EVOLUTION OF REPOSITORY, CONTAINER, WASTE FORM CHARACTERIZATION AND DESIGN AT THE PROPOSED US DISPOSAL SYSTEM IN VOLCANIC TUFF

Rob P. Rechard
Nuclear Waste Disposal Research & Analysis
Sandia National Laboratories
Albuquerque, NM 87185-0747, USA

ABSTRACT

Over 30 years, scientists and engineers designed engineered features to complement attributes of the natural barrier of volcanic tuff at Yucca Mountain in southern Nevada such that a proposed repository in the unsaturated zone would safely isolate spent nuclear fuel and high-level radioactive waste over 10^6 years. Initially in the early 1980s, an engineered barrier design applicable to several geologic media was used. With the Congressional direction to characterize Yucca Mountain, the engineered design gradually adapted to conditions in unsaturated tuff in the 1990s. The repository switched from floor emplacement of waste in small, single-walled stainless steel canisters to in-drift emplacement in large, double-layered containers. By 2000, the outer layer was high-nickel alloy to resist corrosion and the inner layer was stainless steel for strength. To avoid localized corrosion during the ~1000-yr thermal period, titanium drip shields were also added above the containers.

In general, flexibility in accommodating various waste forms has been an intended attribute of geologic disposal system designs, rather than tuning the disposal system to specific characteristics of waste durability. The degradation rate of the radioactive waste matrix was an important parameter of the source-term in early modeling analysis. However, by the mid1990s, analyses used fairly rapid degradation rates within the oxygenated environment of the unsaturated zone. Other components of the multiple barrier disposal system compensated for high degradation rates.

INTRODUCTION

Since 1978, the US Department of Energy (DOE) has explored developing a geologic repository at Yucca Mountain (YM), located at the boundary of the Nellis Air Force Range and the Nevada Nuclear Security Site (NNSS and formally known as Nevada Test Site) (Fig. 1).[1] The purpose was to dispose commercial spent nuclear fuel (CSNF), DOE-managed spent nuclear fuel (DSNF), and high-level radioactive waste (HLW) produced when reprocessing a small amount of CSN, prior to 1972, and DSNF until 1992. The design of the engineered barrier to accommodate this waste varied considerably over the life of the Yucca Mountain Project (YMP) as understanding of the natural barrier increased, techniques for excavation advanced, and desires for large containers for high throughput were expressed.[2-4]

To provide perspective on future material issues for radioactive waste management, this paper describes the evolution of the engineered barrier system between 1984, when the YM disposal system was first described, and 2008, when the safety analysis report for the license application (SAR/LA) to construct a repository was submitted to the US Nuclear Regulatory Commission (NRC).

Geologic Disposal System

Similar to all geologic disposal systems, the YM disposal system consists of two main systems: (1) the natural barrier system (NBS) or the geologic setting that hosts the repository, and (2) the engineered barrier system (EBS). The natural barrier must be characterized through scientific study; whereas, parts of the engineered barrier are designed.
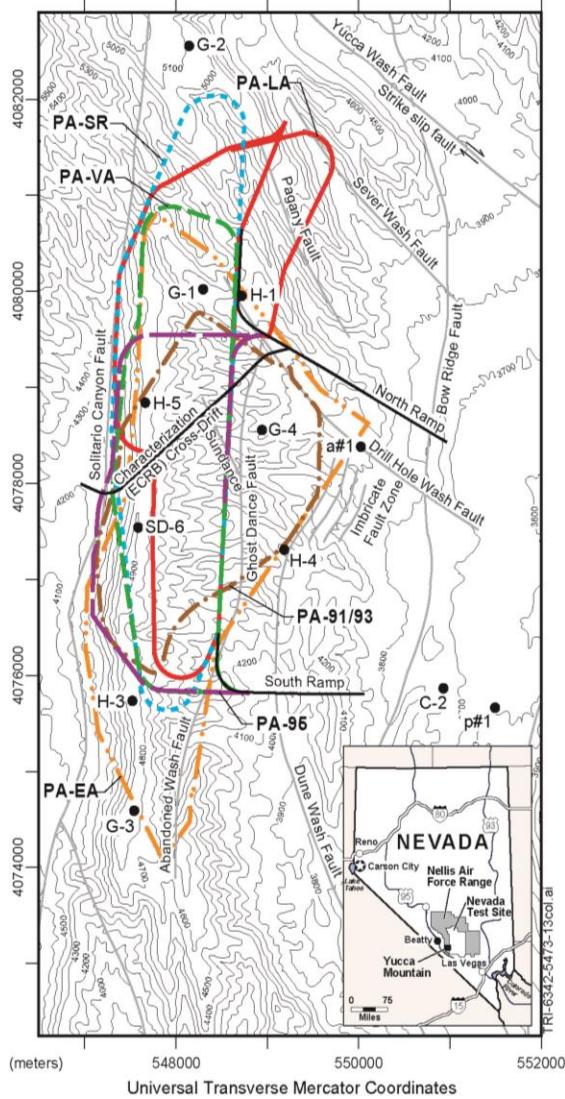
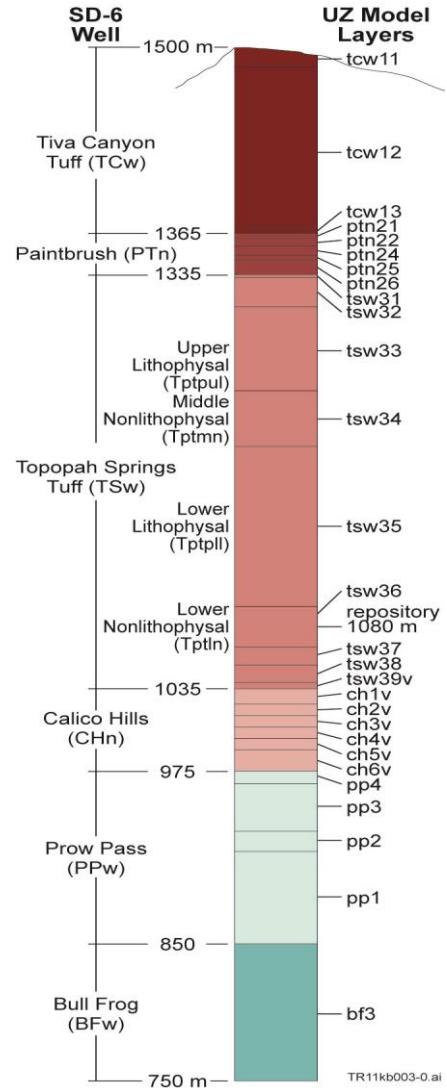Fig. 1. Location and extent of various proposed layouts for YM repository.[2, Fig. 1]

Fig. 2. Stratigraphy of Yucca Mountain volcanic tuff at SD-6 borehole.[2, Fig. 2]

As NRC defined in the 2001 site-specific regulation for the YM repository—10 CFR 63 (and similar to definitions in the earlier 1983 generic regulation—10 CFR 60), the "*engineered barrier system* means the waste packages, including engineered components and systems other than the waste package (e.g., drip shields), and the underground facility" where the "*underground facility* means the underground structure, backfill materials, if any, and openings that penetrate the underground structure (e.g., ramps, shafts, and boreholes, including their seals);" "*waste package* [WP] means the waste form and any containers, shielding, packing, and other absorbent materials immediately surrounding an individual waste container;" and "*waste form* means the radioactive waste materials and any encapsulating or stabilizing matrix." Although much of the engineered barrier is designed, its long-term behavior over 10^4 or 10^6 years must still be determined. For example, waste form designs have not emphasized long-term durability after disposal (e.g., reactor operation dominates design of fuel assemblies and desire for high waste loading dominates design for HLW); hence, waste characterization is also an important activity.

