**Assessment of Degradation
Concerns for Spent Fuel, High-
Level Wastes, and Transuranic
Wastes in Monitored Retrievable
Storage**

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MONITORED RETRIEVABLE STORAGE

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ABSTRACT

A study was performed by Pacific Northwest Laboratory to assess potential degradation mechanisms for spent fuel (SF), high-level wastes (HLW), and transuranic wastes (TRUW) under dry storage conditions in a monitored retrievable storage (MRS) facility. It was concluded that there are no significant degradation mechanisms that could prevent the design, construction, and safe operation of MRS facilities. Long-term mechanisms that could affect the ability to maintain or readily retrieve SF, HLW, and TRUW were identified.

EXECUTIVE SUMMARY

It has been concluded that there are no significant degradation mechanisms that could prevent the design, construction, and safe operation of monitored retrievable storage (MRS) facilities. However, there are some long-term degradation mechanisms that could affect the ability to maintain or readily retrieve spent fuel (SF), high-level wastes (HLW), and transuranic wastes (TRUW) several decades after emplacement. Although catastrophic failures are not anticipated, long-term degradation mechanisms have been identified that could, under certain conditions, cause failure of the SF cladding and/or failure of TRUW storage containers.

Stress rupture limits for Zircaloy-clad SF in MRS range from 300 to 440°C, based on limited data. Additional tests on irradiated Zircaloy (3- to 5-year duration) are needed to narrow this uncertainty. Cladding defect sizes could increase in air as a result of fuel density decreases due to oxidation. Oxidation tests (3- to 5-year duration) on SF are also needed to verify oxidation rates in air and to determine temperatures below which monitoring of an inert cover gas would not be required.

Few, if any, changes in the physical state of HLW glass or canisters or their performance would occur under projected MRS conditions. The major uncertainty for HLW is in the heat transfer through cracked glass and glass devitrification above 500°C.

Additional study of TRUW is required. Some fraction of present TRUW containers would probably fail within the first 100 years of MRS, and some TRUW would be highly degraded upon retrieval, even in unfailed containers. One possible solution is the design of a "100-year" container.

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1.0 INTRODUCTION

This report summarizes the results of a study performed by Pacific Northwest Laboratory^(a) to assess potential mechanisms for degradation of spent fuel (SF), high-level wastes (HLW), and transuranic wastes (TRUW) under dry storage conditions. The purpose of this study was to assess the potential consequences of such degradation on the design and operation of monitored retrievable storage (MRS) facilities.

1.1 OBJECTIVES

The objectives of this study were to identify the principal degradation mechanisms of concern to MRS, evaluate the probable significance and consequences of such degradation, and assess the current state of the art for predicting and accommodating some of these effects in the design of MRS facilities and in MRS operating procedures. A particular objective was to identify areas where knowledge of fuel/waste degradation is incomplete and where additional work might reduce design conservatism or otherwise enhance the ability to design, construct, and operate MRS facilities. The study was commissioned by the U.S. Department of Energy (DOE) MRS Program to aid in the preparation of a proposal for the development of MRS facilities that must be submitted to Congress by June 1, 1985.

1.2 BACKGROUND

The Nuclear Waste Policy Act of 1982 (NWPAct or the Act), PL-97-425, defines the federal government's overall program for management of SF and HLW from commercial nuclear power operation. MRS--the long-term isolation of SF and HLW in facilities that permit continuous monitoring, ready retrieval, and periodic maintenance as necessary to assure containment of radioactive materials--is a potential element of that waste management program. DOE has the primary responsibility for implementing provisions of the NWPAct. Section 141(b) of the Act provides that:

(a) Operated for the U.S. Department of Energy (DOE) by Battelle Memorial Institute.

On or before June 1, 1985, the Secretary [of Energy] shall complete a detailed study of the need for and feasibility of, and shall submit to the Congress a proposal for, the construction of one or more monitored retrievable storage facilities for high-level radioactive waste and spent nuclear fuel.

This proposal will provide Congress with candidate sites and MRS facility designs for consideration on a schedule parallel to that for geologic repository programs.

An initial step in the development of the proposal to Congress was the completion of the MRS research and development (R&D) report (DOE 1983) required by Section 141(b) of the Act. In preparing this report, prior design activities and R&D related to the long-term retrievable storage of SF and HLW were reviewed. This review indicated that passive dry storage technologies, which do not require any external power to provide cooling, are the preferred technologies for long-term MRS applications. In preparing the MRS proposal, DOE has continued to pursue these passive dry storage technologies to minimize the dependence of the operation and safety of MRS facilities on active components.

There have been almost two decades of actual operating experience and research in passive dry storage technology in the United States and abroad. Based on analysis of this experience and data, the R&D report (DOE 1983) concluded that no further R&D activities are needed to develop the June 1985 MRS proposal. It was also observed, however, that as the proposal is developed "...specific R&D activities may be identified that could contribute to a more efficient design, improve safety or facilitate licensing." This study was commissioned based on the judgment of MRS program staff that there may be areas of uncertainty related to fuel/waste degradation where such additional R&D activities could benefit both the near-term and long-range objectives of the MRS Program.

2.0 SUMMARY AND CONCLUSIONS

The general conclusion from this review is that there are no significant degradation mechanisms that could prevent the design, construction, and safe operation of MRS facilities. There are some long-term degradation mechanisms that could affect the ability to maintain or readily retrieve SF, HLW, and TRUW several decades after emplacement. Although catastrophic failures are not anticipated, long-term degradation mechanisms have been identified that could cause failure of SF cladding and/or failure of TRUW storage containers under certain conditions. Principal potential concerns for each waste form are summarized below; the bases for these conclusions are discussed in detail in Section 3.

2.1 SPENT FUEL

If SF rods arrive at an MRS facility without incipient defects, operating temperatures can normally be regulated to prevent stress rupture in most fuel rod cladding. The actual temperature limits for preventing stress rupture of Zircaloy-clad fuel, as derived from different investigators, range from 300 to 440°C based on existing data, depending on the age and condition of the fuel. Additional stress rupture data are needed to verify the assumptions used in deriving these temperature limits for irradiated Zircaloy cladding. However, even the lower estimate of acceptable temperatures (300°C) in an inert cover gas should present no appreciable constraint on MRS system design or operation.

Degradation in stainless steel (SS) rods is not as well characterized. No low-temperature stress rupture data are available for SS cladding, although low-temperature stress corrosion cracking (SCC) data are available for SS other than that used for fuel cladding. A temperature criteria has not been proposed for storage of SS-clad fuel in an MRS facility.

If the cladding is breached (from in-reactor operation, propagation of incipient defects, or stress rupture), then fuel oxidation can occur rapidly during dry storage in air, even at 300°C, since appreciable oxidation rates occur at least as low as 200°C. The change in fuel density due to oxidation can cause increased cladding failure and accelerated oxidation of the fuel,

depending on the availability of oxygen to the interior of the canister. If the canister is sealed, significant fuel oxidation could still occur in an unconsolidated MRS canister filled with air because of the large plenum volume. Some fuel damage could occur in a sealed canister of consolidated fuel rods. However, fuel oxidation can be prevented by storage in inert gas.

Even if there is no stress rupture and incipient defect propagation does not occur during MRS storage, there will probably be some rods with reactor-induced breaches that are placed in storage. Oxidation of fuel in these rods during air storage could cause a problem if the canister is subsequently opened for consolidation at a geologic repository or for other reasons. For air storage, a system for detecting as many of these failed rods as possible may be needed at the MRS facility. In addition, a procedure for handling canisters with ruptured cladding and oxidized fuel may also be required since it cannot be assured that all failed fuel rods will be detected.

2.2 HIGH-LEVEL WASTE

A comprehensive review of the extensive studies on HLW glass and canisters was conducted to identify potential degradation mechanisms that might be active during storage of HLW in an MRS facility. The review indicated that few, if any, changes in the physical state of the glass or canister or their ability to perform would occur under projected MRS conditions. The primary limitations on the storage of HLW are that the glass centerline temperature must be kept below 500°C and the canister surface temperature must be kept below 375°C. The glass limit prevents devitrification of the glass, and the canister limit prevents oxidation and sensitization of the canister. These limits should not significantly constrain MRS system design or specifications. Other degradation mechanisms, such as helium buildup and stress-induced cracking of the glass, do not produce significant limitations.

The primary uncertainties about HLW storage are the heat transfer in the canister and devitrification behavior above 500°C. Additional studies are needed to verify that glass cracking does not impede heat transfer through the glass and to determine the thermal interactions between the HLW and the MRS

facility. New studies are needed to measure devitrification behavior of recently developed glasses at temperatures above 500°C. Assessments can then be made of the devitrification that may occur during off-normal temperature excursions above 500°C. If different wastes (for example, breeder fuel reprocessing) are to be considered, then new studies on their performance should be conducted.

2.3 TRANSURANIC WASTE

A review of the degradations of concern for TRUW indicates that this waste form needs additional study. For present container designs, some fraction of the TRUW containers will probably fail within the first 100 years of MRS. Even if the waste is retrieved before container failure, some TRUW containers will probably be highly degraded and difficult to handle and process. If operating temperatures are significantly above ambient (for example, >100°C), degradation will be exacerbated. Designs should provide for low-temperature storage (<100°C), and provisions for retrieval should assume a dispersible product.

Because of the probable container failure due to internal corrosion during storage, operating procedures may have to provide for repackaging of all TRUW containers at suitable intervals. It may be feasible to schedule repackaging on the bases of TRUW loading, product type (probably unknown), solidification method, drum coating, etc. One alternative would be to incorporate a corrosion detection system to warn of incipient drum failure from either internal or external corrosion. This approach would probably require a significant R&D effort to develop and prove the needed corrosion detection technology. A more direct but more capital-intensive alternative would be to design a "100-year" container.

2.4 DEGRADATION MODELING

Storage temperatures of 300 to 440°C have been calculated as the limiting temperatures for SF cladding breach in dry storage; however, these predictions are based on data generated using unirradiated Zircaloy cladding. Assumptions regarding irradiation effects on cladding properties account for much of the uncertainty. Stress rupture tests on SF cladding could narrow this uncertainty

in calculated temperature limits. The necessary tests should be extended 3 to 5 years since modeling predicts that most of the cumulative stress rupture occurs during peak temperatures existing within this time frame.

Fuel oxidation studies have been conducted primarily on unirradiated UO_2 fuel under limited temperature and atmospheric conditions. Variable temperature tests on irradiated light-water reactor (LWR) fuels are needed to verify models for cumulative damage to fuel by oxidation. These models will allow the determination of temperatures below which monitoring of storage atmospheres would not be required. The duration of the fuel oxidation tests required to develop the necessary model parameters at acceptable levels of uncertainty is estimated to be 3 to 5 years. These longer tests are necessary because extrapolation of the oxidation data to several decades may be required.

3.0 REVIEW AND ASSESSMENT OF WASTE FORMS

Background information on the waste forms that might be stored in an MRS facility is presented in Section 3.1. Operating parameters and degradation mechanisms of concern for SF in an MRS facility are discussed in Section 3.2; and similar discussions for HLW and TRUW are provided in Sections 3.3 and 3.4, respectively.

3.1 WASTE FORM TYPES AND QUANTITIES

This section provides background information on the waste forms that may be placed in an MRS facility by reviewing the current and projected amounts of each waste form. SF refers to both individual fuel rods and intact or consolidated assemblies after they are discharged from the reactor. HLW is generated as a result of reprocessing SF. It is usually defined as the waste product of the first stream of reprocessing and may exist as a liquid or solid depending on subsequent treatment. HLW that will go into MRS is solid, processed, and packaged. TRUW is material that is contaminated to greater than 100 nanocuries per gram (nCi/g) with certain alpha-emitting radionuclides with long half-lives and high specific radiotoxicity. The radionuclides include ^{233}U , plutonium, and transplutonium nuclides (DOE 1982a).

