5/29/01

CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Geological Evidence and Theoretical Bases for Radionuclide-Retention Processes in Heterogeneous Media, organised in the framework of GEOTRAP, the Organisation for Economic Co-operation and Development (OECD) Nuclear Energy Agency (NEA) Project on the Transport of Radionuclides in Geologic, Heterogeneous Media and hosted by the Swedish Nuclear Fuel and Waste Management Company, Sweden and a visit to the Äspö underground laboratory (20.01402.561/761)

Chaired a Session—Geologic and Field Evidence for Retention Processes and Their Representation in Models

Presentation by Dr. Budhi Sagar titled The Consideration and Representation of Retention Processes in Performance Assessment—Regulatory Perspective

DATE/PLACE: Workshop held in Oskarshamn, Sweden—May 6 through 9, 2001
Äspö underground laboratory visit—May 10, 2001

AUTHOR(S): Budhi Sagar

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AUTHOR(S): Budhi Sagar

ATTACHMENTS: Copies of the meeting agenda and attendee list are attached

BACKGROUND, PURPOSE, SUMMARY OF MEETING

Hosted by the Swedish Nuclear Fuel and Waste Management Company (SKB), the fifth Nuclear Energy Agency (NEA) GEOTRAP workshop on Geological Evidence and Theoretical Bases for Radionuclide-Retention Processes in Heterogeneous Media was held at Oskarshamn from 7-9 May, 2001. A tour of the underground laboratory at Äspö was organized on May 10.

The purpose of the workshop was to review the theoretical bases and supporting evidence for the characterization and modeling of retention processes, with emphasis on geological and field evidence, and the treatment of retention processes in performance assessment. In particular, the aims were to identify (i) issues that have been sufficiently resolved for current and future performance assessment purposes, and (ii) future trends that can be foreseen and, in particular, unresolved issues that should be (or are going to be) addressed.

Two categories of processes were considered. The first category of processes were those that retard the migration of radionuclides but are reversible. These processes included sorption and matrix diffusion. The second category of processes were those irreversible processes that potentially immobilize the radionuclides; these included colloid formation and isotope fractionation.

The workshop participants came to the conclusion that significantly more understanding has been obtained for the reversible processes of sorption and matrix diffusion than for the irreversible processes. Several participants suggested that at low concentrations expected to be prevalent in the far field of repository systems, the reversible retention processes will be dominant. The academicians among the participants

generally wanted to extend the understanding of all processes to greater detail and at smaller scales (e.g., atomic scale) while the practitioners generally believed that given the great emphasis on long-term containment in waste packages in almost all national programs, the current understanding of the retention processes was sufficient for current safety evaluations.

MEETING DETAILS

The workshop was organized into four sessions. Invited presentations comprised the three oral sessions and all uninvited presentations were formed into a poster session. Ahead of the workshop, the organizers had provided a series of questions to the invited speakers, which helped the speakers to focus their talks. See the attached agenda, which includes the questions for each session. Abstracts for all invited papers were made available to workshop attendees, copies of which can be obtained from the author of this report. NEA plans to publish workshop proceedings in a few months.

Session 1 dealt with Fundamental Understanding of Individual Retention Processes. The first paper on "How Geologists View Retention" was presented by Mike Heath of University of Exeter. After reviewing the nature of retention processes, he presented examples of geologic evidence of retention processes operating in nature at a scale of 10^5–10^6 year scale. His primary concern was that many processes operate simultaneously and it was not always possible to interpret the data to determine the effect of individual processes. Continuing with this theme, Jim Davis (U.S. Geological Survey) suggested that the distinction between reversible and irreversible processes was only a matter of time scale, irreversible processes being reversible at very long time scales. His assessment was that current understanding for sorption was very good at the molecular level but that at higher (macro) scales, many uncertainties are introduced because of the heterogeneity of the natural media. He suggested that uncertainties can be reduced in the future with the use of semi-empirical site-binding models, which will require site specific characterization of the natural mineral assemblages. Roy Haggerty of Oregon State University presented his research on matrix diffusion. His hypothesis was that non-Fickian matrix diffusion in heterogeneous media gives rise to a heavy-tailed (power-law) residence time distribution for tracers and that such behavior may lead to non-conservative estimates of radionuclide retention. He recommended that future efforts be directed towards both the experimental and modeling aspects of heavy-tailed matrix residence time distributions. Various aspects of colloid mediated transport were addressed by Bruce Honeyman of Colorado School of Mines. He indicated that colloid-facilitated contaminant transport can take place only under a relatively narrow set of conditions such as when colloid concentration is high. According to him, the role of scale and heterogeneity on colloid transport have not yet been studied. Susan Stipp of the University of Copenhagen spoke about immobilization as a retention process. Her emphasis was on using ultra-high-resolution techniques (e.g., scanning probe microscopy, X-ray photoelectron spectroscopy, and transmission electron microscopy) to understand the geochemical processes at the atomic scale. She emphasized that ions can be adsorbed to a solid even while it is dissolving in a solution that is highly undersaturated. She recommended further study of precipitation, co-precipitation and solid state diffusion as processes for immobilization. The final paper is the first session was given by Bill Murphy of California State University at Chico. This paper dealt with the subject of isotopic fractionation with examples from the Peña Blanca natural analog studies where the U-234/U-238 activity ratio was found to be as high as five, which was ascribed to the effect of alpha recoil. Murphy stated that isotopic fractionation effects are generally neglected in performance assessments, which in his opinion was acceptable because of their relatively small magnitude.