Regulatory Performance Criteria

In the site-specific radiation protection standard for the YM disposal system, first promulgated in 2001, the US Environmental Protection Agency (EPA) requires a performance

2

assessment (PA), which is a quantitative stochastic estimate of dose to a hypothetical individual.[1] EPA limits the arithmetic mean of the various estimates of dose from the highest concentration in a hypothetical contaminant plume no further south than the southern edge of NNSS (~18 km from the repository) to 0.15 mSv/yr for 10^4 years after closure ($t \leq 10^4$ yr) and to 1 mSv/yr for $10^4 < t \leq 10^6$ yr. The earlier generic EPA standard, promulgated in 1985, had limited the cumulative release, rather than dose.[1] The use of dose as a health indicator increases the necessary fidelity of estimating the *rate* of radionuclide release to the accessible environment and, thereby, the importance of the EBS, which can presumably be more readily and accurately characterized for analysis.

EBS Modeling and Analysis Iterations

Two types of models are typically developed for analyzing a disposal system, in general, and the EBS, in particular: (1) a detailed mechanistic model to develop better designs through understanding of EBS behavior, and (2) an EBS model as a subsystem in the PA to understand the EBS role in overall performance. Whether the same model is used for both roles depends upon computational capabilities and the influence of the EBS in the overall PA performance. This paper describes the greatly simplified EBS model used in the PA of the YM disposal system.[3, 4] The YM EBS model evolved to include 3 primary modeling parts: EBS chemistry, container, and waste form. The waste form eventually consisted of 12 components:[4, Figure 18] EBS flow, EBS transport, waste water chemistry, radioelement solubility, sorption on container rust, reversible colloid formation, irreversible colloid formation, inventory decay, DSNF degradation, CSNF cladding failure, CSNF degradation, and HLW degradation. Only the modeling evolution of the container, CSNF cladding, CSNF degradation, and HLW degradation is summarized here.

Seven PA iterations, of the many performed by YMP, provide convenient points to discuss evolution of the EBS.[1-4] The 1984 deterministic analyzes, conducted for Environmental Assessment (EA) required by the *Nuclear Waste Policy Act of 1982* (NWPA) and designated herein as PA-EA, provides the initial marker for the paper. The first stochastic PA, conducted in 1991 (PA-91), serves as the second marker. PA-93 and PA-95, which serve as the third and fourth markers, respectively, provided preliminary guidance on site characterization and EBS design. The congressionally requested viability assessment (PA-VA), completed in 1998, serves as the fifth marker. The conclusion of natural barrier characterization culminated with an analysis in late 2000 for the site recommendation—PA-SR, which serves as the sixth marker. PA-LA, which forms the basis of SAR/LA, serves as the final marker.

ENGINEERED BARRIER FOR 1984 PA-EA

Repository Design

A repository concept was developed for the 1984 PA-EA with a stair-step design to remain horizontal in the welded tuff of the Topopah Spring Formation (TSw of Fig. 2).[2] The YM repository was set >200 m below the surface and >100 m above the water table. These vertical limits remained a design goal for the repository through 2008 PA-LA, 24 years later.

A repository for 70,000 metric tons of heavy metal (MTHM), the maximum size legally allowed in NWPA for the first repository (until a second repository was constructed) required 6 km^2, based on an initial areal power loading of 12 W/m^2. The design criteria was to keep the tuff temperature in the repository < 200 ^{o}C and, thereby, avoid altering zeolitic tuff in layers under the repository.[2] The latter criterion, expressed as a drift wall temperature <200 ^{o}C, remained a design goal through PA-LA.

In PA-EA, the repository disposed 3000 MTHM/yr, a rate ~50% higher than the annual rate of generation of CSNF in the US. This rate would guide repository designs for the next 25 years to ensure that CSNF storage at reactors would decrease once the repository was operating.[2]

Both vertical and horizontal emplacement were considered during the mid-1980s.[2] Vertical emplacement was in the floor of a 6.7 m tall by 6.1 m wide disposal drift with one container placed in a 0.7-m diameter borehole spaced every 9 m. Horizontal emplacement was in the pillars of a 3.7 m tall by 6.1 m wide drift with 35 containers per borehole spaced every 14 m.

Package Design

Initially, a generic small container was used for SNF and HLW, which maintained flexibility for disposal in various geologic media being considered in the early 1980s (Fig. 3). A small container with a low thermal load can be disposed in many media. By 1983, stainless steel and high-nickel alloys were candidate container materials for a container in salt, basalt, or tuff repositories.[2, 3] Copper alloys were also evaluated for the anoxic, saturated zone (SZ) of a basalt repository but were not necessary in salt and not effective in the oxidizing, unsaturated zone (UZ) for the tuff repository.

YMP anticipated both CSNF and HLW in the repository. However, only a 9.5-mm thick 304 stainless-steel container of CSNF was modeled in PA-EA, although other materials and designs were under consideration. At this time, the EBS did not include other components of the EBS that would delay release of radionuclides, such as a corrosion-resistant overpack, borehole liner, or adsorptive backfill. The containers for CSNF from either pressurized water reactors (PWRs) or boiling water reactors (BWRs) were assumed to be 0.65 m in diameter and between 4.0 and 4.75 m long (Fig. 4). About 33,000 containers were anticipated, if CSNF was not consolidated, or ~18,000, if CSNF was consolidated at the repository by removing hardware surrounding the fuel rods.[2, Table 2]

