

APPENDIX C

SCREENING OUT COUPLED THERMAL-HYDROLOGIC-CHEMICAL EFFECTS (RESPONSE TO ENFE 4.03 AND GEN 1.01 (COMMENTS 35 AND 37))

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX C

SCREENING OUT COUPLED THERMAL-HYDROLOGIC-CHEMICAL EFFECTS (RESPONSE TO ENFE 4.03 AND GEN 1.01 (COMMENTS 35 AND 37))

This appendix provides a response for Key Technical Issue (KTI) agreement Evolution of the Near-Field Environment (ENFE) 4.03 and general agreement (GEN) 1.01, comments 35 and 37. These KTI agreements relate to providing the technical basis for the screening of thermal-hydrologic-chemical effects on colloids and colloid transport.

C.1 KEY TECHNICAL ISSUE AGREEMENTS

C.1.1 ENFE 4.03 and GEN 1.01 (Comment 35 and 37)

Agreement ENFE 4.03 was reached during the U.S. Nuclear Regulatory Commission (NRC)/U.S. Department of Energy (DOE) Technical Exchange and Management Meeting on Evolution of the Near-Field Environment held January 9 through 12, 2001, in Pleasanton, California. ENFE KTI subissues 1, 2, 3, and 4 were discussed at that meeting (Reamer 2001).

Agreement GEN 1.01 was reached during the NRC/DOE Technical Exchange and Management Meeting on Range of Thermal Operating Temperatures held September 18 through 19, 2001. At that meeting, NRC provided additional comments relating to ENFE 4.03 (GEN 1.01, comments 35 and 37), and DOE provided an initial response (Reamer and Gil 2001).

The wording of these agreements and of DOE's initial response to the general agreement comments is as follows:

ENFE 4.03

Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids. The DOE will provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids in a new AMR or in a revision to an existing AMR, expected to be available in FY 02.

GEN 1.01 (Comment 35)

The SSPA recommends new values for EBS colloid transport parameters. If these are adopted by TSPA in the future, the technical basis for the new distributions will require close scrutiny. Relevant KTI agreements are RT 3.07, ENFE 4.03, ENFE 4.04, and ENFE 4.06.

DOE Initial Response to GEN 1.01 (Comment 35)

The new values for EBS colloidal transport parameters were designed to evaluate unquantified uncertainty for the SSPA. DOE understands that prior to any potential LA, a stronger technical basis must be provided for EBS colloidal transport parameter values carried forward to the base case analysis.

GEN 1.01 (Comment 37)

The discussion of THC effects on UZ transport does not address chemical effects of the repository. This concern is related to KTI agreements ENFE 4.03 and ENFE 4.06, and TSPAI FEPs item J-8.

DOE Initial Response to GEN 1.01 (Comment 37)

DOE acknowledges this comment and notes that some limited studies were documented in Section 11.3.5.4.2 of SSPA Volume 1. Work is underway, consistent with the cited agreements, to study the effects of alkaline plumes generated by the cement-seepage interactions on rock properties (such as porosity and permeability) and thereby effects on radionuclide transport from the waste placement drifts, with preliminary results expected in FY03.

C.1.2 Related Key Technical Issues

GENERAL 1.01: "For NRC comments 3, 5, 8, 9, 10, 12, 13, 15, 16, 18, 21, 24, 27, 36, 37, 41, 42, 45, 46, 50, 56, 64, 69, 75, 78, 81, 82, 83, 93, 95, 96, 97, 98, 102, 103, 104, 106, 109, 110, 111, 113, 116, 118, 119, 120, 122, 123, 124, and 126, DOE will address the concern in the documentation for the specific KTI agreement identified in the DOE response (Attachment 2). The schedule and document source will be the same as the specific KTI agreement."

TSPAI 2.02: "Provide the technical basis for the screening argument, as summarized in Attachment 2. See Comment #3, 4, 11, 12, 19 (Parts 1, 2, and 6), 25, 26, 29, 34, 35, 36, 37, 38, 39, 42, 43, 44, 48, 49, 51, 54, 55, 56, 57, 59, 60, 61, 62, 63, 64, 65, 66, 68, 69, 70, 78, 79, J-1, J-2, J-3, J-4, J-7, J-8, J-9, J-10, J-11, J-12, J-13, J-14, J-15, J-17, J-20, J-21, J-22, J-23, J-24, J-25, J-26, and J-27.

NRC Comment J-7 on FEP #2.2.08.01.00 (Groundwater chemistry/composition in UZ and SZ). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, RT Subissue 1 Agreement 5, and RT subissue 2 Agreement 10). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-8 on FEP #2.2.08.02.00 (Radionuclide transport occurs in a carrier plume in geosphere). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-10 on FEP #2.2.08.06.00 (Complexation in geosphere). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-11 on FEP #2.2.08.07.00 (Radionuclide solubility limits in the geosphere). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 4 Agreement 3). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-12 on FEP #2.2.10.01.00 (Repository-induced thermal effects in geosphere). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-14 on FEP #2.2.10.07.00 (Thermo-chemical alteration of the Calico Hills unit). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work. DOE also stated that alteration of vitric rock has not been addressed and will need to be included in the overall thermal-hydrological-chemical analyses."

NRC Comment J-15 on FEP #2.2.10.09.00 (Thermal-chemical alteration of the Topopah basal vitrophyre). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

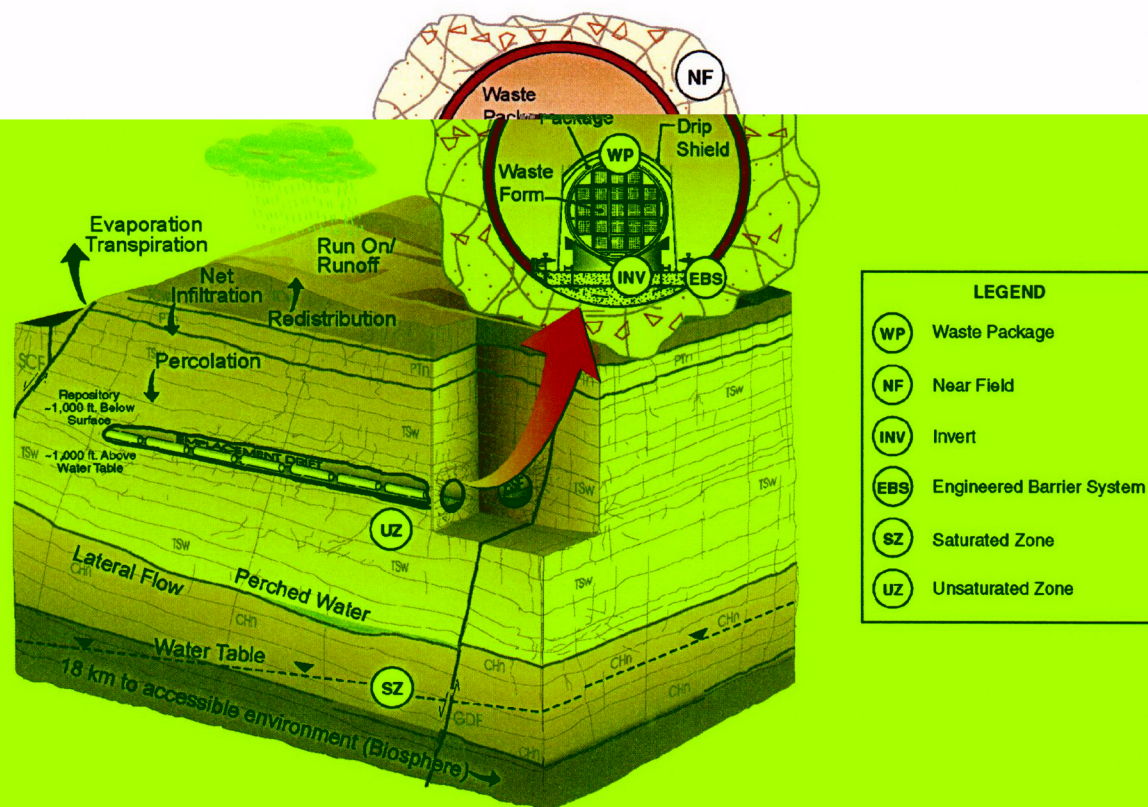
NRC Comment J-21 on FEP #2.2.11.02.00 (Gas Pressure Effects). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreements 5 and 7, and ENFE Subissue 4 Agreement 3). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

NRC Comment J-22 on FEP #1.2.04.02.00 (Igneous activity causes changes to rock properties). NRC/DOE Agreed Path Forward: "This issue is addressed by existing agreements between DOE and NRC (ENFE Subissue 1 Agreement 4, ENFE Subissue 4 Agreements 3 and 4, and RT Subissue 1 Agreement 5). Features, Events, and Processes in UZ Flow and Transport, ANL-NBS-MD-000001 will be revised upon completion of this work."

C.2 RELEVANCE TO REPOSITORY PERFORMANCE

Radionuclide transport as either dissolved species or as colloids represents the means for the release of radionuclides from the repository system to the biosphere. The time and amount of radionuclide release to the biosphere will depend on the transport properties of the dissolved radionuclides and of radionuclide-laden colloids. The transport properties may be dependent on coupled thermal-hydrologic-chemical effects.

The technical basis for the response for this KTI is presented in Sections 3, 4, and 5 of this technical basis document. This KTI is related to the engineered and near-field environment as shown in Figure C-1.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|--|
| NF | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | INV | TSPA1 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| NF | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | EBS UZ SZ | TSPA1 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| NF | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | INV UZ SZ | TSPA1 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| WP | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | SZ | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| WP | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| WP | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure C-1. Mapping of Colloid-Related Key Technical Issue Agreements to Repository System Components

C.3 RESPONSE

Thermal effects may affect radionuclide transport directly by causing changes in radionuclide speciation and solubility in the unsaturated zone or indirectly by causing changes in the host rock mineralogy that affect the flow path. Relevant processes include volume effects associated with silica phase changes, precipitation and dissolution of fracture-filling minerals (including silica and calcite), and alteration of zeolites and other minerals to clays. The effects of colloid formation are accounted for in the colloid source term. Colloids are expected to be formed from the degradation of the high-level radioactive waste and spent nuclear fuel waste forms, engineered barrier system materials, and rock. Radionuclides associated with colloids are assumed to be either irreversibly or reversibly attached to colloids (CRWMS M&O 2000a, Section 6; CRWMS M&O 2000b, Section 6). The near-field thermal-chemical analysis indicates only small changes in hydrologic properties and mineralogy as a result of these coupled processes (BSC 2003a, Section 6). Therefore, far-field changes are likewise expected to be small, including mineral precipitation/dissolution and alteration of minerals such as zeolites and clays. Therefore, coupled thermal-hydrologic-chemical effects on radionuclide transport properties and colloids are excluded from TSPA on the basis of low consequence.

Cementitious materials will not be used in the emplacement drift ground support (BSC 2003b). Therefore, the effects of alkaline plumes generated by cement-seepage interactions on rock properties and radionuclide transport in the vicinity of the emplacement drift are no longer an issue that needs to be considered.

Coupled thermal-hydrologic-chemical effects are likely to result in unstable colloid suspensions and, therefore, reduce the concentration of colloids in suspension. With increasing temperature and increasing ionic strength, both conditions expected as a result of thermal-hydrologic-chemical effects, colloid suspensions become less stable. High temperatures and high ionic strengths are not favorable to colloid transport. Therefore, the screening out of coupled thermal-hydrologic-chemical effects on the transport of radioactive colloids in the TSPA is justified.

New values for the diffusivity of dissolved species through the invert are presented in *EBS Radionuclide Transport Abstraction* (BSC 2003c, Section 6.3.4.1). From the dissolved species diffusivity values, diffusivities for colloids are estimated.

The information in this report is responsive to agreements ENFE 4.03 and GEN 1.01 comments 35 and 37 made between the DOE and NRC. The report contains the information that DOE considers necessary for NRC review for closure of these agreements.

C.4 BASIS FOR THE RESPONSE

Features, Events and Processes in UZ Flow and Transport (BSC 2003d, Section 6.8.7) presents the screening argument for excluding coupled thermal-hydrologic-chemical effects on radionuclide transport properties and colloids (FEP 2.2.10.06.0A; FEP 2.2.08.03.0B) in the unsaturated zone (solubility, speciation, phase changes, and precipitation/dissolution). This FEP is conservatively ignored with respect to solubility reduction in the far field, and the effects of radionuclide precipitation are ignored in the unsaturated zone transport for TSPA. If solubility

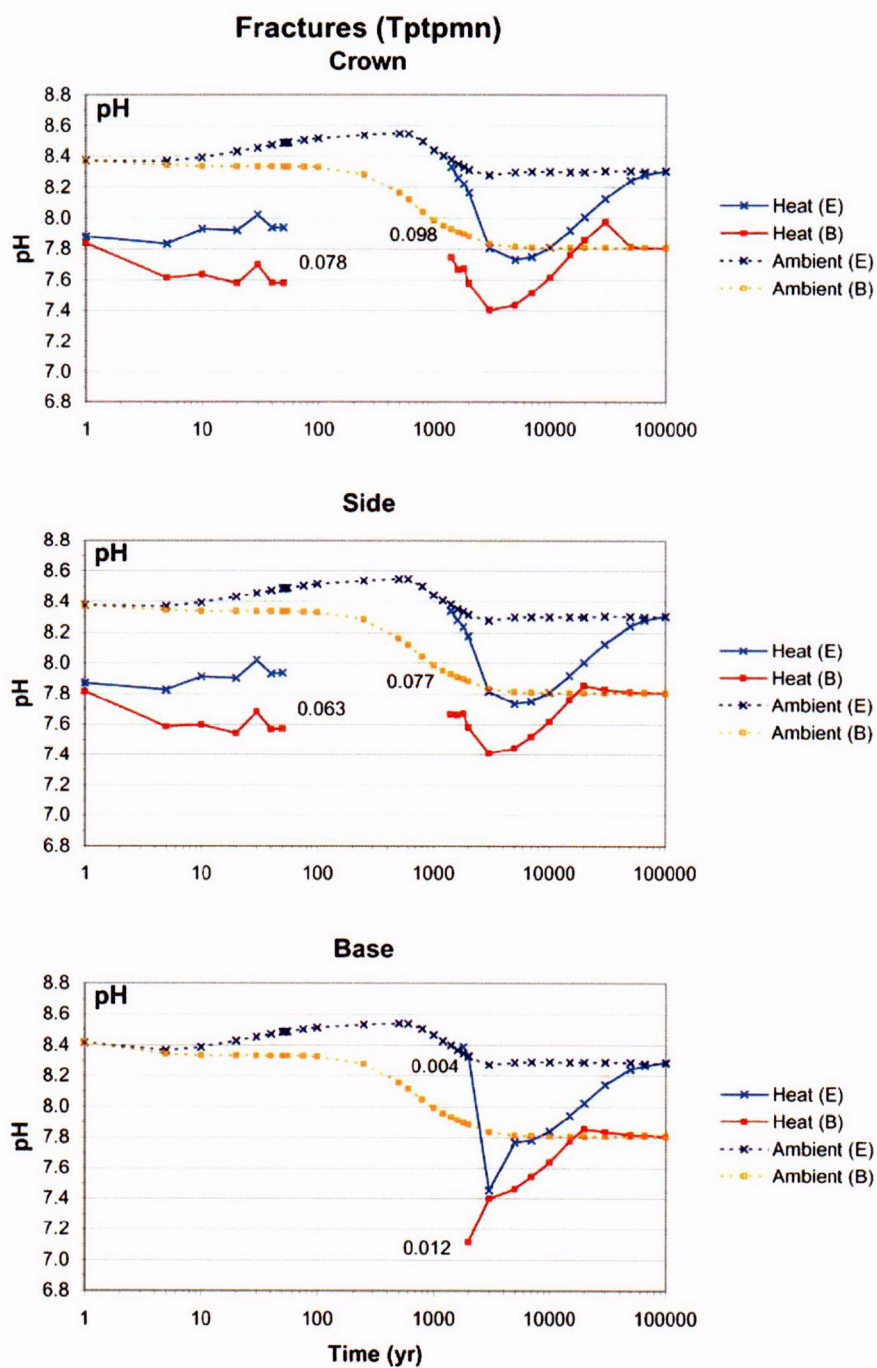
limits increase in the geosphere compared with the waste emplacement drift, there is no effect on transport because all available radionuclides from the source at the waste emplacement drift are already aqueous species. The thermal-chemical effects on colloid formation have already been accounted for in the colloid source term (BSC 2003e). Therefore, any impact from thermal-chemical alteration on colloid entrainment will be insignificant with respect to total repository system performance.

This screening argument is reasonable because in the rock mass around the repository the temperature may be relatively high even at 10,000 years. For example, in Section 6.5.2 in *Drift-Scale Coupled Processes (DST and THC Seepage) Models* (BSC 2003a) it is shown that after 10,000 years the temperature is 45° to 50°C, which is significantly higher than the initial temperature of around 25°C. This higher temperature tends to decrease the stability of a colloidal suspension, as discussed in *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003e).

The diffusion coefficient for dissolved species has been determined specifically for crushed tuff invert materials as a function of volumetric moisture content (m^3 water/ m^3 bulk rock), using electric conductivity measurements (BSC 2003c).