The second session of the workshop was devoted to geologic and field evidence for retention processes and their representation in models. The first paper in this session was presented by Anders Winberg of Contera AB, Sweden. His presentation (and another one by Martin Mazurek) described the *in-situ* migration

experiments using conservative and sorbing radio tracers at various space scales (from < 10 m to > 100 m) in the Äspö underground laboratory. The interpretations of the experiments indicated that diffusion and sorption are the most important retention processes, although opinions differed on how much pore space was involved in these processes. The effective diffusivity and sorption coefficients obtained in *in-situ* experiments were much greater than those obtained in the laboratory. Martin Mazurek from the University of Bern expanded on Winberg's presentation by describing the modeling done to interpret the tests. The breakthrough curve interpreted using a dual porosity model clearly indicated the existence of matrix diffusion into fault gouge at the scale of few meters. Mazurek stated that extrapolation to larger scales however was problematic. Juhani Suksi of the University of Helsinki presented the results of a Finnish natural analog study in which he applied a selective extraction technique to estimate the sorption on to a boulder from old glacial flow systems. Several questions were raised on whether the selective extraction system represented only the sorption processes or a mixture of processes. Gunnar Buckau from the Institute for Nuclear Waste Management in Germany spoke about the impact of colloid-mediated transport on performance assessment. Irreversible actinide binding on colloids and their unretarded transport can enhance risks, but Buckau stated that the present level of process understanding is not sufficient to determine if indeed the binding was irreversible. John Smellie of Conterra AB suggested that evidence of immobilization present at the Oklo site was not relevant to the Swedish repository as processes at Oklo occurred at much higher temperatures than are expected at the repository. He recommended analog studies with respect to redox processes accompanying present-day ground water recharge (e.g., weathering fronts). Ed Sudicky, University of Waterloo described his team's current efforts on developing three dimensional transient models of the coupled surface and ground water processes at a basin scale. This model will be capable of predicting the development of the surface drainage patterns along with changes in the ground water system.

The third session consisted of nine posters related to matrix diffusion, sorption, and colloidal transport. On matrix diffusion, the primary point appeared to be that the entire matrix porosity may not participate in the diffusion process and that it may be difficult to estimate the porosity that does participate in diffusion without conducting large-scale site specific *in-situ* tracer tests. A paper described the development of a numerical flow and transport model incorporating variable density flows along with nonlinear retention processes.

The fourth session of invited papers included only three papers. The first one was presented by Paul Smith (SAM Ltd., UK) who reviewed the incorporation of retention processes in performance assessments of various national programs. He concluded that with respect to sorption and matrix diffusion, the level of current understanding and information is generally adequate to allow performance assessments to take credit for these retention processes, although a reduction in key uncertainties, for example regarding potentially detrimental processes and events, is desirable. Next, I provided a regulatory perspective on representation of retention processes in performance assessments. I related the regulatory perspective to the regulator's mission of acquiring reasonable assurance regarding safety, protection of public health, and the environment. I also pointed out that the regulatory requirement of multiple barriers should also be considered in determining the adequacy of the current knowledge. I concluded by saying that with the current repository concepts incorporating long container lives, the understanding of the sorption and the matrix diffusion processes was sufficient to make initial licensing decisions and that studies to enhance understanding will continue during the operational periods. The last paper in this session was on the WIPP project presented jointly by Larry Brush (Sandia) and Chuck Byrum (EPA) describing the role retention processes played in the certification of WIPP. Larry emphasized that EPA required the DOE to justify the sorption coefficients for Culebra aquifer used in performance assessment and that EPA independently verified some of those values.

