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CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Workshops on Alternative Models and Interpretations Waste Form Degradation and Radionuclide Mobilization Expert Elicitation Project (20-1402-561)

DATE/PLACE: November 18–19, 1997
Millbrae, CA
December 15–16, 1997
South San Francisco, CA
January 27–28, 1998
South San Francisco, CA

AUTHORS: William M. Murphy
Tae Ahn
Richard B. Codell
Sitakanta Mohanty

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PERSONS PRESENT:

In addition to the authors of this report and the speakers noted in the report, the meetings were attended by 10's of others mostly representing the Department of Energy (DOE) Yucca Mountain Project (YMP). The expert panel was made up of Michael Apted (QuantiSci), John Bates (Argonne National Laboratory), Lawrence Johnson and David Shoesmith (AECL), and William Bourcier and Henry Shaw Lawrence Livermore National Laboratory (LLNL). The elicitation was conducted by Kevin Coppersmith and Roseanne Perman (Geomatrix Consultants).

BACKGROUND AND PURPOSE OF TRIP:

The DOE has been conducting a series of expert elicitations as part of the YMP to provide constraints for understanding a number of key issues related to the performance of the proposed high-level radioactive waste (HLW) repository. This series of three meetings on waste form alteration and radionuclide mobilization was intended to cover sequentially (i) introduction of experts to the project, summary of significant issues, and summary of available data; (ii) review of issues and uncertainties, consideration of alternative models, and identification of additional data requirements; and (iii) elicitation of preliminary conclusions from the experts and training in the elicitation process. Following the meetings the experts were to be interviewed individually, and a final report will be prepared summarizing the elicitation.

WORKSHOP ACTIVITIES: PRESENTATION SUMMARIES

WORKSHOP 1

Kevin Coppersmith (Geomatrix) provided background on the elicitation process, and noted in response to a question that the process is conducted in accord with NUREG-1563. Results are to be useful in Total System Performance Assessment-Viability Assessment (TSPA-VA). The experts are to identify issues and evaluate data.

David Stahl (M&O-FCF) presented the reference repository design and waste inventory. The site, drifts, emplacement example, engineered barrier system (EBS), and waste inventory and burnup were described briefly. Viability assessment design, license application design, and procurement and construction design are to be developed. Concrete inverts, presently considered in the EBS design, may be replaced with two carbon steel piers filled in between with nuclide- absorbing material. It was also pointed out that the void space inside a waste package (WP) is approximately 40–50 percent for spend fuel (SF) waste and 33 percent for glass waste and the corrosion products will not completely fill the void space.

Robert Andrews (M&O-TRW) addressed the role of the elicitation in TSPA-VA. Relevant parameters in the performance assessment (PA) relate to cladding and waste form degradation. Particular aspects of TSPA-VA that reach beyond TSPA-1995 include cladding degradation rate, geochemical dependence of waste form degradation, solubility limits as a function of aqueous chemistry, and possibly the role of waste form alteration products. Information is required on timing of pinhole breach of cladding, rate of cladding unzipping, waste form surface area in contact with water (i.e., wet portions of waste form and reacting surface area), waste form alteration rate, form of release of radionuclides, solubilities, and colloid concentrations. The experts are to evaluate and assess knowledge available to provide this information and address uncertainties. More detailed elicitation on solubilities will be made in separate meetings.

Bill Halsey (LLNL) described a general conceptual model for degradation and mobilization: oxidation and dissolution of SF, diffusion limiting gel formation hydration, and dissolution of HLW glass. Priority issues identified are dissolution rate, solution and alteration product evolution, evolution of the near-field environment, exposed surface area of SF, and cladding degradation. Apted pointed out that for gap fraction release, less reliance on a bounding model is appropriate because peak dose could come from 2 percent gap fraction rather than 98 percent matrix dissolution.

Christine Stockman [Sandia National Laboratory,(SNL)] presented a fairly comprehensive program to characterize the near-field environment including fluid fluxes, temperature dependencies of phase stabilities and kinetics, variations in gas and water chemistry, colloid generation, microbial effects, and intradrift variability of materials evolution, but no results were presented. It was noted that the RIP code accommodates complex chemistry but avoids transient modeling by using a batch reactor cell concept. Regarding the question on oxygen availability, her current results show that oxygen will be abundant in failed containers.

