

DRAFT
UMBRELLA SITE TECHNICAL POSITION
GEOCHEMICAL ISSUES FOR A HIGH
LEVEL RADIOACTIVE WASTE
DISPOSAL FACILITY
IN BASALT

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1.0 BACKGROUND

In the review of an application for Construction Authorization for a HLW geologic repository, the Nuclear Regulatory Commission (NRC) is required to determine whether the site and design meet the Technical Criteria of 10 CFR Part 60 (Subpart E). The NRC staff determination will be based on the answers to, and supporting analyses of, technical questions concerning the performance of the geologic setting and engineered barrier system (groundwater flow, geochemical retardation, waste form and package performance, geologic stability, and facility design). During the process of site characterization, the Department of Energy (DOE) will perform the laboratory and field investigations necessary to develop the information needed to address these basic technical questions.

The investigations needed to characterize a geologic repository are complex and require long lead times. The Nuclear Waste Policy Act of 1982 (NWPA) has established a schedule for site characterization and selection. Specifically, NWPA requires publication of Site Characterization Plans (SCP's) by the DOE at an early stage of the process. Subsequent to receipt of an SCP the NRC must prepare a formal Site Characterization Analysis (SCA) for each site. NRC single-issue technical position papers, documented site reviews, and interagency technical meetings will precede and supplement the SCA's. Because of the complexity and long lead times for site characterization investigations, it is essential that activities be organized to make possible an NRC determination of site acceptability. Proper organization necessitates early identification of technical questions (specific issues) relevant to the specific site. Therefore, this document establishes the NRC position as to the essential technical questions relevant to the geochemistry of a repository in basalt at the Basalt Waste Isolation Project (BWIP). Other Site Technical Positions will address both NRC staff concerns regarding selected specific issues and acceptable technical approaches for addressing those specific issues.

In identifying these essential issues, the staff has used a performance analysis approach. In that approach, three terms, site issues, performance issue and significant conditions and processes, have their special meanings described in the paragraphs below.

A Site Issue is a question about a specific site that must be addressed and resolved to complete the licensing assessment of site suitability and/or design suitability in terms of 10 CFR 60. Site issues are not necessarily controversial questions.

A Performance Issue is a broad question concerning the operation and long-term performance of the various components of the repository system. A set of performance issues are derived directly from the performance objectives in 10 CFR 60.

Significant Conditions and Processes, including potential adverse conditions of 10 CFR 60, are those that must be considered in the assessment of a performance issue and either (1) exist before repository disturbance, (2) could cause future changes, or (3) result from change. They may be natural (e.g., faulting), repository-induced (e.g., thermal buoyancy), and human-induced (e.g., withdrawal of water resources).

In its performance analysis approach, the NRC staff first breaks down the performance objectives of 10 CFR 60 into a set of performance issues corresponding to the individual performance of the various components of the repository system. As developed in NUREG-0960, performance issues for a geologic repository are:

1. How do the design criteria and conceptual design address releases of radioactive materials to unrestricted area within the limits specified in 10 CFR 60?
2. How do the design criteria and conceptual design accommodate the retrievability option?
3. When and how does water contact the backfill?
4. When and how does water contact the waste package?
5. When and how does water contact the waste form?
6. When, how, and at what rate are radionuclides released from the waste form?
7. When, how, and at what rate are radionuclides released from the waste package?
8. When, how, and at what rate are radionuclides released from the backfill?
9. When, how, and at what rate are radionuclides released from the disturbed zone?
10. When, how, and at what rate are radionuclides released from the far field to the accessible environment?
11. What is the pre-waste emplacement groundwater travel time along the fastest path of radionuclide travel from the disturbed zone to the accessible environment?
12. Have the NEPA Environmental/Institutional/Siting requirements for nuclear facilities been met?

The next step in the performance analysis approach is identification of the significant conditions and processes that bear on assessment of each of the performance issues. Judgment is involved in determining which conditions and processes are considered significant. Knowledge gained from the staff's review of various related technical data and documents, site visits, technical meetings and research efforts contributed heavily to the particular selection of significant conditions and processes used in developing of this STP. Questions about the significant conditions and processes as they pertain to site geochemistry constitute the site issues identified in this position.