Although HLW was not modeled in PA-EA, designs were available. The pour canisters for defense HLW were to be 0.61 m in diameter and 3 m long (the standard canister used in the vitrification plant at the Savannah River Site); the pour canisters for commercial HLW were to be 0.32 m in diameter and 3 m long (Fig. 3).
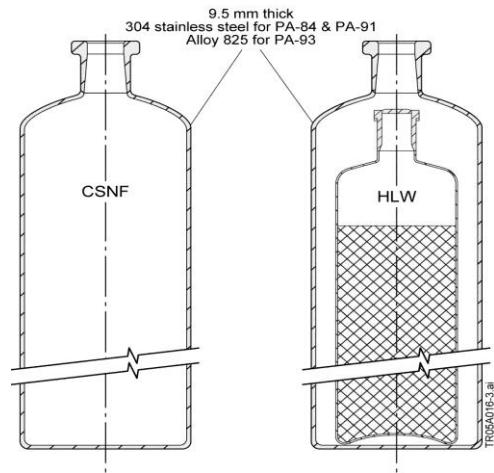

Fig. 3. Small, thin-walled container design of stainless steel or high-nickel Alloy 825 considered for 1984 PA-EA, PA-91, and PA-93 can easily adapt to geologic media that cannot accomodate high heat loads.[3,Fig.2]
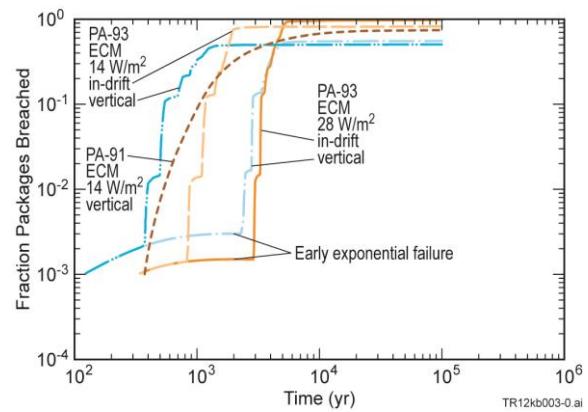

Fig. 4. Fraction of packages breached ($F^{WP}(t)$) for vertically emplaced containers with single Alloy 825 layer (using defined distribution for pitting) and in-drift container with double layer of Alloy 825 and carbon steel (using parabolic equation for general corrosion) at two heat loads in PA-93; comparable release curve shown for PA-91.[3, Fig. 4]

The cumulative fraction of packages breached ($F^{WP}(t)$) was modeled either as (1) instantaneous failure of all packages, or (2) exponential degradation. The time of instantaneous failure was either 300 yr or 1000 yr, which corresponded to the range for the minimum lifetime required in the 1983 NRC implementing regulations (10 CFR 60, §60.113).[1] Instantaneous failure accounted for the susceptibility of stainless steel to stress corrosion cracking.

For exponential degradation, the cumulative fraction of container failure was

$$F^{WP}(t) = 1 - e^{-\lambda_{WP}t} \tag{1}$$

The mean time to failure ($1/\lambda_{WP}$) was 10^4 years. The λ_{WP} was assumed to include both the degradation of the stainless steel container and the degradation of CSNF cladding (i.e., $\lambda_{WP} = \lambda_{can} + \lambda_{clad}$). Exponential degradation accounted for a portion of containers that failed early and the limited ability of water to initially enter containers through stress corrosion cracks.

Waste Characterization

In PA-EA, only advective release of radionuclides from waste ($\Gamma_{CSNF,r}$) (kg/yr-WP) was modeled and assumed to be controlled by the fractional degradation rate of the CSNF fuel matrix ($\Lambda_{matrix}(t)$) (yr^{-1}) and the radionuclide mass per container ($\bar{M}_{CSNF,r}(t)$) (kg/WP). The $\Lambda_{matrix}(t)$ was modeled as a function of uranium solubility (S_U) (kg/m^3), U mass per container (M_U), $F^{WP}(t)$ from Eq. (1)), and quantity of water entering a package (Q^{WP}) (m^3/yr-WP). That is,[4]

$$\Gamma_{CSNF,r}(t) = \Lambda_{matrix}(t)\bar{M}_{CSNF,r}(t) \tag{2}$$

where

$$\Lambda_{matrix}(t) = \frac{S_U}{M_U}Q^{WP}F^{WP}(t) \tag{3}$$

The Q^{WP} was limited to a fraction of the percolation ($f^{perc}q^{perc}$) (m/yr) passing through Yucca Mountain and the intercept area of a package (repository area/number of WP), yet, all containers experienced some seepage. PA-EA did not account for the short time temperatures might exceed 100 $^\circ$C near the boreholes, thus, Q^{WP} was not a function of temperature. The source-term for CSNF did not consider sorption and transport within the EBS domain; hence, $\Gamma_{CSNF,r}$ was used directly in the transport model outside the EBS. The equivalent dissolution rate was quite low (Fig. 5a).
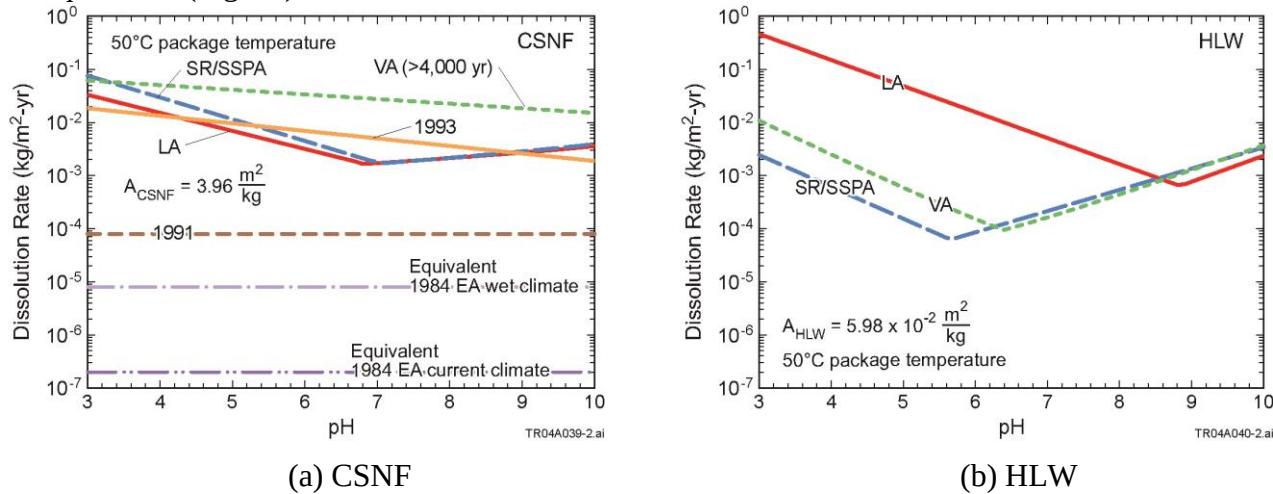

(a) CSNF   (b) HLW

Fig. 5. Dissolution of CSNF and HLW matricies eventually set at similar rapid rates in YMP PAs, but alteration rates differed because of differences in specific surface area.[4, Fig. 20]

ENGINEERED BARRIER FOR PA-91

Repository Design

Similar to PA-EA, the repository design for PA-91 considered either vertical floor emplacement or horizontal pillar emplacement using small waste containers. While ramps were to be constructed using a tunnel boring machine, most of the underground facility was to be constructed with conventional drill and blast methods.

