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Reference Waste Forms and Packing Material for the Nevada Nuclear Waste Storage Investigations Project

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Abstract

The Lawrence Livermore National Laboratory (LLNL), Livermore, Calif., has been given the task of designing and verifying the performance of waste packages for the Nevada Nuclear Waste Storage Investigations (NNWSI) Project. NNWSI is studying the suitability of the tuffaceous rocks at Yucca Mountain, Nevada Test Site, for the potential construction of a high-level nuclear waste repository. This report gives a summary description of the three waste forms for which LLNL is designing waste packages: spent fuel, either as intact assemblies or as consolidated fuel pins, reprocessed commercial high-level waste in the form of borosilicate glass, and reprocessed defense high-level waste from the Defense Waste Processing Facility in Aiken, S.C. Reference packing material for use with the alternative waste package design for spent fuel is also described.

Introduction

The Nevada Nuclear Waste Storage Investigations (NNWSI) Project is studying the suitability of the tuffaceous rocks at Yucca Mountain, Nevada Test Site, for the potential construction of a high-level nuclear waste repository. Lawrence Livermore National Laboratory (LLNL), Livermore, Calif., has been given the task of designing and verifying the performance of waste packages for the NNWSI Project.

The waste forms for which waste packages are being designed are spent fuel, either as intact assemblies or as consolidated fuel pins, reprocessed commercial high-level waste in the form of borosilicate glass, and reprocessed defense high-level waste from the Defense Waste Processing Facility (DWPF), Aiken, S. C. The purpose of this document is to provide a summary description of

these three waste forms. The borosilicate glass that will be produced at West Valley, N.Y., and the reference packing material that is under consideration for use with spent fuel waste packages are also described.

The reference spent fuel for NNWSI waste form testing is pressurized water reactor (PWR) fuel of moderate burnup and low fission gas release from the Turkey Point and H. B. Robinson reactors. The reference waste form for Defense High-Level Waste is borosilicate glass based on Frit 165. The Commercial High-Level Waste testing materials are based on a borosilicate glass formulation PNL 76-68. The reference packing material is Topopah Spring from Yucca Mountain, Nev., crushed to pass through 100-mesh screen and compressed isostatically at 20,000 psi.

Spent Fuel

Spent fuel, for the purposes of this report, is fuel material contained in cladding that has been irradiated in a light water commercial power reactor (LWR). All fuel that was discharged from LW

reactors prior to 1969 was reprocessed. Table 1 gives the accumulated spent fuel population at the end of 1982 as a function of discharge date and burnup. The total accumulation at the end of 1982

Table 1. Burnup and time of discharge of commercial spent fuel in inventory.^a

Year	Burnup (MWd/MTIHM) ^b															
	0-5,000		5,000-10,000		10,000-15,000		15,000-20,000		20,000-25,000		25,000-30,000		30,000-35,000		35,000-40,000	
	BWR	PWR	BWR	PWR	BWR	PWR	BWR	PWR	BWR	PWR	BWR	PWR	BWR	PWR	BWR	PWR
1969			0.2		0.9		7.1		0.2		7.8					
1970							40.5			7.8						
1971			13.9		5.5	4.6						40.0				
1972	141.6		11.8		31.4	7.8	4.0	45.0				9.2		21.7		
1973	9.1		16.3		35.6		40.1	24.2				39.7				
1974	59.9		6.2	7.1	121.5	77.0	31.0	39.6	3.8	13.9		74.0				
1975			0.7	9.3	45.7	186.3	155.2	11.9	21.9	70.8		41.2		19.8		
1976	0.1		49.4		108.0	16.5	149.3	147.0		83.8		91.3		37.2		
1977			52.1		34.2		174.2	131.8	102.2	126.4		134.1		85.9		18.4
1978				1.8	24.5	51.1	118.8	78.3	266.4	45.3	28.4	401.5		72.5		62.1
1979					19.6	26.0	91.6	123.8	161.3	55.3	178.2	278.5		238.9		31.6
1980	15.4				2.7	9.0	45.7		352.4	55.3	110.0	303.7		226.0		28.8
1981						25.5	73.2		255.9		161.0	303.9		428.1		16.0
1982					14.5		40.8	79.5	30.0	88.7	300.0	161.4		360.6		13.2

^a From DOE/NE-0017/2 (1983).

^b MTIHM = metric tons initial heavy metal.

was 3732.5 MTIHM (metric tons initial heavy metal) of boiling water reactor (BWR) spent fuel and 5256.1 MTIHM of PWR spent fuel.

Figure 1 shows a typical fuel pin design used in a PWR and Fig. 2 is a typical PWR assembly.

Figure 3 represents a fuel pin and assembly geometry for a typical BWR. Assembly sizes, fuel pin dimensions, and cladding type vary with reactor design. Table 2 lists typical characteristics; for more detailed discussions consult Woodley (1983).

Figure 1. Westinghouse PWR fuel rod. Specific dimensions depend on design variables, such as prepressurization, power history, and discharge burnup (Woodley, 1983). Figure provided by Hanford Engineering Development Laboratory (HEDL), Richland, Wash., Report HEDL-TME 83-28.

Figure 2. Combustion Engineering, Inc. PWR fuel assembly (Woodley, 1983). Figure provided by Hanford Engineering Development Laboratory (HEDL), Richland, Wash., Report HEDL-TME 83-28.

Most LWR fuel pins consist of a stack of UO_2 pellets enclosed in Zircaloy cladding. Three PWRs and one BWR use fuel pins with stainless steel (SS) cladding. Spent fuel from these reactors represents about 3% of the total inventory. The initial internal atmosphere of the fuel pins is helium at 1 atm (up to 1977) or 3 atm (1977 on) pressure in BWR pins and 1 to 30 atm pressure in PWR pins (Woodley, 1983).

Loss of cladding integrity occurs for a small percentage of fuel pins during irradiation. Figure 4 shows estimates of the failure rate for PWR and BWR pins as a function of time (Garzarolli et al., 1979). The failure rate for current and future fuel pins during in-reactor service seems likely to continue at approximately 0.01% of the pins (Woodley, 1983). Typical cladding defects are small cracks or pin holes.

Figure 3. General Electric Company BWR fuel assembly (Woodley, 1983). Figure provided by Hanford Engineering Development Laboratory (HEDL), Richland, Wash., Report HEDL-TME 83-28.

During irradiation some of the original uranium in the fuel is transformed into fission products or higher actinides. The exact inventory of these nuclides at the end of irradiation depends on the reactor operating characteristics and the

duration of irradiation. Estimates of the isotopic and chemical composition of spent fuel have been made with the ORIGEN2 code (DOE/NE-0017/2, 1983). These estimates were presented for the total inventory accumulation of spent fuel.