Of all the waste forms, SF is the primary source of radioactivity during storage: SF contributed ~86% of the total storage radioactivity in 1981, and HLW comprised nearly the balance. Volumetrically, SF consumed only 0.1% of the space occupied by all (commercial and defense) nuclear wastes stored in 1981. HLW, including both liquid and glass, consumed 9.7% of the space; TRUW consumed 11.7% of the total volume of nuclear wastes; and low-level wastes (LLW) made up the remainder. The volume percentages of SF, HLW, and TRUW are expected to decrease because of the higher accumulation rate for LLW, which is not a candidate waste form for MRS. Disregarding the volume of LLW, the relative volumes of just SF, HLW, and TRUW should remain about the same (DOE 1982a). The mass, radioactivity, and thermal power of SF in storage as of 1981 as well as the projected amounts up to the year 2020 are given by reactor type in Table 1.

TABLE 1. Historic and Projected Mass, Radioactivity, and Thermal Power of Spent Fuel by Reactor Type Assuming No Future Reprocessing^(a,b) (DOE 1982a)

End of Calendar Year	Mass, MTHM		Radioactivity, 10 ⁶ Ci		Thermal Power, 10 ⁶ W	
	Annual Rate ^(c)	Accumu- lation	Annual Rate	Accumu- lation	Annual Rate	Accumu- lation
Boiling Water Reactors						
1970	0.9	9.4	3.0	12.0	0.0	0.0
1980	575.4	2914.4	2,343.2	3,628.8	9.3	13.7
1990	1,009.8	10,855.5	4,167.0	9,000.0	16.6	32.5
2000	1,666.4	24,841.2	6,910.6	16,199.1	27.5	57.2
2010	2,225.5	45,628.2	9,286.1	25,672.9	37.0	90.1
2020	2,951.5	73,368.7	12,315.4	35,464.4	49.1	123.2
Pressurized Water Reactors						
1970	38.8	38.8	190.0	190.0	0.8	0.8
1980	639.1	3,812.1	3,506.7	6,150.1	14.1	23.6
1990	2,082.3	18,173.2	11,542.8	22,317.3	46.4	83.7
2000	2,366.4	40,127.5	13,246.9	32,149.1	53.3	115.9
2010	4,032.3	72,858.1	22,902.9	55,230.3	92.3	200.7
2020	4,112.9	112,812.4	23,709.8	68,991.4	95.8	246.7
Total						
1970	39.7	48.2	193.0	201.9	0.8	0.8
1980	1,214.5	6,726.9	5,849.9	9,778.8	23.4	37.4
1990	3,092.1	29,029.1	15,709.8	31,317.2	63.0	116.3
2000	4,032.8	64,969.1	20,157.5	48,348.2	80.8	173.1
2010	6,257.8	118,486.7	32,189.0	80,903.3	129.3	290.8
2020	7,064.4	186,181.5	36,025.2	104,455.8	144.9	369.9

(a) Based on 167.4 GWe-installed in the year 2000 and 280.7 GWe-installed in the year 2020.

(b) Spent fuel reprocessed at the Nuclear Fuel Services (NFS) plant is not included in the historic discharges.

(c) Annual rate is for year indicated.

The relatively small amount of commercial HLW (nonglassified) is compared with the amount of defense HLW in Table 2. Unless reprocessing begins in 1989, as assumed for Table 3, the quantity of commercial HLW will remain at 2200 m³. More germane to MRS are the projected amounts of HLW glass for commercial wastes indicated in Table 3. It is noteworthy that the amount of commercial HLW glass is expected to exceed that of defense HLW glass between the years 2005 and 2010 if reprocessing begins in 1989. The current MRS mission does not include any responsibility for storage of defense wastes.

TRUW currently represents a small percentage of the total waste volume, but the commercial component of that volume is expected to increase nearly 50-fold from 1990 to 2020. Current inventories and cumulative projections for commercial and defense TRUW are listed in Table 4.

In the following sections, the existing literature and available data from ongoing research are used to define the operating parameters for SF, HLW, and TRUW and to determine the degradation mechanisms of concern in MRS. Correlations are developed for predicting oxidation of irradiated UO₂ and stress rupture of fuel cladding under thermal histories anticipated in dry storage. Areas where data are lacking are identified, and recommendations are made for obtaining the needed data.

3.2 SPENT FUEL

In the United States, SF comes from pressurized water reactors (PWRs) and boiling water reactors (BWRs). Of the ~30,000 SF assemblies in storage in 1981, about one-third were from PWRs and two-thirds were from BWRs (DOE 1982a). The projected accumulation of SF assemblies, assuming no future reprocessing, is shown in Table 5 for both types of reactors. The relative numbers of assemblies expected in each mode of storage are shown in Figure 1. In 1981, ~95% of the SF assemblies contained fuel rods with Zircaloy cladding while the rest had SS cladding (Bailey et al. 1982). Additional SF with SS-clad fuel will be discharged because three reactors--San Onofre (PWR), Connecticut Yankee (PWR), and La Crosse (BWR)--continue to use SS cladding (Johnson, Gilbert, and Guenther 1982). Examples of typical PWR and BWR assemblies and a typical fuel rod are shown in Figure 2. Typical parameters for the various rod and assembly designs

TABLE 2. Current Inventories and Annual and Cumulative Projections of HLW Stored in Tanks, Bins, and Capsules (DOE 1982a)

End of Calendar Year	Volume, 10^3 m^3		Radioactivity, 10^6 Ci		Thermal Power, 10^3 W	
	Annual Rate ^(a)	Accumu- lation	Annual Rate ^(a)	Accumu- lation	Annual Rate ^(a)	Accumu- lation
DOE/Defense						
1981	15.5	300.6	696.1	1579.4	2763.8	5373.5
1985	13.6	292.8	651.8	1593.0	2633.2	5418.3
1990	16.9	310.7	792.1	1952.9	3101.2	6519.8
1995	6.0	299.6	665.3	1773.5	2672.9	5921.1
2000	13.3	302.8	707.4	1809.1	2796.8	6042.7
2005	12.2	299.6	712.8	1846.8	2812.7	6156.1
2010	10.1	300.7	734.7	1914.6	2877.2	6352.8
2015	6.5	300.3	670.4	1895.1	2687.9	6291.8
2020	8.6	305.3	719.1	1968.5	2831.2	6505.1
Commercial						
1981		2.2		39.4		114.9
1985		2.2		35.5		103.0
Total						
1981	15.5	302.8	696.1	1618.8	2763.8	5488.5
1985	13.6	295.0	651.8	1628.5	2633.2	5521.4
1990	16.9	310.7	792.1	1952.9	3101.2	6519.8
1995	6.0	299.6	665.3	1773.5	2672.9	5921.1
2000	13.3	302.8	707.4	1809.1	2796.8	6042.7
2005	12.2	299.6	712.8	1846.8	2812.7	6156.1
2010	10.1	300.7	734.7	1914.6	2877.2	6352.8
2015	6.5	300.3	670.4	1895.1	2687.9	6291.8
2020	8.6	305.3	719.1	1968.5	2831.2	6505.1

(a) Annual rate is for year indicated.

TABLE 3. Annual and Cumulative Projections of HLW Glass Acceptable for Repositories (DOE 1982a)

End of Calendar Year	Volume, m ³		Radioactivity, 10 ⁶ Ci		Thermal Power, 10 ³ W	
	Annual Rate ^(a)	Accumu- lation	Annual Rate	Accumu- lation	Annual Rate	Accumu- lation
DOE/Defense ^(b)						
1990	312.7	312.7	15.0	15.0	43.0	43.0
1995	321.0	1,902.8	15.4	85.5	44.1	245.3
2000	330.1	3,536.5	15.9	149.9	45.4	430.7
2005	270.8	5,145.8	13.0	205.7	37.2	591.7
2010	269.5	6,493.2	12.9	243.6	37.0	701.2
2015	269.5	7,840.8	12.9	277.5	37.0	798.7
2020	269.5	9,188.5	12.9	307.6	37.0	885.4
Commercial						
1990	163.4	302.1	269.6	445.8	855.9	1,420.2
1995	138.8	996.4	344.6	1,914.0	1,121.5	6,253.0
2000	277.5	1,828.9	666.7	3,602.7	2,179.4	11,789.7
2005	416.3	3,493.9	1,013.4	6,988.7	3,305.4	22,902.8
2010	555.0	5,991.4	1,511.3	12,194.5	4,914.7	39,909.5
2015	555.0	8,766.4	1,586.9	18,007.7	5,170.7	58,846.3
2020	555.0	11,541.3	1,653.8	23,429.3	5,415.0	76,520.3
Total						
1990	476.1	614.9	284.6	460.9	898.9	1,463.1
1995	459.8	2,899.2	360.0	1,999.5	1,165.6	6,498.3
2000	607.6	5,365.4	682.5	3,752.5	2,224.7	12,220.4
2005	687.1	8,639.7	1,026.4	7,194.4	3,342.6	23,494.6
2010	824.5	12,484.6	1,524.3	12,438.1	4,951.7	40,610.8
2015	824.5	16,607.2	1,599.9	18,285.1	5,207.8	59,645.0
2020	824.5	20,729.8	1,666.8	23,736.9	5,452.0	77,405.7

(a) Annual rate is for year indicated.

(b) Incorporation of HLW into glass begins in 1989 for future commercial HLW and in 1990 for NFS and DOE/defense HLW.

TABLE 4. Current Inventories and Annual and Cumulative Projections of TRUW (DOE 1982a)

End of Calendar Year	Volume, 10 ³ m ³		Radioactivity, 10 ⁶ Ci		Thermal Power, 10 ³ W		Mass of TRU Elements, kg	
	Annual Rate ^(a)	Accumu- lation	Annual Rate	Accumu- lation	Annual Rate	Accumu- lation	Annual Rate	Accumu- lation
DOE/Defense ^(b)								
1981	4.9	364.6	0.1	1.5	2.5	36.7	74.4	1,903.0
1985	5.9	385.5	0.2	2.0	4.8	53.8	135.0	2,350.2
1990	3.8	405.6	0.2	2.8	5.1	77.5	100.1	2,863.1
1995	3.8	424.6	0.2	3.5	5.1	100.0	100.1	3,363.3
2000	3.8	443.6	0.2	4.2	5.1	121.8	100.1	3,863.4
2005	3.8	462.6	0.2	4.8	5.1	142.7	100.1	4,363.6
2010	3.8	481.7	0.2	5.4	5.1	163.0	100.1	4,863.7
2015	3.8	500.7	0.2	6.0	5.1	182.5	100.1	5,363.7
2020	3.8	519.7	0.2	6.6	5.1	201.3	100.1	5,863.8
Commercial ^(c)								
1990	2.3	4.5	2.3	3.8	18.3	28.7	45.1	83.2
1995	2.3	15.9	3.7	16.8	30.7	125.5	53.3	345.5
2000	4.5	29.5	6.8	29.1	54.1	205.5	105.7	663.3
2005	6.8	56.8	10.5	55.1	84.5	378.2	159.4	1,298.7
2010	9.1	97.7	19.0	100.8	162.3	702.9	218.6	2,261.2
2015	9.1	143.2	22.0	155.3	184.8	1093.5	219.7	3,356.7
2020	9.1	188.6	23.2	200.2	198.4	1399.4	225.2	4,473.5
Total								
1981	4.9	364.6	0.1	1.5	2.5	36.7	74.4	1,903.0
1985	5.9	385.5	0.2	2.0	4.8	53.8	135.0	2,350.2
1990	6.1	410.2	2.5	6.6	23.4	106.2	145.2	2,946.3
1995	6.1	440.5	3.9	20.3	35.8	225.6	153.3	3,708.8
2000	8.3	473.2	6.9	33.3	59.2	327.3	205.7	4,526.7
2005	10.6	519.5	10.7	59.9	89.6	520.9	259.5	5,662.2
2010	12.9	579.4	19.2	106.2	167.4	865.9	318.7	7,124.9
2015	12.9	643.8	22.2	161.3	189.9	1,275.9	319.7	8,720.4
2020	12.9	708.3	23.4	206.8	203.5	1,600.7	325.3	10,337.3

(a) Annual rate is for year indicated.

(b) Includes buried and retrievably stored (contact and remotely handled) TRU waste. TRU waste from remedial action programs is not included.

(c) Assumes that fuel reprocessing begins in 1989.

