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Review of Waste Package Verification Tests

Semiannual Report Covering the Period October 1983 - March 1984

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ABSTRACT

The current study is part of an ongoing task to specify tests that may be used to verify that engineered waste package/repository systems comply with NRC radionuclide containment and controlled release performance objectives. Work covered in this report includes crushed tuff packing material for use in a high level waste tuff repository. Ranges of repository conditions relevant to its testing and other factors important for its performance are discussed.

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EXECUTIVE SUMMARY

A review has been conducted of anticipated repository conditions relevant to crushed tuff packing material testing requirements. Current repository designs specify the location of the repository horizon in the unsaturated zone. Data previously obtained were concerned with emplacement of waste in the saturated zone and may no longer be fully relevant. Key parameters in test evaluations for packing material performance include temperature, radiation dose and dose rate, pressure, hydraulic properties of tuff packing, and steam/groundwater flow rates and chemistries.

Current estimates of groundwater flow rates from the surface of the repository site show that only 8 mm of water per year will approach the repository horizon. The high temperature and low pressure (~1 atmosphere) in the porous tuff will cause boiling of the water so that for several hundreds of years the emplaced waste packages will be subjected to an air/steam environment. Therefore, during this period, alteration of the packing by this gaseous medium needs to be predicted using appropriate test procedures. The type and magnitude of packing material alteration will in turn influence the rate of sorption and transport of radionuclides after the waste container has been breached. An important factor with respect to steam formation is that deposited salt remaining within the host rock will likely redissolve, to a greater or lesser extent, and give concentrated groundwater solutions when steam conditions subside. Packing material interactions with such concentrated groundwaters will determine surface alteration effects which must then be correlated with radionuclide sorption behavior for both steam and liquid water conditions.

Preliminary estimates for the gamma irradiation dose in the vicinity of a waste package emplaced in tuff lie in the range of 10^9 - 10^{10} rads. Since this dose will likely occur during the early steam/air period in the repository there is a potential for nitric acid to be formed due to the formation of nitrogen oxides by radiation, and their dissolution in the steam. No data appear to be available to quantify the effects of this acid on packing material performance or container corrosion.

With respect to anticipated pressures on the waste packages the hydrostatic stress will be negligible since they are emplaced in the unsaturated zone. Atmospheric pressures will prevail unless the host rock settles, in which case a lithostatic stress approximately 8.6 MPa is expected at the repository horizon.

Under the anticipated repository conditions described above alteration of crushed tuff/clay packing will occur. Some zeolites which will be present in tuff may be altered at temperatures as low as 95°C. However, recent studies show that tuff reacted with J-13 well water at 150°C causes no major changes in the principal phases but a $(Ca,Mg)CO_3$ secondary mineral will precipitate. This reaction is accompanied by decreases in Ca and Mg concentrations in the water.

1. INTRODUCTION

The NRC Rule for the Disposal of High Level Waste in Geologic Repositories (10 CFR 60) dated June 1983, specifies two main performance objectives for the engineered system:

a. Containment of HLW within the waste packages will be substantially complete for a period to be determined by the Commission taking into account the factors specified in subsection 60.113(b) (of 10 CFR 60) provided, that such period shall be not less than 300 years nor more than 1,000 years after permanent closure of the geologic repository; and

b. The release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure, or such other fraction of the inventory as may be approved or specified by the Commission; provided, that this requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit. The calculated total release rate limit shall be taken to be one part in 100,000 per year of the inventory of radioactive waste, originally emplaced in the underground facility, that remains after 1,000 years of radioactive decay.

In order to show compliance with these performance objectives the license applicant will need to provide a data base and analyses to quantify anticipated behavior of the waste package/repository system after permanent closure. This will necessarily involve research and testing programs to evaluate the likely modes by which engineered system components will degrade or fail by chemical or mechanical means. Knowledge of the ways in which the engineered barriers fail will permit estimates to be made regarding the containment capability of the waste package and the radionuclide release rate from the engineered system. Below is listed a logical sequence of events leading to loss of containment and the release of radionuclides:

a. Groundwater enters the engineered barrier system
b. Groundwater penetrates the geologic packing material (discrete backfill)
c. Groundwater penetrates the containment system
d. Groundwater leaches radionuclides from the waste form
e. Radionuclides are transported through the failed container system, packing material and disturbed host rock to the near field environment.

Figures 1.1 and 1.2 outline the failure/degradation modes which may occur during the sequence of events leading to loss of containment and the release of radionuclides. Chemical (corrosion) and mechanical failure modes are given for each of the barriers in the engineered system. The ones ultimately found to be applicable will depend on specific waste package/repository designs, temperatures, water chemistry and flow rate, design geometries and sizes, and materials selected, etc.

The objective of the current study is to specify the types of test that may be used to demonstrate compliance with the containment and radionuclide release performance objectives for engineered barrier systems. In particular it will be necessary to address how groundwater chemistry, pH and redox potential change as a result of high temperature reaction with package components and irradiation since these parameters influence the nature of container corrosion. After containment has failed, and radionuclides are leached from the waste form, the effects of radionuclide sorption by packing material, water chemistry, water flow rate, and temperature, etc. on the rate of radionuclide release from the engineered barrier system need to be quantified to determine whether regulatory criteria can be met.

