MOBILITY MISSION REPORT

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MISSION TITLE

Attendance training course Geochemical and Reactive Transport Modelling for Geological Disposals

DESCRIPTION

Concerned organisations

Research entities

Concerned infrastructures or facilities

University of Bern, Switzerland

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
- Salt backfills
- EBS system understanding

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, Aqueous Speciation Reactions, Reactive transport code

EXECUTIVE SUMMARY

One of the key research topics in the nuclear field field is the geological disposal of radioactive waste. The most probable approach is the multi-barrier principle, for which interaction between the disposed waste and the encapsulating material can cause changes to the surrounding natural materials. To ensure safety and minimise risk during waste storage, geochemical modelling helps to recreate experimental data and exploit the results to time spans, incapable of being researched (>4 years). The mission aims to provide the theory of geochemicals, including thermodynamics and kinetics, and reactive transport modelling and its implementation in specific software. During training, the models vary in difficulty, from basic up to advanced codes, which are specifically applied for radioactive waste disposal and related statistical analyses.

In this mission, the main focus is on the software GEM-Selektor (GEMS), an open-source Gibbs free energy (GEM) code and thermodynamic data base kit for geochemical modelling. Participating the training course enables Andrea Kozlowski to learn geochemical modelling to apply to her PhD research. Her research focuses on the development of sustainable substitutes for pulversized fly ash in cement and concrete and geochemical models should extend experiments on the chemical behaviour analysis of the substitutes. This is within the applicability of the software. So far, she has not applied the code yet to her research, as other experimental outcomes have to be performed before.

1. MISSION BACKGROUND

1.1. R&D background

Geological disposal systems for radioactive materials comprises 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) 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 of simulating 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 for the containment.

Given the complexity of the system and the long-time scales, models typically have large computational times and many uncertainties associated with it. 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) 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 will continue from the state-of-the-art and introduce the new developments acquired in these work packages.

1.2. Mission objectives

- 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

1.3. Mission request

The mobility grant application is proposed to participate in the “Geochemical and Reactive Transport Modelling for Geological Disposals” one-week training course in Bern, Switzerland.

The purpose of the training is to learn the software “GEMS” for Andrea Kozlowski’s PhD. In her PhD, she compares the use of various supplementary cementitious materials (SCM) replacing pulverised fly ash (PFA), mechanically, chemically, and physically, and identifies sustainable, cost-effective substitutes. The currently used grout formulation, used for the safe encapsulation of low-level waste, cannot be continued in a few years’ time resulting from political decisions shutting down all unabated coal-fired power plants by 2025. Investigating the various properties of SCMs include long-term studies, which can only be represented experimentally to a limited extent. Using GEMS enables long-term behavioural predictions. Hence, modelling extends the experimental data and is part of Andrea’s research.

1.4. Mission composition

Host organisation

eurad: WP13, WP ACED, WP DONUT, WP FUTURE

Host facility

University of Bern, Switzerland

Mission dates

06. – 10. February 2023

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

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

Software: GEMS

Description

GEMS has been developed at PSI for 23 years. To date, GEMS is the most frequently freely available open-source GEM code and TDB kit for geochemical modelling, with >10'000 downloads and ca. 1'000 active users, and about 1'400 publications where it has been used. Main parts of GEMS are

- GEM-Selektor code
- GEMS3K numerical library for use in coupled codes
- GEMSFITS coupled code for inverse modelling and parameterization.

The GEM-Selektor GUI code is used for the forward thermodynamic modelling (in Equilibria mode) and for manipulation with input thermodynamic and reaction formats (in Database mode). The forward problem is to find mole amounts n and activities a of chemical species in an equilibrium state defined by pressure p, temperature T and bulk elemental composition b. This is done in the GEMS3K code library by applying the IPM-3 GEM algorithm, now available also with C, C++11 and Python 3 interfaces. From GEM results, any species concentrations, activity coefficients in any phase, partition and fractionation coefficients of elements between phases, and saturation indices can be obtained. The applicability of GEM is limited only by the knowledge of standard-state molar properties of end members and interaction parameters, and interaction parameters for phase-solutions.

If some of the GEM outputs are known from the experiments or from observations, then some input thermodynamic properties or interaction parameters of models of mixing can be retrieved by solving inverse thermodynamic modelling problems. This is the purpose of the GEMSFITS code that has been successfully used recently in a number of funded projects.

Usage

Geochemical softwares, such as GEMS, are used as an addition to experimental studies to better understand the underlying processes and to support the interpretation of experimental results. Cementitious interactions are hard to understand and a lot of research is still ongoing. But all research so far is implemented in the GEMS database, making it possible to improve the understanding of the interactions and enable the development of alternative binders.

Benefits

The code has a high input and good database, which is regularly updated with new research findings. The GUI reduces the effort and expert knowledge to write scripts, but for easier usage, an online version (nanocem CemGEMS) is available, providing templates for different main

types of cement. Regarding the technical needs, the code is written executable on most Linux, Windows, and Mac OS operating systems. Even though GEMS is a code, the provided GUI enables the operation controlled only by mouse-clicking using toolbar and menu icons. For specific numbers, spreadsheet-like screen forms can be used. All results are automatically stored in the project database, making them available for further calculations or plotting.