The thermal-hydrologic-chemical model simulations have been performed for both the base case and the extended case and for both Tptpmn and Tptpll geologic formations (BSC 2003a). The extended case includes the major solid phases (minerals and glass) encountered in geologic units at Yucca Mountain, together with a range of possible reaction product minerals, CO_2 gas, and the aqueous species necessary to include these solid phases and the pore-water composition within the thermal-hydrologic-chemical model. The base case is a subset of the extended case excluding aluminum silicate minerals, which form or dissolve much less easily than minerals such as calcite or gypsum. The results for the Tptpmn rock unit are shown in Figures C-2 and C-3. The pH in fracture waters varies within 7.4 to 8.6 but still remain within a neutral to mildly basic range over the 10,000-year regulatory time period (Figure C-2), and thus its effect on colloid stability is expected to be negligible. The concentrations of soluble components (e.g., Na and Cl) and therefore the ionic strength in the waters are predicted to increase over the time period of thermal event (Figure C-4), which will reduce colloid stability. The THC effect may also change the transport properties of rock. However, Figure C-3 shows that this change is small and can be negligible for colloid transport.

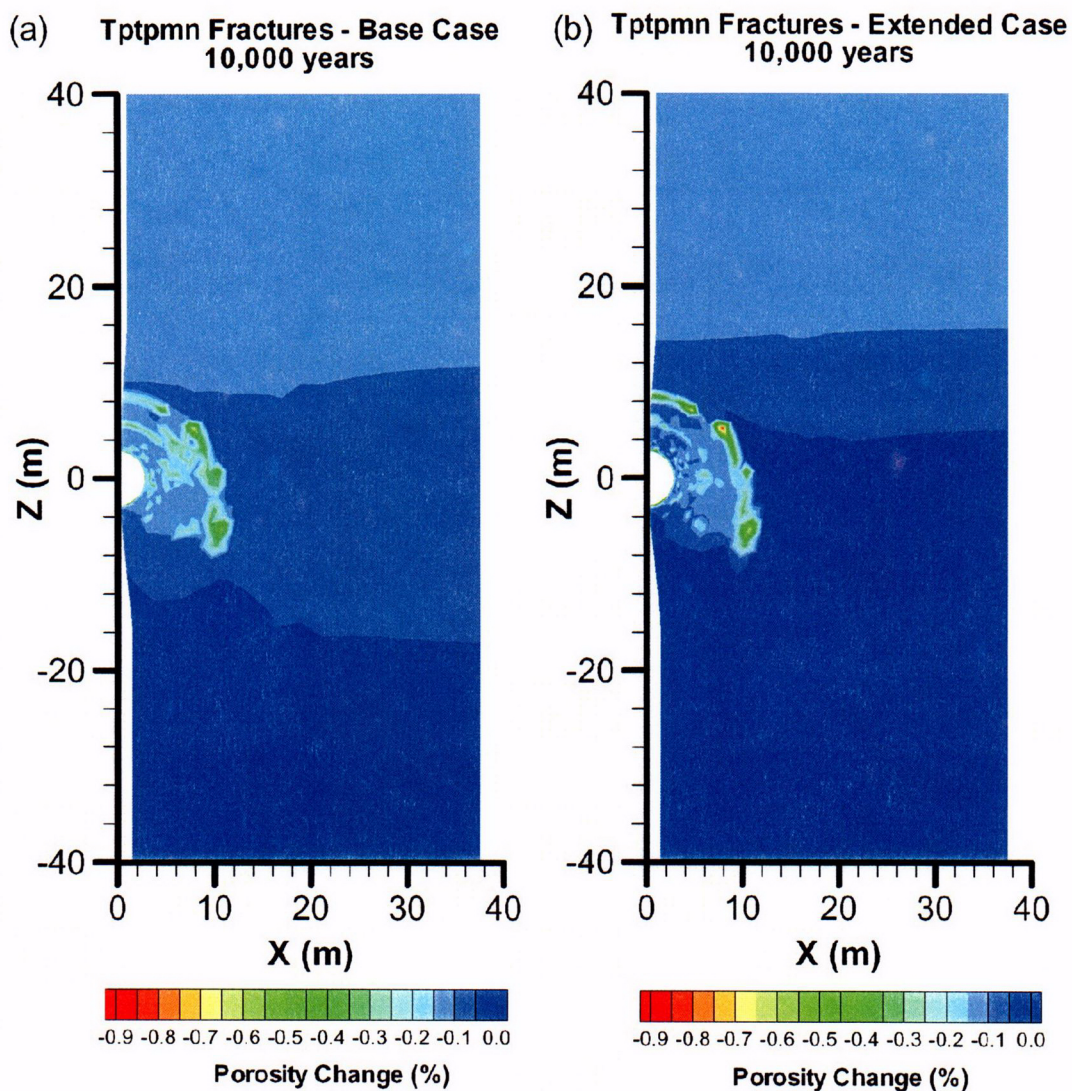
Figure C-5 presents the uncertainty in the statistical fit for the diffusion coefficient.



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Source: BSC 2003a.

Figure C-2. Thermal-Hydrologic-Chemical Simulation (Tptpmn Model): Time Profiles of the Modeled pH of Fracture Water at Three Drift-Wall Locations under Heating (Heat) and Nonheating (Ambient) Conditions for the Extended (E) and Base-Case (B) Geochemical Systems

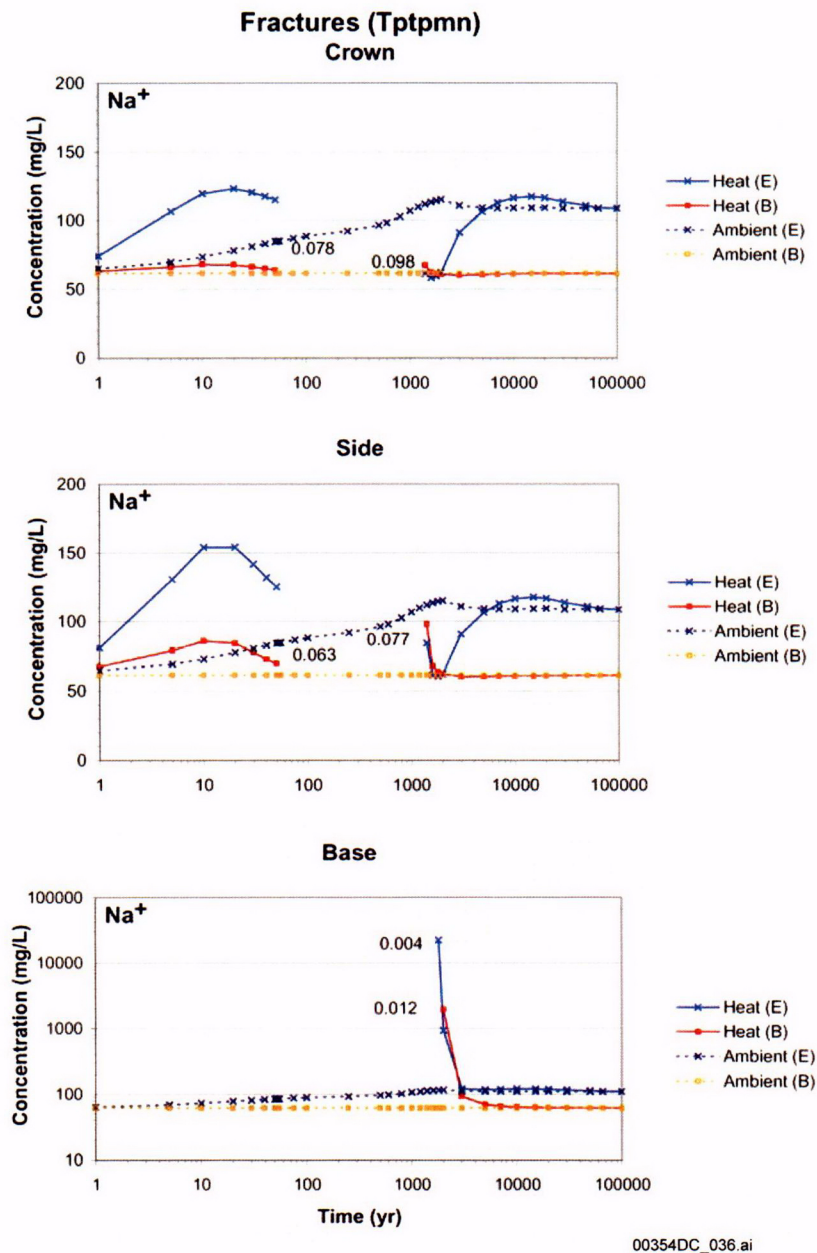


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Source: BSC 2003a.

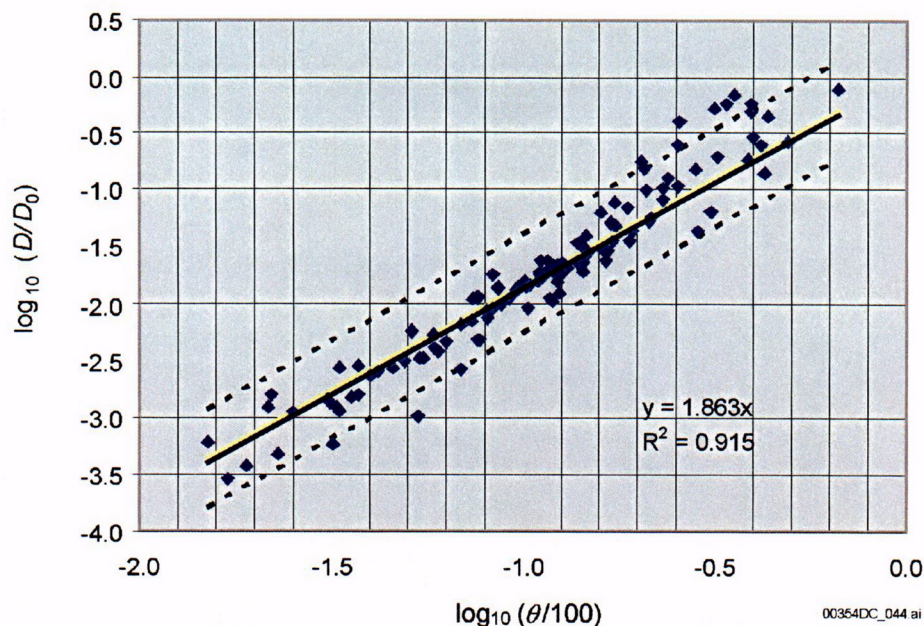
NOTE: Decrease in porosity is primarily due to the precipitation of calcite and amorphous silica.

Figure C-3. Thermal-Hydrological-Chemical Model Simulation (Tptpmn Model): Contour Plot of Modeled Fracture Porosity Change at 10,000 Years for (a) Base-Case and (b) Extended Geochemical Systems



Source: BSC 2003a.

Figure C-4. Thermal-Hydrologic-Chemical Simulation (Tptpmn Model): Time Profiles of Modeled Total Aqueous Sodium Concentrations in Fracture Water at Three Drift-Wall Locations under Heating (Heat) and Nonheating (Ambient) Conditions for Extended (E) and Base-Case (B) Geochemical Systems



Source: BSC 2003c, Figure 7.

NOTE: The dashed lines correspond to two standard deviations above and below the statistical fit to the data. D is the diffusion coefficient of the crushed invert materials, D_0 is the diffusion coefficient of the fully saturated crushed invert materials, and θ is the percentage of saturation.

Figure C-5. Uncertainty in the Statistical Fit for the Diffusion Coefficient

The colloidal diffusion coefficient can be estimated from the following Stokes-Einstein relationship (BSC 2003c):

$$D_{coll} = D_{ion} \left(\frac{r_{ion}}{r_{coll}} \right) \quad (\text{Eq. C-1})$$

where D_{coll} is the diffusion coefficient for a colloidal particle of radius r_{coll} and D_{ion} is the diffusion coefficient of a ion of radius r_{ion} . Given a typical ion radius and colloidal particle radius of 0.1 and 1 nm, respectively, the diffusion coefficient of a colloidal particle is generally 100 times smaller than that of a dissolved ion.

C.5 REFERENCES

BSC (Bechtel SAIC Company) 2003a. *Drift-Scale Coupled Processes (DST and THC Seepage) Models*. MDL-NBS-HS-000001 REV 02. Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20030804.0004.

BSC 2003b. *Repository Design Project, Repository/PA IED Emplacement Drift Committed Materials*. 800-IED-EBS0-0030-000-00A. Las Vegas, Nevada: Bechtel SAIC Company. ACC: ENG.20030311.0021.

BSC 2003c. *EBS Radionuclide Transport Abstraction*. ANL-WIS-PA-000001 REV 01F. Las Vegas, Nevada: Bechtel SAIC Company. ACC: MOL.20030922.0199.

BSC 2003d. *Features, Events and Processes in UZ Flow and Transport*. ANL-NBS-MD-000001 REV 02B. Las Vegas, Nevada: Bechtel SAIC Company. ACC: MOL.20030922.0198.

BSC 2003e. *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary*. MDL-EBS-PA-000004 REV 00. Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20030626.0006.

CRWMS M&O (Civilian Radioactive Waste Management System Management and Operating Contractor) 2000a. *Particle Tracking Model and Abstraction of Transport Processes*. ANL-NBS-HS-000026 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000502.0237.

CRWMS M&O 2000b. *UZ Colloid Transport Model*. ANL-NBS-HS-000028 REV 00. Las Vegas, Nevada: CRWMS M&O. ACC: MOL.20000822.0005.

Reamer, C.W. 2001. U.S. Nuclear Regulatory Commission/U.S. Department of Energy Technical Exchange and Management Meeting on Evolution of the Near-Field Environment (January 9–12, 2001). Letter from C.W. Reamer (NRC) to S. Brocoum (DOE/YMSCO), January 26, 2001, with enclosure. ACC: MOL.20010810.0033.

Reamer, C.W. and Gil, A.V. 2001. Summary Highlights of NRC/DOE Technical Exchange and Management Meeting of Range on Thermal Operating Temperatures, September 18-19, 2001. Washington, D.C.: U.S. Nuclear Regulatory Commission. ACC: MOL.20020107.0162.

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APPENDIX D

**CONTRASTING COLLOID CONCENTRATIONS
IN THE ENGINEERED BARRIER SYSTEM AND SATURATED ZONE
(RESPONSE TO TSPAI 3.30 AND GEN 1.01 (COMMENTS 43 AND 46))**

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX D

CONTRASTING COLLOID CONCENTRATIONS IN THE ENGINEERED BARRIER SYSTEM AND SATURATED ZONE (RESPONSE TO TSPAI 3.30 AND GEN 1.01 (COMMENTS 43 AND 46))

This appendix provides a response for Key Technical Issue (KTI) agreement Total System Performance Assessment and Integration (TSPAI) 3.30 and general agreement (GEN) 1.01, comments 43 and 46. These agreements relate to providing the technical basis for concentrations of colloids available for reversible attachment.

D.1 KEY TECHNICAL ISSUE AGREEMENTS

D.1.1 TSPAI 3.30 and GEN 1.01 (Comments 43 and 46)

Agreement TSPAI 3.30 was reached during the NRC/DOE technical exchange and management meeting on total system performance assessment and integration held August 6 through 10, 2001, in Las Vegas, Nevada. TSPAI KTI subissues 1, 2, 3, and 4 were discussed at that meeting (Reamer 2001).

Agreement GEN 1.01 was reached during the NRC/DOE technical exchange and management meeting on range of thermal operating temperatures held September 18 through 19, 2001. At that meeting, NRC provided additional comments (GEN 1.01, comments 43 and 46) relating to TSPAI 3.30, and DOE provided an initial response to those comments (Reamer and Gil 2001).

The wording of these agreements and of DOE's initial response to the general agreement comments is as follows:

TSPAI 3.30

Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on K_c . Update, as necessary, the K_c parameter as new data become available from the Yucca Mountain region ...

DOE will provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the engineered barrier system and the saturated zone. The sensitivity analyses planned in response to RT Agreement 3.07 will address the effect of colloid concentration on the K_c parameter. The technical basis will be documented in the Waste Form Colloid Associated Concentration Limits: Abstractions and Summary (ANL-WIS-MD-000012) in FY 2003. The K_c parameter will be updated as new data become available from the Yucca Mountain region in the Uncertainty Distribution for Stochastic Parameters AMR (ANL-NBS-MD-000011) in FY 2003.

GEN 1.01 (Comment 43)

The SSPA presents a new distribution for retardation of colloids with irreversibly-attached radionuclides. The distribution takes into account new site-specific alluvium data. However, any future use of this distribution in TSPA will require comparison with results of field and laboratory tests. This concern is indirectly related to agreement TSPAI 3.30.

DOE Initial Response to GEN 1.01 (Comment 43)¹

DOE acknowledges that any future use of this distribution in TSPA will require comparison with results of field and laboratory tests 1,2. This concern is indirectly related to KTI agreements RT 3.07 and RT 3.08. Laboratory testing of microsphere and silica colloid retardation in alluvium-packed columns is in progress. Microspheres will be used as colloid tracers in ATC cross-hole tracer testing.

GEN 1.01 (Comment 46)

The analysis of sensitivity to increased uncertainty in the reversible colloid parameter K_c (Section 12.5.2.4) yielded "somewhat longer transport times" in the saturated zone. This analysis does not illustrate the effect of possibly underestimating K_c , because it is not clear that the mean value of K_c is significantly different from the base case. This concern is related to agreements RT 3.07 and TSPAI 3.30.

DOE Initial Response to GEN 1.01 (Comment 46)

This issue will be handled as part of agreements RT 3.07 and TSPAI 3.30.

D.1.2 Related Key Technical Issue Agreements

None.