Figure 1.1 serves as the basis for specifying containment verification tests for waste package components. Currently, borosilicate glass waste forms, carbon steel, titanium-based and Type 304L stainless steel containers, and bentonite-based, tuff-based, and crushed salt packing materials are emphasized for evaluation in basaltic, tuffaceous, and salt repositories. Initially, the tests that were described in this effort for verification of barrier performance assumed that the individual barriers (containers and packing material) are designed to meet the 300- to 1,000-year radionuclide containment requirement. The recommended tests specified to show compliance outlined the test variables to consider and the test methodology. Where appropriate, tests being developed by the Materials Characterization Center (MCC) were evaluated and any limitations specified. Future research to develop additional testing methodologies was also given.

In the current report, test conditions for evaluating the performance of crushed tuff packing material will be discussed. In the next Biannual Report recommendations will be made regarding tests that may be used to quantify crushed tuff packing under the test conditions specified.

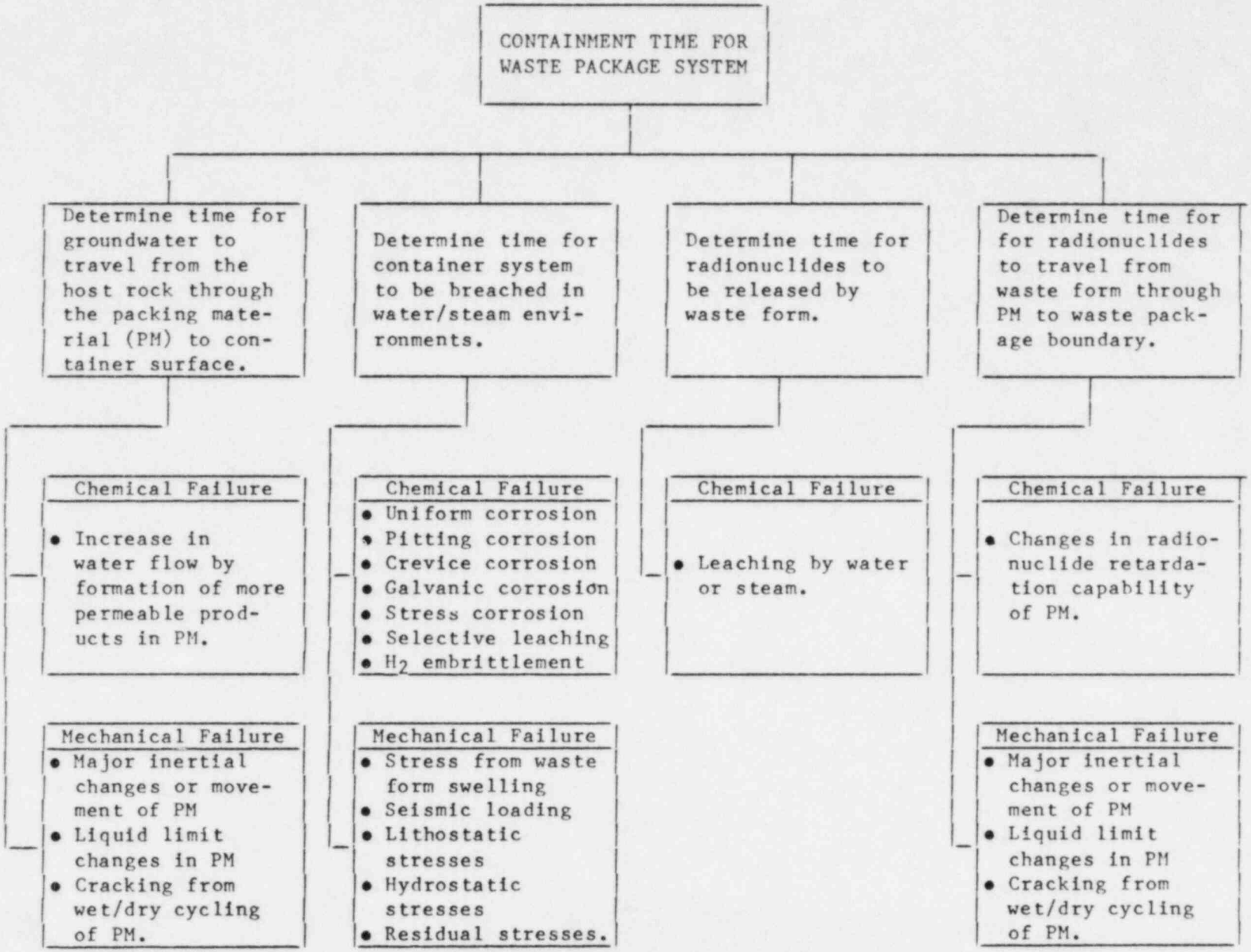

Figure 1.1 Chemical and mechanical failure/degradation modes affecting containment of radionuclides by the waste package system.

