CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT Attendance at the Workshop on Sorption: Conclusion of the Nuclear Energy Agency (NEA) Sorption Project Phase II on Use of Thermodynamic Sorption Models (TSM) in Performance Assessment (AI: 20.06002.01.232.501)

DATE/PLACE: October 10–11, 2005, Paris, France

AUTHORS: David Turner [Center for Nuclear Waste Regulatory Analyses (CNWRA)], Budhi Sagar (CNWRA)
William Ott [U.S. Nuclear Regulatory Commission/Office of Nuclear Regulatory Research (NRC/RES)]

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TRIP REPORT

Subject

Attendance at the Workshop on Sorption: Conclusion of the Nuclear Energy Agency (NEA) Sorption Project Phase II on Use of Thermodynamic Sorption Models (TSM) in Performance Assessment

Dates of Travel and Countries/Organizations Visited

October 10–11, 2005, Paris, France/NEA Offices

Authors, Title, and Agency Affiliation

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Sensitivity

Not applicable

Background/Purpose

Starting in 1991 in Interlaken, Switzerland, the Nuclear Energy Agency (NEA) has supported a series of cooperative international initiatives to support the development and application of TSM in performance assessment. Phase I of the NEA Sorption Project was conducted in 1997 and 1998 to develop an assessment of the current state-of-the-art for TSMs, and identify the type of parameterization that would be necessary to apply these models in performance assessment. Based on the results reported in Phase I, Phase II was established in September 2000, with teams sponsored by 18 funding organizations from 13 countries with an objective of benchmarking TSMs against seven separate test cases. Phase II was completed in 2003, and the final report was published in the summer of 2005. The October 10–11, 2005, workshop was held to summarize the major results of the Phase II modeling exercises, describe strengths and weaknesses of the different modeling approaches, and identify future modeling needs. The workshop agenda is included as Attachment A. A total of approximately 45 scientists and engineers from eight European Union countries, Switzerland, Australia, Japan, and the United States attended the meeting. A participant list is included as Attachment B.

NRC and CNWRA have participated in all phases of the NEA Sorption Project since 1991.

Abstract: Summary of Pertinent Points/Issues

There were three main objectives for this workshop: (i) summarize the results from the Phase II modeling studies, identifying strengths and weaknesses of different modeling approaches; (ii) describe how sorption modeling is currently handled in performance assessment and identify methods to incorporate information from TSMs in radionuclide migration abstractions; and (iii) identify potential future activities for the NEA Sorption Project.

As described in the following sections, all three of these objectives were covered during the workshop. Although the design and site characteristics of many of the European repository programs are different from the current U.S. design (e.g., bentonite backfill versus no backfill and reducing conditions versus oxidizing conditions), the continued opportunity to exchange information and interact with the international radioactive waste management community provides valuable feedback on NRC approaches to sorption modeling and also information on the state-of-the-art in other national radioactive waste management programs. In addition to supporting the development of the Total-system Performance Assessment (TPA) code for the NRC High-Level Waste Repository Safety program, participation in this international program may also support a number of NRC programs where radionuclide migration is an area of concern. NRC has supported the NEA Sorption Project since its inception in 1991, and will consider continuing support dependent on continued and timely relevance to NRC programs.

Discussion

The following are key points of interest from the NEA Sorption Workshop.

Day 1 (October 10, 2005), Session I

The first day of the workshop began with opening remarks by Mehdi Askarieh (Nirex, United Kingdom) who provided a brief history of the NEA Sorption Project, beginning with the meeting in Interlaken in 1991. He outlined the objectives of Phases I and II, and briefly discussed the anticipated outcomes from the workshop.

