Appendix F

The format of this appendix and the criteria it contains follow that of earlier versions of the Radionuclide Transport Issue Resolution Status Report. Progress since the last revision of the RT IRSR is described here relative to the criteria of Revision 1.

5.0 STATUS OF ISSUE RESOLUTION AT THE STAFF LEVEL

The status of resolution of subissues for the RT KTI is summarized in table 5.1.

Table 5.1. Summary of status of subissue resolution

Summary of Subissues	Status of Resolution at the Staff Level
• Radionuclide transport through porous rock	Several methodologies for acquiring retardation coefficients for important nuclides are acceptable. Approach to retardation and transport is acceptable. Additional data required for important nuclides such as U and Pu. Status: **Open**.
• Radionuclide transport through fractured rock	Demonstration of capability to predict breakthrough curves of reactive, nonreactive, and colloidlal material needed. Justification of length of flow path affected by fracture dominated transport needed. No sorption credit within fractures in current PA models. Status: **Open**.
• Radionuclide transport through alluvium	Significant data gaps exist with respect to important transport characteristics of the alluvium. The potential for preferential pathways is insufficiently studied, and confirmation that the alluvium behaves as a homogeneous porous medium is not well supported. Several methodologies for acquiring retardation coefficients for important nuclides are acceptable. Resolution likely to be based on field testing and characterization. Status: **Open**.
• Nuclear criticality in the far field	Status: **Closed pending confirmation** of DOE approach as described in topical report.

Since Revision 1 of the RT IRSR, new information to be considered by the NRC staff has been provided in the letter from Brocoum to Reamer, March 22, 2000 describing DOE's Review of the RT IRSR Rev 1, and the NRC/DOE Technical Exchange on Yucca Mountain Pre-Licensing Issues on April 26, 2000. The production of this revision of the IRSR predates the availability of the official copies of the two PMR's dealing with transport, limiting progress in this time period toward resolution.

5.1 STATUS OF SUBISSUE 1—RADIONUCLIDE TRANSPORT THROUGH POROUS ROCK

5.1.1 Summary of the Status of Subissue 1

Most of the YM geochemical work in the past twenty years has been directed toward determining the retardation of radionuclides in porous rock. Significant progress has been made to address this subissue that is important to waste isolation and repository performance. However, in that time, there have been major changes in the conceptualization of the geologic setting of the repository that impact the relative importance of this subissue on performance. The major changes include the recognition that average infiltration is one or two orders of magnitude greater than original estimates, and the consideration of the point of compliance up to 20 km away from the repository. The greater average infiltration results in a greater proportion of the flux bypassing the sorptive porous rock by flow in fractures. A 20 km point of compliance would result in the need to consider the alluvium along with porous and fractured rock. These major changes reduce the relative importance of radionuclide transport in porous rock on PA.

The NRC staff finds that the approach adopted by DOE (Triay, et al., 1992) to validate K_d values from batch sorption tests is logical and defensible. By performing batch sorption tests using site-specific materials, followed by confirmatory tests to establish the validity of the assumptions needed for the constant K_d approach, and then selecting the minimum K_d from all the tests, an acceptable value can be obtained.

Overall, the NRC staff considers that the subissue can be closed for certain radionuclides but not for others. Some of the radionuclides for which the subissue has not been resolved on the staff level may be important to performance. Three radionuclides are chosen as examples to highlight successes and areas needing further work. They are neptunium, plutonium, and uranium. The staff recognizes that multiple tests have been performed to establish reasonable K_d values for neptunium. However, initial neptunium concentrations in some batch sorption tests ranged from 1×10^{-7} to 3×10^{-5}. Efurd et al., 1997 found that neptunium solubility approached from undersaturation was only $(1.5 \pm 0.6) \times 10^{-5}$ at pH = 8.5 and 25°C. Consequently, sorption experiments carried out at this condition may have precipitated a neptunium phase. Also, although both batch sorption tests and flow-through column tests have been performed to determine a minimum K_d for plutonium, significant inconsistencies have been observed. The NRC staff recognizes plutonium as problematic and encourages further work to establish defensible K_d values. For uranium, geochemical modeling suggests that a uranyl silicate phase, soddyite, could precipitate from solution, given the initial groundwater composition. Eliminating the possibility that processes other than sorption may be contributing to the removal of a radionuclide from solution is necessary for establishing a valid K_d. On the other hand, the thermodynamic modeling could be in error based on parameter uncertainties.

To date, it does not appear that flow-through column tests were performed with uranium. Consequently, this subissue has not been resolved at the staff level.

5.1.2 Criterion-By-Criterion Analysis

Table 5-2, along the left side, lists radioelements that have been considered in previous performance assessments of the proposed YM HLW repository as well as radioelements that have been used in sorption experiments that are chemical analogues for those radionuclides that could affect performance but are difficult to handle in laboratory experiments. Shaded rows indicate those radioelements with isotopes (^{14}C, ^{99}Tc, ^{129}I, ^{79}Se, ^{231}Pa, ^{234}U, ^{237}Np, ^{239}Pu, and ^{242}Pu) considered in DOE TPSA-VA (U.S. Department of Energy, 1998c). Across the top of the table are possible methodologies that may be used to address this AC. Check marks indicate the methodologies have been used. For example, a check mark in column m2 indicates batch sorption experiments were performed using site-specific materials. It is not intended to indicate that all sets of solid/groundwater conditions expected for the YM system have been tested. However, one should not expect that all boxes will have to be checked for issue resolution. Some boxes are mutually exclusive. For example, by assuming K_d is zero (provided that assumption is conservative), as in the case for C, I, and Cl, the AC is satisfied for those radionuclides.

The key to table 5-2 is as follows:

AC component: Estimated parameters based on techniques that may include laboratory experiments, field measurements, natural analog research, and process-level modeling studies conducted under conditions relevant to the UZ at YM;

Methodology	Description
1a	Radioelement has been included in PA
1b	K_d is assumed to be zero
m1	K_d determined experimentally in batch sorption tests
m2	Used site-specific materials in batch sorption tests
m3	Performed crushed tuff flow-through column experiment
m4	Performed intact rock flow-through column experiment
m5	Performed diffusion experiment
m6	Performed unsaturated flow experiment
m7	Used process model to determine constant K_d

AC component: Demonstrated that the assumptions implicit in a K_d (or K_A) approach to radionuclide transport in the UZ are valid if process-level models are to yield a constant retardation parameter

linear	—	Demonstrated linear isotherm by determining K_d as a function of radionuclide concentration
fast	—	Demonstrated fast ion exchange or adsorption reaction

AC component: Demonstrated use of appropriate parameters for process models.

Only those components and methodologies that are specific to a given radionuclide are listed in table 5-2. Consequently, some assumptions implicit in the K_d approach, such as demonstration that the medium acts as a single continuum porous medium are not included.

Only those acceptance criteria that are specific to a given radionuclide are listed in table 5-1. Consequently, Criteria 2a and 3a (Demonstrated single continuum porous medium) are not included nor is the portion of Criteria 2c and 3c involving the demonstration of constant bulk chemistry

Table 5-2. Status of subissue resolution for selected radionuclides

Nuclide	used in PA	Kd=0	Estimated parameters based on appropriate techniques such as laboratory experiments, field measurements, natural analog research, and process-level modeling studies							Demonstrated valid Kd assumptions		Appropriate process model parameters
			m1	m2	m3	m4	m5	m6	m7	linear	fast	
Am	√		√	√			√		√			√
Pu	√		√	√	√		√		√	√	√	√
U	√		√	√			√		√			√
Th	√		√	√								
Ra	√		√	√								
Pb	√		√									
Np	√		√	√	√		√		√		√	√
Pa	√		√	√								
Sn	√		√	√								
Ni	√		√	√								
Cs	√		√	√			√		√	√		√
Sr	√		√	√			√			√		
C	√	√							√			√
Ac			√									
Sm			√									
Nb	√		√	√								
Zr			√									
I	√	√							√			√
Tc	√	√	√	√	√		√				√	
Cl	√	√										
Se	√		√	√		√		√	√			√
Ce			√	√						√		
Cm												
Eu			√	√						√		

5.1.2.1 **CRITERION 1a.** For the estimation of radionuclide transport through porous rock, DOE has Determined, Through Sensitivity Analysis, Whether Radionuclide Attenuation Processes Such as Sorption, Precipitation, Radioactive Decay, and Colloidal Filtration Are Important to Performance

The radionuclides for which the boxes are checked have been evaluated in previous PAs (see Section 3.2). This criterion is included to allow DOE to determine the importance of a given radionuclide without having to perform laboratory experiments. For example, radionuclides with short half-lives and small inventories may contribute so little to the dose that they need not be considered. The use of sensitivity analysis to eliminate from further consideration the retardation of certain radionuclides requires greater reliance on other aspects of the repository performance. It is anticipated that this approach, however, will not be used to the extreme, whereby all the reliance is placed on one aspect of repository performance to the exclusion of all others.