Table 2. Physical characteristics of LWR fuel assemblies.^{a,b}

Characteristics	BWR	PWR
Overall assembly length	4.470 m	4.059 m
Cross section	13.9 x 13.9 cm	21.4 x 21.4 cm
Fuel element length	4.064 m	3.851 m
Active fuel height	3.759 m	3.658 m
Fuel element (outer diameter)	1.252 cm	0.950 cm
Fuel element array	8 x 8	17 x 17
Fuel elements/assembly	63	264
Assembly total weight	319.9 kg	657.9 kg
Uranium/assembly	183.3 kg	461.4 kg
UO ₂ /assembly	208.0 kg	523.4 kg
Zircaloy/assembly	99.5 kg ^c	108.4 ^d
Hardware/assembly	12.4 kg ^e	26.1 kg ^f
Total metal/assembly	111.9 kg	134.5 kg
Nominal volume/assembly	0.0864 m ^{3k}	0.186 m ^{3k}

^a From DOE/NE-0017/2.^b Characteristics are based on 1976-1977 General Electric design data and 1975-1977 Westinghouse design data.^c Includes Zircaloy fuel-element spacers and fuel channel.^d Includes Zircaloy control-rod guide thimbles.^e Includes SS tie-plates, Inconel springs, and plenum springs.^f Includes SS nozzles and Inconel-718 grids.^k Based on overall outside dimension.

Table 3 gives a selection of data from DOE/NE-0017/2 (1983) recalculated to a basis of per MTIHM. The basis for selection of fission products and activation products was half-life longer than 1 yr, and a selection based on activity per MTIHM, which varied according to the half life. The fission products and activation products were divided into three groups:

Group I—Half-life between 1 and 5 yr;
activity greater than 0.5 Ci/MTIHM

Group II—Half-life between 5 and 31 yr;
activity greater than 0.3 Ci/MTIHM

Group III—Half-life over 31 yr;
activity greater than 0.01 Ci/MTIHM

Table 3 also includes three short-lived nuclides that are in secular equilibrium with longer lived parents and that contribute substantially to the total activity of the spent fuel.

Assuming 10-yr-old fuel as a reference case, nuclides with half-lives of less than 1 yr will have decayed to less than 0.1% of their original (at discharge) activities and will constitute only a small

proportion of the activity in the spent fuel. Group I and Group II nuclides are the main contributors to thermal power generation and radiation field during the containment period, up to 300 to 1000 yr after disposal. At 300 yr after disposal, Group I and II nuclides have decayed to less than 0.1% of their original activities. Decay heat generation and radioactivity in the post-300-yr period will be dominated by Group III fission products and actinides. Table 4 gives the dominant actinides in spent fuel recalculated to a basis of per MTIHM. It should be emphasized that the data in Tables 3 and 4 do not represent any individual spent fuel material but are the average of the present inventory calculated by dividing total inventory estimates by total inventory weight of initial heavy metal.

Figure 5 plots the radioactivity and thermal power of PWR and BWR spent fuel as a function of burnup and decay time since discharge. The middle curve, for 25,000 MWd/MTIHM burnup, is the most representative of the current spent fuel inventory average.

The distribution of radionuclides within spent fuel is of interest when behavior of the fuel under geologic disposal conditions is considered. Some of the fission products and most of the actinides are present as oxides dissolved in the

Figure 4. Estimates of the failure rate for (a) fuel rods in PWR plants and (b) fuel rods in BWR plants. (From Garzarolli et al., 1979.)

Table 3. Inventory of dominant fission products and activation products in spent fuel, cladding, and fuel assembly structural material with half-lives of 1 yr or longer.^a

Isotope	Half life (yr)	g/MTIHM	Ci/MTIHM
Group I			
Fe-55	2.7	0.91	2.27×10^1
Ru-106	1.01	27.4	9.17×10^4
[Rh-106]	[29.8s]	2.6×10^{-5}	9.17×10^4
Sb-125	2.7	6.12	6.32×10^1
Cs-134	2.06	29.9	3.88×10^4
Pm-147	2.62	54	5.01×10^4
Eu-155	4.9	6.0	2.79×10^1
Group II			
H-3	12.33	0.054	516
Co-60	5.27	3.28	3.72×10^1
Kr-65	10.7	13.6	5.32×10^1
Sr-90	28.8	346	4.73×10^4
[Y-90]	[64 h]	0.09	4.73×10^4
Cd-113m	14	0.15	33.3
Cs-137	30.17	780	6.79×10^2
[Ba-137m]	[2.55m]	1.2×10^{-4}	6.42×10^4
Eu-152	13.6	0.022	3.8
Eu-154	8.5	18.9	5.09×10^1
Group III			
C-14	5730	0.33	1.47
Ni-59	7.5×10^4	42.9	3.25
Ni-63	100	7.1	438
Se-79	6.5×10^4	4.1	0.29
Zr-93	1.5×10^6	592	1.49
Nb-94	2.0×10^4	4.37	0.82
Tc-99	2.14×10^5	556	9.44
Pd-107	6.5×10^6	156	0.08
Sn-121	55	0.017	1.02
Sn-126	$\sim 1 \times 10^5$	19.5	0.55
I-129	1.6×10^7	128	0.023
Cs-135	3×10^6	218	0.25
Sr-151	90	11.0	288

^a Calculated from data in DOE/NE-0017/2 (1983); half-lives from Friedlander et al. (1981).

UO₂. Some fission products form oxides that are insoluble in UO₂ or remain as metals; these materials tend to form small-scale segregations in the UO₂ matrix. Some fission products form nonoxide compounds either with the fuel or other fission-product elements, e.g., CsI. Finally, some fission products are present as inert gases.

During irradiation, particularly while temperature is changing rapidly, or at high fuel temperatures (>1000°C), gaseous fission products may diffuse out of the UO₂ matrix and collect in the gap between the pellets and the cladding. If

the cladding subsequently fails, any gas in the gap can easily escape. Figure 6 shows estimates of fission gas release from the pellet matrix to the pellet-cladding gap. Gas release is much lower in the newer type pressurized fuel pins than in the older, unpressurized pins. Volatile fission products behave in a manner similar to the gaseous fission products if a high gas release occurs (Woodley, 1983).

For fuel pins operated under conditions that do not lead to high gas release, which includes most of the pins presently in use, fission products

(a)

(b)

Figure 5. Radioactivity and thermal power of 1 metric ton of heavy metal of (a) PWR and (b) BWR spent fuel as a function of burnup and time from reactor discharge. (From DOE/NE-0017, 2, 1 83.)