Because the geochemistry of BWIP site will significantly affect repository performance, information on geochemistry during site characterization will be part

of the total repository system information needs of the NRC staff required to assess the performance elements of 10 CFR 60. Issues identified in the following section delineate information on geochemistry issues on the BWIP site needed by the NRC staff to assess adequately the performance issues. The sequential order in which issues are identified should not be interpreted as the order of relative importance.

Issues STPs have been developed in five technical areas (1.0 GROUNDWATER; 2.0 WASTE FORM/WASTE PACKAGE; 3.0 GEOCHEMISTRY; 4.0 REPOSITORY DESIGN; and 5.0 GEOLOGY). The site issues developed in each STP are designated and numbered in accordance with the above numbering system. The sequential order in which technical areas have been numbered should not be interpreted as the order of relative importance.

2.0 TECHNICAL POSITION

It is the position of the NRC staff that, based on our current level of knowledge of the Basalt Waste Isolation Project (BWIP) investigations, assessments of the Technical Criteria (Subpart E) in 10 CFR Part 60 requires that, at a minimum, the following issues (and associated sub-issues) concerning site geochemistry be addressed.

3.0 GEOCHEMISTRY

3.1 WHAT ARE THE PRESENT GEOCHEMICAL CONDITIONS?

3.1.1 What are the present geochemical conditions of the geologic setting?

3.1.1.1 What are the present temperature and pressure conditions in the host rock and adjacent beds?

3.1.1.2 What is the present mineralogy/petrology/chemistry of primary and secondary minerals of the host rock and adjacent beds?

3.1.1.3 What are the present geochemical conditions of the groundwater in the host rock and adjacent beds?

3.1.2 What is the present mineralogy/petrology/chemistry of the backfill/seals/packing to be emplaced?

3.2 WHAT ARE THE CHANGES IN GEOCHEMICAL CONDITIONS/PROCESSES AS THE RESULT OF WASTE EMPLACEMENT?

3.2.1 What are the changes in the geochemical conditions of the geologic setting?

3.2.1.1 What are the changes in temperature and pressure conditions in the host rock and adjacent beds?

3.2.1.2 What are the changes in the mineralogy/petrology/chemistry of primary and secondary minerals of the host rock and adjacent beds?

- 3.2.1.3 What are the changes in the geochemical conditions in the groundwater?
- 3.2.1.3.1 How does the rock/groundwater ratio in the host rock and adjacent beds change with time?
- 3.2.1.3.2 How does diffusion affect groundwater chemistry?
- 3.2.2 What are the changes in the geochemical conditions of the backfill/seals/packing as a result of waste emplacement?
- 3.2.2.1 What are the changes in temperature and pressure conditions in the backfill/seals/packing?
- 3.2.2.2 What are the changes in the mineralogy/petrology/chemistry of primary and secondary minerals of the backfill/seals/packing as the result of waste emplacement.
- 3.2.2.3 What are the changes in the geochemical conditions of the groundwater moving through backfill/seals/packing as the result of waste emplacement?
- 3.2.2.3.1 How does the rock/groundwater ratio in the backfill/seals/packing change with time?
- 3.3 WHAT ARE THE FUTURE GEOCHEMICAL PROCESSES/CONDITIONS THAT WILL AFFECT RELEASE AND TRANSPORT OF RADIONUCLIDES TO THE ACCESSIBLE ENVIRONMENT?
- 3.3.1 How does solubility/concentration of radionuclides affect radionuclide release and transport?
- 3.3.1.1 How does precipitation/co-precipitation affect radionuclide solubility concentration?
- 3.3.1.2 How does speciation affect radionuclides solubility/concentration?
- 3.3.1.3 How do colloids/particulates/organics affect radionuclide solubility/concentration?
- 3.3.2 How do changes in backfill/seals/packing/disturbed zone/far field mineralogies influence radionuclide migration/retardation?
- 3.3.3 What is the importance of reaction and sorption kinetics on radionuclide release and transport?
- 3.3.4 How do redox conditions of the groundwater, and redox buffering potential of the minerals affect radionuclide speciation/migration/retardation?
- 3.3.5 How does gamma and alpha radiolysis affect radionuclide release and transport?

- 3.3.6 How do colloids/particulates affect radionuclide migration/retardation in the disturbed zone/far field?
- 3.3.7 How do organics/microbes affect radionuclide migration/retardation in the disturbed zone and far field?
- 3.3.8 How does vapor/aerosol transport affect radionuclide migration/retardation?