The staff considers that this acceptance criterion has been met for radionuclides important to performance. However, as processes, conditions, and conceptual models are revised based on new site characterization information and repository designs, it is expected that new sensitivity analyses would be needed. The only situation where this criterion would not be met would occur if DOE failed to consider a radionuclide in its PA that the NRC staff considered to be important.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "The processes of hydrodynamic dispersion, matrix diffusion, sorption (solutes), filtration (colloids), and radioactive decay are considered important to performance for the UZ and are all explicitly modeled in the UZ transport model abstraction." In response, the NRC staff considers this criterion has been met as previously described in Revision 1 of the RT IRSR. However, the list of processes DOE considers important to performance includes matrix diffusion, which, by definition, is a process that may occur in fractured, not porous rock.

5.1.2.2 **CRITERION 1b.** For the estimation of radionuclide transport through porous rock, DOE has (i) Assumed K_d Is Zero and Radionuclides Travel at the Rate of Groundwater Flow, If it Has Been Found That Radionuclide Attenuation Is Unimportant to Performance and it Can Be Demonstrated That this Assumption Is Conservative; No Other Acceptance Criteria for this Subissue Need Be Met, Or, (ii) Demonstrated That Criterion 2 or 3 Has Been Met, If Radionuclide Attenuation in Porous Rock Is Important to Performance or If an Assumption That K_d Is Zero in Porous Rock Is Not Conservative.

This criterion has been used for carbon, technetium, iodine, and chlorine, which occur predominately as anions and, as such, experience limited attraction to the negatively charged surfaces of the minerals along the flowpath. The staff agrees with the selection of C, Tc, I, and Cl as having a K_d of zero, for use in PAs. The staff recognizes, however, that there may be other elements or species that, under certain situations, have a K_d of zero.

There are examples where multiple species of a single radionuclide exist in solution at the same time. Some species can be cations and others, anions. The retardation of the species varies as a function of their charge and size. A classic example, occurred at the Cambric site where ruthenium, with a nonzero K_d, was observed at a sampling well before it was expected. It was later discovered that ruthenium is present in both cationic and anionic forms, and the negatively-charged ruthenate ion contributed to the early breakthrough.

Plutonium is another radionuclide with multiple species, including cationic as well as anionic and colloidal forms. Triay, et al. (1997) and Thompson (1989) show that some plutonium travels unretarded in crushed tuff column experiments. The plutonium and ruthenium examples demonstrate the importance of performing flow-through column tests for identifying mobile species (see status of Methodology 2b3 below).

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "K_d is assumed to be zero for C, Tc, I, and Cl." In response, the NRC staff considers this criterion has been met as previously described in Revision 1 of the RT IRSR. Caveats identified in Revision 1 remain.

5.1.2.3 **CRITERION 2a.** For the valid application of the constant K_d approach, equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Demonstrated That the Portion of the Flow Path to Which Equation (1) Applies Acts as an Single Continuum Porous Medium

This criterion is generally independent of the radionuclide migrating through the medium. However, radionuclide size and charge can exclude certain radionuclide species from the isotropic homogeneous medium in favor of the heterogeneous features. The C-Well tests provide an example that suggests a portion of the flow path is not acting as a single continuum porous medium.

Currently, the only portions of the flowpath considered to be a single continuum porous medium in the NRC's TPA are the nonwelded vitric unit of the Calico Hills and the alluvium (See Subissue 2, Radionuclide Transport through Alluvium). The reason the vitric unit is considered a porous medium is because its matrix permeability is considered high enough to accommodate the percolation rates expected for YM. On the other hand, the nonwelded zeolite unit of the Calico Hills has a matrix permeability that may accommodate only a relatively small portion of the percolation rate. Consequently, most of the water bypasses the zeolite unit in fractures. Finally, it remains to be demonstrated that the Calico Hills nonwelded vitric unit and the alluvium behave as a single continuum porous media.

Phase II of the UZ Field Transport Test at Busted Butte involve identifying possible "fast pathways" within the Calico Hills block and performing cross hole tracer tests using conservative and reactive tracers. The effect of variable flow rates on tracer breakthrough will be determined. The NRC staff considers this type of experiment important for demonstrating transport through porous rock.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "The vitric portion of the Calico Hills hydrogeologic units behaves as a porous medium and provides an effective barrier to radionuclide transport. The upper units of the Prow Pass hydrogeologic

unit also behaves as a porous medium." In response, the NRC staff notes the DOE comment does not include the term "single continuum." If this omission is intentional, and the units identified are not expected to act as single continua but as dual continua, then DOE would have to demonstrate its capability to predict breakthrough curves.

5.1.2.4 **CRITERION 2b.** For the valid application of the constant K_d approach, equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Determined Appropriate Values for Parameters, K_d, n or θ, and ρ_b

Criterion 2b has been subdivided in Table 5-1 into seven methodologies to record the multiple approaches that can be used to determine and validate appropriate values for K_d. The NRC staff considers the batch sorption experiment using site-specific materials along with the crushed tuff column experiment constitute the minimum set of experiments to demonstrate an appropriate value for K_d. Experiments involving intact tuff, diffusion, unsaturated, and fractured tuff can provide useful supplemental information. This strategy for the validation of batch sorption data is described by Triay et al., 1992.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Details of the experimental program are provided in the Yucca Mountain Site Description." The NRC instead reviewed the Summary and Synthesis Report on Radionuclide Retardation for the Yucca Mountain Site Characterization Project (Triay et al., 1997) for details of the experimental program. If the Yucca Mountain Site Description contains additional details of the experimental program that would eliminate NRC concerns, the staff requests DOE identify this supporting information. In the letter from Brocoum to Reamer, Mar. 22, 2000, on DOE's Review of the RT IRSR, there were a total of 23 comments. Only one comment pertained to Section 5 of the RT IRSR Rev 1, where the NRC staff describes the status of the DOE experimental program in light of the acceptance criteria. The NRC staff interprets the relatively small number of comments on Section 5 to mean DOE did not find fault with the NRC observations on the status of this subissue.

5.1.2.4.1 Methodology 2b1. Sorption Coefficient Has Been Determined Using a Batch Sorption Test

In Table 5-1, any radionuclide for which an experimentally determined K_d has been determined using a batch sorption test has this box checked in the table above. This includes radionuclides whose batch sorption tests have not involved site-specific materials, along with radionuclides whose batch sorption tests have. The radionuclides for which nonsite-specific batch sorption tests have been performed are lead, actinium, samarium, and zirconium. The NRC staff considers the sorption coefficients determined from nonsite-specific batch sorption experiments much less certain for application to YM PA calculations than those determined using site-specific materials.

5.1.2.4.2 Methodology 2b2. Sorption Coefficient Has Been Determined From Sorption Test Using Site-Specific Materials

Batch sorption experiments using site-specific materials have been performed yielding K_d values for americium, plutonium, uranium, thorium, radium, neptunium, protactinium, tin, nickel, cesium, strontium, niobium, technetium, selenium, cerium, and europium. Triay, et al. (1997)

states that batch sorption experiments involving protactinium and site-specific materials have only been performed at pH values 6.3 to 6.7. Consequently, the report recommends that sorption experiments involving protactinium be performed at pH values between 7 and 9 to match the expected pH of waters at YM. The NRC staff agrees with this recommendation if protactinium is shown to be important to performance.

Plutonium batch sorption experiments have been performed using site-specific materials. These materials included rocks such as zeolitic tuff, vitric tuff, devitrified tuff, minerals such as clinoptilolite, albite, quartz, synthetic hematite, montmorillonite, calcite, gibbsite, and groundwaters including J-13 and UE-25p#1.

Sorption experiments involving selenium have only used J-13 water. Triay, et al. (1997) states that changes in divalent ion concentration may affect selenium sorption more than changes in pH. The NRC staff agrees with the recommendation in that report that additional experiments using groundwaters with higher divalent ion concentrations (e.g., UE-25p#1) should be performed if sensitivity analysis indicates selenium sorption could be important to performance.

