OFFICE OF NUCLEAR MATERIAL SAFETY AND SAFEGUARDS
DIVISION OF HIGH-LEVEL WASTE REPOSITORY SAFETY
TRIP REPORT

SUBJECT: Report of Staff Exchange with the Center for Nuclear Waste Regulatory Analysis on Colloidal Radionuclide Transport and Drift Collapse

DATE: October 31, 2005 - November 4, 2005

PLACE: San Antonio, Texas

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PERSONS PRESENT: Researchers and scientists from the Center for Nuclear Waste Regulatory Analyses and U.S. Nuclear Regulatory Commission staff including Aladar Csontos, and Allen Fetter. Other U. S. Nuclear Regulatory Commission staff, including Christopher Grossman, Tae Ahn, Randall Fedors, among others, also participated through teleconferencing.

BACKGROUND AND PURPOSE OF THE TRIP

The most recent revision of Total-system Performance Assessment code (TPA) 5.0 has suggested the possibility of large doses as a result of the newly implemented colloidal radionuclide generation and transport modules. While it is well known that colloids may facilitate the transport of radionuclides and heavy metals in the subsurface, it remains to be determined that the relatively large doses in the recent TPA revision are physically based. The first purpose of our staff exchange with the Center for Nuclear Waste Regulatory Analyses (CNWRA hereafter Center) was to discuss with the researchers the causes of the large dose predictions, to formulate strategies and to design future research work to resolve the issues.

It is likely that drift collapse would modify the near-field thermohydrology and percolation flow paths, affect the corrosion rate of the waste packages, and the release of radionuclides. Initial literature reviews at the U.S. Nuclear Regulatory Commission (NRC) and the Center did not resolve the issues and the staff and Center scientists have not yet reached consensus regarding the modified flow paths and their effects on the engineered barrier system.

Our second purpose for the staff exchange was to formulate strategies and to design further laboratory experiments to obtain important information and model parameters. These experiments will be used to refine the model abstraction of drift collapse and near-field thermohydrology in TPA and also to provide bases for reviewing the Total System Performance Assessment (TSPA) License Application (LA).

SYNOPSIS OF DISCUSSION AND ACTION ITEMS

Colloid Formation and Radio-Colloidal Transport

We held two meetings for the purpose of resolving the colloid generation and transport issues. Participants of the first meeting include Tae Ahn, Richard Codell, Aladar Csontos, Allen Fetter, Christopher Grossman, and Jin-Ping (Jack) Gwo from NRC and Paul Bertetti, Jude McMurry, Olufemi Osidele, Scott Painter, English Pearcy, Osvaldo Pensado, David Pickett, Jim Winterle, and Gordon Wittmeyer among others from the Center. Participants of the second meeting include Richard Codell, Allen Fetter, and Jack Gwo from NRC and Paul Bertetti, Scott Painter, and David Pickett from the Center. All work on the generation and effects of colloids in performance assessments will be incorporated under SZ2, but much of the work will actually be carried out in other ISI's.

The following relevant topics and points were raised and discussed in the first colloid meeting:

- There are 16 FEPs that apply to colloids, and that 6 of those are screened-in presently.

- The participants agreed to conduct an in-depth literature survey to acquire more knowledge of the state-of-art of colloid formation and radionuclide adsorption/desorption on colloidal surfaces. A work plan or outline will also be drafted to address the issues related to the fate and transport of colloidal species in the near-field and the subsurface.

- A revised released model has been formulated at the Center, and will be placed into the TPA code. The model assumes that colloidal surface sites will be fully occupied by plutonium, americium, neptunium and uranium. Staff reviews of the open literature and U.S. Department of Energy (DOE) reports were used to estimate the affinity factors for the radioelements. Plutonium has a very high affinity compared to competing cations. The model will assume 2.2 sites per square nanometer of specific surface area of the colloids.

- One of the effects of colloids will be to effectively increase the solubility limit of actinides released from the fuel. The actinides may be immediately removed from the water phase if colloids are in close proximity to the fuel. This could be represented directly in the TPA model as an increase in solubility. The rate of sorption from the water to the colloids appears to be fast compared to the rate of flushing of the waste package by infiltrating water. NRC staff questioned whether 100% site saturation was too conservative. For example, would the activity of the surface sites decrease at high saturation and hydrolysis states of the actinides change their affinity to colloidal surface sites?

