

## **Reference Cases for Use in the Cementitious Barriers Partnership Project - 9446**

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### **ABSTRACT**

The Cementitious Barriers Partnership Project (CBP) is a multi-disciplinary, multi-institution cross cutting collaborative effort supported by the US Department of Energy (DOE) to develop a reasonable and credible set of tools to improve understanding and prediction of the structural, hydraulic and chemical performance of cementitious barriers used in nuclear applications. The period of performance is >100 years for operating facilities and > 1000 years for waste management. The CBP has defined a set of reference cases to provide the following functions: (i) a common set of system configurations to illustrate the methods and tools developed by the CBP, (ii) a common basis for evaluating methodology for uncertainty characterization, (iii) a common set of cases to develop a complete set of parameter and changes in parameters as a function of time and changing conditions, and (iv) a basis for experiments and model validation, and (v) a basis for improving conceptual models and reducing model uncertainties. These reference cases include the following two reference disposal units and a reference storage unit: (i) a cementitious low activity waste form in a reinforced concrete disposal vault, (ii) a concrete vault containing a steel high-level waste tank filled with grout (closed high-level waste tank), and (iii) a spent nuclear fuel basin during operation. Each case provides a different set of desired performance characteristics and interfaces between materials and with the environment. Examples of concretes, grout fills and a cementitious waste form are identified for the relevant reference case configurations.

### **INTRODUCTION**

The Cementitious Barriers Partnership (CBP) Project is a multidisciplinary effort supported by the US DOE to develop a set of tools to improve prediction of the structural, hydraulic and chemical performance of cementitious barriers used in nuclear applications over extended time frames (e.g., >100 years for operating facilities and > 1000 years for waste management) [1]. The CPB partners, in addition to the US DOE, are the U.S. Nuclear regulatory Agency (NRC), the National Institute of Standards and Technology (NIST), the Savannah River National Laboratory (SRNL), Vanderbilt University (VU) / Consortium for Risk Evaluation with Stakeholder Participation (CRESP), Energy Research Center of the Netherlands (ECN), and SIMCO, Technologies, Inc.

The project is focused on reducing uncertainties associated with current methodologies for assessing cementitious barrier performance and increasing the consistency and transparency of the assessment process. The results of this project will support long-term performance predictions and performance-based decision making and are applicable to several of the strategic initiatives in the U. S. Department of Energy (DOE) Environmental Management Engineering & Technology Roadmap [2].

Performance assessments (PAs) for low-level waste facilities consist of 1) ground water flow and contaminant transport models, 2) air and radon transport pathway models, 3) inadvertent intruder analyses, and 4) all path ways human health risk analyses. The CBP project is focused on understanding and predicting the physical (hydraulic), chemical (contaminant retention and matrix evolution) and mechanical (structural) performance of cementitious barriers including waste zones for the subsurface flow and contaminant transport modeling. The set of simulation tools and data developed by this project will be applicable to near surface engineered waste disposal systems, e.g., waste forms, containment structures, entombments and environmental remediation, including decontamination and decommissioning (D&D) activities. The simulation tools will also support analysis of chemical degradation of concrete used in nuclear facilities containment structures (spent fuel pools, dry spent fuel storage units, and recycling facilities, e.g., fuel fabrication, separations processes).

Three prototype reference systems / configurations described in this paper were defined to capture the essential features of the various types of engineered cementitious barriers. The reference cases are intended to provide:

- Full descriptions of the engineered structures that are sufficient to support Performance Assessment (PA) modeling;
- Simplified descriptions for 1- and 2-D analyses with representative materials and interfaces that will be used to evaluate time and spatially dependent evolution of performance in response to dynamic boundary conditions;
- Material descriptions and boundary conditions for experimental programs designed to support property-based chemical and physical constitutive models (non spatially dependent);
- Focused experimental programs that will be designed to reduce uncertainties associated with assumptions about material performance in interfacial regions between the waste, engineered materials, and environmental media.

## REFERENCE CASES

Key information required as inputs for defining systems and scenarios for PA modeling includes:

- Geometry
- Initial conditions
- Boundary conditions (e.g., fluxes, concentrations, etc.)
- Material properties that control matrix durability and contaminant leaching including:
  - a. Physical
  - b. Hydraulic
  - c. Structural
  - d. Chemical
  - e. Mineralogical

In addition, meaningful temporal and spatial scales must be selected to best address the modeling needs.