Sorption of uranium in J-13 well water onto YM tuffs has been described by Triay, et al. (1997). The concentrations of uranium in the initial solutions range from $8x10^{-8}$ to $1x10^{-4}$M (19 ppb to 24 ppm). The silica concentration is 57 ppm, and the final pH ranges from 6.9 to 8.6, depending upon whether the experiments were performed in a glovebox maintaining elevated CO_2 pressure or at atmospheric pressure. The NRC staff performed equilibrium calculations using PHREEQC with the WATEQ4F.DAT database and the log K for the dissolution reaction of soddyite from the EQ3/EQ6 database. The results of these calculations suggest that the initial solutions for the sorption experiments may have been supersaturated with respect to soddyite. Appendix D includes the output of the PHREEQC simulation in which uranium is added to J-13 water resulting in a groundwater with a uranium concentration of $1x10^{-4}$ M. The pCO_2 is maintained at $10^{-2.99}$ atm. The species $(UO_2)_2CO_3(OH)_3^-$, and $UO_2(OH)_2$ were added to the database in an attempt to match the species considered by Triay et al., 1997. Triay, et al. (1997) explain the importance of ensuring that a sorption experiment involves sorption and not precipitation. If precipitation has occurred, the resulting partition coefficient is not a K_d to be used in equation (1).

It is possible that the precipitation of soddyite is inhibited. Sorption of uranium onto the solid could be occurring without the precipitation reaction also occurring. However, if precipitation reactions are kinetically inhibited, the use of solubility limits calculated using thermodynamic data to constrain radionuclide concentrations would be brought into question. The Generic Technical Position on Radionuclide Solubilities (Brooks, et al., 1984) recommends constraints on radionuclide concentrations be based on solubility experiments that approach equilibrium from both undersaturation and oversaturation, but neglected the possibility of supersaturated solutions. The staff recognizes that supersaturated solutions are possible, especially at low temperatures, but reasoned that if solubility experiments are performed using site-specific materials under site conditions, kinetic effects could be identified and factored into radionuclide concentration estimates. In a batch sorption experiment, the nucleation of a phase with which the solution is saturated should be enhanced by the presence of nucleation sites on the crushed tuff.

Sorption of uranium as a function of radionuclide concentration was determined resulting in sorption isotherms. Since the dissolution of soddyite depends on pH, uranyl, and silica concentration in solution, the nonvertical isotherm for uranium does not necessarily preclude the possibility of precipitation of soddyite. If silica activity and pH are held constant, and the initial uranium concentration is increased, precipitation of soddyite would produce a vertical isotherm (i.e., constant final uranium concentration in the liquid). However, variations in uranyl concentration could be compensated for by variations in pH and silica concentration, and the solution could still be in equilibrium with soddyite. Chemical analyses of the steady-state groundwater could be used to eliminate the precipitation process as a cause for uranium partitioning between the solid and the liquid.

Another possible explanation for the suggestion of supersaturation with respect to soddyite is that the thermodynamic parameters used for calculating the dissolution of soddyite are incorrect. The database of EQ3/EQ6 uses the estimated free energy of formation of soddyite from Hemingway (1982). Two more recent studies have yielded much higher free energies of formation of soddyite. Nguyen, et al. (1992) and Moll, et al. (1996) experimentally determined synthetic soddyite solubility. These solubility experiments approached equilibrium from undersaturation. The final equilibrated solutions were analyzed for their uranyl and silica concentrations and the Gibbs free energy of formation was calculated. However, the measured silica concentrations (300 ppm) in the final equilibrated solutions exceed solubility of amorphous silica. Consequently, the results of these studies can be challenged.

Triay, et al. (1997) note that unsaturated zone water that is relatively high in calcium and magnesium, may produce low K_d values for uranium, due to competing ions for sorption sites. The NRC staff agrees with the recommendation in the Triay, et al. (1997) report that experiments using high calcium and magnesium, low carbonate, unsaturated zone water composition with pH ranging from 6 to 9, and batch sorption experiments involving high total carbonate and high calcium and magnesium (e.g., UE-25p#1) should be performed.

Thorium sorption experiments involving site-specific materials were reported by Rundberg, et al. (1985) and Thomas (1988). Triay, et al. (1997) suggests that fine colloidal particles in solution may have contributed to uncertainty in the K_d values for thorium. Such a suggestion would indicate confirmatory tests are needed.

Wolfsberg (1978) describes sorption-desorption studies of NTS alluvium that included experiments with niobium. The NRC staff considers these experiments involved material that may approximate material expected to be encountered in the alluvial portion of the flowpath from the repository. Sorption of niobium on other site-specific material using appropriate groundwaters has not been performed. Triay, et al. (1997) suggests that fine colloidal particles in solution may contribute to uncertainty in the K_d values for niobium. Such a suggestion would indicate that confirmatory tests are needed.

Tin sorption experiments have been performed using site-specific rock samples and J-13 and UE-25p#1 groundwaters. Consequently, Criteria 2b1 and 2b2 have been met for tin. However, as with thorium, Triay, et al. (1997) states the large range in sorption coefficients may reflect the presence of colloidal-size particles in the solution phase. Such a suggestion would indicate that confirmatory tests are needed.

Triay, et al. (1997) groups actinium, americium, and samarium together, along with cerium and europium, due to the similarities in their chemical speciation in aqueous solution and their sorption reactions. All exist in the +3 oxidation state and form carbonate, phosphate, and hydroxide complexes. Sorption coefficients for cerium, europium, and americium were determined for a variety of Yucca Mountain rock samples and J-13 and UE-25p#1 groundwaters. Triay, et al. (1997) states that "the main uncertainty regarding the surficial behavior of americium appears to be the degree to which it is mobilized through colloidal transport (for example, Penrose, et al., 1990)." Such a suggestion would indicate confirmatory tests are needed.

Triay, et al. (1997) groups cesium, radium, and strontium together because they exist as simple ions. Their sorption reactions largely involve ion exchange. The batch sorption tests for these radionuclides were performed using site-specific solids and groundwaters including J-13 and UE-25p#1. The NRC staff considers this criterion has been met for these radionuclides.

Nickel sorption has been determined on YM materials including devitrified, zeolitic and vitric tuff from several groundwaters in the pH range from 8.3 to 9.0. The limited range in pH for the nickel experiments is the reason the NRC staff considers this criterion only partially met for nickel. However, it may be possible to calculate the sorption coefficient at lower pH values.

Sorption of lead, samarium, zirconium, and actinium have not been determined using site-specific materials. Consequently, the criterion has not been met for these elements.

Neptunium sorption has been determined using site-specific materials. They include devitrified, vitric and zeolitic tuff, clinoptilolite, hematite, calcite, albite and quartz, J-13 and UE-25p#1 well waters at pH values of 7, 8.5 and 9.0. Initial neptunium concentrations in some batch sorption tests ranged from 1×10^{-7} to 3×10^{-5}. However, Efurd et al., 1997 found that neptunium solubility approached from undersaturation was only $(1.5 \pm 0.6) \times 10^{-5}$ at pH = 8.5 and 25ºC. Consequently, sorption experiments carried out at this condition may have precipitated a neptunium phase.

A general concern of the NRC staff is that although site-specific materials may be used in the batch sorption tests, certain combinations of groundwaters and solids may be unnatural and not expected in the YM environment. For example, it is reasonable to assume that the carbonate groundwater, UE25p#1, is in equilibrium with the Paleozoic carbonates, but would that water ever be found in equilibrium with zeolitic tuff? What is the composition of the initial groundwater in the batch sorption experiment, when 20 mL of UE25p#1 is contacted with 1 g of pre-treated zeolite? If the UE25p#1 and the pre-treated zeolite are out of equilibrium initially, what affect might this have on the partitioning of radionuclide between solid and groundwater?

5.1.2.4.3 Methodology 2b3. Performed Crushed Tuff Flow-through Column Experiment

Column experiments have been performed to confirm batch sorption K_d values for plutonium, neptunium, and technetium. The criterion has not been met for any other elements listed in the table 5-1. The DOE Site Characterization Plan (SCP) (U.S. Department of Energy, 1988) originally called for these tests to be performed with plutonium, americium, neptunium, uranium, technetium, cesium, strontium and barium.

For those radionuclides that may be mobilized as colloids (e.g., americium, plutonium, thorium, neptunium, tin, actinium, samarium, niobium, cerium, and europium), meeting this criterion has added importance, since the K_d from the batch test may have provided an inaccurate measure of the mobility of the radionuclide. Batch experiments need to be designed and conducted, however, to analyze for the presence of colloids. Additionally, for radionuclides that exist as multiple species in solution, the crushed tuff column experiment can be used to determine if the different species can migrate at different rates in an advective system.