Session 1 was chaired by James Davis (U.S. Geological Survey) and provided a summary of the test cases and results from the Phase II modeling study. Dr. Davis described the two main modeling approaches employed during Phase II: (i) the "bottom-up" component additivity (CA) approach that is a more complete representation of the mineral-radionuclide-water system, but requires much more parameterization, and (ii) the "top-down" generalized composite (GC) model that is mathematically simpler and can be used to model a narrower range of relevant field conditions. He noted some of the difficulties encountered by some of the modeling teams, such as inconsistent use of model parameters. He also noted several key advances in the application of TSMs, including the development of the internally consistent NEA thermodynamic database.

Tim Payne (ANSTO, Australia) provided a summary of findings from the first three test cases for sorption on single, simple end-member oxide minerals (Fe-oxide, quartz, and alumina). For these systems, the review teams selected surface complexation TSMs, both with and without electrostatic double layers. Overall, the findings demonstrated that the different TSMs were capable of modeling the observed sorption behavior as a function of chemistry, provided they were used in a consistent manner. In general, the identification of key chemical parameters and

setting up the models to address these parameters are more important for modeling results than the choice of a specific model. Most modeling groups chose chemically plausible surface species, but there is only limited independent information [e.g., Extended X-Ray Absorption Fine Structure (EXAFS) spectroscopy] to confirm the existence of a given surface species. For this reason, the sets of surface reactions selected by individual teams were quite different in some cases. Dr. Payne also covered two test cases for sorption on complex materials, noting that although none of the teams for these test cases used either a pure GC or CA approach, the models that were based on the GC approach were generally more successful at reproducing observed sorption behavior than those teams that used a CA modeling approach.

Michael Ochs (BMG Engineering Ltd., Switzerland) described the results of two test cases that addressed sorption on more complicated aluminosilicate clays (Ni and Np sorption on smectite). He pointed out that the most successful modeling approaches were those that made use of the information provided in the test cases, including isotherm data and sorption data as a function of pH. He stressed that not considering the complete range in experimental data may lead to models that leave out key sorption processes. For example, he noted that in some cases, ternary surface complexes involving carbonate improve the model results at higher pH, but there is a general lack of experimental data over broad enough ranges in chemical conditions to fully evaluate these effects.

The results of the Phase II modeling study have been compiled in a report entitled, NEA Sorption Project Phase II: Interpretation and Prediction of Radionuclide Sorption onto Substrates Relevant for Radioactive Waste Disposal Using Thermodynamic Sorption Models. This report was published by NEA during the summer of 2005 and may be purchased through the NEA website <www.nea.fr>.

Day 1 (October 10, 2005), Session II

Session II, Part I. During the first part of Session II, presentations focused on developing TSM approaches for complex materials. David Turner (CNWRA) presented a talk focusing on planning and conducting sorption experiments to identify key geochemical parameters (pH, P_{CO2}, and mineral surface area) that influence actinide sorption behavior for single end-member mineral systems, and developing TSMs that account for these effects. Michael Ochs presented modeling considerations for complex bentonite materials. Compacted bentonites are proposed as backfill in many repository programs. These materials have special challenges because of uncertainties in porewater chemistry at high densities. James Davis (U.S. Geological Survey) presented sorption modeling results from the remediated uranium mill tailings site in Naturita, Colorado. This research has been funded by the NRC RES and has provided insights on applying the nonelectrostatic GC approach to simulate uranium sorption and transport at a field scale of several kilometers. Uranium sorption experiments with natural materials have focused on relatively narrow ranges in pH and P_{CO2} that are relevant to the geochemical conditions at the site. A simplified GC approach with two surface reactions and three site types resulted in good simulations of the experimental data over a range in uranium concentrations.

