

CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Meeting on the NRC Office of Research Geochemistry Program
(20.01402.871)

DATE/PLACE: September 26–28, 2001
Rockville, Maryland

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PERSONS PRESENT:

F.P. Bertetti (CNWRA)	W. Ott (NRC ORS)
D.A. Pickett (CNWRA)	R. Cady (NRC ORS)
D.R. Turner (CNWRA)	T. Nicholson (NRC ORS)
J. Davis (USGS)	J. Randall (NRC ORS)
G. Curtis (USGS)	L. Veblen (NRC ORS)
R. Cygan (SNL)	J. Bradbury (NRC NMSS)
J. Serne (PNNL, by telephone)	

BACKGROUND AND PURPOSE OF THE TRIP:

The U.S. Nuclear Regulatory Commission (NRC) Office of Research currently funds several different projects related to radionuclide sorption and transport. Contractors include Jeff Serne at Pacific Northwest National Laboratory, Jim Davis and Gary Curtis at the U.S. Geological Survey, and Randy Cygan at Sandia National Laboratories. NRC Research-funded projects range from laboratory investigations of the effects of organics on plutonium sorption, analog studies of uranium migration at Naturita, Colorado (U.S. Geological Survey and Sandia National Laboratories), microanalysis and molecular dynamic simulation of the mineral/water interface (Sandia National Laboratories), to uncertainty and sensitivity analyses of performance assessment results to sorption model parameters (Sandia National Laboratories).

Partly from a desire expressed by the Advisory Committee on Nuclear Waste to provide better integration of NRC's various research programs, the Center for Nuclear Waste Regulatory Analyses (CNWRA) was invited to participate in this meeting. CNWRA staff presented an overview of the current technical assistance work on radionuclide sorption and transport undertaken as part of the Office of Nuclear Material Safety and Safeguards, Division of Waste Management, High-Level Waste program.

An additional focus of the meeting was organizing NRC participation in the upcoming Sorption Modeling exercise being coordinated by the Nuclear Energy Agency.

MEETING SUMMARY

The following is a brief summary of presentations made by the meeting participants. The authors of this trip report have copies of presentation materials, and a copy of the draft research project plan developed for radionuclide transport in the environment.

R.J. Serne and K.J. Cantrell (Pacific Northwest National Laboratory)

R.J. Serne of Pacific Northwest National Laboratory presented the results of batch and column sorption experiments conducted with plutonium and neptunium. The Pacific Northwest National Laboratory sorption data for actinides and complexing ligands may be of direct benefit to the high level waste program. Outside of the data presented by Pacific Northwest National Laboratory, there are few data available on the effects of organic complexing on neptunium and plutonium sorption. Serne also suggested a look-up table (similar to the response surface proposed by CNWRA staff) method may be an efficient mechanism for incorporating variation in K_d into performance assessment codes. Another interesting point was the possibility that the complexing ligands might act to dissolve or degrade the Fe-oxide coatings of soil/mineral grains over time. This could lead to breakthrough or release of much larger than expected nuclide concentrations, and is evidence for the potential nonconservative predictions inherent in the constant K_d approach.

J. Davis, G. Curtis et al. (U.S. Geological Survey)

J. Davis and G. Curtis presented a summary of about four years of work at the Naturita, Colorado uranium mill tailings site. Curtis described the hydrologic characterization of the site. A shallow (10–20 ft thick), unconfined aquifer of alluvial deposits along the San Miguel River has been characterized and instrumented with about 60 monitoring wells. Uranium concentrations have been measured for groundwater, and hydraulic conductivity has been determined based on several methods, including groundwater age dating, chloride migration, slug tests, and tracer tests. A general conceptual model includes recharge into the aquifer at a bend in river upstream from the site, groundwater flow parallel to the river, and discharge back to the river about 2 km downstream. There is an assumed no flow boundary at a road along the site and some leakage between the river channel and the aquifer.

Davis presented batch sorption results using natural materials from the site, showing the significant effects of pH, uranium concentration, and CO_2 partial pressure on uranium sorption. He also presented information regarding techniques of measuring *in situ* K_d values through field-based experimentation and assessing the “true” sorption capacity of mineral grains through measurements of isotopic exchange. Both techniques appeared to be promising approaches to the determination of site-specific sorption characterization data. He also discussed laboratory methods for measurement of “*in situ*” uranium sorption, using an alkaline sodium carbonate extractant. These methods may prove useful at other analog sites for comparison with laboratory sorption measurements. Field-based “push-pull” methods for carbonate extraction are also underway at Naturita.

Davis has calibrated a nonelectrostatic surface complexation model to the batch sorption data and used it to investigate the key geochemical parameters controlling uranium sorption at Naturita. Calculated *in situ* K_d values showed good agreement in pH space (over a range of CO_2 partial pressures) with the range of surface complexation model results. Because of the limited pH range of Naturita waters, calculated K_d showed correlations only with uranium concentration and alkalinity, and model sensitivity to pH was not really tested. Point-to-point comparisons between calculated K_d and measured *in situ* K_d were not shown, but would provide another good test of both models and experimental approaches.