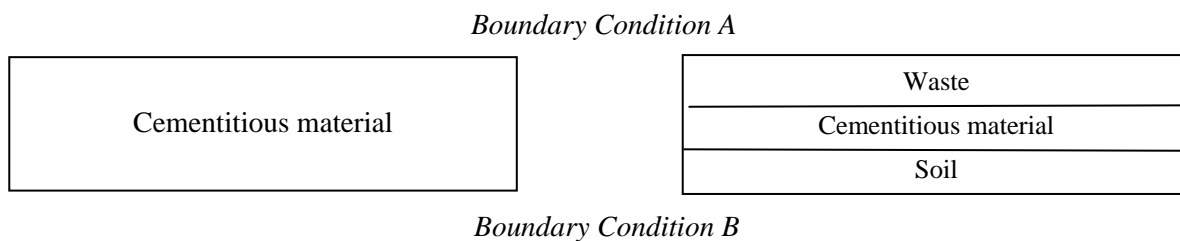
Key outputs required for cementitious barrier performance modeling include:

- Moisture and gas flow and constituent (contaminant) transport (leaching) function of time and spatial relationships,
- Changes in the physical / hydraulic properties of the barrier and waste as a function of time and spatial relationships.

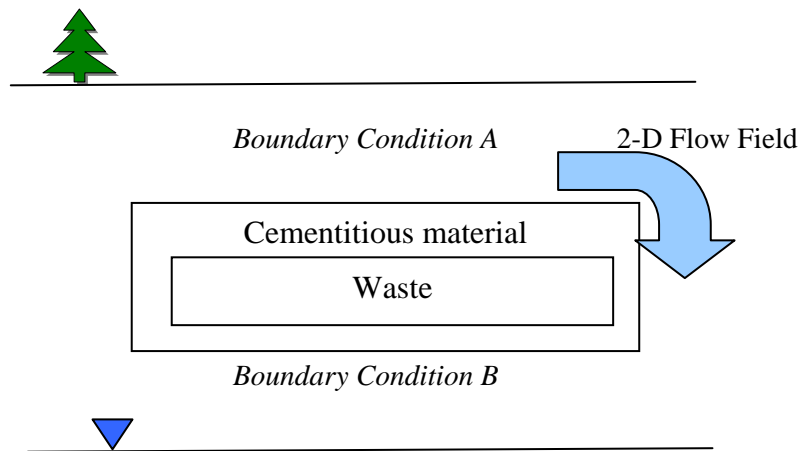
## Reference Case Geometry

Actual structures, engineered barriers, process equipment, and waste packages, etc. are three dimensional (3-D) and typically geometrically complex. For computational convenience, most low-level waste PAs reduce the 3-D complexities to 2-D cross sections that are considered to be reasonable approximations sufficient for addressing the geometrical issues. When cementitious barriers are present, 1-D approximations must be applied with caution and are rarely adequate due to the contrast in the hydraulic conductivities between the barrier and environmental media and / or waste zone.

The proposed CBP progression for the reference cases is illustrated in Figures 1 and 2. Mechanistic understanding will be obtained initially from 1-D phenomenological modeling and supporting experiments as shown in Figure 1. This information will be used as input to multi-dimensional PA flow and transport models, which are schematically illustrated in Figure 2, or in 1-D relative uncertainty analyses, such as those obtained with the Goldsim environmental transport modeling. The process for incorporating the phenomenological information into the multi-dimensional PA codes will be via algorithms developed from the 1-D experimental and associated modeling effort.



