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May 28, 2019

MEMORANDUM TO: Stephen S. Koenick, Chief
Low-Level Waste and Projects Branch
Division of Decommissioning, Uranium Recovery
and Waste Programs
Office of Nuclear Materials Safety and Safeguards

THRU: Christepher A. McKenney, Chief //RA//
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FROM: George W. Alexander, Risk Analyst //RA//
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Office of Nuclear Materials Safety and Safeguards

SUBJECT: TECHNICAL REVIEW: SALTSTONE WASTE FORM PHYSICAL DEGRADATION (DOCKET NO. PROJ0734)

The U.S. Nuclear Regulatory Commission (NRC) staff performed a technical review of the U.S. Department of Energy (DOE) documents related to the Savannah River Site (SRS) Saltstone Disposal Facility (SDF) for saltstone waste form physical degradation as part of the NRC monitoring of the DOE disposal actions to determine compliance with the performance objectives (POs) set forth in Subpart C of Title 10, Part 61, of the Code of Federal Regulations (10 CFR Part 61) at the SRS SDF pursuant to Section 3116(b) of the Ronald W. Reagan National Defense Authorization Act for Fiscal Year 2005 (NDAA).

The NRC review was performed in accordance with monitoring activities described in the NRC 2013 SDF Monitoring Plan, Rev. 1 (Agencywide Documents Access and Management System (ADAMS) Accession No. ML13100A113). The NRC 2013 SDF Monitoring Plan contains monitoring areas and each monitoring area contains one or more monitoring factors. This NRC Technical Review Report (TRR) is related to Monitoring Area (MA) 4, “Waste Form Physical Degradation” and MA 10, “Performance Assessment Model Revisions.” Within MA 4, this TRR is related to Monitoring Factor (MF) 4.01, “Waste Form Matrix Degradation” and MF 4.02, “Waste Form Macroscopic Fracturing.” Within MA 10, this TRR is related to MF 10.02, “Defensibility of Conceptual Models” and MF 10.05, “Moisture Characteristic Curves.”

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The NRC staff's conclusions are the following:

The NRC staff recommends keeping MF 4.01, MF 4.02, MF 10.02, and MF 10.05 open under both §61.41 and §61.42. The NRC staff also recommends keeping MF 4.01, MF 4.02, and MF 10.02 as high priority monitoring factors and increasing the priority of MF 10.05, from low to medium under both §61.41 and §61.42.

Enclosure: Technical Review: Saltstone Waste Form Physical Degradation
Appendix: Additional Information Regarding Potential Degradation Mechanisms

cc: (w/ Enclosure):
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SUBJECT: TECHNICAL REVIEW: SALTSTONE WASTE FORM PHYSICAL DEGRADATION (DOCKET NO. PROJ0734) **May 28, 2019**

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ADAMS Accession No.: ML19031B221 ***via email**

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Appendix

Technical Review: Saltstone Waste Form Physical Degradation

Date:

May28, 2019

Reviewers:

George Alexander, Risk Analyst, U.S. Nuclear Regulatory Commission (NRC)
A. Christianne Ridge, Sr. Risk Analyst, NRC

Purpose:

The purpose of this NRC staff Technical Review Report (TRR) is to describe recent research results related to saltstone degradation, evaluate the U.S. Department of Energy (DOE) revised assumptions about saltstone degradation that is in both the DOE Fiscal Year (FY) 2013 and DOE FY 2014 Saltstone Disposal Facility (SDF) Special Analysis Documents[1], and describe the potential effects of degradation on Savannah River Site (SRS) SDF performance. Release of iodine and technetium (Tc) are discussed in more detail in radionuclide-specific TRRs (ADAMS Accession Nos. ML16342C575 and ML18095A122, respectively).

Background:

As described in the NRC 2013 Monitoring Plan for the SRS SDF, Rev. 1, (Agencywide Documents Access and Management System (ADAMS) Accession No. ML13100A113) (NRC 2013 SDF Monitoring Plan), saltstone degradation is important to SDF performance because it affects the hydraulic and chemical properties of saltstone. Water flow and contaminant transport though saltstone are affected because degradation can result in an increase in hydraulic conductivity and diffusivity of the saltstone matrix as well as introducing fractures into saltstone, through which water and dissolved radionuclides can flow. Chemical properties are affected because degradation could increase the flow of oxygen and carbon dioxide into saltstone as dissolved constituents in water and in the gas phase. An increased flow of oxygen and carbon dioxide into saltstone could result in changes to the chemical properties of the system, such as the oxidation-reduction potential and the pH. These changes could lead to changes in the chemical form of the radionuclides, which could affect the release of the radionuclides from saltstone.

Matrix degradation of saltstone can result in an increase in the release of radionuclides to the groundwater through several mechanisms. Most simply, as the rate of water flow through saltstone increases, more water is available to carry radionuclides from the waste form. An increase in flow and diffusion through the saltstone matrix also can further increase radionuclide release due to changes in the chemistry of the saltstone. Specifically, oxygen dissolved in the infiltrating water can react with the saltstone grout and consume some of the reducing capacity of the saltstone, thereby promoting the release of certain redox-sensitive radionuclides, such as Tc. In addition, depending on the degradation mechanisms, an increase in water flow through the matrix can result in a positive degradation feedback loop. An increase in the hydraulic properties may result in an increase in the transport of deleterious species into the saltstone grout or an increase in the leaching of saltstone (e.g., decalcification).

Fractures are potentially risk significant primarily because they could lead to gas-phase transport of oxygen into the saltstone grout and an increased release of certain redox-sensitive

[1] Hereafter, referenced as the DOE 2013 SDF SA and DOE 2014 SDF SA, respectively

radionuclides. Gas-phase transport of oxygen represents a potentially significant increase in the rate of oxidation of saltstone versus aqueous-phase transport of oxygen because of the relatively high concentration of oxygen in the air compared to the low aqueous solubility. Fractures also can increase water flow through saltstone, potentially providing fast pathways for radionuclides through saltstone while minimizing the potential for re-reduction of redox sensitive species that have been oxidized. However, because such fast pathways also would limit the contact of water with the inventory of radionuclides in the matrix, the potential effects of fast pathways on radionuclide release are complex.

In the DOE SDF 2009 SDF Performance Assessment (PA), the DOE assumed that no physical degradation of saltstone was expected based on a thermodynamic and mass balance analysis (SRR-CWDA-2009-00017; WSRC-STI-2008-00236). Accordingly, the DOE Base Case[2] (Case A) did not include a modeled representation of saltstone degradation, such as an increase in hydraulic conductivity or fracturing of the saltstone matrix. In Section 2.6.4.2 of the NRC 2012 SDF Technical Evaluation Report (TER) (ADAMS Accession No. ML121170309), the NRC indicated that the DOE assumption that saltstone will be hydraulically undegraded for 20,000 years was unrealistically optimistic. The NRC staff also indicated that the DOE assumption was inconsistent with observations of existing cracks in saltstone grout in Saltstone Disposal Structure (SDS) 4, although it was not clear how far those cracks extended into the saltstone grout, or the extent to which saltstone may be subject to additional cracking in the future.

The DOE included alternative scenarios designed to demonstrate the effects of a range of potential saltstone degradation assumptions in the DOE 2009 SDF PA. Those alternative scenarios included: fractures in the saltstone matrix (Case C), increased saltstone matrix hydraulic conductivity (Case E), and a synergistic sensitivity analysis to evaluate the impact of changing multiple material parameters due to several potential increased degradation mechanisms. However, as the NRC staff described in the 2012 SDF TER, there were aspects of those alternative scenarios (e.g., unexpected modeled barrier performance, redundant barriers) that limited the usefulness of the information that the alternative scenarios provided. In the 2012 SDF TER, the NRC concluded that the DOE non-mechanistic analyses of fracturing in the DOE PA included assumptions that were not representative of the expected performance of the SDF.

In response to the second NRC Request for Additional Information (RAI) Questions (ADAMS Accession No. ML103400571) on the DOE 2009 SDF PA, the DOE provided Case K[3] (SRR-CWDA-2011-00044, Rev. 1), which modeled an ingrowth of fractures represented as a log-linear increase in hydraulic conductivity and diffusivity of saltstone grout with time. Complete physical degradation of saltstone was assumed to occur in 10,000 years with the hydraulic

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[2] In the DOE 2009 PA, the DOE referred to Case A as the Base Case, which was described as the scenario that the DOE most expected for the duration of the performance period. In both the DOE SDF FY 2013 and FY 2014 SAs, the DOE did not use the term “Base Case;” but, instead developed an “Evaluation Case” by selecting parameter values that the DOE considered to be most probable and defensible. The DOE also ran a variety of sensitivity analysis cases with “Best Estimate” values. Although there were slight differences in how the DOE described the Base Case and Evaluation Case, the DOE used the Base Case and Evaluation Case results for comparison to the 10 CFR Part 61 performance objectives.

[3] The DOE provided the NRC three cases related to Case K: Case K, K1, and K2. The only differences between those three cases were the K_d values used to represent Tc sorption in oxidizing and reducing cementitious materials (saltstone and disposal structure concrete). When that distinction is not important (e.g., when discussing hydraulic properties, which are the same in all three cases), the NRC staff uses the term “Case K” to refer to all three cases.

conductivity and diffusivity reaching values of 1.0 x 10^{-6} cm/s and 5.0 x 10^{-6} cm^2/s, respectively. However, the NRC indicated in the 2012 SDF TER that there was significant uncertainty in the rate and extent of fracturing of cementitious materials over thousands of years. Consequently, the timing and magnitude of the projected dose from radionuclide releases from the SDF were uncertain.

In the NRC 2012 SDF TER (ADAMS Accession No. ML121170309), the NRC staff also raised concerns related to the DOE assumed moisture characteristic curves (MCCs) in the DOE 2009 SDF PA Base Case. However, in Case K, the DOE revised the modeling approach to unsaturated flow through saltstone and assumed that the relative permeability was 1.0 (i.e., the DOE assumed that the relative permeability was independent of saltstone saturation and that the MCC did not diminish the hydraulic conductivity from the saturated hydraulic conductivity).

To address the NRC staff concerns related to saltstone degradation, the NRC included the following two monitoring factors in the NRC 2013 SDF Monitoring Plan under Monitoring Area 4 (Waste Form Physical Degradation):

- MF 4.01 - “Waste Form Matrix Degradation”
- MF 4.02 - “Waste Form Macroscopic Fracturing”

In addition, the NRC staff included the following two monitoring factors in the NRC 2013 SDF Monitoring Plan under MA 10 (Performance Assessment Model Revisions) related to support for MCCs that may be used in future DOE SDF PAs and DOE SDF SAs:

- MF 10.02 – “Defensibility of Conceptual Models”
- MF 10.05 - “Moisture Characteristic Curves”

FY 2013 and FY 2014 SDF SAs and Recent Research

In both the FY 2013 and the FY 2014 SAs, the DOE revised the approach to modeling saltstone degradation by assuming that decalcification[4] via advective flow controls the degradation of saltstone (SRR-CWDA-2013-00062; SRR-CWDA-2014-00006; SRR-CWDA-2013-00064). As water migrates through the saltstone grout, calcium is assumed to leach from saltstone at a concentration equal to the pore solution concentration measured from a saltstone simulant (SRNL-STI-2013-00118, Rev. 1). The time to complete degradation was calculated according to the following equation:

$$t = \frac{c_{Ca}}{c_{Ca^{2+}}} \cdot \frac{h}{U}$$

Where:
$t = Elapsed\ time, yr$
$c_{Ca} = Concentration\ of\ calcium\ in\ saltstone, mol/cm^3 total$
$c_{Ca^{2+}} = Concentration\ of\ calcium\ in\ solution, mol/cm^3 liquid$
$h = Height\ of\ the\ saltstone\ monolith, cm$
$U = Darcy\ velocity, cm/yr$

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[4] The DOE assumed in the Evaluation Cases in both the FY 2013 and FY 2014 SAs that degradation of the upper (i.e., 3.45 feet) of saltstone grout and clean cap in SDS 4 will be controlled by carbonation due to the presence of steel roof trusses.

The DOE considered best estimate, nominal value, and conservative scenarios with U = 1, 10, and 100 times the saturated hydraulic conductivity, respectively, to account for the potential head build up on top of saltstone. The time for complete degradation of saltstone from decalcification for the three scenarios is shown below, in Table 1.

Table 1: DOE Projected Years until Complete Degradation of Saltstone (Adapted From Table 4.2-7 in SRR-CWDA-2014-00006)

	SDS 1	SDS 4	46-m (150-ft) diameter disposal structures	114-m (375-ft) diameter disposal structures
Best Estimate (U=1)	2,495,499	2,317,249	2,240,856	4,379,855
Nominal Value (U=10)	249,550	231,725	224,086	437,985
Conservative Estimate (U=100)	24,955	23,172	22,409	43,799

The DOE assumed that saltstone would degrade linearly with time from the initial hydraulic conductivity of 6.4×10^{-9} cm/s to that of the surrounding soil (SRR-CWDA-2014-00006). Accordingly, fully degraded saltstone would not act as a barrier or conduit to flow relative to the surrounding soil. In the Evaluation Cases in both the FY 2013 and FY 2014 SAs, the diffusion coefficient of saltstone was also assumed to increase linearly from an initial value of 1.0×10^{-8} cm^2/s to 5.3×10^{-6} cm^2/s after the saltstone was assumed to be completely degraded.

In the FY 2013 and FY 2014 SAs, the DOE also revised the MCCs for saltstone such that the MCCs varied in time to simulate degradation (SRR-CWDA-2013-00062, SRR-CWDA-2014-00006). The revised MCCs were a weighted arithmetic average of the initial intact saltstone matrix hydraulic properties and those of backfill soil, which represents the assumed end state (SRNL-STI-2013-00280). The hydraulic properties (i.e., hydraulic conductivity, saturation, relative permeability, porosity) at each time interval was based on a linear weighting of the extent of degradation (e.g., for a time when the degradation front is expected to have traveled 5 percent (%) of the distance through saltstone, the calculated MCC is taken to be an arithmetic average of the MCC for un-degraded saltstone weighted by a factor of 0.95 and the MCC for soil weighted by a factor of 0.05). As the modeled degradation proceeds, the proportion of these two materials are blended according to the DOE projected degradation of saltstone from intact to fully degraded (i.e., saltstone with the hydraulic properties of soil) for that modeled time period. Figure 1 and Figure 2 below from the FY 2014 Special Analysis Document provide illustrations of the MCCs at different times for saltstone grout in 46-m (150-ft) and 114-m (375-ft) diameter disposal structures.