Session II, Part II. During the second part of Session II, presentations focused on determining the parameters necessary for the application of TSMs. These included presentations on a Monte Carlo approach used to determine surface protolysis constants (Tim Heath, SERCO Assurance, United Kingdom) that can be used to identify both sensitive and insensitive parameters for describing the mineral-water interface. Johannes Lützenkirchen (Karlsruhe,

Germany) provided an overview of potentiometric titration data, with a particular emphasis on potential experimental artifacts that need to be considered in evaluating data quality for model parameterization. Tim Payne and Vinzenz Brendler (Rossendorf Research Center, Germany) presented an overview of model reliability and defensibilty of modeling results. They focused on evaluating the correctness of the model with regard to physical and chemical terms, and the need to evaluate the accuracy of model predictions through blind tests. They also discussed the electrostatic surface complexation model parameter database developed at Rossendorf. The RES^3-T database provides a compilation of model-specific parameters for a large number of chemical-mineral systems. It contains information on different surface complexation models for the sorption of both radionuclides and non-radionuclide elements, and provides a good initial source of data. Dr. Brendler noted that the database is a compilation only; the next step will be to evaluate the adequacy of the parameters in the database for broader applications.

Day 2 (October 11, 2005), Session III

Session III, Part I. The second day of the workshop began with an overview by Scott Altmann (ANDRA, France) focused on how sorption information is used in performance assessment, with specific examples from different national programs. He indicated that, depending on the site and the chemical system under consideration, both the GC and CA approaches should be maintained as possible tools, because the strengths of each approach are applicable to different aspects of sorption modeling. He also noted that, based on the presentations from the previous day, the participants in the meeting are close to being able to write procedural guidelines for how to employ TSMs in performance assessment.

The first presentation of Session III was from Eric Giffault (ANDRA, France) on the selection of sorption coefficients (K_ds) for bentonite systems. Typical uses of TSMs to support performance assessment include developing an understanding of phenomena that control radionuclide sorption under site-specific conditions, estimates of the effects of trace minerals (e.g., calcite, Fe-oxide) on sorption behavior, confidence building for performance assessment parameter values, evaluating parameter sensitivity to chemical and physical conditions, and an investigation of the effects of temperature on sorption. He also summarized the current TSM characteristics that limit their application in performance assessment, including data sparseness, use of different modeling conventions, and the gap with regard to multicomponent systems.

Patrik Sellin (SKB, Sweden) gave an overview of the Swedish SR-Can safety assessment for building and constructing an encapsulation plant. His presentation focused on modeling radionuclide migration through a compacted bentonite buffer. Basic sorption coefficient information is taken from batch experiments with MX-80 montmorillonite (smectite), with diffusivity measurements from tritiated water. The discussion of effective diffusivities focused on monovalent (Cs^+) and divalent (Ra^{2+}, Sr^{2+}) species, where the dominant sorption process is modeled as ion exchange with the clay fraction in the bentonite. The uncertainties are handled using probability density functions (pdfs) based on expert judgement. A sensitivity analysis showed that doses are generally sensitive to sorption (expressed as K_d), especially for plutonium. One interesting outcome is that the assumption of no retardation for Ra-226 is not conservative. A conceptual model where Ra-226 is not sorbed flushes the contaminant through the system; where sorption is assumed (i.e., K_d>0), Ra-226 is retained in the system and the calculated dose increases because of radioactive decay into Rn-222.

Anna Diercx(ONDRAF/NIRAS, Belgium) presented the current status and future plans for performance assessment in the case of the Boom Clay proposed as a repository host formation. She also focused on several potential sources of uncertainty in sorption models, including experimental uncertainty, conceptual model uncertainty, and errors attributable to data transfer (scaling). She noted a limited application of TSM approaches to support K_d selections for a subset of key radionuclides. She identified the need for high quality data to support TSM model development, demonstrations of scaling to the repository scale, and a critically reviewed TSM database similar to the NEA thermodynamic database project.

Bernhard Schwyn (NAGRA, Switzerland) presented an overview of the sorption databases used for the Swiss safety assessment for the Opalinus Clay site. In general, for compacted bentonite buffers, sorption information is scaled from TSM models to reference conditions through a series of conversion factors. These conversion factors include cation-exchange capacity, pH, and aqueous speciation. This approach is only applied to a subset of radionuclides of interest, with the other K_d distributions based on expert judgement and chemical analogs.