Ray Stout (LLNL) described primary spent-fuel inventory and classifications, temperature, and relative humidity predictions. SF responses were described as degradation, oxidation, dissolution, alteration, and release. Some theoretical relationships that may be used to describe these processes were presented, but none seem to have been applied or to be amenable to PA implementation.

Walt Gray [Pacific Northwest Laboratory, (PNL)] also presented results from a suite of flow through SF dissolution tests conducted at a range of T, pH, carbonate, and pO_2. An empirical rate equation was derived as a function of CO_3, pH, and T. Measurements of gap and grain boundary inventories were presented. Gray pointed out that his experiments were not intended to represent repository conditions but were meant only for studying reaction kinetics of reactor fuel. Johnson commented that the Swedish PA used 10 percent gap fraction though experimental data showed only 1 percent. A report is currently under preparation that shows the Swedish approach to scaling the laboratory scale gap-fraction release to the repository scale.

Steven Steward (LLNL) advocated a statistical test array to determine dependencies of SF dissolution rate on environmental parameters. Data analysis leads to an equation of the form used by Gray with an additional T, pO_2 cross term, which Steward attributed possibly to radiolysis.

John Bates (ANL) described drip tests on SF with a "high" drip rate (which is nevertheless low, i.e., 0.75 ml/3.5 days). Through-grain and grain boundary reaction modes were observed. Secondary uranyl phases were identified by XRD and incorporate Cs, Ba, Ru, Mo, and Np. Pu appears to concentrate on SF surfaces. Bates et al. have presented their data in the literature with units that do not require knowledge of SF surface area.

Bob Finch (ANL) presented analytical data on reacted SF. Na-boltwoodite, uranophane, schoepite, and a Cs-Mo uranate were identified. Grain boundaries of SF are attacked, a replacement layer occurs exterior to unaltered grains and is enclosed in a hydrous layer of precipitates. Schoepite containing Np forms in vapor tests. Secondary uranyl silicates can contain other radionuclides. The phases containing Tc and Mo were dissolved preferentially. The secondary minerals have about 20 percent porosity and, therefore, they are unlikely to be diffusion barriers.

Ananda Wijesinghe (LLNL) presented an attempt to interpret the kinetics of the ANL SF tests under no-drip, low-drip, and high-drip rates. A rate equation was derived based on a transport-limited process and calibrated with the data. It provides a reasonably good fit for long times and no-drip and low-drip rates, but not for high-drip rates. Film concentrations are orders of magnitude smaller than radionuclide solubilities assumed in TSPA-95. The current results indicate transport-limited release of radionuclides under no-drip and low-drip rates. In NRC TPA3 exercises, these models need to be considered for drip case (not immersion). Nonetheless, it has been discussed if no-drip or low-drip rates are relevant to the Yucca Mountain (YM) repository.

Eric Siegmann (M&O-DE&S) offered information on cladding characteristics and corrosion and strain studies. In general zircalloy is highly resistant to corrosion. A cladding model is to be implemented in TSPA-VA involving creep failure, surface oxidation, and splitting due to uranium oxidation. Cladding protection is expected to reduce SF exposed to the WP environment. Since this meeting, DOE has dropped cladding protection in the base case of TSPA-VA.

Bob Einziger (PNL) presented data on SF oxidation in air with variable water content. Oxidation to U_4O_9 is rapid on a repository time scale. After formation of U_4O_9 there is a period of no further oxidation. Then there is renewed oxidation to U_3O_8 which involves a volume increase, powdering of SF, and splitting of cladding.

Cynthia Palmer (LLNL) discussed thermodynamic data needs. Inventory considerations indicate that data will be required for Am, Pu, Np, U, Tc, Zr, and Ni with high priority. Aqueous solution species thermodynamics are fairly well known except for phosphates and silicates. Solid characterization and

solubilities are incomplete. Data at elevated temperatures are limited. NEA review data (for U, Am, Np, Pu, and Tc) will be considered qualified by the YMP. Data base reviews including organics are being undertaken. Interim results show that Zr and Ni could be promptly precipitated.