Triay, et al. (1996a) demonstrated that the K_d from batch sorption experiments could be used to predict breakthrough for neptunium through crushed zeolitic, devitrified, and vitric tuffaceous material. The apparent dispersivity of the neptunium elution curves through the zeolitic and vitric tuffs led Triay, et al. (1996a) to suggest that "sorption is either nonlinear, irreversible, or noninstantaneous, which means the transport cannot be completely described using a sorption distribution coefficient." However, the report continues that “the use of a batch sorption distribution coefficient to calculate neptunium transport through YM tuffs would result in conservative values for neptunium release.” Consequently, the NRC staff considers that this methodology has been successful for determining the retardation of neptunium.

Triay, et al. (1997) describes crushed tuff column experiments illustrating plutonium migration. Figures 98 through 109 from that summary and synthesis report illustrate elution curves for plutonium and tritium through devitrified, zeolitic, and vitric crushed tuff with J-13 and UE-25p#1 well waters. This document lacks some details needed for the correct interpretation of the elution curves. The report states that “we inject an aliquot of the radionuclide solution (tritiated water or $^{239}PuO_2^+$ in the appropriate groundwater) and then used a syringe pump to elute the radionuclide through the column.” This statement is interpreted to mean that radionuclide is not added continuously, but instead, as a pulse, which is then flushed through the column with uncontaminated groundwater. The ordinate is labeled “Cumulative Concentration (C/C_o)” and the abscissa is labeled “Cumulative Volume (mL).” From the shape of the curves and the assumption that the radionuclide is added as a pulse, it is assumed that the y-axis should be labeled “Cumulative Relative Mass.” That way the curves approach unity with increased volume, as they do.

The elution curve for plutonium through devitrified tuff with J-13 well water (Figure 98) shows that approximately 85% of the plutonium added to the column moves through the system unretarded. Of the remaining plutonium, 5% elutes and 10% stays on the column until the experiment is terminated (20 hrs and 55 mL through column). (The elution curve for tritium reaches a maximum 1.2 in Figure 100 in Triay, et al., (1997). The explanation for C/C_o for tritium exceeding 1 is unknown.)

The elution curve for plutonium through zeolitic tuff is similar to that through devitrified tuff. Figure 106 of Triay, et al. (1997) illustrates that 50% of the plutonium is eluted shortly after the unretarded tritium peak with UE-25p#1 well water. With J-13 water, only 20% of the added plutonium is eluted in the course of the experiment. When vitric tuff is present the elution curves are very shallow indicating gradual release. The amounts eluted range from 8% to 30% for the experiment’s duration. Figures 108 and 109 (Triay, et al., 1997) indicate that flow rates affect the shape of the elution curve, with slower flow rates resulting in significantly reduced percentage of plutonium eluting.

The NRC staff concludes that, although crushed tuff flow-through column experiments have been performed, as recognized in Table 5-1, the criterion to confirm the K_d determined in static tests are appropriate for calculating a retardation in dynamic systems has not been met for plutonium. This conclusion is based on the observations that the thermodynamic calculations using EQ3 do not agree with observed speciation, the recognition that multiple species do exist, including colloidal forms, and K_d values from batch sorption tests ranging from 20 mL/g (for devitrified tuff) to 2,000 mL/g (for vitric tuff) are inconsistent with crushed tuff column experiments where significant portions of plutonium are eluted unretarded. Triay, et al. (1997) state (page 144) that "The shape of the elution curves for plutonium indicates that use of K_d values to predict plutonium transport through YM tuffs will predict plutonium releases conservatively." On the same page, Figure 98 shows 85% of the plutonium eluting unretarded. Therefore, the proposal that batch sorption tests provide information for conservatively estimating plutonium transport appears to be unsupported.

5.1.2.4.4 Methodology 2b4. Performed Intact Rock Flow-through Column Experiment

Conca and Triay (1996) describes a study of selenium migration through unsaturated intact rock. It was demonstrated that the migration of selenium could be estimated conservatively from batch sorption tests. The advantage of this type of experiment over that of batch sorption test is that the sorbing surfaces are likely to be in a more natural state. This methodology successfully demonstrated that the potential effect of disturbed sorption sites on crushed material was not observed for selenium. Similar experiments have not been performed using any of the other radionuclides.

5.1.2.4.5 Methodology 2b5. Performed Diffusion Experiment

Diffusion experiments have been performed for tritium, technetium, neptunium, americium, strontium, cesium, and barium. The advantage of this experimental approach is that the sorbing surfaces are likely to be undisturbed. Consequently, there should be little concern normally associated with crushing, such as generating greater surface areas, and creating fresh surfaces that may be unnaturally sorptive. Figure 132 of Triay, et al. (1997) illustrates a comparison between calculated cesium concentration in a system in which diffusion and sorption are occurring and the actual observed concentrations. The calculated concentrations assumed reversible, instantaneous, linear sorption. The discrepancy between the observed and calculated concentrations suggests that one or more of the assumptions are incorrect. Since isotherms have been measured for cesium (See Criterion 2c1), it may be possible to adjust this comparison.

5.1.2.4.6 Methodology 2b6. Performed Unsaturated Flow Experiment

Conca and Triay (1996) describe solid-rock column unsaturated flow experiments performed using the unsaturated flow apparatus (UFA). The methodology has been successfully applied to the retardation of selenium. The Busted Butte Unsaturated Transport tests are expected to provide additional important information on unsaturated flow and transport in the Topopah Spring and Calico Hills tuffs at the field scale (Bussod and Turin, 1999). Tracers injected during Phase 1 and Phase 2 have included fluorescein and bromide as conservative elements, lithium, and five separate polyfluorinated benzoic acids. Polystyrene latex microspheres are being injected to investigate colloid movement, and a number of reactive tracers such as nickel, cobalt, manganese, samarium, and cerium are being injected as analogs for radionuclide transport (Bussod and Turin, 1999; U.S. Department of Energy, 1998c, Volume 1, Appendix C).

5.1.2.4.7 Methodology 2b7. Used Process Model to Determine Constant K_d

Experimental evidence indicates that sorption behavior is dependent on the physicochemical conditions in the system of interest. Geochemical sorption models to describe these dependencies, and the necessary model parameters, have been summarized and described in several reports (e.g., Davis and Kent, 1990; Serne, et al., 1990; Turner, 1993; Paviet-Hartmann and Triay, 1997). These include ion exchange and surface complexation approaches to sorption, both of which have been used to describe radionuclide sorption mechanisms and characterize the geochemical dependence of radionuclide sorption (e.g., Turner, 1995; Triay, et al., 1997; Paviet-Hartmann and Triay, 1997). Process models have been applied to some extent to sorption of several radioelements of interest in PA, including: neptunium, plutonium, uranium, americium, carbon, cesium, iodine, and selenium (Bradbury and Baeyens, 1993; Dzombak and Morel, 1990; Turner, 1995; Triay, et al., 1997; Paviet-Hartmann and Triay, 1997). The model parameters are calibrated against a set of well-constrained experiments. Process models typically use thermodynamic data for the radioelement of interest to calculate aqueous speciation and determine the amount of radioelement dissolved in solution and the amount sorbed onto the solid. This output can then be used to calculate K_d as a function of chemistry.

Calibrated process models can be used to calculate K_d values under conditions beyond the limited set of conditions for which batch or column experimental sorption data are available (Turner, 1998; Turner, et al., 1998b, 1999). Even in extrapolating model results, however, the application of the K_d approach is subject to the same conditions as the use of batch sorption results. As discussed previously, the assumption of a constant K_d in PA implies that the physicochemical system does not change significantly over the time period of interest. It also requires that the physicochemical conditions remain constant at the level of discretization of the PA model abstraction.

5.1.2.5 **CRITERION 2c.** For the valid application of the constant K_d approach, using equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Demonstrated That the Three Implicit Assumptions (i.e., Linear Isotherm, Fast Reversible Sorption Reaction, and Constant Bulk Chemistry) Are Valid

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "The AMR on radionuclide transport models under ambient conditions provides the supporting rationale for these assumptions."

The DOE comment suggests that there is information in the AMR that would demonstrate these assumptions are valid. To demonstrate the first two assumptions are valid, multiple sorption experiments would have to be performed where initial radionuclide concentration were varied and the duration of the sorption experiments varied, respectively. The NRC staff intends to review this AMR in FY 01.

5.1.2.5.1 Methodology 2c1. Demonstrated Linear Isotherm by Determining K_d as a Function of Radionuclide Concentration

Sorption isotherms have been determined for plutonium, uranium, neptunium, cesium, strontium, cerium, and europium. In comparison, the SCP (U.S. Department of Energy, 1988) had listed only four radioelements for which isotherms would be determined. These radioelements were plutonium, uranium, neptunium, and americium. The sorption isotherm for

europium, a homologue for americium, but with a lower specific activity, and, thus, less of a problem experimentally, has been determined instead of the isotherm for americium.

