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ThermoChimie project

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

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Thermodynamic data are an essential input to geochemical modelling and often to the assessment of radionuclides and other pollutants behaviour in the Performance Assessment (PA) of a geological disposal facility for radioactive waste.

The thermodynamic database ThermoChimie (TC) was initially created by the French National Agency for radioactive waste management (Andra) in 1996, especially designed and qualified for systems of interest for the French repository concepts. TC is supported by an experimental program on clay minerals, cementitious phases, actinides and fission products, and it has been continuously updated to undertake geochemical calculations related to the PA of different waste packages, backfill/buffer materials and/or geological formations considered in nuclear waste disposal facilities.

In 2014 Andra and Radioactive Waste Management Ltd. (RWM) established the TC consortium to collaboratively work to develop the TC database. Development priorities (2015-2019) were set for TC focusing especially on:

- Ionic strength corrections considering specific ion theory (SIT) (saline waste);
- Temperature corrections considering both major and radionuclide applications;
- Organic complexes considering the inventory of initial waste compounds, natural systems, cement admixtures;
- A chemical model for cementitious materials including a large variety of compositions;
- Stability of clay minerals considering both buffer and natural systems and chemical pathways under near-field conditions;
- Radionuclides and chemotoxic element data sets considering a wide range of conditions (pH, ligands, temperature, ionic strength…).

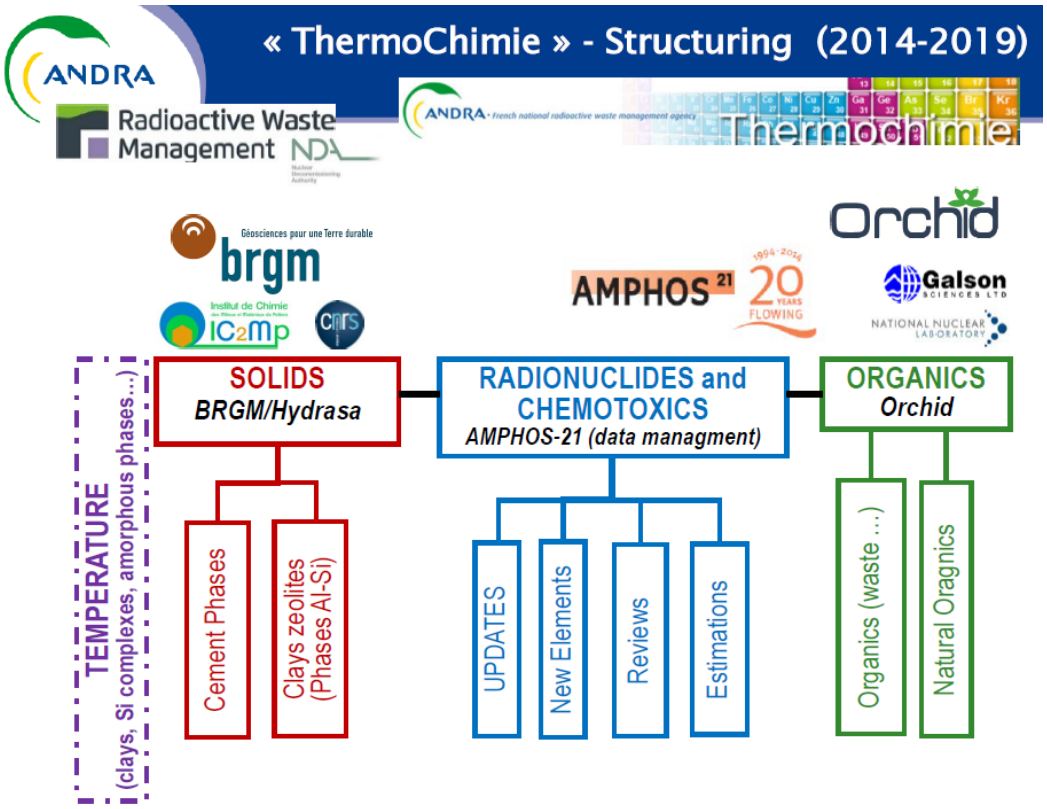

Figure - *Organization of "ThermoChimie" database in Andra-RWM consortium with Orchid consortium, Amphos-21 and BRGM partners.*

TC meets the requirements of completeness, accuracy and consistency for numerous radionuclides and chemotoxic elements and various major components of a geological repository: solid phases constitutive of the host-rock, bentonites, concretes, cement phases and corresponding secondary minerals with respect to their long term evolution. The accuracy and credibility of any thermodynamic modelling output depends heavily on the quality and completeness of the thermodynamic database (TDB) which is used. In the TC project, the selection of thermodynamic data is based on critical reviews of the literature, estimation models and an experimental program. Developments in the international NEA-TDB project are recognized in TC through cross referencing of data, development of the TC data selection procedure and the evaluation of the uncertainties, although TC doesn't rely solely on the NEA selected data.

With respect to the specific geochemical conditions within the near field of the GDF and the host-rock formation TC allows: (i) determination of radioelement and chemotoxic element aqueous speciation and solubility ; (ii) study of geochemical evolution of both the near- and the far-field of the disposal system, covering the stability of the constituents of clay minerals and secondary minerals of the aluminosilicate systems ; (iii) assessment of the process of cement degradation to account for the stability of cementitious phases including a broad composition

range with respect to concrete formulae ; (iv) assessment of the evolution of metallic/steel components present in the containers, liners, reinforced waste packages, with special emphasis to of iron/steel corrosion processes and secondary phases formed ; (v) assessment of the role of complexing agents derived from the degradation of Natural Organic Matter (NOM) present in the clayrock as well as the impact of the degradation products of organic compounds within wastes on the mobilization of radionuclides and (vi) tools and applications of the TC database under different disposal scenarios.

It is of the utmost importance to ensure consistency and quality of the database regarding its application in different numerical codes. A highly relevant additional topic is then the implementation of each new data set using a robust procedure for cross-checking, traceability and validation going from the data acquisition up to the release of new validated version of TC.

TC database is extracted into compatible formats with different geochemical codes used in the scientific community. The TC database is available and open to the public community (http://www.thermochimie-tdb.com/).

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