**Figure 1. Examples of a One-Dimensional Reference Case Configuration for Evaluating Chemical and Physical Phenomena and Mechanisms.**



**Figure 2. Example of a Two-Dimensional Reference Case Configuration that Incorporates a Flow Field for PA Calculations.**

**Reference Case Initial and Boundary Conditions**

Initial conditions typically defined in PAs that are important to the performance of disposal units, including engineered cementitious barriers are:

- Waste inventory and characteristics
  - Radioactive and chemical species of interest
- Influxes of water, gas, and chemical species at the system boundaries (fluxes across material interfaces)
  - Water (% saturation, pH, Eh, dissolved O<sub>2</sub> and CO<sub>2</sub>)
  - Air (% O<sub>2</sub>, CO<sub>2</sub>, H<sub>2</sub>O – relative humidity)
  - Corroderent chemicals such as Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, alkalis, organic and inorganic acids
- Infiltration rates and flow along material interfaces (flow fields)
- Temperature and temperature cycling
- Structural condition
  - Initial cracks from thermal stresses and drying shrinkage stresses
  - Structure penetrations, construction joints, and other construction details
  - Steel reinforcements (rebar and other)
- Episodic events such as seismic events and structural settlement or failure.

The CBP effort will focus primarily on the consequences of the influxes and fluxes across material interfaces and through materials of moisture, gas, and chemical species on the cementitious barrier materials as functions of long-term exposure. Temperature and temperature cycling will also be considered. The radioactive species of interest for the CBP reference cases are primarily Cs<sup>+</sup>, Sr<sup>2+</sup>, and the long lived mobile isotopes, Tc-99, I-129, C-14 and selected actinides, such as U and Pu or suitable surrogates. Degradation of structural penetrations, e.g., construction joints and other construction details will not be included in the mechanistic or phenomenological investigations except for the potential to provide fast pathways.

### **Reference Case Time Periods**

Time periods over which performance predictions are required are 100 years for storage structures and 1000 to 10,000 years for disposal units. Consequently, the phenomenological models will be run to estimate corresponding time periods. Laboratory experiments for mechanistic or validation studies are not expected to exceed a 5-year time period. If specific data are required for older (aged) materials (5-50+ years) cores from existing structures will be collected and analyzed.

### **Reference Case Outputs**

The parameters required for PA modeling are the reference case outputs for the CBP experimental and phenomenological modeling efforts. These parameters are typically chemical, hydraulic, and physical properties of the engineered barrier materials and of the barriers themselves and evolution of the properties as a function of time, influx of chemicals and physical conditions that modify the properties. Examples of important properties for cementitious barriers include: bulk composition, mineralogy, hydraulic conductivity, solubilities and diffusivities of the matrix phases and contaminant species (leaching properties), porosity and pore size distribution, moisture retention curves (function of pore size distribution and pore structure), bulk density and particle density.

### **Reference Case Cementitious Materials**

Three types of cementitious materials were selected as reference cases. Each of the reference case materials have been used as barriers in actual waste disposal units. These materials are listed below:

- Reinforced Concrete (carbon steel rebar with three inch cover)
  - Type I/II Binary Blend (portland cement + blast furnace slag binder)
  - Type I/II Ternary Blend (portland cement + blast furnace slag + Class F fly ash binder)
  - Type V Sulfate Resistant Quaternary Blend (portland cement + slag + Class F fly ash + silica fume binder)
- Flowable, Stable (zero-bleed) Infill / Back Fill Grout
  - Three chemically reducing ternary blends
    - High water to cementitious material ratio
    - Medium water to cementitious material ratio
    - Low water to cementitious material ratio with 3/8 inch stone and sand
  - Non reducing binary blend
    - Low water to cementitious material ratio with 3/8 inch stone and sand
- Salt waste form.

These materials are described in more detail in Tables 1 to 3, respectively.

Table 1. Reference Case Binary, Ternary, and Quaternary Concrete Formulations.