In Naturita soils, there is evidence for association of sorbed uranium with both clays and Fe oxides. Microanalysis of uranium-contaminated soils suggests the clay association, and experimental U(VI) sorption onto uncontaminated soils favors Fe oxides.

J. Davis' presentations stressed the difficulty with assigning a constant K_d when simulating uranium transport at Naturita. The constant K_d , even if a low value was used, did not present a reasonable simulation of the observed variability in uranium concentration, and could lead to a misleading evaluation of "in-place" remediation techniques proposed for groundwater cleanup at the site. The U.S. Geological Survey results for the Naturita site also suggested that the experimental approach of CNWRA sorption projects has been appropriate, with the possible exception of a general lack of data regarding variation in radionuclide concentration. Although expected concentrations of radionuclides of any interest are likely to remain low (relative to available mineral surface sorption sites) as a result of solubility limits and other factors, it would be useful to have more data on the concentration effects on sorption behavior as a test of model robustness. Davis also discussed the usefulness and applicability of spectroscopic studies in providing realism to the selection of sorbed species for surface complexation models. A final point made by Davis stressed the "maturity" of the surface complexation model approach to sorption modeling, and discussed the well established and technically sound basis for its inclusion into performance assessments.

R. Cygan et al. (Sandia National Laboratories)

Characterization of Naturita sediments suggests similarities of the coatings to other Fe-oxide and clay mineral coatings observed in other coastal plain sediments. The apparent ubiquity of Fe-oxide coatings may not mean that Fe-oxides necessarily play a dominant role in sorption of dissolved species on natural materials, as has been suggested by several investigators in the past. Additional fine-scale characterization conducted by Cygan and coworkers seems to indicate that sample pre-treatments can possibly remove or alter natural coatings to skew observations in favor of Fe-oxide dominated coatings. It would be useful to compare several unconsolidated sediment regimes to examine surface coatings more closely. In other work presented by Cygan, fine-scale spectrographic techniques and molecular dynamics simulations offered insight into the mechanisms of sorption processes. This type of information may help to constrain application of surface complexation models and provide a more defensible position for their application in performance assessment or other risk-informed licensing proceedings.

Nuclear Energy Agency Sorption Project

During Phase I of the Nuclear Energy Agency Sorption Project, the technical direction team has established a group of seven sorption data sets for testing the applicability of detailed geochemical models in simulating radionuclide sorption. Two of the data sets (uranium-quartz and neptunium-montmorillonite) are from the CNWRA. Each data set was selected to investigate one or more specific aspects of geochemical sorption modeling, and the strengths and weaknesses of different approaches. J. Davis (U.S. Geological Survey) is a member of the technical direction team that identified and developed the data sets, and L. Veblen (NRC ORS) is a member of the management advisory board for the exercise.

NRC is sponsoring three teams (NRC staff, U.S. Geological Survey, CNWRA), and about 12 to 15 teams from Europe and Japan also plan to participate in the Phase II modeling of the data

sets selected by the technical direction team. It is interesting to note that no team from the DOE has committed to the project. The data have been gathered but not distributed yet. Pending the resolution of some contractual issues with the Nuclear Energy Agency, Davis indicated that the data will be provided to the modeling teams in the near future, with about six months scheduled for the first phase of modeling.

SUMMARY AND CONCLUSIONS:

The NRC Office of Research has an active program in radionuclide sorption and transport. The work at Sandia National Laboratories and at Naturita is reaching a close. It was not clear if there would be an extension to the effort, although it was clear that both NRC and the U.S. Geological Survey/Sandia National Laboratories team wanted to continue.

Because the NRC Office of Research does not sponsor research in the area of high level waste disposal, the results of the work presented at this meeting are currently under investigation for application in NRC non-high level waste programs (e.g., uranium recovery, decontamination and decommissioning, low-level waste disposal). The Office of Research staff, however, indicated both in discussions and in a draft research program plan in circulation for comment, that they anticipate future tasking at a small level in high-level waste. There is increased sensitivity in the NRC with regard to conflict-of-interest. The CNWRA, with its freedom from conflict-of-interest and familiarity with the high-level waste program, is well-positioned to provide technical support in geochemical research.

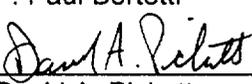
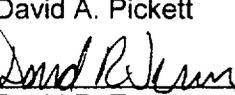
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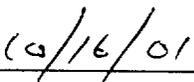
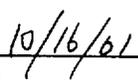
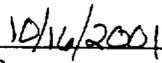
None.

PENDING ACTIONS:

CNWRA staff will participate in Phase II of the Nuclear Energy Agency Sorption Project. This activity has been approved by the NRC Project Manager for the Radionuclide Transport key technical issue and is identified in the fiscal year 2002 CNWRA Operations Plan.

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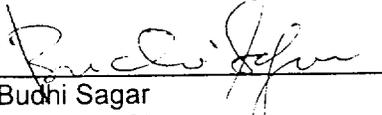

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