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U.S. DEPARTMENT OF ENERGY (DOE) REVIEW OF U.S. NUCLEAR REGULATORY COMMISSION'S (NRC) RADIONUCLIDE TRANSPORT ISSUE RESOLUTION STATUS REPORT, REVISION 1

The DOE has reviewed Revision 1 of the Issue Resolution Status Report (IRSR) on the Key Technical Issue of Radionuclide Transport. The enclosed comments are directed primarily at the acceptance criteria for the subissues associated with radionuclide transport and related discussions of the technical bases supporting those criteria.

In general, DOE agrees with the risk-informed, performance-based approach that the NRC staff has adopted in its development of the proposed 10 CFR Part 63. However, we are concerned that some of discussions in the subject IRSR contain implicit or explicit requirements beyond those in the acceptance criteria. A number of these requirements appear to be more prescriptive than is the intent of the performance-based proposed 10 CFR Part 63. They appear to remove the flexibility contained in the proposed regulations and are not clearly linked to repository performance. These concerns are discussed in our comments.

DOE appreciates the opportunity to review the IRSRs and provide comments for your consideration. The enclosure contains both general and specific comments and additional comments. We request that our comments be considered in the preparation of the next revision of the IRSR.

If you or your staff have any questions regarding our comments, please contact Deborah Barr at (702) 794-5511 or Carol Hanlon at (702) 794-1324.

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OL&RC:TCG-0837

Enclosure:
Comments on Issue Resolution Status
Report, Revision 1, Key Technical Issue:
Radionuclide Transport

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YMP-5

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COMMENTS ON ISSUE RESOLUTION STATUS REPORT. REVISION 1, KEY TECHNICAL ISSUE: RADIONUCLIDE TRANSPORT

General Comments

The Radionuclide Transport (RT) Issue Resolution Status Report (IRSR) addresses only radionuclide mobility in groundwater. Issues related to groundwater flow though the unsaturated zone and saturated zone are evaluated as part of the Unsaturated and Saturated Flow under Isothermal Conditions IRSR. The IRSR acknowledges that transport of gaseous radionuclides is not considered to be an issue because of the recommendations of the National Academy of Science (National Research Council, 1995), to adopt a dose-based standard. Gaseous radionuclides are not expected to be significant contributors to dose.

Consistent with NRC's view that the processes and conditions that disperse the fissile material will dominate over processes and conditions that would tend to concentrate fissile material into critical geometries in the far field (NRC 1999, p. 6), and results of preliminary DOE studies (as discussed in comment #2 below), which indicate that the probability of criticality in the far field is below the threshold of regulatory concern, DOE agrees with the assessment by the NRC staff that this subissue is resolved

In light of the above, DOE recommends combining the remaining subissues of the RT KTI in the discussions of Unsaturated and Saturated Flow under Isothermal Conditions (USFIC) IRSR. Combining discussions of radionuclide transport in porous rock, alluvium, and fractured rock in the USFIC IRSR would allow the flow and transport mechanisms to be discussed together. Such a discussion would require expanding the scope of the USFIC IRSR, but the modifications would more closely match the way the issues are being discussed and developed by DOE in the process model reports (PMRs). Similar changes have previously been found acceptable (e.g., including consideration of radionuclides in volcanic ejecta in the Igneous Activity KTI).

The recommended change would retain the remaining subissues and at the same time allow the Radionuclide Transport KTI to be eliminated as a separate key technical issue.

Specific Comments

1. Section 3.0, page 6, bullet 1: The last sentence in this paragraph states that "the occurrence of cut and fill structures formed in the alluvium by braided streams as evident in the Fortymile Canyon may suggest that preferred pathways exist in the alluvium with the potential to reduce mixing and dilution." This statement is not well supported by information provided in the paragraph.

   If the NRC staff can provide information to the contrary, we would be glad to examine the same. DOE believes the existing wording implies existence of geologic processes that are not consistent with conditions generally found in alluvial fan structures in the arid southwest.