TABLE 5. Projected Number of Fuel Assemblies by Reactor Type Assuming No Future Reprocessing (DOE 1982a)^(a)

End of Calendar Year	BWR		PWR		Total	
	Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
1981		18,685		10,930		29,615
1982	2,718	21,403	1,727	12,657	4,445	34,060
1983	3,104	24,507	2,421	15,078	5,525	39,585
1984	3,128	27,635	2,726	17,804	5,854	45,439
1985	4,982	32,527	3,073	20,877	7,965	53,404
1986	5,332	37,859	3,781	24,658	9,113	62,517
1987	5,020	42,879	3,923	28,581	8,943	71,460
1988	5,876	48,755	4,279	32,860	10,155	81,615
1989	6,040	54,795	4,896	37,756	10,936	92,551
1990	5,627	60,422	4,858	42,614	10,485	103,036
1991	6,528	66,950	4,583	47,197	11,111	114,147
1992	7,055	74,005	5,182	52,379	12,237	126,384
1993	7,077	81,082	5,067	57,446	12,144	138,528
1994	7,825	88,907	4,775	62,221	12,600	151,128
1995	7,872	96,779	4,957	67,178	12,829	163,957
1996	7,164	103,943	5,122	72,300	12,286	176,243
1997	8,483	112,426	4,793	77,093	13,276	189,519
1998	8,223	120,649	5,602	82,695	13,825	203,344
1999	8,255	128,904	5,585	88,280	13,840	217,184
2000	9,280	138,184	5,525	93,805	14,805	231,989
2001	9,587	147,771	6,215	100,020	15,802	247,791
2002	8,607	156,378	6,724	106,744	15,331	263,122
2003	11,004	167,382	6,191	112,935	17,195	280,317
2004	10,443	177,825	7,113	120,048	17,556	297,873
2005	10,199	188,024	7,076	127,124	17,275	315,148
2006	11,668	199,692	7,462	134,586	19,130	334,278
2007	13,399	213,091	8,474	143,060	21,873	356,151
2008	12,611	225,702	8,840	151,900	21,451	377,602
2009	15,440	241,142	8,898	160,798	24,338	401,940
2010	12,323	253,465	9,177	169,975	21,500	423,440
2011	12,975	266,440	9,154	179,129	22,129	445,569
2012	14,624	281,064	8,902	188,031	23,526	469,095
2013	14,435	295,499	9,092	197,123	23,527	492,622
2014	14,923	310,422	9,504	206,627	24,427	517,049
2015	17,040	327,462	9,336	215,963	26,376	543,425
2016	15,191	342,653	9,204	225,167	24,395	567,820
2017	15,387	358,040	9,568	234,735	24,955	592,775
2018	17,936	375,976	9,553	244,288	27,489	620,264
2019	14,391	390,367	9,549	253,837	23,940	644,204
2020	16,255	406,622	9,654	263,491	25,909	670,113

(a) Based on 167.4 GWe-installed in the year 2000 and 280.7 GWe-installed in the year 2020.

FIGURE 1. Distribution of Projected U.S. LWR Spent Fuel Inventory in Various Storage Modes (based on data from DOE/RLO 1983; Engel and White 1982)

fabricated by U.S. LWR vendors are shown in Table 6 (Bailey and Tokar 1982), and data for the three reactors using SS cladding are presented in Table 7 (Simnad 1971; Viebrock and Vogt 1974).

Present designs for SF storage in an MRS facility include enclosing one to three fuel assemblies in a carbon steel canister (DOE 1981; General Atomic 1982; Rasmussen 1982). Typically, one PWR or two to three BWR assemblies would be placed in one canister, although some canisters may contain consolidated PWR rods. Each canister is designed with a wall thickness sufficient to accommodate a minimum of 100 years in the facility assuming a uniform corrosion rate of 0.001 in./yr. A representative design for a SF canister is illustrated in Figure 3.

a) LWR Fuel Rod

b) PWR Fuel Assembly

c) BWR Fuel Assembly

FIGURE 2. Typical Geometries for an LWR Fuel Rod and PWR and BWR Fuel Assemblies (Johnson, Gilbert, and Guenther 1982)

TABLE 6. Typical Fuel Assembly Parameters (Bailey and Tokar 1982)

	VENDOR	B&W	B&W	C-E	C-E	W	W	W	ENC	ENC	GE	GE	GE
	Fuel Rod Array	15x15	17x17	14x14	16x16	14x14	15x15	17x17	15x15	8x8	7x7	8x8	8x8 R
	Reactor Type	PWR	PWR	PWR	PWR	PWR	PWR	PWR	PWR	BWR	BWR	BWR	BWR
	Assemblies per Core	177	205	217	177	121	193	193	193	560	764	560	560
	Fuel Rod Locations Per Assembly	225	289	196	256	196	225	289	225	64	49	64	64
	Fuel Rods Per Assembly	208	264	176	236	179	204	264	204	60	49	63	62
	Empty Locations Per Assembly	17	25	5	5	17	21	25	21	4	NONE	1	2
	Rod Pitch, mm (in.)	14.4 (0.568)	12.8 (0.502)	14.7 (0.580)	12.9 (0.5063)	14.1 (0.556)	14.3 (0.563)	12.6 (0.496)	14.3 (0.563)	16.3 (0.842)	18.7 (0.738)	16.3 (0.640)	16.3 (0.640)
	System Pressure, MPa (psia)	15.2 (2200)	15.5 (2250)	15.5 (2250)	15.5 (2250)	15.5 (2250)	15.5 (2250)	15.5 (2250)	15.5 (2250)	7.14 (1035)	7.14 (1035)	7.14 (1035)	7.14 (1035)
16	Core Average Power Density, kW/liter	91.4	107.3	78.5	96.4	95.6	98.1	104.7	98.1	40.57	50.732	50.51	49.15
	Average LHGR, (a) kW/M (kW/ft)	20.3 (6.20)	18.8 (5.73)	20.0 (6.09)	17.5 (5.34)	20.3 (6.20)	22.0 (6.70)	17.8 (5.44)	22.0 (6.70)	15.2 (4.63)	23.1 (7.049)	17.9 (5.45)	17.7 (5.38)
	Axial Peak LHGR, in an Average Rod, kW/M (kW/ft)	24.41 (7.44)	22.57 (6.88)	24.00 (7.31)	21.00 (6.41)	24.36 (7.44)	26.40 (8.04)	21.36 (6.53)	26.40 (8.04)	18.24 (6.02)	27.72 (9.16)	21.48 (7.09)	21.24 (6.99)
	Max. Peak LHGR, kW/M (kW/ft)	53.0 (16.16)	49.9 (15.20)	53.5 (16.3)	42.7 (13.0)	56.8 (17.3)	61.7 (18.8)	44.6 (13.6)	51.9 (15.83)	47.6 (14.5)	60.2 (18.35)	44.0 (13.4)	44.0 (13.4)
	Max. Fuel Temp., °C (°F)	2340 (4245)	2090 (4155)	2140 (3890)	1880 (3420)	2260 (4100)	2340 (4250)	1870 (3400)	2200 (3997)	2040 (3700)	2440 (4430)	1830 (3325)	1890 (3435)
	Core Average Enrichment, wt% ²³⁵ U	3.10(b)	3.15(b)	2.35	2.36	2.90	2.80	2.60	3.02	2.65	2.19	1.80	1.99
	Max. Local Burnup, (c) MWd/MTU GJ/kgU (d)	55,000 4752	55,000 4752	50,000 4320	55,000 4752	50,000 4320	50,000 4320	50,000 4320	47,500 4104	35,000 3024	40,000 3456	40,000 3456	45,000 3888
	Cladding Material	Zry-4	Zry-4	Zry-4	Zry-4	Zry-4	Zry-4	Zry-4	Zry-4	Zry-2	Zry-2	Zry-2	Zry-2

TABLE 6. (Contd)

VENDOR	B&W	B&W	C-E	C-E	W	W	W	ENC	ENC	GE	GE	GE
Fuel Rod Length, m (in.)	3.90 (153.7)	3.88 (152.7)	3.71 (145.9)	4.09 (161.0)	3.87 (152.4)	3.80 (149.7)	3.85 (151.6)	3.86 (152.0)	3.99 (156.9)	4.09 (161.1)	4.09 (161.1)	4.20 (185.4)
Active Fuel Height, m (in.)	3.602 (141.8)	3.632 (143)	3.47 (136.7)	3.81 (150)	3.66 (144)	3.66 (144)	3.65 (143.7)	3.66 (144)	3.66 (144)	3.66 (144)	3.71 (146)	3.81 (150)
Plenum Length, m (in.)	0.298 (11.72)	0.242 (9.52)	0.22 (8.6)	0.25 (10.00)	0.18 (6.99)	0.21 (8.2)	0.16 (6.3)	0.17 (6.8)	0.27 (10.63)	0.41 (16.0)	0.36 (14.0)	0.25 (10.0)
Fuel Rod OD, mm (in.)	10.922 (0.430)	9.627 (0.379)	11.18 (0.440)	9.70 (0.382)	10.72 (0.422)	10.72 (0.422)	9.50 (0.374)	10.77 (0.424)	12.74 (0.5015)	14.30 (0.563)	12.52 (0.493)	12.27 (0.483)
Cladding ID, mm (in.)	9.576 (0.377)	8.407 (0.331)	9.86 (0.388)	8.43 (0.332)	9.48 (0.3734)	9.48 (0.3734)	8.36 (0.329)	9.25 (0.364)	10.91 (0.4295)	12.68 (0.499)	10.80 (0.425)	10.64 (0.419)
Cladding Thickness, mm (in.)	0.673 (0.0265)	0.610 (0.0240)	0.660 (0.026)	0.635 (0.025)	0.617 (0.0243)	0.617 (0.0243)	0.572 (0.0225)	0.762 (0.030)	0.914 (0.036)	0.813 (0.032)	0.864 (0.034)	0.813 (0.032)
Diametral Gap(e), micron (mil)	213.4 (8.4)	198.1 (7.8)	216 (8.5)	178 (7.0)	190 (7.5)	190 (7.5)	165 (6.5)	190 (7.5)	254 (10.0)	305 (12.0)	229 (9.0)	229 (9.0)
Fuel Pellet Diameter, mm (in.)	9.362 (0.3686)	8.209 (0.3232)	9.64 (0.3795)	8.26 (0.325)	9.29 (0.3659)	9.29 (0.3659)	8.19 (0.3225)	9.06 (0.3565)	10.66 (0.4195)	12.37 (0.487)	10.57 (0.416)	10.41 (0.410)
Fuel Pellet Length, mm (in.)	15.240 (0.600)	9.525 (0.375)	16.51 (0.650)	9.91 (0.390)	15.24 (0.600)	15.24 (0.600)	13.46 (0.530)	6.93 (0.273)	8.13 (0.320)	12.70 (0.500)	10.67 (0.420)	10.41 (0.410)
Fuel Pellet Density, %TD(f)	95	95	94.75	95	94	95	95	94	95	95	95	95

(a) LHGR = Linear heat generation rate.

(b) Reload batch average enrichment.

(c) MWd/MTU = number of megawatt days of thermal energy released by fuel containing one metric ton (10^3 kg) of heavy-metal atoms (e.g., U = uranium).

(d) GJ/kgU = gigajoule/kilogram of heavy metal (e.g., U = uranium).

(e) Diametral gap = cladding ID - pellet diameter.

(f) Theoretical density (TD) of stoichiometric UO_2 is 10.96 g/cm^3 .

TABLE 7. Fuel Assembly Parameters for Reactors with SS Cladding
(Simnad 1971; Viebrock and Vogt 1974)

Parameter	San Onofre	Connecticut Yankee	La Crosse
Fuel rod array	14 x 14	15 x 15	10 x 10
Reactor type	PWR	PWR	BWR
Assemblies per core	157	157	72
Fuel rods per assembly	180	204	100
Empty locations per assembly	26	21	0
Rod pitch, in. (mm)	0.556 (14.1)	0.563 (14.3)	--
Coolant pressure, psig (MPa)	2065 (14.2)	2150 (14.8)	1350 (9.3)
Average core enrichment, wt% ²³⁵ U	3.5	3.2	3.6
Refuel burnup, MWd/MTHM	13,500	20,000	15,000
Cladding material	304 SS	304 SS	348H SS
Rod length, in. (m)	126.686 (3.22)	126.686 (3.22)	--
Active fuel height, in. (m)	120.0 (3.05)	120.0 (3.05)	81.9 (2.08)
Plenum length, in. (mm)	5.312 (134.92)	5.312 (134.92)	--
Fuel rod OD ^(a) , in. (mm)	0.422 (10.72)	0.422 (10.72)	0.390 (9.91)
Cladding ID, in. (mm)	0.389 (9.88)	0.389 (9.88)	0.350 (8.89)
Cladding thickness, in. (mm)	0.0165 (0.42)	0.0165 (0.42)	0.02 (0.51)
Diametral gap, mils (μm)	5.5 (140)	5.5 (140)	5.9 (150)
Fuel pellet OD, in. (mm)	0.3835 (9.7)	0.3835 (9.7)	0.3445 (8.75)
Fuel pellet length, in. (mm)	0.600 (15.2)	0.600 (15.2)	1.024 (26)
Fuel pellet density, % TD	-	94	--

(a) OD - outside diameter; ID - inside diameter.