Table 4. Inventory average at the end of 1982 of dominant actinides in spent fuel.^a

isotope	Half-life (yr)	g/MTIHM	Ci/MTIHM
U-234	2.45×10^5	166	1.04
U-235	7.04×10^8	9.39×10^3	0.02
U-236	2.34×10^7	2.84×10^3	0.18
U-238	4.47×10^9	9.57×10^6	0.32
Np-237	2.14×10^6	276	0.20
Th-238	87.74	77.6	1.34×10^3
Pu-239	2.41×10^4	4.76×10^3	296
Pu-240	6.57×10^3	1.74×10^3	395
Pu-241	14.1	816	8.42×10^4
Pu-242	3.76×10^5	305	1.16
Am-241	433	170	584
Am-242m	152	0.44	4.29
Am-243	7.37×10^3	55.3	11.01
Cm-242	0.45	0.85	2.80×10^3
Cm-243	28.5	0.19	9.77
Cm-244	18.1	14.0	1.74×10^3
Cm-245	8.5×10^3	0.57	0.10
Cm-246	4.7×10^3	0.07	0.02

^a Calculated from data in DOE/NE-0017/2 (1983); half-lives from Friedlander et al. (1981).

are uniformly distributed in the UO₂ matrix. Figure 7 shows the distribution of Pu, Cs, Te, and I in a fuel pellet with a burnup of 28,000 MWd/MTIHM. The fission products are uniform within the pellet while the Pu concentration is enhanced near the pellet surface. This variation in Pu as a function of position is due to enhanced production of actinides near the pellet surface (Woodley, 1983).

Spent fuel waste testing for NNWSI was focused at first on Turkey Point spent fuel, which had an initial enrichment of 2.56 wt% ²³⁵U, was burned to 25,500 to 27,000 MWd/MTIHM, and has an average fission gas release to the cladding gap of 0.3%. The spent fuel used in current experiments was discharged from Turkey Point on Nov. 25, 1975 (Wilson, 1983) and is representative of modern design fuel pins operated to moderate burnup under reactor conditions that produced

limited fission gas migration from the fuel matrix to the cladding gap.

Future spent fuel testing work will use H. B. Robinson PWR fuel pin samples in addition to the Turkey Point samples. Characteristics of the H. B. Robinson spent fuel are similar to those of the Turkey Point spent fuel (Barner, 1984). The desirability of including samples of spent fuel with higher burnup (>35,000MWd/MTIHM), high fission gas release, and of older pin design (nonpressurized pins) in the testing program will be evaluated in FY 85.

Spent fuel from BWRs may have different radioactive release characteristics than that from PWRs. The potential for differences will be determined by testing samples of moderate burnup BWR fuel. The specific BWR fuel to be used has not yet been chosen.

Figure 6. Comparison of fission gas release from (a) unpressurized and (b) pressurized LWR fuel rods (Woodley, 1983). Figure provided by Hanford Engineering Development Laboratory (HEDL), Richland, Wash., Report HEDL-TME 63-28.

Figure 7. Microprobe measured x-ray intensities for Plutonium, Cesium, Tellurium, and Iodine (Woodley, 1983). Figure provided by Hanford Engineering Development Laboratory (HEDL), Richland, Wash., Report HEDL-TME 83-23.

Defense High-Level Waste

The Nuclear Waste Policy Act of 1982 contains a requirement that not later than two years after the date of enactment of the Act (Jan. 7, 1983), the President shall evaluate the use of a commercial waste repository for disposal of high-level radioactive waste resulting from defense activities. Unless the President finds (on or before Jan. 7, 1985) that development of a separate repository for defense wastes is required, DOE is charged by the Act to arrange to dispose of Defense High-Level Waste (DHLW) in the civilian waste repository.

A Defense Waste Processing Facility (DWPF) is being constructed at the Savannah River Plant (SRP) in Aiken, S.C. This facility will solidify the waste presently held in storage tanks at SRP. The waste form will be borosilicate glass cast into 304L SS canisters (Baxter, 1983). Table 5 outlines the dimensions of the waste canisters. The volume of glass in the canister is approximately 85% of the available canister volume (Baxter, 1983). The canister will be sealed with an upset resistance weld, the sealing plug will be 5 in. in diameter and 0.5 in. thick (Baxter, 1983).

Table 5. Dimensions of DWPF waste canisters.^a

Overall length	9 ft 10 in. (300 cm)
Outside diameter	24 in. (61 cm)
Wall thickness	0.375 in. (0.95 cm)
Bow	0.25 in. (max) (0.64 cm)
Inside volume	26.1 ft ³ (739 l)
Weight, empty	1000 lb (454 kg)
Weight, full (Frit 131)	4260 lb (1932 kg)
Weight, full (Frit 165)	4660 lb (2114 kg)

^a From Baxter (1983).

Several frit compositions have been considered for the DWPF glass. Table 6 presents the chemical composition of four frit materials. Frit 131 is the current reference frit for DWPF design purposes; Frit 165 is a composition that more closely represents the expected frit composition for the final DWPF (J. Plodinec, Savannah River Laboratory (SRL), pers. comm.). Frits 18 and 21 were used earlier in the glass development process.

Waste stored at SRP is in the form of sludge that contains most of the radionuclides and supernatant material that contains radioactive cesium. The waste is stored in tanks, which vary in terms of the age of the waste contained and the chemical composition of the waste.

Tables 7-11 provide a profile of a sludge-supernatant representative glass made with Frit 131 and a blend of 5-yr-old sludge and 15-yr-old supernatant. The anticipated thermal neutron fissionable nuclide concentrations per canister for the reference case waste loading (in units of grams per canister) are U-233 = 0.002, U-235 = 635.7, Pu-239 = 139.2, and Pu-241 = 10.8 (Baxter, 1983).

All parameters in Tables 7-11 are nominal values that were calculated based on 5-yr-old sludge and 15-yr-old supernatant. Much of the waste at Savannah River Plant is substantially older than that. Preliminary estimates provided to us by Savannah River Laboratory indicate that at the start-up time of the DWPF, the waste canisters will have a thermal power of only about 75 W/can, and that approximately 3600 cans of this power level will be filled over the first 7 yr of DWPF operation (Bibler, SRL, pers. comm.). Subsequent canisters will have a higher thermal power, with the design basis level being reached after approximately 14 yr of operation. The radionuclide content of the early produced glass and

Table 6. Chemical composition of glass frits.^a

Frit No.	Oxide (wt%)								
	SiO ₂	Na ₂ O	TiO ₂	B ₂ O ₃	Li ₂ O	MgO	ZrO ₂	La ₂ O ₃	CaO
18	52.5	22.5	10.0	10.0	..	—	—	—	5.0
21	52.5	18.5	10.0	10.0	4.0	—	—	—	5.0
131	57.9	17.7	10.0	14.7	5.7	2.0	0.5	0.5	—
165	68.0	13.0	—	10.0	7.0	1.0	1.0	—	—

^a From Baxter (1983).