2. Section 3.0, page 6, bullet 3: "Criticality in the far field... For example, Bates, et al. (1992) found that plutonium (Pu) released from glass waste forms exists predominantly as colloids. If colloidal plutonium could be efficiently filtered in nonwelded bedded units below fractured strata of the repository horizon, it could accumulate sufficient mass for criticality."
The intent of the IRSR appears to be to use the example from Bates et al. (1992) to support plausibility of far field criticality. However, the mechanism Bates et al. outline is extremely improbable. The colloids observed by Bates et al. (1992 and subsequent articles: Feng et al., 1994, pp. 197-205; Buck and Bates, 1999, pp. 635-653) contained only trace Pu; overwhelmingly, the particles were Pu-bearing aluminosilicates or Ca-REE-Th-phosphates. An accumulation of these particles would constitute a dilute, non-critical accumulation of Pu (especially when one considers that the clays could only fill a fraction of the available pore space). If such an efficient clay filtration mechanism existed, the interstices of the non-welded bedded units would already be filled with natural clays. Pu-containing Ca-REE-phosphates will also likely contain neutron poisons (such as gadolinium). In addition, we do not expect future flow patterns to be radically different from flow patterns that have existed over the last 10^5 years or so.

   DOE believes that the largest possible accumulations of 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. Such zones are generally found only in sedimentary rocks, and not in the volcanic rock of Yucca mountain. Nevertheless, an estimate of an upper bound to the probability of occurrence of such a deposit (under very conservative assumptions) has been made and incorporated into an estimate of the upper bound for probability of far-field criticality for commercial spent nuclear fuel (Table 7.6-4 of CRWMS M&O, 1996). This probability leads to an upper bound for the expected number of criticalities of 1.8×10^{-7} for all commercial spent nuclear fuel waste packages for all time.

   DOE plans to confirm the results of this preliminary estimate in future reports. While the study did not address DOE-owned spent nuclear fuel, the amount of such material that could be emplaced in Yucca Mountain is relatively small. DOE expects that similar analyses for DOE-owned SNF will show its emplacement to also be very unlikely to result in far-field criticality, even though some of it is of higher enrichment than commercial spent nuclear fuel.

   Based on the extremely conservative assumptions in the preliminary study and the results showing external criticality to be highly unlikely, DOE suggests that the NRC close the far-field criticality subissue as not being an important part of a Key Technical Issue. If the NRC disagrees regarding the significance of the subissue and chooses to maintain criticality in the far field as a subissue, DOE recommends that text be added to better define the proposed criticality mechanism. A brief scoping calculation in terms of the expected concentration of plutonium in the colloid accumulation would be helpful.

3. Section 3.1, page 7: The Repository Safety Strategy (RSS) (CRWMS M&O) has recently been revised (Revision 3, 2000). The third bullet in this section of the IRSR paraphrases the RSS, Revisions 1&2, by stating an attribute as "slow rate of release of radionuclides from the waste form."

DOE suggests that the NRC revise the bullet to be consistent with the description in Revision 3 of the RSS, which states the attribute as "Low Rate of Radionuclide Release from the EBS." (RSS, Revision 3, Table 3-1, page 3-2)

4. Section 3.2.2.2, page 15, paragraph 2: The sentence at the bottom of the page states that "Movement into these 'dead-end' features in the UZ can be via advection or diffusion." It is unclear from the discussion how advection can occur if the feature is dead-end.

   DOE recommends that this sentence be clarified to specify the role of the dead-end features as they affect the movement of radionuclides by advection.

5. Section 3.2.2.2.2, page 19, last paragraph: A calculation described in this paragraph concludes that peak mean Total Effective Dose Equivalent (TEDE) from NRC colloidal models increases by more than a factor of 60 from the base calculations. However, the description does not state:
   - whether reversibility or irreversibility was assumed.
   - the assumptions made regarding energies of formation.
   - the assumptions made regarding stabilities along the flow path.

   Without knowing the consideration given to these items, it is difficult to evaluate whether the increases of more than a factor of 60 could result from the assumptions described in the text. Increases might also result from not considering formation energies, stability factors, and the potential for reversible sorption onto colloids.

   DOE suggests that additional information on the bases for these calculations be provided. DOE also suggests clarification be provided of how these calculations will be used during the NRC's review of Total System Performance Assessment – Site Recommendation (TSPA-SR) and Total System Performance Assessment – License Application (TSPA-LA).