3.2.1 MRS Operating Parameters for SF

Factors that affect the safety and retrievability of SF in MRS include initial characteristics of the fuel and storage conditions at the facility. The initial characteristics are determined by physical and chemical attributes such as crystalline makeup, thermal and radiation characteristics, and mechanical properties. Reactor power histories, subsequent handling and transportation operations, and dry storage also affect the initial characteristics of SF

FIGURE 3. Canister for an Open-Field Dry Well MRS Concept (General Atomic 1982)

at an MRS facility. Storage conditions at the MRS facility affect SF behavior through the heat transfer ability of the waste package, the choice of cover gas, and the effect that the surrounding media has on storage temperatures and

chemical reactivity. Safety and retrievability are also affected by transportation and handling operations at the MRS facility; however, these operations are not directly assessed in this review.

Initial Characteristics

The following discussion on initial characteristics of SF upon arrival at an MRS facility consists of: 1) as-fabricated fuel and rod design and characteristics, 2) crud on the cladding, 3) postirradiation condition of the fuel and typical burnups, 4) decay heat, 5) reactor-induced defective fuel rods, 6) incipiently defective rods, 7) effect of dry storage on initial characteristics, 8) effect of handling and transportation on fuel rod integrity, and 9) characteristics of SS fuel rods.

As-Fabricated Characteristics. SF may arrive at an MRS facility after water pool or interim dry storage of possibly up to several decades (Bailey et al. 1982; Johnson, Gilbert, and Guenther 1982). Regardless of the length or previous method of storage, the as-fabricated properties of SF rods are similar. Properties for unirradiated Zircaloy cladding and UO_2 fuel are listed in Tables 8 and 9, respectively. A Zircaloy fuel rod is typically 3.9 to 4.2 m long with an active fuel length of ~ 3.7 m. Fuel pellets are 8.2 to 12.4 mm in diameter, depending on reactor type, with typical densities of 95% of theoretical density (TD) (10.96 g/cm^3). The number of fuel rods in an assembly depends on the reactor type and when the assembly was designed: more recently designed assemblies have more rods (as indicated in Table 6). Typically, a PWR assembly has 0.46 MTHM and a BWR assembly has 0.18 MTHM. Rods are pressurized at room temperature with helium to 15 to 30 atm (1.5 to 3 MPa) for PWRs and 1 to 6 atm (0.1 to 0.6 MPa) for BWRs. Fission gas increases pressures in a PWR rod to as high as 65 atm (6.6 MPa). Typical end-of-life (EOL) room temperature pressures are 30 to 40 atm (3 to 4 MPa) for PWR fuel and 15 atm (1.5 MPa) for BWR fuel (Johnson, Gilbert, and Guenther 1982).

Crud. Corrosion products deposit on the cladding during reactor operation and accumulate to roughly 0.2 to 1 kg/BWR assembly and 0.15 kg/PWR assembly (Johnson, Gilbert, and Guenther 1982). Crud thicknesses range from 0.001 to

TABLE 8. Physical Properties of Zircaloy-2 and Zircaloy-4 (ASTM 1978)

Density, kg/m ³ (lb/in. ³)	6560 (0.237)
Hardness annealed, Rockwell Hardness B Scale	170 to 187 87 to 95
Electrical resistivity	74 $\mu\text{m}/\text{cm}$
Elastic modulus	
Tension, MPa (psi)	96,530 (14,000,000)
Torsion or shear, MPa (psi)	35,854 (5,200,000)
Impact	
Charpy V-notch at room temperature, ft/lb	7 to 15
Coefficient of thermal expansion, in./in./°F	
At 27 to 100°C (80 to 212°F)	3.24×10^{-6}
At 27 to 538°C (80 to 1000°F)	3.6×10^{-6}
Crystal structure	Hexagonal close packed a = 3.2312; c = 5.1477

TABLE 9. Properties of Uranium Dioxide (Greene 1980)

Melting point	2840°C (5144°F)
Crystal structure	Face centered cubic
Lattice parameter	5.468 Å
Theoretical density	10.96 g/cm ³
Thermal conductivity	0.02 cal/s-cm-°C
Thermal expansion coefficient (per °C)	1×10^{-5} (0 to 1000°C)
Fracture strength	69.0 MPa (10,000 psi)
Modulus of elasticity	172×10^3 MPa (25×10^6 psi)
Specific heat	0.0590 Btu/lb-°F

0.003 cm (General Atomic 1982). These values can vary significantly as a result of reactor and water pool chemistry or dry storage conditions. Crud may be loose or tenacious. There is some evidence that dry conditions at elevated temperatures may contribute to crud spallation, but there is no systematic assessment of conditions that promote spallation (Johnson, Gilbert, and Guenther 1982).

Postirradiation Condition. After irradiation, the chemical makeup of SF becomes more complex. Cladding ductility decreases as a result of irradiation,

but strength increases. Along with the original UO_2 , the fuel pellet contains gaseous fission products, solid fission products, metallic inclusions, decay products, and reaction products between the cladding and fission products. The fuel pellet is no longer a solid cylinder; it is cracked into many pieces that are closely spaced depending on fuel behavior and original gap sizes. For example, in CANDU fuel with a burnup of 8000 MWd/MTHM, 98 wt% of the fuel particles were larger than 2 mm in diameter and 0.5 wt% were smaller than 100 μm (Johnson 1982). For fuel with a burnup of 28,000 MWd/MTHM (H. B. Robinson reactor), 86 wt% of the fuel particles had diameters from 2 to 6.73 mm and less than 0.08 wt% had diameters smaller than 0.1 mm (Katayama, Bradley, and Harvey 1980). Aside from the isotopic changes that result from decay, the amount of volatile fission products in the free void volume of the fuel rod remains constant after discharge as long as the fuel temperature remains below 1000°C (Einziger and Himes 1982). The total nuclide inventory for all SF stored as of 1981 is listed in Table 10. Burnups of 25,000 to 35,000 MWd/MTHM are typical for current assemblies; however, burnups have ranged from 5,400 to 23,200 MWd/MTHM for BWRs since the first discharge in 1969 and from 17,400 to 29,500 MWd/MTHM for PWRs since the first discharge in 1970 (DOE 1982a). Thus, the relative amount of each nuclide will vary from rod to rod depending on the specific burnup and power history.

Decay Heat. The decay heat and fuel temperature during storage depend primarily on fuel enrichment, discharge burnup, power history, and age. Temperatures also depend on the heat transfer characteristics of the waste package. For a typical PWR burnup of 33,000 MWd/MTHM and a specific power of 37.5 MW/MTHM, the decrease in decay heat with time can be approximated as shown in Figure 4, using an equation developed by Malbrain, Lester, and Deutch (1981). After ~20 years, the decay heat and, thus, the rod temperature will decrease very little because of the much smaller absolute decrease in heat generation. The much slower decrease in decay heat is shown more distinctly in Figure 5, which is taken from Johnson, Gilbert, and Guenther (1982) and is calculated for a range of typical PWR and BWR burnups. The distribution of decay heat for U.S. LWR spent fuel discharged as of January 1983 and projected to 1986 is shown in Figure 6. The time in wet storage for LWR fuel discharged as of

TABLE 10. Inventory of Nuclides in LWR Spent Fuel at the End of 1981^(a) (DOE 1982a)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Hydrogen	Stable	5.474E+03	3.927E+04	0.0	0.0	0.0	0.0
Hydrogen	3	8.249E+01	4.145E+02	7.963E+05	4.001E+06	2.681E+01	1.347E+02
Helium		2.613E+03	1.441E+04	0.00	0.00	0.00	0.00
Lithium	Stable	1.260E+03	8.651E+03	0.00	0.00	0.00	0.00
Beryllium	Stable	1.018E+00	5.781E+00	0.00	0.00	0.00	0.00
Boron	Stable	1.149E+03	7.822E+03	0.00	0.00	0.00	0.00
Carbon	Stable	2.062E+05	1.421E+06	0.00	0.00	0.00	0.00
Carbon	14	4.326E+02	2.494E+03	1.929E+03	1.112E+04	5.656E-01	3.261E+00
Nitrogen	Stable	1.422E+05	9.844E+05	0.00	0.00	0.00	0.00
Oxygen	Stable	1.551E+08	1.062E+09	0.00	0.00	0.00	0.00
Fluorine	Stable	1.232E+04	8.428E+04	0.00	0.00	0.00	0.00
Neon	Stable	4.302E-01	2.426E+00	0.00	0.00	0.00	0.00
Sodium	Stable	1.723E+04	1.179E+05	0.00	0.00	0.00	0.00
Sodium	24	1.814E-06	1.814E-06	1.580E+01	1.580E+01	4.378E-01	4.378E-01
Magnesium	Stable	2.339E+03	1.596E+04	0.00	0.00	0.00	0.00
Aluminum	Stable	9.377E+04	6.196E+05	0.00	0.00	0.00	0.00
Silicon	Stable	5.511E+05	3.799E+06	0.00	0.00	0.00	0.00
Phosphorus	Stable	2.589E+05	1.652E+06	0.00	0.00	0.00	0.00
Phosphorus	32	1.401E-01	1.401E-01	4.000E+04	4.000E+04	4.055E+02	4.055E+02
Sulfur	Stable	3.079E+04	2.164E+05	0.00	0.00	0.00	0.00
Chlorine	Stable	5.723E+03	3.959E+04	0.00	0.00	0.00	0.00
Argon	Stable	4.131E+02	2.353E+03	0.00	0.00	0.00	0.00
Potassium	Stable	1.772E+00	1.001E+01	0.00	0.00	0.00	0.00
Calcium	Stable	2.299E+03	1.573E+04	0.00	0.00	0.00	0.00
Scandium	Stable	1.155E-01	6.538E-01	0.00	0.00	0.00	0.00
Titanium	Stable	9.571E+04	6.237E+05	0.00	0.00	0.00	0.00
Vanadium	Stable	1.912E+04	1.244E+05	0.00	0.00	0.00	0.00
Vanadium	50	5.920E+01	3.736E+02	1.059E-11	6.681E-11	1.167E-13	7.366E-13
Chromium	Stable	1.258E+07	8.601E+07	0.00	0.00	0.00	0.00
Chromium	51	3.975E+01	3.976E+01	3.675E+06	3.675E+06	7.864E+02	7.865E+02
Manganese	Stable	1.052E+06	7.274E+06	0.00	0.00	0.00	0.00
Manganese	54	8.976E+01	1.577E+02	6.948E+05	1.221E+06	3.460E+03	6.079E+03
Iron	Stable	3.833E+07	2.644E+08	0.00	0.00	0.00	0.00
Iron	55	2.334E+03	7.543E+03	5.837E+06	1.887E+07	8.027E+03	2.594E+04