Waste Characterization

Although only degradation of the CSNF matrix was again modeled in PA-91, the conceptual model of CSNF degradation had changed. The radionuclide source rate was dependent on either (1) the solubility of the radioelement, as in PA-EA, or (2) the potentially rapid degradation of CSNF matrix in the UZ. Each radionuclide was assigned, based on analyst judgment, to either the first or second situation in PA-91. In later PAs, however, the release rates from both situations were evaluated and the minimum rate chosen (i.e., $\Gamma_{CSNF,r}(t) = \min\{\Gamma_{CSNF,r}^{\deg}(t), \Gamma_{CSNF,r}^{sol}(t)\}$).

For 5 of the 9 radionuclides tracked, the source rate ($^{sol}\Gamma_{CSNF,r}(t)$) was the product of the solubility of the radioelement e (S_e) and the $Q^{WP}(t)$ entering a package:

$$^{sol}\Gamma_{CSNF,r}(t) = S_e Q^{WP}(t) \text{ for } e \sim {}^{243}\text{Am}, {}^{237}\text{Np}, {}^{239}\text{Pu}, {}^{126}\text{Sn}, {}^{234}\text{U}. \tag{4}$$

For the other 4 radioelements, the source rate $\Gamma_{CSNF,r}(t)$ was a combination of the degradation release from the CSNF matrix (Λ_{matrix}) and instantaneous release (Λ_{prompt}) of a small portion of radionuclides trapped between the CSNF matrix and cladding or trapped in grain boundaries within the CSNF matrix:

$$\Gamma_{CSNF,r}^{\deg}(t) = \bar{M}_{CSNF,r}(t) [(1 - f_r^{prompt})\Lambda_{CSNF}^{matrix}(t) + f_r^{prompt}\Lambda_{CSNF}^{prompt}(t)] \tag{5}$$

for $r \sim {}^{135}\text{Cs}, {}^{129}\text{I}, {}^{99}\text{Tc}, {}^{79}\text{Se}$ where $\bar{M}_r(t)$ was the time varying mass of radionuclide r in a single CSNF package and f_r^{prompt} was instantaneous release fraction. The f_r^{prompt} was not an important parameter even when increased and varied in later PAs.

ENGINEERED BARRIER FOR PA-93

Repository Design

PA-93 considered vertical floor emplacement but also considered in-drift emplacement with large packages. Layout of the underground facility was redesigned to facilitate construction with a tunnel boring machine and, thereby, reduce disturbance of the tuff near the disposal drifts.

Package Design

Both vertical and in-drift configurations in PA-93 and thereafter considered corrosion resistant material for the containers (Fig. 6). General corrosion of carbon steel container under wet conditions (outside the dry-out zone for <100 ^{o}C) for the in-drift package was represented by a parabolic equation. PA-93 also included early failure of containers where a container could fail at early times from manufacturing defects, as represented by an exponential function (Fig. 4).[3]

PA-93 examined the implications of higher temperatures on container degradation for more closely packed packages and higher heat loads, which necessitated introducing a thermal module. That is, PA-93 made the first steps toward evaluating the interaction between the EBS and NBS.
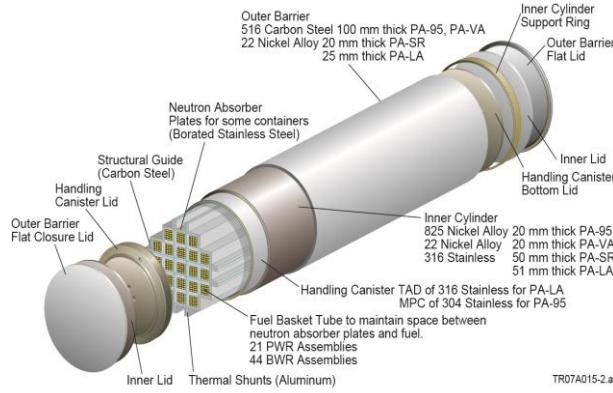
Fig. 6. In-drift container considered for PA-93 and thereafter included nner handling canister for PA-95 (multipurpose canister or MPC) and for PA-LA (transportation, aging, and storage or TAD). [3, Fig. 3]
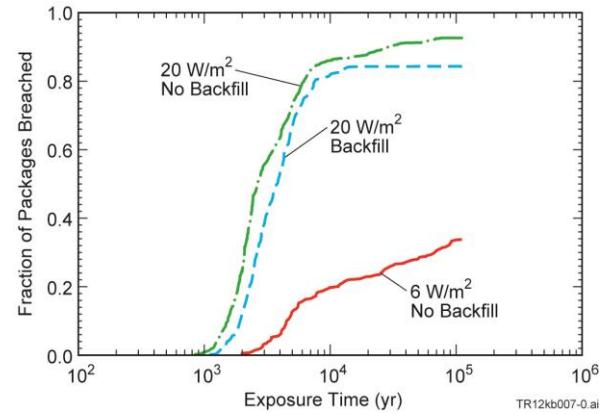

Fig. 7. Fraction of in-drift containers breached ($F^{WP}(t)$) via pits through Alloy 825 layer (using exponential function of temperature) for hot and cool repository with and without backfill in PA-95. [3, Fig. 5]

Waste Characterization

The degradation release rate $\Gamma_{CSNF,r}^{\deg}(t)$ was evaluated with Eq. (5). However, the time dependent evaluation of the matrix fractional releases rates ($\Lambda_{CSNF}^{matrix}(t)$) became more complicated because of the need to account for the breach time of the waste container (τ_{fail}) in addition to $\dot{r}_{CSNF}$ (and the breach time of CSNF cladding in PA-VA and PA-SR). These breach times and degradation rates, which varied with temperature, were estimated at each analysis time step.

The CSNF matrix dissolution rate ($\dot{r}_{CSNF}$) (or fractional alteration rate–yr^{-1} $\dot{a}_{CSNF} = \dot{r}_{CSNF} A_{surf}$ where A_{surf} is the specific surface area) was an important input to the analysis algorithm. The dissolution rate $\dot{r}_{CSNF}(t)$ (kg/m^2-yr) was evaluated using an Arrhenius relationship except that the rate was also a function of the total carbonate concentration ($C_{CSNF}^{CO_3}$) and pH (Fig. 5a):

$$\log \dot{r}_{CSNF}(t) = \kappa_0 - \frac{\kappa_1}{T_{CSNF}^{WP}(t)} + \kappa_2 \log C_{CSNF}^{CO_3} - \kappa_3 pH_{CSNF} \qquad (6)$$

Experimental data were used to evaluate the regression coefficients κ_0, κ_1 (activation energy), κ_2, and κ_3 for PA-93 and thereafter.