<b>Ingredient</b>	<b>Type I/II Binary Blend [3]</b> <b>(kg/m<sup>3</sup>)</b> <b>(lbs/yd<sup>3</sup>)</b>	<b>Type I/II Ternary Blend [4]</b> <b>(kg/m<sup>3</sup>)</b> <b>(lbs/yd<sup>3</sup>)</b>	<b>Type V Quaternary Blend [3]</b> <b>(kg/m<sup>3</sup>)</b> <b>(lbs/yd<sup>3</sup>)</b>
Type I/II Cement (ASTM C 150)	239 (419)	71.3 (120)	0
Type V Cement (ASTM C 150)	0	0	133.5 (225)
Blast Furnace Slag (ASTM C 989)	158 (278)	163 (275)	178 (300)
Type F Fly Ash (ASTM C 618)	0	80.1 (135)	103.8 (175)
Silica Fume (ASTM C 1240)	0	0	29.7 (50)
Quartz Sand (ASTM C 33)	646 (1133)	756.7 (1270)	540.7 (911)
No. 67 Granite Aggregate (maximum ¾ in) (ASTM C 33)	1025 (1798)	1038.6 (1750)	1098 (1850)
Water (maximum)	152 268 (32.1 gallons)	142.4 240 (28.8 gallons)	168.6 284 (34 gallons)
Water to cementitious material ratio	0.385	0.38	0.38
Grace WRDA 35 (ml /100 kg cement + pozzolan) (oz/cwt cement + pozzolans)	32.6 (5.0)	32.6 (5.0)	32.6 (5.0)
Grace Darex II (ml /100 kg cement + pozzolan) (oz/cwt cement + pozzolans)	2.6-3.3 (0.4-0.5)	2.6-3.3 (0.4-0.5)	2.6-3.3 (0.4-0.5)

Grace Adva 380 (ml /100 kg cement + pozzolan) (oz/cwt cement + pozzolan)	19.6 - 26.1 (3 - 4)	19.6 - 26.1 (3 - 4)	19.6 - 26.1 (3 - 4)
Unit Weight (kg/m <sup>3</sup> ) (lbs/yd <sup>3</sup> )	2220 (3896)	2156 (3790)	2162 (3795)
Compressive strength at 28 days (MPa) (psi)	27.6 (4000)	27.6 (4000)	34.5 (5000)

Table 2. Reference Case Flowable, Stable Infill / Backfill Grout Formulations.

<b>Ingredient</b>	<b>Type I/II Ternary Blend [5] (kg/m<sup>3</sup>) (lbs/yd<sup>3</sup>)</b>	<b>Type I/II Ternary Blend 2 [5] (kg/m<sup>3</sup>) (lb/yd<sup>3</sup>)</b>	<b>Type I/II Ternary Blend 3 [5] (kg/m<sup>3</sup>) (lbs/yd<sup>3</sup>)</b>	<b>Type I/II Binary Blend [6] (kg/m<sup>3</sup>) (lbs/yd<sup>3</sup>)</b>
Type I/II Cement (ASTM C 150)	44.5 (75)	109.8 (185)	109.8 (185)	267 (450)
Grade 100 Blast Furnace Slag (ASTM C 989)	124.6 (210)	163.2 (275)	154.3 (260)	0
Type F Fly Ash (ASTM C 618)	222.6 (375)	344.2 (580)	504.5 (850)	267 (450)
Quartz Sand (ASTM C 33)	1365 (2300)	1118.7 (1885)	559.1 (942)	746.6 (1258)
No. 8 Granite Aggregate (maximum 3/8 in) (ASTM C 33)	0	0	561.5 (946)	741.9 (1250)
Water (maximum) (kg/m <sup>3</sup> ) (lbs/yd <sup>3</sup> )	297 501 (60 gallons)	297 501 (60 gallons)	302 509 (61 gallons)	207.7 350 (42 gallons)
Water to cementitious material ratio	0.76	0.49	0.39	0.39
Viscosity Modifier (Welan Gum) Kelco-Crete (grams/m <sup>3</sup> ) (grams/yd <sup>3</sup> )	360 (275)	283 (216)	283 (216)	0
High Range Water Reducer (HRWR) (L/m <sup>3</sup> ) (fl oz/yd <sup>3</sup> )	3.48 90*	2.88 54**	2.88 54***	2.88 – 2.707 54-70
Sodium Thiosulfate (optional)	1.25 (2.1)	1.25 (2.1)	1.25 (2.1)	0
Set Regulator (W. R. Grace Recover (fl oz/yd <sup>3</sup> )	as needed	as needed	as needed	as needed
Unit Weight (kg/m <sup>3</sup> ) (lbs/yd <sup>3</sup> )	1972 (3461)	1952 (3426)	2104 (3692)	2141 (3758)
Compressive strength at 28 days MPa (psi)	27.6 (4000)	27.6 (4000)	34.5 (5000)	27.6 (4000)

\* W. R. Grace Adva flow \*\* Sika ViscoCrete 2100 \*\*\* W. R. Grace Advaflex

The reference case salt waste form is prepared from a premix of cementitious reagents and a low-level radioactive solution containing dissolved sodium salts. The formulation for the premix is provided in Table 3. The formulation for a typical DOE salt waste solution stabilized with the reference premix is provided in Table 4.