Triay, et al., (1997) state that the sorption isotherms for plutonium "are generally nonlinear." The nonlinearity in the isotherms is expressed as a curve with the convex side up. As discussed in Section 4, isotherms with convex side up produce self-sharpening fronts and can be used to predict breakthrough. Consequently, the results of this methodology do not preclude the constant K_d for plutonium retardation.

The isotherms for neptunium are generally linear. Only near the solubility limit for neptunium is there some nonlinearity. Uranium isotherms are nonlinear but convex up and so can be used to estimate retardation. Isotherms for cesium, strontium, cerium, and europium are best fitted to a nonlinear Freundlich isotherm, with convex side up.

5.1.2.5.2 Methodology 2c2. Demonstrated Fast Ion Exchange or Adsorption Reaction

Triay, et al. (1996a) investigated the kinetics of sorption of neptunium and found these reactions to be fast when the sorbing materials are tuff or clinoptilolite. When the solid was calcite or hematite, steady state had not been reached in 30 days of contact (experiment duration). It is suggested by Triay, et al. (1996a) that coprecipitation is the process that removes neptunium from solution when in contact with calcite. The NRC staff considers that, although the coprecipitation process will remove neptunium from the groundwater, it is inappropriate to use a K_d from the batch tests in formulating a retardation factor.

A comparison of batch tests and column tests for neptunium sorption (Triay, et al., 1996b) indicated that the batch sorption distribution coefficient could be used to predict arrival time. However, the elution curves showed dispersivity, which may suggest the isotherm is nonlinear, sorption is irreversible, or noninstantaneous. These column experiments did not involve significant calcite.

Batch sorption tests for plutonium failed to reach steady state in 32 days (experiment duration). Likewise, crushed tuff column experiments resulted in some of the plutonium eluting unretarded. Variation in the flow rate resulted in different proportions of the plutonium moving unretarded through the column. At fast flow rates, larger proportions moved unretarded; at slow flow rates, smaller proportions moved unretarded.

Triay, et al. (1997) states that "It is noteworthy that the experimentally determined redox behavior of plutonium in solution was quite distinct from the behavior predicted on the basis of EQ3 calculations (Nitsche, 1991)." The report goes on to say "the uncertainties in our knowledge of the solution behavior of plutonium will make it difficult to properly interpret the sorption behavior of that element." The NRC staff considers that this criterion has not been met for plutonium. Neither has the removal of plutonium from solution been shown to be the result of an ion exchange or adsorption reaction nor has the reaction been shown to be fast.

The crushed tuff column experiments for studying the elution of plutonium involved adding a pulse of plutonium-contaminated groundwater to the system and flushing through with uncontaminated groundwater. Some unknown reaction reduced the amount of plutonium exiting the column. It is possible that a precipitation reaction could have produced this result. If the experiments were performed with a continuous source instead of a pulse, the experiment

might better simulate the situation expected at YM, where the plutonium source term should be continuous after the waste packages are breached.

Technetium has been eluted through crushed tuff columns at different flow rates. The flow rate did not affect breakthrough. There was little or no interaction with the solid as technetium moved through the column unretarded, so flow rates should have had no effect as observed.

5.1.2.5.3 Methodology 2c3. Demonstrated Constant Bulk Chemistry

As discussed previously (Section 4.1.2.2.3.3), an implicit assumption in using K_d to describe radionuclide sorption is that the chemistry of the groundwater is constant at the scale of discretization used in the transport model. Many of the sorption experiments discussed previously (e.g., Meijer, 1990) are performed using water from tuff aquifers in well J-13 in Jackass Flat and carbonate aquifers in well UE-25p#1. A number of studies, however, have demonstrated that water chemistry in the vicinity of Yucca Mountain can vary significantly with regard to parameters such as pH, ionic strength, and CO_3^{2-} concentration that may affect radionuclide retardation (Yang et al., 1996a,b; Perfect, et al., 1995; Turner, et al., 1999). Both DOE and NRC have made an effort to account for the effects of this changing chemistry in developing the PDFs used in TSPA-VA (Triay et al., 1997; Civilian Radioactive Waste Management System, Management and Operations, 1998a; U.S. Department of Energy, 1998c; Turner, et al., 1999). It also may be possible to demonstrate, through the use of process models, that the observed variability in geochemical conditions will produce minimal changes in K_d. Another approach is to simulate transport using a dynamic reactive transport system model (e.g., PHREEQC, MULTIFLO, HYDROGEOCHEM, etc.) to evaluate the effects of chemical heterogeneity on transport.

5.1.2.6 **CRITERION 3a.** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: Demonstrated That a Portion of the Flow Path Acts as an Isotropic Homogeneous Porous Medium [see USFIC IRSR (Nuclear Regulatory Commission, 1998b) on Deep Percolation]

Same as Criterion 2a.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that “The vitric portion of the Calico Hills hydrogeologic units behaves as a porous medium and provides an effective barrier to radionuclide transport. The upper units of the Prow Pass hydrogeologic unit also behaves as a porous medium.” The NRC staff notes that the DOE comment does not include the term “single continuum.” If this omission is intentional, and the units identified are not expected to act as single continua but as dual continua, then DOE would have to demonstrate its capability to predict breakthrough curves.

5.1.2.7 **CRITERION 3b.** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: Demonstrated That Appropriate Values Are Used in Process Models

Table 5-2 shows that process models have been applied to some extent to sorption of several radioelements of interest in PA including: neptunium, plutonium, uranium, americium, carbon,

cesium, iodine, and selenium (Bradbury and Baeyens, 1993; Dzombak and Morel, 1990; Turner, 1995; Triay, et al., 1997; Paviet-Hartmann and Triay, 1997). These models typically use thermodynamic data for the radioelement of interest to calculate aqueous speciation, and to determine the amount of radioelement dissolved in solution and the amount sorbed onto the solid. This output can then be used to calculate K_d as a function of chemistry. Although process models can be applied over a wide set of geochemical conditions, it is recognized that determining the necessary model parameters relies on calibration against a set of well-constrained experiments. To provide additional confidence in the model results, the experiments used to calibrate the model should cover a range in critical parameters such as pH, ionic strength, and radionuclide concentration. The experimental variation in these parameters should span the range expected over the time period of interest at the scale of discretization in the PA abstraction.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "UZ Transport parameters are summarized in Section 3.11.2 of the UZ F&T PMR." In response, the NRC staff intends to review the UZ F&T PMR in FY01.

5.1.2.8 **CRITERION 3c.** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: (i) Demonstrated That the Three Implicit Assumptions (as in 2c) are Realized, If Process Models Are Intended to Yield a Constant K_d for Use in the Retardation Equation (Equation 1); Otherwise, Determined Transport in a Dynamic Reactive Transport System Model (e.g., PHREEQC, MULTIFLO, HYDROGEOCHEM, etc.)

Turner (1998) uses groundwater compositions from Perfect, et al. (1995) to suggest the possibility of a significant range of K_d values for neptunium and uranium for the YM region. However, the variation in groundwater composition in the portion of the pathway that is considered to be an isotropic homogeneous porous medium may be less than that over the whole region, which includes significant fractured rock. The limited availability of groundwater analyses for the porous rock units will likely result in a limited range of calculated K_d values. One additional concern stems from the evidence that in some wells, groundwater compositions have varied over time.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "The AMR on radionuclide transport models under ambient conditions provides the supporting rationale for these assumptions." The NRC staff intends to review this AMR in FY01.

5.1.2.9 **CRITERION 4.** Where Data Are Not Reasonably or Practicably Obtained, Expert Judgement Has Been Used and Expert Elicitation Procedures Have Been Adequately Documented. If Used, Expert Elicitations Were Conducted and Documented in Accordance with the Guidance in NUREG-1563 (Nuclear Regulatory Commission, 1996) or Other Acceptable Approaches

The NRC staff considers K_d values from batch sorption tests, confirmed by crushed tuff column or other experiments, can be reasonably or practicably obtained. Consequently, expert elicitation would not appear to be appropriate, as a general matter, to use in place of

experimentally determined sorption coefficients. This applies only to those radionuclides whose sorption characteristics affect performance.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "No expert elicitation was conducted." With regard to expert elicitation, the NRC staff considers this acceptance criterion has been met. However, the staff needs clarification on the use of expert judgement in radionuclide transport. For example, how are the distributions of sorption parameters determined? Triay et al., 1997 state that "sorption coefficient distributions are inferred from data..." NRC staff requests that the term "inferred" be clarified.