6. Section 4.1.2.1.2, page 22: The second sentence states that "Only in those rare cases where the radionuclides travel faster than the groundwater would this assumption not be conservative." Based on the results of test results from the C-wells, colloids do not move faster than groundwater, but they may move faster than the average flow rate, as indicated by the following information (Reimus 1999):

   > "The microsphere breakthrough curve, which is corrected for the 3.5 hr delay in their injection time relative to the solutes, is clearly attenuated compared to the solutes. However, it is interesting that their first arrival preceded the solutes, although if the delay in their injection time is not accounted for, the first arrival time is very close to the first solute arrival time. The early microsphere arrival behavior has been observed in other field and laboratory tests in fractured systems (Reimus 1995; Becker et al. submitted 1998), and it may be attributed to a small fraction of the spheres moving rapidly through high velocity streamlines in fractures without having the opportunity to diffuse into low-velocity or stagnant water."

   DOE suggests that the description in the IRSR text be revised to reflect this additional information.

7. Section 4.1.2.2.1, pages 23 and 24, last paragraph with bullets: The paragraph contains inaccuracies in the description of the information in the Viability Assessment (DOE 1998, Vol. 1, App. C). It is not a goal of the unsaturated zone transport test (UZTT) at Busted Butte to develop testing capabilities for possible future experiments beneath the repository horizon (i.e. East-West drift extension). In addition, other UZTT goals stated in these bullets contain different representations of the test goals from those stated in the reference.

   DOE suggests that the NRC clarify the description in the paragraph.

8. Section 4.1.2.2.2.1, page 25, bullet 1: It is not clear from the accompanying text why batch sorption experiments should be carried to steady state. Nor does the text state whether the discussion refers to open- or closed-batch sorption experiments. DOE believes that the description should indicate that K_d has been determined from experiments carried to equilibrium conditions rather than steady-state conditions and that the experiments should be described as closed system experiments. It is important to clarify these conditions in order to determine if previous work is sufficient for meeting this criterion.

   DOE suggests that the NRC clarify the description of conditions under which batch sorption experiments are adequate to describe K_d. DOE suggests the K_d be determined from experiments carried to equilibrium conditions and that the experiments be described as closed system experiments.

9. Section 4.1.2.2.2.1, page 25, bullet 4: DOE does not believe that bounding chemistry of groundwaters from an ambient or perturbed system can be "proved" for a heterogeneous groundwater system that is subject to the potential perturbations from repository operations.

   DOE suggests that the NRC revise this criterion to change "proved" to "demonstrated to be reasonable," or similar language that is more consistent with proposed 10 CFR Part 63 (64 FR 8640).

10. Section 4.1.2.2.2.1, page 25, bullet 7: The meaning of the term *experimental inconsistencies* is not clear. The term could refer to ability to duplicate experiments, inconsistencies in experimental conditions relative to potential site conditions, or uncertainties inherent in the experiment and results.

    DOE suggests that the NRC describe what the term *experimental inconsistencies* is intended to mean and provide examples.

11. Section 4.1.2.2.2.1, pages 26 and 27, Site-Specific Groundwaters: While this section provides a good summary of the variations in groundwater chemistry that exist in a broad region around Yucca Mountain, it tends to overemphasize chemical variations in waters that are quite atypical of the Saturated Zone (SZ) and Unsaturated Zone (UZ) beneath Yucca Mountain. For example, on page 27 is the statement "Total inorganic carbon ranged from 6.8 to more than 10,000 mg/L." The samples from the high end of this range are overwhelmingly brines from playas, and are not representative of deep groundwaters at Yucca Mountain (Turner and Pabalan 1999, p. 380). In addition, many of the seemingly broad chemical variations are really systematic covariations, and are easily handled by sensitivity studies using the EQ6, EQ3NR or PHREEQC codes. For example, if there are obvious covariations among the pH, alkalinity, and

CO_2 fugacity, it is not necessary to vary all three in a sensitivity analysis. The variations in assumed aqueous silica contents are also best handled in sensitivity calculations, since it is often difficult to predict, *a priori*, the conditions that lead to "conservatism."
DOE suggests that text be added to acknowledge (1) that the probable variation in Yucca Mountain groundwater chemistry is much smaller than the southern Nevada ranges cited by the authors; and (2) that many of the variables used to describe groundwater chemistry are dependent (e.g., CO_2 fugacity, alkalinity, and pH). It is somewhat misleading to cite pH, dissolved inorganic carbon, and silica ranges as if they were independent.

12. Section 4.1.2.2.2.1, page 27, paragraph 2: The sentence that begins "Aquifers in the Paleozoic carbonate aquifer..." should be changed to "Water(s) in the Paleozoic carbonate aquifer..." to avoid implying that multiple aquifers have been defined in a single designated aquifer.

