# CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

## TRIP REPORT

**SUBJECT:** Seventh International Conference on the Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere (Migration '99)
20.01402.871

**DATE/PLACE:** September 26–October 1, 1999
Lake Tahoe, Nevada

**AUTHORS:** R.T. Pabalan, D.R. Turner, J.D. Prikryl, and L. Browning

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**PERSON(S) PRESENT:**

In addition to the authors, D. Esh and J. Contardi of the Nuclear Regulatory Commission (NRC) participated. The meeting was attended by scientists from various U.S. and international organizations.

**BACKGROUND AND PURPOSES OF TRIP:**

The Migration conferences focus on recent developments in the fundamental chemistry of actinides and fission products in natural aquifer systems, their interactions and migration in the geosphere, and the processes involved in modeling their geochemical behavior. The purpose of the trip was to present two papers discussing the results of NRC-funded work. The papers are "Uranium(VI) Sorption Behavior on Mixed Silicate Minerals," co-authored by J.D. Prikryl, A. Jain, D. Turner, and R. Pabalan, and "Modeling Colloid Transport for Performance Assessment," co-authored by J.S. Contardi (NRC), D.R. Turner, and T. Ahn (NRC). The former paper was presented by J.D. Prikryl in a poster session, and the latter was orally presented by J.S. Contardi. Another objective of the trip was to gather technical information that would be useful for CNWRA technical assistance activities in the NRC high-level waste program.

**SUMMARY OF PERTINENT POINTS:**

The primary mode of dissemination of technical information at the 5-day Migration '99 conference was by evening poster sessions designed to encourage intensive communication between the authors and participants, and daily oral sessions with invited and contributed papers. About 211 papers were presented in the poster sessions and 50 papers were presented in the oral sessions. The sessions covered the following technical areas:

(A) Chemistry of actinides and fission products in natural aquatic systems
- Solubilities and dissolution reactions
- Complexation with inorganic and organic ligands

- Redox reactions
- Colloid formation
- Experimental methods

(B) Geochemical interactions and transport phenomena
- Diffusion and migration in geologic media
- Sorption/desorption phenomena
- Natural analog studies
- Effects of biological activities and organic materials
- Colloid transport
- Radionuclides in soils
- Soil-remediation chemistries

(C) Data base development and modeling
- Data selection and evaluation
- Data base management
- Geochemical models and modeling
- Application of models
- Validation of modeling results

The conference was very successful as judged by the large attendance (>300) and generally high caliber of scientific papers presented. Many oral and poster presentations have relevance to geologic disposal of nuclear waste and performance assessments (PA) for geologic repositories. Most of the talks and posters were focused on laboratory experiments, including imaging techniques at the atomic level and spectroscopic measurements on aqueous and solid phases. Much of the research presented was funded by international radioactive waste management programs, and concerns of these programs have apparently driven experimental design. Nevertheless, despite the presentation of excellent research results, there was not much discussion of end-use of the laboratory results, or site-specific applications to waste management issues. The final day of the conference was dedicated to sessions on current status within different repository and remediation programs; this would perhaps be more appropriate earlier in the conference.

Papers submitted to the conference organizers are to be published in the peer-reviewed journals Radiochimica Acta or the Journal of Contaminant Hydrology. Brief summaries of some of the papers presented at the conference are given in the following paragraphs. A copy of the conference abstracts can be borrowed from the authors of this report.

J.A. Davis (USGS) opened the conference with an excellent presentation on solute transport modeling in which different methods for describing radionuclide sorption (e.g., $K_d$, surface complexation) were compared. Site specific data from a contaminated uranium mill tailings pile was used to derive hydrologic parameters and chemical conditions used in the modeling. Results indicated that surface complexation modeling is an effective tool for estimating $K_d$ values at sites with variable chemical conditions. J. Davis also proposed that sorption data be normalized to the surface area of the sorbent phase. He showed a figure in which he plotted his data on U(VI) sorption on three minerals (ferrihydrite, quartz, and kaolinite) in terms of $K_A$ ($K_d$ normalized to the mineral surface area) as a function of pH. The $K_A$ values for the different minerals plot on top of each other. He supported his proposed surface-area normalization scheme by showing two figures taken from CNWRA publications that showed uranium(VI) and neptunium(V) sorption on different minerals are essentially the same if normalized to the mineral's effective surface area.

A. Tompson (LLNL) reported the results of reactive transport simulations used to evaluate the near-field migration of radionuclides in groundwater away from underground nuclear tests located under Frenchman Flat at the Nevada Test Site (NTS). The cavity created by the nuclear explosion contains melt glass and rubble containing high radionuclide concentrations. Transport simulations illustrated that radionuclide release is particularly sensitive to the available surface area of the melt glass and rubble within the cavity and that transport is sensitive to the groundwater flow configuration of the system. An interesting aspect of this work was the technical approach—the authors represented 3D geochemical evolution across the site by considering reactions within a collage of 1D flow lines through the mountain. This streamline modeling approach allows detailed characterization across large areas, although it does not allow the flow pathways to evolve as a result of diffusion or permeability changes.