5.1.2.10 CRITERION 5. Data and Models Have Been Collected, Developed, and Documented under Acceptable QA Procedures (e.g., Altman, et al., 1988), or If Data Were Not Collected under an Established QA Program, They Have Been Qualified under Appropriate QA Procedures

The Total System Performance Assessment—Viability Assessment (TSPA-VA) indicates that the K_d values for plutonium, uranium, and neptunium are Q-listed. All other K_ds and K_d distributions have Not Qualified (NQ) QA status.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Activities associated with this work were determined to be subject to the quality assurance program as described in the Quality Assurance Requirements Description (QARD) document." In response, the NRC staff considers the status unchanged. Values of K_d for plutonium, uranium, and neptunium are Q-listed. All other K_ds and K_d distributions have Not Qualified QA status.

5.2 STATUS OF SUBISSUE 2 -- RADIONUCLIDE TRANSPORT THROUGH ALLUVIUM

5.2.1 Summary of the Status of Subissue 2

The status of this subissue is tied closely to that of the previous subissue. However, additional uncertainty is a result of the very limited information collected to date on the mineralogy, groundwater chemistry, and flow systems of the alluvium. Past efforts by the DOE have focused on characterizing the geologic media within 5 km of the repository because of the provisions of the then applicable 40 CFR Part 191. With the resultant increase in the length of the flowpath to the biosphere to 20 km, consistent with the proposed 10 CFR Part 63 (Nuclear Regulatory Commission, 1999c), a significant portion of relatively uncharacterized geologic media has been added to the system.

Although the NRC staff currently assumes in its TPA code that the alluvium acts as a homogeneous porous medium, it is recognized that little or no information is available to support that assumption. Furthermore, it is recognized that the staff's current assumption may be nonconservative.

The NRC staff expects that the series of boreholes to be drilled in the alluvium as part of the Nye County EWDP will provide significant information concerning its geologic, and hydrologic characteristics. It is expected that the mineralogy will reflect that used in batch sorption experiments for determining K_ds for radionuclides in tuff. If that is so, the laboratory work

needed to address Subissue 1 will also address Subissue 2. Through early 1999, the EWDP had drilled eight wells to the south of YM. Lithologic logs available through the Nye County website (Nye County, 1999) indicate that much of the alluvium consists of valley fill deposits of gravel, silt, and sand varying in thickness from 33 m (110 ft) in well NC-EWDP-3S to more than 490 m (1618 ft) in well NC-EWDP-2D.

The determination of the modes of flow in the alluvium will require field tests. If the alluvium is a composite of cut and fill structures resulting from the accretion of anastamosing channels, preferred pathways limiting water-rock interaction may result. Criteria associated with the subissue on radionuclide transport in fractured rock would then apply. If on the other hand, the alluvium is homogeneous, the application of experimentally determined K_ds to calculate retardation factors would be appropriate. Resolution of this subissue will await the geologic and hydrologic information to be collected in the Nye County EWDP.

5.2.2 Criterion-By-Criterion Analysis

Same as Section 5.1.2.

5.2.2.1 **CRITERION 1a** For the estimation of radionuclide transport through porous rock, DOE has determined through PA calculations whether radionuclide attenuation processes such as sorption, precipitation, radioactive decay, and colloidal filtration are important to performance.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ performance assessment calculations indicate these processes are important." In response, the NRC staff considers this acceptance criterion has been met.

5.2.2.2 **CRITERION 1b** For the estimation of radionuclide transport through porous rock, DOE has (i) Assumed K_d is zero and radionuclides travel at the rate of the groundwater flow, if it has been found that radionuclide attenuation is unimportant to performance and it can be demonstrated that this assumption is conservative in which case, Acceptance Criteria 2 and 3 do not have to be met or, (ii) demonstrated that Criterion 2 or 3 has been met, if radionuclide attenuation in porous rock is important to performance, or if an assumption that K_d is zero in porous rock is not conservative.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "K_d is assumed to be zero for C and Cl." The NRC staff notes that Tc and I are not assumed to have K_d value of zero as was assumed in the porous rock. The presentation at the Technical Exchange included results of recent batch sorption experiments for Np, Tc, and I. The values of K_d were nonzero for all three elements. The staff requests that DOE describe plans to confirm the K_d values.

5.2.2.3 **CRITERION 2a** For the valid application of the constant K_d approach, equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Demonstrated that the flow path acts as a single continuum porous medium. If the flow can not be shown to be a single continuum porous medium, then the acceptance criteria for radionuclide transport in fractured rock apply.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ fluid flow in the alluvium is likely to be well represented using a porous continuum conceptual model. Data to quantify the alluvial portion of the flow path are sparse, and hydrologic parameters used in numerical models should be considered to be bounding."

Additionally, this acceptance criterion was identified in the Technical Exchange presentation as an area of potential disagreement. This point was also made in the letter from Brocoum to Reamer, 03/22/00. In the presentation, it is stated that "homogeneity of porous rock or alluvium does not need to be demonstrated at all scales." Instead DOE suggests that "Homogeneity only needs to be addressed at the level of assumed homogeneity in the model, for example, the grid discretization or level of heterogeneity modeled."

In the letter from Brocoum to Reamer, it is stated that "DOE believes that, consistent with the intent of proposed 10 CFR Part 63, IRSR discussions should not impose prescriptive requirements and should not call for additional DOE work not clearly linked to repository performance. We believe in this case that qualitative arguments should suffice (e.g., alluvium is unconsolidated sediment) to justify treating alluvium as a porous medium."

In response, the NRC staff is unconvinced that the alluvium can be considered homogeneous on the scale of grid discretization, which has been reported to 500 m. Logging of Nye County EWDP holes suggests heterogeneity in a vertical sense. Cross-hole correlation of the lithologic logs from the EWDP could provide information on heterogeneity in a horizontal sense on scales of tens of meters and greater. Furthermore, recent pump tests suggest heterogeneity of hydraulic parameters in single well. The NRC staff agrees with DOE that homogeneity only need be demonstrated on the scale modeled. However, the method by which this can be accomplished on blocks 500 m on a side is unclear.

Qualitative arguments will not suffice if retardation in the alluvium is important to performance. Performance assessment calculations by the NRC staff suggest the alluvium can be an important component of the geologic barrier.

5.2.2.4 **CRITERION 2b** For the valid application of the constant K_d approach, equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Demonstrated that appropriate values for the parameters, K_d, n or θ, and ρ_b have been adequately considered (e.g., experimentally determined or measured).

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ transport of sorbing solutes in porous media is a subject that has been well studied. Therefore, it is reasonable to assume that the transport velocities of sorbing radionuclides in the alluvium can be conservatively represented using an equilibrium sorption coefficient. Sorption coefficients onto alluvium from the Nye County wells have been measured for a few key radionuclides; for the remaining radionuclides, sorption coefficients have been estimated based on the corresponding values measured for crushed tuff. Recent evaluations of K_d for Np, Tc, and I have been accomplished for alluvium." The NRC staff agrees that sorption of solutes in porous media has been the subject of considerable study. Sorption onto alluvial materials is new. It is likely that information from crushed tuff experiments can be applied to the alluvial system. However, this would require comparing the solid phases present in the alluvium with phases in the crushed tuff experiments. The fact that Tc and I have nonzero K_d values for the alluvium and not the porous rock suggest some differences.

5.2.2.5 **CRITERION 2c** For the valid application of the constant K_d approach, using equation (1) $Rf = 1 + \rho b K_d/n$, (or $Rf = 1 + \rho b K_d/\theta$), DOE has: Demonstrated that the following assumptions (i.e., linear isotherm, fast reversible sorption reaction, and constant bulk chemistry) are valid.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Sorption is incorporated into the site-scale SZ flow and transport model using a linear isotherm model." In response, the NRC staff considers the constant K_d approach should be confirmed through the use of column experiments. This approach is recommended by Triay et al., 1996b.

5.2.2.6 **CRITERION 3a** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: Demonstrated that the flow path acts as a single continuum porous medium.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ fluid flow in the alluvium is likely to be well represented using a porous continuum conceptual model. Data to quantify the alluvial portion of the flow path are sparse, and hydrologic parameters used in numerical models should be considered to be bounding."

In response, the NRC staff considers this the same as for 2a.

5.2.2.7 **CRITERION 3b** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: Demonstrated that appropriate values are used in the process models.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ transport of sorbing solutes in porous media is a subject that has been well studied. Therefore, it is reasonable to assume that the transport velocities of sorbing radionuclides in the alluvium can be conservatively represented using an equilibrium sorption coefficient. Sorption coefficients onto alluvium from the Nye County wells have been measured for a few key radionuclides; for the remaining radionuclides, sorption coefficients have been estimated based on the corresponding values measured for crushed tuff. Recent evaluations of K_d for Np, Tc, and I have been accomplished for alluvium." In response, the NRC staff considers this the same as for 2b.

