SORPTION WORKSHOP REPORT

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Prepared by

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: None.

ANALYSES AND CODES: None.

1 INTRODUCTION

A one-day workshop on radionuclide sorption in repository performance assessment (PA) was held on August 16, 2011, at the U.S. Nuclear Regulatory Commission (NRC) in Rockville, Maryland, with a video link to the Center for Nuclear Waste Regulatory Analyses (CNWRA®) offices in San Antonio, Texas. The seminar was led and facilitated by Dr. Tim Hicks from Galson Sciences, Ltd (GSL), with support from Dr. Daniel Galson (GSL).

The main purpose of the workshop was to support the NRC Integrated Spent Nuclear Fuel Regulatory Activities (ISFR) program by reviewing and discussing the treatment of radionuclide sorption for a range of geologic disposal projects in different geologic media worldwide. The aim was to provide the NRC and CNWRA staffs a high-level summary of approaches to this issue and the treatment and justification of radionuclide sorption in PAs in other projects. The workshop was organized such that background information on the relevant concepts was presented first, followed by presentation and discussion of selected PAs (as a set of "case studies"), followed by discussion of several relevant international projects.

Attendees

NRC: J. Bradbury, R. Fedors, M. Fuhrmann, J. Gwo, B. Leslie, C. Markley, and J. Rubenstone

CNWRA: P. Bertetti, R. Janetzke, J. McMurry, R. Pabalan, O. Pensado, D. Pickett, and J. Prikryl

Galson Sciences, Ltd.: T. Hicks and D. Galson

The agenda for this workshop is included as an appendix.

Introductory Session of Seminar: Participants' Concerns

In the initial session, participants were surveyed to raise any issues beyond the stated objectives that were important to consider at the meeting. Participants stated that they wanted to improve their understanding of the following issues:

- The use of sorption coefficients (K_ds) in PA versus the uses of thermodynamic sorption modeling together with K_ds in PA, and whether these approaches or others were being used in other programs.

- The approach to modeling sorption in the near-field and the geosphere versus its consideration in the biosphere, and the general topic of model integration. In particular, it was noted that an approach to parameter uncertainty that was considered conservative for one model component might not be conservative when applied to another model component.

- The use of risk-informed performance-based regulation in considering the modeling of sorption in PA and, in this context, the value of a more realistic approach to PA versus the use of a simpler model.

- The importance of radionuclide sorption in dose assessment (i.e., the extent to which uncertainties in radionuclide retardation in particular subsystems could affect calculated dose).

- Features, events and processes relevant to radionuclide sorption and contaminant transport accounted for in PA, and those screened out in calculations of system performance.

2 CASE STUDIES: SWEDISH SPENT FUEL DISPOSAL CONCEPT (GSL)

An overview of the Swedish spent fuel disposal program was presented. In the Swedish concept, spent fuel, high-level waste (HLW), and intermediate-level waste (ILW) are disposed within fractured crystalline bedrock in copper canisters surrounded by a bentonite clay buffer. The Swedish program is at an advanced stage and an application to construct a repository has recently been submitted. Sorption in the Swedish PA models is represented using a K_d approach. The K_ds were derived from an extensive set of batch experimental data. Experiments utilized various water chemistries and bentonite and rock types to consider the range of conditions expected for the repository. Supporting analyses including thermodynamic modeling, diffusion experiments, and literature data reviews were used to increase confidence in the K_ds selected. Within the PA models, several conversion factors were used to adjust K_d values for varying conditions, such as pH, rock type, ion-exchange capacity, and mechanical damage to bedrock. These conversion factors were coupled with uncertainty factors to generate the bounds and ranges for K_ds. K_d values used for the biosphere models were derived similarly using site-specific materials and uncertainty factors. PA model calculations include a deterministic best-estimate and probabilistic analyses.