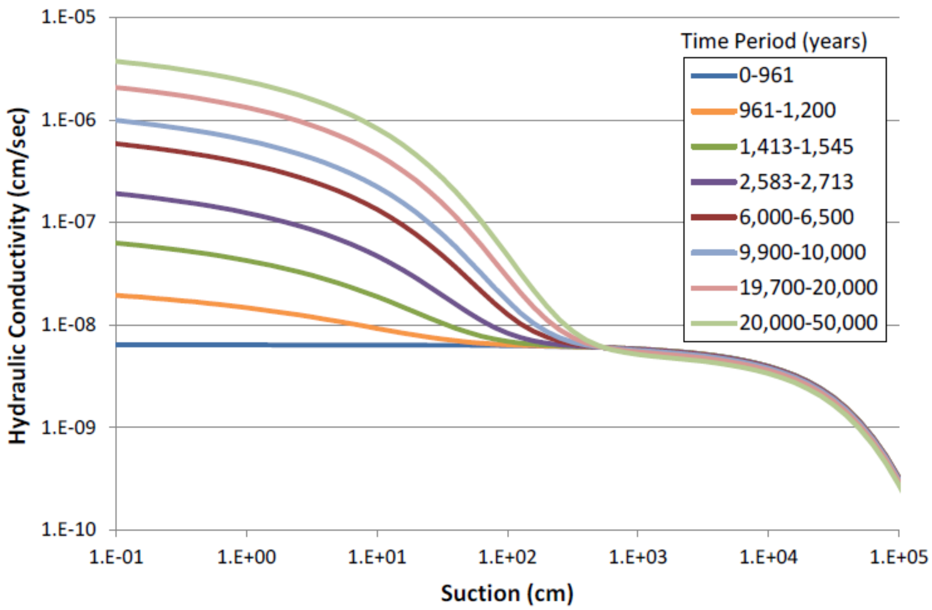

Figure 1: Blended MCC for 46-m (150-ft) Diameter Disposal Structure Assuming Nominal Degradation (Reproduced from Figure 4.2-26 in SRR-CWDA-2014-00006)

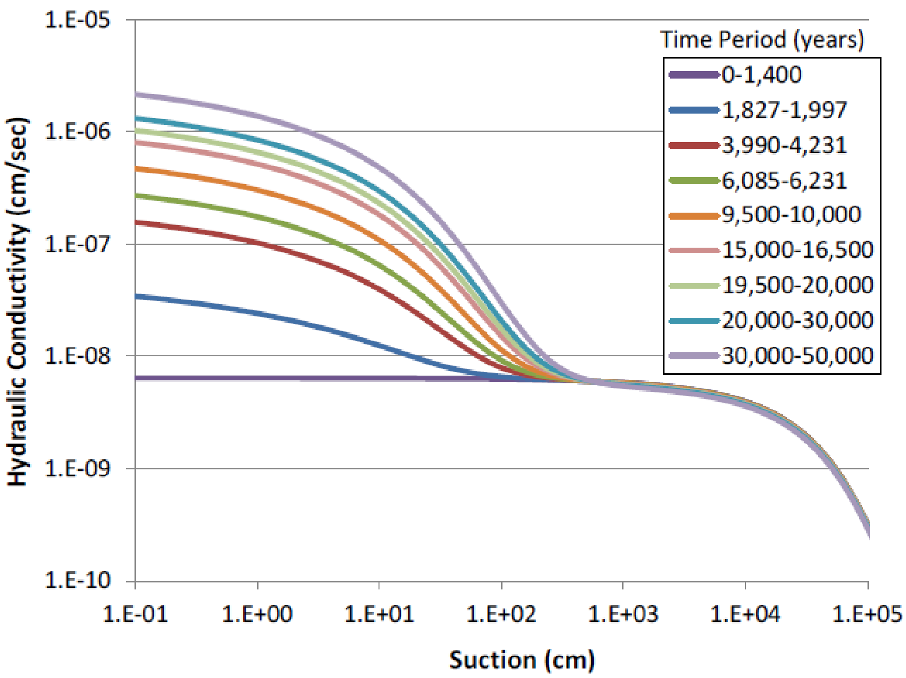

Figure 2: Blended MCC for 114-m (375-ft) Diameter Disposal Structure Assuming Nominal Degradation (Reproduced from Figure 4.2-30 in SRR-CWDA-2014-00006)

NRC Evaluation

In the NRC 2012 SDF TER, the NRC staff concluded that the constant physical and hydraulic properties assumed for the entire 20,000-year analysis period in the Base Case in the DOE SDF 2009 PA were unrealistically optimistic. In that TER, the NRC staff also described that, of the cases analyzed by the DOE, the NRC staff considered the non-mechanistic modeling of saltstone degradation in Case K to best reflect the current and future expected conditions of the waste form based on available data. However, the NRC staff further described that there is significant uncertainty in the rate and extent of fracturing in Case K.

In the NRC 2013 SDF Monitoring Plan, MA 4, "Waste Form Physical Degradation," had two monitoring factors: MF 4.01, "Waste Form Matrix Degradation" and MF 4.02, "Waste Form Macroscopic Fracturing." Degradation of the matrix can result in increased diffusivity and water flow through the pores. Accordingly, matrix degradation can result in an increase in aqueous-phase transport of oxygen, which increases the amount of saltstone oxidation, and faster release of dissolved radionuclides. Saltstone fracturing can result in increased water flow through saltstone, a shortened diffusive length, and increased oxidation due to an increase in surface area and gas-phase transport of oxygen. The distinction between these two monitoring factors is important because of differences in experiments and measurement methods to quantify hydraulic properties. Also, matrix degradation and macroscopic fractures are often represented separately in models. MA 10, "Performance Assessment Model Revisions," included two monitoring factors related to saltstone degradation: MF 10.02, "Defensibility of Conceptual Models" and MF 10.05, "Moisture Characteristic Curves." The representation of saltstone degradation has a significant effect on the projected release of radionuclides from saltstone and is therefore a risk-significant part of the conceptual model of how saltstone functions. MCCs are important to long-term projections of saltstone performance because they govern, in part, how much water is projected to flow through unsaturated saltstone as it is projected to degrade.

Due to the potential for degradation processes to affect cementitious materials, the NRC staff considers the inclusion of saltstone degradation in the Evaluation Cases in both the DOE FY 2013 and FY 2014 SAs to be a significant improvement over the DOE 2009 SDF PA. Those Evaluation Cases provided insight into the risk associated with the gradual degradation of saltstone due to decalcification. However, there is considerable uncertainty associated with saltstone degradation. During the development of the DOE FY 2013 SA, the DOE and the NRC held a public meeting (see meeting summary in ADAMS Accession No. ML13025A038) and discussed the modeling activities that the DOE conducted since issuance of the NRC 2012 SDF TER. In that meeting, the NRC staff discussed the need for consideration of the following processes:

- mechanical degradation mechanisms as well as a complete range of potential saltstone chemical degradation mechanisms (MF 4.01 and 4.02),
- a positive feedback loop (i.e., coupling) between multiple degradation mechanisms (MF 4.01 and MF 4.02), and
- saltstone fracturing and gas-phase transport of oxygen in addition to aqueous-phase transport of oxygen from infiltrating water (MF 4.02).

After the NRC staff's initial review of the FY 2013 SDF SA, the NRC staff discussed in a public meeting in 2014 with the DOE (see meeting summary in ADAMS Accession No. ML14057A578) the need to account for mechanical degradation mechanisms. Both the DOE and the NRC staff have continued that discussion in several forums since that time. During these discussions, the NRC staff expressed concerns that mechanical degradation mechanisms such as, but not limited to, shrinkage, thermal gradients, and differential settlement were not adequately accounted for in the DOE analysis. Over time, the DOE provided several reasons to support their position that mechanical degradation mechanisms, while not explicitly represented in the DOE analysis, were adequately accounted for by other conservatisms. The NRC staff addressed most of those reasons in RAI Comment SP-4 in the NRC RAI Comments (ADAMS Accession No. ML14148A153) on the DOE FY 2013 SDF SA and in RAI Question SP-3 in the NRC RAI Questions (ADAMS Accession No. ML15161A541) on the DOE FY 2014 SDF SA. As is described in those RAIs, the NRC staff still remains concerned that the DOE degradation modeling does not adequately address the potential extent and effects of mechanical degradation mechanisms.

The DOE document entitled "Conceptual Model Development for the Saltstone Disposal Facility Performance Assessment" (SRR-CWDA-2018-00006) indicated that, mechanical degradation mechanisms including: (1) cracking due to seismic events, (2) settlement due to overburden, (3) subsidence due to calcareous zones, (4) cracking due to static loading, (5) cracking due to expansive chemical reactions with ions in the soil, (6) increased porosity and/or cracking due to soil corrodents, and (7) cracking due to freeze thaw processes will be considered in a future SDF PA in an "early release" scenario and a "fast flow paths" scenario. However, in that document the DOE indicated that those mechanical degradation mechanisms will not be included in the central scenario. That document also indicated that saltstone was not expected to undergo significant mechanical degradation because it will be surrounded by disposal structure concrete. However, the document does not indicate how the disposal structure concrete would prevent shrinkage cracking or cracking due to thermal gradients. In addition, the document did not indicate how the disposal structure concrete would prevent cracking due to static or dynamic settling, which could affect both disposal structure concrete and saltstone.

MF 4.01, "Waste Form Matrix Degradation"

In MF 4.01 of the NRC 2013 SDF Monitoring Plan, the NRC staff concluded that the assumed values for the saturated hydraulic conductivity and diffusivity during the performance period need to be adequately supported. Examples of adequate model support described in MF 4.01 included mechanistic modeling and laboratory experiments intended to simulate accelerated aging. Since the issuance of the 2013 SDF Monitoring Plan, the DOE developed a mechanistic decalcification model for the evolution of saltstone physical and hydraulic properties over time. The DOE also conducted accelerated leaching tests that provided some insight into the properties of saltstone during several pore volume flushes.

In the FY 2014 SA, the DOE indicated that a linear degradation rate in the Evaluation Case was conservative because it was expected to overestimate the amount of degradation that occurs in early time (i.e., much less than 10,000 years), thereby, providing some compensation for the potential effects of mechanical degradation. The NRC staff agrees that at early times a linear degradation rate projects a greater extent of fracturing than the non-linear approach, as described in the DOE Document SRR-CWDA-2016-00004 Rev. 1. However, it is not clear to the NRC staff that a linear degradation rate can account for mechanical degradation because there is not sufficient information available to understand the risk significance of those mechanisms or to exclude additional degradation mechanisms[5]. Those additional potential degradation mechanisms could result in degradation that exceeds the DOE projections.

In addition, although the linear degradation rate exceeds the non-linear degradation rate at early times, as shown in Figure 3 below, the hydraulic conductivity for the non-linear model increases

[5] Appendix to this TRR provides additional information regarding potential degradation mechanisms, in addition to the previously discussed decalcification process.

more rapidly at later times. The NRC staff expects that flow through freshly-degraded saltstone would result in greater radionuclide releases than the same amount of flow through previously-degraded saltstone because previously-degraded saltstone would be expected to be more depleted in inventory. Therefore, annual radionuclide releases may be more sensitive to the rate of degradation than the total amount degraded at any time. If that conceptual model is accurate, then a non-linear degradation rate could result in greater annual radionuclide releases and therefore greater dose. In other words, because the release of radionuclides from a linear degradation model is the most gradual release rate possible for a given extent of degradation, it would not be the most conservative model if radionuclide releases are more sensitive to the total annual new degradation rather than to the cumulative amount of saltstone that is degraded. However, if the contaminants of concern are depleted prior to the rapid increase in hydraulic conductivity at later times associated with the non-linear model, then the linear model with the higher initial release would result in a greater dose. Based on the available information, there is not sufficient support for the assumption that the degradation rate will be linear or that a linear degradation rate is conservative.

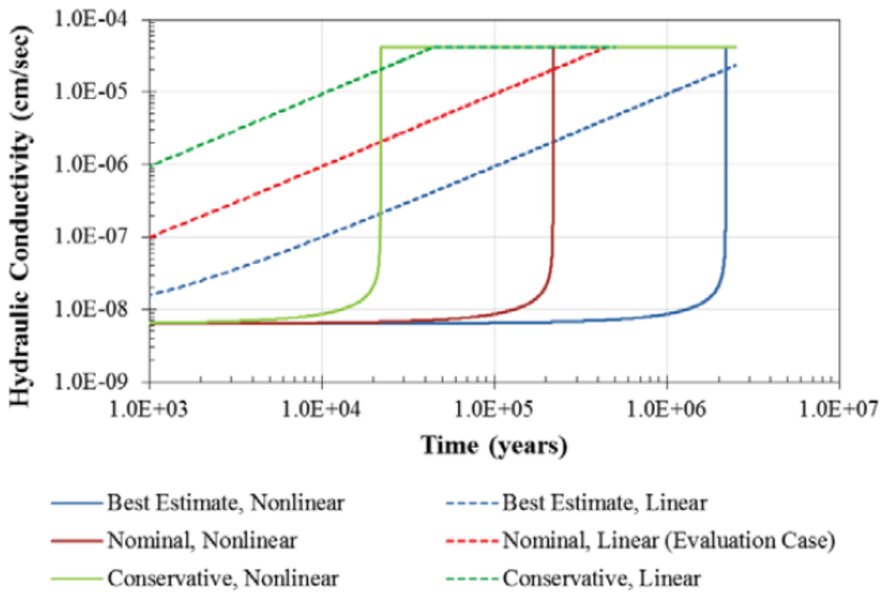

Figure 3: Log-Log Plot of the Modeled Hydraulic Conductivity of Saltstone (Reproduced from SRR-CWDA-2016-00004 Rev. 1)

The NRC staff is also concerned that multiple degradation mechanisms could act concurrently. Degradation mechanisms tend to act in concert with each other rather than in isolation. For example, mechanical degradation, such as shrinkage cracking, can cause fracturing that will allow more water to migrate into the wasteform, which could increase the formation of expansive phases in localized regions resulting in additional cracking. Accordingly, there is a potential for a positive feedback between multiple degradation mechanisms, which could result in a non-linear rate of degradation. Therefore, if releases are sensitive to the degradation rate as well as the cumulative amount of degradation as previously discussed, then a greater-than-assumed release rate of radionuclides could occur.

The NRC staff is also concerned about the potential for degradation to begin earlier than when the DOE projected because of the possibility of additional degradation mechanisms occurring that were not considered in both the DOE FY 2013 and FY 2014 SAs (see RAI Comment SP-5 in ADAMS Accession No. ML14148A153 and RAI Question SP-5 in ADAMS Accession No.

ML15161A541). In the Evaluation Cases in both the FY 2013 and FY 2014 SAs, the DOE assumed that saltstone did not degrade until the overlying barriers (i.e., the roof of all the disposal structures and the clean cap of SDS 4) were degraded. As indicated in Table 2 below, that resulted in the delay of initiation of saltstone degradation. In both the NRC RAI Comments on the FY 2013 SA and the NRC RAI Questions on the FY 2014 SA, the NRC staff described that degradation mechanisms that were not included in the DOE analyses (e.g., shrinkage cracking, differential volume changes due to thermal gradients) that could result in the degradation of saltstone shortly after placement in the disposal structures.

Table 2: Delay in Saltstone Grout Degradation (Adapted from SRR-CWDA-2014-00006)

Saltstone Disposal Structure	Delay to Initiation of Degradation
SDS 1	486 years
SDS 4	2,112 years
46-m (150-ft) Diameter Disposal Structure	961 years
114-m (375-ft) Diameter Disposal Structure	1,413 years

In response to the NRC RAI Comments and NRC RAI Questions, the DOE described that the nominal degradation rate, after the delay to initiation of degradation of saltstone, was assumed to be one order of magnitude greater than the DOE best estimate of the hydraulic degradation rate. The DOE described that this provided for greater defensibility (DOE Response to NRC RAI Comment SP-3 in SRR-CWDA-2016-00004, Rev. 1) and to account for a head gradient greater than one (SRNL-STI-2013-00118, Rev. 1). Because of the uncertainty in infiltration through the closure cap as discussed in NRC TRR *Hydraulic Performance and Erosion Control of the Planned Saltstone Disposal Facility Closure Cap and Adjacent Area* (ADAMS Accession No. ML18002A545), the likelihood of a hydraulic head greater than one on top of the saltstone grout is not clear to the NRC staff. Therefore, it is not clear to the NRC staff the extent to which increasing the hydraulic degradation rate by an order of magnitude from the DOE best estimate to the nominal value would account for a head gradient greater than one, and how much, if any, of the increase is left to account for additional and coupled degradation mechanisms.

In January 2017, the DOE provided additional analyses related to recent laboratory results of Tc release (SRR-CWDA-2016-00134). Several of those analyses evaluated the dose impact of assuming a Tc solubility greater than what is assumed in both the FY 2013 and 2014 SAs in combination with the DOE best estimate for the cementitious degradation rate. As shown in Table 1 above, the DOE best estimate of degradation for saltstone assumed a hydraulic gradient of 1 through saltstone (i.e., no buildup of head) and a resultant time to complete degradation of greater than 2 million years for saltstone grout in all of the disposal structures. The NRC staff is still concerned that there is not adequate support for the DOE best estimate of degradation based on the limited consideration of additional and coupled degradation mechanisms.