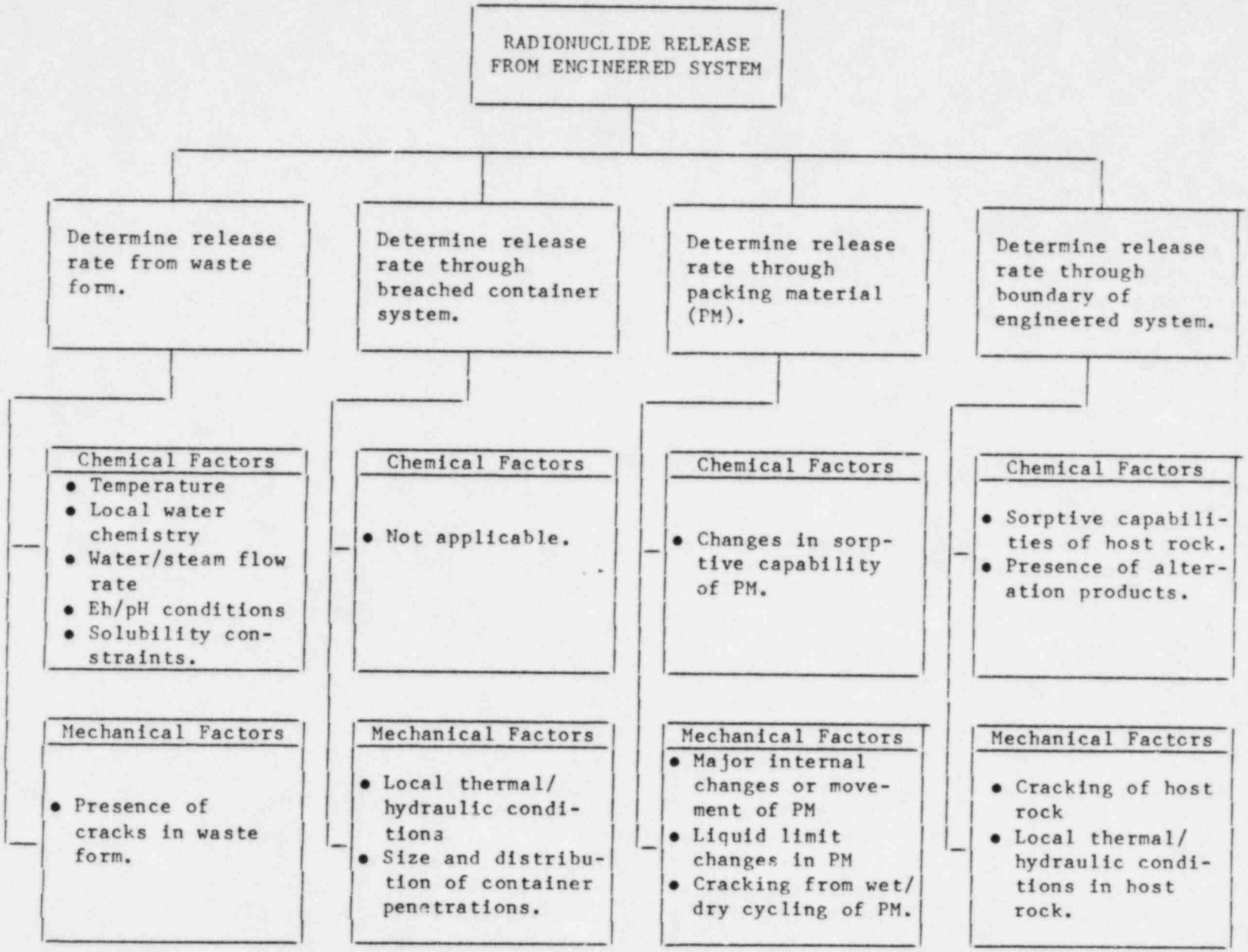

Figure 1.2 Factors affecting radionuclide release from the engineered repository system.

2. WASTE FORM TESTING REQUIREMENTS FOR DEMONSTRATION OF COMPLIANCE WITH THE CONTAINMENT AND CONTROLLED RELEASE CRITERIA

This part of the program has been completed and is reported in a prior Biannual Report entitled "Review of Waste Package Verification Tests" (NUREG/CR-3091, Vol. 2, BNL-NUREG-51630, 1983).

3. CARBON STEEL CONTAINER SYSTEM TESTING REQUIREMENTS FOR DEMONSTRATION OF COMPLIANCE WITH THE CONTAINMENT CRITERION

This part of the program has been completed and is reported in a prior Biannual Report entitled "Review of Waste Package Verification Tests" (NUREG/CR-3091, Vol. 3, BNL-NUREG-51630, 1983).

4. CRUSHED TUFF PACKING MATERIAL TESTING REQUIREMENTS TO DEMONSTRATE COMPLIANCE WITH THE RADIONUCLIDE CONTROLLED RELEASE CRITERION

The Yucca Mountain area in Nevada is the principal site for a HLW tuff repository. The geology, hydrology and geochemistry of this area have been described in prior BNL work (NUREG/CR-2937, 1983). The information on design and other characteristics of this repository is sparse compared to basalt and salt repositories. In early designs, the repository horizon was situated in the saturated zone. However, it was recently decided to place HLW packages in the unsaturated zone where container corrosion and waste form leaching is expected to be much lower. Specifically, the proposed repository horizon is located 400 m below the surface and 170 m above the static water table, which is in the lower portion of the densely welded (but highly fractured) Topopah Spring Member of the Paintbrush tuff formation and just above the basal vitrophyre layer (Bish, D. L., 1983). Therefore, some of the data obtained earlier for the saturated zone may no longer be relevant. In locating the repository in the unsaturated zone, DOE expects container corrosion and waste form leaching to be significantly lower.

The waste package design for tuff has yet to be finalized and the reference repository conditions have not been fully determined. According to presentations made at a recent NRC-NNWSI Workshop (Pescatore, C., 1983) the radionuclide release rates from a borosilicate glass waste form in an unsaturated tuff repository may be sufficiently low so that the packing material component can be eliminated from the waste package design. The reasons cited against the use of packing material are its low thermal conductivity, higher cost and post-emplacement problems. However, to meet the controlled release rate criterion given in 10 CFR 60, a spent fuel package may need packing material to reduce the radionuclide migration rate in one of the proposed designs. It is in this respect that packing material testing requirements are considered in the current work.