- All agreed that the proposed model would be conservative, yet more realistic than the previous colloid model, which did not take into account the availability of surface sites. These calculations have shown, in general, reduction of release by one to two orders of magnitude compared to the previous model. If the decrease is also reflected in the transport of these radionuclides and final dose calculations, one may expect a large reduction in dose as well. Should this be the case, the importance of colloid generation and transport might be diminished to a lower level, making the refinement of the model less of a priority. Before the new release model is implemented, the participants of the meeting suggested that geochemical speciation modeling of the solid interface and some laboratory experiments be planned.

- We discussed several other processes that could affect the importance of colloids. Filtration and straining of colloids are difficult to defend. Although these phenomena are included in the TPA model, they are currently not being used. Generation of iron oxyhydroxide and oxide from corroding carbon steel and stainless steel is potentially the most important source of highly sorptive colloids. There is a large quantity of steel in the engineered barrier (inner waste package, ground support, rails, nuclear fuel bundle). However, it is not clear that the generation of colloids from this steel would occur coincidently with the release of radionuclides from the waste form, whether stainless steel corrosion leads to appreciable colloids, or that the colloids produced would be sufficiently stable (both in terms of the stability field of colloids or the crystallization to more stable goethite and hematite). Furthermore, rusting steel may form insoluble oxides that would not be easily transportable from the engineered barrier, and would tend to tie-up radionuclides in place rather than creating a vector for their transport to the geosphere.

- Some of the experimental evidence the Center staff used to justify their model of colloid-facilitated transport was based on DOE experiments using a variety of colloids. One of the colloidal materials was hematite powder obtained from a chemical company. It is not clear that this material is sufficiently similar to colloids that could form in the repository from the corrosion of carbon steel or stainless steel. This consideration led to discussions about an integrated experiment looking at generation of colloids from rusting carbon steel and transport of the colloids in flowing columns packed with Yucca Mountain (YM) materials.

- We discussed the special problems posed by colloids emanating from vitrified waste forms. The current TPA model ignores glass colloids, even though there is experimental and anecdotal evidence that they could be important. Nearly all plutonium emanating from vitrified waste, and probably from melt glass in underground nuclear weapon testing, is in colloidal form, and seems to be irreversibly bound. By contrast, relatively few colloids are found in spent fuel dissolution tests, and the radionuclides must attach themselves to existing colloids to escape the engineered barrier in colloidal form. We concluded that more in-depth literature survey will need to be conducted and incorporated into a revised TPA before a clearer path forward can be factored into the current research plan. There may be additional information on the migration of plutonium-containing colloids (e.g., additional wells) at the Benham test. Peter Lichtner from Los Alamos National Laboratory, and formerly from the Center, might know more.

- There may be an OR visit on colloids in January.

- The Center might be able to conduct experiments with colloids tagged with isotope-enriched iron or a rare earth metal.

- DOE had planned experiments with colloids in alluvium, but the permit for the facility was withdrawn by the State of Nevada. The Nye County early warning wells may be a suitable test facility for colloids, but this is out of DOE and NRC hands. Paul Bertetti offered to check with Nye County to see if any such tests are planned.

The second meeting regarding colloids consisted of a smaller working group that discussed colloid scoping calculations and their implementation in the TPA code. We also discussed previous and possibly ongoing DOE experiments and reports regarding observations near and in the Nevada Test Site. The colloidal release models of TPA and TSPA-LA were compared with regard to colloid generation and filtration, colloidal size distribution, input parameter distributions, and the impact of longer compliance time.

We discussed the possibility of an intermediate scale colloid experiment in a block of alluvium carefully removed from the site, but the advantages and disadvantages were not clear. Scott Painter and Oleg Povetko could scope the process. One concern was that alluvium easily excavated from the site might not be representative of the alluvium nearer to the water table, possibly hundreds of meters below. The deeply incised banks of Fortymile Wash might show the condition of deeper alluvium. The alluvium is an immature soil, with some calcite cementation. There might be iron oxide coatings in some areas, since there was some pyrite found in the deeper materials. Clays and zeolites are more common in the alluvium, but there are not much of these materials in the tuff rocks. Iron found in the alluvium however does not seem to add too much to the sorptive capacity, and may be "passivated". There is some manganese oxide lining fractures, but it is present in small amounts only. Iron coatings seem to be most associated with silica particles.

