

UNITED STATES NUCLEAR WASTE TECHNICAL REVIEW BOARD

2300 Clarendon Boulevard, Suite 1300 Arlington, VA 22201

June 19, 2001

MEMORANDUM

To: Participants in International Workshop on Long-Term Extrapolation of Passive

Behavior

From: Carl Di Bella

Subject: Background Information on Waste Package Environment

Below please find a brief background statement for the workshop. The primary purpose of the statement is to define the possible range of environments that waste packages might be subject to after being emplaced in a proposed repository in Yucca Mountain. This statement complements a separate statement (which we hope to send Friday) containing background information on corrosion and the two questions that will be addressed at the workshop.

Background — Repository

The United States, through the U.S. Department of Energy (DOE), is characterizing a site at Yucca Mountain, in the state of Nevada, for its suitability as the site of a repository for high-level nuclear waste. If the site is determined to be suitable and the U.S. Nuclear Regulatory Commission licenses subsequent construction and operation plans, a repository would be built at Yucca Mountain and would begin operating in approximately 2010. Yucca Mountain is about 150 km northwest of Las Vegas (Figure 1).

As currently conceptualized by DOE, the repository would consist of an underground area of approximately 500 hectares containing approximately 60 km of 5.5m-diameter, near-horizontal tunnels (Figure 2). The tunnels all would be essentially on the same plane, approximately 1,100 m above sea level. The distance from the tunnels to the surface would vary because of variations in surface topography but would be at least 200 m.

The waste would be emplaced horizontally in the tunnels in sealed cylindrical waste packages. Although the dimensions of the waste packages would vary somewhat, depending on the waste in them, they would be approximately 1½-2 m in diameter and 3½-6 m long (Figure 3). The repository would contain more than 10,000 such packages. The outer shell of each waste package would be 20-mm-thick Alloy 22 (UNS No. N06022; nominal composition: Ni59, Cr20.4, Mo14.1, W3.2, Fe2.3). A 50-mm-thick inner shell of type 316NG stainless steel is included in the design for mechanical strength.

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¹ This document is an effort to simplify some very complex topics and does not necessarily represent the views of the U.S. Nuclear Waste Technical Review Board (Board).

Nuclear waste generates heat by radioactive decay. After emplacement of the waste packages and before closure of the repository, most of the heat would be removed by ventilation with outside air (Figure 4). After the last waste package is emplaced, there would be a period of ventilation that is yet to be determined (at least 25 years and as much as 300 years or more). At the end of the ventilation period, a decision would be made to close the repository, titanium (grade 7) drip shields would be placed over the waste packages (See Figure 5.), and all penetrations (e.g., ventilation shafts, entrances, exits) from the surface to the repository would be sealed permanently.

Waste Package Environment

Yucca Mountain is in a desert environment. Average yearly precipitation is about 170 mm. Runoff and evaporation rates are high, and only a fraction of the precipitation infiltrates into the mountain. The water table is approximately 300 m below the tunnels of the conceptual repository. Thus, the repository would be located in what is known as the "unsaturated (vadose) zone." The pores in the rock in the unsaturated zone contain water and air, and the relative humidity at the level of the repository under undisturbed conditions (i.e., before any tunneling operations take place) is higher than 90%. Gas pressure in the vadose zone is essentially atmospheric. The climate at Yucca Mountain has been wetter in the past, and it is likely to become wetter at some time in the future. However, although extended periods of higher average yearly precipitation at Yucca Mountain are likely in the future, such higher precipitation is unlikely to cause the water table to rise sufficiently to flood the repository.

The rocks of Yucca Mountain, from the surface down to and somewhat below the water table, are varieties of tuff. Tuff is a high-silica rock deposited as a fine ash from volcanic eruptions and then welded together to varying degrees by heat and weight. The rock is fractured, and many of the fractures can transmit water. The water below the water table has been well characterized. It is a slightly oxidizing, slightly basic, bicarbonate-type water. (See table below, which shows major species in water below the water table before and after evaporation to a concentrated brine and water in rock pores at the repository level before and after evaporation to a concentrated brine. The many minor species in these two waters are not shown.).

