MOBILITY MISSION REPORT

This work has been partially supported by the EURAD project that has received funding from H2020-EURATOM 1.2 under grant agreement ID 847593.

MISSION TITLE

Participation in the EURAD training course on "Geochemical and Reactive Transport Modelling for Geological Disposal".

DESCRIPTION

Concerned organisations

University of Bern (Switzerland), SCK CEN (Belgium), PSI (Switzerland), Petten & Delft University of Technology (Netherlands), EMPA (Switzerland), Amphos 21 (Spain), Mines Paris (France), University of A Coruña (Spain).

Concerned infrastructures or facilities

- Geological disposal

Concerned phases

- Phase 5: Post-closure

Themes and topics

Theme 3: Engineered barrier system (EBS) properties, function and long-term performance

  - Spent Fuel and high-level waste disposal canisters

    - Containers for long-lived intermediate and low level wastes
    - Clay-based backfills, plugs and seals
    - Cementitious-based backfills, plugs and seals
- Theme 4: Geoscience to understand rock properties, radionuclide transport and long-term geological evolution
    - Long-term stability (uplift, erosion and tectonics)
    - Perturbations (gas, temperature and chemistry)
    - Aqueous pathways and radionuclide migration
- Theme 7: Performance assessment, safety case development, and safety analyses
    - Integration of safety-related information
    - Performance assessment and system models
    - Treatment of uncertainties

Keywords

Geochemical modelling; thermodynamic modelling; surface reactions; cation exchange; transport modelling.

EXECUTIVE SUMMARY

The topic of the training is Geochemical and Reactive Transport Modelling for Geological Disposal. The geological disposal systems for radioactive materials consist of different engineering and natural materials following a multi-barrier principle. As the geochemical evolution is involved with physical and chemical perturbations, the safety and performance analyses of the repository are important in the aspects of (1) the durability of different materials, and (2) the speciation and mobility of radionuclides. The objective of this training is to assess the evolution via numerical models in which geochemistry is linked to transport and reactive transport codes. Three main codes were presented and introduced during the training, and the geochemical code PHREEQC was chosen as my target one. The code PHREEQC is capable of calculating aqueous speciation, mineral equilibria, multi-site cation exchange, complex surface modelling, exchange with a gas phase, solid solutions and kinetic reactions. The flow and transport modelling was implemented by coupling PHREEQC to HYDRUS HPx software which is applicable for 1D and 2D unsaturated transient-variable water flow, multicomponent transport and heat transfer with different geochemical solvers. During the training, several hands-on exercises have been implemented including (1) modelling a cementitious system with SiO2 and CaO present, (2) modelling the speciation of uranium at different pH conditions in which COUNTER and REPEAT functions were practised, (3) modelling the dissolution and precipitation of minerals with kinetics, and (4) modelling of reactive transport in 1D with a given solution infiltering a material. Through these exercises, we are able to transform specific research questions related to geochemical properties or evolution into a conceptual model. Moreover, the principles of geochemical thermodynamic and kinetic modelling and reactive transport modelling have been understood, and their application in the field of radioactive waste disposal is foreseen. Last but not the least, interesting lectures were given during the training, the topics of which include geochemical and reactive transport modelling, properties and evolution of materials in the engineered barrier system of geological disposal, geochemical properties of host matrix, sorption of radionuclides, sensitivity and uncertainty studies and surrogate learning, demonstration of state-of-the-art codes for implementing geochemical and reactive transport models.

1. MISSION BACKGROUND

Klikněte nebo klepněte sem a zadejte text.

1.1. R&D background

Geological disposal systems for radioactive materials consist of different engineered and natural materials; this is in view of the multi-barrier principle. The interaction between the contrasting materials in the near field of a disposal system (both the engineered barrier system and the host rock) will induce geochemical changes in these materials. The geochemical evolution as a consequence of physical and chemical perturbations needs to be part of the safety and performance analyses of the repository as it will influence (i) the durability of the different materials, and (ii) the speciation and mobility of radionuclides. Given the time scales involved (ten thousand to hundred thousand years), assessing the evolution can only be done with numerical models in which geochemistry is linked to transport, thus with reactive transport codes.

A key aspect of these models is the geochemical model in which the geochemical state variables are calculated based on thermodynamic equilibrium and kinetic processes. These geochemical models account for aqueous speciation reactions, dissolution/precipitation based on saturation state, sorption based on mechanistic sorption models (exchange reactions, surface complexation) and possible kinetic processes (related e.g. to the dissolution of glass or clay minerals or the corrosion of steel canisters).

Reactive transport codes typically couple these chemical models to solute transport equations. State-of-the-art reactive transport codes may couple this also the water flow or heat transport. Geochemical solvers in state-of-the-art reactive transport codes are also capable of handling some (micro)biological reactions. Therefore, these codes are capable to simulate coupled thermal, hydraulic, chemical and biological (THCB) processes and possible feedback between the processes. They became a powerful tool for understanding and assessing these coupled processes and the consequences of containment.