Drift Collapse and Unsaturated Zone Thermohydrology

Two meetings were held for this topic on Tuesday, November 1 and Thursday, November 3. Participants were Richard Codell, Randall Fedors and Jack Gwo from NRC and Ronald Green, Chandrika Manepally, Kevin Smart, Stuart Stothoff, and Gordon Wittmeyer, among others, from the Center. The objective of the meeting was to come to a consensus about conducting laboratory experiments to test the effects of drift degradation on near-field thermohydrology and to improve calculations of the amount of water that may come in contact with the waste package in the drift. The effects of rubble piles from a collapsed drift on the flow paths of water are currently not well understood. Limited laboratory and field experiments and modeling efforts, including those conducted at the Center, have not conclusively identified the effects of the rubble piles on drips from the ceiling of the drifts. The composition of the rubble is poorly characterized at YM, thus presenting a large uncertainty in the near-field thermohydrology and the design of laboratory experiments.

Recent papers have addressed water infiltration through packed rubbles. Experiments in packed rock columns shows that infiltrating water that was dispersed uniformly at the top tends to concentrate along pathways into rivulets. This is an interesting observation, but does not necessarily alleviate our concern that the presence of rubble could lead to less water contact with the spent fuel rather than more. Two reasons why we believe that these experiments may not be completely relevant are: (1) the infiltration rate used in the experiments was thousands of times greater than expected infiltration rates at YM. Conditions under very low flow might be quite different; .e.g., dominated by Van der Waals forces rather than gravity; and (2) a concentrated drip into an unfilled drift is not the same as infiltrating water uniformly entering the top of the column. It is not clear that the experimental results could be de-convoluted to gain the necessary information for the dispersal of a single drip.

Several modeling ideas were discussed in the first meeting. The staff suggested that more information from the geomechanical submodel of TPA (Mechfail) needs to be obtained and more understanding of the size distribution of the rubble is needed before a laboratory experiment can be designed.

The second meeting discussed in details the design of the laboratory experiments after a preliminary plan to acquire rubble material had been proposed by Randall Fedors and possible laboratory apparatuses were identified at the Center. The discussion included not only the design of the in-drift hydrological experiments but also how the laboratory experiments could be coordinated to meet the needs of seepage chemistry calculations, estimating seismic effects on the packing of the rubble, the contact of the rubble with the waste packages and its effects on waste package corrosion, and finally the scale-up of the laboratory experimental results to the field. We also discussed the idea of setting up experiments with an engineered drift fill, or a Richards barrier to simulate the case that DOE decides to go with an engineered backfill situation. We were interested in using an existing model waste package in an approximately 3 ft wide plastic pipe, or a new experiment in a shorter length of a similar pipe. The existing experiment would have to be modified to introduce dripping water at the top and collect infiltrating water at the bottom.

We discussed the possibility of a series of experiments, both isothermal and heated, that would demonstrate the effect of rubble on the path of dripping water. The simplest and most straightforward experiments would consist of a column packed with typical rubble with water infiltrating at a single point and collected at the bottom at very low flow rates typical of YM infiltration. Several obvious concerns with the experimental design are: (1) picking the right size rock so they would be typical of a rubble column in a drift, (2) placing the rubble in an orientation typical of how rock would fall in the drift, (3) carefully controlling the environment of the experiment (e.g., temperature and humidity) so that very low infiltration rates could be demonstrated, and (4) introducing and collecting the water in the experiment to provide a useful analog to flow in the drift impinging on the drip shield or waste package. Rock typical of drift collapse could come from present rock fall in drifts, spalled rock between the drift and the ground support net, rock fall in similar tunnels like those at Ranier Mesa, and talus from similar lythophysal rock units near the site. How rock falls into a drift was not obvious to anyone, and may have to be explored by experts in rock mechanics and mining operations. Thermal spallation during the heating period might affect the type of rock fall expected. The Center has several 55 gallon drums filled with cuttings from the YM tunnel boring machine, both unwashed/unsorted and washed/sorted, but it’s unclear if this is appropriate rock for the experiments.

Other topics discussed were the effect of rock in direct contact with either the drip shield, or in the case of an engineered backfill, the waste package. Would this condition lead to enhanced corrosion because of enhanced moisture at the contact point, or mechanical stress applied to the surface because of the weight of the rubble? Furthermore, would fill in contact with a failed (i.e., with a hole in it) drip shield or waste package divert water away, or concentrate flow into openings?