K.E. Roberts (LLNL) presented results of a study on the precipitation of crystalline Np(IV) oxide from near-neutral aqueous solutions at high temperatures (200 °C). The authors stressed that this is the first time that crystalline Np(IV) oxide has been observed to precipitate from near-neutral solution. The authors hypothesized that Np(IV) oxide is the thermodynamically stable solid phase in near neutral solutions at low temperatures, as is indicated by existing thermodynamic data at 25 °C, but it is slow to form at the time scale of laboratory experiments.

R. Finch (ANL) described the solid alteration products of corroded nuclear waste forms (e.g., spent $UO_2$ fuel and borosilicate nuclear waste glasses) and their role in limiting the release of U, Pu, Np, and lanthanides. Uranium released from corroded nuclear wastes forms uranyl oxyhydroxides and silicates. Pu, Am, and REE tend to precipitate as amorphous residue on the surfaces of corroded spent fuel, whereas Pu, Am, and REE commonly form crystalline phosphates in corrosion experiments on nuclear waste glass. Neptunium enters the structure of the uranyl oxyhydroxides formed from corrosion of spent fuel but uranyl silicates formed from spent fuel corrosion do not incorporate significant Np.

M.C. Duff (SREL) investigated the distribution of sorbed Pu on tuff from Yucca Mountain, (YM) NV. Thin sections of the tuff material were exposed to a Pu-bearing synthetic groundwater and then analyzed by microprobe x-ray fluorescence. Microprobe x-ray fluorescence maps showed sorbed Pu to be highly localized and to be spatially associated with Mn oxides and smectites, not with iron oxides or zeolites.

Colloid formation and transport were the subject of several presentations. A. Kersting (LLNL) presented evidence for colloid migration of Pu at the NTS. These findings are one of the main reasons that colloid transport has resurfaced as an issue in the YM Project. Pu associated with colloids has been found in groundwaters at Pahute Mesa, about 1.3-km downgradient from the BENHAM detonation cavity. Isotopic ratios support BENHAM as the source of the Pu, and analysis of the materials suggest a low colloid concentration (0.2 mg/L) of mostly inorganic Ca-aluminosilicate minerals.

P. Vilks (AECL Whiteshell Laboratories) presented results of laboratory experiments designed to study colloid transport of Am and Sr through fractured granite. Without colloids, no Am was transported on a scale of 1-meter. With the introduction of colloids, there was early breakthrough of Am, but only 1–2 percent recovery. Strontium transport was not strongly affected by colloids, and increasing colloid concentration to ~3 g/L lead to early breakthrough, but even lower recovery.

H. Geckeis (FZK, Karlsruhe, Germany) presented an overview of colloidal transport of actinides. He presented a general empirical linear relationship between colloid diameter and particle concentration based

on a review of natural systems around the world. Kinetically controlled desorption from colloids suggests that irreversibility may be significant with regard to colloid transport.

W. Schubler (Inst. Nukleare Entsorgungstechnik) discussed the results of column experiments and models to identify the processes related to humic colloid borne Am(III) migration. It was found that interactions between humic acids and AM are kinetically controlled.

Studies presented by C. Mertz (ANL) support association of Pu with a colloidal phase during glass dissolution. Increases in ionic strength induced aggregation and settling, reducing colloid concentration.

A few presentations focused on the use of laboratory results in assessment calculations. S. Serkiz (WSR) presented a means of using sequential extraction to simulate U(VI)-sorption on composite soils. By separating out the relative influence of different types of mineral coatings on sorption, the authors were able to match laboratory sorption results extremely well using a relatively simplified model.

D. Kaplan (WSR) examined different ways to correct soil Sr(II)-sorption parameters ($K_d$) for the presence of low surface area/low sorption gravels. The most accurate simulation of Sr sorption assumed a proportional composite of the fine soil $K_d$ and the coarse gravel $K_d$. This additive approach offers a more accurate and more conservative measure of sorption parameters, as opposed to standard approach of using sorption parameters determined using the fine (diameter < 2 mm) soil component.

D. Esh (NRC) presented reactive transport simulations of thermo-hydro-chemical (THC) processes at YM using the computer code TOUGH2. Several conceptual models of fractures were evaluated to determine the effects of THC processes on the concentration of chloride. A maximum chloride concentration of 100,000 mg/L was predicted under extreme conditions caused by evaporation/condensation processes due to heating.

**IMPRESSIONS/CONCLUSIONS**

Migration '99, the seventh in a series of international conferences, offers perhaps the best opportunity to meet and discuss current research in the chemistry and migration behavior of actinides and fission products in the geosphere. The meeting was dominated by discussions of detailed experimental techniques, and suggestions about how to relate this data to the development of process-level or PA models were seldom offered. Nevertheless, the information presented at the conference and the opportunity for interactions with high caliber scientists from different countries and nuclear waste programs are invaluable resources for CNWRA technical assistance activities in the high-level waste program.

**PROBLEMS ENCOUNTERED:**

None.

**PENDING ACTIONS:**

A manuscript on U(VI) sorption on mineral mixtures was submitted for publication in the Journal of Contaminant Hydrology. Revisions will be made to the manuscript upon receipt of comments from peer reviewers.

**RECOMMENDATIONS:**

Continued participation by CNWRA staff in future Migration conferences is highly recommended.

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