D.2 RELEVANCE TO REPOSITORY PERFORMANCE

Radionuclide transport as either dissolved species or colloids represents the means for the release of radionuclides from the repository to the biosphere. The concentration of colloids released from the engineered barrier system provides a source term for colloid transport within the unsaturated zone. In turn, the colloid concentration reaching the water table provides the source term for colloid transport in the saturated zone. The colloid concentration (m_c) and the distribution coefficient (K_d) are two important parameters controlling colloid-facilitated radionuclide transport because K_c , the ratio of radionuclide mass in colloids to the concentration in solution, is defined as the product of K_d and m_c .

¹ "[F]ield and laboratory tests 1, 2" refers to test conducted using CML microspheres as colloid surrogates. These tests will be discussed in detail in Appendix M (Response to KTI Agreement RT 3.08) of the saturated zone technical basis document.

The technical basis for the response for this KTI is presented in Sections 3, 4, 5 and 6 of this technical basis document. This KTI is related to the engineered and near-field environment, the unsaturated and the saturated zones as shown in Figure D-1.

D.3 RESPONSE

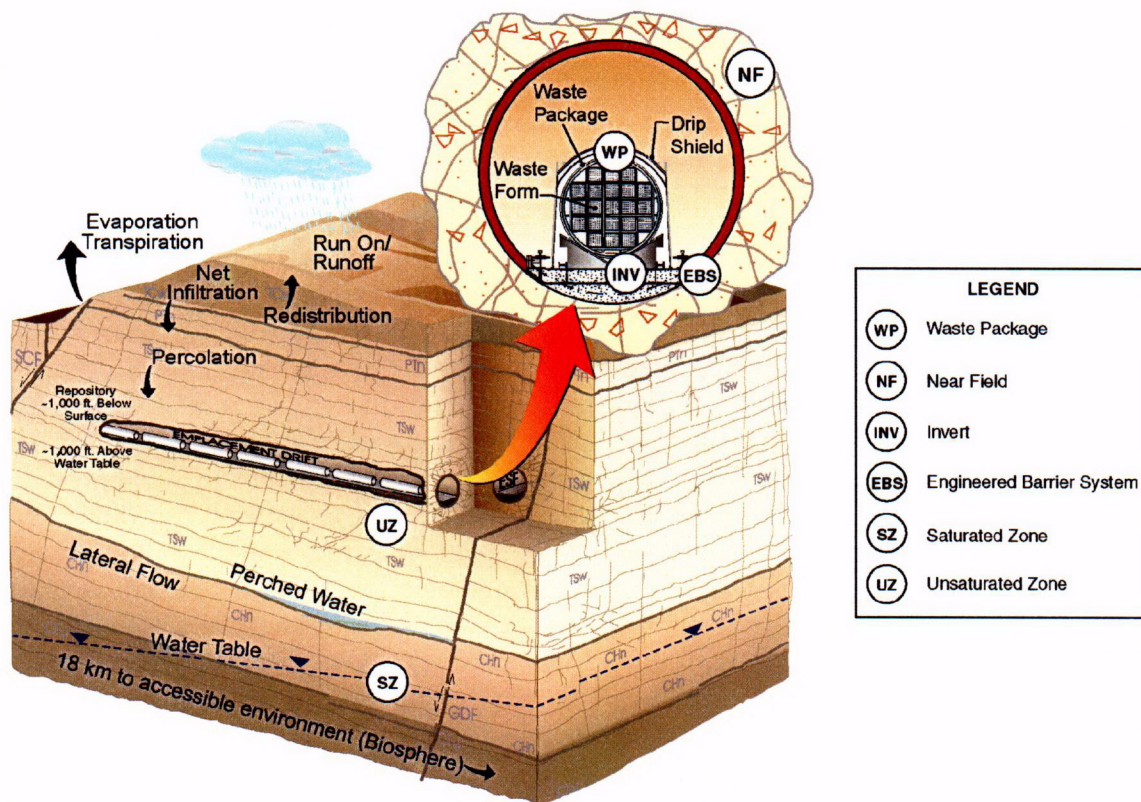
D.3.1 Response to Key Technical Issue Agreement TSPAI 3.30

In contrast to the constraints on colloid concentrations used in the total system performance assessment (TSPA) for the site recommendation to represent reversible radionuclide attachment, the engineered barrier system and the saturated zone colloid models use the same distribution of colloidal concentration for natural system colloids. This distribution has been revised since the TSPA for the site recommendation with additional data that raise the upper limits of colloid concentration by about two orders of magnitude over the previous values considered. This translates to about a four-order-of-magnitude increase for the saturated zone colloid treatment from the TSPA for the site recommendation that used only the mean value of colloid concentration. This change in colloid concentration is reflected directly in changes to the K_c parameter used in the saturated zone colloid treatment. Such increased concentration limits for colloids that can reversibly sorb radionuclides in the saturated zone provides a higher level of confidence that the evaluation of saturated zone radionuclide transport by reversible colloids is bounding. This increased level of consistency for colloid concentrations in the saturated zone does not mean that the treatment is identical to that for the engineered barrier system. The differences and the respective technical bases for the colloid concentrations in the engineered barrier system and saturated zone are discussed below.

Within the engineered barrier system, the concentration of colloids available for reversible sorption, m_c , is evaluated for the effects of temperature and the chemical environment. Such effects are not expected within the saturated zone, and therefore the colloid concentration is unaffected by them. In the saturated zone, colloid concentration is affected by attachment/detachment of the colloids to the immobile rock surfaces.

It is expected that under the high-temperature and high ionic strengths conditions within the engineered barrier system, particularly in the first 2,000 to 3,000 years after closure, the concentration of colloids will not be sufficiently high to effectively compete with the immobile rock surfaces for the reversible attachment of dissolved radionuclides. Colloids leaving the engineered barrier system will travel through the unsaturated zone to reach the saturated zone. Within the unsaturated zone, colloid transport is expected to be primarily through fractures, and only those colloids with characteristic dimension considerably smaller than the film coating the fracture surfaces are expected to be transported. Colloids will be subjected to retardation effects, thus increasing their transport time through the unsaturated zone. Larger colloids are expected to be subjected to filtration by film straining. As colloids travel through the unsaturated zone within the Topopah Spring Tuff, they will reach the Calico Hills Formation, which has a significantly different porosity. The interface between the Topopah Spring and the Calico Hills units presents a barrier to colloid transport. At that interface colloids may move laterally until they find another fracture system to continue traveling vertically towards the saturated zone. Therefore, the concentration of colloids reaching the saturated zone is expected to be significantly lower than that within the engineered barrier system. Because the only direct

source of colloids in the saturated zone is the natural system materials, they are the focus of the reversibly sorbing colloids in the treatment for this subsystem.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|--|
| (NF) | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | (INV) | TSPAI 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| (NF) | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | (EBS) (UZ) (SZ) | TSPAI 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| (NF) | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | (INV) (UZ) (SZ) | TSPAI 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| (WP) | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | (SZ) | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| (WP) | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| (WP) | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure D-1. Mapping of Colloid-Related Key Technical Issues Agreement to Repository System Components

D.3.2 Response to Agreement GEN 1.01 (Comments 43 and 46)

The saturated zone transport simulations of radionuclides that are irreversibly attached to colloids are conducted for radioisotopes of plutonium and americium. The retardation of colloids with irreversibly attached radionuclides is a kinetically controlled process, which approaches equilibrium behavior for long transport times. For transport of colloids through the saturated zone, equilibrium behavior is nearly achieved. However, nonequilibrium behavior results in unimpeded migration of some of the colloids. Consequently, a small fraction of these colloids are transported through the saturated zone with no retardation; whereas the larger fraction is delayed by a retardation factor. The saturated zone transport simulations of radionuclides that are reversibly attached to colloids are conducted for radioisotopes of plutonium, americium, thorium, protactinium, and cesium. In the simulations, the distribution coefficient (K_c), a product of sorption coefficient K_d and colloid concentration, is sampled based on the distributions of both K_d values and colloid concentrations. The simulations show a significant retardation of radionuclide transport in the saturated zone as compared to the case where radionuclides irreversibly sorbed on colloids.

The information in this report is responsive to agreements TSPAI 3.30 and GEN 1.01 comments 43 and 46 made between the DOE and NRC. The report contains the information that DOE considers necessary for the NRC to review for closure of these agreements.

D.4 BASIS FOR THE RESPONSE

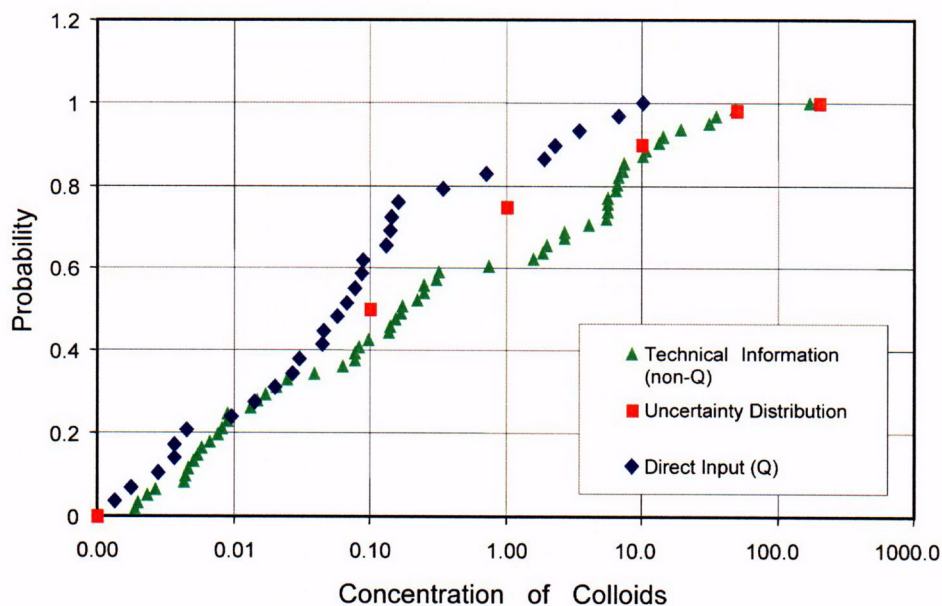
D.4.1 Response to Key Technical Issue Agreement TSPAI 3.30

Reversible Colloid Concentrations in the Engineered Barrier System—The technical basis for selection of typical colloid concentrations in the engineered barrier system and the saturated zone are discussed in *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003a) and summarized below:

- The static-saturated tests for Defense High-Level Radioactive Waste Glass at Argonne National Laboratory indicated that colloids developed and increased in concentration with time, up to the point where the colloid concentration reached a maximum value and then the colloidal suspension became less stable because of high ionic strengths resulting in agglomeration and settling (BSC 2003a). The maximum colloid concentration from the experimental degradation of high-level radioactive waste glass is estimated to be 5 ppm (BSC 2003a, Figure 3).
- The concentration range of iron oxyhydroxide colloids generated from the defense high-level radioactive waste glass was estimated based on experiments performed at the University of Nevada at Las Vegas and on professional judgment. In these experiments, scaled-down miniature waste packages were exposed to J-13 groundwater in either a bath-tub mode or a flow-through mode. The cumulative results have yielded average concentrations of colloidal size materials in the range of 20 mg/L within the initial four weeks of the experiments (BSC 2003a, p. 56).

- The range of colloid concentration in seepage/groundwater was derived based on literature data and groundwater sampling from the Yucca Mountain area and Idaho National Engineering and Environmental Laboratory (BSC 2003a, Figures 11 and 12). Practically no colloids were detected for the ionic strength above 0.05 M. The upper limit of colloid concentration is about 200 ppm, with average values of approximately 0.1 ppm (Figure D-2).
- Results from the unsaturated testing of commercial spent nuclear fuel and defense spent nuclear fuel at Argonne National Laboratory indicated the formation of alteration products containing very low concentrations of uranium-based colloids and dissolution of uranium-based colloids in a few months (BSC 2003a, Section 6.3.1.2).

For purposes of this discussion, colloid concentration within the engineered barrier system will consist of colloid concentrations within the waste package (Section 3 of this technical basis document) and the invert (Section 4 of this technical basis document).



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Source: BSC 2003a, Figure 12.

Figure D-2. Cumulative Distribution Function Showing the Probability of Occurrence of Colloid Concentration Levels (ppm or mg/L) in Groundwater Samples in the Yucca Mountain Area and Idaho National Engineering and Environmental Laboratory

Within the waste package, the concentration of three different types of colloids available for reversible sorption are estimated (see Figure 3-11 of this technical basis document): (1) reversible defense high-level radioactive waste glass colloids (also referred to as waste form colloids), (2) reversible iron oxyhydroxide colloids, and (3) reversible groundwater colloids. In general, the concentration of each of these three types of colloids are calculated at each time step

of the simulation in TSPA. At each time step, (1) the colloid mass available for reversible sorption is estimated as a function of ionic strength, (2) the stability of colloids as a function of ionic strength and pH is determined, and (3) the sorption of available mass of the dissolved inventory of cesium, americium, plutonium, protactinium, and thorium onto colloids is calculated. Colloids are assumed to move unretarded from the waste package to the invert.

Colloids are not assumed to form within the invert. First, the invert is divided into two pore spaces (see Section 4.3.2 of this technical basis document). As colloids are released from the waste package into the invert, the released colloids are divided into two fractions (see Figure 4-2 of this technical basis document): (1) the inter-granular pore space fraction and (2) the intragranular pore space fraction. No mass transfer of colloids is assumed to occur within these two pore spaces. The distribution of colloids between the two pore spaces depends on the particular scenario (nominal scenario, seismic scenario, or igneous scenario) being simulated. Colloids entering the intra-granular pore space are transported by diffusion as defined by the chemistry (ionic strength and pH) of the tuff matrix. The colloids entering the inter-granular pore space are transported by advection and diffusion as defined by the chemistry of the fractures. The stability of the colloids is reevaluated using the same ionic strength and pH relationships for the three types of reversible colloids shown in Figure 3-11 of this technical basis document.

The cumulative distribution function of colloid concentration for natural groundwater colloids is shown in Figure D-2. In the calculations summarized above, this cumulative distribution function is sampled and modified based on the chemical conditions within the waste package and the invert.

In the TSPA, the transport model for the engineered barrier system explicitly accounts for the partitioning coefficient (K_d) and the colloid concentration. Updated K_d values are tabulated in Table D-1. From the values of K_d and the colloid concentration, values of K_c are calculated as the product of K_d and colloid concentration.

The K_c approach is used for both the unsaturated zone and saturated zone colloid transport. The same concentration distribution of smectite colloids in Figure D-2 is used for the engineered barrier system, unsaturated zone, and saturated zone colloid models. Note that in the engineered barrier system this distribution of smectite colloid concentration is reevaluated using the pH and ionic strength values, while for the unsaturated zone and saturated zone colloid transport models the distribution is assumed to be unaffected by temperature or chemical effects.

As discussed in Section 3.2.5 of this technical basis document, the thermal and chemical perturbations within the engineered barrier system are likely to destabilize colloid suspensions by increasing Brownian motion and particle collision due to the elevated temperature and increasing ionic strength of the solution. Therefore, the bounding values summarized above are still suitable for the engineered barrier system. In addition, any perturbed colloid concentration inside the drift will ultimately attenuate to the background concentration in the Yucca Mountain groundwater, as the colloidal solution percolates through the invert and the unsaturated zone into the saturated zone, and the water chemistry converges to the ambient groundwater. As a result, the total system performance becomes insensitive to the near-field thermal and chemical perturbations.