Volcanic Ash Redistribution

A brief meeting was also held among Richard Codell, Jack Gwo and Donald Hooper. The objective of the meeting was to determine the path forward for the improvement of the volcanic ash redistribution model in TPA. Concerns regarding the downwind deposition of ash particles larger than current model predictions were raised. The staff suggested that model comparison may be undertaken to identify the knowledge gaps and the possible physical mechanisms that may explain the re-distribution of large ash particles.

TSPA-LA Review, Seepage Geochemistry and Programmatic Concerns

A meeting was held among Jack Gwo, Olufemi Osidele, Osvaldo Pensado, and Jim Winterle regarding the review activities of TSPA-LA. In the off-session meeting between Jack Gwo and Olufemi Osidele, various problems were identified regarding the operating system dependence of TSPA-FEIS runs, the model modifications from TSPA-FEIS to TSPA-LA, and the contrast in the contribution of matrix diffusion to the saturated and unsaturated zone radionuclide transport as suggested by TSPA and TPA calculations. The causes of the contrast, as pointed out by Olufemi, is likely a result of the removal of geologic layers to improve run times in NEFTRAN and may point to possible re-examination of the resulting conservatism in TPA.

Several meetings between Jack Gwo and Alexander Sun were held to discuss planned modeling activities in the unsaturated zone for FY06. Alexander Sun mentioned in particular calculations of drip water chemistry that are to be obtained from the modeling exercise so that corrosion rate calculations of waste package may be better based. Jack Gwo suggested that the planned modeling activity should be coordinated with the drift degradation hydrological experiments to better constrain the model calculations.

A meeting among Richard Codell, Jack Gwo, and Sitakanta Mohanty was held to discuss some programmatic issues, including delivery schedules for the upgrade to the TPA colloid and Mechfail submodels.

Repository Scale-Up Issues

Richard Codell also discussed with Sitakanta Mohanty the upscaling of waste packages in the repository. For the sake of computational expediency, the TPA and TSPA models simulate the behavior of many thousands of waste packages by just a few; e.g., one representative waste package for each of the 7 to 10 subareas in the TPA case for each failure time period specified. This coarse representation of the waste package may lead to errors in the releases of radionuclides from the engineered barrier system because it may not properly account for variations among waste packages (e.g., times of failure, water flow to the waste package, contact of water with the waste form, temperature). We discussed Richard Codell's progress to date using a simplified version of the TPA release model to simulate the failure of thousands of waste packages and compare the results to the original model.

Modification of TPA to Replace NEFTRAN Code

We discussed with Scott Painter a possible replacement for NEFTRAN that would not require the cumbersome work-arounds currently in the TPA code (e.g., skipping layers if the travel time through them is too short in order to save computational time, creation of composite flow paths, long run times when matrix diffusion is implemented, large computer storage capacity needed for "catcher blocks"). The method uses direct simulation of radionuclides by particles, each traversing a layer in one time step. Longitudinal dispersion is accounted for by adding a random component of velocity to the particles. Radioactive chain decay of the particle is simulated directly by a Poisson process. When a particle changes to the next member of the chain, the particle assumes new properties such as half life and retardation. Matrix diffusion is also simulated directly by diffusion of particles from the liquid phase into a semi-infinite solid representing the matrix of the rock. The procedure has been in use for a number of years, but Scott is perfecting it using techniques found to be effective in Monte Carlo treatment of radiation and neutron transport. We also discussed former modeling work by R. Codell on adapting numerical inversion of Laplace transform methods to solving the transport equations for the TPA code.

New sensitivity approach

We discussed a new parameter sensitivity approach put forward by Osvaldo Pensado. This approach is capable of picking up sensitivity of parameters that do not exhibit a monotonic response for the dependent variable; e.g., the parametric importance might be greatest in the middle of the parameter range. His method maps the independent variable to a Gaussian coordinate system and then squares it. This in effect changes the response to mostly monotonic, allowing it to be detected by linear regression, which is the basis of many of the sensitivity methods. It seems like a good addition to the sensitivity analyses we have performed on TPA results.

CONCLUSIONS

We spent a highly productive week at the Center covering a wide variety of topics dealing with performance assessment of the potential YM repository. As a result of our meetings, we expect the Center to be engaged in literature reviews and laboratory experiments that deal with the importance of colloids and the hydrological and thermal effects of backfill in the drifts, either from drift collapse or by the intentional use of engineered backfill.