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Iron	59	1.521E+00	1.526E+00	7.482E+04	7.510E+04	6.977E+02	7.002E+02
Cobalt	Stable	9.121E+04	6.160E+05	0.00	0.00	0.00	0.00
Cobalt	58	4.801E+01	4.930E+01	1.528E+06	1.569E+06	9.147E+03	9.393E+03
Cobalt	60	6.423E+03	2.622E+04	7.265E+06	2.965E+07	1.120E+05	4.572E+05
Nickel	Stable	1.166E+07	7.711E+07	0.00	0.00	0.00	0.00
Nickel	59	5.867E+04	3.262E+05	4.445E+03	2.472E+04	2.830E+01	1.574E+02
Nickel	63	1.004E+04	5.415E+04	6.195E+05	3.341E+06	2.460E+02	1.327E+03
Copper	Stable	2.029E+04	1.372E+05	0.00	0.00	0.00	0.00
Zinc	Stable	4.635E+04	3.171E+05	0.00	0.00	0.00	0.00
Zinc	65	1.156E+01	1.786E+01	9.529E+04	1.471E+05	3.339E+02	5.156E+02
Gallium	Stable	3.399E+01	1.925E+02	0.00	0.00	0.00	0.00
Germanium	Stable	5.992E+02	3.411E+03	0.00	0.00	0.00	0.00
Arsenic	Stable	1.890E+02	1.075E+03	0.00	0.00	0.00	0.00
Selenium	Stable	4.622E+04	2.648E+05	0.00	0.00	0.00	0.00
Selenium	79	5.548E+03	3.164E+04	3.866E+02	2.205E+03	9.626E-02	5.489E-01
Bromine	Stable	2.023E+04	1.163E+05	0.00	0.00	0.00	0.00
Krypton	Stable	3.149E+05	1.818E+06	0.00	0.00	0.00	0.00
Krypton	81	2.131E-02	1.086E-01	4.483E-04	2.284E-03	3.720E-07	1.895E-06
Krypton	85	2.188E+04	1.072E+05	8.589E+06	4.208E+07	1.287E+04	6.303E+04
Rubidium	Stable	9.160E+04	5.280E+05	0.00	0.00	0.00	0.00
Rubidium	86	1.349E+00	1.349E+00	1.099E+05	1.099E+05	4.964E+02	4.964E+02
Rubidium	87	2.278E+05	1.319E+06	1.994E-02	1.154E-01	1.666E-05	9.649E-05
Strontium	Stable	3.262E+05	1.889E+06	0.00	0.00	0.00	0.00
Strontium	89	5.684E+03	5.723E+03	1.652E+08	1.664E+08	5.711E+05	5.751E+05
Strontium	90	4.952E+05	2.696E+06	6.757E+07	3.679E+08	7.842E+04	4.270E+05
Yttrium	Stable	3.958E+05	2.252E+06	0.00	0.00	0.00	0.00
Yttrium	90	1.255E+02	6.775E+02	6.834E+07	3.688E+08	3.787E+05	2.044E+06
Yttrium	91	1.032E+04	1.046E+04	2.533E+08	2.568E+08	9.096E+05	9.222E+05
Zirconium	Stable	4.023E+08	2.896E+09	0.00	0.00	0.00	0.00
Zirconium	93	7.836E+05	4.536E+06	1.970E+03	1.140E+04	2.288E-01	1.325E+00
Zirconium	95	1.917E+04	1.956E+04	4.121E+08	4.204E+08	2.087E+06	2.129E+06
Niobium	Stable	5.900E+05	3.811E+06	0.00	0.00	0.00	0.00
Niobium	93M	5.724E-01	7.576E+00	1.619E+02	2.142E+03	2.868E-02	3.795E-01
Niobium	94	6.018E+03	3.319E+04	1.128E+03	6.220E+03	1.149E+01	6.338E+01

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Niobium	95	1.655E+04	1.701E+04	6.472E+08	6.654E+08	3.105E+06	3.192E+06
Niobium	95M	8.330E+00	8.491E+00	3.174E+06	3.235E+06	4.401E+03	4.495E+03
Molybdenum	Stable	3.417E+06	1.956E+07	0.00	0.00	0.00	0.00
Technetium	99	7.390E+05	4.243E+06	1.253E+04	7.197E+04	6.285E+00	3.609E+01
Ruthenium	Stable	2.053E+06	1.142E+07	0.00	0.00	0.00	0.00
Ruthenium	103	7.246E+03	7.257E+03	2.339E+08	2.343E+08	7.826E+05	7.839E+05
Ruthenium	106	1.220E+05	2.339E+05	4.084E+08	7.829E+08	2.428E+04	4.655E+04
Rhodium	Stable	4.117E+05	2.367E+06	0.00	0.00	0.00	0.00
Rhodium	103M	6.483E+00	6.494E+00	2.110E+08	2.113E+08	4.857E+04	4.864E+04
Rhodium	106	1.147E-01	2.199E-01	4.084E+08	7.829E+08	3.917E+06	7.509E+06
Palladium	Stable	9.443E+05	4.936E+06	0.00	0.00	0.00	0.00
Palladium	107	2.141E+05	1.140E+06	1.101E+02	5.867E+02	6.529E-03	3.478E-02
Silver	Stable	7.502E+04	4.081E+05	0.00	0.00	0.00	0.00
Silver	108	1.146E-09	6.673E-09	8.425E-01	4.904E+00	3.139E-03	1.827E-02
Silver	108M	3.631E-01	2.113E+00	9.466E+00	5.510E+01	9.214E-02	5.363E-01
Silver	110	8.128E-06	1.232E-05	3.390E+04	5.140E+04	2.436E+02	3.693E+02
Silver	110M	5.363E+02	8.132E+02	2.549E+06	3.865E+06	4.256E+04	6.454E+04
Silver	111	1.021E+01	1.021E+01	1.613E+06	1.613E+06	3.614E+03	3.614E+03
Cadmium	Stable	1.312E+05	7.318E+05	0.00	0.00	0.00	0.00
Cadmium	109	4.036E-01	9.147E-01	1.042E+03	2.363E+03	6.636E-01	1.504E+00
Cadmium	113M	2.424E+02	1.151E+03	5.259E+04	2.496E+05	8.853E+01	4.202E+02
Cadmium	115M	1.237E+01	1.242E+01	3.152E+05	3.162E+05	1.175E+03	1.179E+03
Indium	Stable	1.217E+03	7.538E+03	0.00	0.00	0.00	0.00
Indium	114	3.316E-05	3.338E-05	4.565E+04	4.596E+04	2.096E+02	2.110E+02
Indium	114M	2.061E+00	2.075E+00	4.771E+04	4.803E+04	6.727E+01	6.773E+01
Indium	115	2.572E+03	1.707E+04	1.601E-08	1.062E-07	2.296E-11	1.524E-10
Indium	115M	3.523E-06	3.535E-06	2.234E+01	2.241E+01	4.454E-02	4.469E-02
Tin	Stable	6.592E+06	4.743E+07	0.00	0.00	0.00	0.00
Tin	117M	7.464E+00	7.464E+00	5.951E+05	5.951E+05	1.104E+03	1.104E+03
Tin	119M	1.721E+03	2.754E+03	7.712E+06	1.234E+07	3.986E+03	6.378E+03
Tin	121M	2.310E+01	1.322E+02	1.366E+03	7.820E+03	2.737E+00	1.567E+01
Tin	123	2.347E+02	2.741E+02	1.930E+06	2.254E+06	6.028E+03	7.038E+03
Tin	125	4.870E+00	4.870E+00	5.279E+05	5.279E+05	3.499E+03	3.499E+03
Tin	126	2.626E+04	1.459E+05	7.453E+02	4.140E+03	9.295E-01	5.164E+00

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Antimony	Stable	2.049E+04	1.161E+05	0.00	0.00	0.00	0.00
Antimony	124	1.635E+01	1.658E+01	2.861E+05	2.902E+05	3.798E+03	3.853E+03
Antimony	125	1.504E+04	4.991E+04	1.554E+07	5.155E+07	4.858E+04	1.612E+05
Antimony	126	5.991E-01	5.991E-01	5.010E+04	5.010E+04	9.257E+02	9.257E+02
Antimony	127	3.388E+00	3.388E+00	9.053E+05	9.053E+05	5.372E+03	5.372E+03
Tellurium	Stable	4.437E+05	2.482E+06	0.00	0.00	0.00	0.00
Tellurium	123	9.713E+00	4.983E+01	2.824E-09	1.449E-08	4.225E-13	2.168E-12
Tellurium	123M	9.299E-01	1.053E+00	8.252E+03	9.346E+03	1.202E+01	1.361E+01
Tellurium	125M	2.026E+02	6.901E+02	3.652E+06	1.244E+07	3.069E+03	1.045E+04
Tellurium	127	2.557E+00	2.801E+00	6.752E+06	7.395E+06	9.117E+03	9.986E+03
Tellurium	127M	6.347E+02	7.043E+02	5.989E+06	6.646E+06	3.221E+03	3.575E+03
Tellurium	129	1.881E-01	1.882E-01	3.942E+06	3.944E+06	1.408E+04	1.409E+04
Tellurium	129M	2.008E+02	2.009E+02	6.052E+06	6.055E+06	1.061E+04	1.062E+04
Tellurium	132	2.741E+01	2.741E+01	8.327E+06	8.327E+06	1.650E+04	1.650E+04
Iodine	Stable	5.144E+04	2.850E+05	0.00	0.00	0.00	0.00
Iodine	129	1.722E+05	9.686E+05	3.041E+01	1.711E+02	1.407E-02	7.913E-02
Iodine	131	1.915E+02	1.915E+02	2.375E+07	2.375E+07	8.067E+04	8.067E+04
Xenon	Stable	4.926E+06	2.780E+07	0.00	0.00	0.00	0.00
Xenon	129M	1.046E-03	1.046E-03	1.324E+02	1.324E+02	1.852E-01	1.852E-01
Xenon	131M	9.227E+00	9.227E+00	7.733E+05	7.733E+05	7.440E+02	7.440E+02
Xenon	133	1.611E+02	1.611E+02	3.017E+07	3.017E+07	3.232E+04	3.232E+04
Cesium	Stable	1.067E+06	6.140E+06	0.00	0.00	0.00	0.00
Cesium	134	8.961E+04	2.424E+05	1.160E+08	3.137E+08	1.818E+06	3.193E+06
Cesium	135	2.967E+05	1.673E+06	3.417E+02	1.928E+03	1.140E-01	6.433E-01
Cesium	136	2.620E+01	2.620E+01	1.921E+06	1.921E+06	2.619E+04	2.619E+04
Cesium	137	1.124E+06	5.986E+06	9.780E+07	5.209E+08	1.082E+05	5.762E+05
Barium	Stable	1.278E+06	7.200E+06	0.00	0.00	0.00	0.00
Barium	136M	1.175E-06	1.175E-06	3.165E+05	3.165E+05	3.828E+03	3.828E+03
Barium	137M	1.719E-01	9.158E-01	9.252E+07	4.928E+08	3.633E+05	1.935E+06
Barium	140	9.632E+02	9.632E+02	7.027E+02	7.027E+07	1.961E+05	1.961E+05
Lanthanum	Stable	1.150E+06	6.545E+06	0.00	0.00	0.00	0.00
Lanthanum	138	5.395E+00	3.289E+01	1.036E-07	6.314E-07	1.784E-10	1.088E-09
Lanthanum	140	1.473E+02	1.473E+02	8.203E+07	8.203E+07	1.375E+06	1.375E+06
Cerium	Stable	1.140E+06	6.431E+06	0.00	0.00	0.00	0.00