Table 3. Reference Case Blended Premix Reagents for DOE Salt Waste Forms [3].

<b>Ingredient</b>	<b>Wt. %</b>
Type I/II Cement (ASTM C 150)	10
Grade 100 Blast Furnace Slag (ASTM C 989)	45

Type F Fly Ash (ASTM C 618)	45
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Table 4. Reference Case Non Radioactive Salt Waste Solution [3].

Ingredient	Molarity (Moles/Liter)	Mass (g/Liter H <sub>2</sub> O)
Sodium Hydroxide, NaOH (50% by weight solution)	2.866	229.28
Sodium Nitrate, NaNO <sub>3</sub>	1.973	167.66
Sodium Nitrite, NaNO <sub>2</sub>	0.485	33.43
Sodium Carbonate, Na <sub>2</sub> CO <sub>3</sub>	0.118	12.46
Aluminum Nitrate Nona-hydrate, Al(NO <sub>3</sub> ) <sub>3</sub> ·9H <sub>2</sub> O	0.114	42.90
Sodium Sulfate, Na <sub>2</sub> SO <sub>4</sub>	0.055	7.84
Sodium Phosphate Na <sub>3</sub> PO <sub>4</sub> ·12H <sub>2</sub> O	0.007	2.76
Density (g/ml)	1.248	
Dynamic Viscosity (cP)	2.78	
Wt.% Water	71.12	
Wt. % Solids	28.88	
Wt. % Salt in Wet Waste Form with a Water to Premix Ratio of 0.60	13.0	

### Reference Disposal and Storage Units

The CBP reference case materials have been used in actual low-level waste (LLW) disposal units in the DOE complex or in commercial nuclear industry process / storage units. Three reference case configurations are listed below:

- a) Cementitious low-level salt waste form in a reinforced concrete disposal vault.
- b) Reinforced concrete vault containing a carbon steel high-level waste tank filled with a chemically and structurally stabilizing cementitious grout and low-level waste residuals.
- c) Reinforced concrete spent nuclear fuel basin with a stainless steel liner.

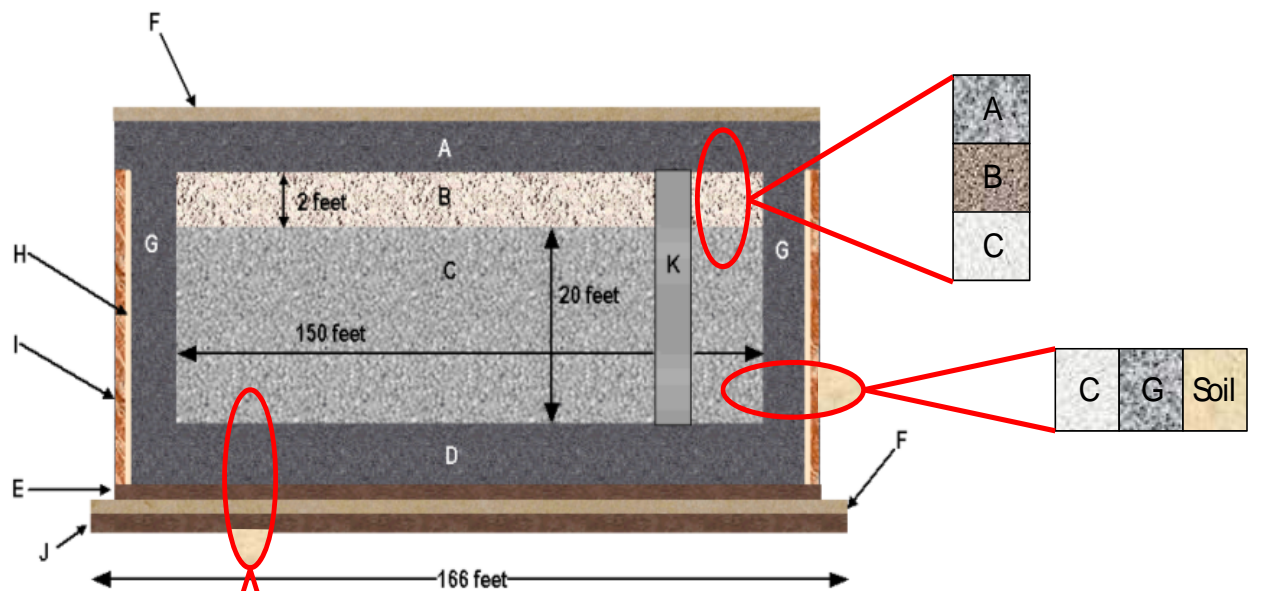
Schematic illustrations of the two disposal units and of the spent fuel basin are provided in Figures 3-5.