Table D-1. K_d Values (mL/g) Used for Reversible Radionuclide Sorption on Colloids in Total System Performance Assessment-License Application Calculations

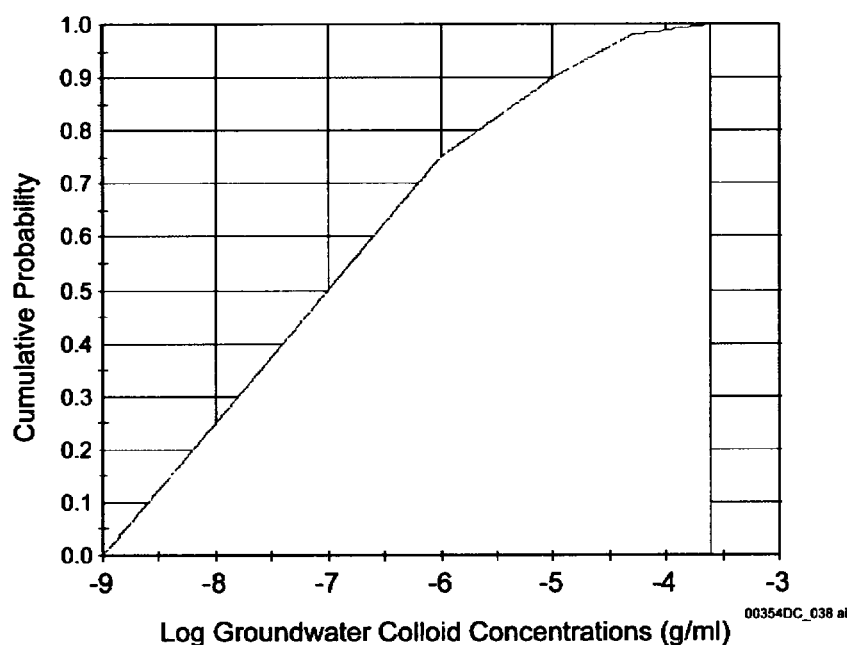
| Radionuclide | Colloid | K_d Value Range (mL/g) | K_d Value Intervals (mL/g) | K_d Value Interval Probabilities |
|--------------|-------------------|--------------------------|--|---|
| Pu | Iron Oxyhydroxide | 10^4 to 10^6 | $< 1 \times 10^4$ 1×10^4 to 5×10^4 5×10^4 to 1×10^5 1×10^5 to 5×10^5 5×10^5 to 1×10^6 $> 1 \times 10^6$ | 0 0.15 0.2 0.5 0.15 0 |
| | Smectite | 10^3 to 10^6 | $< 1 \times 10^3$ 1×10^3 to 5×10^3 5×10^3 to 1×10^4 1×10^4 to 5×10^4 5×10^4 to 1×10^5 1×10^5 to 5×10^5 5×10^5 to 1×10^6 $> 1 \times 10^6$ | 0 0.04 0.08 0.25 0.2 0.35 0.08 0 |
| Am, Th, Pa | Iron Oxyhydroxide | 10^5 to 10^7 | $< 1 \times 10^5$ 1×10^5 to 5×10^5 5×10^5 to 1×10^6 1×10^6 to 5×10^6 5×10^6 to 1×10^7 $> 1 \times 10^7$ | 0 0.15 0.2 0.55 0.1 0 |
| | Smectite | 10^4 to 10^7 | $< 1 \times 10^4$ 1×10^4 to 5×10^4 5×10^4 to 1×10^5 1×10^5 to 5×10^5 5×10^5 to 1×10^6 1×10^6 to 5×10^6 5×10^6 to 1×10^7 $> 1 \times 10^7$ | 0 0.07 0.1 0.23 0.2 0.32 0.08 0 |
| Cs | Iron Oxyhydroxide | 10^1 to 10^3 | $< 1 \times 10^1$ 1×10^1 to 5×10^1 5×10^1 to 1×10^2 1×10^2 to 5×10^2 5×10^2 to 1×10^3 $> 1 \times 10^3$ | 0 0.13 0.22 0.55 0.1 0 |
| | Smectite | 10^2 to 10^4 | $< 1 \times 10^2$ 1×10^2 to 5×10^2 5×10^2 to 1×10^3 1×10^3 to 5×10^3 5×10^3 to 1×10^4 $> 1 \times 10^4$ | 0 0.2 0.25 0.5 0.05 0 |

Source: BSC 2003a, Table 10.

NOTE: The K_d values for Tc and I are very low and not listed here.

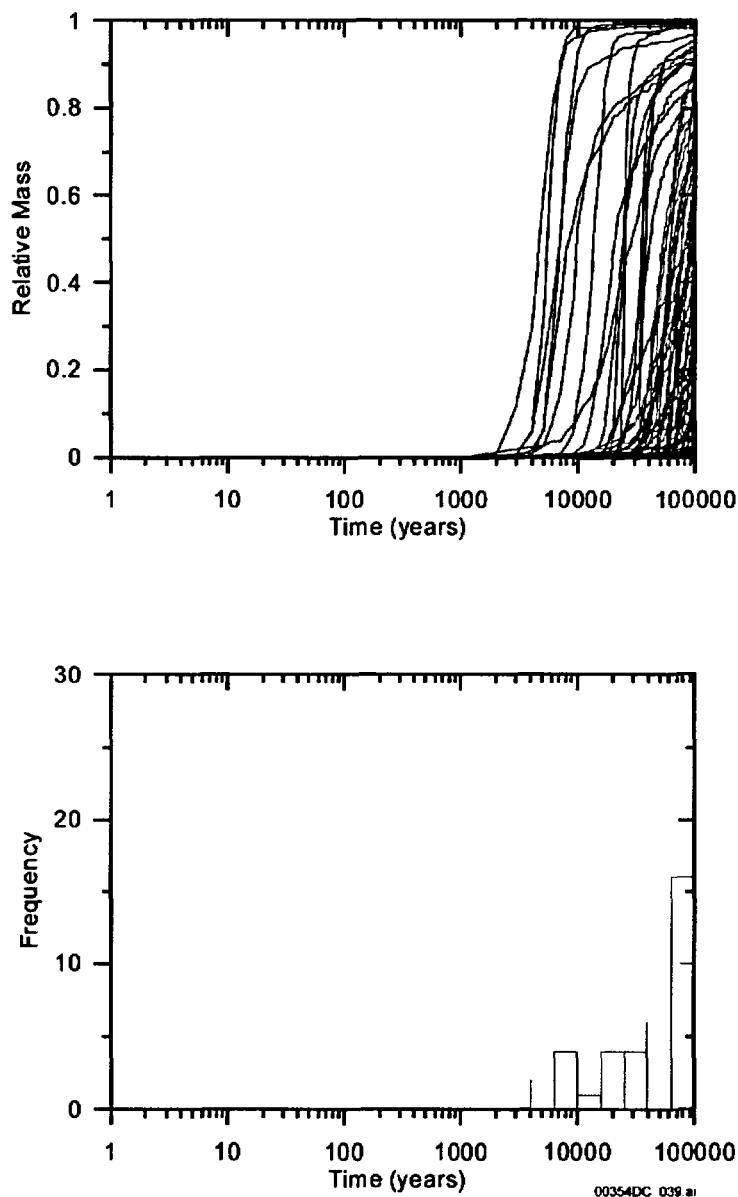
Concentration of Reversible Colloids in the Saturated Zone—Radionuclides that are reversibly sorbed onto colloids are modeled to be temporarily attached to the surface of colloids. Thus, these radionuclides are available for dissolution in the aqueous phase and their transport characteristics are a combination of the transport characteristics of solute and colloids. The saturated zone transport simulations of radionuclides that are reversibly attached to colloids are conducted for radioisotopes of plutonium, americium, thorium, protactinium, and cesium (BSC 2003b). (Note that a major fraction of plutonium and americium is transported as irreversibly sorbed onto colloids.)

The distribution coefficient (K_c), a product of sorption coefficient K_d and colloid concentration, represents the equilibrium partitioning of radionuclides between the aqueous phase and the colloidal phase. The sorption coefficient distributions are listed in Table D-1. The distribution of colloid concentration is shown in Figure D-3. The same K_c applies to transport of a radionuclide in both the volcanic units and the alluvium. The simulation results are shown in Figure D-4.



Source: BSC 2003b.

Figure D-3. Cumulative Distribution Function of Uncertainty in Groundwater Colloid Concentrations



Source: BSC 2003b.

Figure D-4. Mass Breakthrough Curves (Upper) and Median Transport Times (Lower) for Plutonium on Reversible Colloids at 18-km Distance

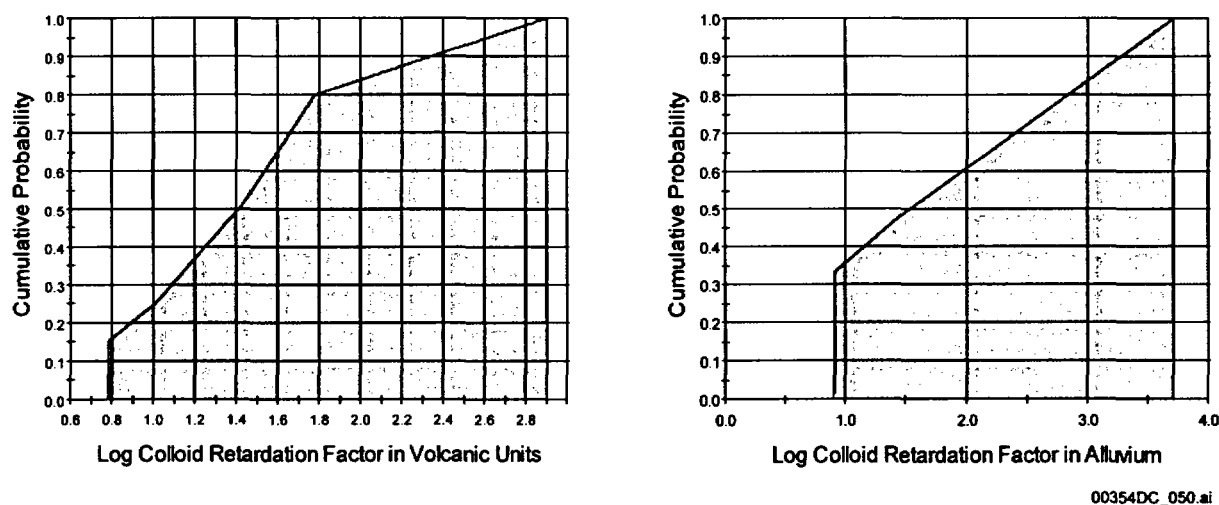
D.4.2 Response to Agreement GEN 1.01 (Comments 43 and 46)

Transport of Irreversibly Sorbed Radionuclides in Saturated Zone—For the TSPA for the license application, colloid-facilitated transport of radionuclides in the saturated zone is simulated to occur by two basic modes (BSC 2003b). In the first mode, radionuclides that are irreversibly attached to colloids are transported at the same rate as the colloids, which are themselves retarded by interaction with the aquifer material. In the second mode, radionuclides that are reversibly attached to colloids are in equilibrium with the aqueous phase and the aquifer

material. In this mode of transport, the effective retardation of these radionuclides during transport in the saturated zone is dependent on the sorption coefficient of the radionuclide onto colloids, the concentration of colloids, and the sorption coefficient of the radionuclide onto the aquifer material.

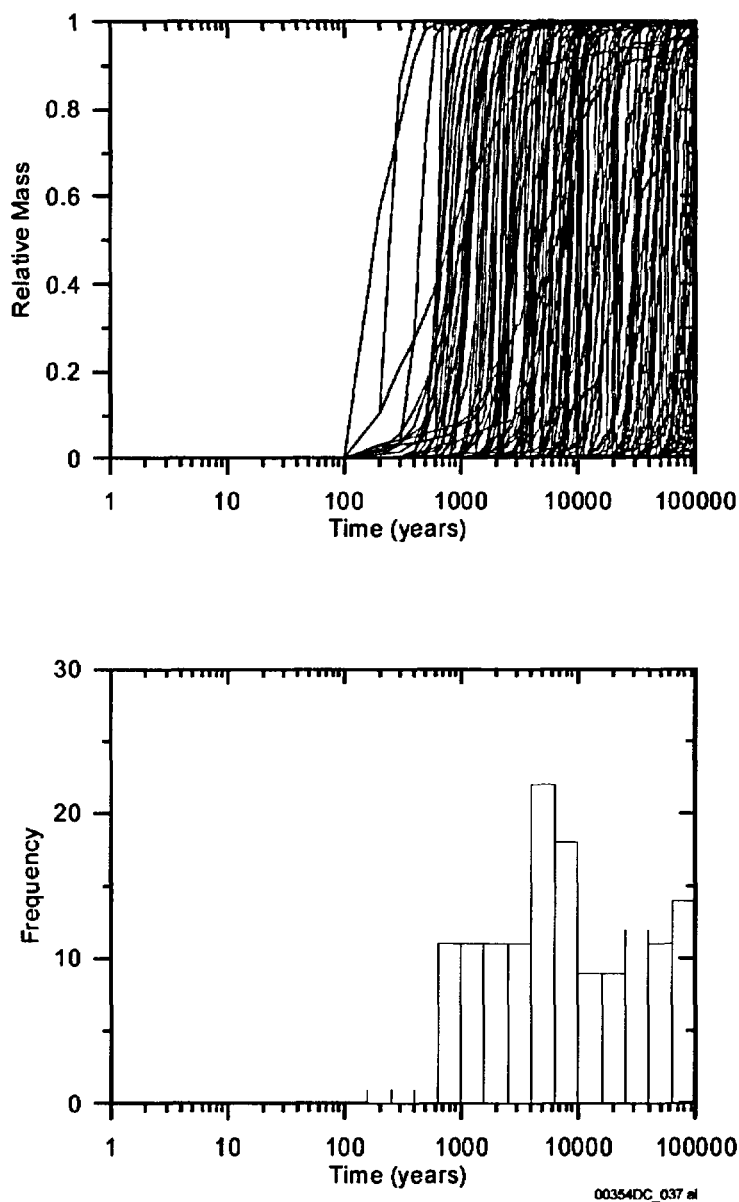
The saturated zone transport simulations of radionuclides that are irreversibly attached to colloids are conducted for radioisotopes of plutonium and americium (BSC 2003b). The retardation of colloids with irreversibly attached radionuclides is a kinetically controlled process, which approaches equilibrium behavior for long transport times. For transport of colloids through the saturated zone, equilibrium behavior is nearly achieved. However, nonequilibrium behavior results in unimpeded migration of some of the colloids. Consequently, a small fraction of these colloids is transported through the saturated zone with no retardation; whereas the larger fraction is delayed by a retardation factor. The processes important to the transport of irreversible colloids in the volcanic units of the saturated zone are as follows: advection and dispersion of colloids in the fracture water, exclusion of the colloids from the matrix waters, and chemical filtration or adsorption of the colloids onto the fracture surfaces. The processes modeled for irreversible colloids in the alluvium are the same as those modeled for irreversible colloids in the volcanic units, with the exception of matrix exclusion, because the alluvium is modeled as a single porous medium.

Figure D-5 shows the cumulative distribution function used for retardation factors for the saturated zone transport abstraction model. The simulation results for plutonium and americium are shown in Figure D-6.



Source: BSC 2003b.

Figure D-5. Cumulative Distribution Function Used for Retardation Factors for the Saturated Zone Transport Abstraction Model



Source: BSC 2003b.

Figure D-6. Mass Breakthrough Curves (Upper) and Median Transport Times (Lower) for Plutonium and Americium on Irreversible Colloids at 18-km Distance

D.5 REFERENCES

BSC (Bechtel SAIC Company) 2003a. *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary*. MDL-EBS-PA-000004 REV 00. Las Vegas, Nevada: Bechtel SAIC Company. ACC: DOC.20030626.0006.

BSC 2003b. *Saturated Zone Flow and Transport Model Abstraction*. MDL-NBS-HS-000021 REV 00. Las Vegas, Nevada: Bechtel SAIC Company. ACC: MOL.20030612.0138.

Reamer, C.W. 2001. "U.S. Nuclear Regulatory Commission/U.S. Department of Energy Technical Exchange and Management Meeting on Total System Performance Assessment and Integration (August 6 through 10, 2001)." Letter from C.W. Reamer (NRC) to S. Brocoum (DOE/YMSCO), August 23, 2001, with enclosure. ACC: MOL.20011029.0281.

Reamer, C.W. and Gil, A.V. 2001. Summary Highlights of NRC/DOE Technical Exchange and Management Meeting of Range on Thermal Operating Temperatures, September 18-19, 2001. Washington, D.C.: U.S. Nuclear Regulatory Commission. ACC: MOL.20020107.0162.

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APPENDIX E

**SENSITIVITY STUDIES TO TEST IMPORTANCE
OF COLLOID TRANSPORT PARAMETERS AND MODELS
(RESPONSE TO RT 3.07 AND GEN 1.01 (COMMENTS 35, 43, AND 46))**

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX E

SENSITIVITY STUDIES TO TEST IMPORTANCE OF COLLOID TRANSPORT PARAMETERS AND MODELS (RESPONSE TO RT 3.07 AND GEN 1.01 (COMMENTS 35, 43, AND 46))

This appendix provides a response for Key Technical Issue (KTI) agreement Radionuclide Transport (RT) 3.07 and general agreement (GEN) 1.01, comments 35, 43 and 46. These KTI agreements relate to providing sensitivity studies for colloid transport parameters and models.

E.1 KEY TECHNICAL ISSUE AGREEMENTS

E.1.1 RT 3.07 and GEN 1.01 (Comments 35, 43, and 46)

Agreement RT 3.07 was reached during the U.S. Nuclear Regulatory Commission (NRC)/U.S. Department of Energy (DOE) Technical Exchange and Management Meeting on Radionuclide Transport held December 5 through 7, 2000, in Berkeley, California. RT KTI subissues 1, 2, and 3 were discussed at that meeting (Reamer and Williams 2000).

Agreement GEN 1.01 was reached during the NRC/DOE Technical Exchange and Management Meeting on Range of Thermal Operating Temperatures, held September 18 through 19, 2001. At that meeting, NRC provided additional comments (GEN 1.01, comments 35, 43, and 46) relating to RT 3.07, and DOE provided an initial response to those comments (Reamer and Gil 2001).

The wording of these agreements and of DOE's initial response to the general agreement comments is as follows:

RT 3.07

Provide sensitivity studies to test the importance of colloid transport parameters and models to performance for UZ and SZ. Consider techniques to test colloid transport in the Alcove 8/Niche 3 test (for example, microspheres). DOE will perform sensitivity studies as the basis for consideration of the importance of colloid transport parameters and models to performance for the unsaturated and saturated zones and will document the results in updates to appropriate AMRs, and in the TSPA-LA document, all to be available in FY 2003. DOE will evaluate techniques to test colloidal transport in Alcove 8/Niche 3 and provide a response to the NRC in February 2001.

GEN 1.01 (Comment 35)

The SSPA recommends new values for EBS colloid transport parameters. If these are adopted by TSPA in the future, the technical basis for the new distributions will require close scrutiny. Relevant KTI agreements are RT 3.07, ENFE 4.03, ENFE 4.04, and ENFE 4.06.

DOE Initial Response to GEN 1.01 (Comment 35)

The new values for EBS colloidal transport parameters were designed to evaluate unquantified uncertainty for the SSPA. DOE understands that prior to any potential LA, a stronger technical basis must be provided for EBS colloidal transport parameter values carried forward to the base case analysis.