3.0 DISCUSSION

Issue 3.1 covers the initial geochemical environment of the repository, ie. the geochemical baseline for the repository. Issue 3.2 covers changes to the initial geochemical environment, which will be caused by mining and waste emplacement, and then changed further during heating due to decaying waste. Issue 3.3 deals with waste package/geological environment interactions and the transport of waste radionuclides to the accessible environment. The rationale for each geochemistry issue is described in the subsequent discussion. In the discussion, the broadest issues, those that would appear in the first tier of a hierarchy of issues and sub-issues, are related directly to the performance issues that are listed in the Background Section above. Sub-issues are related by technical argument to the broad issue(s).

3.1 WHAT ARE THE PRESENT GEOCHEMICAL CONDITIONS?

An understanding of the geochemical conditions preceding mining and waste emplacement is necessary in order to evaluate the release of radionuclides from the disturbed zone to the accessible environment. Adverse conditions within the far field are likely to remain unchanged after waste emplacement, whereas favorable prewaste emplacement conditions in the disturbed zone may change to potentially adverse conditions or may become even more favorable. For these reasons, knowledge of the geochemical conditions prior to waste emplacement is necessary to provide the baseline (or framework) for assessments of future repository performance and evaluation of perturbations caused by construction and waste placement. These "baseline" conditions are needed to evaluate Performance Issues 3, 7, 8, 9, and 10.

3.1.1 What are the present geochemical conditions of the geologic setting?

The host rock and adjacent beds are an important component of the geologic HLW isolation system. Knowledge of the mineralogy, petrology, and chemical composition will contribute to the necessary understanding of the future geochemical stability of the host rock, aid in the evaluation of the effects of waste/rock interactions, and provide information for interpreting groundwater chemistry.

3.1.1.1 What are the present temperature and pressure conditions in the host rock and adjacent beds?

Temperature and pressure are important parameters for determining mineral/rock chemical stability, and the geochemical evaluation of groundwater. Therefore, baseline measurements are necessary for determining the magnitude of changes for these parameters under the stressed environment induced by a repository.

3.1.1.2 What is the present mineralogy/petrology/chemistry of primary and secondary minerals of the host rock, and adjacent beds?

The Grande Ronde basalts have a fractured/jointed structure interspersed with vugs, and possess relatively porous flow tops and bottoms. Interbed regions represent a period of inactive magmatic deposition, and weathering. Groundwater movement most likely occurs along flow pathways found in four groups of geologic features: (1) discontinuities within the basalt layers, (2) contacts between layers, (3) fault and fracture zone, and (4) interbed regions between flows.

Openings in the basalt host rock are generally filled with secondary minerals that are derived mostly from the alteration of basalt by circulating ground water. These joints, fractures, and vesicular flow regions are pathways for groundwater carrying radionuclides leached from the waste. The secondary minerals are expected to be a primary sorption medium in the retardation of radionuclides. Thus, information concerning alteration and secondary minerals will aid (1) in interpreting the groundwater chemistry and defining the retardation properties of the host rock prior to waste emplacement, and (2) as a baseline for predicting any alteration that may occur as the result of waste emplacement. Further, information concerning the existing distribution of alteration products may indicate potential release pathways of radionuclides.

3.1.1.3 What are the present geochemical conditions of the groundwater in the host rock and adjacent beds?

Groundwater geochemical conditions, in particular temperature, pressure, pH, redox conditions, ionic strength, and presence of complexing ligands and cations, determine which chemical species of radionuclides are most likely to form and determine what other reactions are likely to occur. Reactions of radionuclides in solution with components of the backfill, the disturbed zone and far field host rock and associated beds, including adsorption and precipitation, will determine the limiting concentrations of soluble species. Also, knowledge of the present groundwater conditions will serve as a baseline for predicting changes in groundwater chemistry resulting from increased temperature and pressure in the disturbed zone.

3.1.2 What is the present mineralogy/petrology/chemistry of the backfill/seals/packing to be emplaced?

"Backfill, seals and packing," as discussed here, refer to materials used to fill drillholes, emplacement holes, shafts, tunnels, and disposal rooms. The large man-made cavities and holes, including fracturing around these cavities or holes, represent broad and potentially short pathways to the accessible environment for the radionuclides released from waste packages. The pathways must be blocked with engineered barriers that provide a means of geochemical retardation of radionuclide migration to the biosphere. The petrology/mineralogy/chemistry of these barriers will influence their ability to affect water ingress, and egress, and radionuclide migration.