John Bates (ANL) described colloid characterization in glass and SF dissolution tests. For the glass tests most Am and Pu releases occur as colloids. A colloidal hydrous phosphate mineral, brockite, contains Pu and Am. Fe silicate and smectite colloids were also observed.

Bill Bourcier (LLNL) summarized HLW glass dissolution processes, data, and rate equations. For TSPA-VA glass alteration depends on T, pH, and silica concentration and the rate equation is calibrated empirically. Incorporation of released radionuclides in secondary phases is neglected. Bourcier considered the area for the dissolution of glass waste to be 25 times that of the geometric surface area. The way the leaching rate is parameterized in Bourcier's work, the long-term dissolution rate would stabilize at a value equivalent to the rate of formation of secondary phase. However, this is compensated by decreasing surface area as a function of time.

John Bates (ANL) discussed glass testing in vapor hydration tests and unsaturated (drip) tests. In some drip tests all actinide release was colloidal, in others actinides were dissolved, (whereas about half of Pu releases from SFs are colloids). He said that Pu releases from SF and HLW glass are of approximately equal amounts, when normalized over inventory. Release rates can accelerate abruptly after hydrated secondary phases begin to precipitate in long term tests. Also, releases can be abrupt when hydrated phases dissolve.

Lee Bendixsen (Idaho National Engineering Laboratory,INEL) summarized the dozens of different types of DOE-EM SF including N-Reactor SF (uranium metal), INEEL SF (uranium oxide, metal, carbide, thorium oxide, etc.), high U-235 enriched oxide and mixed oxide, melted Three Mile Island SF, TRIGA (U-Zr hydride), U-Al alloy, etc.

Walt Gray (PNL) described preliminary dissolution studies on Hanford N-Reactor SF as a function of T, carbonate, and pH. Hanford SFs were found to dissolve much faster than LWR SF. However, when dissolution rates are normalized over exposed surface area, the two rates are comparable. Further, if the inventory is considered, the contribution of Hanford N-Reactor SF in PA exercises would be minimal. Johnson showed concern about the high rate at which the materials would come out of the WP (i.e., ~100 yr). Gray agreed, but indicated that the experiments were conducted at a much higher rate than one would anticipate in the repository conditions.

WORKSHOP 2

Bill Halsey (LLNL) outlined options for the spent-fuel source term for TSPA-VA. Walt Gray's empirical rate model will be expanded to include dependence on oxygen concentration and burnup. Temperature, pH, carbonate, and some cross terms including radiolysis and effective surface area will be considered. Observed concentrations in long-term tests are notably below solubilities used in TSPA-95 for many radioelements. It is proposed to sample solubility values between revised solubilities and observed aqueous film concentrations. In some of the commentary on this presentation, it was noted that there are only a few experiments on the effect of silicates on solubility, and no appropriate conceptual model. Water chemistry in contact with the SF is expected to be dominated by the effects of the SF itself and secondary uranium phases. L. Johnson (AECL) commented that solubility values may be distorted in some of the experiments because of the gamma radiation in relatively fresh SF. He expects most of this radiation to diminish to insignificance in hundreds of years. Halsey (LLNL) commented that it cannot be demonstrated that the rate

will drop, so the TSPA models will have to assume conservatively high values. It seems to us that the abundant oxygen masks the effect of gamma radiation in the YM repository, unlike the anoxic Canadian repository.

Dave Sassani (M & O, DE&S) observed that Np concentrations in dissolution tests are consistently below, by orders of magnitude, solubilities determined in supersaturation studies which form the basis for solubilities in TSPA-95. He suggests that the high-solubility phases of Np are metastable, and actual solubilities should be lower. However, no definitive values were provided for use in TSPA-VA. In some batch data with SF, U and Np concentrations reach a steady state, while others like Cs and Tc keep increasing, suggesting that secondary U phases are limiting the Np releases.