Table 7. Chemical composition of sludge-supernatant glass.^a

Component	wt%	Component	wt%
Ag ₂ O	0.75E - 02	MnO	1.73
Al ₂ O ₃	3.35	Na ₂ B ₄ O ₇	0.21E - 03
B ₂ O ₃	10.59	Na ₂ O	17.59
BaO	0.39E - 01	NiO	0.62
Ca ₃ (PO ₄) ₂	0.15	PbO	0.41E - 01
CaO	1.07	PuO ₂	0.15E - 01
CoO	0.16E - 01	RhO ₂	0.56E - 02
Cr ₂ O ₃	0.11	RuO ₂	0.25E 01
Cs ₂ O	0.73E - 01	SiO ₂	45.09
CuO	0.36E - 01	SrO	0.29E 01
Fe ₂ O ₃	6.09	ThO ₂	0.23
FeO	2.70	TiO ₂	0.72
Group A ^b	0.11	UO ₂	1.20
Group B ^c	0.36	Y ₂ O ₃	0.17E 01
K ₂ O	0.19E 03	Zeolite	1.47
La ₂ O ₃	0.36	ZnO	0.95E 01
Li ₂ O	4.13	ZrO ₂	0.36
MgO	1.59		

^a From Baxter (1983).^b Group A: Radionuclides of Mo, K, Se, Te, and Tl.^c Group B: Radionuclides of Ag, Am, C, Ce, Cm, Co, Cr, Eu, La, Nb, Nd, Np, Pd, Pm, Pr, Sb, Sm, Sn, Tb, Tl, and Zr.

the surface dose rate from those canisters will also be substantially less than the design basis.

The DHLW glass that is being tested for the NNWSI Project is based on Frit 165, which more likely represents the actual product of the DWPF than the reference Frit 131 glass. The glass that is currently being tested at LLNL was supplied to us by SRL in the form of a "mini-pour" can (SRL #171). The glass was melted at 1150°C and poured into a 500-cc SS beaker. No annealing procedures were performed (N. Bibler, SRL, letter to V. Oversby, June 6, 1983).

Table 12 lists the nominal composition of the glass as supplied to LLNL by SRL and the average of the results of electron microprobe analyses performed at LLNL on five samples of the glass. For each sample, 10 spot analyses were performed and the data were averaged. The uncertainty estimates given in Table 12 are based on the standard deviation of the average of the five sample averages. The SRL data giving the results of a partial analysis by x-ray fluorescence are shown for comparison.

Zeolites are aluminosilicate minerals; thus, the slightly higher SiO₂ vs the nominal composition in the analysis is easily explained. Only a portion of the higher Al₂O₃ can be explained by allowing for the zeolite component. Given the estimated precision of the analyses, it seems that the as-fabricated glass contains more Al than the nominal composition. There also seems to be slightly more Mn in the finished product than in the nominal composition. The lower Na₂O in the electron microprobe analysis may be due to volatilization of Na during analysis. Data for MgO, ZrO₂, Fe₂O₃, CaO, and NiO are in excellent agreement among the two analyzed values and the nominal composition.

The testing of fully active samples of DHLW glass is in progress at SRL under a cooperative testing program agreement between LLNL (representing NNWSI) and SRL. There are currently no plans to test fully active DHLW glass at LLNL.

Table 8. Radionuclide content of sludge-supernate glass.^a

Isotope	Ci/lb	Isotope	Ci/lb	Isotope	Ci/lb
⁵¹ Cr	2.45E 20	¹²⁵ Sb	2.29E 01	¹⁶⁰ Tb	3.09E 10
⁶⁰ Co	4.72E 02	¹²⁶ Sb	5.83E - 07	²⁰⁶ Tl	2.90E 07
⁵⁴ Ni	5.48E 04	^{126m} Sb	4.16E - 05	²³² U	4.03E 05
⁶¹ Ni	6.80E 02	^{125m} Te	4.16E 02	²³⁴ U	4.79E 09
⁷⁶ Se	2.84E 05	¹²⁷ Te	1.82E 05	²³⁴ U	1.27E 04
⁸⁷ Rb	1.89E 09	^{127m} Te	1.86E 05	²³⁵ U	4.21E 07
⁸⁶ Sr	2.13E 08	¹²⁹ Te	4.62E - 16	²³⁶ U	9.18E 06
⁹⁰ Sr	1.21E - 01	^{129m} Te	7.19E - 16	²³⁸ U	2.35E 06
⁹⁰ Y	7.35E - 00	¹³² Cs	1.05E 01	²³⁶ Np	4.81E 12
⁹¹ Y	2.28E 07	¹³⁵ Cs	2.33E 05	²³⁷ Np	2.44E 06
⁹¹ Zr	3.11E 04	¹³⁷ Cs	1.01E - 01	²³⁸ Pu	2.32E 05
⁹⁴ Zr	2.79E 06	^{137m} Ba	9.60E - 00	²³⁹ Pu	1.70E 15
⁹¹ Nb	2.58E 07	¹⁴¹ Ce	9.91E 15	²³⁸ Pu	2.81E - 01
⁹⁵ Nb	5.88E 06	¹⁴² Ce	2.63E - 09	²³⁹ Pu	2.65E 03
^{95m} Nb	3.45E 08	¹⁴⁴ Ce	2.71E + 00	²⁴⁰ Pu	1.68E 03
⁹⁸ Tc	5.18E 04	¹⁴¹ Pr	2.71E - 00	²⁴¹ Pu	3.18E 01
¹⁰¹ Ru	5.67E 12	^{144m} Pr	3.27E - 02	²⁴² Pu	2.32E - 06
¹⁰⁰ Ru	7.54E 01	¹⁴⁴ Nd	1.33E - 13	²⁴¹ Am	2.98E 03
^{101m} Rh	5.08E 12	¹⁴⁷ Pm	6.65E - 00	²⁴² Am	3.93E 06
¹⁰⁰ Rh	6.84E 01	^{147m} Pm	1.91E 14	^{242m} Am	3.96E 06
¹⁰⁷ Pd	2.58E 06	^{148m} Pm	2.77E 13	²⁴³ Am	1.58E 06
^{110m} Ag	3.78E 03	¹⁴⁷ Sm	5.40E - 10	²⁴² Cm	9.65E 06
¹¹³ Cd	2.42E 18	¹⁴⁹ Sm	1.57E 15	²⁴³ Cm	1.53E 06
^{115m} Cd	5.85E 14	¹⁵⁰ Sm	4.82E 16	²⁴⁴ Cm	4.48E 05
^{121m} Sn	8.22E 06	¹⁵¹ Sm	6.72E 02	²⁴⁵ Cm	1.83E 09
¹²² Sn	7.29E 05	¹⁵² Eu	1.02E 03	²⁴⁶ Cm	1.46E 10
¹²⁶ Sn	4.14E 05	¹⁵⁴ Eu	1.72E 01	²⁴⁷ Cm	1.79E 16
¹²⁵ Sb	1.97E 11	¹⁵⁵ Eu	1.32E - 01	²⁴⁸ Cm	1.87E 16
Total activity		5.42E + 01 Ci/lb			
Decay heat					
Total primary		1.05E 01 W/lb			
Total gammas		3.93E 02 W/lb			

^a From Baxter (1983).