Given the complexity of the system and the long-time scales, models typically have large computational times and many uncertainties associated with them. Recent developments in new couplings between different solvers, faster methods to solve equations including methods based on machine learning, and efficient algorithms for uncertainty analysis are crucial in the framework of the analysis of the long-term evolution, optimization and performance assessment of a radioactive waste repository.

In EURAD, the work packages ACED (Assessment of Chemical Evolution of ILW and HLW Disposal cells) and DONUT (Development and improvement of numerical methods and tools for modelling coupled processes) improve and implement codes and models for assessing the geochemical evolution in the near field of a repository. In view of that, this training continues from the state-of-the-art and introduces the new developments acquired in these work packages.

1.2. Mission objectives

This training aims to enlarge the knowledge and expertise of the participants concerning geochemical and coupled reactive transport modelling, in the framework of the disposal of radioactive waste, with a focus on geological disposal. The expected development of skills as a result of this training will be: (1) an understanding of the principles of the geochemical and reactive transport modelling; (2) the applications for processes and evolution of materials in geological disposal, especially for cementitious materials in my case; (3) the speciation and migration of radionuclides; (4) to identify advanced methods for sensitivity analysis, uncertainty analysis, and the integration of machine learning techniques; (5) the application of the PHREEQC code, and the knowledge of other similar codes, e.g., GEMS, ORCHESTRA, OPENGEOSYS, CHESS, etc; (6) case studies on simulating the phases variations in cementitious materials, the uranium sorption on clay materials, the precipitation/dissolution of minerals, and the reactive transport modelling.

1.3. Mission request

Upon completion of the training course, the participants should be able to:

- Understand the principles of geochemical thermodynamic and kinetic modelling and reactive transport modelling;
- Use these principles for application in the field of radioactive waste disposal;
- Transform specific research questions related to geochemical properties or evolution into a conceptual model;
- Implement simple conceptual models into numerical codes for geochemical and reactive transport modelling;
- Identify advanced methods for sensitivity analysis, uncertainty analysis and integration of machine learning techniques.

1.4. Mission composition

Host organisation

University of Bern, Bern, Switzerland.

Host facility

Institute of Geological Sciences.

Mission dates

6th, February, 2023 – 10th, February, 2023.

2. MAJOR PRACTICES, TECHNIQUES, METHODS, TOOLS OR SYSTEMS OPERATED OR STUDIED

Klikněte nebo klepněte sem a zadejte text.

2.1. Practice, technique, method, tool or system operated or studied during the mission

The first practice was to model cement systems with the code PHREEQC.

Description

This practice was to consider a primitive cement system including only Ca and Si.

Usage

The components, solution species, and phases in the database were defined.

Benefits

A basic understanding of the code PHREEQC and the included functions were obtained.

Limitations

No limitations were related to this practice.

Applicability

It is useful to model the hydration of cementitious materials.

2.2. Practice, technique, method, tool or system operated or studied during the mission

The second practice was to calculate the speciation of Uranium at different pH conditions.

Description

In this practice, the speciation of U under different pH conditions was calculated.

Usage

This practice was implemented by a sliding activity reaction path using specified functions.

Benefits

I learned how to use COUNTER together with Global Variables, and the REPEAT functions.

Limitations

No limitations were related to this practice.

Applicability

COUNTER and REPEAT functions can significantly simplify some simulation scenarios.

2.3. Practice, technique, method, tool or system operated or studied during the mission

This practice was to model the cation exchange isotherm.

Description

In this practice, the ion exchange in a solution was modelled.

Usage

The exchange capacity for montmorillonite and its equilibrium solution were defined.

Benefits

This is useful for simulating the ion exchange, e.g., cation exchange in clay rocks.

Limitations

No limitations were related to this practice.

Applicability

The ion exchange isotherm shows the preference of a solution for different ions.

2.4. Practice, technique, method, tool or system operated or studied during the mission

This practice was for 1-D reactive transport modelling.

Description

This practice aimed to simulate the transport of a solution into a material.

Usage

The pressure head and transport properties were defined in HYDRUS HPx software.

Benefits

I learned to simulate the transport of a solution with corresponding chemical reactions.

Limitations

No limitations were related to this practice.

Applicability

It is useful to simulate the chemical assessment of the repository with groundwater flow.