J. McNeish (M & O) presented an overview of water ingress to the WP. The bathtub scenario was discussed, but it was not clear that DOE would adopt it. McNeish also mentioned a case in which the exposed SF was in contact with corrosion products and rock rubble. There will be three periods of infiltration: present, long-term, and superpluvial. DOE does not appear to be contemplating episodic flow or explicit hydrothermal flow in their VA models, and the extent of failure does not increase with time. The drift seepage models indicate there would be no seepage during heating. It is assumed that corrosion holes form in patches that can move with time, and water entering the patches comes into contact with the SF. The fraction of WPs wetted is a function of infiltration, but is determined from an abstraction of a 3-D transient model of flow in a fractured porous medium from Lawrence Berkeley National Laboratory (LBNL). This model alternatively considers advective transport from the waste form by flowing water, or if patches are not on top, diffusion is the only operative mechanism for radionuclide release. One interesting distinction with the NRC TPA3 models is that DOE can have multiple failure modes for canisters in each subarea; e.g., diffusion releases and advective releases simultaneously. NRC models assume all variability must be captured in a single WP representative of each subarea.

Dave Sassani (M&O-DE&S) outlined a strategy to characterize the near-field chemical environment accounting for thermohydrologic evolution, introduced materials, and gas phase composition variations. 100,000 yr of repository future, was subdivided in 6 periods, and abstracted T, gas compositions, and water chemistries provided for each, based on EQ3 modeling of gas-water-rock interactions. These incoming waters were then modeled to react with concrete components. The modeling studies presented indicate oxygen depletion initially, but return to an oxygenated environment within 4 to 5 thousand yrs.

D. Franklin (Bettis) described zircalloy corrosion. Repository applications are a long extrapolation in time and temperature from laboratory data. In reactors a protective oxide film forms and grows by oxygen diffusion, and eventually the layer breaks down resulting in cyclic kinetics. Irradiation leads to dissolution and amorphization. Autoclave testing data at temperatures near 300 °C extend for 30 yr showing maximum oxide thickness of 4.5 mils. An empirical rate equation was provided which is only a function of T. The best estimate for oxide thickness in the repository at 1,000,000 yr is about 1.5 mils. Tests are planned to look at high Cl and Fe concentrations. Tae Ahn (NRC) pointed to some data suggesting that very low pH would be necessary for pitting, even at high concentrations. Gamma radiolysis of thin films could produce acidic conditions, but it is not known if such conditions could exist in the WPs in the time frame of interest.

Eric Siegmann (M&O-DE&S) reiterated a point on zircalloy made in the previous meeting and described cladding failure modes in TSPA-VA including perforations, unzipping, and mechanical breaking. The study concludes that limiting cladding temperature to 350 °C limits strain perforations. Also, delaying WP failures to after 200 yrs eliminates cladding unzipping by UO_2 oxidation to U_3O_8. Cladding failure scenarios included

the fall of rubble blocks on assemblies after most of the WP has already failed by corrosion. Siegmann predicts only about 0.5 percent of SF would be exposed in this way.

Lawrence Johnson (AECL) summarized the estimation of the instant release fraction for the SR-97 (Canadian) PA study. He noted that the data from dripping experiments on SF by Bates at Argonne did not indicate protection by secondary phases of uranium, but batch immersion data indicate that dissolution is controlled by schoepite solubility in some cases. He sent Tae Ahn (NRC) SKB Technical Report (97-18) which contains Canadian test results of instant release from BWR SF. The report was prepared for Sweden and discussed instant release in PWR and CANDU SFs.

Yueting Chen (M&O-DE&S) presented results of reactive transport modeling inside a WP using AREST-CT. UO_2 is modeled to dissolve kinetically and four secondary uranyl minerals precipitate. Schoepite precipitation is abundant, uranophane precipitates at the top and subsequently dissolves. Porosity decreases from 65 percent to 40 percent. Results compare remarkably well with laboratory data from ANL. In a preliminary study Chen observed that in localized reducing environments TcO_2 may precipitate and noted that TcO_4- is reported to inhibit WP corrosion. He predicted that SF will be totally dissolved after 10,000 yrs, using the geometry of SF pellets.