Table 9. Physical properties of glass waste forms based on Frit 21 (unless otherwise noted).^a

Property	Value
Thermal conductivity at 100°C	0.95 W/m-°K
Heat capacity at 100°C	0.22 cal/(g)(°C)
Fractional thermal expansion ^b	$1.22 \times 10^{-5}/^{\circ}\text{C}$
Young's modulus ^c	62 GPa
Tensile strength	62 MPa
Compressive strength	690 MPa
Poisson's ratio ^d	0.2
Density at 100°C ^b	2.75 g/cc
Softening point ^b	502 C

^a From Baxter (1983).

^b Experimentally determined for Frit 131 glasses.

^c Young's modulus, or the modulus of elasticity, measures the stiffness of the material.

^d Poisson's ratio is equivalent to the ratio of equatorial-to-axial strain under an applied axial stress.

Table 10. Canister decay heat and activity of sludge-supernatant glass.^a

Year	Sludge-supernatant glass	
	Ci/can	W/can
5	176,700	470
10	137,900	397
15	118,100	360
20	103,700	320
25	91,680	285
30	81,290	255
35	72,140	228
40	64,050	204
45	56,890	183
50	50,540	164
60	39,910	132
70	31,540	107
80	25,250	87
90	19,760	71
100	15,670	58
200	1728	12
300	314	4.7
400	119	2.6
500	71	1.7
600	51	1.3
700	41	1.1
800	35	0.93
900	31	0.83
1000	29	0.76

^a From Baxter (1983).

Table 11. Radiation from sludge-supernatant glass canisters.^a

Basis	Case I	Case II	Case III
Fill height	91 in.	91 in.	107 in.
Glass density	2.37 g/cm ³	2.56 g/cm ³	2.56 g/cm ³
Waste loading	28%	28%	35%

Distance (ft)	Radiation level (rems/h)		
	Case I	Case II	Case III
Contact	5500	5275	6550
1/3	4150	3895	4825
1	1785	1660	2135
3	610	565	735
4	425	395	520
5	315	290	390
10	102	95	135
15	49	45	65
20	28	26	38
30	13	12	17
60	3.3	3	4.4
100	1.2	1.1	1.6

^a From Baxter (1983).

Table 12. Composition of DHLW glass (simulated waste) used in NNWSI testing.

Component	Nominal wt%	LNL wt%	SRL ^a wt%
SiO ₂	53.47	54.9 ± 2.1	
Na ₂ O	11.40	9.74 ± 0.54	
Li ₂ O	5.04		
B ₂ O ₃	7.20		
MgO	0.72	0.72 ± 0.04	0.65
ZrO ₂	0.72	0.66 ± 0.07	0.58
Fe ₂ O ₃	10.53	10.6 ± 0.7	10.2
MnO ₂	2.35	2.72 ± 0.13	2.6
CaO	1.37	1.51 ± 0.32	1.2
NiO	1.06	0.84 ± 0.24	0.83
Al ₂ O ₃	3.78	5.13 ± 0.18	
Zeolite	1.68		
CeO ₂	0.42		
K ₂ O	0.14		
SrO	0.10		
RuO ₂	0.036		
Cs ₂ O	0.0028		

^a Data from Bibler, SRL, pers. comm.

Commercial High-Level Waste: West Valley

Western New York Nuclear Service Center located near West Valley, N.Y., reprocessed nuclear reactor fuel from both Federal Government reactors and commercial power reactors from 1966 to 1972. Liquid wastes resulting from those reprocessing activities are currently stored in steel tanks in underground concrete vaults at the West Valley Site. Congress (Public Law 96-368) has directed the Department of Energy (DOE) to carry out at the Center a demonstration project that will involve solidification of the wastes into a durable high-level nuclear waste form suitable for disposal. DOE is now the site administrator, and the West Valley Nuclear Services Company, Inc. (WVNSC)—a wholly owned subsidiary of the Westinghouse Electric Corporation—is the site operator (WVDP-100).

Two different types of high-level liquid wastes are stored in the tanks at West Valley: (a) neutralized Purex wastes that consist of a layer of sludge covered by a supernate, stored in a carbon steel tank (8D2); and (b) acidic Thorex wastes that are stored in a SS tank (8D4). The neutralized Purex wastes, as will be documented below, are similar in composition to the DHHW at Savannah River.

The fuels processed at West Valley have a minimum "out of reactor" age in 1984 of 14 yr, and an average "age" of 17 yr. Many of the fuels reprocessed had a low reactor burnup (DOE/EIS-0081, 1982). West Valley wastes contain a large amount of inert iron in the neutralized sludge, and the acid wastes from the Thorex process are unusual because that fuel was from an experimental reactor.

To date, there is very little documentation of the actual content of radionuclides in the storage tanks at West Valley. Estimates, based on calculations of burnup of fuel and other relevant parameters, have been made of the expected content of the tanks. A borosilicate glass waste form is being tailored to the expected tank compositions and will be available late in 1984 (I. Pope, WVNSC, pers. comm.).

The neutralized wastes, contained in tank 8D2, consist of approximately 2 million gallons of waste generated from the reprocessing of 625 metric tons of uranium fuel. Approximately 44% of this waste was from the reprocessing of commercial reactor fuel and the remainder from defense-related fuels. The defense fuels had, on average, lower burnup; thus, the present radioactive con-

tent of the tank is derived primarily from the commercial fuels (DOE/EIS-0081, 1982). Table 13 gives the estimated chemical composition of the neutralized wastes contained in the sludge and supernate of tank 8D2. Table 14 gives the estimated radioactivity in the waste as of 1987.

The acidic wastes contained in tank 8D4 were generated during the reprocessing of the fuel from an experimental reactor. The fuel contained an original composition of 93.5% thorium and 6.5% highly enriched uranium (DOE/EIS-0081, 1982). The wastes were retained as acid solutions to avoid precipitation of thorium. There is some indication, however, that thorium precipitation has occurred in the acid solutions (DOE/EIS-0081, 1982, p. B-7). Table 15 lists the estimated chemical composition of the acidic waste and Table 16 lists the estimated radioactivity as of 1987. The estimated fission-product inventory in the acid waste is ten times less than that in the neutralized wastes, and the transuranic content of the acid wastes is expected to be commensurately less (DOE/EIS-0081, 1982).