Limitations

The major limitation of GEMS is its specific usage and hence the need for a good basic understanding of aquatic chemistry and chemical thermodynamics whilst operating the software. Also, whilst contesting codes are capable of easy modelling and calculations, GEMS skips these parts and requires knowledge and some modelling experience before. The GEMS model should only be used to recreate and interpret experimental geochemical data, as the software does not provide any result interpretation nor pre-selectable chemical modelling problem formulations. The modelling results rely solely on the input composition and the underlying thermodynamic data. Plotting phase diagrams is a standard in chemical thermodynamics which GEMS can also rudimentary perform, but due to the complexity of the system, it should not be used. When coming to an issue, help can be barely found online and a problem solution has to be performed alone or in consultation with the current provider.

Applicability

GEMS is a software which is, besides PHREEQC, one of the most used codes to model the behaviour of cement in deep geological disposals. Whilst PHREEQC is a more generic code, GEMS is specifically created for the use of environmental and geochemical problems, such as carbon storage, (radioactive) waste disposal, (ore)geochemistry, geothermics, and cement chemistry.

3. MISSION FINDINGS AND CONCLUSION

3.1. Lessons learned and conclusions

Participating the course taught me the fundamentals of GEMS. The code seems to be very applicable for my PhD and getting to know the people behind the code as well as other users definitely improved my confidence in using the software. Various studies showed the applicability of the code and its feasible sensitivity, providing me with enough insight to proceed with GEMS.

As this code needs fundamental understanding while working with it, the coding cannot be done parallel to experimental work rather than having my full concentration on the coding. Having long-term experiments ongoing and various experimental setups, it might be hard to solely focus on the coding. Further, the underlying complexity of the code and–even though stated as user-friendly–the not-intuitive GUI result in a very time-consuming prepation before modeling. As my PhD is in its last year, I will try the code but at a certain point, I need to switch to a more generic used software (PHREEQC), as there are no online international user community help forums of GEMS.

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

The course was very focused on its educational aim with no social gathering during or after the course provided by the organiser. A round of introductions is not enough to motivate participants to engage in discussions or networking, resulting in a distant atmosphere between the participants.

Speaking only for the practical parts of the software GEMS, the session structure could be improved, as the workshop leader assumed the participants already had experience with the code. Even after stating out all participants being beginners, the workshop continued in the same unstructured and unprepared way as before. Together with the other workshop participants, we figured out it would be helpful if the software owners would create a community forum in which the code users can help each other. Until this point, issues have to be solved by oneself or the provider has to be contacted.

4.2. Potentials for home organisation

4.3. Potentials for host organisation

APPENDICES

Mission journal

The workshop duration was five consecutive days and comprised lectures, practical, and code introducing sessions. Each day, lectures and practical sessions alternated and two brief sessions presented geochemical or thermodynamic software which are additional packages or individual software, their applications, and small coding examples.
The first day provided mainly fundamentals to thermodynamics and cement. After both lectures and first introductions to the softwares HPx/PHREEQC, GEMS, and ORCHESTRA (participants had to decide for one software to focus on during the course), the first Hands-on session took place. Here, the participants got to know the GUI of their selected code and the implementation of primitive cement systems.
The following days started with a lecture, followed by four Hands-on sessions and either ended with another lecture or a code presentation. The training sessions were built around two examples: Leaching of cementitious materials based on ordinary Portland cement and uranium sorption on clay materials, and the lectures provided the required theory to the focused example. As all codes started with the uranium sorption example, the lectures held during day two to four were:

- Geochemistry of the host rock and natural barrier material
- Reactive Transport–Pore to Continuum scale
- Speciation of radionuclides–Including thermodynamic databases
- Molecular aspect and thermodynamic modelling of sorption phenomena
- Modelling of kinetically controlled processes in radioactive waste disposal, from radiolytic corrosion to microbial activity.

In the practical sessions, the modelling included pore water modelling, U speciation with complexation with carbon, and sorption edge, isotherm modelling, and Kd calculation. All these simple reactive transport models were 1D steady-state examples.
On the last day, the second example was performed in the Hands-on sessions, accompanied by the lectures on integration of processes at larger scale–sensitivity (uncertainty) analyses and machine learning for accelerating reactive transport model simulations and analysis.
The brief insights to codes presented during the week included iCP, CRUNCH, MIN3P, Core2D, and PHREEQC-OPENFOAM. Each of them specifies on different specific field, for example connecting the softwares Comsol Multiphysics and PHREEQC to maximise synergies (iCP) or using finite element methods to calculate transient saturated or unsaturated solute transport (CORE).

MISSION BENEFICIARY

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Host organisation experts

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Home organisation experts

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- Katherine Dobson, Lecturer, University of Strathclyde, UK

Other organisations experts

REPORT APPROVAL

Date	Beneficiary	Home mentor/supervisor	Host mentor/supervisor

Date of last signee	Andrea Kozlowski	Joanna Renshaw	Katherine Dobson