David Shoesmith (AECL) offered an authoritative review of SF dissolution mechanisms from an electrochemical perspective. He contends that secondary uranyl phases will insulate the SF surface and form a transport barrier, but they could localize radiolysis effects. Alpha radiolysis might be effective in this gap, but only with a strong source. Alpha radiolysis is insignificant in the Canadian HLW program, but contentious. He is of the opinion that gamma and beta radiation would be unimportant in general after a few thousand years at most. Metallic phases generated in the reactors catalyze the reduction of O_2 and oxidation of UO_2. Carbonate in the water neutralizes acidity and reduces the rate of reaction. He volunteered a rate of SF oxidation after 1,000 yr for a Canadian repository of 10^{-6} to 10^{-8} mol/m^2 yr. This rate is very low compared with DOE data. We should remember that the YM repository is oxidative.

John Tait (AECL) described flow through dissolution tests on the rate of UO_2 Dissolution. The rate depends on oxygen concentration, but the order of reaction is higher than that for SF. The rate also depends on carbonate. The rate depends on gamma field, but the effect is negligible for rates less than 10^2 R/hr or 10^3 R/hr in carbonate solutions. In some of the tests, radiation was from an external beta source using unirradiated UO_2 fuel rather than SF. The rate depends on T, but dependence differs between carbonate-free and carbonate waters. Tests on CANDU used fuel indicate release of Cs and Sr from grain boundaries. Pu and U releases are similar in oxidizing, carbonate solutions. SF dissolution is faster than UO_2 dissolution under deaerated conditions perhaps due to radiolysis effects. Silica and Ca strongly suppress the dissolution rate; this was true for UO_2 and SF. Previously, DOE demonstrated this effect of silica and Ca only for UO_2.

Tae Ahn (NRC) presented considerations related to waste form source-term modeling. Because currently measured dissolution rates of SF are high in PA exercises, potential mechanisms limiting radionuclide mobilization may be significant. A brief summary was made of cladding protection, release from failed cladding/container, and protection by secondary minerals.

Richard Codell (NRC) presented a synopsis of the models for SF wetting and radionuclide release used in the NRC TPA3 code. The presentation covered isothermal water influx to the drift, water ingress and egress from WPs, SF wetting, and radionuclide transport from the WP. Some preliminary results from the code were presented, but without absolute values of dose, highlighting alternative models being considered such as the bathtub, flow-through, pulled plug, and diffusion-only models.

David Wronkiewicz (University of Missouri) presented results of drip tests on UO_2 for conditions like YM. Petrographic observations indicate formation of numerous uranyl minerals palygorskite, Fe- and Ti oxides and amorphous silica. Similar paragenesis is observed at the Nopal I uranium deposit, which is a natural analog. Na-boltwoodite forms as an alteration product on similar tests with SF. Vapor hydration tests on simulated waste glass indicate that alteration is accelerated in a radiation field. Vapor hydration leads to a paragenesis of smectites, zeolites, and feldspars, which is similar to the paragenesis of volcanic glass alteration. He suggested that radionuclide migration may be retarded by incorporation into alteration phases. Examples include Np in dehydrated schoepite, and Cs, Mo, Ba, and rare earth elements in uranyl oxide hydrate.

William Murphy (CNWRA) discussed source term related studies at the Nopal I natural analog site. An overview was presented of geography, geology, petrography of uraninite alteration, mineralogy, uraninite chemistry, paragenesis, and uranium decay series data. Retrograde solubility of uranyl minerals was illustrated, and the possibility was noted of a change in mechanism from dissolution control to transport control with decreasing temperature. The concept of an alternate release rate sensitivity analysis was mentioned, but no results presented.

WORKSHOP 3

Kevin Coppersmith (Geomatrix) announced that the purpose of the workshop was to permit the experts to express their preliminary ideas and to receive feedback primarily from the other experts.