ENGINEERED BARRIER FOR PA-95

YMP conducted PA-95 to again provide guidance on (a) characterizing the site, (b) two options for the heat load, and (c) two options for package placement in the repository. [3, 4, 5, Table 1]

Repository Design

In 1994, DOE formally adopted the use of in-drift emplacement of large packages. The appeal was the ability to more easily receive 3000 MTHM/yr of SNF and HLW since fewer (~⅔ less) packages were emplaced. A mountain site facilitated use of large packages since gently inclined ramps could be used to move large, heavy packages by rail into the repository.

The PA-95 design closely spaced the drifts 28 m apart such that decay heat would dry out the UZ for ~1000 yr.[2] The moderately hot and more compact repository fit west of the Ghost Dance Fault (Fig. 1). The design saved tunneling costs by reducing the repository to ~200 km of drifts (160 drifts up to 1200 m long).

Package Design

For PA-95, the container consisted of an inner layer of 20-mm nickel Alloy 825 for corrosion-resistance, and an outer layer of 100-mm thick carbon steel for structural strength and corrosion sacrificial allowance. The package also included a 25-mm thick multipurpose handling canister (MPC) of 304 stainless steel (Fig. 6).[2, Table 3] The MPC was not considered as a barrier.

PA-95 examined the influence of percolation, seepage, and relative humidity on the thermal regime in an expanded thermal-hydrologic module. With the proposed change to an individual dose performance measure, PA-95 also took the major step of modeling corrosion stochastically to evaluate the diffusive and advective barrier capability of the container.[3] Radionuclides sorption on rust as the container degraded was not include but would be for PA-SR and PA-LA.

Two modes of corrosion were considered for the outer carbon steel corrosion allowance layer: humid air corrosion and wet (aqueous) corrosion. For humid air corrosion, the layer removal was an exponential function:

$$s_{steel}^{air}(t) = e^{\kappa_0 + \frac{\kappa_1}{T_{CSNF}^{WP}(t)} + \frac{\kappa_2}{RH_{CSNF}^{drift}(t)} + \kappa_3 \ln t} \quad (\mu m) \tag{7}$$

where $RH_{CSNF}^{drift}(t)$ was the relative humidity (RH) in the drift, and $T_{CSNF}^{WP}(t)$ was the temperature of a representative CSNF package (evaluated for two heat load cases with and without backfill and two infiltration cases). The parameters κ_0, κ_1, and κ_2 were determined from published literature.

Wet corrosion of the carbon steel outer layer was modeled as

$$s_{wet}^{steel}(t) = e^{\kappa_0 + \frac{\kappa_1}{T_{CSNF}^{WP}(t)} + \kappa_3 \ln t + \kappa_4 T_{CSNF}^{WP}(t)^2} \quad (\mu m) \tag{8}$$

where the parameters κ_0, κ_1, κ_3, and κ_4 were determined from published literature.

Only wet, pitting corrosion was modeled for the Alloy 825 layer. The pit penetration rate was an exponential function of temperature with parameters based on scientific judgment (Fig. 7).[3]

$$\dot{s}_{wet}^{A825\,pit}(t) = e^{\kappa_0 - \frac{\kappa_1}{T_{CSNF}^{WP}(t)}} \quad (mm/yr) \tag{9}$$

Waste Characterization

The degradation rate for CSNF was of the same functional form as PA-93 (Eq. (6)) with a generic CSNF package temperature ($T^{WP}(t)$) calculated for 2 heat loads, 2 backfill options, and 2 moisture infiltration cases.[3]

The degradation rate of HLW encapsulated in borosilicate glass ($\dot{r}_{HLW}(t)$) was derived from transition state theory to obtain a chemical affinity reaction rate equation (Fig. 5b):[3]

$$\dot{r}_{HLW}(t) = \kappa^0(pH(t), T(t)) \cdot \left(1 - \frac{C_{SiO_2}(t)}{K_{SiO_2}}\right) \tag{10}$$

where κ^0 was the forward or intrinsic dissolution rate and an empirical function of time varying temperature and pH, obtained from regression analysis of experimental results.[4]

The second term of Eq. (10) is the chemical affinity, which accounts for reduced glass dissolution as the silica concentration in the solution surrounding the glass ($C_{SiO_2}(t)$) gradually increases to the solubility equilibrium constant for amorphous silica dissolution (K_{SiO_2}). For PA-95, however, the chemical affinity term was a function of temperature (i.e., $C_{SiO_2} / K_{SiO_2} = \hbar\{T_{CoWP}^{WP}\}$), because $C_{SiO_2}(t)$ was not yet available during the calculation; hence, Eq. (10) became a first-order reaction.
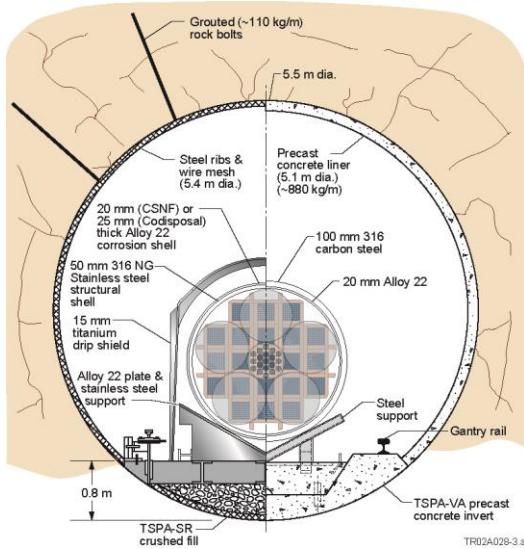
Fig. 8. Although PA-VA used concrete; PA-SR used steel and titanium drip shields to limit altering water chemistry and influence of water drips during thermal period.[2, Fig. 9]
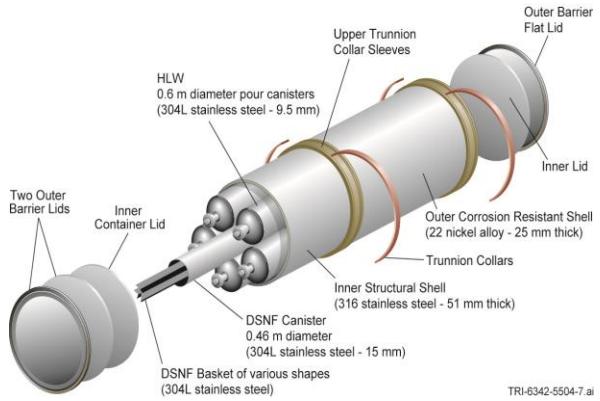

Fig. 9. Co-disposing DSNF with HLW limited fissile content in package yet made efficient use of space for PA-VA and thereafter.[3, Fig. 6]

ENGINEERED BARRIER FOR 1998 PA-VA

PA-VA was conducted to demonstrate the viability of the YM disposal system to Congress.[3, 4, 5, Table 1] YMP used the most current information as interpreted by expert panels prior to completion of experiments. For example, a Waste Package Degradation Expert Elicitation Panel, which consisted of 6 experts with three outside DOE, evaluated the status of current waste package models or proposed for PA-VA, and aggregated disparate data available in the literature.