***Cementitious Waste Form in Reinforced Concrete Vault:*** A brief description of the reference cementitious waste form disposed of in a concrete vault is provided below:

- Reinforced concrete vault filled with a monolithic cementitious low-level radioactive salt waste form. The concrete vault also contains carbon steel columns and trusses to support the roof. The vault is filled in layers typically 15 to 30 cm thick.
- A clean grout cap is placed between the final waste form layer and the top of the vault.
- Upon closure of the disposal facility, which will contain multiple vaults, soil backfill will be placed around vaults constructed on grade and a multi-layer cap will be constructed to limit infiltration. (New vault designs call for the vaults to be constructed below grade.)
- External boundary conditions for the at grade vaults prior to closure include: exposure of the concrete walls and roof to ambient air conditions, for example, free exchange of moisture and air with atmosphere, unsaturated concrete with intermittent wetting, and precipitation diverted away from waste form. The base slab will be exposed to unsaturated soil.

- Internal boundary conditions for the vault walls and base slab are a function of exposure to the salt waste form. The waste form is a highly alkaline material with a very high sulfate content and is therefore a potential source of chemicals that are known to degrade concrete.
- External boundary conditions for the concrete vault after closure include: contact with native soil (sand and clay) with very low, intermittent infiltration and unsaturated moisture content controlled by balance of capillary pressures and pore water-vapor equilibrium that is a function of pore space relative humidity.
- Internal boundary conditions for the vault walls and base slab are a function of exposure to the salt waste form (same as above).

For this system, the CBP will conduct research to improve the understanding of degradation mechanisms and material evolution as a function of long times and develop algorithms that link degradation to changes in hydraulic properties of the cementitious barriers which can be used in the PA models.



Key	Material	Thickness (cm)
A	Reinforced Concrete for Resistance to Moderate Sulfate Exposure	20.5
B	Clean Fill Grout	61
C	Cementitious Salt Waste Form	610
D	Reinforced Concrete for Resistance to High Sulfate Exposure	20.5
E	Upper Mud Mat Concrete for Resistance to High Sulfate Exposure	10
F	Geotextile Material (HDPE / GCL)	0.76
G	Reinforced Concrete for Resistance to High Sulfate Exposure	20.5
H	Carbon Steel Wall Liner	0.047
I	Geotextile Material (HDPE)	0.254
J	Lower Mud Mat Concrete	10
K	Reinforced Concrete Column with Resistance to High	35.6