Lawrence Johnson (AECL) commented on cladding concluding that creep rupture is improbable, stress corrosion cracking is unlikely, delayed hydride cracking is unlikely, general corrosion is inconceivable, no data exist on localized corrosion, unzipping is unlikely at low T due to oxidation to U_3O_8, hydrogen embrittlement is possible but no data are available, and swelling due to oxidation to schoepite is possible. He believes stress corrosion cracking will be negligible and general corrosion of cladding will be very slow at temperatures less than 250 °C.

David Shoesmith (AECL) noted that zircalloy should pit if conditions are sufficiently saline. Hydrogen embrittlement was concluded to be the most likely failure mechanism over a time period of 10^3 to 10^4 yr. He presented data on Ti corrosion implying that Zr corrosion is analogous. Possible sites of pitting corrosion in cladding might be (i) intermittent drip sites experiencing wetting/drying cycles, and (ii) occluded sites where stagnant water can be present for long periods. Once initiated, pitting could penetrate quickly through the thin cladding.

It was noted that much pitting data comes from situations far removed from canister or pit corrosion; e.g., the Alaska pipeline. In that case, most deep pits occurred because of a protective layer used to coat the pipes that allowed occluded space for water accumulation between the layer and the steel. Pits in passivating materials such as Zr and Ti would be narrow.

In the discussion on cladding Tae Ahn (NRC) noted that localized corrosion data are available from Bettis and AECL (Chalk River). Pitting of zircalloy would occur under conditions of high salinity with oxidizers such as ferric ions that are stable at low pH (e.g., less than 4.5). Apted questioned the relevance of the Ti data to Zr. Johnson and Shoesmith asserted that appropriate tests could be conducted. Johnson expressed concern over uncertainty in the chemical environment. Shoesmith noted that sulfate always exceeds chloride (which is good for cladding stability poren), but this observation contradicts the models presented by Sassani.

There was discussion on cladding failure by swelling. This might occur at elevated temperatures by formation of U_3O_8, but not at lower temperatures. Ray Stout (LLNL) discussed some long- term experiments with clad SF in oxidizing steam that indicated no splitting. Corrosion products clog holes in the cladding. Lawrence Johnson (AECL) also indicated that sealing of clad failures was a distinct possibility.

Mick Apted (QuantiSci) was asked to address waste form surface area exposure. He posed a series of general questions suggesting that the problem is difficult to address well. Noting that uncertainties provoke broad ranges in sampled parameter values in PA, the consequence may be to spread releases and diminish peak doses because of uncertainty rather than real variability.

Lawrence Johnson (AECL) noted that the oxidation rate of SF is limited only if aqueous diffusion of oxygen is the rate limiting process. Nevertheless, U oxidation is fast relative to release if limited by diffusion. Small cladding defects can lead to large released from the gap.

Henry Shaw (LLNL) noted that surface area is irrelevant to solubility limited species, and concluded that even vapor phase hydration wets the entire surface. The question of the appropriate surface area was raised, e.g., fragments, grains, role of alteration coatings, without conclusion.

In the discussion on wetted surface area it was generally concluded to assume that if the cladding is breached, then all surface area becomes wetted, but this does not necessarily relate to releases from the SF. An LBNL study showing very fast transport in thin water films was noted. This study however pertained to water films being supplied by a liquid source, not films that form from atmospheric condensation on surfaces. Walter Gray (PNL) noted that he has data on effective surface area for release from SF that showed only 15 times the surface area for crushed spent-fuel samples as compared to whole SF pellets. This indicates roughly that the surface area of SF particles (several millimeters diameter) rather than SF grains (~10 microns diameter) should be used in the models. Some questioned whether it was important to know the exposed surface area of the SF, because many of the radionuclides would be limited by solubility in the thin water film next to the SF rather than release rate.

David Shoesmith (AECL) reiterated information on the intrinsic dissolution rate of SF concluding that the process is fairly well understood.

Henry Shaw (LLNL) noted that the empirical release rate equation used in TSPA reasonably reproduces laboratory data, particularly due to large uncertainties in surface areas. Nevertheless, he stated many reservations regarding further validation, limited range of conditions applicable, few data for high burnup SF, and limited dependence on chemistry.