Repository Design

For the PA-VA, the repository was extended farther north than in PA-95 (Fig. 1). The repository was roughly horizontal and primarily in the lower lithophysal unit (Tptpll) with a small fraction in the middle non-lithophysal unit (Tptpmn) and lower non-lithophysal unit (Tptpln) (Fig. 2). A moderate temperature repository concept with 28-m drift space was used for PA-VA. Spacing down the drift between hot CSNF packages was large and varied between 10.6 and 8.9 m. A cool HLW/DSNF package might be placed between two CSNF packages and in this case spacing between packages was reduced to 2.3 m.

Backfill to create a capillary barrier was studied as an option, but its use was not recommended. Backfill caused operational difficulties in filling the long drift. Also, fine backfill would insulate the waste containers and promote internal temperatures above 350 $^{\circ}$C for the large containers during the first 1000 yr after emplacement, which could prematurely damage CSNF cladding. Steps taken to maintain operational safety and allow package retrieval included pre-cast concrete lining and invert for the 5.5 m diameter disposal drifts (Fig. 8).

Package Design

For PA-VA, the inner 20-mm Alloy 825 layer was replaced with another high-nickel Alloy 22 to more reliably increase life. Also, DSNF was included in PA-VA and envisioned to be mostly co-disposed with HLW. Co-disposal of 5 HLW canisters with usually one DSNF canister reduced the need for criticality control inside the package since the DSNF fissile mass was limited (Fig. 9). Generalized corrosion of the Alloy 22 was added for PA-VA and represented by probability density functions (PDF) at 3 different temperatures at humid and wet conditions, as assigned by expert judgment, though humid corrosion was unimportant (Fig. 10).
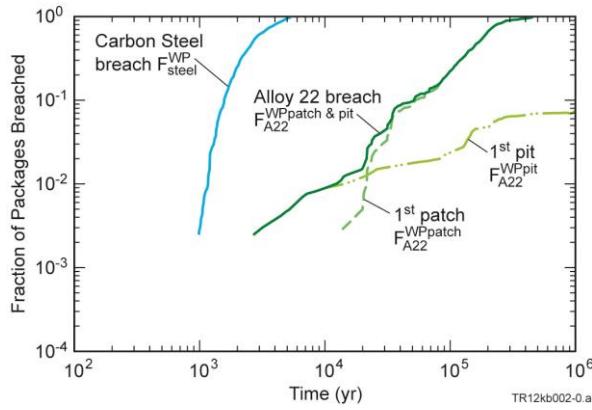
Fig. 10. Fraction of in-drift CSNF containers breached ($F_{CSNF}^{WP}(t)$) for PA-VA; general corrosion of Alloy-22 patches used PDFs set at 3 temperatures and pitting corrosion used exponential function of temperature.[3, Fig. 9]

Fig. 11. Ti drip shield and stainless steel invert and ground support for PA-LA.[2, Fig. 13]

Waste Characterization

Since the 1950s, most CSNF has been clad with between 600 through 900 μm of corrosion resistant Zircaloy, an alloy that is ~98% zirconium with small amounts of tin, iron, nickel, and chromium. The Zircaloy cladding was not a designed engineered barrier for disposal, but rather an existing characteristic of CSNF that could influence the release rate of radionuclides once the container breached. YMP was generally reluctant to account for its presence because of the perceived difficulty in defending its condition at disposal and uncertainty in degradation performance over time. Thus, cladding performance was not considered in early PAs. However, NRC used cladding data as the basis for extending the period of wet storage, for licensing dry storage facilities, and for licensing shipping casks for CSNF. Hence for PA-VA, YMP scientists added a rudimentary component for cladding degradation to explore the impact. At 10^6 years, the total fraction of fuel exposed was between 1.7 and 52%. Including cladding had a major effect on dose because the cladding prevented release of 48% to 97.3% of the inventory,[4, 5] because degradation of the CSNF matrix could not proceed until the cladding had perforated.

Once perforated, the exposed CSNF matrix degraded at the rate ($\dot{r}_{CSNF}$) as determined from an 11-term regression equation dependent on temperature, water chemistry (pH, total carbonate concentration, and oxygen partial pressure) and several cross-products. The coefficients were based on batch experiments with irradiated CSNF.[3]

For HLW, Eq. (10) was used in PA-VA, except the model added a long-term residual reaction rate (k_{long}) when the surrounding water film solution was saturated with silica (Fig. 5b). The wet dissolution rate ($\dot{r}_{HLW}^{wet}(t)$) was used for all environmental conditions because it was surmised that the usually high relative humidity in the drifts would ensure adsorbed water films on the glass.[4] The dissolution rate varied over 5 orders of magnitude during the simulations; yet, the HLW dissolution rate was not a sensitive parameter in PA-VA.[5, Table 2]

ENGINEERED BARRIER FOR 2000 PA-SR

By December 2000, YMP completed PA-SR to support the decision of the Secretary of Energy and President to recommend the YM site for a repository.[1] PA-SR made liberal use of conservatisms in model choices and parameter assignments, and in July 2001, YMP conducted a more realistic perspective of disposal system performance (along with an evaluation of an alternative cooler repository design): the Supplemental Science Performance Analysis (SSPA).
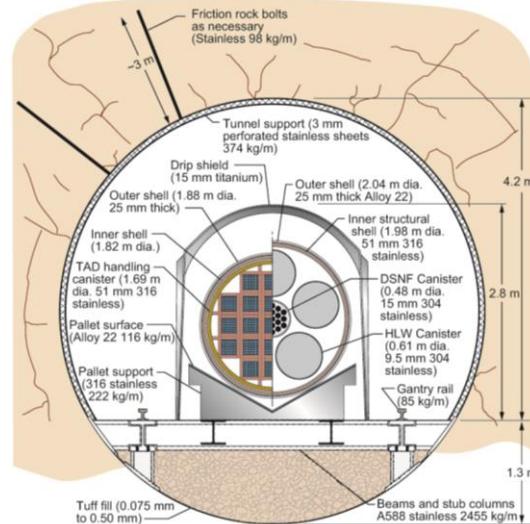
Another analysis was completed for the 2002 final Environmental Impact Statement (EIS) on site-suitability (PA-EIS), which built upon SSPA.