	Nominal Composition (mg/L)			
Major	Water below	Brine formed by	Water in rock	Brine formed by
Species	the water table	evaporating water	pores at	evaporating water
	(based on many samples)	from below the water	repository level	from rock pores to
	samples)	table to near dryness	(based on few samples)	near dryness
Na ⁺	45	850	60	500
K^{+}	5	700	8	500
Ca ⁺⁺	10	1	100	200
Mg^{++}	2	<.1	17	300
SiO_2	60	760	70	60
C1	7	850	120	500
SO ₄	18	850	115	15
HCO ₃	130	300	220	<3
NO_3	9	860	8	500
F ⁻	2	700	1	100?

Evaporating a sample of water from below the water table to concentrated brine yields a high-pH brine rich in Na and K but relatively depleted in other cations. The water in the pores at the level of the conceptual repository has not been well characterized; although it seems to be slightly oxidizing and near-neutral in pH, it also appears to have higher relative concentrations of Mg, Ca, and Cl. (See the table.) Evaporating porewater to a strong brine seems to result in a near-neutral brine of a different composition (rich in Na, K, Ca, and Mg) than would be obtained by evaporating water below the water table to a strong brine.² Of interest from a localized corrosion standpoint, there seems to be on evaporation of both waters appreciable enrichment of not only aggressive anions (e.g., Cl, F) but also beneficial anions (e.g., NO₃).

During the preclosure period (i.e., during ventilation), the relative humidity of the air in the emplacement tunnels would be very low and the waste package surface temperatures would be less than approximately 90°C. Corrosion occurs under these conditions but at such an infinitesimally slow rate that it is not considered an issue in comparison to aqueous corrosion.

After the preclosure period, there are essentially two possible scenarios, depending largely on the duration (and efficiency) of ventilation during the preclosure period. One scenario, known as the "above-boiling" case or sometimes simply the "hot" case, would occur if the ventilation period were comparatively short (say, 25-50 years after emplacement of the last waste package). In this case, representative temperatures at the waste package surface would rise to above the boiling point of water (pure water boils at approximately 96°C at the repository elevation) within a few weeks after closure, peak at approximately 160°C in a few years, and remain above boiling for 500-1,000 years. *Note:* The postclosure temperatures of a waste package depend principally on the thermal power of the waste in the package and of the waste in its immediate neighbors, the distance of the package from its nearest neighbors, and the position of the package in the repository. (For example, packages close to the edge of the repository where ventilating air enters during the preclosure period would be cooler.)

The other scenario, known as the "below-boiling" case or sometimes simply the "cold" case, would occur if the ventilation period were comparatively long (say, 200 years or more after emplacement of the last waste package) and the distance between waste packages were somewhat longer. In this case, waste package temperatures would rise after closure but would peak at no more than 85-90°C. Time-temperature curves for a representative waste package for both a hot repository design and a cold one are depicted in Figure 6.

There is a diversity of opinions on whether a hot or a cold repository would be better. Advocates of a hot repository argue, among other things, that the heat would drive away as vapor substantially all of the water in the pores of the rock at least several meters into the tunnel walls and that the water would condense far away from the waste packages and drain between the tunnels. (The tunnels would be far enough apart that some space between the tunnels would always be below boiling.) Furthermore, they argue, because liquid water would be driven away

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² The brines referred to in this and the previous paragraph and in the table were produced by slowly evaporating synthesized water samples from below the water table or from the rock pores at approximately 80°C in an open system until more than 99% of the water was evaporated.

during the above boiling period, there would be no liquid water on the waste package surfaces, and therefore corrosion rates would be negligible during that period.

Advocates of a cold repository argue that the heterogeneity of the fractures in the rock renders futile any attempts to predict local water movement near the tunnels or between the tunnels, other than in a gross sense. Further complicating the ability to predict water movement, they say, are the inevitable collapse of parts of some tunnels at some time after closure and the difficulty of characterizing fractures between tunnels in other than a gross sense. Cold repository advocates point out that thermomechanical and thermochemical effects could alter the transmissivity of the fractures in ways that are difficult to predict locally. They also argue that in a hot repository the combined effects of boiling-point elevation (due to dissolved salts), deliquescence, capillarity, and superheating indicate the possible presence of liquid water on waste package surfaces at temperatures at least as high as 160°C and that data on the corrosion behavior of Alloy 22 under such conditions at Yucca Mountain are, at best, limited (see later). Finally, these advocates argue, any hot repository will eventually cool down and become a cold one. Thus, they claim, a hot repository would have not only whatever uncertainties might be associated with a cold one but also the additional uncertainties that might be associated with above-boiling repository temperatures.