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Cerium	141	6.761E+03	6.764E+03	1.927E+08	1.927E+08	2.821E+05	2.822E+05
Cerium	142	1.064E+06	6.054E+06	2.554E-02	1.453E-01	0.00	0.00
Cerium	144	2.521E+05	4.242E+05	8.047E+08	1.354E+09	5.337E+05	8.979E+05
Praseodymium	Stable	1.012E+06	5.693E+06	0.00	0.00	0.00	0.00
Praseodymium	143	1.063E+03	1.063E+03	7.159E+07	7.159E+07	1.334E+05	1.334E+05
Praseodymium	144	1.065E+01	1.791E+01	8.047E+08	1.354E+09	5.915E+06	9.950E+06
Praseodymium	144M	5.322E-02	8.953E-02	9.656E+06	1.624E+07	3.304E+03	5.558E+03
Neodymium	Stable	2.541E+06	1.459E+07	0.00	0.00	0.00	0.00
Neodymium	144	9.737E+05	6.376E+06	1.152E-06	7.545E-06	0.00	0.00
Neodymium	147	2.789E+02	2.789E+02	2.241E+07	2.241E+07	5.405E+04	5.405E+04
Promethium	147	1.339E+05	4.608E+05	1.242E+08	4.274E+08	4.455E+04	1.533E+05
Promethium	148	2.245E+01	2.245E+01	3.689E+06	3.690E+06	2.841E+04	2.841E+04
Promethium	148M	2.088E+02	2.093E+02	4.463E+06	4.473E+06	5.658E+04	5.671E+04
Samarium	Stable	4.154E+05	2.370E+06	0.00	0.00	0.00	0.00
Samarium	147	7.154E+04	8.204E+05	1.626E-03	1.865E-02	2.227E-05	2.554E-04
Samarium	148	1.510E+05	7.900E+05	4.561E-08	2.386E-07	5.445E-10	2.848E-09
Samarium	149	3.176E+03	2.155E+04	7.628E-10	5.176E-09	0.00	0.00
Samarium	151	1.352E+04	8.317E+04	3.559E+05	2.189E+06	4.173E+01	2.566E+02
Europium	Stable	1.013E+05	5.474E+05	0.00	0.00	0.00	0.00
Europium	152	3.285E+01	1.779E+02	5.683E+03	3.078E+04	4.308E+01	2.334E+02
Europium	154	3.418E+04	1.424E+05	9.231E+06	3.844E+07	8.257E+04	3.439E+05
Europium	155	1.219E+04	4.617E+04	5.674E+06	2.148E+07	4.127E+03	1.562E+04
Europium	156	1.738E+02	1.738E+02	9.583E+06	9.583E+06	9.889E+04	9.889E+04
Gadolinium	Stable	7.286E+05	5.543E+06	0.00	0.00	0.00	0.00
Gadolinium	152	9.585E+01	1.349E+03	2.088E-09	2.938E-08	2.721E-11	3.828E-10
Gadolinium	153	1.663E+02	2.837E+02	5.866E+05	1.001E+06	5.056E+02	8.626E+02
Terbium	Stable	1.063E+04	6.731E+04	0.00	0.00	0.00	0.00
Terbium	160	8.201E+01	8.534E+01	9.260E+05	9.636E+05	7.542E+03	7.848E+03
Terbium	161	5.327E-01	5.327E-01	6.246E+04	6.246E+04	1.251E+02	1.251E+02
Thallium	Stable	8.772E-10	4.425E-09	0.00	0.00	0.00	0.00
Thallium	207	2.103E-12	4.175E-11	4.007E-04	7.953E-03	1.177E-06	2.335E-05
Thallium	208	3.047E-09	5.083E-08	8.977E-01	1.497E+01	2.113E-02	3.524E-01
Thallium	209	5.081E-15	2.048E-14	2.079E-06	8.378E-06	3.454E-08	1.392E-07
Lead	Stable	1.135E+03	7.766E+03	0.00	0.00	0.00	0.00

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Lead	204	1.583E+01	1.084E+02	1.983E-13	1.357E-12	3.056E-15	2.092E-14
Lead	205	3.323E-02	1.880E-01	1.933E-06	1.094E-05	4.010E-10	2.269E-09
Lead	209	2.121E-11	8.537E-11	9.640E-05	3.880E-04	1.109E-07	4.462E-07
Lead	210	2.846E-08	6.882E-07	2.174E-06	5.256E-05	5.036E-10	1.218E-08
Lead	211	1.627E-11	3.229E-04	4.018E-04	7.975E-03	1.204E-06	2.390E-05
Lead	212	1.797E-06	2.998E-05	2.498E+00	4.168E+01	4.757E-03	7.935E-02
Lead	214	5.635E-13	1.900E-11	1.848E-05	6.233E-04	5.894E-08	1.988E-06
Bismuth	Stable	4.603E+02	3.150E+03	0.00	0.00	0.00	0.00
Bismuth	208	5.338E-03	2.976E-02	2.494E-05	1.390E-04	4.243E-07	2.366E-06
Bismuth	210	4.784E-06	4.785E-06	5.939E-01	5.939E-01	1.369E-03	1.369E-03
Bismuth	210M	3.158E-02	1.787E-01	1.793E-05	1.014E-04	5.628E-07	3.185E-06
Bismuth	211	9.601E-13	1.905E-11	4.018E-04	7.975E-03	1.603E-05	3.181E-04
Bismuth	212	1.705E-07	2.844E-06	2.499E+00	4.168E+01	4.249E-02	7.088E-01
Bismuth	213	4.974E-12	2.005E-11	9.624E-05	3.879E-04	4.046E-07	1.631E-06
Bismuth	214	4.184E-13	1.411E-11	1.848E-05	6.233E-04	2.369E-07	7.988E-06
Polonium	210	4.899E-03	5.855E-03	2.202E+01	2.631E+01	7.058E-01	8.435E-01
Polonium	211	1.178E-17	2.338E-16	1.125E-06	2.233E-05	5.063E-08	1.005E-06
Polonium	212	9.021E-18	1.505E-16	1.601E+00	2.670E+01	8.483E-02	1.415E+00
Polonium	213	7.463E-21	3.008E-20	9.416E-05	3.795E-04	4.765E-06	1.920E-05
Polonium	214	7.284E-20	1.957E-18	2.338E-05	6.281E-04	1.086E-06	2.916E-05
Polonium	215	1.362E-17	2.704E-16	4.018E-04	7.975E-03	1.794E-05	3.560E-04
Polonium	216	7.165E-12	1.196E-10	2.496E+00	4.167E+01	1.022E-01	1.706E+00
Polonium	218	6.534E-14	2.204E-12	1.849E-05	6.234E-04	6.698E-07	2.259E-05
Astatine	217	5.974E-17	2.409E-16	9.620E-05	3.879E-04	4.105E-06	1.655E-05
Radon	218	3.314E-18	3.314E-18	4.902E-06	4.902E-06	2.109E-07	2.109E-07
Radon	219	3.087E-14	6.128E-13	4.018E-04	7.975E-03	1.667E-05	3.309E-04
Radon	220	2.705E-09	4.516E-08	2.496E+00	4.167E+01	9.476E-02	1.582E+00
Radon	222	1.201E-10	4.052E-09	1.849E-05	6.234E-04	6.125E-07	2.066E-05
Francium	221	5.425E-13	2.187E-12	9.620E-05	3.879E-04	3.713E-06	1.497E-05
Francium	223	1.567E-13	2.934E-12	6.064E-06	1.135E-04	1.575E-08	2.948E-07
Radium	222	3.664E-15	3.664E-15	4.902E-06	4.902E-06	1.941E-07	1.941E-07
Radium	223	7.843E-09	1.557E-07	4.018E-04	7.975E-03	1.431E-05	2.840E-04
Radium	224	1.566E-05	2.615E-04	2.496E+00	4.167E+01	8.566E-02	1.430E+00
Radium	225	2.234E-09	9.678E-09	8.763E-05	3.796E-04	6.145E-08	2.662E-07

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Radium	226	1.887E-05	6.331E-04	1.867E-05	6.263E-04	5.389E-07	1.808E-05
Radium	228	1.005E-11	3.388E-10	2.354E-09	7.932E-08	1.814E-13	6.113E-12
Actinium	225	1.657E-09	6.680E-09	9.619E-05	3.878E-04	3.360E-06	1.355E-05
Actinium	227	6.072E-06	1.137E-04	4.394E-04	8.226E-03	2.128E-07	3.983E-06
Actinium	228	1.140E-15	3.546E-14	2.557E-09	7.954E-08	2.210E-11	6.874E-10
Thorium	226	1.826E-13	1.826E-13	4.901E-06	4.901E-06	1.874E-07	1.874E-07
Thorium	227	1.333E-08	2.588E-07	4.097E-04	7.957E-03	1.495E-05	2.904E-04
Thorium	228	3.005E-03	5.050E-02	2.464E+00	4.141E+01	8.059E-02	1.354E+00
Thorium	229	3.607E-04	1.735E-03	7.676E-05	3.691E-04	2.348E-06	1.129E-05
Thorium	230	1.381E+00	2.136E+01	2.789E-02	4.314E-01	7.893E-04	1.221E-02
Thorium	231	4.037E-05	3.325E-04	2.147E+01	1.768E+02	1.205E-02	9.921E-02
Thorium	232	2.032E-01	2.988E+00	2.229E-08	3.278E-07	5.396E-10	7.937E-09
Thorium	234	1.590E-02	1.093E-01	3.683E+02	2.532E+03	1.493E-01	1.027E+00
Protactinium	231	2.394E-01	1.608E+00	1.131E-02	7.597E-02	3.408E-04	2.289E-03
Protactinium	232	6.319E-07	6.319E-07	2.716E-01	2.716E-01	1.776E-03	1.776E-03
Protactinium	233	1.327E-02	7.124E-02	2.756E+02	1.479E+03	6.255E-01	3.357E+00
Protactinium	234	2.397E-07	1.646E-06	4.794E-01	3.292E+00	6.885E-03	4.728E-02
Protactinium	234M	5.361E-07	3.685E-06	3.683E+02	2.532E+03	1.821E+00	1.251E+01
Uranium	230	1.793E-10	1.793E-10	4.895E-06	4.895E-06	1.738E-07	1.738E-07
Uranium	231	1.002E-09	1.002E-09	1.350E-04	1.350E-04	7.126E-08	7.126E-08
Uranium	232	4.254E-01	3.444E+00	9.108E+00	7.374E+01	2.925E-01	2.368E+00
Uranium	233	1.255E+00	9.503E+00	1.215E-02	9.203E-02	3.533E-04	2.675E-03
Uranium	234	1.894E+05	1.369E+06	1.184E+03	8.556E+03	3.411E+01	2.464E+02
Uranium	235	9.779E+06	8.162E+07	2.115E+01	1.765E+02	5.538E-01	4.622E+00
Uranium	236	3.680E+06	2.216E+07	2.382E+02	1.434E+03	6.452E+00	3.886E+01
Uranium	237	1.708E+02	1.708E+02	1.395E+07	1.395E+07	2.639E+04	2.639E+04
Uranium	238	1.095E+09	7.528E+09	3.683E+02	2.532E+03	9.342E+00	6.422E+01
Uranium	240	1.892E-06	1.894E-06	1.753E+00	1.755E+00	1.438E-03	1.439E-03
Neptunium	235	2.602E-03	4.982E-03	3.652E+00	6.993E+00	2.134E-04	4.085E-04
Neptunium	236	3.279E-01	1.652E+00	4.321E-03	2.177E-02	8.717E-06	4.391E-05
Neptunium	236M	2.776E-06	2.776E-06	1.639E+00	1.639E+00	1.295E-03	1.295E-03
Neptunium	237	3.907E+05	2.097E+06	2.755E+02	1.479E+03	8.420E+00	4.521E+01
Neptunium	238	2.818E+00	2.818E+00	7.308E+05	7.308E+05	3.500E+03	3.500E+03
Neptunium	239	3.017E+02	3.019E+02	7.001E+07	7.007E+07	1.692E+05	1.694E+05

TABLE 10. (contd)