5.2.2.8 **CRITERION 3c** For the valid application of process models affecting radionuclide transport such as surface complexation, ion exchange, precipitation/dissolution, and processes involving colloidal material, DOE has: (i) Demonstrated that the three implicit assumptions are valid, if process models are intended to yield a constant K_d approach for use in the retardation equation; or (ii) determined transport in a dynamic reactive transport system (e.g., PHREEQC, MULTIFLO, HYDROCHEM, etc.)

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Sorption is incorporated into the site-scale SZ flow and transport model using a linear isotherm model." In response, the NRC staff considers this the same as 2c.

5.2.2.9 **CRITERION 4** Where data are not reasonably or practicably obtained, expert judgement has been used and expert elicitation procedures have been adequately documented. If used, expert elicitations were conducted and documented in accordance with the guidance in NUREG-1563 (Nuclear Regulatory Commission, 1996) or other acceptable approaches. However, the NRC staff considers K_d values from batch sorption tests, confirmed by crushed tuff column or other experiments can be reasonably or practicably obtained. Consequently, expert elicitation would not appear appropriate to use in place of experimentally determined sorption coefficients.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "No expert elicitation was conducted." With regard to expert elicitation, the NRC staff considers this acceptance criterion has been met. However, the staff needs clarification on the use of expert judgement in radionuclide transport. For example, how are the distributions of sorption parameters determined? Triay et al., 1997 state that "sorption coefficient distributions are inferred from data..." the term "inferred" needs to be clarified.

5.2.2.10 **CRITERION 5** Data and models have been collected, developed and documented under acceptable QA procedures (e.g., Altman et al., 1988), or if data were not collected under an established QA program, they have been qualified under appropriate QA procedures.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Activities associated with this work were determined to be subject to the quality assurance program as described in the Quality Assurance Requirements Description (QARD) document." In response, the NRC staff considers the status unchanged. Values of K_d for plutonium, uranium, and neptunium are Q-listed. All other K_ds and K_d distributions have a Not Qualified QA status.

5.3 STATUS OF SUBISSUE 3—RADIONUCLIDE TRANSPORT IN FRACTURED ROCK

5.3.1 Summary of the Status of Subissue 3

In current PA calculations (Nuclear Regulatory Commission, 1999a,b; U.S. Department of Energy, 1998c), no retardation is assumed in fractures in PA calculations, and radionuclides are transported through the fractures at the same velocity as groundwater. Under these conditions, Criteria 2 and 3 do not need to be satisfied, and flow issues related to fracture/matrix interaction and fracture flow velocity are the critical aspects of radionuclide transport. These flow issues are considered as part of the USFIC KTI, and the reader is referred to the USFIC IRSR (Nuclear Regulatory Commission, 1998b) for an analysis.

Experiments have been performed using fractured rock (Triay, et al., 1997). Whereas the retardation factor in fractures is typically assumed to be 1 (i.e., no sorption) in performance assessments, due to the uncertainty with regard to radionuclide transport in fractured rock,

preliminary experiments suggest that some retardation occurs. For example, neptunium experiments have been performed and show reduced recovery and a delay in the breakthrough relative to tritium and technetium. Field scale experiments (30 to 100 m) being conducted at the C-Wells complex (Reimus and Turin, 1997; Reimus, et al., 1998) result in bimodal breakthrough curves for nonreactive tracers (polyfluorinated benzoic acids, bromine), reactive solutes (lithium), and microspheres. Reimus, et al. (1998) suggest that fast pathways and diffusion from the fracture into the matrix may play a role in saturated zone transport (Reimus, et al., 1998). Resolution of this subissue will depend on a combination of laboratory experiments by DOE and additional tracer tests like those at the C-holes and Busted Butte (Bussod and Turin, 1999) field sites.

5.3.2 Criterion-By-Criterion Analysis

5.3.2.1 CRITERION 1: Estimation of Radionuclide Transport Through Fractured Rock

5.3.2.1.1 CRITERION 1a. For the estimation of radionuclide transport through fractured rock, DOE has: Determined, Through PA Calculations, Whether Radionuclide Attenuation Processes such as Sorption, Precipitation, and Radioactive Decay, and Colloidal Transport are Important to Performance

DOE has used performance assessment calculations to determine whether radionuclide attenuation processes are important to performance. Consequently, this criterion has been met.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Performance assessment calculations have determined that such processes are important. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." In response, the NRC staff considers this criterion has been met.

5.3.2.1.2 CRITERION 1b. For the estimation of radionuclide transport through fractured rock, DOE has: Assumed K_d (or K_A) is Zero and that Radionuclides Travel at the Velocity of Groundwater Flow Through Fractures, If it has been Found that Radionuclide Attenuation in Fractures is Unimportant to Performance, and it Can Be Demonstrated that this Assumption Is Conservative

Current performance assessment models assume radionuclides are not sorbed in fractures. However, it appears that matrix diffusion will be considered as an attenuation processes affecting radionuclide transport in fractured rock. Consequently, this criterion has not been met.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "K_d is assumed to be zero for C and Cl. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." The NRC notes that K_d for I and Tc are assumed to be zero in the porous rock but here in fractured rock their K_ds are considered to be nonzero. This apparent discrepancy needs to be clarified.

5.3.2.1.3 **CRITERION 1c.** For the estimation of radionuclide transport through fractured rock, DOE has: Justified the Length of Flowpath to Which These Fracture Transport Conditions Apply

Geostatistical analysis of tracer tests as suggested in Criterion 2a can provide support for establishing the length of the flowpath to which fracture transport conditions apply. Lacking a geostatistical analysis, estimation of the fractured pathway length should err on the side of conservatism. This criterion has not been met.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Chloride hydrochemical groundwater data, as well as other chemical and isotopic data and the potentiometric surface map have been used to constrain SZ flowpaths. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." The NRC staff will review the AMR describing the use of hydrochemical data to constrain SZ flowpaths in FY01. This acceptance criterion is intended to apply to both the SZ and UZ fracture rock flow systems.

5.3.2.2 CRITERION 2: Radionuclide Attenuation in Fractured Rock

5.3.2.2.1 **CRITERION 2a.** If credit is to be taken for radionuclide attenuation in fractured rock, DOE has: Demonstrated Capability to Predict Breakthrough Curves of Reactive, Nonreactive, and Colloidal Tracers in Field Tests

Unlike the estimation of transport in porous media, which is supported by 50 years of chemical engineering experience in chromatographic separation techniques, the estimation of transport through fractured rock is relatively untested. The C-hole reactive tracer test is the only field test the NRC staff is aware of in the SZ at YM that provides direct information on the transport of reactive, nonreactive, and colloidal material. The C-hole breakthrough curves (concentration and travel time) could not be quantitatively predicted using the laboratory experiments including batch sorption, crushed tuff column, diffusion, and fractured rock column tests, alone, or in concert with the hydraulic pump tests in the C-holes. Consequently, this criterion is not met.

The NRC staff considers the cross hole reactive tracer tests, like those at the C-hole complex and the Busted Butte facility to be crucial to demonstrate the capability to predict transport. The use of field tests to compare back to the laboratory experiments is a logical extension to the strategy proposed by Triay et al., 1992 to validate the batch sorption data. Geostatistical analysis of multiple tracer tests could be used to demonstrate the capability to predict radionuclide transport in fractured rock.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ model of fracture flow and matrix diffusion consistent with data. Sorbing tracer (Li) breakthrough consistent with combined matrix diffusion/sorption model. Microspheres breakthrough slightly earlier than solutes, but are attenuated. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." The NRC staff questions the appropriateness of developing an SZ model that extends tens of kilometers based on a test that extends only tens of meters. The C-well test was an excellent experiment that provided important information on transport in a fractured system on the scale of 30 meters. However, extrapolation to larger scales is untested. If processes other than pure matrix diffusion are occurring in a tracer test (e.g., Becker and

Shapiro, 2000), extrapolation using inappropriate abstract models could lead to miscalculations of breakthrough curves.

The NRC staff is unclear how credit can be taken for matrix diffusion but sorption in fractures is neglected in PA transport evaluations because of limited data. In the TSPA VA it is stated that "surfaces of fractures, often lined with minerals that differ from the bulk of the rock matrix may be capable of sorbing many of the radionuclides (Triay et al., 1997). However, there has been limited characterization of the distribution of the fracture-lining materials and sorptive interactions with these minerals." The NRC staff requests that DOE explain this apparent inconsistency.