David Turner (CNWRA) presented a summary of the evolution of the use of TSMs to support TPA code development. These include parameter distribution support through the simulation of sorption in regional groundwaters. A subsequent refinement through the use of K_d response surfaces is the current representation of actinide sorption in the TPA code. With this access port into the TPA code, sorption TSMs can now be further refined off line and the information fed back into the performance assessment calculations.

Les Knight (Nirex, United Kingdom) discussed the use of expert elicitation to develop sorption and solubility parameter distributions. Almost 300 elicitations were conducted using a formal process, but many of them were conducted more than 10 years ago. The information used by the experts to develop the distributions is not well preserved, and Nirex is beginning the process of updating prior elicitations.

James Davis (U.S. Geological Survey) presented the development of reactive transport models for the Naturita uranium mill tailings site. One of the main results of this work is that it highlights the importance of capturing the spatial and temporal variability of site geochemistry and the subsequent effects on sorption behavior.

Hans Wanner (HSK), Bo Stromberg (SKI), and Budhi Sagar (CNWRA) each spoke briefly on the role of TSMs in regulatory reviews. They emphasized that any analysis that helped build confidence in models was helpful to regulatory reviews. The TSMs discussed in this workshop, when properly used, together with other information, could enhance the confidence in the K_d values used in performance assessment.

Day 2, Closeout Discussion

The workshop ended with an extended discussion of the usefulness of the results from the Phase II modeling studies, and proposals for the next step in the NEA Sorption Project.

One general comment was that the participants in the Phase II modeling exercise should have been more constrained in setting up their models. There were 18 different modeling teams, and they chose a number of widely different approaches to model development. In addition, the

modeling teams generally did not make extensive use of spectroscopic information, and tended to use a hybrid rather than a pure GC or CA approach to modeling complex materials.

In general, however, participants in the meeting were encouraged by the progress made in the development of thermodynamic sorption models since the inception of the NEA Sorption Project in 1991, and particularly since the Phase I organizational meeting in 1997. There was a belief that while sorption models are not yet at the same level as aqueous thermodynamics, the participants in the sorption project have identified several key geochemical parameters that exert the most control on radionuclide sorption, and are focusing modeling efforts on what is most important to simulating sorption behavior. In addition, this workshop was distinctive in the involvement of performance assessment experts to explore potential ways to transfer the results from detailed process modeling to performance assessment.

During the discussion, some participants stressed the need to emphasize transparent and traceable sorption modeling approaches that are accessible to stakeholders and increase public confidence in waste management decisions. A key part of this effort should focus on establishing a solid scientific basis for all modeling decisions that is understandable and generally accepted by the scientific community. Particular emphasis was placed on using sorption models that "make chemical sense." One means of enhancing the realism of TSMs is to use spectroscopic information to constrain the selection of surface complexes in the models. Improvements in analytical techniques are bringing this information to the point where it can be applied in more relevant solution chemistries, especially with regard to more dilute radionuclide solutions.

Discussions of potential Phase III follow-up activities included (i) preparing a guidance document that would provide instructions for model setup, (ii) holding a workshop on TSM development and applications, (iii) reviewing available spectroscopic information to constrain surface reactions, and (iv) developing a sorption parameter database through a focused effort similar to the NEA Thermochemical Database Project for aqueous speciation.

Pending Actions/Planned Next Steps for NRC

The participants in the meeting expressed general agreement for a Phase III to follow-up the 2005 NEA Sorption Workshop. Although the exact nature of the tasks to be proposed by the workshop organizing committee has not been determined, the NRC will participate in discussions of any proposed activities and consider continuing its participation in this international working group.

Points for Commission Consideration or Items of Interest

Thermodynamic sorption models are more mature now and have been used in a variety of ways to support radionuclide migration abstractions in performance assessment. These advances in modeling environmental systems are also beginning to be applied to decommissioning problems and other analytical situations where subsurface contamination is found. NRC has been a strong supporter of the development and use of these models and should consider continuing this support in the future. No Commission action is required.