Repository Design

For the 2000 PA-SR and 2008 PA-LA, spacing between drifts was increased to 81 m to allow water to percolate through cool pillars. However, some flow percolating through fractures could become drips, even though a large drift cavity provided a capillary barrier. Titanium drip shields were added to avoid drips on the package and, thereby, reduce the potential for localized corrosion of Alloy 22 during the ~1000-yr thermal period (Figs. 8 and 11).

Package Design

Because the repository was in the UZ, the drifts did not need to be backfilled (at least for an extended period); hence, convection and radiative heat transfer in the open drifts lowered temperatures on the large package surface. Yet, temperatures in the drift exceeded boiling, which prompted questions about the coupling of thermal, hydrologic, and chemical processes during the ~1000 yr thermal period. Hence, YMP conducted much experimental work and code development to advance coupled thermal-hydrologic-chemical modeling. Eventually, concepts from the coupled process modeling were incorporated into a simplified EBS chemistry module for PA-SR and PA-LA to evaluate the possibility of localized corrosion of the Alloy 22 outer layer. However, in both PA-SR and PA-LA, the range of chemical conditions was generally mild and not conducive to localized corrosion; thus, the containers were quite long lived (Fig. 12).
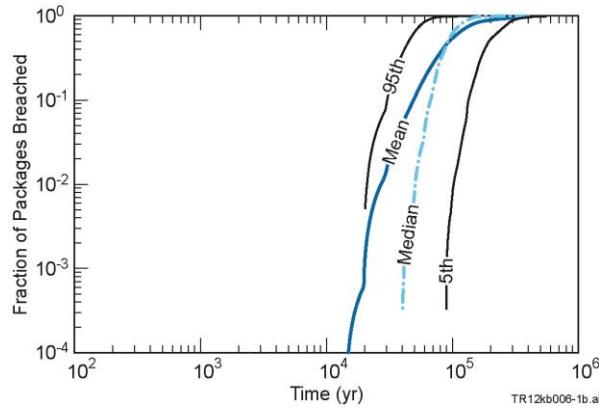

Fig. 12. Calculated distribution for fraction of Alloy 22 outer layer breached ($\mathcal{G}\{F^{WP}(t)\}$) from generalized corrosion (since pitting omitted by drip sheilds) in PA-SR.[3, Fig. 11]

Fig. 13. Wide distribution for fraction of CSNF containers breached was calculated when possibility of seismic damage added in PA-LA ($\mathcal{G}\{F_{CSNF}^{WP}(t)\}$).[3, Fig. 13]

Waste Characterization

The cladding component was refined for PA-SR .Two steps for cladding degradation were included, perforation and unzipping, rather than just perforation as in PA-VA. Three perforation modes were defined:

$$F^{clad}(t) = F_{initial}^{clad}(0) + F_{creep}^{clad}(^{\max}T_{CSNF}^{WP}) + F_{corr}^{clad}(t; \tau_{CSNF}^{fail}(t)) \tag{11}$$

where $F_{initial}^{clad}(0)$ was the fraction of cladding initially failed when CSNF was placed in containers; $F_{creep}^{clad}(^{\max}T_{CSNF}^{WP})$ was the fraction of cladding failed by creep and stress corrosion cracking during storage, transportation, and emplacement (~8%);[4] $F_{corr}^{clad}(t; \tau_{CSNF}^{fail}(t))$ was the fraction of cladding failed by localized corrosion from some unknown chemical species such as iodide or fluoride in
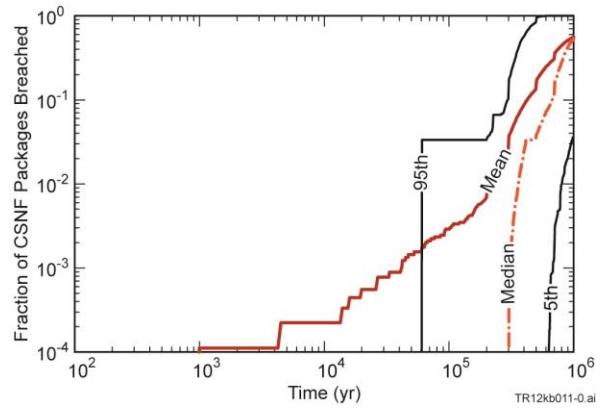
water entering a failed container much later in time. F^{clad} was set to 1 when a seismic event caused sufficient ground motion to perforate all the cladding.

Once cladding was perforated, unzipping of the cladding could occur as the UO_2 of CSNF formed schoepite with its larger volume. The alteration rate was assumed proportional to the degradation rate of the UO_2 matrix. The unzipping of the cladding was quite rapid in an oxic environment. Hence, CSNF cladding was not important in PA-SR and not considered for PA-LA.[5, 13] While the container was structurally intact, the container provided substantial diffusive and advective resistance to flow even without the presence of the cladding.[4, 5]

For PA-SR and PA-LA, the generic rate of CSNF degradation in a container that failed at time τ^{fail} was expressed similar to PA-95 (Eq. (6)) (Fig 5a):[4]

$$\log\{\dot{r}_{CSNF}(t;\tau_{CSNF}^{fail}(t))\} = (\kappa_0 U^{a_0} + \kappa_1/T_{CSNF}^{WP}(t) + \kappa_2 \log C^{CO_3}(t;\tau_{CSNF}^{fail}(t))$$
$$+ \kappa_3 pH(t;\tau_{CSNF}^{fail}(t)) + \kappa_4 \log P^{O_2}) \tag{12}$$

where U^{a_o} was a scale factor to account for overall uncertainty, P^{O_2} was the partial pressure of oxygen, and κ_0, κ_1, κ_2, κ_3 and κ_4 were pH-dependent regression coefficients determined from experimental results.[3] For PA-LA, more data was available, especially at low pH, and the 5 coefficients were adjusted slightly.

For PA-SR and PA-LA, the HLW degradation model was simplified to an Arrhenius rate equation.[4] The model of glass dissolution rate ($\dot{r}_{HLW,b}(t)$) was purposefully biased to bound the initial rate when immersed in liquid water when the relative humidity was > 5% (Fig. 5b):

$$\dot{r}_{HLW}(t) = \kappa_{pHseg}^0 \bullet 10^{\eta_{pHseg} pH_{CoWP}(t)} \bullet e^{-\frac{E_{pHseg}^a}{\Re T_{CoWP}(t)}} \tag{13}$$

where $\dot{r}_{HLW}(t)$ was a piece-wise linear function with two pH segments (pH_{low}, pH_{high}); κ_{pHseg}^0 was the intrinsic glass dissolution rate constant in either the low or high pH segment and no longer a function of temperature. It was dependent on pH only as a segment switch. The 2nd power term represented the catalyzing influence of either H^+ of OH^- where η_{pHseg} was a coefficient dependent upon either the low or high pH segment, and $pH_{CoWP,b}(t)$ was the time-varying pH in a co-disposal package. The 3rd exponential term represented the increase in SiO_2 hydrolysis as the temperature increased where E_{pHseg}^a was the activation energy in either low or high pH segment, $T_{CoWP}(t)$ was the time-varying temperature in a co-disposal package, and $\Re$ was the universal gas constant. As a bounding equation, the reaction affinity term (Eq. (10)) was omitted since its influence was to decrease the dissolution rate. Also, a long-term residual reaction rate (k_{long}) was not necessary with the omission of the affinity term. Parameter uncertainty was still large for PA-SR but reduced for PA-LA.