The NRC staff had several questions related to the rate of the DOE assumed degradation due to decalcification (RAI Questions SP-1 and SP-4 in ADAMS Accession No. ML15161A541). In RAI Question SP-1, the NRC staff described the concern that the DOE assumed change in hydraulic conductivity with time did not account for the feedback of increasing hydraulic conductivity as decalcification progresses. As the NRC staff stated:

> *In Section 4.2.2.3 of the DOE FY14 SDF Special Analysis document, decalcification via advection was assumed to control the degradation of saltstone. The rate of*

decalcification was assumed to be constant and was based on the initial hydraulic conductivity of saltstone. However, decalcification would result in an increase in the hydraulic conductivity of saltstone and therefore, the rate of decalcification would increase as the hydraulic conductivity in saltstone increases in time. That dependency would create a feedback loop that was not accounted for by the DOE. Including that feedback loop of increasing hydraulic conductivity with decalcification would significantly decrease the amount of time required for complete degradation of saltstone to occur.

In response to RAI Question SP-1 (SRR-CWDA-2016-00004, Rev.1), the DOE provided a degradation analysis based on decalcification that compared the linearly increasing hydraulic conductivity (i.e., linear) approach in the DOE FY 2014 Special Analysis to a two-layered system that included a feedback loop (i.e., non-linear) in determining the hydraulic conductivity. As shown in both Figure 4 and Figure 5 below, that analysis showed that including a feedback loop resulted in the fully degraded hydraulic conductivity being reached in about half the time of the constant hydraulic conductivity approach. However, the hydraulic conductivity of the case including feedback remained approximately two orders of magnitude lower than the constant hydraulic conductivity case for more than 10,000 years. That non-intuitive result is due to the inhibition of flow by the non-degraded fraction of saltstone ahead of the decalcification front.

The DOE also considered a more spatially rigorous simulation where the hydraulic conductivity of overlying layers increased relatively quickly with less permeable underlying layers. That conceptual model would result in more lateral flow with water being diverted around the underlying saltstone grout. That process would tend to reduce the peak dose because more water flows around, rather than through the un-degraded saltstone matrix. That process also would tend to move the peak dose earlier in time than in the Evaluation Case because, in the Evaluation Case, Tc-99 is released from overlying cells in the saltstone grout into underlying saltstone grout where it is re-reduced. The bulk of the Tc-99 in the Evaluation Case is not released until the oxidation front migrates through most of the saltstone. The DOE concluded that the simpler conceptual model used in the FY 2014 SA was therefore conservative with respect to the peak dose. The NRC staff agrees with the DOE assessment of the simpler conceptual model of decalcification being conservative compared to the multi-layer model of decalcification with respect to the magnitude of the peak dose. However, the NRC concludes that there is uncertainty in the timing of the peak dose. Recent dynamic leaching research from both the Savannah River Ecology Laboratory (SREL) and the Center for Nuclear Waste Regulatory Analyses (CNWRA) has provided additional understanding of the initial conditions of flow and contaminant transport through saltstone grout. That information, in combination with a more complete degradation analysis, would improve the defensibility of the DOE conceptual model for flow and contaminant transport.

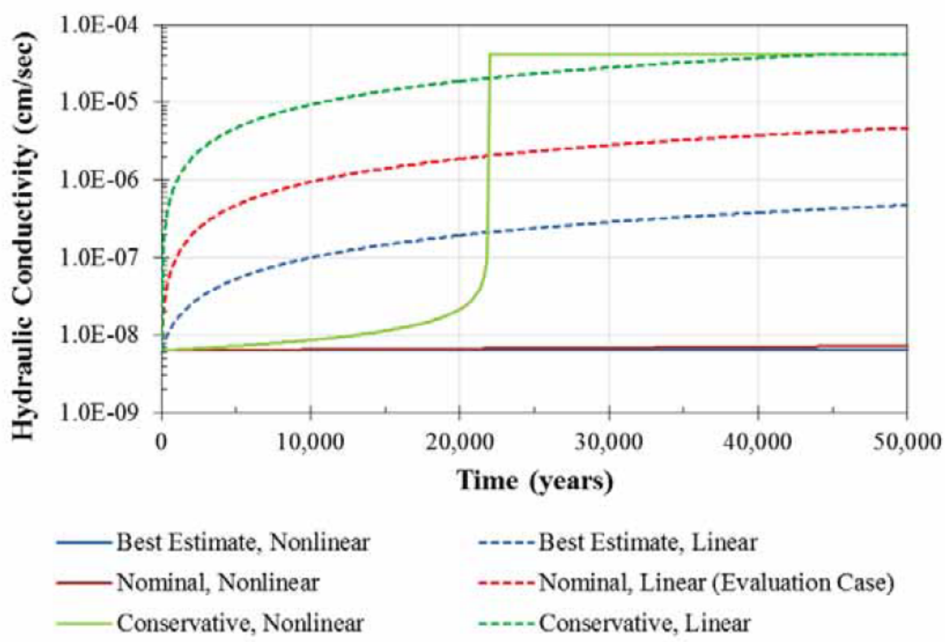

Figure 4: Semi-Log Plot of the Hydraulic Conductivity Over Time Used in the DOE Response to the NRC RAI Questions on the DOE FY 2014 SDF SA (Reproduced from Figure SP-1.1 in SRR-CWDA-2016-00004, Rev. 1)

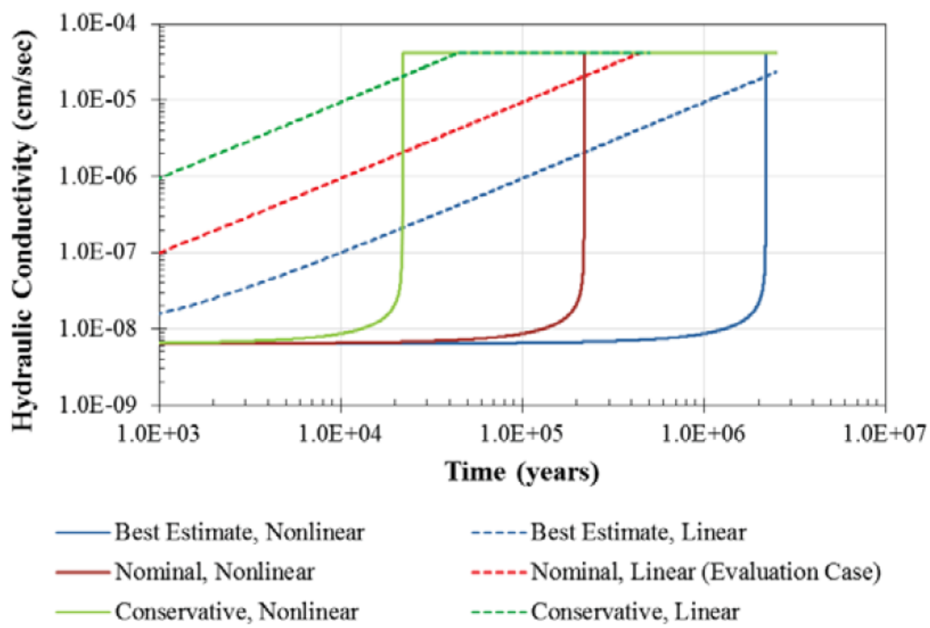

Figure 5: Log-Log Plot of the Hydraulic Conductivity Over Time Used in the DOE Response to the NRC RAI Questions on the DOE FY 2014 SDF SA (Reproduced from Figure SP-1.1 in SRR-CWDA-2016-00004, Rev. 1)

In 2015, the DOE collected five saltstone core samples from SDS 2A (SRR-CWDA-2016-00053). The results from the physical and hydraulic property measurements were reviewed in the NRC TRR *Saltstone Waste Form Hydraulic Performance* (ADAMS Accession

No. ML17018A137). In addition to the determination of hydraulic properties from actual saltstone core samples, the DOE conducted dynamic leaching tests on the release of Tc from the saltstone core samples as well as release of Tc from the simulated saltstone specimen at SREL, as shown in Figure 6 below. The results from those leaching tests were reviewed in the NRC TRR *Update on Projected Technetium Release from Saltstone* herein referred to in this TRR as the NRC Tc Release TRR (ADAMS Accession No. ML18095A122).

The CNWRA also evaluated the hydraulic conductivity of simulated saltstone samples under higher pressures. Although those accelerated tests are ongoing, they have provided some insight into the change in hydraulic conductivity with successive pore volumes of simulated groundwater migrating through the saltstone grout. As shown below in both Figure 6 and Figure 7, the hydraulic conductivity of the saltstone specimen generally decreased with additional pore volume exchanges in both the DOE and CNWRA experiments. That observation is significant because if this phenomenon occurs under field conditions, then the flow through saltstone, which directly affects radionuclide release, could be lower than is currently projected. Research is still ongoing to determine the mechanism(s) responsible for the apparent decrease in hydraulic conductivity. An understanding of the mechanism(s) causing the decrease is important to understand if those results will translate to field conditions.

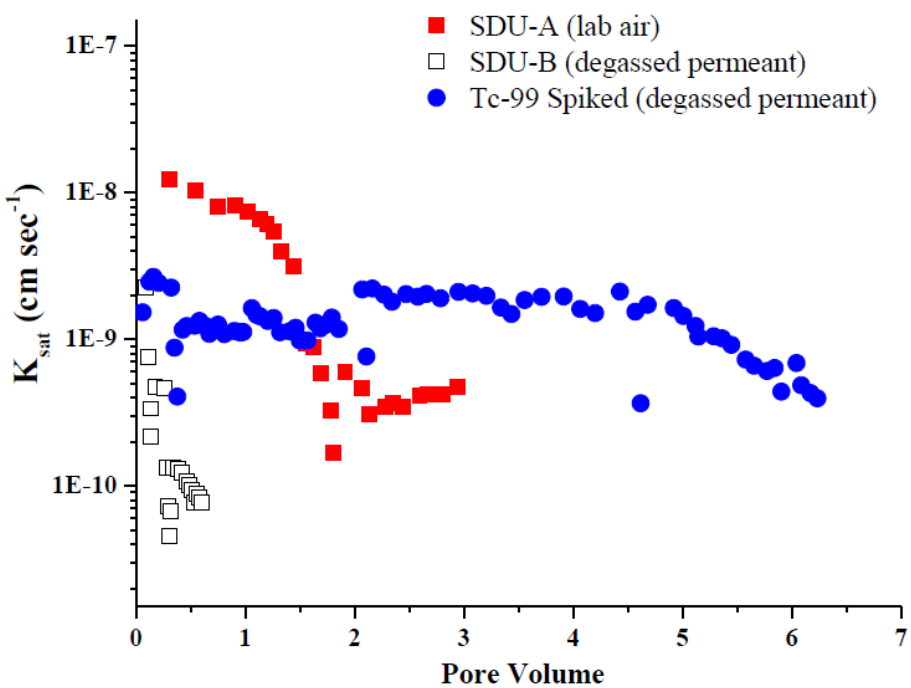

Figure 6: Saturated Hydraulic Conductivity of Dynamic Leach Method Samples (Reproduced from Figure 9 in SREL DOC No. R-16-0003)

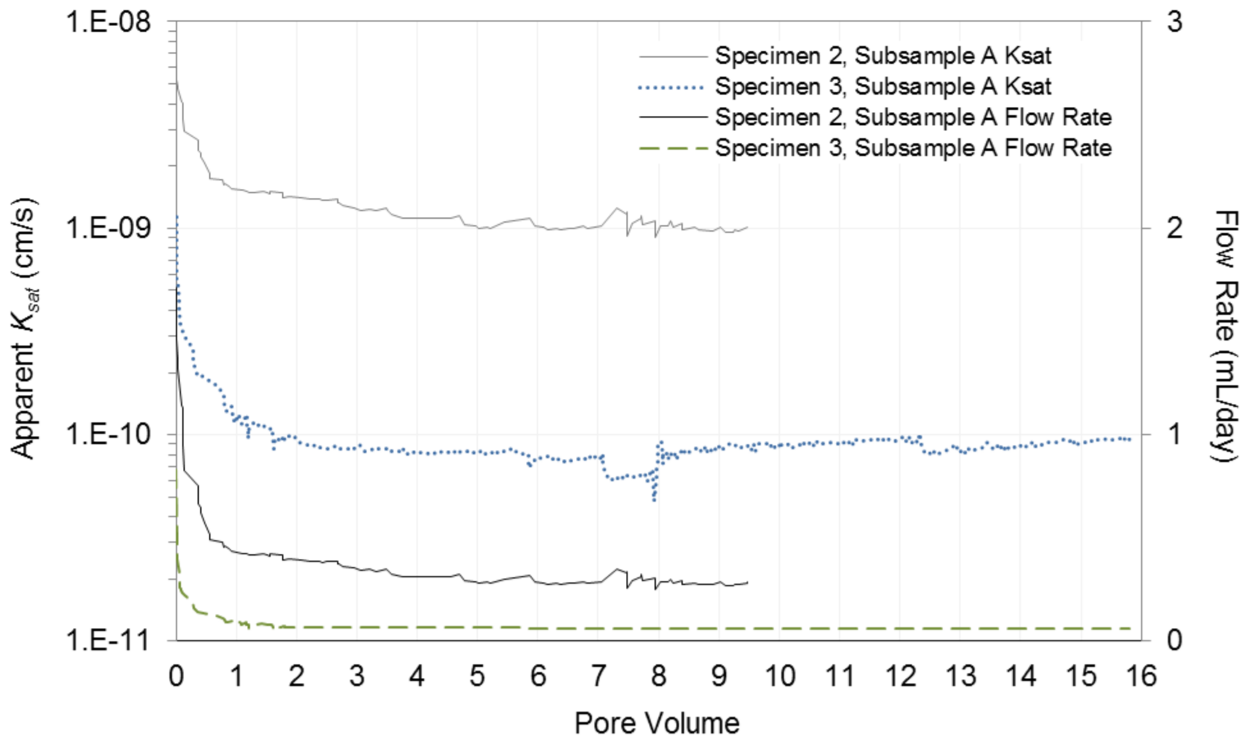

Figure 7: Comparison of Flow Rates and Apparent Saturated Hydraulic Conductivity of Two Simulated Saltstone Samples from the CNWRA (Reproduced From CNWRA Report in ADAMS Accession No. ML17221A038)

As discussed above, results from recent research appear to indicate that the initial hydraulic conductivity of field-emplaced saltstone may be lower than the DOE assumed in the Evaluation Case in the DOE SDF FY 2014 SA. However, the NRC staff notes: (1) the preliminary and ongoing nature of that research, (2) uncertainty in the mechanisms controlling the change in hydraulic properties, (3) uncertainty in the applicability of this research to field-emplaced saltstone due to differences in hydraulic pressure or head, and (4) uncertainty in the potential for degradation mechanisms in addition to decalcification to impact flow and contaminant transport. The NRC staff expects that continuation of the research at both the SREL and the CNWRA will reduce the uncertainty in those areas.

Although the degradation analyses in the Evaluation Cases in the DOE FY 2013 and FY 2014 SAs and the sensitivity cases provide insights into several future potential degradation states, it is unclear to the NRC staff whether those cases cover an adequate range of possible degraded states of saltstone. Based on the uniqueness and limited operational experience with saltstone, coupled with the long timeframes of interest, there is significant uncertainty in what degradation mechanism or mechanisms may affect saltstone performance and the corresponding timing and magnitude of degradation that could occur during the period of performance. Because of that uncertainty and the risk significance of saltstone performance as a barrier to radionuclide release, a more complete analysis of potential degradation mechanisms should be considered by the DOE. That analysis could screen out degradation mechanisms due to limited dose impacts or if there is a sufficient technical basis to conclude that a particular mechanism is unlikely to occur. However, any degradation mechanisms that cannot be screened out due to a limited dose impact or a low probability of occurrence should be considered in a degradation analysis. The degradation analysis may be simplified if multiple degradation mechanisms can be described with a similar mathematical representation.

To evaluate the conservatism of the assumed scenarios, additional information is needed to understand what reasonably foreseeable degradation scenarios could occur. In the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs, the DOE assumed that saltstone degradation would primarily be controlled by decalcification. The NRC staff is concerned that the DOE has not provided adequate support to exclude other degradation mechanisms, which could result in saltstone degrading more quickly than what is assumed in both the DOE Evaluation Cases. The NRC staff provided information in the Appendix to this TRR regarding additional degradation mechanisms that may affect saltstone performance.