The reference process for management of West Valley wastes involves the treatment of the supernatant liquid to separate and concentrate Cs-137 and a trace amount of Sr-90, leaving a low-level radioactive sodium salt to be disposed of separately. The sludge from neutralized wastes would be washed with water to reduce the sodium concentration. The rinse solutions from the sludge would be treated with the low-level portions of the supernate. Washed sludge and fission products from the supernate would be combined with the acid wastes and glass frit and would be blended. The blended product would be fed to an electrically heated, ceramic-lined melter. The average product is expected to consist of 75% by weight of alkaline (neutralized) wastes and 25% by weight of acid wastes with respect to the radioactive components. The final waste form will be approximately 28% by weight of waste and 72% by weight of glass formers (WVDP-100).

Table 17 shows the average expected characteristics of waste glasses that the West Valley Demonstration Project will produce. The reference does not state the time for which the curie content per canister is calculated; however, comparison of the values in Tables 14 and 16 divided by the total canister estimate of 300 suggests that the curie content is approximately the value in 1983. The curie content per canister is less than

Table 13. Estimated composition of neutralized Purex wastes (Tank 8D2).^a

Compound	In Sludge		In Solution	
	10 ³ moles	Kilograms	10 ³ moles	Kilograms
(Na, K) ₂ SO ₄	—	—	630	89,800
(Na, K) NO ₃ /NO ₂	—	—	12,300	1,090,000
(Na, K) OH	—	—	386	15,400
(Na, K) Cl	—	—	8.4	500
Fe (OH) ₃	460	49,500	—	—
Fe PO ₄	190	28,500	8.3	1,250
Cr (OH) ₃ , ^b	38	3,900	21	2,100
Ni (OH) ₂ , ^c	18	1,700	8.3	750
Al (OH) ₃	11	830	—	—
AlF ₃	—	—	18.7	1,570
Rare earth [M (OH) ₃]	8.3	1,620	—	—
Fission product, remainder	—	—	2.9 ^d	550
(M'SO ₄)	5.5	1,240	—	—
[M' (OH) ₃]	9.6	1,580	—	—
(M ₂ O ₃) - (MO ₃) ^e	.07	210	1.2 ^e	220
Mo, as Na phosphomolybdate (Na ₂ PO ₄ 12MoO ₃)	7.8	980	4.2	500
Mn, as MnO ₂ (estimated)	25	2,200	—	—
Pu, as PuO ₂	—	35	—	—
U, as Na ₂ U ₂ O ₇	21	6,700	—	—
Actinides, total as oxides	—	19	—	—
TOTALS		99,000		1,203,000
	~28,300 liters (at assumed dry density of 3.5 g/mL)			557 g/L

^a Source: U.S. Department of Energy (1978).^b Assuming that the ratio of chromium to nickel is 19:9 (the ratio in which it occurs in 304 SS).^c The average atomic weight is 143.9.^d (Rb, Cs) NO₃, the average molecular weight is 189.52.^e Technetium as NaTcO₄. It is very doubtful that this compound of Tc is present; it is most likely to be present as a pertechnetate, coprecipitated with BaSO₄ (Martin & Miles, "Chemical Processing of Nuclear Fuels," Academic Press, 1958, p. 77).

the DWPF estimates for design purposes but is similar to the average anticipated output of the DWPF (see preceding section). The chemical composition of average West Valley wastes is within the anticipated range for the DWPF or can be made so by the addition of inert elements to the glass frit (DOE/EIS-0081, 1982).

The expected glass composition for West Valley waste will fall within the range expected for DHIW glasses. A decision on the reference glass

composition for West Valley will not be made until late 1984 (J. Pope, WVNSC, pers. comm.). For these two reasons, NNWSI has decided not to initiate testing of glass specifically related to West Valley wastes until a glass composition is chosen by West Valley Nuclear Services Company, Inc., after which testing will be coordinated to derive maximum benefit from previous experience with DHIW glasses of similar compositions.

Table 14. Estimated 1987 radioactivity of neutralized Purex wastes (Tank 8D2).^a

Fission product	Radioactivity (curies)			Actinide	Radioactivity (curies)		
	Sludge	Supernate	Total		Sludge	Supernate	Total
Tritium (H-3)	0	2.4×10^1	2.4×10^1	Uranium-235	8.0×10^{-2}	0	8.0×10^{-2}
Selenium-79	0	5.0×10^1	5.0×10^1	Uranium-238	8.2×10^{-1}	0	8.2×10^{-1}
Strontium-90	6.6×10^0	6.7×10^1	6.7×10^0	Neptunium-237	2.3×10^1	- 1	2.3×10^1
Yttrium-90	6.6×10^0	6.7×10^1	6.7×10^0	Neptunium-239	2.2×10^2	- 1	2.2×10^2
Zirconium-93	2.5×10^2	- 1	2.5×10^2	Plutonium-238	1.5×10^3	1.5	1.5×10^3
Niobium-93m	2.4×10^2	1	2.4×10^2	Plutonium-239	1.8×10^3	1.8	1.8×10^3
Technetium-99	0	1.9×10^3	1.9×10^3	Plutonium-240	9.7×10^2	- 1	9.7×10^2
Ruthenium-106	1.0×10^2	1.0×10^1	1.1×10^2	Plutonium-241	7.0×10^1	7.0×10^1	7.0×10^1
Rhodium-106	1.0×10^2	1.0×10^1	1.1×10^2	Plutonium-242	- 1	0	- 1
Palladium-107	6	- 1	6	Americium-241	2.0×10^1	2.0×10^1	2.0×10^1
Antimony-125	6.0×10^1	6.0×10^1	6.1×10^1	Americium-242	1.8×10^2	- 1	1.8×10^2
Tellurium-125m	0	6.1×10^1	6.1×10^1	Americium-242m	1.8×10^2	1	1.8×10^2
Irin-126	4.0×10^1	- 1	4.0×10^1	Americium-243	2.2×10^2	- 1	2.2×10^2
Antimony-126m	4.0×10^1	- 1	4.0×10^1	Curium-242	- 1	0	- 1
Antimony-126	4.0×10^1	- 1	4.0×10^1	Curium-244	8.8×10^3	8.8	8.8×10^3
Iodine-129 ^b	0	4.7	4.7	Curium-245	1	0	1
Cesium-134	0	2.1×10^1	2.1×10^1	Curium-246	1	0	- 1
Cesium-135	0	3.5×10^1	3.5×10^1	Total			1.0×10^4
Cesium-137	0	8.9×10^0	8.9×10^0				
Barium-137m	0	8.4×10^0	8.4×10^0				
Cerium-144	1.1×10^1	- 1	1.1×10^1				
Praseodymium-144	1.1×10^1	1	1.1×10^1				
Promethium-147	6.0×10^1	6.0×10^1	6.0×10^1				
Samarium-151	2.0×10^2	2.0×10^1	2.0×10^2				
Europium-152	4.1×10^2	1	4.1×10^2				
Europium-154	1.3×10^3	1.4×10^1	1.3×10^3				
Total			3.1×10^4				

^aSource: U.S. Department of Energy (1978), with appropriate radioactive decay taken into account.