Although the composition of the packing material to be used with a spent fuel waste package has not been defined, presumably it will consist of crushed

tuff with a small fraction of binding material such as clay. Typically, this may be a mixture of crushed tuff and 15 percent or less of iron-bearing smectite clay consolidated to about 75 percent of theoretical density (UCID-19926, 1983). To evaluate the effect of clay addition, it will be useful to compare available results on tuff/clay compositions with those on crushed tuff alone. In an earlier report (NUREG/CR-3091, Vol. 2, 1983), packing material testing methods pertinent to a basalt repository environment have been discussed in detail. Due to some similarities in the properties of basalt and tuff, much of the discussion regarding basalt repository packages would also be applicable to the tuff repository waste package, although the test conditions may be different in the two cases. Therefore, the present report emphasizes the differences expected for packing material tests for tuff systems, and only outlines other pertinent tests which have been discussed in detail in earlier BNL work (NUREG/CR-3091, Vol. 2, 1983).

4.1 Repository Conditions Relevant to Packing Material Testing

The repository parameters which should be known to conduct tests on packing material are temperature, gamma radiation dose and dose rate, pressure, hydraulic properties and the groundwater chemistry. Some of the information regarding these parameters has been reviewed earlier (NUREG/CR-2482, Vol. 4, 1983) and is summarized in this section. New information made available since the publication of this review is also included.

4.1.1 Temperature

Thermal history is an important factor with respect to packing material performance since it will have a strong influence on the degree of mineralogic alteration, water content, radionuclide sorption, water chemistry and water pH and redox condition. The temperature profile across the packing material depends on the thermal conductivity of various components of the waste package/host rock system and the thermal loading of the package. The variation of temperature with time will depend on the thermal properties of the packing materials as well as the decay profile of various radionuclides in the waste. The most recent information from thermal analyses on a tuff repository is summarized in Table 4.1 for different waste package designs (Hockman, J. N., 1984). In each case the waste is assumed to have aged for ten years before burial. Note that only three spent fuel package designs include a packing material component. Time dependence of the temperature of various waste package components is shown in Figure 4.1. The packing material may be exposed to temperatures as high as the peak container temperature which in Figure 4.1 is 250°C (note that the data in Figure 4.1 are not calculated for the design which includes packing material). The peak waste form temperature for the waste package has been calculated in Table 4.1 assuming six fuel assemblies per container. Since this leads to temperatures higher than the maximum design value of 350°C, further calculations are in progress assuming four fuel assemblies per container (O'Neal, W. C., 1984). These results may reduce the temperature below the 250°C design limit for the packing material. Since the thermal conductivities of compacted basalt and clays are not very different, the addition of a small amount of clay as a binder will not significantly

Table 4.1. Significant input parameters and output results for the thermal analyses completed to date (Hockman, J. N., 1984).

	DHLW		CHLW		BWR			SF PWR			
Emplacement Mode	H	H	H	V	V	H	H	H	H	H	H
Package Power Level [W]	380	680	2210	2210	3420	3300	3300	3300	3300	3300	3300
Gross Thermal Loading [kW/acre]	50	50	50	50	50	50	50	50	50	50	50
Age at Burial [yr]	10	10	10	10	10	10	10	10	10	10	10
Canister Diam [cm], Material	61 SST	81 SST	32.4 SST	32.0 SST	57 SST	50 SST	50 SST	45 SST	50 SST	45 SST	45 SST
Overpack Diam [cm], Material	66 SST	86 SST	37.5 SST	---	---	---	---	---	---	---	---
Borehole Liner Diam [cm], Material	72.4 CS	92.4 CS	43.8 CS	---	---	62.2 CS	62.2 CS	51.4 CS	86.0 CS	86.4 CS	86.4 CS
Packing Outer Diam [cm], Material	---	---	---	---	---	---	---	---	82.4 CT	80 CT	80 CT
Borehole Diam [cm]	81.3	101.6	54	42.2	67.2	69	69	61	92.8	96.5	96.5
Borehole Pitch [m]	8.8	11.4	44	---	---	52	52	48.9	52.0	48.9	48.9
Package Pitch [m]	3.5	3.5	4.0	5.9	9.1	4.5	4.5	4.8	4.5	4.8	4.8
Drift Pitch [m]	---	---	---	30.5	30.5	---	---	---	---	---	---
Number of Internal Fins, Material	---	---	---	---	*	6 CS	12 CS	6 SST	24 CS	6 SST	---
Temperature limit [oC]	500	500	400	400	350	350	350	350	350	350	350
Peak Waste Temp [°C], Time [yr]	121 17.5	165 17.5	340 2.0	327 1.0	336 1.0	343 3.0	327 3.0	342 2.5	374 2.5	379 2.0	449 2.5
Waste Temp at 300 yr [°C]	62	73	112	92	101	153	144	148	156	155	185
Waste Temp at 1000 yr [°C]	44	50	62	60	62	109	105	109	112	111	129
Peak Borehole Temp [°C] Time [yr]	101 35	135 30	206 4.2	215 2.0	212 19.0	253 4.0	253 3.5	237 4.0	242 7.0	215 7.0	223 7.5
Peak Temperature ~1 m From Borehole Surface [°C]	96	127	145	103	130	175	175	161	178	158	167

*17 horizontal fins with the equivalent volume as could be found in the reference vertically finned canister.