GEN 1.01 (Comment 43)

The SSPA presents a new distribution for retardation of colloids with irreversibly-attached radionuclides. The distribution takes into account new site-specific alluvium data. However, any future use of this distribution in TSPA will require comparison with results of field and laboratory tests. This concern is indirectly related to agreement TSPAI 3.30.

DOE Initial Response to GEN 1.01 (Comment 43)¹

DOE acknowledges that any future use of this distribution in TSPA will require comparison with results of field and laboratory tests 1, 2. This concern is indirectly related to KTI agreements RT 3.07 and RT 3.08. Laboratory testing of microsphere and silica colloid retardation in alluvium-packed columns is in progress. Microspheres will be used as colloid tracers in ATC cross-hole tracer testing.

GEN 1.01 (Comment 46)

The analysis of sensitivity to increased uncertainty in the reversible colloid parameter K_c (Section 12.5.2.4) yielded "somewhat longer transport times" in the SZ. This analysis does not illustrate the effect of possibly underestimating K_c , because it is not clear that the mean value of K_c is significantly different from the base case. This concern is related to agreements RT 3.07 and TSPAI 3.30.

DOE Initial Response to GEN 1.01 (Comment 46)

This issue will be handled as part of agreements RT 3.07 and TSPAI 3.30.

E.1.2 Related Key Technical Issue Agreements

None.

E.2 RELEVANCE TO REPOSITORY PERFORMANCE

Transport parameters and models for colloid transport in the unsaturated zone and saturated zone will determine (1) the ability of colloids to move through the unsaturated zone and saturated zone and into the accessible environment, and (2) the ability of the unsaturated zone and

¹ "[F]ield and laboratory tests 1, 2" refers to test conducted using CML microspheres as colloid surrogates. These tests will be discussed in detail in Appendix M (Response to KTI Agreement RT 3.08) of the saturated zone technical basis document.

saturated zone to effectively filter colloids or retard colloid transport. The interplay among several key colloid transport parameters and mechanisms will determine the significance of colloid transport through the unsaturated zone and saturated zone. Based on several pertinent studies, it has been concluded that significant filtration and retardation of colloids within both the unsaturated zone and saturated zone will take place. The technical basis for the response for this KTI agreement is presented in Sections 3, 4, 5 and 6 of this technical basis document. This KTI agreement is related to the engineered and near-field environment, the unsaturated and the saturated zones as shown in Figure E-1.

E.3 RESPONSE

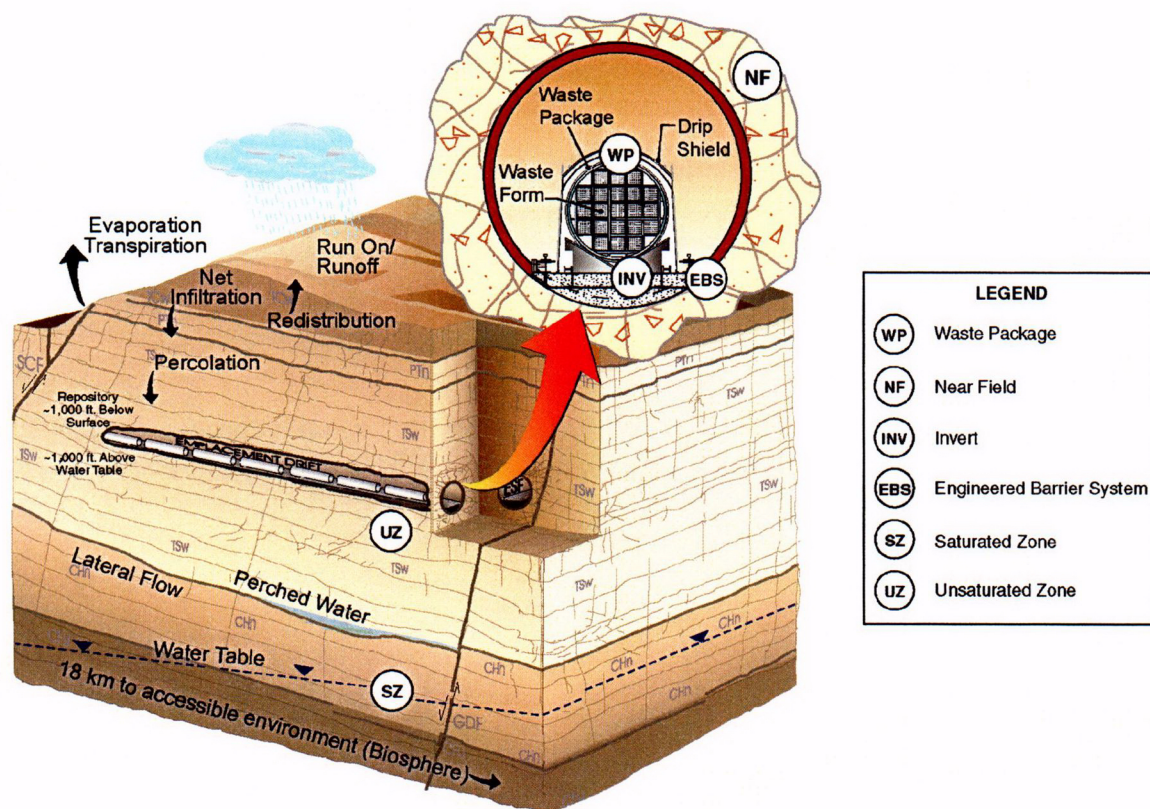
There are a number of factors associated with the transport of colloids through the unsaturated zone and saturated zone. These include: (1) where (into fractures or into the matrix) colloids are released from the repository, (2) the degree of colloid resuspension or declogging, (3) colloid retardation factors, (4) colloid reversible sorption, (5) the degree of matrix diffusion, (6) the application of extrapolating saturated zone determined colloid parameters to the unsaturated zone, (7) colloid particle size, (8) the specific discharge rate, (9) the use of linear equilibrium isotherms to describe the sorption of radionuclides onto colloids, and (10) the use of linear kinetics models to describe the retardation of colloids. Sensitivity analyses have been done that address many of these uncertainties. Details of the analyses can be found in *Radionuclide Transport Models under Ambient Conditions* (BSC 2003a, Section 6). The impact of these uncertainties on colloid transport varies from negligible (items 1 and 2) to important (items 7 and 8). The uncertainties are dealt with in two ways: 1) distributions describing the parameter uncertainty are established and propagated through the modeling systems leading to a quantified uncertainty in the results and 2) choosing a conservative conceptual model (one which results in a larger release) when two or more conceptual models are potentially possible.

The information in this report is responsive to agreements RT 3.07 and GEN 1.01 comments 35, 43, and 46 made between the DOE and NRC. The report contains the information that DOE considers necessary for NRC review for closure of these agreements.

E.4 BASIS FOR THE RESPONSE

Sensitivity analyses for colloid transport in the unsaturated zone have been performed.

Four colloid transport simulations using TOUGH2 V1.11MEOS9NTV1.0 (Module EOS9nTV1.0) were conducted, referred herein as Cases 1, 2, 3, and 4. Details and presentation of the results for these four cases appear in *Radionuclide Transport Models under Ambient Conditions* (BSC 2003a, Section 6.18, Attachment VI). It is useful to define two terms for the purpose of facilitating the analysis of the results. These are (1) the relative mass fraction of a species in the fractures or matrix, (X_R), defined as the ratio of the mass fraction in the liquid in a fracture or matrix to the mass fraction of that species in liquid released from the repository, and (2) the relative filtered concentration, (F_R), defined as the ratio of the concentration of species sorbed or filtered onto a fracture or the matrix to the concentration of that species released in the liquid from the repository.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|--|
| NF | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | INV | TSPAI 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| NF | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | EBS UZ SZ | TSPAI 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| NF | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | INV UZ SZ | TSPAI 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| WP | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | SZ | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| WP | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| WP | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure E-1. Mapping of Colloid-Related Key Technical Issue Agreements to Repository System Components

The simulation for Case 1 assumes that no declogging occurs once colloids are attached to the matrix. Two observations appear particularly important.

1. The fast breakthrough of the larger colloids (characterized by a rapid rise of the breakthrough curve), the rapidity of which is about the same for all the larger colloids.
2. Contrary to expectations, the smallest colloid (6 nm) exhibits the slowest breakthrough. It is noteworthy that this colloid never reaches the 0.1 fraction level, indicating that it is sufficiently small to enter the matrix or become sorbed to the fracture walls. This behavior results from a combination of the following factors:
 - a. The larger transport velocity of larger colloids, which, by virtue of their size, can only move in the center of pores/fractures where velocities are larger than the average water velocity.
 - b. The inability of larger colloids to penetrate into the matrix from the fractures because of size exclusion. Thus, the colloid mass in the fractures is not reduced through colloidal diffusion and/or hydrodynamic dispersion, and practically all of it moves exclusively in the fractures.

The simulation for Case 2 assumes that there is a strong declogging factor. The difference in breakthrough curves between Case 1 and Case 2 is very small for all colloids, and is most prominent for 6 nm colloids. The relative insensitivity to the clogging model in the matrix indicates the dominant role of the fractures in the three-dimensional site scale system, with the matrix appearing to have a negligible contribution.

A comparative analysis based on Case 2 (Section 5.4.3) was conducted and reported in *Radionuclide Transport Models Under Ambient Conditions* (CRWMS M&O 2000, Section 6.17) using an earlier version of the unsaturated zone transport model. This analysis examined the transport of colloids with and without colloid matrix diffusion. The analysis concludes that diffusion is less significant in colloid transport than in solute transport, because (1) colloid diffusion is smaller than solute molecular diffusion because of the larger colloid size, and (2) size-exclusion effects at the interfaces of different geologic units further limit entry by diffusion into the matrix (especially for larger colloids). However, diffusion effects become increasingly important for a decreasing colloid size. Uncertainty in colloid matrix diffusion is treated by sampling the diffusion coefficient over a range of values in the total system performance assessment for the license application unsaturated zone transport model (BSC 2003b, Section 4.2).

The simulation for Case 3 shows that the change in the magnitude of the reverse kinetic filtration coefficient \hat{e}^- (as a fraction of \hat{e}^+) has a very small effect on colloid transport, attesting to the fact that the role of advection through fracture flow (the same in Cases 1 through 3) is by far the dominant mechanism of colloid transport. The effect of a change in the reverse filtration coefficient on transport is apparent only in the case of the 6 nm colloid (which can enter the pores, and thus be subject to filtration). The insensitivity of transport of the larger colloids to the sorption model in the matrix indicates that the matrix has practically no participation in the

retardation, which is attributed to straining at matrix/fracture interfaces. The pattern that emerges is the same as the one discussed in Case 1.

In Case 4, assigning a porosity of 1 percent to the fractures, and making the fracture filtration properties equal to those of the matrix (assumed to be those of Case 2), is equivalent to creating a system of minor partial fracture filling. The effect of limited diffusion on transport (because of pore-size exclusion and filtration) becomes more obvious in this case. While the more-freely diffusing 6 nm colloids exhibit a behavior similar to the one in Cases 1 through 4, the effect on the larger colloids is more dramatic. The occurrence of even a minor fracture filling retards the colloid transport.

The potential points of release from the repository are uncertain and may vary with time; however, this does not impact the transport of colloids. Because of the importance of transport and flow within faults on the migration of dissolved radionuclides and colloids, the impact of releasing radionuclides directly into faults was assessed (BSC 2003a, Section 6.20). Despite the importance of faults (particularly the Drill Hole Wash, the Pagany Wash, and the Sundance faults) identified in the analyses of *Radionuclide Transport Models Under Ambient Conditions* (BSC 2003a, Section 6), eliminating potential sources from the immediate vicinity of the faults has a small (even negligible) effect on transport and arrivals at the water table. This is not to suggest that faults are unimportant but only that they continue to dominate whether radionuclides are directly released into them or not.

Sensitivity Analysis for Colloid Transport in the Saturated Zone—Uncertainties associated with model parameters are explicitly captured by using probability distributions of model parameter values. These uncertainties are propagated through the model to the output breakthrough curves. The results are presented in *Saturated Zone Colloid Transport* (BSC 2003c, Section 7.3). Radionuclide transit times are most sensitive to groundwater specific discharge. This is because increasing the specific discharge not only increases the advective velocity but also reduces the time available for matrix diffusion to be effective. In assessing the sensitivity of breakthrough times to the specific discharge through the model, permeabilities of the various units are scaled along with the specific discharge to preserve the model calibration. Other parameters of importance to the breakthrough times are the retardation factor for irreversible colloids in the alluvium, matrix diffusion, the sorption coefficient in the matrix as well as the alluvium, the effective fracture porosity in the volcanics, the effective porosity in the alluvium, and the sorption coefficients for reversible colloids.

Quantification of the sensitivity of the model output breakthrough curves for various radionuclides of concern to parameter uncertainties are further evaluated in *SZ Flow and Transport Model Abstraction* (BSC 2003d). There are uncertainties associated with scaling parameter values from the scale of measurements to the scale of interest. Much of the data used for deriving parameter values in this report is from laboratory or field experiments conducted on spatial and temporal scales much smaller than those expected to occur in site-scale saturated zone model.

The sensitivity of the output breakthrough curves to each of the uncertain input parameters is summarized as follows.

Specific Discharge: The results show that the output is sensitive to this parameter. The upper limit of specific discharge, 6 m/year, with extremely fast fluid flow (such as would be expected for the unlikely case of a high permeability channel going continuously over the distance of 18 kilometers in a highly faulted region), results in breakthrough as fast as 50 years. The lower limit of specific discharge, 0.02 m/year, leads to breakthrough times greater than 10,000 years.

Colloid Retardation Factor in Alluvium for Irreversible Colloids: Colloid retardation is not modeled in the base case. Compared to that, both the low and high limiting values of the colloid retardation factor for irreversibly sorbed radionuclides lead to significant retardation, resulting from retardation of the colloids themselves. The time for the 50 percent breakthrough for the lower limit of the retardation factor ($R = 7.9$) is 5,041 years, and that for the upper limit of the retardation factor ($R = 5188$) exceeds 10,000 years.

Reversible Sorption onto Colloids in the Alluvium: The range of values for the modified sorption coefficient used in this case is the same range of values as the original sorption coefficient in the alluvium. Thus, the results for the lower limiting case are identical to the base case, and those for the upper limiting case show no breakthrough at all within 10,000 years.

Colloid Retardation Factor in Volcanics for Irreversible Colloids: The colloids are modeled as having no diffusion into the volcanic matrix, and the retardation arises from the reversible filtration of the colloids by attachment onto the surfaces of the fractures. The range of uncertainty in the retardation factor of 6 to 794 translates into an output uncertainty range of 1,420 to more than 10,000 years for the breakthrough time for 50 percent of the colloids.

Reversible Sorption onto Colloids in the Volcanics: The modified sorption coefficient for this case has the same range of values as the original sorption coefficient in alluvium. In this case, the diffusion coefficient is also modified for the upper limit. Thus, the results for the lower limiting case are identical to the base case, and those for the upper limiting case show no breakthrough at all within 10,000 years. A small fraction of colloids traveled unretarded.

Alcove 8/Niche 3 Test—The matrix diffusion model is an important component of the unsaturated zone transport model. This model is corroborated with data from the Alcove 8/Niche 3 fault test (BSC 2003a, Section 7.4). The test was carried out in the upper lithophysal and middle nonlithophysal subunits in the Yucca Mountain unsaturated zone. These geological subunits correspond to model layers TSw33 and TSw34, respectively, in the site-scale model for the Yucca Mountain unsaturated zone. The TSw33 has some lithophysal cavities that may intersect fractures. Liquid water with and without tracers was released at the floor of an alcove along the fault (about 5 m long) within TSw33. Seepage from the fault into a niche and tracer concentrations of seeping liquid as a function of time were monitored. The niche is located within TSw34, about 20 m below the floor of the alcove; the interface between TSw33 and TSw34 is about 15 m below the floor of the alcove. A water-pressure head of 2 cm was applied at an infiltration plot along the fault at the alcove. The plot consists of four trenches that have different infiltration rates as a result of subsurface heterogeneity along the fault. Seepage from the fault into the niche was measured during the test, with a number of trays used to cover the areas where seepage might occur. Seepage was found to be highly spatially variable. Several boreholes were installed around the niche. Water arrival times at these boreholes were monitored through electrical resistivity probes. After 209 days, two tracers with different

molecular diffusion coefficients (Br and pentafluorobenzoic acid, or PFBA) were introduced into infiltrating water at the infiltration plot. Tracer concentrations in three of the trays (at the niche) capturing seeping water from the fault were measured. For technical reasons, seepage rates corresponding to these three trays were not measured during the period of tracer concentration measurement. In this study, a flux-averaged breakthrough curve (concentration as a function of time) from these trays was used to represent the average breakthrough curve for all trays at the niche where seepage was captured. A constant flux value for each of the three trays was used for calculating the flux-averaged breakthrough curve. The constant flux values for the three trays were determined as the averaged value over 56 days before tracers were introduced. This flux-averaged breakthrough curve is comparable to the simulation results.

Use of CML Microspheres—All of the field measurements have involved fluorescent carboxylate-modified polystyrene latex (CML) microspheres ranging in size from 250 to 500 nm diameter. Laboratory fracture experiments have been conducted using silica, montmorillonite, and clinoptilolite colloids in addition to CML microspheres. In one study, silica colloids (approximately 100 nm diameter) were compared directly with CML microspheres transport (330 nm diameter), and it was found that the microspheres transported conservatively relative to silica colloids. This result suggests that colloid filtration and retardation parameters derived from CML microsphere responses in field tracer tests should be conservative if used to predict natural inorganic colloid transport in fracture systems. See detailed discussions in *Saturated Zone Colloid Transport* (BSC 2003c).