No iodine should remain after Purex reprocessing of the fuel; however, the iodine was included to yield conservative doses.

Table 15. Estimated composition of acidic Thorex wastes (Tank 8D4).^a

Compound	Weight (metric tons)
H ₂ (NO ₃) ₂	32 ^b
Al(NO ₃) ₃	2.4
NaI	0.2
HNO ₃	3.0
H ₃ PO ₄	0.2
H ₂ BO ₃	0.4
M(NO ₃) ₃ (M = Fe, Cr, Ni)	0.8

^aSource: U.S. Department of Energy (1978).^bApproximately 15,500 kg of thorium.Table 16. Estimated 1987 radioactivity of acidic Thorex wastes (Tank 8D4).^a

Fission product	Radioactivity (Ci)
Cobalt-60	1.5×10^1
Strontium-90	6.5×10^1
Yttrium-90	6.5×10^1
Cesium-134	5.4×10^2
Cesium-137	6.9×10^3
Barium-137m	6.4×10^3
Europium-154	4.2×10^3
Total	2.6×10^4

^aSource: U.S. Department of Energy (1978), with appropriate radioactive decay taken into account.

Table 17. Features of the West Valley Demonstration Project waste form and canister.^a

Feature	West Valley Reference
Waste loading	23 wt%
Borosilicate glass density	2.75 g/cm ³
Borosilicate glass weight	1480 kg
Canister material	304L SS
Canister dimensions	0.61 m diam 3.0 m length 0.95 cm wall
Total canister weight (with borosilicate glass)	1930 kg
Heat generation	365 W
Heat generation, after 1000 yr	~ 1 W
Radionuclide content	130,000 Ci
Radionuclide content, after 1000 yr	500 Ci
Number of canisters	300

^a Source: WVDP-100.

Commercial High-Level Waste: Potential Waste Forms

There is presently no commercial reprocessing facility for spent fuel in the United States; thus, there are no high-level wastes being generated from commercial sources. Should a reprocessing facility be built and begin producing high-level wastes, the waste form for solidification of those wastes would be tailored to the details of the facility process flow sheet.

In the absence of a clear candidate for commercial high-level waste (CHLW) forms, the NNWSI Project has chosen as a reference waste form a borosilicate glass based on the PNL 76-68 formulation. NNWSI chose PNL 76-68 because an extensive data base on its physical and chemical properties and on the leaching of glass of that general composition in deionized water exists. This data base can be used as a starting point for determining appropriate test conditions that will aid in estimating the long-term performance of CHLW glasses in the Yucca Mountain environment.

Mendel et al. (1977) give the reference composition for PNL 76-68 glass. The glass was based on Frit 76-101 and waste composition PW-8a. Table 18 lists the chemical composition of those components and the resulting glass. The waste composition represents the combination of high- and intermediate-level wastes from a Purex re-

processing plant that uses mild steel basket liners to transfer chopped fuel to the dissolver. The basket is dissolved along with the spent fuel giving a high iron content to the waste.

Waste glasses used for performance testing are frequently based on a simplification of the original formulation. The usual variations include elimination of the noble metals (Ru, Pd, Rh), which are expensive, a simplification of the suite of rare earth elements to allow use of a relatively inexpensive rare earth mixture in the glass fabrication, and elimination of the actinides and technetium to simplify handling of the glass. Table 19 outlines the chemical analysis results of glasses labeled MCC-76-68 glass, which are being used by the Materials Characterization Center (MCC), Battelle Pacific Northwest Laboratory, Richland, Wash., as feed stock for the preparation of two glass testing materials for NNWSI. The first glass was prepared by doping the base glass with approximately 4% UO_2 . The second glass is being fabricated to include all of the components in the first glass plus NpO_2 (0.4%), PuO_2 (0.1%), Tc_2O_7 (0.5%), RuO_2 (1.0%), Rh_2O_3 (0.17%), and PdO (0.57%). NNWSI requested a third glass formulation from MCC with a composition specified to represent a glass sample 300 yr after disposal.

Table 18. Chemical composition of PNL 76-68 borosilicate glass, Frit 76-101, and waste PW-8a.^a

Component	Frit 76-101 (wt%)	Glass PNL 76-68 (wt%)
SiO ₂	59.7	40.0
B ₂ O ₃	14.2	9.5
Na ₂ O	11.2	7.5
ZnO	7.45	5.0
CaO	3.0	2.0
TiO ₂	4.45	3.0
PW-8a waste		33.0

Component	PW-8a (wt%)	Component	PW-8a (wt%)
Inerts		Actinides	
Na ₂ O	15.08	U ₃ O ₈	12.54
Fe ₂ O ₃	29.21	NpO ₂	0.93
Cr ₂ O ₃	1.24	PuO ₂	0.19
NiO	0.61	Am ₂ O ₃	0.19
P ₂ O ₅	1.44	Cm ₂ O ₃	0.04
Fission products		Fission products (cont.)	
Rb ₂ O	0.38	TeO ₂	0.78
SrO	1.14	Cs ₂ O	3.09
Y ₂ O ₃	0.64	BaO	1.68
ZrO ₂	5.31	La ₂ O ₃	1.59
MoO ₃	5.55	Ce ₂ O ₃	3.57
Ta ₂ O ₅	1.39	Pr ₆ O ₁₁	1.59
RuO ₂	3.19	Nd ₂ O ₃	4.85
Rh ₂ O ₃	0.52	Pm ₂ O ₃	0.13
PdO	1.59	Sm ₂ O ₃	0.99
Ag ₂ O	0.09	Eu ₂ O ₃	0.21
CdO	0.10	Gd ₂ O ₃	0.15

^a Source: Mendel et al. (1977).

The appellation "PNL-76-68" originally indicated a specific melt number (Mendel et al., 1977); its subsequent usage has carried a more general meaning and usually indicate a glass that has a composition similar to the original PNL 76-68

melt run. The composition in Table 19 differs from the original 76-68 primarily due to the simplifications mentioned above and the addition of Al₂O₃ to the glass formulation.