3. MISSION FINDINGS AND CONCLUSIONS

3.1. Lessons learned and conclusions

The principles of geochemical thermodynamic and kinetic modelling and reactive transport modelling have been well introduced and explained during the training. Through this training, I learned how to apply these principles in the field of radioactive waste disposal and transform specific research questions related to geochemical properties or evolution into a conceptual model. As a project member working on the EURAD project (Task 4.2 and 4.3 in ACED work package), this training is beneficial for me to complete my work on the project. The objective of my work within the project is to simulate the chemical assessment of the ILW (Intermediate Level Wastes) disposal cell with the surrounding field environment and the host rock, which perfectly matches the content of this training. Moreover, this training course is useful and meaningful for me personally and for my home organisation -- VTT. The training content matches perfectly with two topics that we are working closely on – nuclear power plant safety and nuclear waste management. The practices implemented during the training showed me how to build up the conceptual model for a specific question and simulate the corresponding chemical reactions involved. For example, one of my upcoming projects aims to simulate the deterioration of concrete structures in nuclear power plants exposed to a certain aggressive water environment, e.g., leaching, sulfate attack, and chloride ingress. The knowledge and modelling skills I learned during this training definitely help me to accomplish the project. In a summary, this training is useful and beneficial to me.

3.2. Relevant findings and conclusions for home organisation

3.3. Relevant findings and conclusions for host organisation

3.4. Relevant findings and conclusions for other organisations

4. POTENTIALS FOR IMPROVEMENT OR DEVELOPMENT

4.1. Generic potentials

4.2. Potentials for home organisation

4.3. Potentials for host organisation

APPENDICES

Mission journal

Monday 06/02:

The day started with a section on Welcome and Introduction, followed by Lecture 1 -- <Modeling geochemical systems -- Equilibrium, Thermodynamics, Reaction Progress (database)>. Then the introduction to the course with some hands-on examples was presented, followed by the introduction of three codes: GEM-Selektor, ORCHESTRA, and PHREEQC. Lecture 2 was given in the afternoon, with a topic on the <Thermodynamic modelling of cementitious systems and their evolution>. The day was finished with a second hands-on session, in which a practice of modelling a primitive cement system including only Ca and Si was implemented.

Tuesday 07/02:

Two lectures were given during the day, and the topics were the <Modelling of slow processes (cement hydration, steel corrosion, glass dissolution, mineral dissolution (TST))> and <Geochemistry of the host rock and natural barrier material (pore water chemistry, mineralogy, fracture-matrix)>. Moreover, two codes (iCp and Crunch) were introduced, and two hands-on sessions were implemented.

Wednesday 08/02:

Two lectures were given at the beginning of the morning and afternoon, with topics of <Speciation of radionuclides> and <Molecular aspects and thermodynamic modelling of sorption phenomena>. One more code Min3P was introduced as well. Hands-on sessions following the topics of lectures were performed: sorption of uranium at different pH conditions.

Thursday 09/02:

The day started with one lecture on the topic of <Introduction to reactive transport modelling> was given, followed by an introduction to the code Hytec. Two hands-on sessions were given during the day modelling the dissolution/precipitation of crystals considering kinetics. The day finished with a laboratory tour at the University of Bern, where the set-up devices to measure the composition of pore water in rocks were shown.

Friday 10/02:

The day started with an introduction session to the code PHREEQC-OpenFoam, followed by two lectures on topics of <Integration of processes at a larger scale – sensitivity (uncertainty) analyses> and <Model abstraction – Surrogate modelling – Machine learning>. In the last two hands-on sessions, we learned how to implement 1-D reactive transport modelling where a given solution was infiltered into a system.

MISSION BENEFICIARY

Yushan GU
Research Scientist, Materials Science, PhD
Structural Materials Group
VTT, Technical Research Center of Finland, Espoo, Finland.

PARTNER EXPERTS CONTRIBUTING TO THE MISSION

Host organisation experts

- Diederik Jacques, SCK CEN (Belgium);
- Hans Meeussen, Nuclear Research Group (NRG) Petten & Delft University of Technology (Netherlands) ;
- Dmitri Kulik, PSI (Switzerland);
- Eric C. Gaucher, Rock-Water-Interaction group. University of Bern (Switzerland).

Home organisation experts

- Miguel Ferreira, VTT (Finland).

Other organisations experts

- Francis Claret, BRGM (France);
- Barbara Lothenbach, EMPA (Switzerland);
- Emilie Coene, Amphos 21 (Spain);
- Sergey Churakov, PSI (Switzerland);
- Ulrich Mayer, University of British Columbia (Canada);
- Laurent De Windt, Mines Paris (France);
- Javier Samper, UDC (Spain);
- Nikoloas Prasianakis, PSI (Switzerland).

REPORT APPROVAL

Date	Beneficiary	Home mentor/supervisor	Host mentor/supervisor
01/ 03/	Yushan GU	Miguel Ferreira	Diederik Jacques
	谷雨珊 27/02/2023	28/2/2023	Diederik Jacques Digitally signed by Diederik Jacques Date: 2023.03.01 06:57:02 +01'00'

eurad