In the letter from Brocoum to Reamer, 3/22/00, on reviewing this acceptance criterion, the DOE comment states that "The TSPA models are not designed to be predictive or to represent transport on the scale of a few meters and over the timeframe of days. The models are designed to bound the potential transport rates over some 20 km and over periods pf 10,000 years or more. Given these constraints, it would be unrealistic to require the same models to predict tracer transport on much smaller time scales." The NRC staff agrees that the TSPA models are not suited for predictions of small scale systems. However, DOE made predictions of transport in column tests from the results of batch tests. The characterization program has been based on a principle of testing simple systems first and using the results to predict more complex systems. Can DOE predict breakthrough curves of field experiments?

Also, in the letter it was stated that this work is not clearly related to repository performance. The staff disagrees. The C-well work should be repeated in other localities with efforts to predict the breakthrough preceding the tests.

5.3.2.2.2 **CRITERION 2b.** If credit is to be taken for radionuclide attenuation in fractured rock, DOE has: Demonstrated Nonradioactive Tracers Used in Field Tests Are Appropriate Homologues for Radioelements

Considerable effort has been expended in identifying reactive tracers that mimic the properties of radionuclides. The use of "cocktails" of tracers is an innovative approach at characterizing transport processes in the field. The NRC staff considers this criterion is being met with regard to the Busted Butte and C-hole complex tests. The NRC staff will continue to track the tests to assure this criterion is met.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "SZ model of fracture flow and matrix diffusion consistent with data. Sorbing tracer (Li) breakthrough consistent with combined matrix diffusion/sorption model. Microspheres breakthrough slightly earlier than solutes, but are attenuated. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." The NRC staff considers this criterion has been met, but will continue to track the tests to assure reasonable tracers are used.

5.3.2.2.3 **CRITERION 2c.** For the estimation of radionuclide transport through fractured rock, DOE has: Justified the Length of Flowpath to Which These Fracture Transport Conditions Apply

Same as Criterion 1c. Geostatistical analysis of tracer tests as suggested in Criterion 2a can provide support for establishing the length of the flowpath to which fracture transport conditions

apply. Lacking a geostatistical analysis, estimation of the fractured pathway length should err on the side of conservatism. This criterion has not been met.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Chloride hydrochemical groundwater data, as well as other chemical and isotopic data and the potentiometric surface map have been used to constrain SZ flowpaths. Sorption in the fractures is neglected in the PA transport evaluations because of limited data and the conservative nature of the assumption." In response, the NRC staff consider this is the same as 1c.

5.3.2.3 **CRITERION 3:** Where Data Are Not Reasonably or Practicably Obtained, Expert Judgement Has Been Used and Expert Elicitation Procedures Have Been Adequately Documented. If Used, Expert Elicitations Were Conducted and Documented in Accordance with the Guidance in NUREG-1563 (Nuclear Regulatory Commission, 1996) or Other Acceptable Approaches. Expert Elicitation and Sensitivity Analyses Should Not Be Used as a Replacement for Experimental and Field Data, Where Such Data Can Be Reasonably Obtained

If expert judgement or elicitation is used to address radionuclide transport through fractured rock, the NRC staff would expect those people elicited would have a proven track record of predicting tracer breakthrough curves in fractured rock. Further, it would have to be demonstrated that this expertise can be applied to the fracture systems in the YM region. This approach has not been taken by the DOE so the NRC staff has no comment. A discussion of the status of meeting this criterion is not applicable.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "No expert elicitation was conducted." With regard to expert elicitation, the NRC staff considers this acceptance criterion has been met. However, the staff needs clarification on the use of expert judgement in radionuclide transport. For example, how are the distributions of parameters important to transport determined?

5.3.2.4 **CRITERION 4:** Data and Models Have Been Collected, Developed and Documented under Acceptable QA Procedures (e.g., Altman, et al., 1988), or If Data Were Not Collected under an Established QA Program, They Have Been Qualified under Appropriate QA Procedures

The NRC has expressed concerns about the DOE's QA program (U. S. Nuclear Regulatory Commission, 1999). Status of this criterion is to be determined.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "Activities associated with this work were determined to be subject to the quality assurance program as described in the Quality Assurance Requirements Description (QARD) document." In response, the NRC staff considers the status is open.

5.4 STATUS OF SUBISSUE 4—NUCLEAR CRITICALITY IN THE FAR FIELD

5.4.1 Summary of the Status of Subissue 4

NRC staff has recently completed a review of the methodology report (U.S. Department of Energy, 1998d) and prepared a safety evaluation report (Nuclear Regulatory Commission, 2000) to document the review findings. Therefore, a detailed review of the far-field portions of the methodology report is not presented here. Based on the outlined methodology (U.S. Department of Energy, 1998d) and on previous work in the viability assessment (U.S. Department of Energy, 1998c), NRC staff believes DOE is on a path to resolution of this subissue. This subissue is closed pending confirmation that the methodology is being implemented appropriately. The NRC will continue to review any AMR or PMR describing DOE's analysis supporting their screening of disposal criticality in the far field as part of the safety evaluation report process. DOE will need to provide a technical basis for its probability screening. If probabilities for far-field criticality events exceed 10^{-8}, DOE will need to provide an adequate demonstration that the TSPA abstraction has bounded the effect on repository performance.

5.4.2 Criterion-By-Criterion Analysis

5.4.3 Criterion 1 DOE has determined the probabilities of scenarios that lead to the accumulation of a critical mass of fissile material into a critical configuration within 10,000 years in the far field using appropriate site characteristics.

The proposed DOE methodology logically outlines the steps to evaluate the probability of accumulating a critical mass of fissile material into a critical configuration in the UZ far field at YM. The DOE methodology report (U.S. Department of Energy, 1998d) anticipates that the likelihood of nuclear criticality in the UZ is low, and screens criticality on that basis. Although the methodology appears sound (Nuclear Regulatory Commission, 2000), DOE will need to provide a sufficient technical basis for this screening.

DOE commented at the Technical Exchange on Yucca Mountain Pre-Licensing Issues on April 26, 2000 that "The Disposal Criticality Analysis Methodology Topical Report has been developed and submitted to the NRC. A draft NRC Safety Evaluation Report has been released. FEPs screening considers event of farfield occurrence to be of low probability. Analyses show no significant effect on calculated performance in first 10,000 years, even if waste packages are breached." The NRC has not seen these calculations**.**

In the letter from Brocoum to Reamer on 03/22/00 consisting of DOEs review of the RT IRSR Rev 1, Comment 2 states that plutonium-bearing colloids from glass waste form contain only trace plutonium and thus would not contribute greatly to the mass transfer of the fissile material. The NRC staff agrees that the reference cited does not support a substantially increased radionuclide load. It is recognized that low concentrations of fissile material in the leachate, make it difficult to move a critical mass. Conditions that could accentuate the mass transfer would increase probability of a farfield criticality. If the water passing through the repository is muddy, carrying a large amount of particulates/colloids, it is possible that a larger quantity of fissile material could be mobilized. A system that has been boiling for hundreds of years may be characterized by muddy water (e.g. Yellowstone). A pot in which sediment (dirt) has been allowed to settle, has a relatively low quantity of particulates and colloids in the supernatant. If the pot is heated, the fluid will begin to convect entraining more particulates and colloids. At a rolling boil, the water is turbid. Can this water come in contact with the waste form? The NRC staff has not done the analysis. At this point, this scenario is just speculation.

With regard to the accumulation process, DOE states in Comment 2 that if such an efficient clay filtration mechanism existed, the interstices of the nonwelded bedded units would already be filled. This statement requires the assumption that the flow system of the site will remain unchanged throughout the repository lifetime. This assumption has not been demonstrated to be valid.

The comment goes on to say that "DOE believes the largest possible accumulations or uranium in the far field would take place in natural deposits analogous to those that have produced the richest uranium ore deposits, namely organic reducing zones. The comment references CRWMS M&O, 1996. In this document it is stated that "From the results of this analysis it is recommended that future studies should:

> Determine the geochemistry of the invert and analyze the potential for precipitation of a critical mass.
> Perform an in-depth screening of geochemical processes and geologic scenarios to identify any which could produce a sufficiently concentrated reducing zone in tuff (either organic or otherwise) to precipitate a critical mass of commercial SNF uranium (VLEU).
> Evaluate the potential for external criticality for highly enriched DOE owned SNF.

5.4.4 **CRITERION 2** For those scenarios that have probabilities greater than 10^{-8}, the DOE has determined their effects on performance.

DOE commented in the Technical Exchange on Prelicensing Issues on April 22, 2000 that "The Disposal Criticality Analysis Methodology Topical Report has been developed and submitted to the NRC. A draft NRC Safety Evaluation Report has been released. FEPs screening considers event of far field occurrence to be of low probability. Analyses show no significant effect on calculated performance in first 10,000 years, even if waste packages are breached." In response, the NRC staff has not seen the calculations addressing consequences.