Summary for MF 4.01

In the NRC 2013 SDF Monitoring Plan, the NRC staff expected to close MF 4.01 under both §61.41 and §61.42 after the NRC determines that support for modeled changes in the saturated hydraulic conductivity and diffusivity of saltstone during the performance period is sufficient.

The saltstone degradation model in the Evaluation Cases in both the DOE FY 2013 and 2014 SDF SAs is more defensible than in the Base Case in the DOE 2009 SDF PA and the model provided some additional risk insight. The mechanistic decalcification model provides insight into the potential doses due to gradual leaching of calcium and subsequent degradation to the hydraulic properties of the surrounding soils. Dynamic leaching studies also provided some insight into the potential evolution of saltstone hydraulic properties with several additional pore volume exchanges. Additional insights from those accelerated test studies should continue to be provided with additional pore volume exchanges. The development of information supporting the applicability of the results to long-term field conditions would greatly enhance the utility of the data.

Although the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs represent an improvement over the representation of saltstone degradation in the DOE 2009 SDF PA, sufficient bases have not been provided for excluding additional degradation mechanisms and excluding the coupling of degradation mechanisms in those two Evaluation Cases. The DOE document that described the development of a conceptual model for a future revised SDF PA (SRR-CWDA-2018-00006) indicated that several mechanical degradation mechanisms will be considered in “early release” and “fast flow path” scenarios in a future revised SDF PA. However, in that document (SRR-CWDA-2018-00006), the DOE indicated that those mechanical degradation mechanisms will not be included in the central scenario of the future revised SDF PA. The NRC staff will evaluate the technical bases for excluding those mechanisms from the central scenario, as well as any risk insights gained from including those mechanisms in the alternative scenarios, when it reviews the DOE future revised SDF PA. Similarly, the NRC staff will evaluate the coupling of saltstone degradation mechanisms in the future revised SDF PA. The DOE conceptual model development document (SRR-CWDA-2018-00006) indicated coupling of degradation mechanisms will be included in the central scenario in the future revised SDF PA.

Based on the limited information regarding the potential for additional and coupled degradation mechanisms to result in greater-than-projected degradation of saltstone, the NRC staff recommends keeping MF 4.01 open. Also, based on the expected risk significance of degradation on the rate of release of radionuclides, the NRC staff recommends that MF 4.01 remain prioritized as high priority under §61.41 and §61.42.

MF 4.02, "Waste Form Macroscopic Fracturing"

In the NRC 2013 SDF Monitoring Plan, the NRC staff described in MF 4.02 the NRC concern that the DOE Base Case assumption that the saltstone would remain intact (i.e., not fractured) for thousands of years was unrealistic. The NRC staff indicated that the DOE assumption was inconsistent with already observed fractures in emplaced saltstone. In Case K associated with the DOE 2009 SDF PA, the DOE assumed that saltstone would fracture significantly during the performance period. However, the NRC concluded in the 2012 TER that none of the sensitivity cases, including Case K, provided reasonable reassurance that the performance objectives would be met.

In the NRC 2013 SDF Monitoring Plan, the NRC staff described that saltstone fracturing was important to site performance because it: (1) increases flow through the saltstone, (2) shortens the diffusive length for radionuclide release, and (3) provides additional surface area for the progression of saltstone oxidation, which increases Tc release. In the NRC Tc Release TRR, the NRC staff stated:

> *The NRC staff was concerned that gas-phase transport of oxygen into saltstone fractures could lead to greater Tc oxidation than the DOE had predicted along fractures because of studies that appeared to show sensitivity of Tc releases to trace quantities of oxygen. The NRC staff also was concerned that fractures could subsequently become fast pathways for water transport and Tc release. A conceptual model in which Tc oxidation is sensitive to fracturing could cause large uncertainty in dose projections because the extent of fracturing that will occur in saltstone as it ages is highly uncertain.*

In the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs, the DOE assumed saltstone degradation was controlled by decalcification of the matrix. In the DOE document, *Crosswalk for Select Documents Related to the Monitoring Programs for the Saltstone Disposal Facility* (SRR-CWDA-2014-00002), the DOE stated, "Various mechanical influences that could lead to physical degradation (i.e., fracturing) were not explicitly modeled in the FY2013 SDF SA. However, the linear degradation model approach utilized is expected to bound any influences from fracturing." As NRC staff described above, it is not clear that the assumed linear degradation of the saltstone matrix due to decalcification in the two Evaluation Cases bounds influences from fracturing because other physically reasonable assumed degradation rates (e.g., non-linear) have periods over which the degradation rate increases more rapidly, exposing fresh saltstone to fractures and, potentially, water flow, at a greater rate. The DOE provided several sensitivity analyses in the FY 2013 and FY 2014 SAs and in response to the NRC RAI comments and questions that provided additional understanding of the risk significance of some of the processes associated with fractures.

In Section 4.4.1.3 of the DOE FY 2014 SDF SA, the DOE described that it only expected oxygen to enter the disposal structures via infiltrating water (i.e., aqueous-phase transport of oxygen). However, the DOE provided PORFLOW sensitivity analyses in both the FY 2013 and FY 2014 SAs and associated responses to the NRC RAI comments and questions that evaluated the effects of non-depleting oxygen sources on Tc release. Those sensitivity analyses provided insight into the potential significance of gas-phase transport of oxygen through fractured saltstone grout on the release of Tc. Those non-depleting oxygen sources were a non-mechanistic approach to modeling additional oxidation, which could represent oxidation due to gas-phase transport of oxygen in unsaturated fractures. However, the results did not entirely represent the effects of fracturing because the oxidized areas do not coincide with degraded hydraulic properties, as they would if the sources of oxygen represented fractures.

The modeled regions of non-depleting oxygen sources comprised 5%, 10%, and 20% of the saltstone volume and hasten the modeled consumption of the reducing capacity of the slag and consequently increase the Tc release rate. Both Figure 8 and Figure 9 below show that the non-depleting oxygen-source cases for the 46-m (150-ft) and 114-m (375-ft) diameter disposal structures result in the projected release of Tc occurring earlier in time than the Evaluation Case. However, the modeled peak rate of release (i.e., the maximum slope of the curves in those two figures), which is directly related to the magnitude of the peak dose, is more gradual in the oxygen-source cases than in the Evaluation Case. In response to NRC RAI Question SP-10, the DOE provided a dose approximation to the Member of the Public for each of the cases with varying percentages of non-depleting oxygen sources (SRR-CWDA-2016-00004, Rev.1). Figure 10 shows that the projected peaks from the non-depleting oxygen-source cases occur thousands of years earlier in time than the projected peak dose from the Evaluation Case and that the projected peak dose within 50,000 years of site closure decreased by more than a factor of 4 for the non-depleting oxygen sources sensitivity cases relative to the Evaluation Case. However, within 10,000 years of site closure, assuming that 20% of the saltstone was a non-depleting oxygen source, there was an increase in the projected peak fractional release by approximately a factor of three as compared to the Evaluation Case for the 46-m (150-ft) disposal structures (i.e., 3.0×10^{-2} versus 8.8×10^{-3}) and by approximately a factor of five (i.e., 1.4×10^{-1} versus 2.6×10^{-2}) for the 114-m (375-ft) disposal structures.

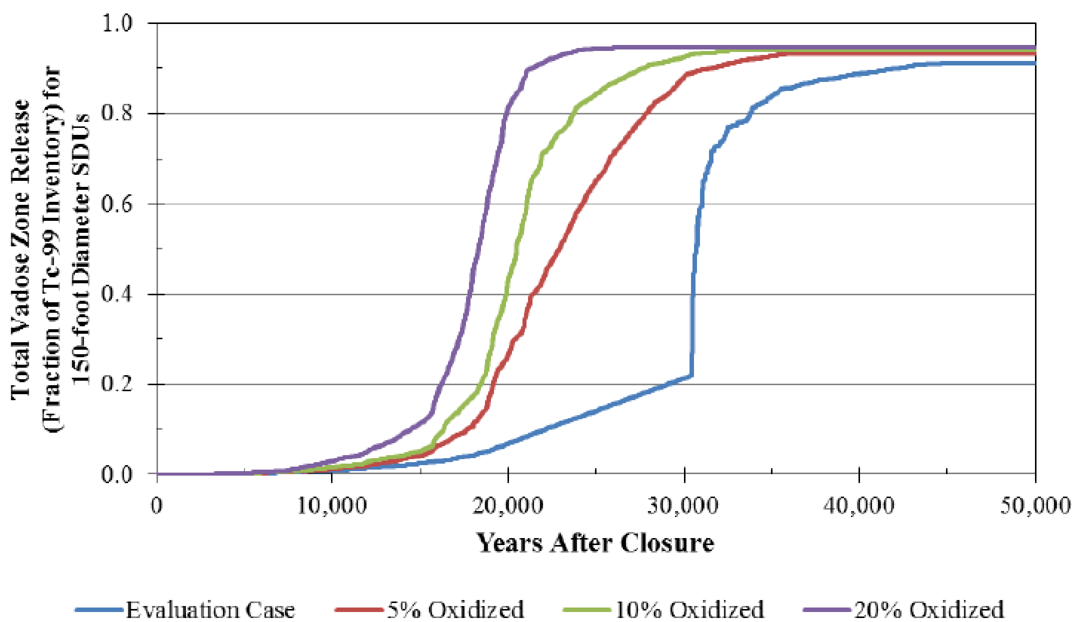

Figure 8: Projected Release of Tc to the Water Table from a 46-m (150-ft) Diameter Disposal Structure for Varied Percentages of Non-Depleting Oxygen Sources within Saltstone (Reproduced from Figure 5.6.7-12 in SRR-CWDA-2014-00006)

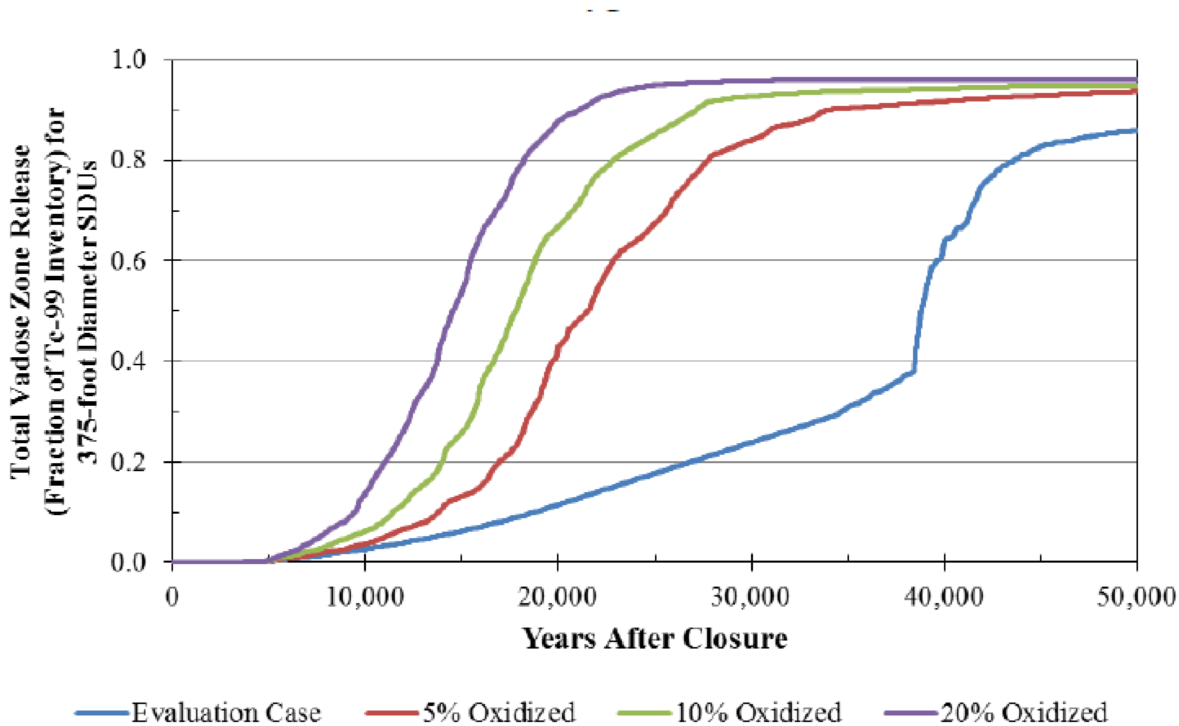

Figure 9 Projected Release of Tc to the Water Table from a 114-m (375-ft) Diameter Disposal Structure for Varied Percentages of Non-Depleting Oxygen Sources within Saltstone (Reproduced from Figure 5.6.7-12 in SRR-CWDA-2014-00006)

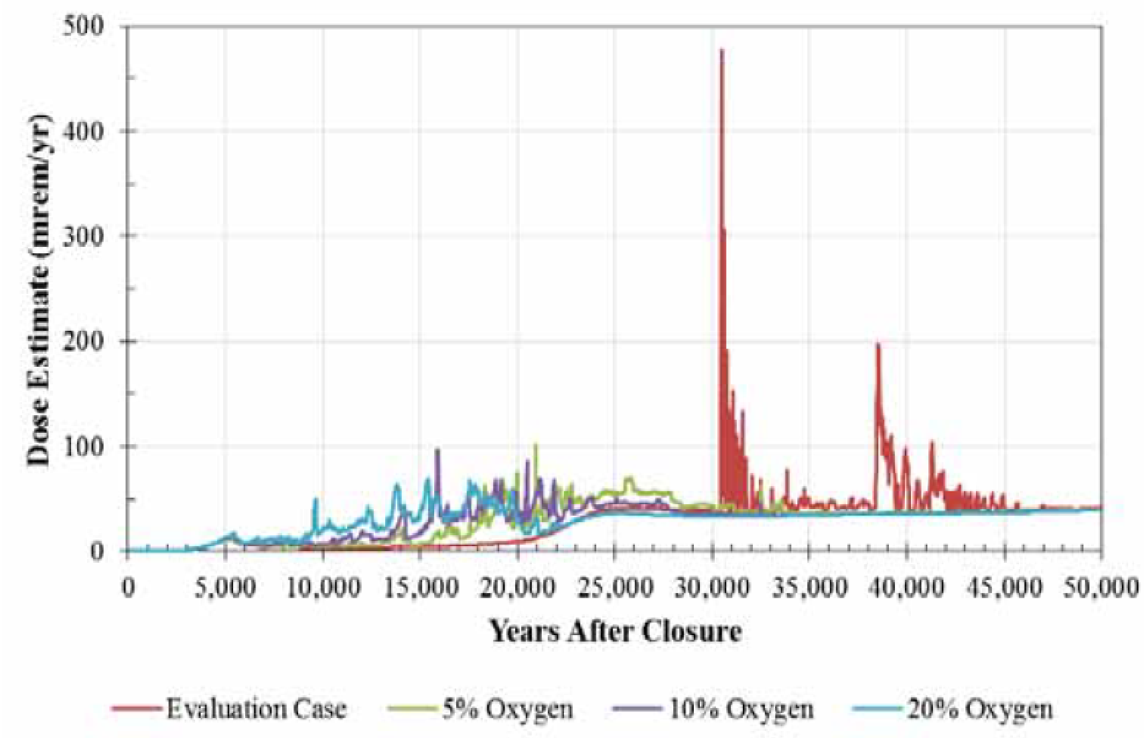

Figure 10: Projection of Peak Dose to the Member of the Public with Varying Percentages of Non-Depleting Oxygen Sources (Reproduced from Figure SP-10.4 in SRR-CWDA-2016-00004, Rev. 1)

In the NRC Comment SP-1 (ADAMS Accession No. ML14148A153) the NRC staff described that the non-depleting oxygen source sensitivity cases may not adequately represent oxidation in unsaturated fractures, in part, because of modeled re-reduction of Tc throughout the wasteform. The random placement of the oxygen sources also might not be representative of flow and transport of redox-sensitive radionuclides through a network of fractures in which Tc might encounter fast flow pathways and might not encounter reduced areas of saltstone.