ENGINEERED BARRIER FOR 2008 PA-LA

A compliance analysis, to support an NRC license application to construct the repository, was started after Congressional authorization in July 2002.[1, 5, Table 1] Following two interim PAs in 2004 and 2005, PA-LA was completed in June 2008.

Repository Design

For PA-LA, YMP embraced a staged construction approach for the repository and surface facilities as tentatively proposed in the 1986 EA. Also, YMP used a modular design strategy for the repository underground and surface facilities to be (a) consistent with the annual funding available, and (b) able to flexibly adapt to design changes in the container and waste form.

Drift disposal without backfill increased the importance of the disruptive events such as seismic and igneous intrusion. The effects of seismic ground motion on in-drift disposal, as modeled in the 2008 PA-LA, included fault displacement; rockfall damage from a collapsed drift; degraded internal supports, which allowed spent fuel assemblies to puncture the container; and containers hitting other containers and the invert supports. The possibility of seismicity broadened the time when containers might breach (Fig. 13).

Package Design

For PA-LA, the temperature dependence of general corrosion and early container failure was reintroduced and the stress corrosion cracking model for the Alloy 22 outer layer of the container updated. PA-LA added a 25-mm thick transport, aging, and disposal (TAD) handling canister loaded and sealed at the utility, which eliminated handling assemblies whenever transporting, storing, or disposing CSNF (Fig. 6). The TAD was functionally similar to the proposed MPC in PA-95. The TAD handling canister made CSNF containers less susceptible to seismic damage.

GUIDANCE FOR FUTURE DIRECTION OF EBS STUDY

The influence of waste form behavior and its uncertainty is diminished in geologic disposal because other barriers often control the release, whether by design in the case of a robust container or by existing conditions in the natural barrier. Indeed, a benefit of geologic disposal is to provide sufficient flexibility to accommodate a variety of SNF from commercial reactors, experimental reactors, and HLW from reprocessed SNF from current and future fuel cycles.

The role of the EBS differs with the host geologic media. For all repositories, the container provides important short-term radionuclide confinement for operations and when retrieval might be necessary. However, long-term container and waste form performance is less important for a mined repository in salt and clay/shale because the NBS provides substantial long-term isolation. In these geologic media, the long-term EBS function consists primarily of drift and shaft seals.

Long-term radionuclide isolation is important for a mined repository in crystalline rock and volcanic tuff, when using a dose standard. In these media, the NBS functions to establish a long-term stable environment for the important EBS barrier. In the YM disposal system, for example, the container provides substantial diffusive and advective resistance to radionuclide transport while the container is structurally intact; thus, disposal performance relies less on degradation rates of HLW glass, CSNF cladding, and CSNF/DSNF matrix. To fully accounting for the performance of the Alloy 22 corrosion-resistant waste container in the YM disposal system modeling evolved from basic assumptions of complete failure of the container in early PAs, when using a cumulative release performance measure, to a stochastic description of container degradation in PAs after 1993, when using an individual dose performance measure..[5, Table 2]

The interaction between the EBS and NBS is also important when long-term EBS performance is important. PA-93 introduced a thermal model to evaluate this interaction. Each successive PA would evaluate this EBS-NBS interaction in more detail. In the later PAs, container degradation was a complex function of both the thermal and chemical history and provides a motivation for developing efficient EBS mechanistic models and using high performance computing in the future.[4]

Although disposal performance influenced EBS design, repository operations did also. Flexibility in the size of the handling container is useful in order to accommodate various thermal loads when searching for a new geologic disposal site (as is the current situation in the US). Specifically, the thermal load of the waste container determines when it can be disposed and the requirements for drift spacing, long-term ventilation prior to closure of the repository in various geologic media. A small container with a low thermal load can be disposed in many media, while a large container with a high thermal load cannot (such as crystalline rock with a

surrounding clay engineered barrier) unless the container is cooled in long-term storage (either on the surface or in the repository with ventilation). For applications in saturated clay or crystalline environments, enhancing natural materials in order to withstand high heat loads from large containers, without extensive surface or drift ventilation, would facilitate operations and increase disposal options.

After the host medium has been selected, a waste management organization can optimize the EBS design. For example, in-drift disposal of a large container with a high heat load improved the operation of the YM repository by reducing repository size and number of containers emplaced. The YMP could, thereby, more easily accommodate a high 3000 MTHM/yr rate of disposal for the large amounts of CSNF in the US. Similarly, a TAD handling canister eased surface loading operations.

Crystalline and tuff host rocks, currently depend on the long-term isolation provided by the container to flexibly dispose of a wide variety of waste forms without fully modeling waste behavior. However, thorough knowledge of waste form behavior in a variety of environments, conditioning the EBS environment to lower radioelement solubility, and/or developing waste forms with an emphasis on long-term disposal durability may make container longevity using expensive alloys less important in the future. An example of a durable waste form includes silicon-carbide coatings on reactor fuels, such as on pellet fuel for high-temperature gas reactors.

ACKNOWLEDGEMENTS

REFERENCES
[1]R.P. Rechard, Milestones for Selection, Characterization, and Analysis of the Performance of a Repository at Yucca Mountain, SAND2015-1060 Sandia National Laboratories (2015).(465 refs)
[2]R. Rechard, and M.D. Voegele, Evolution of Repository and Package Designs for Yucca Mountain Disposal System, Reliability Eng Sys Safety, **122**(2), 53-73 (2014). (150 refs)
[3]R.P. Rechard, J.H. Lee, E. Hardin, and C.R. Bryan, Waste Package Degradation from Thermal and Chemical Processes in Performance Assessments of the Yucca Mountain Disposal System, Reliability Eng Sys Safety, **122**(2), 145-64 (2014). (71 refs)
[4]R.P. Rechard, and C.T. Stockman, Waste Degradation and Mobilization in Assessments of Yucca Mountain Disposal System, Reliability Eng Sys Safety, **122**(2), 165-88 (2014). (91 refs)
[5]R.P. Rechard, Results from Past Performance Assessments of the Yucca Mountain Disposal System, Reliability Eng Sys Safety, **122**(*2*), 207-22 (2014). (62 refs)