3.2 WHAT ARE THE CHANGES IN GEOCHEMICAL CONDITIONS PROCESSES AS THE RESULT OF WASTE EMPLACEMENT?

The geochemical conditions/properties of the host rock surrounding the engineered facility will be affected by construction, and the emplacement of nuclear waste. Construction and increased temperatures in the vicinity of the repository may alter the properties of the basalt/secondary mineralogy to the extent that water is more/less accessible to the waste package and backfill (performance issues 3,4), affecting the release and transport of radionuclides to the accessible environment (performance issues 8,9,10).

3.2.1 What are the changes in the geochemical conditions of the geologic setting?

Rock and mineral stabilities may be affected by repository construction and changes induced by waste emplacement. Many minerals exist in metastable states and the changes in temperature, pressure, and/or degree of saturation may alter the stability of the minerals in a rock. Stability changes will influence the sorptive properties of the host rock, and its ability to prevent water ingress or egress.

3.2.1.1 What are the changes in temperature and pressure conditions in the host rock and adjacent beds?

The magnitude of thermal and pressure gradients over time and space must be determined in order to anticipate mineral/rock stability, and the geochemical evolution of groundwater.

3.2.1.2 What are the changes in the mineralogy/petrology/chemistry of primary and secondary minerals of the host rock, and adjacent beds?

The secondary mineralogy associated with basalt is cited by DOE as a favorable condition for the retardation of radionuclides due to the sorptive capacity of zeolites and clays. Many minerals exist in metastable states and the change of temperature and/or pressure may alter the stability of the minerals in the basalt rock. The alteration products, although often pseudomorphic after the original minerals, generally have different physical/chemical properties which can affect the initial retardation capacity of the rock/minerals. The effects will depend on the amount of water present, and may vary significantly depending on the amount of water present.

3.2.1.3 What are the changes in the geochemical conditions in the groundwater?

Geochemical conditions, in particular temperature, pressure, pH, redox conditions, ionic strength, and the presence of complexing ligands, determine which chemical species of radionuclides are most likely to form and determine what reactions are likely to occur. Reactions of radionuclides in solution with the existing components of the backfill, the disturbed zone and far field host rock, will determine the limiting concentrations of soluble species. Changes in temperature and pressure alter the geochemical conditions of the groundwater which determine the mineral stabilities and may affect radionuclide migration. Also, there is evidence that radiolysis may affect redox conditions, causing generation of hydrogen, oxygen and other species. These altered conditions

will influence waste package and host rock/minerals stability and the ability of released radionuclides to migrate. In addition, repository construction may cause new groundwater pathways to be formed, which would provide fresh rock surfaces for groundwater/rock interaction.

3.2.1.3.1 How does the rock/groundwater ratio in the host rock and adjacent beds change with time?

The mineral stability and induced alteration will depend on the amount of water present in the disturbed zone and far field. Changing rock/water ratios, due to heating and cooling effects, within the disturbed zone may generate mineral zonations that could have a significant effect on the performance of the repository with regard to radionuclide migration.

3.2.1.3.2 How does diffusion affect groundwater chemistry?

The mineralogy associated with the basalt rock, as well as secondary minerals, have been cited by DOE as controlling the redox conditions and pH of the groundwater. However, the host rock is generally armored or coated with secondary minerals. Thus, it has to be demonstrated that both minerals in the host rock and the secondary minerals are in communication with circulating groundwater in order to demonstrate that the mineralogy has the capacity to buffer the redox conditions of the system.

3.2.2 What are the changes in the geochemical conditions of the backfill/seals/packing as a result of waste emplacement?

As the temperature of the disturbed zone and far field host rock, and adjacent beds increases with the time, minerals and ionic solubilities will change in an attempt to re-equilibrate with the new conditions. Minerals may dissolve or precipitate thereby altering the mineral distribution in materials used for backfill/seals/packing. The resultant change will depend on temperature, groundwater conditions and fluid flow regime (i.e., diffusion/convection or fracture flow). Precipitation of minerals may in turn alter the fluid flow path, and ultimately the migration of radionuclides. Also, backfill/seals/packing will be affected by geochemical changes induced by waste emplacement, such as radioanalysis effects, and changes in temperature, pressure, and degree of saturation. Changes in mineral stability may provide pathways for increased groundwater movement, and changes in sorption characteristics affecting repository performance.