E.5 REFERENCES

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APPENDIX F

**TRANSPORT OF DISSOLVED AND COLLOIDAL RADIONUCLIDES
THROUGH INVERT
(RESPONSE TO TSPAI 3.17 AND GEN 1.01 (COMMENTS 36 AND 38))**

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX F

TRANSPORT OF DISSOLVED AND COLLOIDAL RADIONUCLIDES THROUGH INVERT (RESPONSE TO TSPAI 3.17 AND GEN 1.01 (COMMENTS 36 AND 38))

This appendix provides a response for Key Technical Issue (KTI) agreement Total System Performance Assessment and Integration (TSPAI) 3.17 and general agreement (GEN) 1.01, comments 36 and 38. These KTI agreements relate to providing an uncertainty analysis of the colloid transport diffusion coefficient.

F.1 KEY TECHNICAL ISSUE AGREEMENTS

F.1.1 TSPAI 3.17 and GEN 1.01 (Comments 36 and 38)

Agreement TSPAI 3.17 was reached during the U.S. Nuclear Regulatory Commission (NRC)/U.S. Department of Energy (DOE) Technical Exchange and Management Meeting on Total System Performance Assessment and Integration held August 6 through 10, 2001, in Las Vegas, Nevada. TSPAI KTI subissues 1, 2, 3, and 4 were discussed at that meeting (Reamer 2001).

Agreement GEN 1.01 was reached during the NRC/DOE Technical Exchange and Management Meeting on Range of Thermal Operating Temperatures held September 18 through 19, 2001. At that meeting, NRC provided additional comments (GEN 1.01, comments 36 and 38) relating to TSPAI 3.17, and DOE provided an initial response to those comments (Reamer and Gil 2001).

The wording of these agreements and of DOE's initial response to the general agreement comments is as follows:

TSPAI 3.17¹

Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.4.1). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis will include uncertainty in the modeled invert saturation. The uncertainty analysis will be documented in the EBS Radionuclide Transport Abstraction AMR (ANL-WIS-PA-000001) expected to be available to NRC in FY 2003.

GEN 1.01 (Comment 36)

The discussion of uncertainty in the saturation level of the invert does not consider the possibility of higher saturation. This comment is related to KTI agreement TSPAI 3.17.

¹ ENG4.4.1 in this agreement refers to NRC integrated subissue ENG 4 (NRC 2002, Table 1.1-2).

DOE Initial Response to GEN 1.01 (Comment 36)

Studies with the MSTH model, as reported in Chapter 5 of the SSPA Volume 1, investigated the sensitivity of invert liquid saturation to a variety of repository parameters. These parameters included bulk permeability, host-rock thermal conductivity, lithophysal porosity, and invert thermal conductivity. Predicted liquid saturation remained within a narrow range, between four percent and 10 percent, for all parameter variations. In addition, the diffusive breakthrough time for the invert is already relatively rapid, so any increase in saturation levels is expected to have a negligible impact.

DOE will provide an uncertainty analysis of diffusion in the invert. This analysis will include uncertainty in invert saturation per KTI agreements TEF 2.05 and TSPAI 3.17.

GEN 1.01 (Comment 38)

The effect of the drift shadow assumption on invert transport needs to be evaluated. Also, as mentioned in the chapter, any adoption of a drift shadow model will require additional justification. This concern may be related to agreement TSPAI 3.17.

DOE Initial Response to GEN 1.01 (Comment 38)

The invert transport abstraction does not incorporate any direct assumptions related to a drift shadow effect. The hydrologic inputs to the invert transport calculation come primarily from the MSTH model that tracks water and gas within the near-field rock and the drift. The specific inputs from the MSTH model to invert transport are the temperature of the invert and liquid saturation of the invert. DOE acknowledges that models carried forward to support a potential license application will be qualified and documented, and may require supplemental justification or analysis.

Also, see response to comment 29.

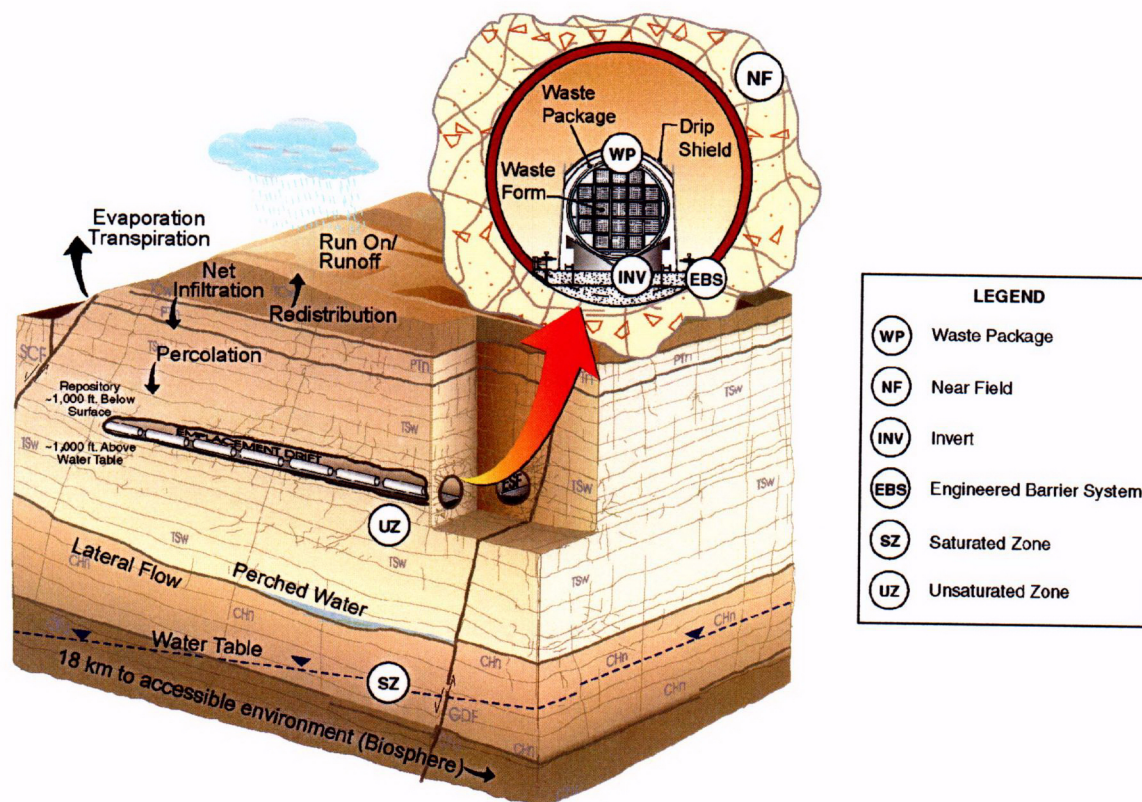
F.1.2 Related Key Technical Issue Agreements

GEN 1.01 (comment 36), TSPAI 3.30, and RT 3.07.

F.2 RELEVANCE TO REPOSITORY PERFORMANCE

Radionuclide transport through the invert is expected to be primarily dominated by diffusion of both dissolved and colloidal species. One of the key parameters is the diffusion coefficient for both types of species. The diffusion coefficient for dissolved species is a function of degree of saturation, with higher values at the higher moisture contents. The diffusion coefficient for colloids is estimated from diffusion coefficients for dissolved species multiplied by the ratio of ionic radius to colloid radius.

The technical basis for the response for this KTI agreement is presented in Section 4 of this technical basis document. This KTI agreement is related to the invert as shown in Figure F-1.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|---|
| NF | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | INV | TSPA 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| NF | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | EBS UZ SZ | TSPA 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| NF | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | INV UZ SZ | TSPA 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| WP | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | SZ | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| WP | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| WP | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure F-1. Mapping of Colloid-Related Key Technical Issue Agreements to Repository System Components

F.3 RESPONSE

Using electric conductivity measurements, the diffusion coefficient for dissolved species has been measured for crushed tuff invert material as a function of volumetric moisture content and presented in *EBS Radionuclide Transport Abstraction* (BSC 2003). From these values, diffusion coefficients for colloids can be estimated using the ratio of ionic size (molecular size) to colloid size. These measurements show that for typical moisture and saturation levels expected in the invert, diffusion of dissolved species would take more than 10,000 years, and much longer for colloids.

Uncertainty of the dissolved species diffusivity is discussed in *EBS Radionuclide Transport Abstraction* (BSC 2003, Section 6.3.4.1).

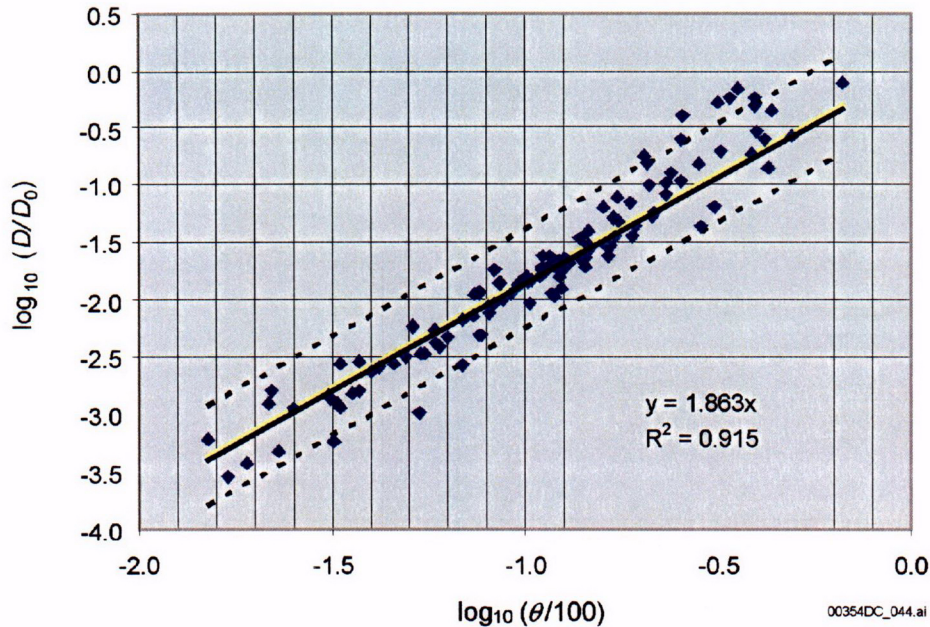
As discussed in *EBS Radionuclide Transport Abstraction* (BSC 2003, Section 7.1.2), the invert diffusion coefficient has a low impact on performance because the response of the unsaturated zone generally dominates the response of the invert. The unsaturated zone is dominant because the invert is filled with crushed tuff that is similar to the host rock in the unsaturated zone and because the thickness of the invert is quite small in relation to the travel distance in the unsaturated zone.

The information in this report is responsive to agreements TSPAI 3.17 and GEN 1.01 comments 36 and 38 made between the DOE and NRC. The report contains the information that DOE considers necessary for the NRC to review for closure of these agreements.

F.4 BASIS FOR THE RESPONSE

The diffusion coefficient for dissolved species has been determined specifically for crushed tuff invert materials as a function of volumetric moisture content (m^3 water/ m^3 bulk rock), using electric conductivity measurements (BSC 2003). Figure F-2 presents the uncertainty in the statistical fit for the diffusion coefficient.

For the nominal scenario evaluated out to 10,000 years, releases only occur because a small fraction of the waste packages have small manufacturing defects. Hence, all the releases occur via diffusive pathways through the waste package and invert. In this scenario, the only source of water is vapor from evaporation and condensation. Based on corrosion analysis, the drip shield will remain intact for the regulatory period and, thus, any seepage water that enters the drifts will contact the drip shield and be diverted away from the package. This diversion creates a drip shadow zone underneath the waste package. Therefore, no advective transport will occur through the waste package or invert even in seepage environments. Within this drip shadow, all radionuclides will be transported via diffusion through the intragranular pore space. Diffusive transport through the intergranular pore space will be insignificant since water saturations in the intergranular pore space will be near zero. Hence, transport of colloidal material through the invert will likely be negligible, since the diffusion coefficient for colloids is at least 100 times lower than for dissolved species based on the relative size of colloids and dissolved species. Any mixing that occurs between waters in this shadow will be by the slow process of diffusion of anions and cations. The water chemistry in this shadow will remain similar to matrix water as ions migrate and react with the water constituents and minerals in the matrix. Thus, the chemistry for evaluating the stability of the colloids corresponds to the matrix water chemistry.



Source: BSC 2003, Figure 7.

NOTE: The dashed lines correspond to two standard deviations above and below the statistical fit to the data. D is the diffusion coefficient of the crushed invert materials, D_0 is the diffusion coefficient of the fully saturated crushed invert materials, and θ is the percentage of saturation.

Figure F-2. Uncertainty in the Statistical Fit for the Diffusion Coefficient

The lack of diffusion can be shown as follows. The characteristic time (T) for dissolved radionuclides to diffuse through the invert of thickness L can be calculated by:

$$T = L^2 / D \quad (\text{Eq. F-1})$$

where D is the diffusion coefficient. With $L = 0.8$ m, the calculated T values are listed in the fourth column of Table F-1. As shown in Table F-1, it will take 100 to 1.65×10^6 years for a dissolved species to diffuse through the invert, depending on the saturation degree.

The colloidal diffusion coefficient can be estimated from the following Stokes-Einstein relationship (BSC 2003):

$$D_{coll} = D_{ion} \left(\frac{r_{ion}}{r_{coll}} \right) \quad (\text{Eq. F-2})$$

where D_{coll} is the diffusion coefficient for a colloidal particle of radius r_{coll} and D_{ion} is the diffusion coefficient of a ion of radius r_{ion} . Given a typical ion radius and colloidal particle radius of 0.1 and 1 nm, respectively, the diffusion coefficient of a colloidal particle is generally 100 times smaller than that of a dissolved ion. Thus, it takes at least 10,000 years for colloids to diffuse through the invert.

As discussed in *EBS Radionuclide Transport Abstraction* (BSC 2003, Section 7.1.2), the invert diffusion coefficient has a low impact on performance because the response of the unsaturated zone generally dominates the response of the invert. The unsaturated zone is dominant because the invert is filled with crushed tuff that is similar to the host rock in the unsaturated zone and because the thickness of the invert is quite small in relation to the travel distance in the unsaturated zone.

Table F-1. Diffusion Coefficient of and Time of Diffusion through Crushed Tuff Invert Materials

| Volumetric Moisture Content (%) | Diffusion Coefficient (cm ² /s) | Saturation Degree ^a (%) | Time for Diffusion through Invert ^b (year) |
|---------------------------------|--|------------------------------------|---|
| 32.13 | 2.02E-06 | 58.95 | 1.02E+02 |
| 18.15 | 5.40E-07 | 33.30 | 3.81E+02 |
| 9.26 | 4.05E-08 | 16.99 | 5.09E+03 |
| 8.29 | 2.24E-09 | 15.21 | 9.20E+04 |
| 7.54 | 6.81E-09 | 13.83 | 3.02E+04 |
| 7.36 | 6.21E-09 | 13.50 | 3.32E+04 |
| 7.22 | 4.38E-09 | 13.25 | 4.70E+04 |
| 7.03 | 6.75E-09 | 12.90 | 3.05E+04 |
| 6.97 | 7.45E-09 | 12.79 | 2.77E+04 |
| 6.89 | 6.73E-09 | 12.64 | 3.06E+04 |
| 6.84 | 2.19E-09 | 12.55 | 9.41E+04 |
| 6.75 | 5.42E-09 | 12.39 | 3.80E+04 |
| 6.63 | 4.39E-09 | 12.17 | 4.69E+04 |
| 6.63 | 3.76E-09 | 12.17 | 5.48E+04 |
| 6.23 | 3.40E-09 | 11.43 | 6.06E+04 |
| 6.11 | 1.55E-09 | 11.21 | 1.33E+05 |
| 6.00 | 3.43E-09 | 11.01 | 6.01E+04 |
| 5.55 | 2.04E-09 | 10.18 | 1.01E+05 |
| 5.46 | 2.04E-09 | 10.02 | 1.01E+05 |
| 5.41 | 9.97E-10 | 9.93 | 2.07E+05 |
| 4.45 | 6.19E-10 | 8.17 | 3.33E+05 |
| 3.64 | 5.00E-10 | 6.68 | 4.12E+05 |
| 0.29 | 1.24E-10 | 0.53 | 1.66E+06 |
| 0.20 | 1.25E-10 | 0.37 | 1.65E+06 |

Source: BSC 2003, Table 43. DTN: MO0002EBSDDC02.003

NOTE: ^a Saturation degree = volumetric moisture/porosity. The porosity of crushed tuff invert material is assumed to be 0.545 (BSC 2003, Table 1).

^b Time for diffusion is calculated using Equation F-1 with $L = 0.8$ m.

F.5 REFERENCES

F.5.1 Documents Cited

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Reamer, C.W. and Gil, A.V. 2001. Summary Highlights of NRC/DOE Technical Exchange and Management Meeting of Range on Thermal Operating Temperatures, September 18-19, 2001. Washington, D.C.: U.S. Nuclear Regulatory Commission. ACC: MOL.20020107.0162.