The Board's position on the hot-versus-cold repository issue is that DOE should perform a technically defensible and quantitative evaluation and comparison of hot and cold repository designs to clarify advantages and disadvantages for performance and uncertainties about that performance.

From a corrosion perspective, it is important to know whether there is *liquid* water on the waste package surface and, if so, the temperature of the water and the chemical species in it. Also important are the solid materials that may be in contact with waste packages, because these solids could influence the composition of the water and could form crevices and local occluded zones. For example, during the preclosure period, dust would be deposited on the waste packages. The composition of the dust is unknown and is a matter being studied by DOE. (Although suggestions have been made to wash emplaced waste packages just before repository closure or to scrub or filter inlet air, or both, no such steps are in the current design.)

If the dust contains deliquescent salts and the relative humidity is above the deliquescence point of the dust, deliquescence could cause liquid water to form on the waste package at temperatures well above 96°C. For example, if the dust had a composition similar to the brine that would result by evaporating water from below the water table, then liquid water could form on the waste package at temperatures as high as 120°C (a temperature corresponding closely to the deliquescence point of sodium nitrate). If the dust had a same composition similar to the brine resulting from evaporating water found in rock pores at the repository level, then liquid water could form at temperatures as high as 160°C (a temperature that may correspond to the deliquescence points of calcium chloride or magnesium chloride).

Besides dust, other solid materials close to the waste package would include carbon steel (used to support the walls of the tunnels), hydrated cement (used in grout for rock bolts), stainless steel (used in the pallet that supports the waste package), titanium (used in the drip

shield), and tuff. (See Figure 5, which portrays all constructed elements that would be in the emplacement tunnels. Collectively, these elements are known as the "engineered barrier system.") Over time, as degradation of the engineered barrier system progresses, all of these materials are likely to come into contact with some parts of some waste packages for extended periods. A particular concern is very high pH (e.g., 12-13+) deriving from water in the pores of cementitious grouts. Another concern is low pH, oxidizing environments caused by iron corrosion products. (The buffering nature of and reactivity of carbon dioxide in the air would bring both high and low pH's closer to neutral if there is chemical equilibrium.)

As mentioned above, titanium drip shields would be placed over the waste packages shortly before closure of the repository. The primary purpose of the drip shields is to deflect water that may drip from the roof of the emplacement tunnels. However, there are at least two reasons that drip shields would not necessarily ensure that liquid water and waste package surfaces would not be in contact. One reason is that any deliquescent salts in the dust on waste package surfaces could form brines if the relative humidity is higher than the deliquescence point of the salt. The other reason is that very thin layers of liquid water can form on clean metal surfaces in less than 100% relative humidity if the relative humidity is high enough.

For the purposes of the workshop, assume that waste package temperatures follow the upper (red) curve of <u>Figure 6</u>. Further assume that some parts of the surfaces of some waste packages may be in contact with liquid water at temperatures up to 160°C and that the composition of the water may range from pure condensate to concentrated brine. Contact between liquid water and waste package surfaces may be episodic, particularly at temperatures above 96°C. Anions in the water would include those in the table on page 2 but not necessarily in the same ratios.

For purposes of the workshop, assume that the bulk gas phase composition in the tunnels after closure would be as follows: When temperatures are at or below 96°C, the composition of the gas phase would be essentially that of atmospheric air that has a relative humidity in the range of 90-100% and slightly elevated carbon dioxide content. Above 96°C, equilibrium considerations would indicate that the gas phase is essentially entirely steam, with only traces of air. However, the mountain is permeable, and convective forces due to heating could lead to gases (air) from another part of the mountain flowing to the repository area. Another complicating factor is that repository tunnels, although sealed from the surface, would remain connected to each other. The differences in temperatures in and between tunnels would cause convective flow in and between tunnels. (Backfilling the tunnels just before closure would significantly reduce convective flow, but the insulating effect from the backfill would lead to unacceptably high waste package temperatures, so no backfill is planned for the repository.)