Table 19. Composition of MCC-76-68 glass.^a

Oxide	Compositions (wt%) ^b					
	Lot 1		Lot 2		Lot 3	
	wt%	Standard deviation ^c	wt%	Standard deviation ^c	wt%	Standard deviation ^c
Al ₂ O ₃	0.84	0.03	0.63	0.03	0.59	0.02
B ₂ O ₃	8.65	0.06	8.99	0.15	9.06	0.01
BaO	0.62	0.01	0.54	0.02	0.55	0.00
CaO	2.29	0.04	2.22	0.11	2.37	0.04
CdO	0.05	0.01	0.05	0.01	0.05	0.01
CeO ₂	0.02	—	0.82	0.11	0.92	0.01
Cr ₂ O ₃	0.50	0.02	0.47	0.01	0.47	0.01
Cs ₂ O	1.21	0.08	1.14	0.01	1.11	0.06
Dy ₂ O ₃	0.01	0.00	0.01	0.00	0.01	0.00
Eu ₂ O ₃	0.01	0.00	0.01	0.00	0.01	0.00
Fe ₂ O ₃	9.08	0.12	9.52	0.17	9.56	0.13
Gd ₂ O ₃	0.03	0.01	0.02	0.00	0.02	0.00
K ₂ O	0.09	0.03	0.27	0.06	0.06	—
La ₂ O ₃	4.89	0.06	4.33	0.24	4.16	0.05
MgO	0.18	0.01	0.14	0.01	0.16	0.01
MnO ₂	0.06	0.00	0.06	0.01	0.06	0.01
MoO ₃	2.17	0.01	1.96	0.05	1.97	0.03
Na ₂ O	11.60	0.95	11.30	0.20	12.77	1.21
Nd ₂ O ₃	1.65	0.01	1.47	0.08	1.41	0.02
NiO	0.25	0.03	0.22	0.02	0.23	0.01
P ₂ O ₅	0.56	0.08	0.66	0.11	0.80	0.16
SiO ₂	40.33	0.81	40.73	0.32	42.00	0.26
SrO ₂	0.49	0.01	0.42	0.01	0.42	0.01
TiO ₂	2.86	0.04	3.04	0.03	3.08	0.05
ZnO	4.75	0.08	4.76	0.05	5.10	0.46
ZrO ₂	2.33	0.04	1.79	0.07	1.89	0.04
TOTAL	95.50		95.57		98.76	

^a Source: Mellinger and Daniel (1983).^b Based on analysis of three bars for each lot.^c Standard deviation of a single analysis.

Packing Material

Preliminary estimates of release rates from waste forms indicated that for porousilicate glass waste forms in the Topopah Spring, the release of radionuclides for unsaturated conditions would be less than 1 part in 100,000/yr for all relevant species. For spent fuel, the situation is less clear and may depend on the condition of the Zircaloy cladding. To increase our options with respect to the control of release of radionuclides from the engineered barrier system, we have been investigating the potential use of a packing material as part of the waste package for spent fuel.

Chemical interactions become increasingly complex as the number of components in the system is increased. To minimize the potential for deleterious interactions among the system components and at the same time gain the positive features of packing material, we have chosen one based on crushed tuff. The reference packing material is Topopah Spring tuff crushed to pass 100-mesh screen and compressed isostatically at 20,000 psi. Alternative packing materials involve the reference material plus additives to improve the quality of the final product. Additives under

investigation include iron-smectite clays and silica gel.

The primary purpose of packing material in the spent fuel waste package would be to provide a region around the spent fuel where the water flow mechanism is known to be porous media flow. Daniel et al. (1982) used crushed devitrified Topopah Spring tuff with similar mineralogy to that expected for the repository horizon in batch sorption studies. They found sorption ratios (in ml/gm) of 59 for Sr, 365 for Cs, 2500 for Am, 1330 for Pu, 1.2 for Tc, and 33 for Np. Consequently, the packing material can be expected to provide some retardation of radionuclide release due to sorption.

A fabricated packing material will probably have a thermal conductivity that is substantially less than that of the rock from which the material was made and of the repository host rock. Consequently, the packing material would cause waste form temperatures to be higher than for waste packages that did not include packing material. If the contrast in thermal conductivity between the host rock and the packing material is too great, the thermal loading of the waste packages might have to be decreased, which would result in an increased total number of waste packages and commensurately increased costs.

Because the thermal conductivity of the packing material may be a limiting factor in the waste package design, we have directed the initial efforts in packing material development toward determining the fabrication techniques that will provide reliable high-density products. This will help to provide high thermal conductivity in the finished product. Mechanical properties of the packing material are also important because of the handling operations involved in assembling the waste package. Initial fabrication trials used two sizes of crushed material to determine whether grain size affected the density of the compressed samples and their mechanical stability.

Crushed Topopah Spring tuff was prepared in two size ranges: less than 50 mesh and less than 100 mesh. An attempt to compact material crushed to less than 18 mesh was unsuccessful because the sample was too fragile to handle following compaction. Samples are compacted using isostatic pressure. A thin Teflon liner is used between the outer Tygon jacket and the crushed rock to prevent the sample from sticking to the jacket. If the Teflon liner had not been used, the

finished sample would have pulled apart as "poker chips" when the pressure was released. Table 20 gives the density of the finished samples as a function of confining pressure. For confining pressures of 15,000 psi or more, the density increases only slightly as the compaction pressure is increased. This is a favorable characteristic from the point of view of process control during fabrication, since rather large variations in pressure would have only a small effect on sample quality. Samples prepared from the inner-grained material had better mechanical stability than those prepared from the coarser material.

To determine the thermal conductivity of the packing material, a large sample was prepared using approximately 1 kg of crushed tuff. The sample was isostatically pressed at 25,000 psi, removed from the pressing jacket, and placed in an aluminum canister. A central hole was drilled in the sample and a line heater assembly was placed in the hole. Thermocouples were placed on the sample surface and in the heater assembly. Thermal conductivity was measured at room temperature (approx. 20°C), 60°, 100°, 130°, 165°, and 200°C. Data points were taken on the heating cycle, with two measurements at each temperature; Fig. 8 plots the results. Duplicate measurements at 20°C and 100°C gave the same value and are shown as a single point in each case. The agreement between results at the same temperature is very good, and the thermal conductivity seems to be essentially independent of temperature.

The measured conductivity for the fabricated sample is approximately one-third of that for the rock that was used as the starting material. In an

Table 20. Test results of the fabrication of the packing material.

Mesh size of powder	Compacting pressure (Kpsi.)	Density (g/cm ³)
60	15	1.73 ± 0.06
60	25	1.75 ± 0.06
100	10	1.55 ± 0.03
100	15	1.64 ± 0.03
100	20	1.65 ± 0.03
100	25	1.71 ± 0.03
100	30A	1.65 ± 0.04
100	30B	1.69 ± 0.04
100	35	1.72 ± 0.03

Figure 8. Thermal conductivity vs temperature of compressed Topopah Spring crushed tuff packing material.

attempt to improve the sample density and thermal conductivity, a sample was prepared using a mixture of 95% crushed tuff plus 5% bentonite clay. The finished sample was considerably less friable than the samples made from crushed tuff alone, but the density and thermal conductivity of

the mixture were the same as that for samples made with crushed tuff alone. Future work on packing material development will focus on the use of other additives to attempt to improve the density, mechanical stability, and thermal conductivity of the fabricated materials.

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