13. Section 4.1.2.2.3.2, page 30, paragraph 1: This paragraph addresses the need for demonstrating that ion exchange and adsorption reactions are fast relative to flow velocities. This paragraph does not address the relationship of dose with sorption reaction time. There may be radionuclides with slow sorption times whose contribution to the peak dose is well within the regulatory limits.

    DOE suggests that this paragraph be revised to reflect the fact that the contribution to the peak dose of some radionuclides with slow sorption times is well within the regulatory limits.

14. Section 4.1.2.3, pages 31 and 32: In the discussion of review methods, the IRSR indicates that DOE must demonstrate that the flowpath must act as an isotropic, homogeneous porous medium when process models are used to justify TSPA parameter values. The discussion indicates that the NRC is assuming that the process model applies only to homogeneous, isotropic conditions. If the process model simulates transport through a heterogeneous, anisotropic medium and the properties of the system modeled are represented in the model, there is no need to demonstrate homogeneity and isotropy. Additionally, even if the process model is representing a heterogeneous anisotropic system as a homogeneous, isotropic system and the model can be used to provide bounds on the effective model parameter value, the model could still provide justifiable parameter values (e.g., minimum effective K_d).

    DOE suggests that the NRC relax the guidance to demonstrate that the flowpath must act as an isotropic, homogeneous porous medium when process models are used to justify TSPA parameter values or that further rationale for the requirement be provided.

15. Section 4.1.2.3.2.2, page 38, Ion Exchange...Thermodynamic constants: "These data should be consistent with data used in other aspects of the HLW repository program and internally consistent."

    A unified EQ6 database (data0.ymp), which will also be used for calculations that use the PHREEQC code, is undergoing qualification by a group of scientists. However, care is required in expressing a requirement of "internally consistent," because strict thermodynamic internal consistency is unlikely to be achieved in any database derived from many sources. Furthermore, it is not necessary to demonstrate strict internal consistency to demonstrate conservatism, and at times rejection of "inconsistent data" leads to loss of conservatism. For example, Osthols et al. have determined maximum stability constants for aqueous Th-

phosphate complexes. These constants are not necessarily thermodynamically consistent, but they allow the user to calculate a maximum solubility of Th in phosphate-bearing solutions. In addition, some "consistent" databases (e.g. SUPCRT database) have entries that are consistent but inaccurate (especially at lower temperatures), which cause indirect loss of conservatism. For example, the SUPCRT database information for $BaCO_3$ probably gives witherite an artificially high stability at 25 °C (Stockman et al. 1995), which in turn decreases the stability of $BaCrO_4$, which in turn increases the potential acidity of systems that include weathered stainless steel.

Because strict consistency is often impossible to demonstrate and is at times undesirable for non-academic calculations, a proposed alternative is to use sensitivity studies to determine the effects of database uncertainty. The EQ6 and PHREEQC codes allow testing effects of variations in log K, from the input file, without requiring alterations in the database.

DOE suggests that the NRC consider removing "internally consistent" from the section or note that thermodynamic internal consistency, while desirable in a general sense, is not necessary for a conservative analysis, and is unlikely to be achieved.

16. Section 4.1.2.3.2.2, page 46, first paragraph: The last sentence states that > 99 percent of the Pu was associated with particles ranging from >1μm to <7 nm. This would encompass essentially all sizes.

    DOE suggests that the description be corrected. It appears that the values should be:....>7nm to <1μm.

17. Section 4.2.2.3.1, page 55, criterion 3a: The review method guidance for this criterion, to demonstrate that a portion of the flow path acts as a single continuum porous medium, is somewhat unclear and suggests that the staff may have more extensive work in mind than does DOE. Also, the reference back to criterion 2a in Section 4.1.2.2.1 opens the possibility that the NRC would want results from tracer tests to demonstrate a lack of preferential flow and transport pathways in the alluvial aquifer. The back reference to demonstrations of flow in porous rock could indicate a desire for workscope that DOE believes is unnecessary.

    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. There is no need for quantitative data from tracer tests to demonstrate a lack of preferential flow and transport pathways in the alluvial aquifer. DOE suggests that the discussion be revised to remove the prescriptive guidance.