Abbreviations:
H = Horizontal
SST = 304L Stainless Steel
AS = 20 Axisymmetric
IC = Infinite Cylinder
CT = Compressed Tuff
V = Vertical
CS = Carbon Steel

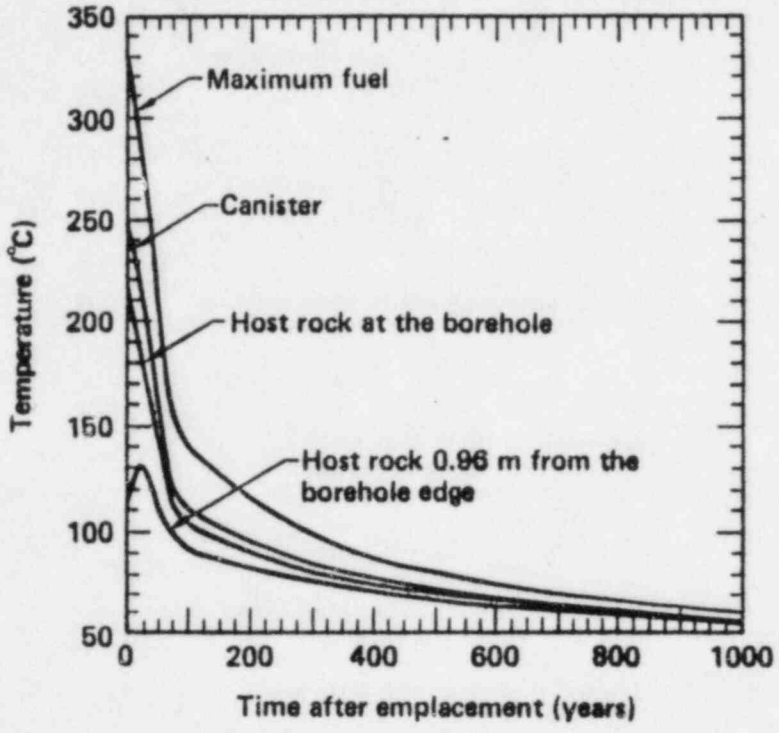

Figure 4.1. Temperature histories of waste package components and host rock for vertically emplaced BWR spent fuel (Hockman, J. N., 1984).

change the results of the thermal analysis (O'Neal, W. C., 1984). Other work supports this claim since well-dried bentonite with a density of 2.0 gm/cc has a thermal conductivity of 0.67 W/m°K. When wet the value rises to 0.75 W/m°K (ONWI-439, 1983).

4.1.2 Radiation

Radiation can change the desirable properties of a packing material either directly by structural damage and loss of crystallinity, or indirectly by changing the pH/Eh of the surrounding groundwater during radiolysis (PNL-3873, 1981). Figure 4.2 shows the radiation dose rate as a function of distance from a reference spent fuel container which has a thermal loading of 0.55 kW (DOE/NWTS-12, 1981). Similar plots for a CHLW package are approximately one order of magnitude higher. Note that the spent fuel package designs included in Table 4.1 have a package power level of 3.3 kW/canister which implies that if one of these designs is used, the dose rate may be considerably higher than shown in Figure 4.2. A cumulative dose of 10^9-10^{10} rads is expected before the fission product radionuclides have decayed to background levels. Since this dose will be administered during the early steam/air period in the repository, there is a potential for the formation of nitrogen oxides. These may interact with steam to form nitric acid (SAND81-1677, 1981). No data appear to exist to quantify the effects on packing material integrity or container corrosion.