3 CASE STUDIES: SWISS GEOLOGIC DISPOSAL CONCEPT (GSL)

The Swiss radioactive waste disposal concept is emplacement of stainless steel canisters (HLW) or grouted concrete containers (ILW) into clay host rock. The HLW containers would be surrounded by a bentonite buffer, while the ILW containers would be surrounded by a cement buffer. The Swiss program anticipates that transport of radionuclides would be dominated by aqueous diffusion. Consideration of sorption in the program is focused on the bentonite and cement buffers. Sorption data were primarily developed using results of in-house batch and diffusion experimental data with support from literature data. Corrections to the sorption values are made to account for variations in pH, mineralogy, and aqueous speciation. The program identified the phyllosilicate mineral content of the host clay rock as the major factor influencing sorption. Conversion factors and measured data are combined to generate a sorption database for each site under consideration for the repository.

4 CASE STUDIES: WASTE ISOLATION PILOT PLANT (WIPP) (GSL)

At the WIPP site waste containers are emplaced in bedded salt. Sorption is considered in PA models only for the Culebra Dolomite and is based on a linear K_d approach. K_d vales were derived from batch sorption experiments and considered a range of chemical conditions. K_d distribution functions, which were originally specified as uniform distributions, were later changed to log-normal distributions to better reflect experimental data. K_d values are adjusted to account for potential colloid transport.

5 CASE STUDIES: UNITED KINGDOM GEOLOGIC DISPOSAL CONCEPT (GSL)

The United Kingdom radioactive waste disposal concept is generic in nature and several options for disposal of HLW and ILW are under consideration. Sorption is included in scoping PA models as a linear K_d model. Sorption distribution functions are currently derived based on expert elicitations using data from batch experimental results in programs in the United Kingdom and other countries (e.g., the Swedish program). The sorption distributions span rather large ranges to account for uncertainties.

6 CASE STUDIES: SAVANNAH RIVER SITE AND IDAHO NATIONAL LABORATORY (CNWRA)

D. Pickett described development of a database for site-specific sorption data at the Savannah River Site and the Idaho National Laboratory. The goal of the work was to create a review tool and recommend appropriate single value K_ds to consider in evaluating deterministic analyses. Sorption data for multiple radionuclides were evaluated and recommendations considered important sorptive characteristics such as mineralogy and water chemistry.

7 REVIEW OF INTERNATIONAL COLLABORATIVE PROJECTS RELATED TO SORPTION (GSL)

T. Hicks covered information from the Nuclear Energy Agency (NEA) and sorption and thermodynamic database projects, in addition to the following European Commission projects: FUNdamental processes of radionuclide MIGration (FUNMIG), Understanding and physical and numerical modeling of the key processes in the Near Field and their coupling for different host rocks and repository strategies (NF-PRO), and Performance Assessment Methodologies IN Application to guide the development of the safety case (PAMINA).

The NEA Sorption Project comprised three phases that have progressed from identification of the diversity and gaps associated with thermodynamic sorption models in Phase I, to a set of benchmarking modeling exercises with well defined data sets in Phase II, and establishment of guidelines for developing sorption parameters and using thermodynamic sorption models in Phase III. Final reports for Phase III activities, which concluded in 2011, are expected to be published soon. Based on reviews of draft materials, the main outcomes of Phase III are guidelines for application of thermodynamic sorption models that emphasize the importance of:

- Defining model purpose (scoping, prediction, interpolation)
- Using well-constrained geochemical and mineralogical data for the system
- Having thermodynamic data of sufficient quality and completeness
- Generating sorption data of sufficient quality to test model applicability, and
- Iterating between experiments and model development

Conclusions from Phase III suggest that thermodynamic sorption models work well for simple systems and that simple system models can be a starting point to models for complex systems and to understand fundamental processes. Unfortunately, large uncertainties remain in the ability to make predictions of sorption on larger temporal and spatial scales than can be simulated in the lab or *in situ*. These uncertainties include the need to make many empirical decisions to implement and parameterize sorption models due to the difficulties in adequately

characterizing the myriad geological surfaces and geochemical conditions along a transport pathway. The work to date suggests that clay and sediment dominated geologic systems may be easier to upscale than fractured crystalline rock systems.