Additional Information

The term "barrier" refers to any repository element that helps to protect human health and safety by reducing or retarding the flow of radionuclides from the repository to the human environment. There would be many natural and engineered barriers in a repository at Yucca Mountain. Besides the Alloy 22 outer shell of the waste package, other engineered barriers that depend on the phenomenon of passivity include the titanium drip shield, the 316NG stainless

steel inner shell of the waste package, and the zirconium cladding that encases spent-fuel pellets. Because of the short duration of the Board's workshop, only the long-term extrapolation of the corrosion resistance of the Alloy 22 outer shell will be addressed. The expectation, however, is that many of the observations at the workshop will apply in a generic sense to any metals that depend on passivity for corrosion resistance.

Recently, DOE commissioned a peer review of the technical basis for predicting long-term behavior of the waste package. The peer review is being conducted by a panel of seven materials scientists. The panel began its work in May and plans to issue an interim report in September and a final report in February 2002. The DOE's peer review and the Board's workshop are completely independent (but we hope complementary) efforts. The Board's focus for this workshop is on a critical but narrow issue: the extrapolation of the body of knowledge consisting of short-term data (i.e., data from experiments with durations of a few years or less) and slightly longer-term experience (i.e., experience of relatively few decades with Alloy 22 or similar high-nickel alloys) over very long time periods (i.e., ten thousand years and longer).

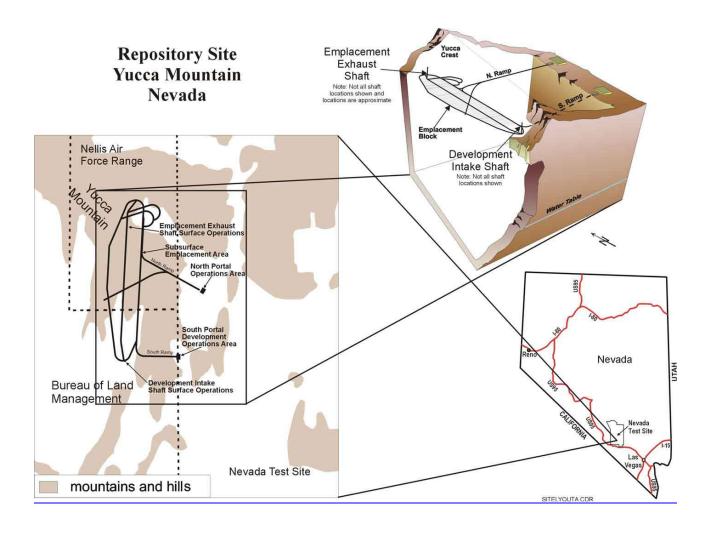
The DOE peer review panel's charge is much broader than the focus of the Board's workshop. For example, the DOE panel will examine stress corrosion cracking issues. Such issues are not in the scope of the Board's workshop. Presumably the DOE panel also will review the data from the LLNL test facility and from other DOE-sponsored corrosion-testing activities. The Board's workshop, however, will accept the data as given. Finally, the DOE panel will examine corrosion issues for the titanium drip shield; the Board's workshop will not.

The background information on repository and waste package design and on waste package environment that is in this document, together with background information on corrosion that that is in a separate document along with two questions to be addressed at the workshop, should be enough preparatory material for the purposes of the workshop. At least, that is our intent. We realize that some may wish additional information about the DOE program. To them we recommend a look at the Yucca Mountain Project Web site, http://www.ymp.gov/, where the documents listed in the next two paragraphs, and many more, can be found.

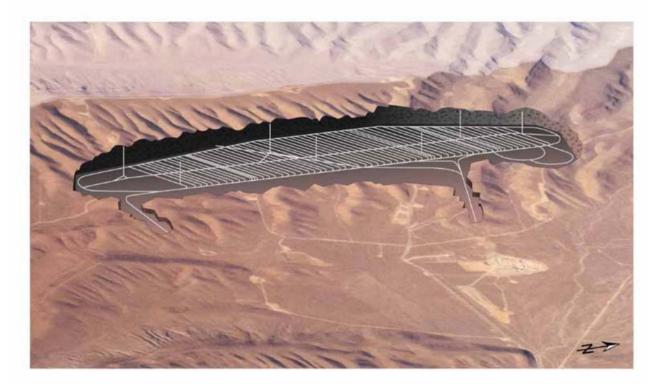
Figures 1-5 were taken from "Reference Design Description for a Geologic Repository" which is at the following DOE Web site: http://www.ymp.gov/documents/seda06m3 b/index.htm.