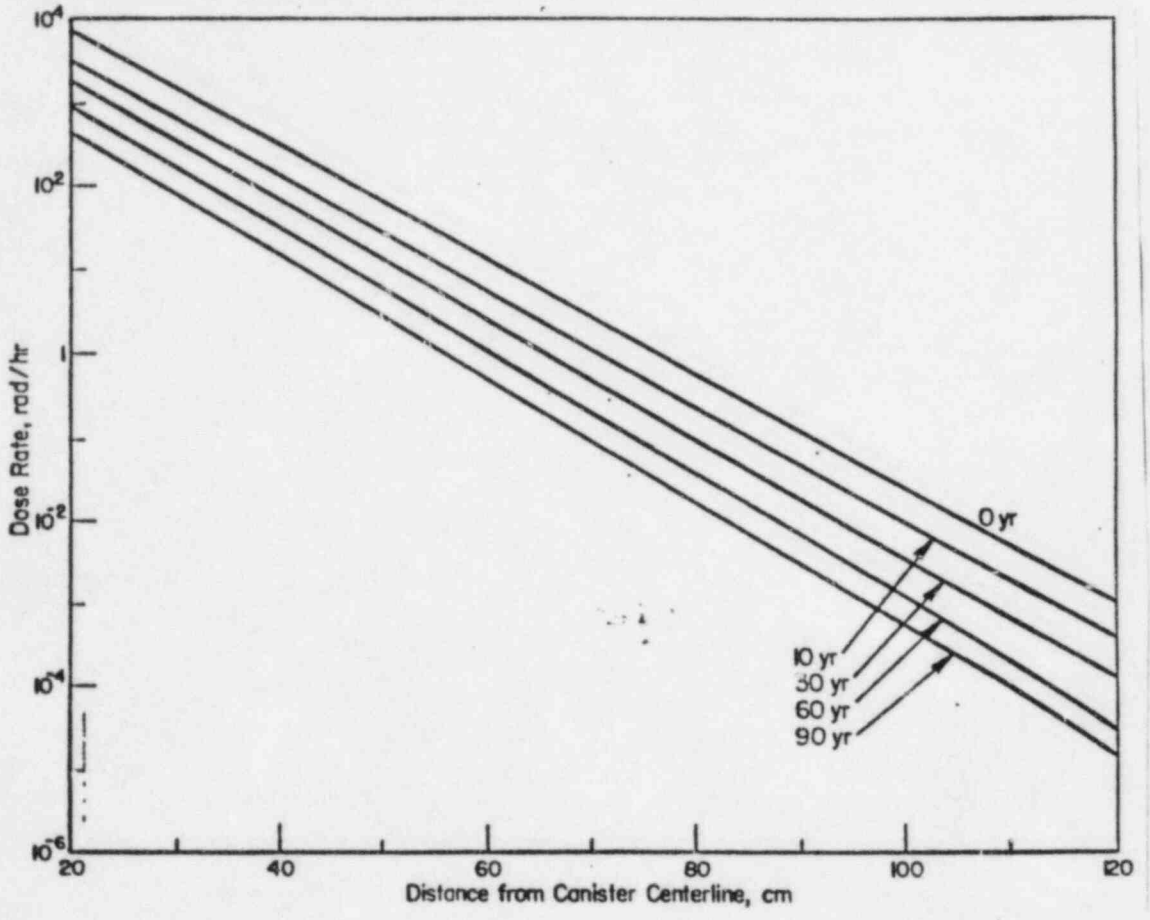

Figure 4.2. Dose rate in tuff from reference spent fuel canister (single PWR assembly 10 years out of the reactor; thermal loading of 0.55 kW/canister) as a function of distance into the rock and time after emplacement (DOE/NWTS-12, 1981).

4.1.3 Pressure

Since the presently proposed tuff repository site is in the unsaturated zone, there will not be any significant hydrostatic pressure on the waste package unless for some unexpected reason the repository is inundated after emplacement. In general, the pressure within the repository would remain at approximately one atmosphere. On the other hand, the repository may collapse and thus subject the packing material to lithostatic stresses. Assuming a bulk density of tuff (welded) of 2.2 g/cm^3 and repository level 400 m below the surface, a lithostatic pressure of 8.6 MPa will be acting on the waste package. When groundwater contacts the packing material, the clay constituent may swell and also exert pressure on the waste container. Nevertheless, the vertical component of the stress on the waste package should not exceed the lithostatic stress since this depends on the rock overburden load.

4.1.4 Hydraulic Properties

The hydraulic behavior within tuff deposits depends on its structure, composition, degree of welding, and the presence of cracks, faults, etc. through which rainwater migrates to aquifers. The matrix porosity of welded tuff is approximately 13 percent and the rock fracture density is 0.8-3.9 fracture/m (Dudley, W. W., 1982). Guzowski and others (NUREG/CR-2937, 1983) have compiled the available information on hydraulic properties in the saturated zone in the Yucca Mountain area. However, the amount of data for the unsaturated zone is very sparse. According to an estimate (Vieth, D. L., 1983), the annual rainfall at Yucca Mountain is 15 cm/yr and only five percent of this, i.e. 8 mm/yr, percolates down through the unsaturated zone to the water table. Until the repository temperature falls below ~95°C, which is the boiling point of water at the repository horizon, this rainwater is expected to fill the repository environment with a mixture of steam and air.

In a recent draft report (813-1179, D278/A, Task 4, 1983) Golder Associates and others have suggested a way to improve the function of the packing component by taking advantage of the difference in capillary forces acting between the pores of packing material and host rock. A complex design with layers of packing material of different pore size has been proposed. At present, however, it is not clear how much benefit can be derived from capillary forces since they could concentrate water around the waste rather than allow it to dissipate (P. Soo, 1983). At this stage it will be useful to first demonstrate by analytical modeling that capillary forces can play an important role even if the pore size is hard to control (such as due to material inhomogeneity, cracking, etc.). Then, hydraulic conductivity and other tests may be conducted to support the conclusions.

4.1.5 Groundwater Chemistry

In order to predict waste package performance over extended time periods it is necessary to fully characterize the chemical composition, pH and redox state of groundwater and ascertain how these parameters change as a result of repository excavation and waste emplacement. High temperature and radiation

environments will obviously accelerate packing material interactions with steam and water and the effect of these interactions may control, to a large extent, the ability of the packing to sort radionuclides released from the waste.

There is a significant amount of information available on the chemical composition of groundwater in the Yucca Mountain area, but part of it may be unreliable because of contamination from the well drilling process (NUREG/CR-2937, 1983). A field filtration apparatus (Figure 4.3) has been used to collect groundwater while minimizing changes in composition due to atmospheric oxygen and drilling fluids (LA-9577-PR, 1983). It was found that there is a range of concentrations for various species, depending upon the location of the well and the depth of the water table. Table 4.2 gives the range of chemical compositions recently reported by Bish and others (1983) for groundwater from the vicinity of Yucca Mountain. The last column in this table gives the composition which has been designated as the reference compositon for tuff groundwater (LA-9328-MS, 1982). Generally, the upper limits in Table 4.2 were found for a well which was drilled deeper to intersect the Paleozoic rocks below the tuff. Note that the groundwater under consideration is oxic in nature with a measured redox potential range of -40 to +402 mV (SHE).