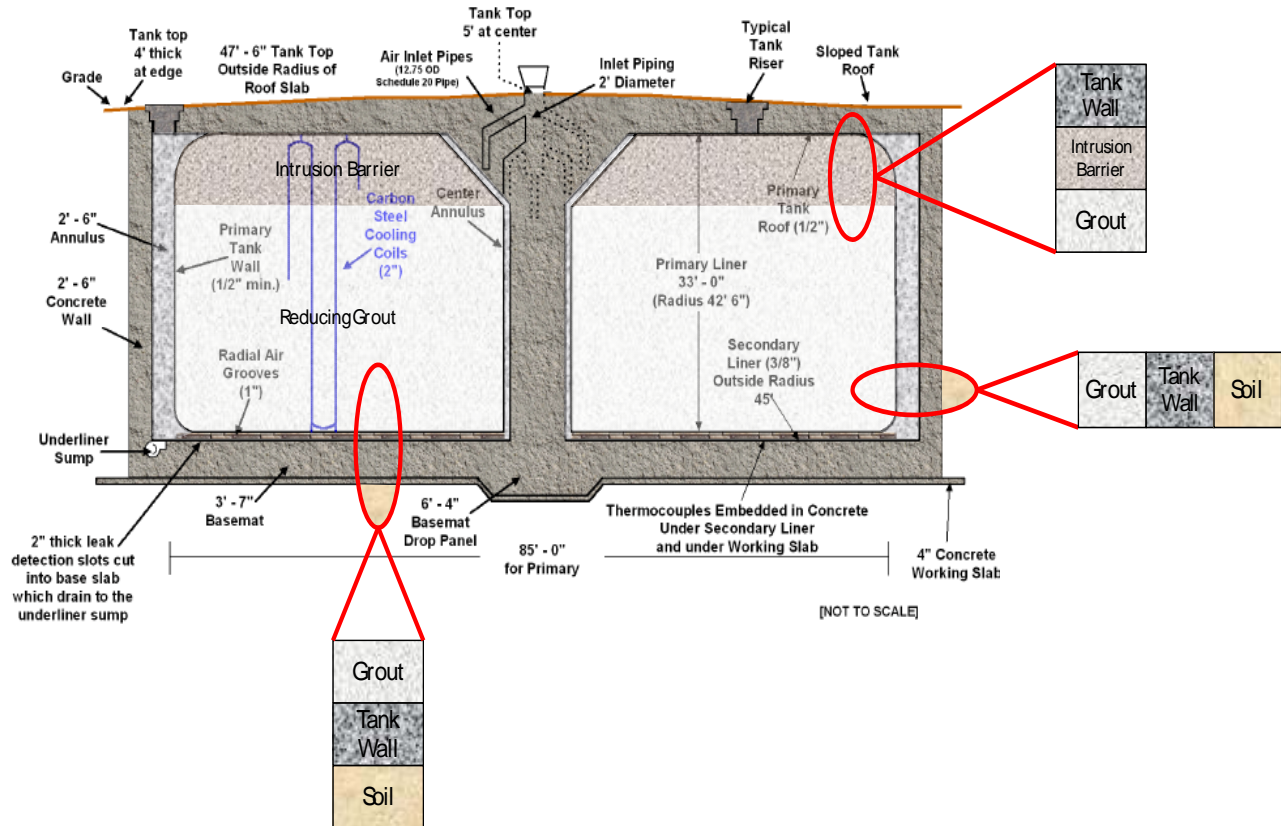
	<i>Sulfate Exposure</i>	
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**Figure 3. Schematic illustration of a reinforced concrete vault containing a cementitious low activity waste form. Examples of multi-layer material and interfaces relevant to 1-D mechanistic studies are illustrated.**

***Closed High-Level Waste Tank:*** A brief description of the reference closed carbon steel high-level waste tank surrounded by a concrete vault and filled with a cementitious grout is provided below:

- Carbon steel liner (HLW tank) in a reinforced concrete vault will be filled with a cementitious grout to physically stabilize the structure and prevent collapse and to also chemically stabilize residual waste and contaminants. The annulus space between tank and concrete vault will also be filled with cementitious grout.
- One or more grout formulations will be used to fill the tank. A chemically reducing formulation (containing blast furnace slag) will be used for grout in contact with waste residuals.
- Tanks typically contain metal piping (e.g., cooling coils) and process equipment (e.g. pumps) which will also be filled with grout where practical.
- Closure includes backfill in some cases and coverage with multi-layer cap to limit infiltration.
- Each engineered barrier has a unique set of boundary conditions. For example, the external boundary conditions for the concrete vault are determined by the surrounding soil with a low, intermittent infiltration and unsaturated moisture content controlled by the balance of capillary pressures and pore water-vapor equilibrium and atmospheric exchange by gas diffusion. For the purposes of estimating the consequences over long performance times, the interfaces between the annulus grout and steel tank and the fill grout and the steel piping in the tank will be assumed to be similar to the interfaces between the reinforcing steel in the vault concrete and the concrete itself.

For this system, the CBP will conduct research to improve the understanding of degradation mechanisms and material evolution as a function of soil saturation, episodic events that may create fast pathways, i.e., cracking and its effect on hydraulic and leaching performance.