SIGNATURES:

David Turner, Assistant Director CNWRA — Date: 10/25/2005

Budhi Sagar, President CNWRA — Date: 10/25/2005

William R. Ott, Section Chief, NRC/RES — Date: 10/26/2005

CONCURRENCE:

English Pearcy, Manager
Geochemistry — Date: 10/25/2005

Wesley Patrick, Vice President
Geosciences and Engineering Division — Date: 10/25/2005

ATTACHMENT A

Organisation de Coopération et de Développement Economiques
Organisation for Economic Co-operation and Development

English - Or. English

NUCLEAR ENERGY AGENCY
RADIOACTIVE WASTE MANAGEMENT COMMITTEE

NEA/RWM/SORPTION(2005)2
Unclassified

Sorption Project

Workshop on Sorption: Conclusion of NEA Sorption Project Phase II and Status Analysis of Sorption Modelling for PA

Final Programme

10-11 October 2005
NEA HQ
Issy-les-Moulineaux, France

English - Or. English

WORKSHOP ON SORPTION:

CONCLUSION OF NEA SORPTION PROJECT PHASE II AND STATUS ANALYSIS OF SORPTION MODELLING FOR PA

NEA BUILDING, 7TH FLOOR,
ISSY-LES-MOULINEAUX (PARIS, FRANCE)

10 - 11 OCTOBER 2005

A workshop organised by the
OECD/Nuclear Energy Agency

Final Programme

1. WORKSHOP AIMS

The goals of the workshop are to:

- provide an overview and illustration of the main project results, with emphasis on merits and limitations of thermodynamic sorption models (TSMs) and recommendations on their use;
- share ideas and stimulate discussion on the best use and practical implementation of "top-down" and "bottom-up" TSM approaches for PA-relevant materials of different complexity;
- achieve a clear picture of the importance of uncertainty in K_d for various performance assessments, of the potential of TSMs for strengthening the respective safety cases, and of the corresponding present and plausible future needs for TSM-based quantification of radionuclide sorption.

2. WORKSHOP STRUCTURE

The workshop is organised into the following main sessions:

- ***Session I:*** Key results of the NEA Sorption Project, Phase II;
- ***Session II:*** Thermodynamic sorption model (TSM) approaches for complex materials;
- ***Session III:*** Implementation of TSMs in PA programmes, present status, future plans, challenges & needs.

Annex 1 presents the final agenda of the workshop.

3. WORKSHOP CHAIRPERSONS

During the sessions, the work of the chairperson will be to introduce speakers, keep the session on schedule, keep in mind the objectives of the workshop, and motivate participants for discussion.

4. PRACTICAL INFORMATION/ORGANISATION

The workshop is an NEA workshop and organised in the framework of the OECD Nuclear Energy Agency, Sorption Project.

The OECD/NEA is kindly hosting the workshop at Issy-les-Moulineaux, France on 10 and 11 October 2005.

5. PARTICIPATION

Participation in the workshop is limited to approximately 50 persons.

6. WORKING LANGUAGE

English will be the working language of the workshop.

7. ORAL PRESENTATIONS

Presenters are requested to provide an electronic version of their oral presentations in advance of the workshop to the NEA Secretariat (Katia-Karina Le Bot, Katia-Karina.lebot@oecd.org with copy to Sylvie Voinis, sylvie.voinis@oecd.org).

8. REPORTING

The workshop will be reported in the form of a short report consisting of a short summary of main findings (about 5 pages), the agenda of the workshop and the list of participants.

9. LOCAL ARRANGEMENTS

The workshop will take place on 7th floor at NEA HQ in Issy-les-Moulineaux, France.
Please look at: http://www.nea.fr/html/general/nea-access.html

Accommodation
Please look at [http://www.nea.fr/html/general/hotels.html] to get a list of hotels near the NEA.