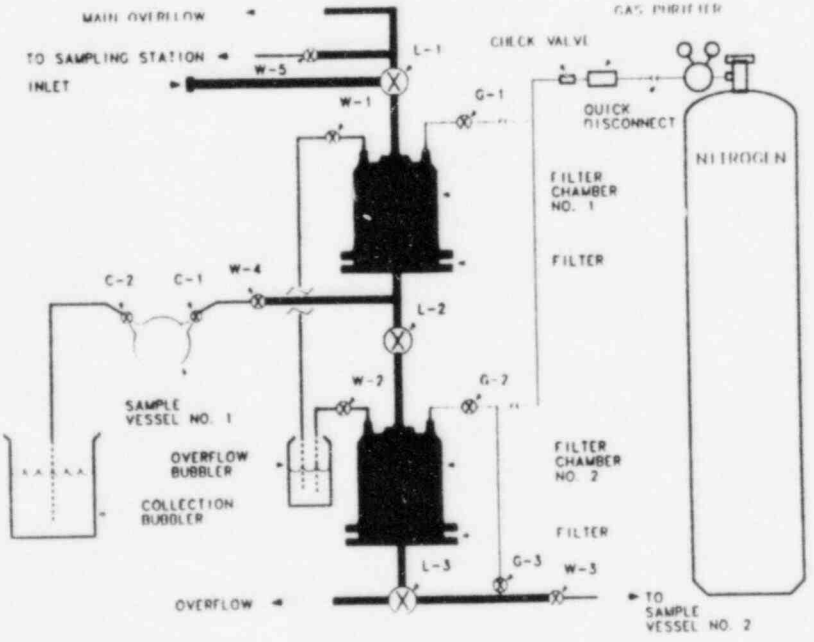

Figure 4.3. Field filtration apparatus (LA-9577-PR, 1983).

Table 4.2. Range of concentration of chemical species in filtered groundwaters from the vicinity of Yucca Mountain (Bish, D. L. and others, 1983; LA-9328-MS, 1982).

Specie	Range (mg/L)	Reference (mg/L)
Ca	1.6 - 87.8	14
Mg	0.05 - 31.9	2.1
Na	37 - 110	51
K	1.2 - 13.4	4.9
Li	0.06 - 0.72	0.05
Fe	0.01 - 0.93	0.04
Mn	0.004 - 0.43	----
Al	0.02 - 0.32	0.03
Si	19.1 - 31.8	31
Sr	-----	0.05
Ba	-----	0.003
F^-	0.6 - 4.5	2.2
Cl^-	5.5 - 37.0	7.5
SO_4^{-2}	14.6 - 129.0	22
PO_4^{-3}	-----	0.12
HCO_3^-	108 - 698	120
NO_3^-	<0.1 - 18.7	5.6
O_2	<0.1 - 7.1	----
Eh	-40 - +402 mV vs SHE	----
pH	6.7 - 7.7	7.1

The presently available information on the groundwater chemistry, such as given in Table 4.2, refers to samples taken below the water table (saturated zone). Since there is little information available on the chemistry of water reaching the waste package after percolating through the rock above, Table 4.2 can only give an idea of the water composition to be used in packing material testing. Table 4.3 lists various processes which will determine the composition of water coming in contact with the waste package. In this regard, Knauss and others (1983) have recently reported that the chemistry of vadose water changes very little when it comes in contact with the tuff rock. These tests were conducted at 90°C and 150°C using surface outcrop material (at Fran Ridge) washed in groundwater to remove soluble salts.

An important feature of the proposed tuff repository is that any water in the vicinity of the repository will boil at 95°C leaving behind previously dissolved salts. At a later time new groundwater will redissolve these salts so that groundwater reaching the waste package may have ionic species at their saturation limits. Therefore, some of the testing should be performed in groundwater containing high concentrations of the salts to be expected under near field conditions.

Table 4.3. Processes important to groundwater chemistry at Yucca Mountain and NTS (NUREG/CR-2937, 1983).

Process	Dissolved Constituent							
	Na^+	K^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	Cl^-	HCO_3^-	SiO_2
Dissolution of:								
vitric tuff	++	0	+	+		++	++	++
devitrified tuff	+	0	++	++		?	++	+
sulfates			++		++			
carbonates			++	++			++	
pyrite					++			
Precipitation of:								
montmorillonite		-	--	--				--
clinoptilolite	-	-	--	--				--
flourite			--					
calcite			--				--	
silica gel								--
Dissociation of carbonic acid							++	
Ion-exchange or fixation onto secondary minerals	+	--	--	--				
Evapotranspiration	++	++	++	++	++	++	++	++

Key: (++) = major source for this species
(+) = minor source for this species
(0) = element is retained in solid phase during this process
(?) = variable effect
(--) = sink for this species
(-) = minor sink.

4.1.6 Wet/Dry Cycling of Packing Material

Finally, due to intermittent water flow in a tuff repository, wet-dry cycling of the packing material is a distinct possibility. As discussed with reference to basalt packing material (NUREG/CR-3091, Vol. 2, 1983), the wet-dry cycling can cause cracking and fracturing of clay-containing packing material as well as some irreversible structural changes. It must be demonstrated that permeability and sorption behavior of the packing, after the early high-temperature steam/air period, does not compromise the ability of the waste package to meet the NRC controlled radionuclide release criterion.

In the next Biannual Report an evaluation of testing procedures to quantify radionuclide release for steam and aqueous phases will be conducted. The usefulness of these procedures and suggestions for additional work will be specified.