In the DOE response to the NRC RAI Comment SP-1 (SRR-CWDA-2014-00099, Rev. 1), the DOE provided several counterpoints:

- The DOE did not expect that saltstone will degrade in such a way as to form a network of interconnected, unsaturated fractures within the 10,000 year evaluation period due to the stability of the local geography.
- The DOE developed the sensitivity analysis to show the potential impact from fracturing carrying increasing amounts of oxygen through the system; but, the cases were not intended to simulate expected conditions.
- The DOE sensitivity case modeled that, as the non-depleted oxygen sources expand, they became interconnected and created continuous paths of oxidation.

- The DOE expected Tc to become re-reduced and re-contained without a continuous source of oxygen (e.g., a fracture extending through the disposal structure roof to the upper mad mat).

- The DOE indicated that the doses associated with the continuous flow path due to sulfate attack on the columns did not exceed the performance objectives.

As previously discussed in this TRR, it is not clear to the NRC staff what degradation mechanisms and/or what coupling of degradation mechanisms may affect saltstone. Accordingly, the potential timing, rate, and extent of saltstone fracturing and, therefore, the potential for the formation of a network of interconnected fractures is not clear to the NRC staff. Although conclusions regarding the applicability of the non-depleting oxygen source sensitivity cases to the potential fracturing of saltstone cannot be drawn at this time, the NRC staff agrees with the DOE that these sensitivity cases do provide some insight into the potential dose impacts due to fractures. Specifically, the sensitivity analyses provided some insight into the potential effects of gas-phase transport of oxygen in unsaturated fractures. However, the sensitivity analyses, to date, have not provided information about the potential hydraulic effects of saltstone fracturing and the resulting impacts on radionuclide release or the effects of a decreased diffusive length due to fracturing.

The DOE conceptual model for the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs assumed that Tc will be re-reduced after it is initially oxidized if it migrates into areas of reducing grout. That conceptual model tends to result in a projected release that is larger than and occurs later than the projected release from cases that include non-depleting sources of oxygen within saltstone. In the non-depleting oxygen source case and the column sulfate attack case, saltstone is still projected to re-reduce Tc for thousands of years; but, not for as long of a period as is projected in the two Evaluation Cases. The DOE described that a continuous source of oxygen was necessary for Tc to not re-reduce. However, in a review of the bypass sensitivity analysis in the Tc Release TRR, the NRC staff concluded that there was not yet experimental evidence to support the DOE assumption that Tc would be re-reduced as it flowed through reducing saltstone grout.

In Section 5.6.6.2 of the DOE FY 2014 SDF SA, the DOE provided a sensitivity analysis to address the uncertainty in the expected extent of re-reduction of Tc in saltstone and disposal structure concrete and the potential effects on Tc release. In that analysis, a fraction of the oxidized Tc (i.e., 25%, 50%, 75%, 100%) was modeled as being transported directly out of the saltstone into the unsaturated zone. In the recent Tc Release TRR, NRC staff stated:

> *Although there is not yet experimental evidence for the assumption that oxidized Tc would be reduced if it flowed through chemically reduced saltstone, the bypass sensitivity analysis performed by the DOE does provide information about the importance of assumptions about Tc re-reduction to projections of Tc release. Model support does not yet exist to support a choice of what fraction of bypass best represents expected SDF field conditions. However, the DOE analysis summarized in Table 3 bounds the effects by providing values for 100% bypass. Therefore, incomplete re-reduction is projected to cause less than a factor of nine increase in peak Tc fluxes from the 150-foot disposal units and less than a factor of five increase in peak Tc fluxes from the 375-foot disposal structures, as compared to the 2014 Evaluation Case results. As shown in Table 3, longer term peak Tc fluxes are projected to decrease if Tc re-reduction is incomplete. That result occurs because increased Tc flux at earlier times is projected to remove*

> *some of the Tc from saltstone before Tc oxidation is projected to accelerate at later times.*

As indicted in the NRC staff RAI Question SP-2 (ADAMS Accession No. ML15161A541) on the DOE FY 2014 SDF SA, additional information was needed regarding the risk associated with gas-phase transport of oxygen into unsaturated fractures. As part of that, the NRC staff described that, although fractures may divert water away from the inventory, fractures have the potential to introduce much more oxygen into the wasteform than oxygen dissolved in infiltrating water. However, recent research and interpretation of earlier research has altered the NRC staff understanding of the risk significance of trace quantities of oxygen. That new understanding is directly related to the factors described in the Tc Release TRR that supported reducing the priority of MF 5.02 "Chemical Reduction of Tc by Saltstone." In the Tc Release TRR, the NRC staff stated:

> *First, the research results that originally prompted the NRC staff to develop MF 5.02 have been reinterpreted by the NRC staff based on more recent research results and the concern that the redox state of Tc is sensitive to trace quantities of oxygen has been reduced. Second, data from experiments conducted by the DOE with cores of field-emplaced saltstone showed that both cores leached with deareated liquid and cores leached with liquid equilibrated with laboratory air released similar concentrations of Tc and those concentrations were consistent with releases from reduced Tc solid phases. Third, based on the current knowledge of the inventory, solubility, and sorption of Tc in saltstone, Tc mobilization in reduced saltstone is expected by the NRC staff to be as or more important to performance than Tc mobilization in oxidized saltstone unless there is significant saltstone oxidation prior to contact with water (e.g., from oxygen transport in unsaturated fractures) or significant channelization of flow in oxidized areas of saltstone. Thus, the extent and timing of fracturing relative to the timing of water flow through saltstone is a significant source of uncertainty in projecting Tc release.*

<u>*Summary for MF 4.02*</u>

The NRC 2013 SDF Monitoring Plan described that the NRC staff expected to close MF 4.02 under both §61.41 and §61.42 after the NRC determines that model support for the assumed formation of macroscopic fractures in saltstone during the performance period was sufficient. DOE has not provided sufficient information to justify the assumed degradation mechanisms. Accordingly, the timing, rate, and extent of saltstone fracturing is not clear. Based on the uncertainty in saltstone fracturing, the NRC staff recommends keeping MF 4.02 open under both §61.41 and §61.42.

Regarding the risk significance of saltstone fracturing, recent research and reinterpretation of earlier research has reduced some of the NRC staff concerns associated with gas-phase transport of oxygen in unsaturated fractures. In addition, if flow through reducing saltstone grout is the risk-controlling source of radionuclide release, then saltstone fracturing could ultimately reduce radionuclide release as fractures may decrease the interaction between infiltrating water and the saltstone grout. However, fracturing could still result in increased radionuclide release relative to the DOE Evaluation Case due to decreased diffusive lengths, oxidation of saltstone grout prior to contact with water, and increased flow through saltstone grout, depending on the type and extent of fracturing (e.g., large-scale network of small fractures). Therefore, based on the potential risk significance of fracturing on radionuclide release, the NRC staff recommends that MF 4.02 remain prioritized as high priority under both §61.41 and §61.42. The DOE document that described its development of a conceptual model for a future revised SDF PA (SRR-CWDA-2018-00006) indicated that the effects of fracturing will be considered in a "fast flow paths" scenario; but, not in the central scenario. When the NRC staff reviews the DOE future revised SDF PA, the NRC staff will evaluate the basis for excluding fracturing from the central scenario, as well as any risk insights gained from including fracturing in an alternative scenario.

MF 10.02, "Defensibility of Conceptual Models"

The assumed conceptual model for saltstone degradation directly affects the modeled water flow through saltstone as well as its rate of oxidation, both of which directly affect projected radionuclide release. Thus, the conceptual model of saltstone degradation has a significant effect on projected SDF performance. As discussed in the context of MF 4.01 above in this TRR, the NRC staff is concerned that insufficient support has been given for excluding a variety of mechanical and chemical degradation mechanisms from the models of SDF performance. Several of those degradation mechanisms are discussed further in the Appendix to this TRR. Although the DOE indicated that those degradation mechanisms will be bounded by the assumption that the degradation rate is linear, as previously described in this TRR, there is insufficient model support to conclude that assuming a linear degradation rate is either realistic or bounding. For example, if the peak dose is sensitive to the peak degradation rate, because the peak degradation rate determines the rate at which fresh saltstone is exposed to water, then a conceptual model of saltstone degradation that results in periods of faster degradation than a linear degradation rate does (e.g., see Figure 3 above) may lead to greater peak dose projections.

The DOE document that described the development of a conceptual model for a future revised SDF PA (SRR-CWDA-2018-00006) indicated that several mechanical degradation mechanisms will be considered in a future revised SDF PA in an "early release" scenario and a "fast flow paths" scenario. However, that DOE document indicated that those mechanical degradation mechanisms will not be included in the central scenario. When the NRC staff reviews the DOE future revised SDF PA, the NRC staff will evaluate the basis for excluding fracturing from the central scenario, as well as any risk insights gained from including fracturing in an alternative scenario. Similarly, potential coupling of degradation mechanisms is an important conceptual feature models of cementitious materials.

Summary for MF 10.02

The NRC 2013 SDF Monitoring Plan described that the NRC staff expected to close MF 10.02 under both §61.41 and §61.42 after the DOE updates the SDF PA and the NRC determines that the conceptual modes are appropriate. Although the conceptual model of saltstone degradation in the Evaluation Cases in the DOE FY 2013 and FY 2014 SDF SAs appear to be more realistic than the conceptual model of saltstone degradation in the DOE 2009 SDF PA (i.e., no saltstone degradation for 20,000 years), the NRC staff continues to have significant concerns about the degradation mechanisms that were included in those Evaluation Case models. Therefore, the NRC staff recommends keeping MF 10.02 open under both §61.41 and §61.42 as a high-priority monitoring factor.

MF 10.05, "Moisture Characteristic Curves"

In the NRC 2012 TER, the NRC staff identified concerns regarding the greater-than-expected reduction in modeled water flow through cementitious materials due to the assumed MCCs in the DOE 2009 SDF PA. In response to many NRC staff concerns, the DOE provided a Case K analysis, which did not assume decreasing hydraulic conductivity with decreasing saturation (i.e., DOE assumed that the relative permeability was equal to 1.0 at all saturations) (ADAMS Accession No. ML13100A076). In the NRC 2013 Monitoring Plan, the NRC staff described in MF 10.05 that the NRC would monitor the DOE development of model support for any MCCs used to modify the assumed hydraulic conductivity of saltstone.

In both the DOE FY 2013 and FY 2014 SDF SAs, the DOE again used MCCs for saltstone. However, the MCCs in these documents, versus the DOE 2009 SDF PA, varied in time to simulate degradation (SRR-CWDA-2013-00062, SRR-CWDA-2014-00006). The assumed saturated and unsaturated hydraulic conductivities in the Evaluation Case in the FY 2014 SA are shown in Figure 11[6].

The unsaturated hydraulic conductivity values are derived in the PORFLOW near-field model as a function of the assumed saturated hydraulic conductivity, MCCs, and saltstone saturation, which all vary in time. The saturated hydraulic conductivity values were direct inputs into the PORFLOW near-field model and were based on the DOE assumed rate of degradation, as discussed previously in this document. The MCCs, which are also direct inputs into the PORFLOW model, were assumed to vary in time to simulate degradation, as discussed previously. Then, based on those model inputs and the saturation calculated in the PORFLOW model, PORFLOW selects the corresponding unsaturated hydraulic conductivity from the assumed MCC tables of saturation versus unsaturated hydraulic conductivity for each time interval. The PORFLOW calculated saturation of saltstone in each of the disposal structures ranged from 99.86% to 100% within 10,000 years. For the unsaturated hydraulic conductivity values shown in Figure 11, the NRC staff cross-referenced the saltstone saturations calculated within PORFLOW with the corresponding relative permeabilities from the assumed MCC look-up tables for several time intervals.

[6]Figure 11 only includes values for several time intervals, although the DOE used additional intermediate MCCs at additional time intervals.

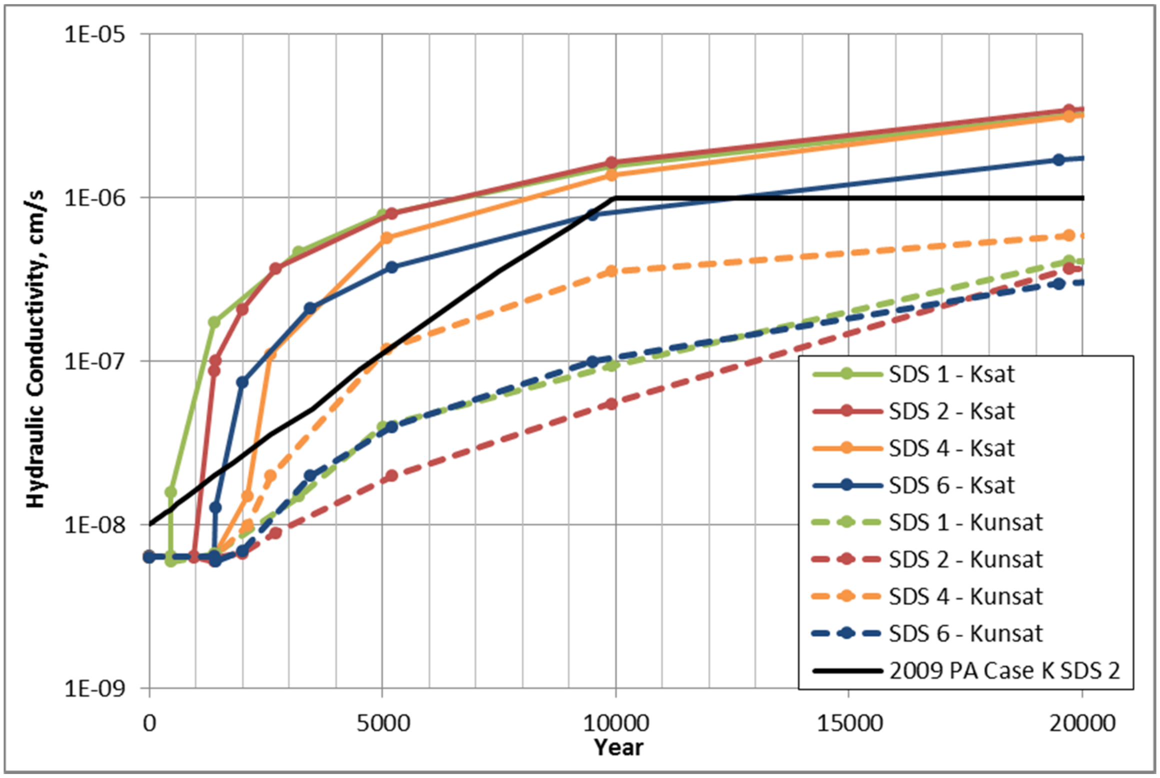

Figure 11: Assumed Saturated and Unsaturated Hydraulic Conductivity of Saltstone in the Evaluation Case from the DOE FY 2014 SA (Prepared from the DOE PORFLOW Data)

Based on the assumed saturated hydraulic conductivity values, MCCs, degree of saturation, and the corresponding relative permeabilities shown in Figure 11, the DOE projected that the hydraulic conductivity of unsaturated saltstone would decrease relative to the saturated hydraulic conductivity by a factor of up to 30 for SDS 2 (and the other 46-m [150-ft] disposal structures) within 10,000 years. For SDS 6 (and the other 114-m [375-ft] disposal structures), the relative permeability was projected to decrease by up to a factor of approximately 10 within 10,000 years. A decrease in permeability of a factor of 30 with a decrease in saturation of 0.14% is not consistent with other cementitious materials, as shown in Figure 2.7-4 of the 2012 NRC SDF TER (ADAMS Accession No. ML121170309).