3.2.2.1 What are the changes in temperature and pressure condition in the backfill/seals/packing?

The magnitudes of thermal and pressure gradients over time and space must be determined in order to assess the possibility of thermally induced stresses on the backfill. Pathways may develop which could alter groundwater movement through the geologic setting. These parameters also significantly affect mineral alterations and groundwater chemistry, and are needed in order to predict changes in mineralogy and groundwater chemistry in the vicinity of waste packages.

3.2.2.2 What are the changes in the mineralogy/petrology/chemistry of primary and secondary minerals of the backfill/seals/packing as the result of waste emplacement?

Mineral assemblages may be affected by changes induced by waste emplacement. Many minerals exist in metastable states and the changes in temperature, pressure, water chemistry, and/or degree of saturation may affect the stability of the minerals in the backfill packing, and seals. These changes can influence the sorptive behavior of the backfill, and the groundwater flow within these materials.

3.2.2.3 What are the changes in the geochemical conditions of the groundwater moving through backfill/seals/packing as the result of waste emplacement?

Geochemical conditions, in particular temperature, pressure, pH, redox conditions, ionic strength, and the presence of complexing ligands, determine which chemical species of radionuclides are most likely to form and determine what reactions are likely to occur. Reactions of radionuclides in solution with the existing components of the backfill/seals/packing including adsorption and precipitation, will determine the limiting concentrations of soluble species. Changes in temperature and pressure alter the geochemical conditions of the groundwater which determine the mineral stabilities and may affect radionuclide migration. An assessment of the geochemical conditions of the groundwater following waste emplacement must be made in order to predict radionuclide release and migration.

3.2.2.3.1 How does the rock/groundwater ratio in the backfill/seals/packing change with time?

The mineral stability of backfill/seals/packing will depend on the amount of water present. The degree of induced mineral alteration may vary significantly, depending on how much water is present. Zones of varying saturation, due to heating and cooling effects, in the vicinity of waste packages may generate mineral zonations that could have a significant effect on the performance of the repository with regard to radionuclide release and migration.

3.3 WHAT ARE THE FUTURE GEOCHEMICAL PROCESSES/CONDITIONS THAT WILL AFFECT RELEASE AND TRANSPORT OF RADIONUCLIDES TO THE ACCESSIBLE ENVIRONMENT?

Geochemical reactions, processes, and conditions at the waste package surface, in the backfill, the disturbed zone, and the far field will affect the release and transport of radionuclides from the repository and the accessible environment; and thus play an important role in assessing performance issues 7, 8, 9, and 10. Release involves waste package degradation and solubilization of the radionuclides in the waste form. Transport involves any mechanical or chemical process which promotes or inhibits radionuclide migration from the repository to the accessible environment. During release and transport, radionuclides will react with the groundwater, the waste container, backfill, the host rock adjacent beds and secondary minerals, and the nature of these reactions will determine the extent of the migration of each radionuclide from the waste form.

3.3.1 How does solubility/concentration of radionuclides affect radionuclide release and transport?

The rate at which radionuclides are transported to the accessible environment is a function of solubility, the rate and path of groundwater movement, and the reactions of radionuclides with minerals in the backfill, in fractures in the host rock and adjacent beds, and in the matrix of the rock itself. Dissolution of radionuclides from the waste form into solution is controlled by the physical characteristics of the waste (e.g., structure and surface area), chemical and radiolytic properties of the waste, composition, redox reactions and the pH of circulating waters, temperature, and pressure. Under slow flow or no flow conditions a conservative estimate of concentrations of radionuclides species released into solution is that they are solubility limited. Therefore, in order to determine the concentrations of radionuclides in the disturbed zone and the far field (under different geochemical conditions) through time, their solubilities need to be assessed.

3.3.1.1 How does precipitation/co-precipitation of radionuclides affect radionuclide release and transport?

Under varying geochemical conditions, radionuclides in solution may precipitate in the presence of certain inorganic ligands (e.g., carbonate, hydroxyl, sulfide). Parameters controlling precipitation include groundwater composition, rock composition, redox reactions, and pH. Certain radionuclides may co-precipitate by substitution with non-radioactive species such as iron.