F.5.2 Data, Listed by Data Tracking Number

MO0002EBSDDC02.003. The Determination of Diffusion Coefficient of Invert Materials. Submittal date: 02/16/2000.

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APPENDIX G

**SCREENING CRITERIA FOR ATTACHMENT OF RADIONUCLIDES TO COLLOIDS
(RESPONSE TO RT 1.03 AIN-1, ENFE 3.05 AIN-1, AND ENFE 4.05 AIN-1)**

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX G

SCREENING CRITERIA FOR ATTACHMENT OF RADIONUCLIDES TO COLLOIDS (RESPONSE TO RT 1.03 AIN-1, ENFE 3.05 AIN-1, AND ENFE 4.05 AIN-1)

This appendix provides a response for additional information need (AIN) requests for Key Technical Issue (KTI) agreements Radionuclide Transport (RT) 1.03, Evolution of the Near-Field Environment (ENFE) 3.05 and ENFE 4.05. These KTI agreements relate to providing a technical basis for selection of radionuclides that are transported via colloids in Performance Assessment analyses.

G.1 KEY TECHNICAL ISSUE AGREEMENTS

G.1.1 RT 1.03 AIN-1, ENFE 3.05 AIN-1, and ENFE 4.05 AIN-1

Agreement RT 1.03 was reached during the U.S. Nuclear Regulatory Commission (NRC)/U.S. Department of Energy (DOE) Technical Exchange and Management Meeting on Radionuclide Transport held December 5 through 7, 2000, in Berkeley, California. RT KTI subissues 1, 2, and 3 were discussed at that meeting (Reamer and Williams 2000).

NRC used four DOE documents in its review of the response to KTI agreement RT 1.03: *Inventory Abstraction* (CRWMS M&O 2000a); *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2001); *Total System Performance Assessment (TSPA) Model for Site Recommendation* (CRWMS M&O 2000b); and *Total System Performance Assessment for the Site Recommendation* (CRWMS M&O 2000c). Additional information was requested by the NRC after the staff's review of these reports was completed, resulting in RT 1.03 AIN-1 (Reamer 2002).

Agreements ENFE 3.05 and 4.05 were reached during the NRC/DOE Technical Exchange and Management Meeting on Evolution of the Near-Field Environment held January 9 through 12, 2001, in Pleasanton, California. ENFE KTI subissues 1, 2, 3, and 4 were discussed at that meeting (Reamer 2001).

NRC used three DOE documents in its review of the response to KTI agreement ENFE 3.05: *Total System Performance Assessment (TSPA) Model for Site Recommendation* (CRWMS M&O 2000b); *Total System Performance Assessment for the Site Recommendation* (CRWMS M&O 2000c); and *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000d). Additional information was requested by the NRC after their review of these reports was completed, resulting in ENFE 3.05 AIN-1 (Schlueter 2002).

NRC used four DOE documents in its review of the response to KTI agreement ENFE 4.05: *Inventory Abstraction* (CRWMS M&O 2000a); *Waste Form Colloid-Associated Concentrations Limits: Abstraction and Summary* (CRWMS M&O 2000d); *TSPA for the Site Recommendation* (CRWMS M&O 2000c); and *TSPA Model for Site Recommendation* (CRWMS M&O 2000b). Additional information was requested by the NRC after their review of these reports was completed, resulting in ENFE 4.05 AIN-1 (Schlueter 2002).

The wording of these agreements is as follows:

RT 1.03

Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. The screening criteria for radionuclides selected for TSPA are contained in the AMR Inventory Abstraction. DOE is documenting identification of radionuclides transported via colloids for TSPA in the AMR Waste Form Colloid-Associated Concentration Limits: Abstraction and Summary, in the TSPA-SR Technical Report, and in the TSPA-SR Model Document. These documents will be available to the NRC in January 2001.

RT 1.03 AIN-1

Provide clarification and justification of radionuclides for which reversible and irreversible colloidal transport is modeled.

ENFE 3.05

Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. The technical basis for the selection of radionuclides released via reversible and irreversible attachments to colloids for different waste forms is provided in Section 3.5.6.1 of the Total System Performance Assessment (TSPA) Model for Site Recommendation (MDL-WIS-PA-000002) Rev 00. This document will be provided to the NRC in January 2001.

ENFE 3.05 AIN-1

1. Provide clarification and justification of radionuclides for which reversible and irreversible colloidal release is modeled.
2. Provide a stronger technical basis that release by irreversible attachment can be neglected for spent nuclear fuel."

ENFE 4.05

Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. The screening criteria for radionuclides selected for TSPA are contained in the AMR Inventory Abstraction (ANL-WIS-MD-000006) Rev 00, ICN 01. The DOE is documenting identification of radionuclides transported via colloids for TSPA in the AMR Colloid-Associated Concentration Limits: Abstraction and Summary (ANL-WIS-MD-000012) Rev 0, in the Total System Performance Assessment for the Site Recommendation (TDR-WIS-PA-000001) Rev 00 ICN 01, and in the Total System Performance Assessment (TSPA) Model

for Site Recommendation (MDL-WIS-PA-000002) Rev 00. These documents will be available to the NRC in January 2001.

ENFE 4.05 AIN-1

1. Provide clarification and justification of radionuclides for which reversible and irreversible colloidal release is modeled.

G.1.2 Related Key Technical Issue Agreements

None.

G.2 RELEVANCE TO REPOSITORY PERFORMANCE

Determining which radionuclides can reversibly and irreversibly attach to colloids is an important consideration in establishing the relative importance of colloid-facilitated radionuclide transport. Once the concentration of colloids and the available surface area per unit volume is determined, the mechanism for the attachment of dissolved radionuclides to colloids will establish the mass of radionuclides that can be transported via colloids.

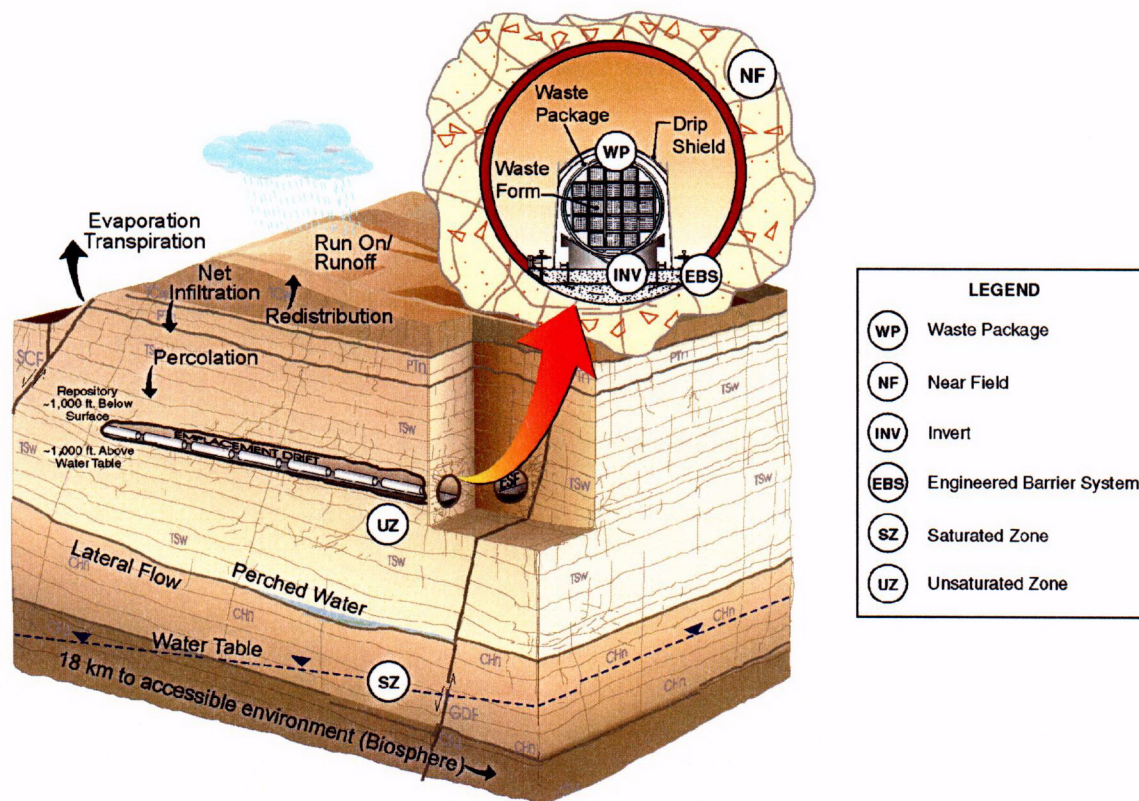
The technical basis for the response for this KTI agreement is presented in Section 3 of this technical basis document. This KTI agreement is related to the waste package as shown in Figure G-1.

G.3 RESPONSE

Section 6.3.3.1 of *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003) provides detailed discussion of the screening criteria for radionuclides undergoing reversible sorption and Section 6.3.3.2 for radionuclides undergoing irreversible sorption.

The rationale for selecting radionuclides was based on a combination of long half-life, relatively strong sorption characteristics, relatively large abundance during the initial 10,000 years in the repository, and observed field and laboratory behavior. The sorption of Pu, Am, Th, Cs, and Pa, are modeled using a linear isotherm model. K_d values for uptake of cesium (Cs), thorium (Th), protactinium (Pa), plutonium (Pu), and americium (Am) by colloids are provided in *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003, Table 10). These radionuclides were modeled because they are considered to be important to dose.

A major fraction of Pu and Am (greater than 90 percent) are assumed to attach to smectite colloids irreversibly. Evidence from sorption experiments with Pu and Am with colloidal hematite and goethite show that the rates of desorption (backward rate) of Pu and Am are significantly slower than the rates of sorption (forward rate). More importantly, over a significant time period (up to 150 days in some experiments), the extent of desorption is considerably less than the extent of sorption. Pu and Am are considered so strongly sorbed to colloids that, for practical purposes, they can be considered irreversibly sorbed and are modeled in this manner within the engineered barrier system.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|---|
| (NF) | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | (INV) | TSPA 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| (NF) | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | (EBS) (UZ) (SZ) | TSPA 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| (NF) | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | (INV) (UZ) (SZ) | TSPA 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| (WP) | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | (SZ) | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| (WP) | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| (WP) | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure G-1. Mapping of Colloid-Related Key Technical Issue Agreements to Repository System Components

Recent long-term corrosion tests with commercial and DOE spent nuclear fuel have demonstrated alteration products containing low concentrations of uranium-based colloids (and low concentrations of actinides), dissolution of the uranium-based colloids over a short time duration (less than several months), and sorption dominating the behavior of the actinides (particularly onto the vessels' metal surfaces).

The information in this report is responsive to agreements RT 1.03 AIN-1, ENFE 3.05 AIN-1, and ENFE 4.05 AIN-1 made between the DOE and NRC. The report contains the information that DOE considers necessary for NRC review for closure of these agreements.

G.4 BASIS FOR THE RESPONSE

G.4.1 Screening of Radionuclides for Reversible and Irreversible Attachment to Colloids

Section 6.3.3.1 of *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003) provides detailed discussion of the screening criteria for radionuclides undergoing reversible sorption and Section 6.3.3.2 for radionuclides undergoing irreversible sorption.

G.4.1.1 Reversible Sorption

Transport of radionuclides on colloids is potentially important for radioisotopes that 1) have low solubility; 2) have long half-lives, 3) can be entrained in waste-form colloids, or can be sorbed onto waste-form colloids, engineered barrier materials colloids, or geologic barrier materials colloids; 4) represent a major portion of the inventory in terms of total activity; and/or 5) can contribute significantly to radioactivity during the 10,000-year regulatory period. Considering these five criteria as part of radionuclide screening in Section 6.3.3.1, *Waste Form and In-Drift Colloids-Associated Radionuclide Concentrations: Abstraction and Summary* (BSC 2003) evaluated eight radionuclides for reversible sorption onto to colloids: Pu, Am, Th, Cs, Pa, Np, U, and Sr. Below is the rationale used in that report to include or exclude these radionuclides from the model for reversible attachment to colloids:

- **Plutonium**—A large quantity of Pu will exist in the repository. Pu is sparingly soluble but sorbs strongly to oxide mineral surfaces (generally less strongly to silicates). Pu is observed to sorb strongly to soil minerals, and laboratory investigations have shown that it sorbs readily to colloids as well.
- **Americium**—Am will be a significant contributor to radioactivity during the 10,000 years. As is Pu, Am is sparingly soluble but strongly sorbs to mineral surfaces, including colloids. Laboratory investigations have shown that it sorbs strongly to colloids.
- **Cesium**—¹³⁵Cs has a long half-life and can attach strongly to certain sheet silicates (including clays) by means of ion exchange. For this reason, ¹³⁵Cs has been observed to sorb to soil minerals, and it could potentially form pseudocolloids particularly with groundwater and defense high-level radioactive waste glass-derived clay colloids.

- **Protactinium**—Pa will be a significant contributor to radioactivity during the first 10,000 years. Because of this, and the fact that relatively little is known of the colloid behavior of Pa, it was included in this analysis.
- **Thorium**—Th will be a significant contributor to radioactivity during the first 10,000 years. Because of this, and the fact that there is evidence that Th sorbs strongly to oxides, it was included in this analysis. There is relatively little known of the colloid-related behavior of Th.
- **Neptunium**—Because Np will be the most significant contributor to radioactivity beyond the first 10,000 years, it was considered for inclusion in this analysis. Np is more soluble under anticipated repository conditions than many of the other important radionuclides, and it sorbs considerably less strongly than, for example, Pu and Am. It would appear then that the mobility of Np is influenced mostly by its solubility. For these reasons, and to simplify the modeling, Np was not included in the reversible-sorption portion of the colloid-associated radionuclide transport analysis.
- **Uranium**—U will be by far the most abundant radioactive element in the repository and primarily for this reason was considered for the analysis. U is more soluble under anticipated repository conditions than many of the other important radionuclides, and it sorbs considerably less strongly than, for example, Pu and Am. As with Np, the mobility of U is influenced mostly by its solubility. Field observations at U deposits and mine sites have indicated that little or no colloid U transport occurs. For these reasons, and to simplify modeling, U was not included in the reversible-sorption of the colloid-associated radionuclide transport analysis.
- **Strontium**—Sr is expected to be a potentially significant contributor to dose but because of its very short half-life was not considered further in this analysis.

G.4.1.2 Irreversible Sorption

Defense high-level radioactive waste glass degradation experiments show that plutonium is probably irreversibly attached to smectite colloids generated during the experiments. Further, evidence from sorption experiments with Pu and Am (Lu et al. 2000) with colloidal hematite and goethite show that the rates of desorption (backward rate) of Pu and Am are significantly slower than the rates of sorption (forward rate). More importantly, over a significant time period (up to 150 days in some experiments), the extent of desorption is considerably less than the extent of sorption. Pu and Am are considered so strongly sorbed to colloids that, in essence, they can almost be considered irreversibly sorbed and are modeled in this manner within the engineered barrier system. Pu transport velocities in soils reflect the fact that Pu binds strongly to soils, leaving very little, if any, soluble Pu available for groundwater transport or plant uptake. Coughtrey et al. (1985) estimated exchangeable Pu to be less than one percent. At Rocky Flats, soil Pu is largely bound to soil metal hydroxides. Litaor and Ibrahim (1996) used 0.01M CaCl₂ as an extractant and measured Pu in Rocky Flats soil to be 0.04 to 0.08 percent exchangeable. Bunzl et al. (1995) measured exchangeable ²³⁹Pu and ²⁴⁰Pu (0.5 to 1 percent) and ²⁴¹Am (1.5 to 15 percent) from fallout-contaminated soils in Germany using 1M C₂H₇NO₂ (ammonium acetate NH₄C₂H₃O₂) as the extractant. Laboratory experiments of Pu sorption onto iron oxides have

shown that only approximately one percent of the initially sorbed Pu can be desorbed into solution, even after months of time have elapsed (Lu et al. 2000), which is broadly consistent with field observations. For these reasons, Pu and Am are modeled as irreversibly attaching to colloids. No other radionuclides are considered to be irreversibly attached to colloids.

In order to accommodate these observations in the colloid abstraction, the TSPA-LA model calculates irreversible and reversible sorption of Pu and Am onto corrosion colloids as functions of specific surface area (S_A), site density (N_A), mass of corrosion colloids, dissolved concentration of Pu and Am, target-flux out ratio, and other parameters (such as flow and diffusion rate) internal to the TSPA-LA model. This is done in such a way that a majority (90 to 99 percent) of Pu and Am are irreversibly sorbed. The reversibly sorbed portion is determined according to an equilibrium K_d value model.

G.4.2 Colloid Formation from Commercial and Defense Spent Nuclear Fuel

Long-term corrosion testing of commercial spent nuclear fuel and defense spent nuclear fuel under unsaturated, oxidizing conditions has been performed to examine the release of radionuclides and, specifically for the purpose of this report, the release in the form of colloids. Testing was designed to simulate a variety of Yucca Mountain repository relevant water-exposure conditions for several spent nuclear fuels with a range of fuel burnups and compositions. Results from the unsaturated testing of commercial spent nuclear fuel and defense spent nuclear fuel indicated spallation of alteration products containing low concentrations of uranium-based colloids (and low concentrations of actinides), dissolution of the uranium-based colloids over a short time duration (less than several months), and sorption dominating the behavior of the actinides (onto clay colloids or onto metal surfaces).