Element	Isotope	Inventory, g		Radioactivity, Ci		Thermal Power, W	
		Annual Rate	Accumulation	Annual Rate	Accumulation	Annual Rate	Accumulation
Neptunium	240M	1.670E-08	1.671E-08	1.769E+00	1.770E+00	1.025E-02	1.026E-02
Plutonium	236	8.303E-01	2.473E+00	4.414E+02	1.315E+03	1.536E+01	4.575E+01
Plutonium	237	7.295E-03	7.321E-03	8.818E+01	8.849E+01	8.468E-03	8.498E-03
Plutonium	238	1.165E+05	5.644E+05	1.995E+06	9.666E+06	6.611E+04	3.204E+05
Plutonium	239	5.555E+06	3.616E+07	3.455E+05	2.249E+06	1.065E+04	6.931E+04
Plutonium	240	2.280E+06	1.308E+07	5.197E+05	2.982E+06	1.618E+04	9.287E+04
Plutonium	241	1.243E+06	6.244E+06	1.281E+08	6.435E+08	3.971E+03	1.995E+04
Plutonium	242	4.414E+05	2.162E+06	1.686E+03	8.257E+03	4.979E+01	2.438E+02
Plutonium	243	4.813E-07	4.814E-07	1.253E+00	1.253E+00	1.446E-03	1.446E-03
Plutonium	244	2.346E+01	1.004E+02	4.163E-04	1.781E-03	1.207E-05	5.164E-05
Plutonium	245	3.149E-09	3.149E-09	3.802E-03	3.802E-03	9.014E-06	9.014E-06
Plutonium	246	1.647E-07	1.647E-07	8.060E-03	8.060E-03	6.785E-06	6.785E-06
Americium	239	1.050E-11	1.050E-11	1.158E-05	1.158E-05	2.629E-08	2.629E-08
Americium	240	2.192E-06	2.192E-06	5.637E-01	5.637E-01	3.626E-03	3.626E-03
Americium	241	6.132E+04	1.175E+06	2.105E+05	4.034E+06	6.994E+03	1.340E+05
Americium	242	1.323E-02	4.410E-02	1.070E+04	3.566E+04	1.243E+01	4.144E+01
Americium	242M	6.454E+02	3.226E+03	6.275E+03	3.136E+04	2.479E+00	1.239E+01
Americium	243	8.350E+04	3.750E+05	1.665E+04	7.478E+04	5.353E+02	2.404E+03
Americium	244	1.579E-05	1.579E-05	2.009E+01	2.009E+01	1.053E-01	1.053E-01
Americium	245	7.642E-10	7.642E-10	4.724E-03	4.724E-03	8.765E-06	8.765E-06
Americium	246	2.640E-10	2.640E-10	8.075E-03	8.075E-03	6.519E-05	6.519E-05
Curium	242	5.755E+03	7.221E+03	1.904E+07	2.388E+07	2.212E+04	2.775E+04
Curium	243	3.088E+02	1.292E+03	1.594E+04	6.674E+04	5.850E+02	2.449E+03
Curium	244	2.364E+04	9.268E+04	1.914E+06	7.502E+06	6.694E+04	2.624E+05
Curium	245	8.836E+02	3.648E+03	1.518E+02	6.266E+02	5.037E+00	2.080E+01
Curium	246	1.134E+02	4.560E+02	3.486E+01	1.401E+02	1.141E+00	4.588E+00
Curium	247	1.096E+00	4.348E+00	1.018E-04	4.036E-04	3.251E-06	1.290E-05
Curium	248	5.908E-02	2.316E-01	2.513E-04	9.849E-04	3.128E-05	1.226E-04
TOTAL		1.779E+09	1.232E+10	5.792E+09	1.032E+10	2.324E+07	3.915E+07

(a) Total of all nuclides in the fuel, cladding, and fuel assembly structural material.

FIGURE 4. Decay Heat for Typical PWR Spent Fuel (Malbrain, Lester, and Deutch 1981)

January 1983 and projected to 1986 is presented in Figure 7. BWR assemblies will have typical decay heats of ~0.2 kW after ~4 to 12 years of storage; PWR assemblies will have 0.3 to 0.7 kW after 4 to 12 years of storage.

Reactor-Induced Defects. Not all SF will arrive intact at the MRS facility. A very small fraction may have failed as a result of reactor power operations or handling in the reactor storage pool. Through 1976, a total of 2290 SF assemblies were known to have from one to several defective rods; less than 300 of these assemblies have been reprocessed (Bailey et al. 1982). The number of unprocessed, defective assemblies represents less than 7% of the assemblies stored as of 1981. The percentage of failed rods out of the total number of SF rods has been less than 1% and, in some instances, may be less than one rod in 10,000 (Strasser and Lindquist 1977). The percentage of defective assemblies may decrease because earlier LWR fuel had a higher incidence of cladding failures during reactor operation than more recent fuel designs

FIGURE 5. Decay Heats for Typical PWR and BWR Assemblies Indicating the Rapid Decrease in Decay Heat During the First Two Decades After Discharge (Johnson, Gilbert, and Guenther 1982)

(Johnson, Gilbert, and Guenther 1982). However, there is strong interest in storing these older assemblies because of their low decay heat. The improved failure statistics of new fuel rod designs could be offset by any additional failures caused by higher burnups.

Incipient Defects. Some Zircaloy fuel rods are known to have partial through-wall cracks (incipient defects). During inspection, it is probable that some of the defective or incipiently defective rods will not be found because inspection techniques are not 100% accurate. Visual methods are purported to detect less than 10% of the failed fuel rods, while out-of-core sipping immediately following reactor discharge may detect as high as 95% of the defects. However, inspection is not routine (many assemblies are not inspected); a substantial effort by the nuclear industry would be required if the inspection frequency were to be increased substantially (Bailey et al. 1980). If all the fuel assemblies are not inspected for defects prior to shipment, then each assembly would have to be inspected at the MRS facility if

FIGURE 6. Distribution of Decay Heats for U.S. LWR Spent Fuel Inventories Discharged Prior to January 1983 and Projected to 1986 (Johnson and Gilbert 1983b)

FIGURE 7. Distribution of Wet Storage Time for U.S. LWR Spent Fuel Inventories Discharged Prior to January 1983 and Projected to 1986 (Johnson and Gilbert 1983b)

only unbreached fuel rods were to be stored. Rods with incipient defects occur relatively infrequently; however, there are no statistics on the number of incipient defects.

Previous Dry Storage. The initial SF characteristics at an MRS facility will also be determined by any previous dry storage. Interim dry storage will be necessary in the 1980s (due to filled wet storage capacities) (see Figure 1) and will probably supply some portion of the SF in an MRS facility (DOE 1982b). A detailed analysis of current knowledge on interim dry storage is given in Johnson, Gilbert, and Guenther (1982). Although operating conditions and equipment design will obviously be optimized, some rods with previously undetected incipient defects could fail during dry storage. Assuming the same detection capability as at the original reactor site, then more than 80% of the newly failed assemblies could be detected before being sent to an MRS facility. However, the sensitivity of sipping tests decreases with the age of the fuel. If the fuel rods do not fail from stress rupture in the first few years of dry storage, then the rods will probably be cool enough that failure will be unlikely (see Section 4). Thus, interim dry storage could very well act as a screening method for incipiently defective rods that are susceptible to failure. The probability of additional failures at an MRS facility would be reduced since storage conditions in interim dry storage and MRS may be very comparable except for the anticipated lower initial fuel temperatures in MRS compared with initial temperatures previously achieved in interim dry storage.

Handling and Transportation. SF rods that do not fail in-reactor could be damaged during handling and transportation operations. Bailey et al. (1982) reviewed the reported cases of damage or suspected damage to fuel rods at reactor sites and those shipped to other facilities. Of the 89 cases reviewed, only a few rods were actually damaged. In most cases involving minor handling damage, no fuel or fission gas was released. Another study on the effect of shipping on cladding integrity concluded that there was no evidence that transportation contributed to loss of cladding integrity, although this could not be validated due to a lack of quantitative measurements (Bailey et al. 1982). Present evidence indicates that fuel rods that are intact when they leave the reactor pool will remain intact during transportation to an MRS facility.

SS-Clad Fuel. The foregoing discussion on SF characteristics upon arrival at the MRS facility was based primarily on Zircaloy-clad fuel rods; however, about 1500 SF assemblies have SS cladding. Although the number is small and there has not been great interest in interim dry storage for SS-clad rods, some SS-clad rods will probably eventually reach an MRS facility. SS cladding defects have been reported; thus, MRS storage of defective SS-clad rods must be considered (Johnson, Gilbert, and Guenther 1982). Aside from cladding characteristics, other SF characteristics for SS-clad fuel rods are comparable with those for Zircaloy-clad fuel rods.

Storage Conditions for SF in MRS

Storage conditions for MRS consist of the environmental and physical/chemical conditions that exist during normal and off-normal situations. During normal operation, cladding temperatures will be in the range of 200 to 400°C at the beginning of storage, based on current designs for interim dry storage (Johnson, Gilbert, and Guenther 1982). Fuel temperatures will decrease depending on the amount of burnup and previous decay as discussed above. Examples of temperature histories for dry storage of rods with different amounts of previous pool storage are given in Figure 8. Data are given for German tests (KWO, KKS, and KWW),^(a) several U.S. tests at the Nevada Test Site (NTS), and Canadian tests (WNRE)^(b) (Johnson and Gilbert 1983b). These peak operating temperatures are not expected to cause either physical changes in the fuel pellets or gas release to the free volume. The temperature oscillations for the NTS tests are caused by seasonal variations.

Plenum gases under normal operation will have pressures that range as high as 65 atm (6.6 MPa) in a spent PWR fuel rod at room temperature; but EOL room temperature pressures are typically 30 to 40 atm (3 to 4 MPa) for PWR fuel rods and 15 atm (1.5 MPa) for BWR rods (Johnson, Gilbert, and Guenther 1982). These pressures will produce hoop stresses in the Zircaloy cladding from ~2000 psi (13.8 MPa) in a BWR rod to 9000 psi (62 MPa) in a 40-atm PWR rod at a storage temperature of 250°C. The isotopic composition of the plenum gases is complex

(a) KWO - Kernkraftwerk Obrigheim, Federal Republic of Germany (FRG); KKS - Kernkraftwerk Stade, FRG; KWW - Kernkraftwerk Wuergassen, FRG.

(b) WNRE - Whiteshell Nuclear Research Establishment, Canada.

FIGURE 8. Zircaloy-Clad Fuel Assembly Temperature Histories for Dry Storage Demonstrations (Johnson and Gilbert 1983b)

because of the power history dependence and radioactive decay; but most of the gas is xenon, krypton, and helium. More detailed information on gas composition for specific power histories would require sampling or calculations based on power history and design data. Additional fission gas will not be released under typical dry storage temperatures unless the fuel rod fails and U_3O_8 forms, creating a much larger fuel surface area. Even if helium from actinide decay diffuses into the plenum of a typical PWR rod with 50,000 MWd/MTHM burnup, the pressure increase would be less than 1 MPa during the 100-year MRS period even at 350°C (see Figure 9).

In previous dry storage tests and demonstrations, air, argon, carbon dioxide, helium, and nitrogen have been used as cover gases (Johnson, Gilbert,

FIGURE 9. Pressure Increase Due to Helium from Actinide Decay (Johnson and Gilbert 1983b)

and Guenther 1982). Temperatures and pressures of these gases will be dictated by the desired heat loading and the design of the storage canister and the rest of the waste package.

The number of off-normal events that could affect storage conditions at an MRS facility is limited once the SF has been safely placed in storage. Accidents during handling and transportation could possibly cause mechanical damage to fuel already stored in the storage system. Significant damage from such incidents might allow the loss of the cover gas, the ingress of air if the cover gas is not air, and/or contact of the canister with ground water. An above-ground fire might conceivably affect the temperature in a silo or other above-ground structure, but below-ground storage (for example, a dry well system) would have a larger thermal resistance to heating from above.

3.2.2 SF Degradations of Concern in MRS

Fuel rod degradation could occur in an air environment at reactor-induced failures, reactor-induced incipient defects, or cladding failures resulting from mechanical overload or other degradation mechanisms during previous handling or dry storage. Such degradation would affect the retrievability and fuel handling requirements of the SF from an MRS facility.

Defects such as through-wall failures or incipient defects in fuel rod cladding can occur during reactor operation. Since not all of these failures (especially incipient defects) will be detected, it is probable that some defective rods will enter dry storage at an MRS facility. If cladding failure has occurred, oxidation of the fuel from UO_2 to U_3O_8 during dry storage in an air atmosphere could cause significant additional degradation of the fuel rod due to increased stresses at the point of failure (Johnson and Gilbert 1983a).

Stress Rupture. Blackburn et al. (1978) examined potential degradation mechanisms for dry storage of SF, including the following:

- rupture from mechanical overload
- stress rupture
- SCC from fission products
- rapid fracture of flawed cladding
- hydrogen-induced cracking
- internal hydriding
- cladding oxidation.

Blackburn concluded that the maximum storage temperature for Zircaloy-clad fuel in dry storage is 380°C, based on stress rupture considerations. Einziger and Atkin (1982) performed tests on PWR fuel rods at 482°C, 510°C, and 571°C and concluded that 400°C is a conservative storage temperature for SF in limited air or inert gas atmospheres, based on a stress rupture mechanism and the presumption that irradiation hardening of the cladding does not reduce the stress rupture properties. They also found no signs of additional cladding hydriding, SCC, or other fuel pellet degradation.

