the ThermoChimie-TDB". Applied Geochemistry, 55, 85-94.
 [3] Blanc, P., et al. (2015) "ThermoChimie database developments in the framework of cement/clay interactions". Applied Geochemistry, 55, 9-107.

[3] Blanc, P., et al. (2015) "ThermoChimie database developments in the framework of cement/clay interactions". Applied Geochemistry, 55, 9-10 [4] Marty, N. C., et al. (2015) "A database of dissolution and precipitation rates for clay-rocks minerals". Applied Geochemistry, 55, 108-118.

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