Bill Bourcier (LLNL) summarized some information presented previously on secondary phases formed in laboratory tests. He noted that as secondary phases recrystallize to more stable crystalline phases, coprecipitated radionuclides may be released.

John Bates (ANL) noted that secondary phases are specific to individual glass waste forms. Much information is lacking with respect to secondary phases associated with glass including actinide distributions between solution and solids, a link between short term tests and long term behavior, and effects of interactions with EBS materials. Many radionuclides are retained in secondary phases formed by SF dissolution. However, data at elevated temperature are limited. Thermodynamic models at 25 °C are reasonably consistent with experimental results. He noted in vapor testings that Tc in grain boundaries could be soluble and washed out readily.

John Bates (ANL) confirmed that colloids exist and have been well characterized particularly in glass alteration studies. However, transport of colloids is speculative. For instance, little is known on colloid transport through pits. He said his experimental data on glass waste form show 100 percent release of Am and Pu is on glass-produced clay colloids.

Bill Bourcier (LLNL) noted that Fe oxide colloids have a pzc of 6 to 8 so changing geochemical conditions could affect their sorption characteristics.

Christine Stockman (SNL) announced that waste form colloids would be included in TSPA-VA and treated like equilibrium species with a diffusivity 1,000 times less than ionic diffusivity, and with stability a function of pH and ionic strength. She also said that solubility limits would apply either the EBS or the geosphere.

The last session concluded with introductory training on probability assessment conducted by Peter Morris of Applied Decision Analysis, Inc.

OBSERVATIONS

Given the diversity of defense wastes, high enrichment, chemical instability in oxidizing conditions, etc., DOE SFs may significantly affect repository performance, and little attention seems to be given to them in PAs. However their inventories and burnups are small compared to power reactor SF. Offline analyses are required to confirm that the net contribution of DOE SFs is not significant. In this regard, the HLW glass contribution can also be treated in the NRC TPA3 code without much complication.

Aspects of the following scenario were described. After breach of the container and cladding, SF oxidation is rapid relative to removal of waste from the disposal horizon. After oxidation occurs, a change in climatic conditions could lead to a flushing of the system. Np and Tc in particular may be available for immediate transport in large quantities. This scenario seems plausible and could lead to spikes in radionuclide concentrations which could correspond to periods of peak dose. There was also some evidence that Np would be captured in schoepite produced by humid air oxidation, and that this would inhibit its release when wetted.

The artificial smearing out of releases because of large parameter sampling ranges due to uncertainty rather than variability could lead to underestimation of peak doses.

The surface area of reacting SF is still under debate. For instance, wetting in humid environments is said to be able to transport radionuclides but it is questionable whether the environment of humid air testing is relevant to the YM repository. Also, details about grain boundary attack are not well understood.

The assessment of cladding protection has uncertainties associated with localized corrosion and hydrogen embrittlement. However, the panel does not seem to have all literature available. The protection by failed cladding also needs more attention.

A more effective method to incorporate the evolution of near-field environment in waste-form degradation is needed. The existing parametric rate equations could be used and extended in a relatively simple manner.

The use of AREST-CT code could be extended to assess the role of secondary minerals in the dissolution of the primary phase.

The HLW glass is hydrated in humid environments. Later, this hydrated phase can be washed out with water contact. We do not know how much radionuclide would be released as a pulse during this washing period.

In a number of cases the experts were being asked to evaluate their own work.

The expert panel consisted of highly qualified individuals, and their recommendations should have an import impact on the DOE program.

A great deal of uncertainty remains in the issue of waste form degradation and radionuclide mobilization.

PROBLEMS ENCOUNTERED:

None

PENDING ACTIONS:

None

RECOMMENDATIONS:

CNWRA and NRC staff should examine the final report of the elicitation.

SIGNATURES:

6/18/98

William M. Murphy
Principal Scientist

Date

6/18/98

Sitakanta Mohanty
Senior Research Scientist

Date

CONCURRENCE:

6/18/98

English C. Pearcy, Manager
Geohydrology/Geochemistry, Element

Date

6/18/98

Budhi Sagar
Technical Director

Date