More information on the environment of the waste package is in the February 2000 document *Environment on the Surfaces of the Drip Shield and Waste Package Outer Barrier*, which can be found at the following Web site: http://www.ymp.gov/documents/amr/21844/21844.pdf.

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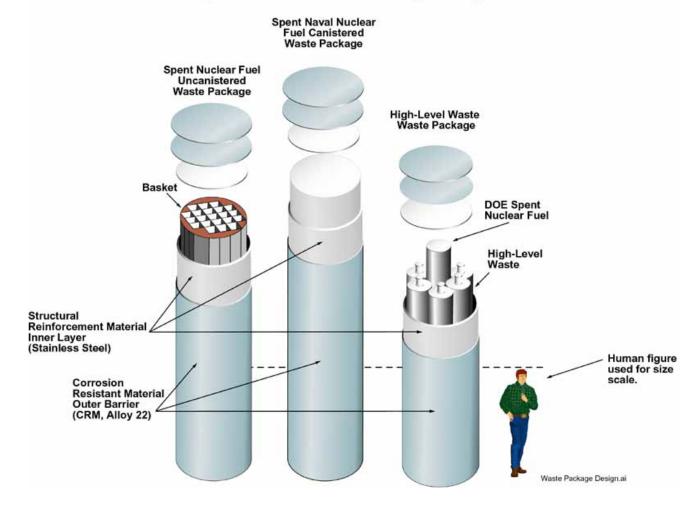
Subsurface Facility



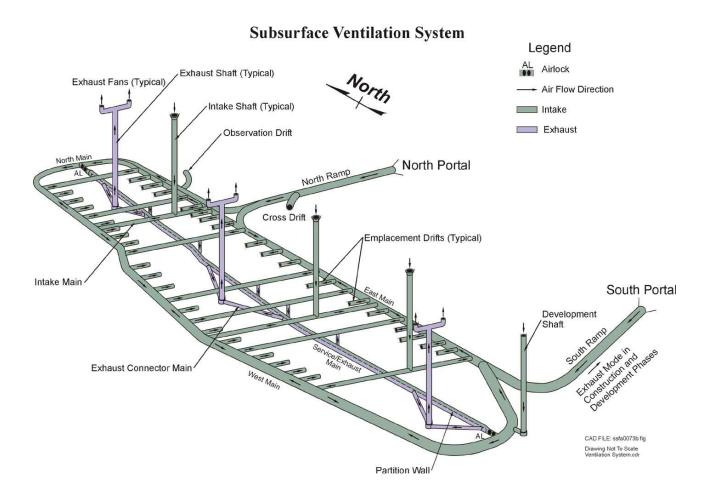
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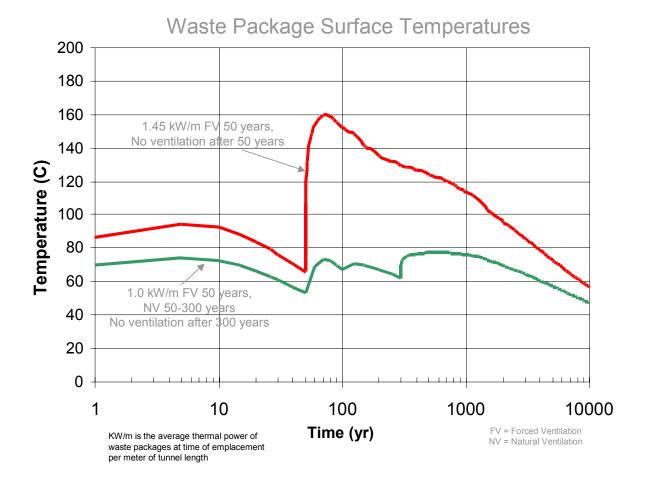
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Representative Waste Package Designs



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Engineered Barrier System