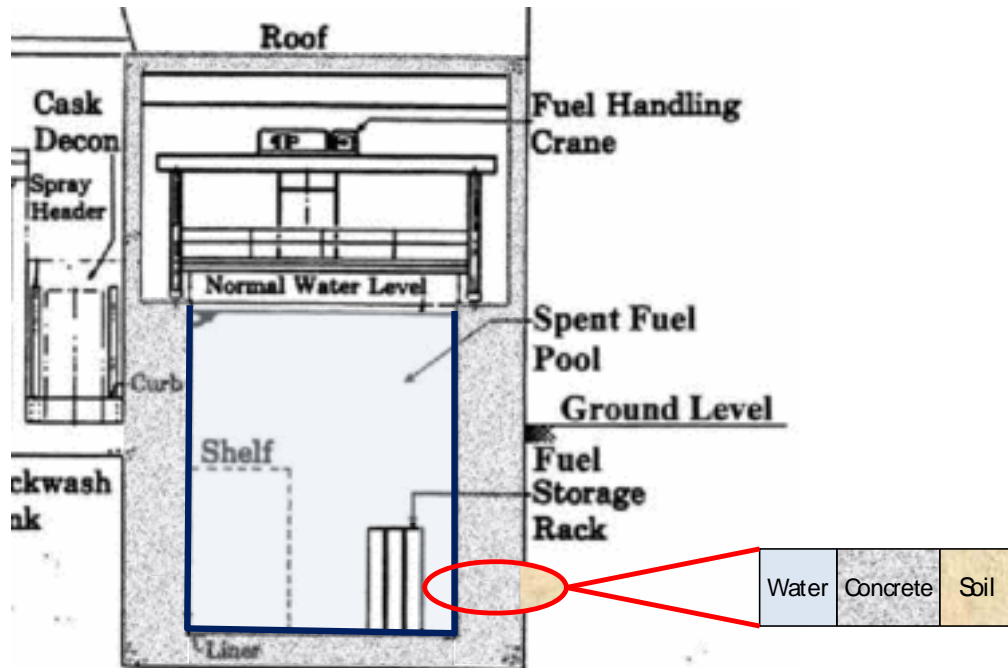


**Figure 4. Schematic illustration of a closed high-level waste tank (carbon steel tank in a reinforced concrete vault) containing a cementitious grout fill. Examples of multi-layer material and interfaces relevant to 1-D mechanistic studies are illustrated.**

*Spent Fuel Basin:* A brief description of the reference case for a stainless steel-lined spent fuel basin is provided below:

- Below grade stainless steel-lined, reinforced concrete basin filled with borated water that results in approximately 6 m (20 ft) of hydraulic head on the basin.
- Internal boundary conditions for the reinforced concrete include complete saturation (water) of concrete pores with water containing borate.
- External boundary conditions include contact with saturated soil.

For this system, the development of through wall cracks due to initial conditions, construction joint failure, or post construction settlement and the resulting impact on flow and transport are of primary interest.



**Figure 5. Schematic diagram of spent fuel pool during operations. Examples of multi-layer material and interfaces relevant to 1-D mechanistic studies are illustrated [7].**

## DISCUSSION AND CONCLUSIONS

Each reference case includes the physical geometry of the engineered system, materials of construction (including wastes and contaminants where applicable), and environmental interfaces. In addition, the description of each system includes a scenario which with multiple reference states over defined time intervals:

- (i) Initial construction,
- (ii) Operations
- (iii) Closure (with maintenance) and
- (iv) Closure (post-maintenance).

The close state may also have multiple evolutionary states, that include fast pathways or other features that will require consideration in the performance modeling.

Initial definition of the reference cases is focused on a single reference state, i.e., closure (post-maintenance) for waste management units, or operations for operating / storage units. For the purpose of developing algorithms that predict changes in parameters as a function of time and conditions, each reference case was selected to have a plausible system configuration and set of characteristics. However, the reference disposal units are not defined to represent a specific field case. This allows for development and testing over a range of field conditions that cover those encountered across the DOE complex.

Each reference case is a simplification of the actual expected disposal or storage unit and is a conceptual model of a unit. The definition of each reference case is expected to evolve over time as more knowledge is obtained and model uncertainties are addressed in addition to parameter and numerical uncertainties.

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