Discussion of the European Commission programs focused on the last round of completed projects (or 6th Framework). The FUNMIG project considered three different rock types under investigation as host rocks for potential European repositories. The FUNMIG program has identified three main uncertainties regarding sorption and PA: (i) conceptual uncertainty in modeling diffusion in clays at different scales, (ii) impact of colloids on transport through crystalline rocks, and (iii) the applicability of thermodynamic sorption models for strongly sorbing radionuclides.

8 CLOSING DISCUSSION OF SEMINAR: POINTS FOR FUTURE CONSIDERATION

In the closing discussion, participants were asked to provide observations from the meeting considering (i) advantages and disadvantages of different modeling approaches, (ii) knowledge base, and (iii) gaps and ongoing research. The purpose of the discussion was to provide direction to subsequent NRC/CNWRA consideration of future work in this area. Participants made the following general observations on what they had heard during the seminar:

- The seminar provided a good overview on approaches and issues considered to be important in other programs and projects.
- In general, there is confidence in the use of K_d for PA for systems or system components that are geochemically well defined. However, there are specific issues for particular rock types and geochemical environments that could require additional investigation.
- One of the valuable aspects of international collaboration is the training provided to staff.
- Participants suggested there would be value in obtaining further specific information and pursuing work on the following topics:
  - — Incorporating spatial and temporal variability in K_d into PA.
  - — The value of field-scale versus laboratory-scale tests (considering upscaling to PA), and the circumstances under which batch or column experiments provide a better representation of sorption at the scale of a PA.
  - — The influence of organic complexants and colloids on sorption.
  - — The role of thermodynamic sorption modeling in extrapolating understanding of K_d in spatially and temporally variable systems.
  - — The importance of changes in chemical conditions and mineralogy in particular system components over long timescales.
  - — Synthesis of results from recently completed large European Commission funded R&D projects (such as FUNMIG).

— An integrated approach to considering spatial heterogeneity in radionuclide retardation at the large scale, e.g., development of an approach to correlate flow field and geochemical variability with variability of radionuclide transport parameters.

APPENDIX

Agenda
NRC Seminar on Sorption in Repository PA
16 August 2011 (Rockville, MD)

The purpose of this seminar to discuss the treatment of radionuclide sorption for a range of geologic disposal projects in different geologic media worldwide. The aim is to provide NRC and CNWRA staff a high-level summary of approaches to this issue and the treatment and justification of radionuclide sorption in performance assessments (PAs) in other projects.

0900–0930 Introduction and purpose of meeting (All)

0930–1030 Overview (Galson Sciences)
- Sorption processes
- Sorption sites for different engineered and geologic media
- Sorption modeling / treatment in PA

1030–1045 Break

1045–1130 Case studies (Galson Sciences and CNWRA)
- Sweden – assessment of spent fuel disposal in crystalline rock
- Switzerland – assessment of spent fuel, HLW and ILW disposal in clay
- US – assessment of transuranic waste disposal in salt (WIPP)
- UK – generic assessment of spent fuel, HLW and ILW disposal for illustrative disposal concepts
- US – assessment of tank closure activities at Savannah River/Idaho National Laboratory (CNWRA)

1230–1330 Lunch

1330–1415 Case studies – continued (Galson Sciences and CNWRA)

1415–1500 International collaborative projects (OECD/NEA and EC) (Galson Sciences)
- NEA Sorption project
- NEA Thermochemical Database (TDB) project
- EC FUNMIG project and other EC projects

1500–1515 Break

1515–1615 General discussion – summing up (All)
- Advantages/disadvantages of different modeling approaches
- Knowledge base
- Gaps and ongoing research

1615–1715 Programmatic discussions and path forward (NRC/CNWRA only)