Recent DOE research (SRR-CWDA-2016-00053) indicated that saturation of the SDS 2A core samples and the laboratory-prepared samples were only 70% - 75% saturated. The NRC staff described in the report for the August 2016 NRC Onsite Observation Visit (OOV) to the SDF (ADAMS Accession No. ML16147A197) that it was not clear if the relatively low saturation of the samples relative to the assumed saturation in the FY 2014 SA reflected actual field-emplaced saltstone conditions or if the observed saturations were due to: (1) desiccation during sample handling; or (2) a difference in how the values were calculated (e.g., by weight versus by volume). If the saturation is in the range of 70% - 75%, then the relative permeability could be significantly less than the saturated hydraulic conductivity. Accordingly, the NRC staff determined that additional information on the actual saturation of saltstone is needed to understand the hydraulic performance of saltstone.

In an effort to provide support for the assumed MCCs, the DOE developed a method for measuring the unsaturated hydraulic conductivity of fractured materials (SRNL-STI-2013-00522;

SRNL-STI-2014-00618). The DOE used an outflow extraction experiment to validate the method using a series of shimmed glass plates with a wedge geometry. Van Genuchten/Mualem parameters were estimated for the study; but, were not in agreement with the analytic solutions for saturation and relative permeability. The authors of the study indicated that the reason for the discrepancy was unknown; but, may be due to the uncertainty in the wetting angle. In addition to uncertainty in the wetting angle, there was also uncertainty in the analytic solution and application of the method to saltstone. That study represented the potential type of information that the NRC staff needs to have in order to have confidence in projections of flow in degraded and unsaturated saltstone, depending on the risk significance of the assumed MCCs. However, that study was indicative of the challenges associated with developing support for unsaturated flow in saltstone. Even in an idealized system using glass plates in the laboratory, there was a discrepancy between the results and the analytic solutions. Applying that approach to a much more complex medium, such as saltstone, adds significant uncertainty. Furthermore, projections of unsaturated flow through a hypothetically degraded saltstone material adds yet more uncertainty.

The assumed saturated hydraulic conductivity values in the Evaluation Case in the FY 2014 SDF SA typically exceeded those of Case K for the 46-m (150-ft) diameter disposal structures by up to approximately a factor of 10, as shown in Figure 12. However, Case K assumed that the relative permeability of saltstone was 1.0 at all saturations, whereas the Evaluation Case included MCCs that decrease the hydraulic conductivity according to the degree of saturation. Intermediate PORFLOW model results from Case K and the Evaluation Case in Figure 13 show that those two assumptions nearly balance each other with respect to the cumulative volumetric flow and the rate of increase in flow through saltstone. However, the NRC staff remains concerned that there is significant uncertainty associated with the degradation of saltstone and, therefore, the saturated hydraulic conductivity, as well with the assumed MCCs and the saturation of saltstone.

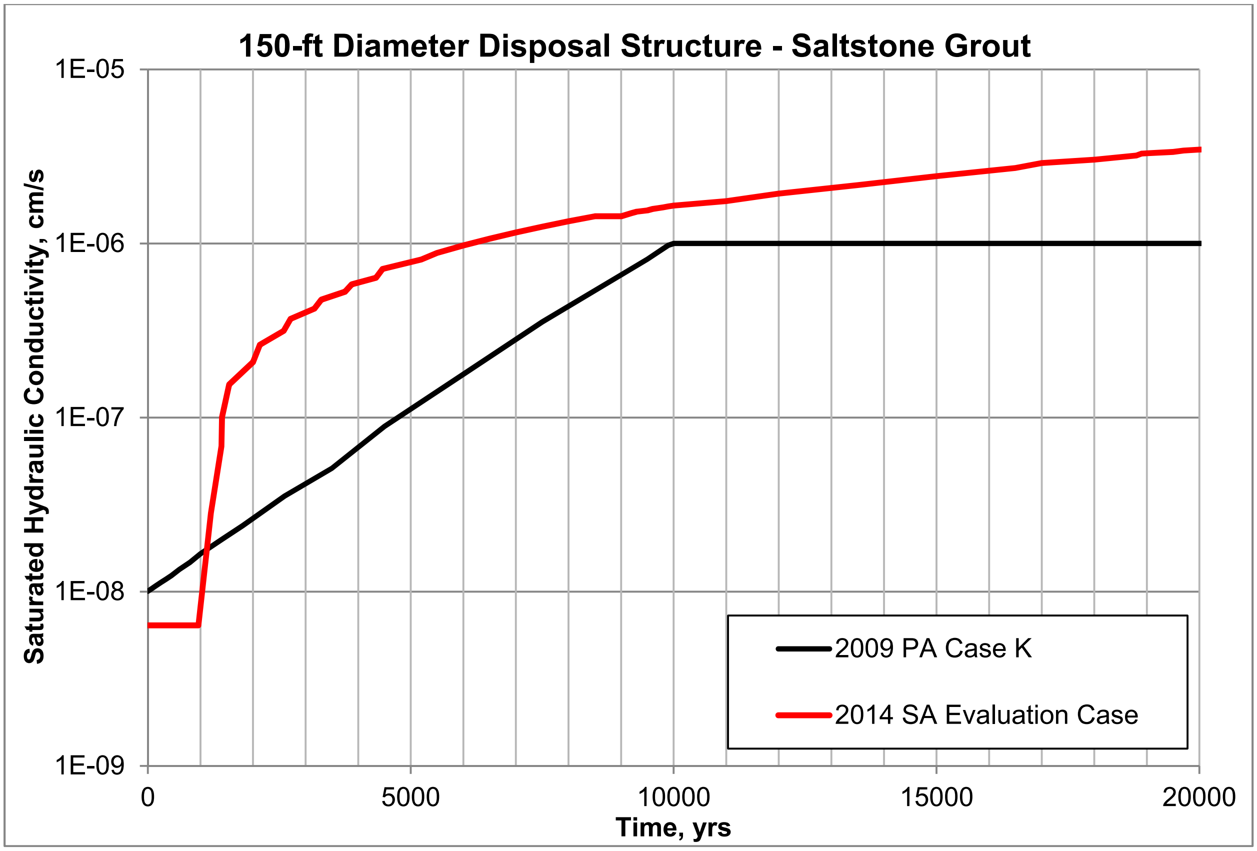

Figure 12: Assumed Saturated Hydraulic Conductivity of Saltstone Grout in 46-m (150-ft) Diameter Disposal Structures (Prepared From the DOE PORFLOW Data)

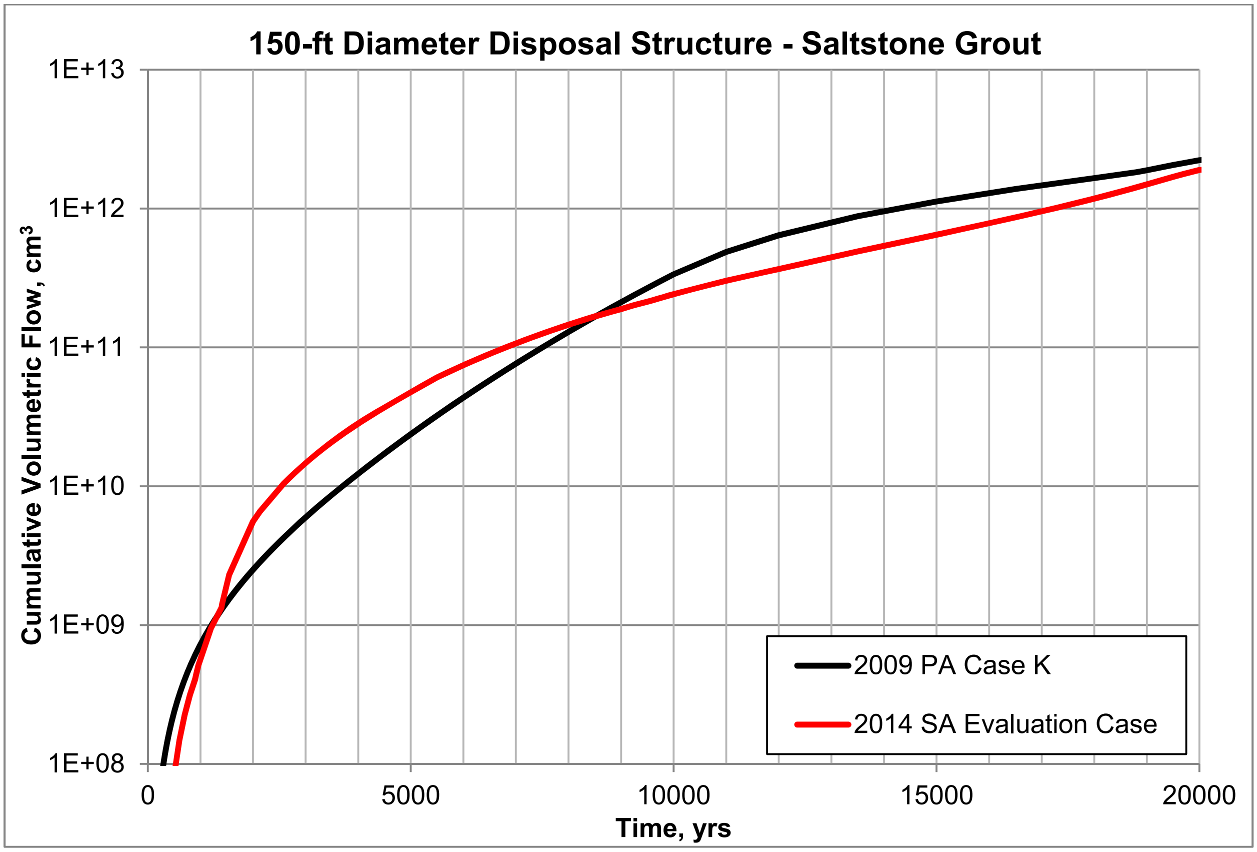

Figure 13: Cumulative Volumetric Flow through Saltstone Grout in 46-m (150-ft) Diameter Disposal Structures (Prepared from the DOE PORFLOW Data)

Summary for MF 10.05

In the NRC 2012 TER, the NRC staff identified concerns regarding the greater-than-expected reduction in modeled water flow through cementitious materials due to the assumed MCCs in the DOE 2009 SDF PA. In response to many NRC staff concerns, the DOE provided a Case K analysis, which did not assume decreasing hydraulic conductivity with decreasing saturation (i.e., DOE assumed that the relative permeability was equal to 1.0 at all saturations) (ADAMS Accession No. ML13100A076). In the NRC 2013 Monitoring Plan, the NRC staff described in MF 10.05 that the NRC would monitor the DOE development of model support for any MCCs used to modify the assumed hydraulic conductivity of saltstone.

The DOE relied on MCCs in the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs to model flow through unsaturated saltstone grout. The assumed MCCs and saturation of saltstone resulted in a significant decrease in flow. Based on that decrease in flow and the uncertainty associated with the assumed MCCs and saturation of saltstone, the NRC staff recommends keeping MF 10.05 open and increasing the priority of MF 10.05 from low to medium under §61.41 and §61.42.

Follow-up Actions

There are no Follow-up Actions related to this TRR.

Open Issues

There are no Open Issues related to this TRR.

Conclusions

Beginning with the NRC 2012 SDF TER and in subsequent public meetings, OOVs, RAI Comments, and RAI Questions, the NRC staff expressed concerns about the range of saltstone degradation mechanisms included in the DOE models of saltstone performance and the coupling of those mechanisms. Relative to the DOE 2009 SDF PA, which assumed that saltstone would not degrade hydraulically for 20,000 years after site closure, the Evaluation Cases in both the DOE FY 2013 and FY 2014 SDF SAs took a more realistic approach by including carbonation and decalcification as saltstone degradation mechanisms. However, the NRC staff concludes that insufficient support has been provided to exclude other degradation mechanisms from the DOE models of saltstone performance. The DOE document that described the development of a conceptual model for a future revised SDF PA (SRR-CWDA-2018-00006) indicated that several mechanical degradation mechanisms will be considered in "early release" and "fast flow path" scenarios in a future revised SDF PA. However, that document indicated that those mechanical degradation mechanisms will not be included in the central scenario of the revised SDF PA. When the NRC staff reviews the DOE future revised SDF PA, the NRC staff will evaluate the basis for excluding fracturing from the central scenario, as well as any risk insights gained from including fracturing in an alternative scenario. Similarly, the NRC staff will evaluate assumptions regarding the coupling of saltstone degradation mechanisms in the future revised SDF PA. The DOE conceptual model development document (SRR-CWDA-2018-00006) indicated coupling of degradation mechanisms will be included in the central scenario in the future revised SDF PA.

In the FY 2014 SDF SA, the DOE indicated that assuming a linear degradation rate of saltstone bounds the potential effects of degradation mechanisms that were not included in that analysis. However, the NRC staff concludes that insufficient support has been provided to determine that assuming the degradation rate is linear bounds the projected peak dose. For that reason and the additional specific reasons provided in the context of specific monitoring factors in this TRR, the NRC staff recommends keeping MF 4.01, MF 4.02, MF 10.02, and MF 10.05 open under both §61.41 and §61.42.

Because saltstone degradation directly affects the ability of saltstone to retain radionuclides and the potential effects of saltstone degradation could be large, MF 4.01, MF 4.02, and MF 10.02 were originally designated as high-priority monitoring factors. Since the creation of those monitoring factors in 2013, no additional evidence supports reducing the priority of those monitoring factors. MF 10.05 was originally designated as a low-priority MF because MCCs were not used in the DOE model that the NRC staff weighted most heavily in the NRC 2012 SDF TER (i.e., Case K). However, the Evaluation Cases in the FY 2013 and FY 2014 SAs models both use MCCs to reduce the projected hydraulic conductivity of saltstone relative to its assumed saturated hydraulic conductivity. For that reason, the NRC staff recommends increasing the priority of MF 10.05, from low to medium under both §61.41 and §61.42.

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APPENDIX: Additional Information Regarding Potential Degradation Mechanisms

In addition to the assumed decalcification degradation mechanism of saltstone, other potential degradation processes exist that could resulting in cracking. In general, cementitious materials are susceptible to cracking due to their inherently low tensile strength. Once the tensile strength of the material is exceeded by the tensile stress, cracks develop. Saltstone has several characteristics that could limit its strength and increase its susceptibility to additional stresses relative to other cementitious materials, including: (1) a high water-to-cement ratio, (2) lack of aggregate, (3) pozzolanic replacement, (4) large, unreinforced monolith (5) method of grout placement, and (6) curing temperature. The influence of these characteristics on degradation is discussed in more detail below with respect to the following potential degradation mechanisms:

- Shrinkage Cracking
- Freeze-Thaw Cycling and Cracking
- Thermal Gradients and Cracking
- Expansive-Phase Formation and Cracking
- Static or Dynamic Settlement-Induced Cracking
- Steel Corrosion-Induced Cracking
- Microbial Degradation

Those mechanisms do not represent an exhaustive list of degradation mechanisms that could be applicable to saltstone; however, they serve as examples of the types of mechanisms that should be considered in a model of the Saltstone Disposal Facility (SDF) performance. In general, degradation mechanisms should either be excluded based on evidence that specifically supports exclusion of that mechanism, or included non-mechanistically based on evidence that the non-mechanistic representation either adequately represents or bounds the mechanism. For example, a statement that a particular overall degradation rate bounds the potential cumulative effect of all of the degradation mechanisms that were considered to be plausible but not explicitly included in the model should be supported by all three of the following types of analyses:

1. A systematic screening of mechanisms that were included in and excluded from the model;

2. An explanation of the assumed maximum rate of degradation of the mechanisms that were considered plausible but were not included in the model; and

3. A demonstration that the assumed overall degradation rate exceeds or represents the cumulative effects of the included and excluded degradation mechanisms.

Shrinkage Cracking

Cementitious materials can undergo volume changes due to changes in moisture content, chemical reactions[7], temperature changes[8], and loading. These volume changes in turn create

[7] The formation of expansive phases such as ettringite and corrosion products are discussed separately under sections on Expansive-Phase Formation and Steel-Induced Corrosion Cracking.
[8] Temperature-related volume changes are discussed further under sections on Thermal Gradients and Freeze-Thaw Cycling below

stresses in the cementitious systems. The likelihood of cracking is a function of the magnitude of the stress and the strength of the cementitious material. The characteristics of saltstone and its placement in the disposal structures can both increase and decrease its susceptibility to cracking relative to ordinary concrete systems, such as:

Water-to-cement ratio: The high water-to-cement ratio for saltstone increases the likelihood of grout cracking due to a combination of reduced tensile strength and increased shrinkage.