4.2 Geochemistry and Water/Rock Interactions

Mineral composition, compaction density/pressure, water content, hydraulic conductivity, thermal conductivity, swelling pressure, thermal and radiation stability, and sorption of radionuclides are the properties of a packing material which should be known to evaluate its performance. Since the reference packing material for a tuff repository has not been formally defined, a complete evaluation of these properties is not possible at this stage. However, the most likely packing material appears to be the crushed tuff excavated during the mining of the repository. Due to a significant variation in the composition of tuff as a function of depth, it is important to know which specific tuff composition will be used for packing. In the event that a specific tuff composition cannot be defined - due to considerable variability present in individual units of tuff - compositions must be at least bounded in order to test under a given range of compositional characteristics. A bounded range of compositions can be adapted for corrosion and leaching tests. Equally important is the amount and type of any clay or other binder which will be added to the crushed tuff. If tuff from the reference horizon is used, the packing material will predominantly comprise tridymite, quartz, cristobalite, alkali feldspars and a small amount of smectite clay (Bish, D. L., 1983). Zeolites which have useful sorption properties and glassy phases will be usually absent. The mineralogic composition, based on studies to date, does not vary greatly (except for smectite which varies in amount from 1 to 6%) within the exploration block evaluated. However, if future work shows that there are likely to be significant compositional differences in the crushed tuff packing material it will be necessary to determine how these will affect radionuclide sorption/desorption behavior. A recent study clearly shows that individual radionuclides are sorbed and eluted at different rates depending on the tuff composition (LA-9329, 1982). Therefore, accurate estimates of radionuclide migration rates through anticipated crushed tuff packing materials will be needed to demonstrate that the engineered barrier system will meet the NRC controlled release criterion.

The use of tuff from the repository horizon in packing has the advantage of being compatible with the host rock and no synergistic effects should be expected at the waste package/host rock boundary. However, to take full advantage of the packing component in improving sorption properties, addition of other minerals may be desirable. Addition of charcoal, bentonite, CaO and/or MgO has been mentioned in the literature for this purpose.

As discussed in an earlier report, tuff is likely to undergo alteration at high temperature (NUREG/CR-2482, Vol. 5, 1984). Since a crushed tuff packing material will be subjected to temperatures as high as 250°C for extended periods, the nature of any changes during service need to be addressed. These include changes in water content, porosity, volume, strength, permeability, radionuclide sorption, as well as changes in mineralogy. Loss of water during heating will likely alter clays, zeolites and glasses to phases of smaller volume. For example, clinoptilolite (a zeolite) may be altered at temperatures as low as 95°C (LA-8612-PR, 1981). Mordenite, also a zeolite, will alter at temperatures in the range 300 to 400°C in the presence of water at 400 bars pressure (LA-9328-MS, 1982). Zeolites and clays, themselves, may be formed from feldspars and silica on addition of water (rehydration).

In recent work, Oversby (1983) and Knauss and others (1983) have examined mineralogic changes in crushed tuff during reaction with J-13 well water at 150°C. Although there was little change in the major phases at this temperature, a significant amount of (Ca, Mg)CO_3 was formed as a precipitated secondary mineral. This was associated with decreases in Mg, Ca, and CO_3+HCO_3 concentrations in the water. Presumably, this calcite phase will modify the radionuclide sorption behavior on crushed tuff packing and long term sorption tests may be required to assess the magnitude of this effect.

Preliminary analysis shows that there is a significant (not specifically quantified) reduction in tensile and compressive strength of Topopah Spring tuffs after 2 to 6 month exposure to water under conditions simulating those anticipated in the near field environment (LA-9328-MS, 1983). These changes were detected even though changes in tuff mineralogy were subtle and thought to be connected with grain boundary and fracture surface modification by water reaction.

One of the primary purposes of using a packing material is to retard the migration of radionuclides from the waste package by sorption, diffusion control or ion exchange. A large amount of data was collected on the sorption properties of various radionuclides by tuffaceous materials (LA-9328-MS, 1982; NUREG/CR-2937, 1983). Many of these experiments were performed on crushed samples which are analogous to the packing material. Some information is also available on the effect of particle size on sorption coefficients (LA-9225-PR, 1982) and rock-to-water ratio (Knauss, K. G., 1983). Once a reference compositon for the packing material has been established, the presently available data will become useful in specifying the test conditions. In this respect, it is important to note that sorption properties strongly depend on the mineral composition of the tuff as well as the concentration and composition of

the ionic species under consideration. In other words, testing will be necessary at least on different classes of radionuclides which include alkali and alkaline earth elements, plutonium, americium and other rare earths (actinides and lanthanides), and various anionic complexes (NUREG/CR-2482, Vol. 5, 1984).

An important concern, however, centers on the possibility that, after the container has been breached, steam/air conditions may still be present. No data appear to be available to quantify radionuclide release behavior in the presence of such an environment, in terms of either the release of radionuclides by steam interactions with the waste form or the transport of radionuclides by steam through packing materials. Such information will be needed to quantify the controlled release rate of radionuclides in the early post-containment period.

The presence of radiation can cause structural changes in the packing material, which may have an adverse effect on its function. However, the presently available information as discussed earlier (NUREG/CR-2482. Vol. 4, 1983) suggests only minor damage to silicate and aluminosilicate materials by high doses of gamma radiation. It is not clear whether the presence of high temperature will reduce the radiation damage by annealing, or will further deteriorate the desirable properties of the packing material. In either case direct radiation damage is not expected to affect the packing performance significantly, and probably more important effects of radiation will be through the changes in solution chemistry during radiolysis of groundwater or the water contained in the pores of the packing material.

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5. COMPONENT INTERACTIONS AND WHOLE PACKAGE TESTING REQUIREMENTS

This part of the program has been completed and is reported in a prior Biannual Report entitled "Review of Waste Package Verification Tests" (NUREG/CR-3091, Vol. 3, BNL-NUREG-51630, 1983).

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13 ABSTRACT (200 words or less)

The current study is part of an ongoing task to specify tests that may be used to verify that engineered waste package/repository systems comply with NRC radionuclide containment and controlled release performance objectives. Work covered in this report includes crushed tuff packing material for use in a high level waste tuff repository. Ranges of repository conditions relevant to its testing and other factors important for its performance are discussed.

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