The workshop was then divided into five discussion groups: (i) Sorption, (ii) Matrix diffusion, (iii) Colloid-Related Retention Processes, (iv) Immobilization, and (v) Synthesis of GEOTRAP and Proposals for

Followup. The summary from these groups will be included in the workshop proceedings. I attended the fifth discussion group. It was generally agreed that the GEOTRAP workshop format had proved to very useful for discussion and information exchange and should be continued in the future. Several proposals including one on formulation of conceptual models through the use of various types of qualitative and quantitative data will be presented to the NEA for consideration.

PENDING ACTION

Complete the paper for submission to GEOTRAP proceedings.

SIGNATURES:

Budhi Sagar — Date: 5-15-2001

CONCURRENCE:

English Pearcy — Date: 5/15/2001

Gordon W. Wittmeyer — Date: 5/18/2001

Wesley C. Patrick — Date: 5/21/2001

AEN
NEA

Fifth GEOTRAP Workshop

Geological Evidence and Theoretical Bases

for Radionuclide-Retention Processes

in Heterogeneous Media

Oskarshamn, Sweden
7-9 May 2001

A workshop organised in the framework of

GEOTRAP,
the OECD/NEA Project on the Transport of Radionuclides in Heterogeneous Geologic Media

and

Hosted by the
Swedish Nuclear Fuel and Waste Management Company
SKB, Sweden

Final Programme

PROGRAMME

Monday, 7th May 2001

08:00 - 08:30 **Registration**

08:30 - 09:00 **Welcome addresses**
SKB, NEA

Session I

Fundamental Understanding of Individual Retention Processes

(25-minute presentations, 15 minutes discussion per paper)

Chairmen: Ivars Neretnieks and Jan-Olof Selroos

This session addresses the mechanistic description of retention processes relevant to transport in the geosphere, the general understanding that is available, and current capabilities to model the processes. The first presentation is intended to survey the topic of retention from the standpoint of geologists outside the specific field of radioactive waste disposal.

09:00 - 09:40 **How Geologists View Retention**
Mike Heath (University of Exeter)

Questions to be addressed:

1. What is the geologic significance of retention?
2. What retention processes are recognised?
3. Under what conditions do these processes occur?
4. On what time and spatial scales are these processes thought to be active, and in what media of interest for waste disposal?
5. Are the processes temporary, permanent, reversible, ...?
6. To what types of elements do these processes apply?
7. What geologic observations support our belief in, and understanding of, these processes?

The following presentations have been chosen to cover a number of broad categories of retention processes. Both present-day knowledge and future developments and trends concerning improvement and extension of the experimental bases and modelling efforts will be taken into account. Questions to be addressed in all presentations are:

1. What is the scope of the process or processes covered?
2. What is the experimental basis (broadly speaking)?
3. What are the possibilities for mechanistic modelling?
4. What are the data needed for modelling?
5. What is the range of applicability of models?
6. Which processes/factors could affect the effectiveness of the retention process?
7. What is your judgement about the reliability of our current understanding?
8. What are the future prospects for resolving uncertainties?

09:40 - 10:20 **Molecular Scale Observations and Models of Sorption Reactions**
James Davis (USGS)

10:20 - 10:50 *Coffee break*

10:50 - 11:30 **Matrix Diffusion: Heavy-tailed Residence-time Distributions and their Influence on Radionuclide Retention**
Roy Haggerty (Oregon State University)

11:30 - 12:10 **The Role of Colloids in Radionuclide Retention by and Transport Through Geologic Media**
Bruce Honeyman (Colorado School of Mines)

12:10 - 13:30 *Lunch break*

13:30 - 14:10 **Immobilisation**
Susan Stipp (University of Copenhagen)

14:10 - 14:50 **Isotopic Fractionation in Radionuclide Transport in Geologic Disposal of Nuclear Waste**
William Murphy (California State University, Chico) and David Pickett (CNWRA)

Session II

Geologic and Field Evidence for Retention Processes and their Representation in Models

(25-minute presentations, 15 minutes discussion per paper)

Chairmen: Budhi Sagar and Peter Jürgen Larue (GRS)

This session addresses the evidence for, and understanding of, retention processes provided by field experiments, nature observations, and natural analogues, and how these processes can be represented in models. The temporal and spatial scales to which the various sources of evidence apply are to be emphasised. Also addressed is the degree to which it is possible to integrate retention processes in transport models[1]. Questions to be addressed by all speakers (except for the final presentation on integration) are:

1. How are the geologic and/or field data related to laboratory information?
2. What are the generic aspects of the information?
3. How have the needs of PA influenced the investigation?
4. What additional processes affect the processes under study? On what time and/or spatial scales?
5. How are the processes conceptualised or modelled?
6. How unique is the interpretation, and how might ambiguities be resolved?
7. What are the key uncertainties in the interpretation or model, including those related to the time and spatial scales of the observations?