Annex 1

FINAL AGENDA

Day 1 10 October 2005		
09:00 9:30	**Introduction** Welcome, purpose and topics of workshop Overview of Sorption Project Phase I & II	*M.Askarieh, S.Voinis*
09:30-10:30	**Session I: Key results from Phase II** • Complexity of PA-relevant substrates and relevance for sorption models • Key results and assessment of sorption models • Key issues in the consistent application of thermodynamic sorption models (TSMs) • Management of uncertainty in K_d • Recommendations for TSM approaches	Chair: *M. Askarieh* *J. Davis, M. Ochs, T. Payne*
10:30-11:00	**Coffee break**	
11:00-12:30	**Session I, cont.**	Chair: *M. Askarieh*
12:30-14:00	**Lunch**	
14:00-15:00	**Session II: Thermodynamic sorption model (TSM) approaches for complex materials** **Part I:** Introduction and examples • Introduction to the session • Examples: o Single minerals in clay/tuff systems o Bentonite o Sediments, weathered rock/fracture filling	Chair: *S. Altmann* *S. Altmann* *D. Turner* *M. Ochs* *J. Davis*
15:30-16:00	**Coffee break**	
16:00-18:00	**Session II, cont.** **Part II:** Application of thermodynamic sorption models: strengths and limitations of "top-down" and "bottom-up" approaches, practical issues • Introduction: Different models for different tasks • Model parameters (fitted, other sources, chemical plausibility), data needs/bases, TDB • Practical usefulness and scientific defensibility • Application to intact systems, link with transport	Chair: *S. Altmann* *M. Ochs* *J. Davis, T. Heath, J.Lützenkirchen* *T. Payne, V. Brendler* *J. Davis, M. Ochs*
18:00	**Closing of sessions I and II**	

Day 2 11 October 2005		
	Session III: Implementation of TSMs in PA programmes, present status, future plans & challenges, needs	Chair: *M. Randall*
09:00-10:30	**Summary of Day 1, outlook on Day 2**	*S. Altmann*
	Presentations and detailed discussions:	
	Selection of K_d for PA as a function of (evolving) conditions: Examples and viewpoints of PA and regulatory agencies	
	• Bentonite EBS	*E. Giffaut, P.Sellin*
	• Clayrock (Boom/Opalinus Clay)	*A. Dierckx/L. Wang B. Schwyn*
10:30-11:00	**Coffee break**	
11:00-12:00	**Presentations and detailed discussions: cont.**	
	• Near-surface formations/US sites	*D. Turner, J. Davis*
12:00-13:30	**Lunch**	
13:30-15:00	**Presentations and detailed discussions: cont.**	
	• Crystalline rock/fractures	*L. Knight*
	• Viewpoint of regulators	*H. Wanner, B. Sagar, B. Stromberg*
15:00-15:30	**Coffee break**	
15:30-17:00	**Session III: Discussion**	Chair: *P. Herman*
	• Critical/less critical issues for using TSMs in PA • Foreseeable requirements for successful future TSM applications in PA • Discussion of priorities for a possible Phase III	*Panel:* M. *Askarieh, S. Altmann, J. Bruno, J. Davis, A. Dierckx, , P. Larue, M. Hakanen, M. Ochs, S.Voinis*
17:00-17:15	**Closing of the session III and workshop**	*M. Askarieh*

ATTACHMENT B

OECD Nuclear Energy Agency

Friday, 30 September 2005

SORPTION Project Phase II - Workshop 10 11 October 05
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RADIOACTIVE WASTE MANAGEMENT COMMITTEE

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Sorption Project

Workshop on Sorption: Conclusion of NEA Sorption Project Phase II and Status Analysis of Sorption Modelling for PA

Final Programme

10-11 October 2005
NEA HQ
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English - Or. English

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