# GEOCHEMICAL ENVIRONMENTS FOR THERMOCHIMIE DATABASE APPLICATION

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## Consortium Andra-RWM

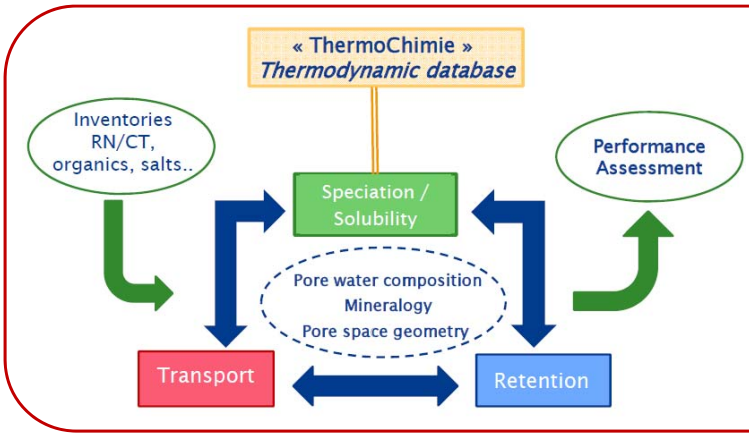
Thermochimie

https://www.thermochimie-tdb.com/

PARTNERS

ThermoChimie (TC) [1], is a thermodynamic database initially created and developed by Andra, the French National Radioactive Waste Management Agency, since 1996.

In October 2014, Radioactive Waste Management Limited (RWM) and Andra formed a **ThermoChimie project consortium** to further develop the thermodynamic database.




## INTRODUCTION - OBJECTIVES

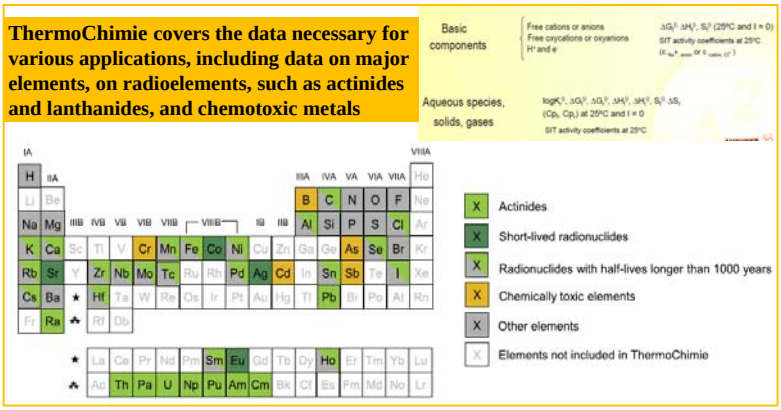
Thermodynamic or geochemical modelling plays a substantial role in the approaches taken in **Performance Assessment of a Geological Disposal Facility (GDF) for radioactive waste.** Thermodynamic modelling is mainly used in assessing the geochemical evolution of the GDF in terms of the performance of 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, 15ºC<T<100ºC, SIT, water domain ...) [1] [2] [3] [4].**

## ThermoChimie database strengths

- **Consistency** : thermodynamic functions are relevant but also consistent when taken together, among thermodynamic functions for one chemical reaction, and between thermodynamic functions for a chemical system
- **Exhaustivity** : ThermoChimie covers the data necessary for various applied purposes ([1][2][3][4]), as:
  - speciation and solubility limits for radionuclides and chemo-toxics,
  - geochemical 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, and if necessary the calculation used.
- **Usability** : data values and organization are compatible with the numerical tools to be used (PhreeqC, Crunch, Chess, Toughreact ...).

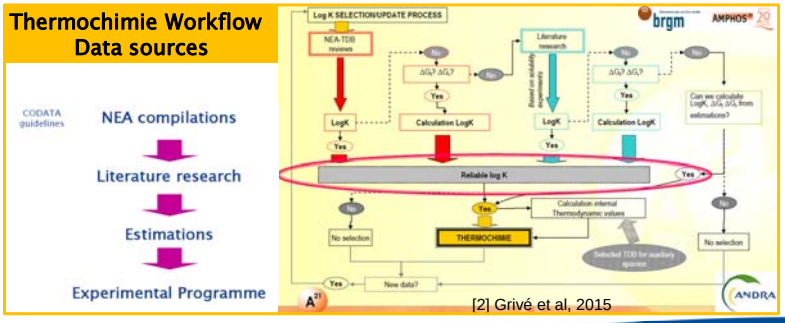
**ThermoChimie covers the data necessary for various applications, including data on major elements, on radioelements, such as actinides and lanthanides, and chemotoxic metals**




## 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 (oxi-hydroxides; carbonates...)
  - Correction van't Hoff / HKF => challenge on ΔHr (RN, TC etc...)
  - « U(VI) Ca-$CO_3$ » 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 Workflow**
**Data sources**




[2] Grivé et al, 2015

[1] Giffaut, E., Grivé, M., Blanc, P., Vieillard, P., Colàs, E., Gailhanou, H., Gaboreau, S., Marty, N., Madé, B., & Duro, L. (2014). "Andra thermodynamic database for performance assessment: ThermoChimie". Applied Geochemistry, 49, 225-236.
[2] Grivé, M., Duro, L., Colàs, E., & Giffaut, E. (2015). "Thermodynamic data selection applied to radionuclides and chemotoxic elements: An overview of the ThermoChimie-TDB". Applied Geochemistry, 55, 85-94.
[3] Blanc, P., Vieillard, P., Gailhanou, H., Gaboreau, S., Marty, N., Claret, F., Madé, B. & Giffaut, E. (2015) "ThermoChimie database developments in the framework of cement/clay interactions". Applied Geochemistry, 55, 9-107.
[4] Marty, N. C., Claret, F., Lassin, A., Tremosa, J., Blanc, P., Madé, B.; Giffaut, E., Cochepin, B. & Tournassat, C. (2015) "A database of dissolution and precipitation rates for clay-rocks minerals". Applied Geochemistry, 55, 108-118.

Radioactive Waste Management

ANDRA French national radioactive waste management agency

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