[1] We note that such detailed transport models may differ from the (often simplified) transport models used in performance assessment, and discussed in Session IV.

14:50 - 15:30 **Evidence for Retention Processes in the TRUE Experiments**
Anders Winberg (Conterra AB)

15:30 - 16:00 *Coffee break*

16:00 - 16:40 **Do We See *In Situ* Sorption? – Can Useful Information Be Derived for Migration Modelling?**
Juhani Suksi (University of Helsinki), Kari Rasilainen (VTT), Cécile Le Guern (BRGM), and Timo Ruskeeniemi (GSF)

16:40 - 17:20 **Evidence for Matrix Diffusion in the TRUE-1 Block at Äspö Based on Fracture Characterisation and Modelling of Tracer Tests**
Martin Mazurek (University of Bern) and Andreas Jakob (PSI)

17:20 - 18:00 **Impact of Colloids on Long-term Safety in Performance Assessment of Nuclear Waste Disposal**
Gunnar Buckau (INE) Claude Degueldre (PSI), and Annie Kersting (LLNL)

Tuesday, 8th May 2001

08:30 - 09:00 Poster set-up

Session II (continued)

09:00 - 09:40 **Solute Immobilisation: Observations from Natural Analogue Studies**
John Smellie (Conterra AB)

09:40 - 10:20 **Integration and Evaluation of Processes in Basin-scale Models of Radionuclide Transport**
Jon Sykes, Ed Sudicky, Stefano Normani, and Robert McLaren (University of Waterloo) and Mark Jensen (Ontario Power Generation)

Questions to be addressed by the presentation on integration:

1. Which processes can be integrated in transport models and which cannot?
2. What simplifications are required for this integration?
3. How well understood is the coupling among processes?
4. How confident can we be that all processes are treated either realistically or conservatively?
5. What are the prospects for improved understanding and more capable models?

Session III

Posters

10:20 - 10:40 Brief oral introductions to posters by authors (approx. 2 min. per poster)

10:40 - 12:00 Viewing of posters, with authors present (including *coffee break*)

Contributions:

Effects of Heterogeneous Porosity on Retention - Results from the TRUE Laboratory Experiments,
J. Byegård, M. Skålberg, H. Widestrand, and E.-L. Tullborg.

Modelling Colloid-facilitated Radionuclide Transport in Groundwater,
V. Cvetkovic and G. Lindgren.

Recent Developments in the Modelling of Transport and Retention of Radionuclides in Porous Media,
E. Fein, U. Noseck, and T. Kühle.

Immobilization and Retention Processes of Uranium in Tertiary Argillaceous Sediments (Czech Republic),
A. Laciok, M. Hercik, U. Noseck, and T. Brasser.

Comparison of Formation Factor Logs Obtained by Electrical Methods *In Situ* and in Laboratory,
M. Löfgen, Y. Ohlsson, and I. Neretnieks.

Modelling of Colloid-Facilitated Contaminant Transport with the Computer Code TRAPIC: Theoretical Basis and Application,
U. Noseck.

Matrix Diffusion - Through Diffusion Versus Electrical Conductivity Measurements,
Y. Ohlsson, M. Löfgren, and I. Neretnieks.

Discordance in Understanding of Isotope Solute Diffusion and Elements for Resolution,
C. Pescatore

Natural Uranium Concentration in Boom Clay: Influences of Carbonate and Natural Organic Matter,
L. Wang, M. De Craen, and N. Maes.

12:00 - 13:30 *Lunch break* (posters on display during lunch break)

Session IV

Consideration and Representation of Retention Processes in Performance Assessment and Justification of Treatment

(25-minute presentations, 15 minutes discussion per paper)

Chairmen: Pedro Hernán and Richard Beauheim

This session addresses how various retention processes are represented in performance assessment. Some processes are included within performance assessment models in a very simple manner (e.g. via the K_d parameter for sorption), or via a more complex approach. Other processes may be omitted from performance assessment models and discussed in a purely qualitative manner. Whatever the approach, its acceptability, e.g. to regulators and to technical reviewers, is likely to depend on careful justification.

The session includes an overview of the representation of retention processes in performance assessments carried out by waste-management organisations and an overview of how such representations (and the justifications for them) have been judged by regulatory authorities. A case study of the representation of retention processes in performance assessments leading up to licensing and for the future recertification of the Waste Isolation Pilot Plant (WIPP) in New Mexico, USA, is also given.