Assessment of the importance of potential colloid formation from commercial spent nuclear fuel was based on four major observations: (1) very low colloid concentrations were observed in the commercial spent nuclear fuel degradation tests, at least an order of magnitude less than concentrations observed in the defense high-level radioactive waste glass degradation tests (based on dynamic light scattering measurements) (Mertz et al. 2003); (2) the fraction of uranium in the colloid mass was uniformly low in the commercial spent nuclear fuel tests, the only deviation from this occurring immediately following one of two changes in vessel configuration in which the uranium fraction increased but rapidly decreased to the approximate level of earlier values (Mertz et al. 2003); (3) suspensions of meta-schoepite and UO_{2+x} colloids in J-13 groundwater appear to exhibit decreasing stability in short-term saturated (with respect to the uranium phases) tests; their stability in unsaturated solutions has not been tested (Mertz et al. 2003); and (4) field studies at uranium-bearing deposits indicate generally that under oxidizing conditions at near-neutral pH, colloid particles contain little uranium and there is little sorption of uranium complexes to colloids.

One of the reasons hypothesized for the low colloid release in the commercial spent nuclear fuel tests was the test configuration in which the Zircaloy-4 support for the fuel fragments had 7-micron holes. However, the results from the unirradiated UO_2 tests also show few colloids after the formation of alteration products (Wronkiewicz et al. 1997). The unsaturated tests on unirradiated UO_2 had a test configuration with large 2 to 3 mm holes at the holder base allowing for the spallation of UO_{2+x} particulate during initial corrosion; however, the formation of a dense

mat of alteration products during the UO_2 corrosion apparently reduced particulate release by trapping particulates in the altered products (Wronkiewicz et al. 1997). A similar mechanism whereby the alteration products minimize particulate release may be applicable to the commercial spent nuclear fuel unsaturated tests.

The concentration of released particulates or colloids from the commercial spent nuclear fuel tests is very low except during test conditions corresponding to disruptive events, such as movement of the fuel from one retainer to another (Mertz et al. 2003). In that case, colloid and particulate concentrations increased temporarily but returned to very low concentrations after the disruption (Mertz et al. 2003). While this indicates that disruptive events may contribute to the release of particulates and colloids from commercial spent nuclear fuel, it also indicates that the longevity of the colloids in the leachate is short.

Of the different types of fuel in the defense spent nuclear fuel inventory, metallic uranium fuel comprises approximately 85 percent (by weight) of that inventory and thus was selected for corrosion testing. An irradiated uranium metal fuel from the N-Reactor at Hanford was tested in an experimental setup similar to that used at Argonne National Laboratory for testing commercial spent nuclear fuel. Additional details on this testing can be found elsewhere (DTN: MO0306ANLSF001.459). Corrosion testing of metallic uranium samples resulted in rapid oxidation (within a few months) of the uranium primarily to an oxide sludge consisting of UO_2 and higher oxides of uranium (DTN: MO0306ANLSF001.459). Although the uranium fuel disintegrated rapidly, corrosion testing was continued to determine the effect of groundwater leaching on the fuel sludge. Results from the corrosion tests showed that the composition of the defense spent nuclear fuel colloids evolve over time from an initially UO_2 -rich population, to a mixed colloid population containing UO_2 and higher oxides of U as well as smectite clays, to a population that appears to be dominated by U-containing smectite clays.

After approximately one year of testing on the defense spent nuclear fuel, the total quantity of uranium in the sludge represented approximately 99 weight percent of the original uranium fuel sample, while that in the colloid size group corresponded to 0.002 to 0.006 weight percent of the original uranium fuel sample. The quantity of uranium in the fraction attached to the stainless steel vessel was 0.1 to 0.3 weight percent of the original fuel sample (DTN: MO0306ANLSF001.459). The attached material is measured by washing the stainless steel vessel in HNO_3 ; the attached material includes sorbed solutes, sorbed colloids, and precipitates (DTN: MO0306ANLSF001.459). The disposition of Pu during defense spent nuclear fuel corrosion showed that Pu is associated predominantly with the colloidal, particulate, and sorbed size fractions. The $^{239}\text{Pu}/^{238}\text{U}$ ratios in the colloid fraction are significantly larger than those in the other fractions (sorbed, particulate, and dissolved) and is the only fraction that showed enrichment of Pu in comparison to that in the fuel prior to corrosion (DTN: MO0306ANLSF001.459). Results from the testing support a model of defense spent nuclear fuel in which Pu is significantly adsorbed to the surface of colloids (such as corrosion products of the waste package or groundwater clays) but does not occur as an embedded radionuclide in waste form colloids (DTN: MO0306ANLSF001.459).

The results of the tests conducted for commercial spent nuclear fuel and defense spent nuclear fuel along with the corroborative field information indicate that the impact of colloids from the degradation of these waste forms is not going to be significant compared to the potential impact of

colloids from defense high-level radioactive waste glass and from pseudocolloids. Therefore, it is reasonable to exclude true colloids from the TSPA modeling.

G.4.3 Response to Additional Information Needed Requests

In response to the AINs listed in Section G.1.1, clarification and justification of radionuclides for which reversible and irreversible colloidal release is modeled has been provided. In addition, recent tests in commercial spent nuclear fuel and defense spent nuclear fuel indicate that colloids forming from the degradation of those waste forms will not be significant; thus, providing a stronger technical basis that colloid-associated radionuclides release can be neglected for commercial spent nuclear fuel and defense spent nuclear fuel.

G.5 REFERENCES

G.5.1 Documents Cited

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G.5.2 Data, Listed by Data Tracking Number

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APPENDIX H
CHANGES IN COLLOID CONCENTRATIONS
DUE TO SHIFTS IN pH AND IONIC STRENGTH
(RESPONSE TO TSPAI 3.42)

Note Regarding the Status of Supporting Technical Information

This document was prepared using the most current information available at the time of its development. This Technical Basis Document and its appendices providing Key Technical Issue Agreement responses that were prepared using preliminary or draft information reflect the status of the Yucca Mountain Project's scientific and design basis at the time of submittal. In some cases this involved the use of draft Analysis and Model Reports (AMRs) and other draft references whose contents may change with time. Information that evolves through subsequent revisions of the AMRs and other references will be reflected in the License Application (LA) as the approved analyses of record at the time of LA submittal. Consequently, the Project will not routinely update either this Technical Basis Document or its Key Technical Issue Agreement appendices to reflect changes in the supporting references prior to submittal of the LA.

APPENDIX H CHANGES IN COLLOID CONCENTRATIONS DUE TO SHIFTS IN pH AND IONIC STRENGTH (RESPONSE TO TSPAI 3.42)

This appendix provides a response for Key Technical Issue (KTI) agreement Total System Performance Assessment and Integration (TSPAI) 3.42. This KTI agreement relates to providing a sensitivity analysis on changes in colloid concentration due to changes in pH and ionic strength.

H.1 KEY TECHNICAL ISSUE AGREEMENT

H.1.1 TSPAI 3.42

Agreement TSPAI 3.42 was reached during the U.S. Nuclear Regulatory Commission (NRC)/U.S. Department of Energy (DOE) technical exchange and management meeting on total system performance assessment and integration held August 6 through 10, 2001, in Las Vegas, Nevada. TSPAI KTI subissues 1, 2, 3, and 4 were discussed at that meeting (Reamer 2001).

The wording of this agreement is as follows:

TSPAI 3.42

DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. This analysis may be combined with plans to address ENFE Agreement 4.06 and RT Agreement 3.07.

DOE will complete sensitivity analyses to investigate the effects of varying colloid concentration due to shifts in model predicted pH and ionic strength across uncertain stability boundaries. These analyses will be documented in TSPA for any potential license application expected to be available to NRC in FY 2003.

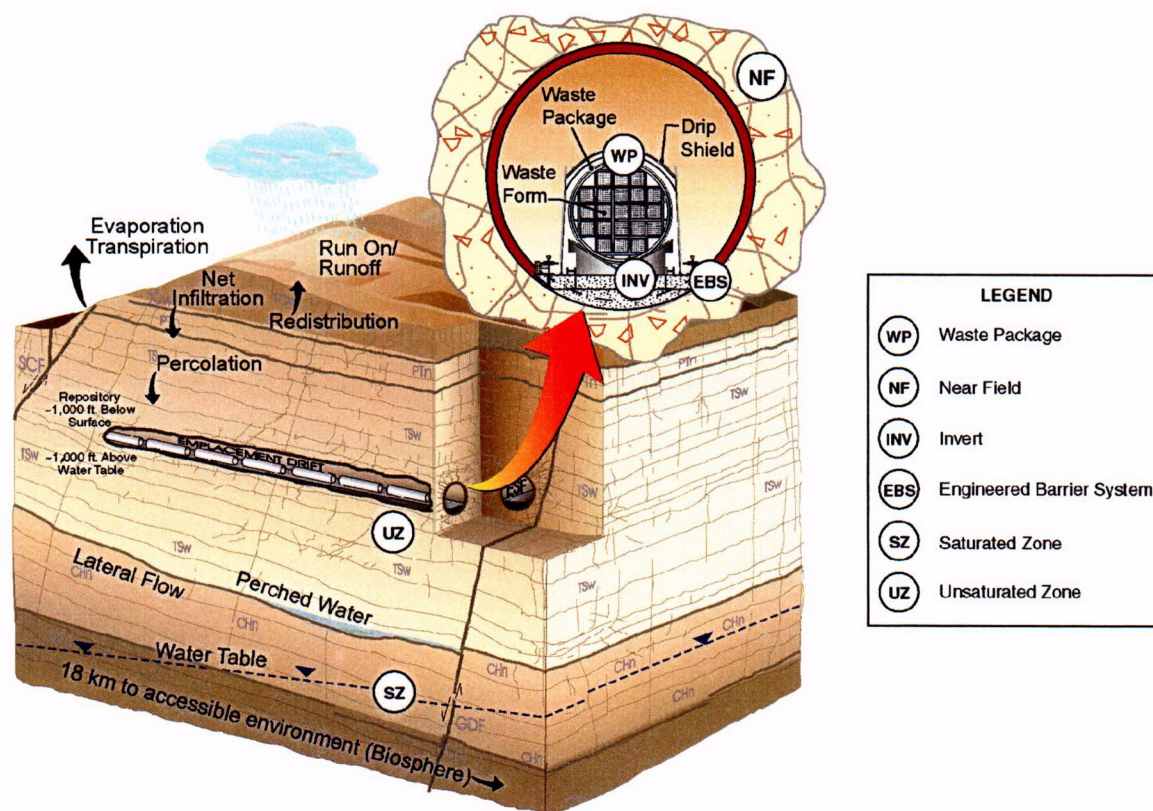
H.1.2 Related Key Technical Issue Agreement

ENFE 4.06 and RT 3.07.

H.2 RELEVANCE TO REPOSITORY PERFORMANCE

The stability of colloidal suspensions impacts the significance of colloid-facilitated radionuclide transport. The pH-ionic strength relationship is one of several phenomena that impact the concentration of colloids in suspension. In addition, temperature and relative humidity also have an impact on the stability of colloid suspensions.

The technical basis for the response for this KTI agreement is presented in Sections 4, 5, and 6 of this technical basis document. This KTI agreement is related to the invert, saturated zone, and unsaturated zone as shown in Figure H-1.



| SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT | SYSTEM COMPONENTS | KEY TECHNICAL ISSUE AGREEMENT |
|-------------------|--|-------------------|---|
| (NF) | ENFE 1.06 and ENFE 4.04: Provide the technical basis for excluding entrained colloids in the analysis of FEP 2.2.10.06.00 (Thermo-Chemical Alteration). | (INV) | TSPA 3.17: Provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. The analysis should include uncertainty in the modeled invert saturation (ENG4.41). DOE will provide an uncertainty analysis of the diffusion coefficient governing transport of dissolved and colloidal radionuclides through the invert. |
| (NF) | ENFE 4.03: Provide the technical basis for screening out coupled THC effects on radionuclide transport properties and colloids- dissolved and colloids. | (EBS) (UZ) (SZ) | TSPA 3.30 and RT 3.07: Provide the technical basis for the contrasting concentrations of colloids available for reversible attachment in the the engineered barrier system and the saturated zone. Sensitivity analyses planned in response to RT Agreement 3.07 should address the effect of colloid concentration on Kc. |
| (NF) | ENFE 4.06: Provide documentation to demonstrate suitability of the bounding values used for colloid transport through the perturbed near-field environment. | (INV) (UZ) (SZ) | TSPA 3.42: DOE should provide a sensitivity analysis on the potentially abrupt changes in colloid concentrations due to shifts in modeled pH and ionic strength across uncertain stability boundaries. |
| (WP) | ENFE 4.05: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of radionuclides that are transported via colloids in the TSPA. | (SZ) | RT 3.08: Provide justification that microspheres can be used as analogs for colloids (for example, equivalent ranges in size, charge, etc.). |
| (WP) | RT 1.03: Provide the screening criteria for the radionuclides selected for PA. Provide the technical basis for selection of the radionuclides that are transported via colloids in the TSPA. | | |
| (WP) | ENFE 3.05: Provide the technical basis for selection of radionuclides that are released via reversible and irreversible attachment to colloids for different waste forms in the TSPA. | | |

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Figure H-1. Mapping of Colloid-Related Key Technical Issue Agreements to Repository System Components

H.3 RESPONSE

The sensitivity analysis required by this KTI agreement was not performed. However, as discussed below, performance of the repository system is not believed to be sensitive to abrupt changes across the pH-ionic strength stability boundaries.

Only during the first 300 years following waste package breach can pH-ionic strength values for both smectite and iron-oxyhydroxide colloids fall within the range for stable colloid suspensions (BSC 2003). After 300 years, all possible pH-ionic strength combination of values would be within the range for which the colloid suspensions are unstable (BSC 2003). As shown in Section H.4, even during the first 300 years of the postclosure period only a very small number (less than 6 percent) of pH-ionic strength values will fall within the stable colloid suspension region.

In addition, within the drift environment, temperature will be high during the early postclosure period (e.g., the first 1,000 years) as shown in part (a) of Figure 3-2 of this technical basis document. These conditions will contribute to making colloid suspensions unstable. Therefore, even if the pH-ionic strength values were to be favorable to stable colloids, the high temperature will destabilize the colloid suspensions.

Based on these considerations, the total repository system performance will not be impacted by abrupt changes across pH-ionic strength stability boundaries. Therefore, a detailed sensitivity analysis as stated in this KTI agreement is not necessary.

The information in this report is responsive to agreement TSPAI 3.42 made between the DOE and NRC. The report contains the information that DOE considers necessary for NRC review for closure of this agreement.

H.4 BASIS FOR THE RESPONSE

Ranges of sampled values of ionic strength and pH for the co-disposal waste package with N-Reactor fuel and no-drip conditions are shown in Table H-1 (BSC 2003). It is shown in the table that for the first 300 years after the waste package breaches there is a small chance (less than 6 percent) of colloids being stable within the waste package.¹ Note that the analysis in *In-Package Chemistry Abstraction* (BSC 2003) assumes no water evaporation during waste package degradation. As discussed in Section 3.1 (Figures 3-2 and 3-3), radiation heat-induced water evaporation will likely concentrate any solution available inside the drift and lead to a high ionic strength environment (greater than 0.05 M, a threshold for colloid instability) environment for a significant portion of the regulatory time period. The evaporation effect will further reduce the chance for the formation of a stable colloid suspension with the drift. In summary, the probability of forming a stable colloid suspension is low (less than 5 percent), and so is the chance for “the potentially abrupt changes in colloid concentration due to modeled pH and ionic strength across uncertain stability boundaries”. This means that changes in pH and ionic

¹ Similar data for a CSNF package under no-drip conditions indicate that there is a similarly small chance of sampling a pH-ionic strength combination that would yield a stable colloid suspension persisting over the entire 10,000-year regulatory period, not just the first 300 years. These data were not presented since it has been concluded that CSNF colloid contribution is negligible.

strengths across uncertain stability boundaries will have little, if any, impact on colloid concentrations.

Table H-1. Ranges of Sampled Values of Ionic Strength and pH for Co-Disposal Packages with N-Reactor Fuel, No-Drip

| Time Period (yr) | Ionic Strength Range (M) | pH Range | Probability | |
|------------------|----------------------------|------------|-------------|----------|
| | | | Stable | Unstable |
| 0 to 55 | 4×10^{-6} to 0.85 | 4.1 to 9.6 | 0.06 | 0.94 |
| 55 to 300 | 0.01 to 0.85 | 4.1 to 9.6 | 0.05 | 0.95 |
| 300 to 10,000 | 0.05 to 0.85 | 4.1 to 9.6 | 0.0 | 1.00 |

Source: BSC 2003.

Within the drift environment, temperature will be high during the early postclosure period (e.g., the first 1,000 years) as shown in part (a) of Figure 3-2 of this technical basis document. The high temperature will make colloid suspensions unstable. Therefore, even if the pH-ionic strength relationship was to be favorable towards stable colloids (first 300 years of the postclosure period), the high temperature will destabilize the colloid suspensions.

H.5 REFERENCES

BSC (Bechtel SAIC Company) 2003. *In-Package Chemistry Abstraction*. ANL-EBS-MD-00037 REV 01D. Las Vegas, Nevada: Bechtel SAIC Company. ACC: MOL.20030617.0024.

Reamer, C.W. 2001. "U.S. Nuclear Regulatory Commission/U.S. Department of Energy Technical Exchange and Management Meeting on Total System Performance Assessment and Integration (August 6 through 10, 2001)." Letter from C.W. Reamer (NRC) to S. Brocoum (DOE/YMSCO), August 23, 2001, with enclosure. ACC: MOL.20011029.0281.

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