3.3.1.2 How does speciation affect radionuclide solubility/concentration?

Information concerning the identities and solubilities of the solid phases, and information concerning identities of the solution species likely to form under anticipated geologic conditions at BWIP are needed in order to determine solution concentrations of radionuclides in a repository groundwater system. Different species of the same element will remain in solution in different concentrations and migrate at different rates.

3.3.1.3 How do colloids/particulates/organics affect radionuclide solubility/concentration?

Some radionuclides, especially hydrolyzable ones, may readily form colloids or pseudocolloids under certain geochemical conditions. These colloids may result from interactions with the waste package. The formation of colloidal species may affect the concentrations and thus the migration of radionuclides in solution. Also, the sorptive capacity of suspended particulated could provide a transport mechanism for radionuclides otherwise considered immobilized due to sorption onto surrounding fixed materials. Finally, the presence of organic ligands can allow some radionuclides to form complexes and remain in solution at concentrations different than uncomplexed species.

3.3.2 How do changes in backfill/seals/packing/disturbed zone/far field mineralogies influence radionuclide migration/retardation?

Chemical changes in the packing material due to temperature, pressure, and the degree of saturation will affect its ability to retard mobile radionuclide

species. Highly sorptive minerals in the backfill, near-field, and far field host rock may cause significant retardation of radionuclides. A good estimate of the location, volume, distribution and accessibility of minerals along the likely flow paths is necessary to assess the effects of mineralogy on radionuclide migration and retardation.

3.3.3 What is the importance of reaction and sorption kinetics on radionuclide release and transport?

The occurrence of reactions is predicted by chemical equilibrium. However, reaction rates are generally not instantaneous as predicted by equilibrium, but kinetically controlled (time dependent). Thus, rate information is necessary in order to predict reaction rates and the steady state conditions expected in the repository system.

3.3.4 How do redox conditions of the groundwater and redox buffering potential of the minerals affect radionuclide speciation/migration retardation?

Reducing redox conditions may be a significant determinant of radionuclide speciation, solubility and migration in a basalt environment. Construction of a repository will allow atmospheric oxygen to enter into the repository horizon and cause oxidizing conditions. After closure, the atmospheric oxygen may be consumed and redox conditions could become reducing (neglecting radiolytic effects). However, there are a number of factors that make predictions based on redox conditions uncertain: (1) the effects of gamma radiation and, after breach of containment, alpha radiation, may result in the continuous generations of oxygen from the radiolysis of water (2) the oxygen buffering capacity of the basalt may be limited; and (3) the reducing capacity of the water may not produce the anticipated radionuclide solubility retardation characteristic desired.

3.3.5 How does gamma and alpha radiolysis affect radionuclide release and transport?

There is evidence that radiolysis may affect redox conditions, through the generation of hydrogen, oxygen, and other species, and thus affect anticipated reactions. Further, radiolysis reactions may generate stable organics. These conditions may influence radionuclide speciation and transport.

3.3.6 How do colloids/particulates affect radionuclide migration/retardation in the disturbed zone/far field?

Under certain geochemical conditions, radionuclides may form colloids, pseudo-colloids, or become attached to particulates. Colloids and particulates are potentially more mobile than aqueous species formed under the same conditions. The stability and mobility of colloids and particulates under changing geochemical conditions need to be addressed in evaluating radionuclide retardation.

3.3.7 How do organics/microbes affect radionuclide migration/retardation in the disturbed zone and far field?

Organics/microbes may be introduced into a repository during construction by contamination from the surface, or could be present in the host rock or

associated beds. Radionuclide organic complexes have different migration behavior than inorganic complexes. The likelihood of significant amounts of organics or microbes being present for complexing or combining with radionuclides, along with their effects on radionuclide migration behavior, should be addressed.

3.3.8 How does vapor/aerosol transport affect radionuclide migration/retardation?

Spent fuel may contain radionuclides in gaseous phases that could be transported to the accessible environment via fractures. Aerosols may be formed due to the drying effects in the disturbed zone, caused by the heat generated by the emplaced waste, and may promote transport of soluble radionuclides in a vapor phase. These processes should be considered and the role of vapor/aerosol transport in radionuclide migration assessed.