18. Section 4.2.2.3.2.2, page 59, paragraph 4, line 1: The basis for the statement "the effect of radionuclide concentration will be the same on alluvial radionuclide transport as it is on porous rock radionuclide transport..." is not clear. Alluvium would be expected to have more surface area than porous rock, and alluvium can be reworked to expose fresh surfaces along the groundwater flow path.

    DOE suggests that the NRC clarify the statement.

19. Section 4.3.2.2.1, page 63: The need for DOE to demonstrate the capability to predict breakthrough curves of reactive, nonreactive, and colloidal tracers in field tests, as described in this section of the IRSR, is not clear. 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 a few days. The models are designed to bound the potential transport rates over some 20 km and over periods of 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.

    DOE suggests that, consistent with the intent of proposed 10 CFR Part 63, the discussion be revised to remove the prescriptive guidance regarding demonstrating capability to predict breakthrough curves. As noted in the comment, this work is not clearly related to repository performance.

20. Section 4.3.2.2.1, page 70, bullet for True Colloid Formation: DOE believes that the cited studies overemphasize "true colloid" formation under extreme conditions. Typically, these colloids are produced, experimentally, from gross oversaturation at either very low pH (followed by neutralization), or very high pH (in the latter case, the conditions are high pH and low CO_2 fugacity). The fact that these colloids can remain "stable" for years is not important if there is no realistic path to their formation. DOE knows of no convincing evidence that significant actinide "true colloids" are produced by dissolution of waste forms, in realistic groundwaters, from undersaturation. Unfortunately, the actinide literature uses ambiguous terminology, rarely defining the grain size or stoichiometry. It is probable that many of the reported "colloids" are simply very fine-grained actinide oxyhydroxides which, upon aging, would become coarser and less mobile (e.g. Rai and Ryan, 1982, pp. 213-216). The ionic strength during degradation of Pu-rich waste-forms is likely to be high, and such conditions will be conducive to aggregation and ripening of colloidal materials.

    DOE suggests that descriptions in this section be revised to distinguish between experiments that produce actinide "true colloids" from gross oversaturation or other extreme conditions and experiments that have produced true colloids under conditions one might reasonably expect at Yucca Mountain.

21. Section 4.4.1, page 72, paragraph 2: Item 2 relates to determining the effect on performance of those scenarios that have probabilities greater than 10^{-7}. It is not clear if the probabilities are annual or in 10,000 years. The frequency of occurrence for scenarios related to nuclear criticality due to radionuclide transport should be similar to those established for other postclosure events.

    DOE suggests that the acceptance criterion be revised to clarify that only events that have at least one chance in 10,000 of occurring in 10,000 years need be considered.

22. Section 4.4.2.1, page 73, Radiocolloid formation: "The EQ3/6 database does not contain thermodynamic parameters for colloidal plutonium. How will the mass of plutonium in the groundwater be determined?"

    The EQ3/6 database has an entry for $Pu(OH)_4$ that is typical of very fine-grained Pu hydroxide, and the log K of this solid is easily varied, to more "colloid-like" properties, via the EQ6 augment command. The database contains entries for some Pu-polymeric species, like

$(PuO_2)_4(OH)_7^+$. Also, there are no explicit entries for more typical Pu(IV) polymers because the interpretation of the data is ambiguous (Fuger, 1992, p. 83-84).

As a more important point, it is unlikely that EQ3/6 will be used to determine directly the proportion of Pu colloids. Current plans are to use empirical information from Argonne National Laboratory drip tests and Los Alamos National Laboratory attachment/detachment experiments to determine colloid transport properties. Any role of EQ3/6 will be supportive (e.g., EQ6 may be used to calculate solution properties that appear as independent variables in the colloid models).

DOE suggests that the section be revised to explain what is meant by "colloidal plutonium" in the context of entries for the EQ3/6 database. If the authors mean small hydroxide particles or polynuclear Pu species, the statement is incorrect and should be corrected.

23. Section 5.4.3, page 98, second full paragraph: The last sentence in this paragraph states that interaction between criticality and coupled processes should be considered in future scenario analyses. The description suggests that all interactions between criticality and coupled processes must be considered in scenario analyses regardless of probabilities relating to criticality events. Such analyses would be excessive and could compel DOE to perform scenario analyses for criticality events with much less than 1 chance in 10,000 of occurring in 10,000 years.

    DOE suggests that the acceptance criterion be revised to clarify the frequency of concern for interactions between criticality and coupled processes. (See comment on Section 4.4.1, page 72, paragraph 2.)

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