13:30 - 14:10 **The Consideration and Representation of Retention Processes in Performance Assessments Carried Out by Waste-management Organisations – What Has Been Done and Why?**
Jürg Schneider, Bernhard Schwyn, and Piet Zuidema (Nagra), Hiroyuki Umeki (NUMO), and Paul Smith (SAM Ltd.)

Questions to be addressed:

1. Which retention processes are normally included?
2. Which retention processes have been included quantitatively and which qualitatively?
3. What simplifications have been made for the purposes of performance assessment modelling; what are the motivations and justifications for these simplifications?
4. To what extent has geologic and field evidence (field experiments, observations of natural systems, etc.) been included in PA model representations of retention processes and in qualitative discussion of retention processes in PA; how is evidence transferred to temporal and spatial scales relevant to performance assessment?
5. What are the strengths and weaknesses of current PA models in terms of the representation of retention processes?
6. What future trends can be foreseen in how retention processes might be represented in PA (additional processes included in models, refinement of existing model representations, etc.)?

14:10 - 14:50 **The Consideration and Representation of Retention Processes in Performance Assessment – A Regulatory Perspective**
Budhi Sagar (CNWRA), Richard Codell (US NRC), and Bo Strömberg (SKI)

Questions to be addressed:

1. Is the current understanding of retention processes adequate for current and future PA purposes?
2. How important is the representation of heterogeneity and temporal variability viewed for the modelling of retention in PA?
3. To what extent should geological evidence (observations of natural systems) be used in PA?
4. How do regulatory bodies view the adequacy of simplifications that are made for the purposes of performance assessment modelling?
5. What are the strengths and weaknesses of current PA models in terms of the representation of retention processes?
6. What future trends can be foreseen in how retention processes might be represented in PA (additional processes included in models, refinement of existing model representations, etc.)?

14:50 - 15:30 **The Consideration and Representation of Retention Processes in the WIPP Performance Assessment: Justification of Adopted Approaches and Interaction with the Regulator**
Laurence Brush, Charles Bryan, Lucy Meigs, Hans Papenguth, and Palmer Vaughn (SNL) and Charles Byrum and Thomas Peake (US EPA)

Questions to be addressed:

1. Which retention processes have been included qualitatively and which quantitatively, in how much detail and with what justification?
2. What is the regulatory view of the treatment of retention processes in WIPP PA?
3. Did intervenors question the treatment of retention processes in PA, and were these questions taken into account?
4. What changes in the representation of retention processes can be foreseen in recertification?
5. How are the changes justified?
6. What is the role of a continuing research programme in recertification?

15:30 - 16:00 *Coffee break*

Session V

In-depth Discussions by Working Groups

16:00 - 16:15 Introduction to working groups and working group tasks (NEA)

16:15 - 18:30 Discussions in groups (generally beginning with a short presentation of topics for discussion by the chairman – coffee break included)

Working group topics have been set to cover all of the broad categories of retention processes described in Sessions I and II:

WG1: Sorption
Chair: Scott Altmann (ANDRA)

WG2: Matrix diffusion
Chair: Ivars Neretnieks (KTH)

WG3: Colloid-related retention processes
Chair: Jörg Hadermann (PSI)

WG4: Immobilisation
Chair: Mike Heath (University of Exeter)

These four working groups will discuss, in the context of each broad category of retention processes:

1. What issues have been sufficiently resolved for current and future PA purposes?
2. What future trends can be foreseen and in particular, what, if any, unresolved issues should still be addressed?

In addition, a special working group will discuss the lessons learnt from the complete GEOTRAP workshop series, and what follow-up actions may usefully be taken.

WG5: Conclusions and synthesis of GEOTRAP and proposals for follow-up
Chair: Alan Hooper (Nirex)

- This working group is to be attended, potentially, by members of the IGSC, GEOTRAP contact persons, and others.
- A discussion paper on lessons learnt from previous GEOTRAP workshops, and future possibilities, is to be prepared and circulated in advance of the fifth workshop.

Wednesday, 9th May 2001

Session V (continued)

09:00 - 11:00 Discussions in groups

11:00 - 12:00 Working group chairmen to prepare presentations of findings for Session V

12:00 - 13:30 *Lunch break*

Session VI

Working Group Presentations and Final Discussions

13:30 - 15:30 Presentations by working group chairmen
(15 minutes + 10 minutes discussion for each group)

15:30 - 16:00 *Coffee break*

16:00 - 17:15 Final discussion of topics arising in the course of the workshop
Moderator: Claudio Pescatore (NEA)

Closing of the workshop

17:15 - 17:30 Closing addresses (SKB/NEA)

Workshop Dinner hosted by SKB

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