Aggregate: The lack of aggregate in saltstone will tend to increase shrinkage relative to aggregate-containing cementitious materials as aggregate restrains shrinkage (American Concrete Institute (ACI) 209.1R-05). That increased shrinkage can then lead to increased cracking.

Pozzolans: The large pozzolanic replacement (i.e., fly ash and blast furnace slag in place of ordinary Portland cement) in saltstone grout will tend to slow the hydration reactions and help limit thermal excursions, but the delayed curing also delays strength development. A large pozzolanic replacement can also increase autogenous shrinkage relative to Portland-cement concrete without pozzolans. (ACI 207.1R-05; ACI 209.1R-05)

Grout Placement: Cementitious materials with a large slump, no vibration applied, and large loading can be susceptible to settlement cracking (ACI 224.1R-07).

Humidity: The humid environment present in the disposal structures and additional grout pours will limit plastic shrinkage during initial curing; however, drying shrinkage over longer periods of time will still occur. The extent of drying shrinkage in saltstone is unclear because of the limited operational history and uniqueness of saltstone.

Admixtures: The addition of water-reducing admixtures can increase shrinkage (ACI 209.1R-05)

Freeze-Thaw Cycling

In the NRC 2007 Monitoring Plan for the SDF (ADAMS Accession No. ML070730363), the NRC questioned whether or not freeze-thaw cycling could result in the degradation of saltstone grout and the disposal structures. In response to the NRC RAI Comments (PA-13 and VP-2) during the NRC review of the DOE 2009 SDF Performance Assessment (PA), the DOE provided several lines of evidence suggesting that freeze-thaw cycling was not a concern for saltstone grout, including: (1) limited durations of subfreezing temperatures, (2) High-Density Polyethylene (HDPE) limiting rainwater intrusion, and (3) the disposal structures being located beneath the freezing zone of soil in South Carolina after the placement of the closure cap (SRR-CWDA-2011-00044, Rev. 1).

The DOE provided grout temperature profiles from nested thermocouples from within the disposal structures from January 2009 to December 2017 (SRR-CWDA-2018-00015). The thermocouples recorded temperatures at varying depths, which illustrate temperature changes that vary with fill height, curing time, and the outside temperature. Those data were obtained from Saltstone Disposal Structure (SDS) 4 Cells D, E, F, J, K, and L; SDS 2A, SDS 2B, SDS 5A, and SDS 5B. Based on the DOE thermocouple data from January 2009 to December 2017, the minimum temperature recorded for saltstone grout was 12˚C in SDS 4 Cell K (SRR-CWDA-

2018-00015), as shown in Figure 14 below. During this period, the minimum temperature recorded[9] in Augusta, Georgia was -11˚C during the month of January, 2014. The minimum temperature recorded[10] since 1881 for Augusta, Georgia was -17˚C in 1985.

Based on the historic record, the potential exists for the ambient temperature to be lower than was observed during this period. However, there are several mitigating factors, which limit the potential for freezing conditions in the grout. Prior to site closure, freeze-thaw cracking is expected to be limited for the following reasons:

1. Lifts lower in the disposal structures will be insulated to some extent by overlying saltstone lifts.

2. The uppermost lift will be exposed to the coldest temperatures. However, the uppermost lifts are also the youngest lifts, which may still be generating heat from exothermic curing reactions.

3. The minimum grout temperature remained above 10˚C during this observational period as outside air temperatures were as low as -11˚C. That was likely due to a combination of insulation from the saltstone and disposal structure as well as ongoing exothermic reactions from the curing process.

4. The high salt content of saltstone would likely lower the temperature required for differential volume changes to occur due to freeze-thaw cycling.

After site closure, freeze-thaw cracking is expected to be limited for the following additional reasons:

5. As the DOE described in the document SRR-CWDA-2011-00044, Rev. 1, the HDPE layer for the cylindrical disposal structures will limit infiltration into the disposal structures. Accordingly, availability of free water, which would be more susceptible to freezing than saltstone pore liquid, will be limited after site closure.

6. The cylindrical disposal structures are backfilled prior to filling which will limit exposure of saltstone to colder temperatures prior to site closure. In addition, all of the disposal structures will be covered with backfill and a closure cap upon closure. In Section 2.5.3.2 of the NRC 2012 SDF Technical Evaluation Report (TER), the NRC described that the soil only freezes to a depth of 25 cm (10 in) in South Carolina. Accordingly, the closure cap will preclude freezing conditions within the disposal structures after site closure.

Based on the above information, the NRC staff has determined that freeze-thaw cycling is unlikely to contribute to the degradation of saltstone.

[9] Minimum recorded temperatures from January 2009 to December 2017 at the Augusta Daniel airport in Georgia were obtained from https://www.wunderground.com/history/airport/KDNL.
[10] Minimum historic temperature for Augusta, GA was obtained from http://www.intellicast.com/local/history.aspx?location=USGA0032

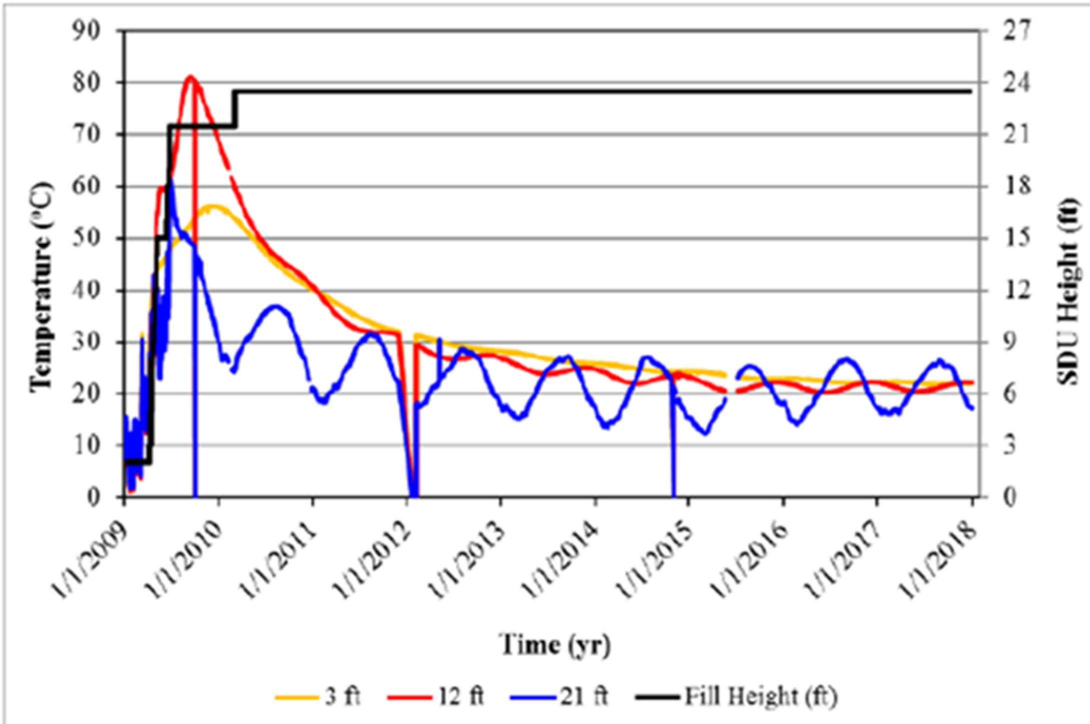

Figure 14 Thermocouple and Fill Height Data from SDS 4 Cell K (SRR-CWDA-2018-00015)

Thermal Gradients

Spatial and temporal thermal gradients develop during curing, which can result in differential volume changes and, consequently, the development of stress. The DOE collected core samples from SDS 2A for determination of saltstone properties. The samples were collected at heights ranging from 4.7 m (15.5 ft) to 5.2 m (17 ft). The thermocouple data in Figure 15 shows that the maximum grout temperature was approximately 68˚C for the thermocouple located at 4.7 m (15.5 ft). As the core samples were collected from 4.7 m (15.5 ft) to 5.2 m (17 ft), the core samples are representative of samples that experienced the maximum curing temperature for SDS 2A. Also, a maximum curing temperature of 68˚C in SDS 2A is similar to the maximum observed curing temperature in the other saltstone-filled disposal structures (i.e., SDS 4 Cells D, E, F, J, and L; SDS 2B, SDS 5A, SDS 5B) with the exception of Cell K in SDS 4, which reached a maximum curing temperature of approximately 80˚C (SRR-CWDA-2018-00015).

For determination of the saturated hydraulic conductivity, samples were taken from intact[11] portions of the core samples (SRNL-STI-2016-00106). The mean saturated hydraulic conductivity of the intact core samples was <1.6×10^{-9} cm/s, which is less than the DOE assumed initial saturated hydraulic conductivity in the Evaluation Cases of 6.4×10^{-9} cm/s (SRR-CWDA-2016-00051). As the saturated hydraulic conductivity is a function of the pore volume,

[11] The report authors stated that through-diameter fractures were observed at and between cold joints due to core-drilling and extraction processes (SRNL-STI-2016-00106)

size, distribution, and connectivity, curing temperatures up to approximately 70˚C do not appear to adversely affect the saltstone pore structure. However, it is not clear from the core sample analysis whether thermal fracturing could have occurred on a larger spatial scale than the core samples. Some of the through-diameter fractures observed at the Savannah River National Laboratory (SRNL) could be related to thermal gradients. It is also not clear how larger fractures due to potential thermal gradients would affect radionuclide release. Fracturing decreases the diffusive length of saltstone; however, fracturing also tends to divert water around the saltstone matrix and radionuclide inventory. Additional information could be developed by the DOE on the risk significance of degradation due to thermal degradation.

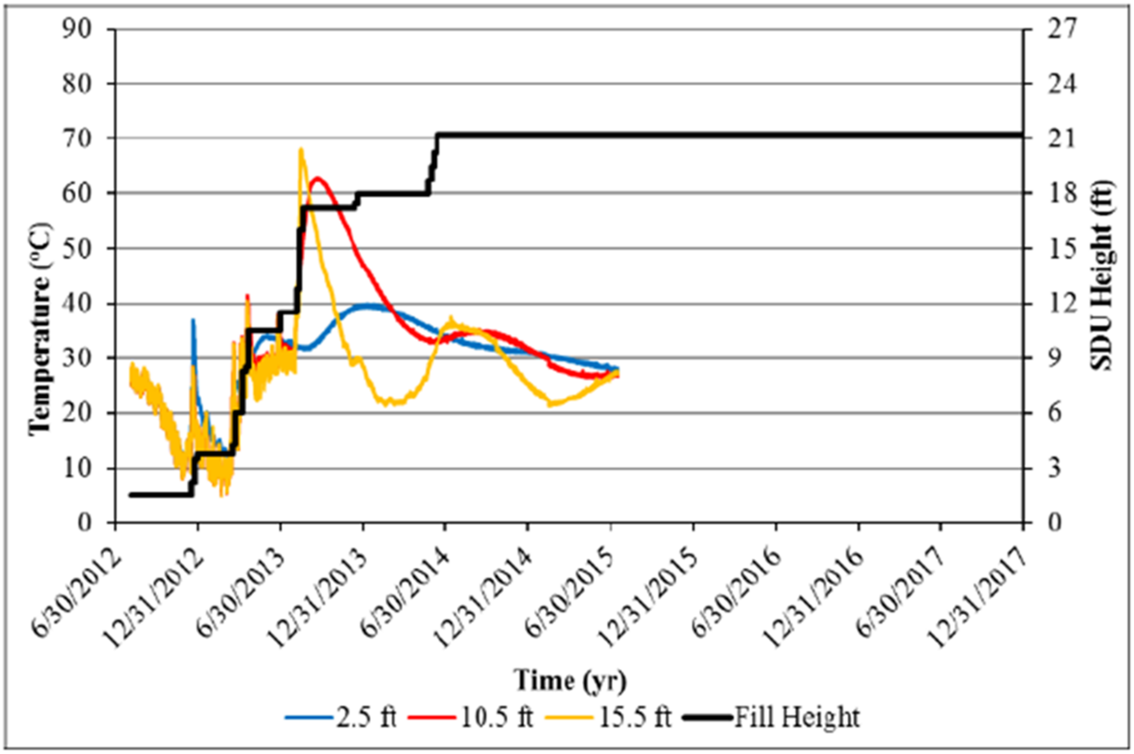

Figure 15 Thermocouple and Fill Height Data from SDS 2A, Train C (SRR-CWDA-2018-00015)

Expansive-Phase Formation

In 2008, the DOE issued a report that analyzed the potential for saltstone cracking due to the formation of expansive mineral phases (WSRC-STI-2008-00236). The report concluded that saltstone fracturing by expansive phases is not probable if chemical equilibrium could be assumed. The NRC staff conducted a technical review (ADAMS Accession No. ML093030220) of the DOE report and provided a series of comments, including:

- The DOE report provided a good first step in developing an understanding of the potential for expansive phase formation.

- In addition to the uncertainties addressed by the DOE report (e.g., reaction kinetics, fraction of porosity that must be filled by expansive phases before fracturing can ensue, homogeneous distribution of minerals), the NRC staff determined that there was also uncertainty in the selection of minerals modeled and the expected effects of pozzolanic materials and additives on the dissolution and precipitation reactions.

- Because of those uncertainties associated with a thermodynamic modeling study, the NRC staff determined that comparisons of the modeling calculations to measured data would be useful, especially in constraining the following key uncertainties:

    - mineralogy of the saltstone waste form;
    - fraction of minerals available for reaction; and
    - fractional porosity that must be filled before fracturing occurs

As the NRC staff described in the appendix of the technical review of the Cementitious Barrier Partnership Toolbox (ADAMS Accession No. ML16196A179), there was significant uncertainty associated with the use of fractional porosity to predict cracking due to expansive mineral formation. Additional information was needed to support the use of volume increase based on equilibrium thermodynamic calculations as the basis for modeling damage due to sulfate attack. If support for the basis of volume increase due to calculated mineralogical changes can be developed, then the NRC staff had the following additional concerns related to model parameterization:

- First, the underlying research that was relied upon for the fractional porosity did not include cementitious materials similar to saltstone. Differences between saltstone and the cementitious materials that were studied would be expected to affect the results of those studies.

- Second, the underlying research was based on a very limited set (i.e., four samples) of cementitious materials. Those samples demonstrated that expansion was sensitive to water-to-cement ratios, cement type, and boundary conditions. Fitting a model to that number of variables with four samples results in a poorly constrained model.

- Third, in addition to fitting the model to fractional porosity, the underlying research relied on diffusivity, tensile strength, and initial modulus of elasticity as fitting parameters. It was not clear to the NRC staff that selecting a fractional porosity based on this underlying research in conjunction with other materials and with different material properties was meaningful.

In response to an NRC RAI Comment on the DOE Fiscal Year 2013 SDF SA, the DOE stated that: "[i]nternal sulfate attack was not observed in the saltstone material and is not predicted based on the very high pH of the saltstone pore solution." Although internal sulfate attack due to the formation of expansive phases was not predicted by the DOE, geochemical conditions within saltstone could evolve with subsequent pore volume exchanges and favor the formation of ettringite. Researchers at Pacific Northwest National Laboratory (PNNL) recently observed ettringite in leached cast stone samples; but, not in unleached samples (PNNL-25578). Cast stone and the experimental conditions from PNNL were not identical to saltstone; however, the conditions of the PNNL study were similar to saltstone and demonstrate the potential for expansive phase formation in saltstone. If fracturing due to expansive phase formation was to occur, then its risk significance would depend on the extent and type of fracturing (i.e., several large discrete fractures, small localized fractures, or large-scale network of interconnected small fractures). Large, discrete fractures could largely channel infiltrating water around the saltstone and radionuclide inventory and the increase in diffusive length might be relatively insignificant.

However, if expansive phase formation results in a large-scale network of interconnected small fractures, radionuclide release from the increased diffusive length could outweigh bypass flow.

Based on the lack of support for several key assumptions related to the DOE thermodynamic analysis, the NRC staff determined that sulfate attack cannot currently be excluded as a potential saltstone degradation mechanism. There is significant uncertainty associated with thermodynamic modeling of cementitious materials due to the complexity and number of chemical reactions that occur over different time scales. In addition to those challenges, saltstone is unique and has a short operational history. The recent collection of saltstone core samples from SDS 2A could provide additional insight by comparing the predicted mineralogy based on thermodynamic calculations to the observed mineralogy in actual saltstone samples. Updating the thermodynamic model with the actual mineralogy from field-emplaced saltstone would reduce the uncertainty in the initial saltstone mineralogy predicted by thermodynamic modeling. That information could then be used as an updated starting point for projections of long-term saltstone mineralogy.

Settlement Cracking

In Section 2.6.4.2 of the NRC 2012 SDF TER, the NRC staff described the DOE research that predicted saltstone fracturing in SDS 4 due to static and dynamic settlement at 9.1 m (30 ft) and 15.5 m (50 ft) intervals, respectively. If it can be demonstrated that settlement cracking is likely to result in that type of isolated fractures instead of an interconnected network, then the DOE may be able to demonstrate that any increase in oxidation and subsequent release of redox-sensitive radionuclides due to the gas-phase transport of oxygen along the fracture area would be outweighed by the channeling of water around the grout. During the NRC review of the DOE 2009 SDF PA, the NRC staff were primarily concerned with the release of technetium (Tc) due to oxidation of the slag. However, as discussed in the NRC staff technical review report, Tc Release TRR (ADAMS Accession No. ML18095A122), recent results of dynamic leaching testing at the Savannah River Ecology Laboratory (SREL) and the Center for Nuclear Waste Regulatory Analyses (CNWRA) have reduced the NRC staff concern about the potential sensitivity of Tc release to trace quantities of oxygen. In addition, iodine has shown to be less sorbing than assumed by the DOE in both the FY 2013 and FY 2014 SDF SAs. Accordingly, fracturing could result in a net reduction of risk due to Tc and I-129 due to a fraction of the water flowing around the saltstone grout instead of through the matrix.

Static and dynamic settlement could be risk significant for the closure cap. Depending on the mobility of Tc and I and the timing and extent of degradation of saltstone, the HDPE/Geosynthetic Clay Liner (GCL) composite layer could be a risk significant barrier for infiltrating water. Depending on the extent to which the DOE relies on this composite layer, additional support for the performance of this barrier including performance under reasonably anticipated settlement conditions may be needed.

Steel Corrosion-Induced Cracking

The formation of expansive phases associated with steel corrosion products can result in cracking of surrounding cementitious materials. In the case of saltstone, steel is generally not embedded within the grout with the exception the roof trusses in the upper grout lifts in SDS 4 (SRR-CWDA-2014-00006). There is also steel embedded in the concrete support columns within all of the disposal structures except SDS 1. Degradation of the support columns due to steel corrosion-induced cracking can result in infiltrating water bypassing the saltstone grout. Table 4.2-5 in the FY2014 SA provides the estimated time for complete degradation of the upper saltstone and clean cap lifts in SDS 4 and the columns. The impact of this assumed degradation results in a delay in degradation in the underlying saltstone lifts in SDS 4 by 1,006 years.

The DOE assumed that the degradation of the underlying saltstone lifts in SDS 4 would be delayed by 1,006 years, until the overlying lifts, which contain steel roof trusses, are degraded. The NRC staff determined that there is insufficient support to assume that the degradation of the underlying layers of saltstone will be delayed until the overlying layers are degraded from corrosion-induced cracking. As previously described, the NRC staff is concerned that additional and coupled degradation mechanisms may result in greater-than-assumed saltstone degradation. These degradation mechanisms could be independent from decalcification and degradation of the overlying materials and therefore there would not be a delay in degradation. It also seems more plausible to the NRC staff that the occurrence of steel corrosion induced cracking from embedded steel trusses would be more likely to accelerate, not delay, the timing of the underlying saltstone grout. A more complete saltstone degradation analysis could show that feedback from multiple degradation processes (e.g., steel corrosion-induced cracking and expansive phase formation) tend to accelerate the degradation process.

The cumulative volumetric flow through several saltstone materials[12] in SDS 4, the 46-m (150-ft) and 114-m (375-ft) diameter disposal structures in Figure 16, Figure 17, and Figure 18 shows that the assumed degradation in the FY 2014 SA results in a portion of the infiltrating water bypassing the saltstone grout. Those figures show that bypass flow through the columns in SDS 4 and the 46-m (150-ft) and 114-m (375-ft) diameter disposal structures is limited and does not exceed 25% of the flow through the saltstone grout and is typically less than 10%. The NRC staff acknowledges that there is uncertainty in the degradation analysis; but, the DOE predicts the effect of bypass flow to be limited and the NRC staff agrees with the DOE. Other assumptions (e.g., saltstone degradation, Tc solubility, I sorption) in the FY 2013 and FY 2014 SAs are much more risk significant. However, if the DOE assumes in future analyses that more water will bypass the saltstone grout, then additional support may be necessary.

[12] NRC staff compiled the cumulative volumetric flow from the DOE PORFLOW near-field model results. The bypass flow was calculated as the fraction of water flowing through the degraded columns relative to the flow through the saltstone grout.

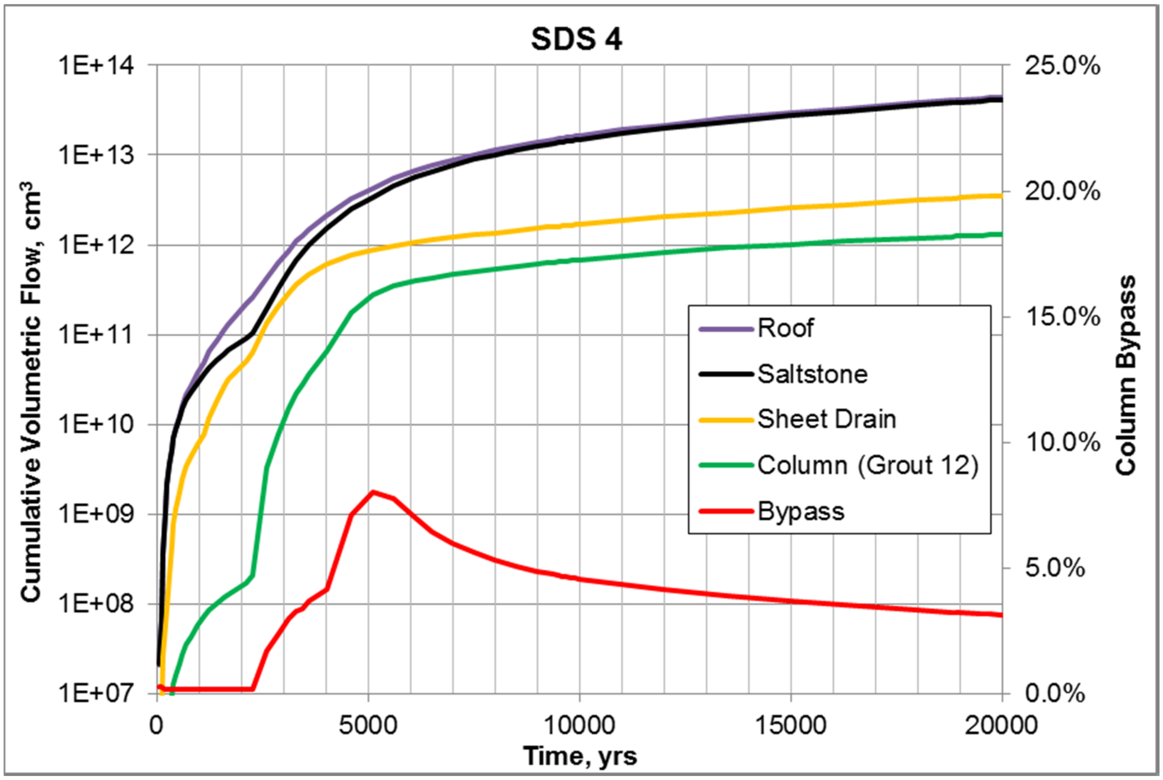

Figure 16: PORFLOW Near-Field Model Cumulative Volumetric Flow through Several Materials in SDS 4 (Prepared from DOE PORFLOW Data)

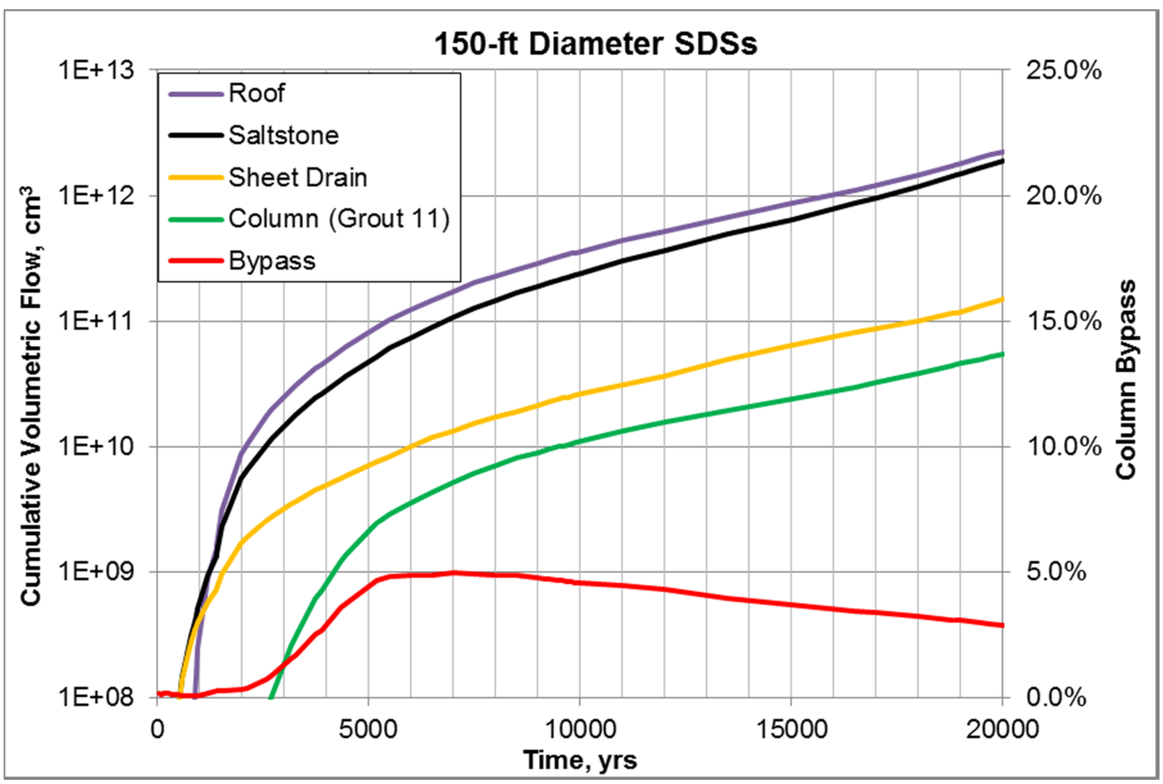

Figure 17: PORFLOW Near-Field Cumulative Volumetric Flow through Several Materials in 46-m (150-ft) Diameter Disposal Structures (Prepared from DOE PORFLOW Data)

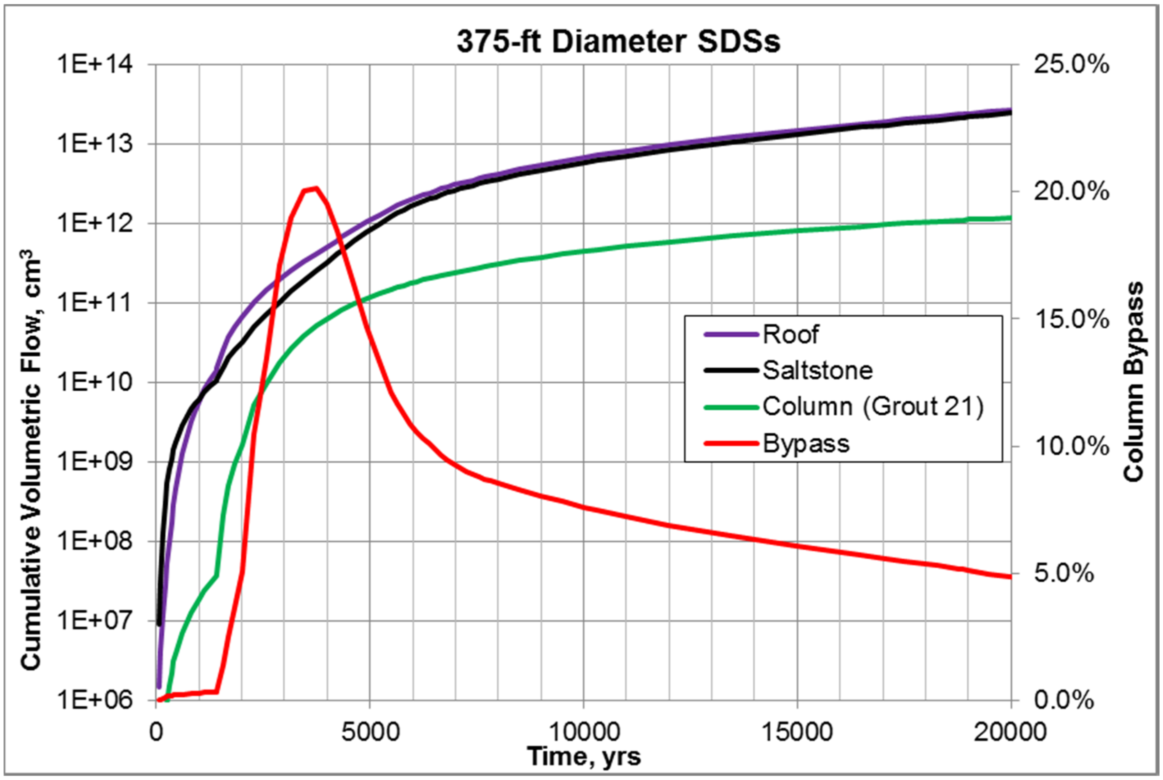

Figure 18: PORFLOW Near-Field Model Cumulative Volumetric Flow through Several Materials in 114-m (375-ft) Diameter Disposal Structures (Prepared from DOE PORFLOW Data)

<u>*Microbial Degradation*</u>

In a 2012 DOE report (SRNL-STI-2012-00435), *Review of Concrete Biodeterioration in Relation to Buried Nuclear Waste,* the authors stated, "[p]revious international efforts related to microbial impacts on concrete structures that house low level radioactive waste showed that microbial activity can play a significant role in the process of concrete degradation and ultimately structural deterioration." For the case of saltstone grout, the authors concluded that, "[t]he results of this review suggest that microbial activity in Saltstone (grouted low level radioactive waste) is unlikely due to very high pH and osmotic pressure." However, a 2016 study (PNNL-25578) on cast stone, which is similar to saltstone, found microbial activity on the surface of cast stone monoliths after leaching in a pore-water solution. A phylogenetic analysis of the samples revealed the presence of several different bacterial phyla. Not surprisingly, several of those bacteria were known to be tolerant of high salt concentrations and alkaline conditions. The authors of the PNNL study concluded that the effects of biological growths on the performance of cast stone was not clear.

Given the observations of microbial growth on cast stone, which also has high pH and high osmotic pressure, the NRC staff determined that it does not appear that microbial degradation can be excluded. Further, as infiltrating water interacts with saltstone, salts will be leached from the grout and it will become more hospitable to additional bacterial strains. In addition, the potential coupling of microbial degradation with other degradation mechanisms should also be considered. Biodegradation of more common cementitious materials might be an appropriate analog for saltstone, because the long-term leaching of saltstone will render it more similar to other cementitious materials.