The Einziger and Atkin (1982) tests were conducted at temperatures and times sufficient to cause annealing of radiation damage, which may have resulted in improved ductility as compared with as-irradiated SF rods. Some

rods strained 15%; SF cladding has less than 5% ductility. As long as the storage temperature remains below 250°C, it is not likely that recovery and annealing of radiation damage in the cladding will occur in dry storage. Sufficiently high temperatures anneal radiation damage, causing an increase in ductility and a decrease in the strength and resistance to failure by creep rupture (Johnson, Gilbert, and Guenther 1982). The increase in ductility may enable gas pressures to be relieved by rod volume increases, although this could be limited by adjacent fuel rods (Guenther 1983). Additional PWR whole-rod experiments are being conducted at 325°C at Battelle Columbus Laboratories (BCL) to investigate rod behavior with residual irradiation. Additional information on predicting stress rupture in Zircaloy cladding is given in Section 4.

Fuel Oxidation. Some fuel rods with large incipient defects could fail even if the storage temperatures are set to prevent stress rupture in a completely intact rod. Other rods might arrive at the MRS facility with failed cladding that may not be detected prior to emplacement. In either case, fuel degradation could occur. Exposure of unirradiated fuel to air at temperatures as low as 200°C causes oxidation (White et al. 1983; Simpson and Wood 1983). Tests of water-logged and breached irradiated fuel rods in air at 325°C after 2100 h indicated rapid oxidation that caused fuel swelling and crack extension in BWR cladding. The limited supply of air prevented complete oxidation of the fuel. Concurrent testing of a similarly breached fuel rod in an argon environment indicated no degradation of the cladding or fuel (Johnson et al. 1983). Additional data and preliminary rate equations for oxidation of irradiated fuel are given in Section 4.

The above tests with water-logged fuel rods indicated that 5 to 10 grams of water were released during heating at 325°C. Based on this amount of water in each rod of a 15 x 15 PWR assembly, ~125 moles of water would be available from the 225 rods. For complete, uniform reaction with just the cladding, less than 6% of the cladding thickness of 0.048 in. (1.2 mm) would corrode, which would probably not cause rapid failure but could cause premature failure of rods stored near their stress rupture limit.

Handling and Transportation. Mechanical deformation of the cladding could occur during handling and transportation operations. Very little information

exists on the response of cladding after sustaining damage, and extensive development of limits applicable to already damaged bundles or structures is required (Bailey et al. 1982). Some tentative limits have been proposed (Bailey et al. 1982). For lateral loading, acceleration limits were set at 30 m/s^2 ; proposed axial loading limits were 70 m/s^2 . During handling, load/trip settings were assumed to be 1.5 times the assembly weight.

SS-Clad Fuel. Degradation in SS-clad SF has not been extensively investigated, but studies that have been completed indicate that SS cladding should behave as well as or better than Zircaloy cladding in storage. Stainless steel is generally more resistant to oxidation than Zircaloy; thus, concern over introducing air into a canister centers on the oxidation of the fuel, which is considered the probable cause of primary defect propagation. Fuel retrievability is not expected to be limited from degradation caused by SCC, fission product attack, hydrogen, or creep-related rupture. Postirradiation creep data for irradiated SS suggest that creep rates are very low below 500°C , but the data are very limited for lower operating temperatures (Johnson, Gilbert, and Guenther 1982). Therefore, the principal concern for SS-clad SF rods will be fuel oxidation in rods that were defective prior to placement in an MRS facility.

Canister Degradation. Carbon steel is the reference canister material for MRS; however, specific data regarding atmospheric corrosion for carbon steel canisters were not found during this brief review. A review of state-of-the-art corrosion data for metallic barriers used in repositories does compare data for stainless and carbon steels in brines but not under any atmospheric conditions (Merz 1982). One study of an MRS facility assumes a uniform corrosion rate of 0.001 in./yr (0.025 mm/yr) and a wall thickness sufficient to support such a rate for 100 years (General Atomic 1982). A review of atmospheric corrosion data for carbon steel in an MRS environment would be appropriate, especially if the integrity of the cladding is not assured or if it is deemed unimportant.

The effect of overpressurization in a canister by premature failure of fuel rods has not been calculated for SF assemblies in a carbon steel canister. Such an analysis was completed for a 304 SS canister used as a repository design for the Dry Surface Storage Demonstration and Climax-Spent Fuel Tests

(Fish and Einziger 1981). A 75% fission gas release was assumed to be the maximum release prior to any fuel rod failure even in transient tests. For this canister design with a 10% plenum volume, the canister stresses were about half of the design strength. Similar calculations could be performed for a typical MRS canister design.

SF canister designs entail placing the rods in a vertical position. If sufficient cladding degradation occurs prior to or during MRS storage, fuel could be lost to the interior of the canister and subsequent removal of the fuel rods would require special handling methods. Some of the designs for geologic repositories include only one PWR spent fuel assembly, but several designs include as many as eight consolidated assemblies, depending on the assembly type. Consolidation enhances the economics of geologic disposal but would require opening the MRS canister to allow consolidation of the fuel rods for geologic disposal. Thus, the integrity of the SF cladding may be important to assuring safe and economical retrievability at the end of storage in an MRS facility.

3.3 HIGH-LEVEL WASTE

HLW canisters produced during commercial SF reprocessing may eventually be stored at an MRS facility. The HLW canister is one of the best candidates for MRS because it is designed specifically to provide containment and stability over a long period.

The HLW glass and canister are described below, and information concerning degradation during some off-normal or accident conditions is given to support MRS facility design selection. Effects from major design-basis accidents were not considered.

3.3.1 MRS Operating Parameters for HLW

There are several potential designs for an MRS facility, ranging from below-ground dry wells with engineered barriers to securely fenced pads with individual storage casks. All facility concepts will be dry with passive cooling (DOE 1981).

The requirements for safe storage during off-normal events or accident conditions such as floods, earthquakes, tornados, handling impacts, and external missiles will evolve as these design-basis accidents are analyzed and the consequences are identified. The descriptions of HLW glass and canisters as well as potential degradation mechanisms that are presented below can be used in these analyses.

Initial Characteristics

The following discussion first considers the initial characteristics of the HLW glass and then the initial characteristics of the canister. Initial characteristics of the glass include the composition, physical properties, mechanical properties, and radiation levels. The description and design of HLW canisters are also provided.

HLW Glass Characteristics. Radioactive waste glasses are formulated by combining a preselected mixture of glass-making additives with the waste. The glass-making additives usually comprise 65% to 85% of the product waste glass. Because of the many different potential formulations possible, HLW glass cannot be thought of as a single substance with set properties. However, many measurements have been made of different glass formulations using a borosilicate glass matrix; and physical and mechanical property ranges are available.

The amount of HLW glass produced as a result of reprocessing 1 MT of SF can range from 100 kg to over 500 kg. The range is determined primarily by the chemicals added to the HLW during reprocessing. Extensive background information on typical glass formulations for HLW arising from commercial LWRs is contained in several reports (Mendel et al. 1977; Ross et al. 1978; Ross and Mendel 1979). A typical glass that could be produced by the fuel reprocessing plant owned by Allied-General Nuclear Services (AGNS) at Barnwell, South Carolina, has been given the code 77-260. Slate, Ross, and Partain (1981) described the composition of this typical glass and characterized a HLW canister filled with this type of HLW glass. The approximate composition of 77-260 and several other HLW glasses is given in Table 11.

Established physical properties for HLW glass include density, thermal conductivity, specific heat, and thermal expansion coefficient. The density of

TABLE 11. Typical HLW Glass Compositions (wt%) (Chick and Turcotte 1983)

Frit	USA 76-68	USA 77-260	France LWR	Sweden ABS-39	Japan POS77	Germany 98-20
SiO ₂	40.0	36.0	47.5	48.5	47.5	40.2
B ₂ O ₃	9.5	9.0	15.0	19.1	13.5	10.8
Na ₂ O	7.5 (12.5) ^(a)	8.0 (11.1) ^(a)	12.0	12.9	2.0 (9.4) ^(a)	21.9
ZnO	5.0	--	--	--	1.0	--
CaO	2.0	1.0	4.0	--	1.0	2.2
K ₂ O	--	2.0	--	--	1.0	--
TiO ₂	3.0	6.0	--	--	--	3.3
Al ₂ O ₃	--	2.0	3.0	3.1	2.5	1.1
CuO	--	3.0	--	--	--	--
Li ₂ O	--	--	--	--	2.0	--
BaO	--	--	--	--	1.0	--
Waste	(b)	(c)				
Fe ₂ O ₃	9.64	(d)	5.0	5.7	--	1.87
Na ₂ O	4.98	3.13	--	--	7.4	--
Gd ₂ O ₃	--	9.84	--	--	--	--
Other inerts	1.08	3.97	0.5	--	8.4	0.82
Fission products	12.71	10.99	13.0	9.0	12.9	16.64
Actinides	4.59	5.25	(e)	1.66	--	1.07

(a) Total Na₂O in waste plus frit.

(b) PW-8a.

(c) PW-7c.

(d) Included in "Other Inerts."

(e) Included with "Fission Products."

HLW glasses can range from 2.5 to 3.3 g/cm³ depending on the constituents. The 77-260 glass described in Table 11 has a density of 3.1 to 3.2 g/cm³. The thermal conductivity of HLW glass varies with temperature as shown in Figure 10. The dotted region bounds the measured values of glass thermal conductivity. The specific heat of waste glass is ~0.2 cal/C-g. Typical thermal expansion coefficients of borosilicate waste glasses are 80 to 100 x 10⁻⁷ per °C.

Because the thermal expansion coefficient of HLW glasses is less than that of the metal canisters, HLW canisters normally have unrelieved cooling stresses that approach the yield strength of the canister material (Slate and Maness 1977). Elongation of the canister wall (i.e., increase in circumference) is less than 1%, which is insignificant when compared with the amount of elongation required before failure. Analyses and tests indicate that the residual stress does not reduce the ability of the canister to contain the glass (Simonen and Slate 1979).

Mechanical properties of HLW glasses include the strength of the glass, Young's modulus, and impact resistance. The strength properties are more a function of surface condition and previous glass thermal history than of composition. Tensile strengths determined by a diametral compression test for two different waste glasses have been found to be 51 MPa (Chick et al. 1980) and 37 MPa (Bunnell 1979). Theoretical calculations of tensile strength of glasses give values of 7000 MPa. This difference between measured and theoretical values as well as work by Slate et al. (1979) demonstrates the effects of surface condition and internal defects on glass strength.

Young's modulus for HLW glass is generally insensitive to composition or surface condition. Values of ~7 x 10⁴ MPa have been found (Shand 1958). Other moduli values can be calculated from this value.

Impact behavior is an important issue in analyzing handling and transportation accidents. Glass fragments smaller than 10 µm in diameter are usually considered respirable (Lippman 1977). Impact behavior is relatively independent of glass composition. Data relating the impact velocity of small glass samples to the weight percent of respirable fragments generated are given in

FIGURE 10. Thermal Conductivity of Waste Glasses (Mendel 1978)

Figure 11 (Smith and Ross 1975). Other work on impact testing of full-scale canisters has also been completed and provides data that compare well with small-scale test results (Simonen and Slate 1979).

Canister Characteristics. The initial characteristics of the canister are also important to safety and retrievability in an MRS facility. The canister contains and isolates the HLW glass during production, storage, transportation, and emplacement in MRS facilities or geologic repositories. Therefore, meeting long-term storage requirements for an MRS facility is one of the generally recognized objectives of canister design. The following discussion includes a description of the canister, the design of the HLW canister, and the decay heat and radioactivity for the HLW canister.

Various canister designs and sizes have been investigated at PNL and Savannah River Laboratories (SRL). One possible design is shown in Figure 12. The top of the canister is formed from a flanged-only tank head. The bottom is a reversed-dished flanged tank head. Extensive structural and thermal analyses on various canister designs have been conducted (Simonen and Slate 1979;

FIGURE 11. Effect of Impact Velocity on Fraction of Respirable Particles Formed (Smith and Ross 1975)

Westcott and Slate 1979). Details on designs for different waste treatment facilities vary according to the specific needs of those facilities. For example, Baxter (1981) described a canister design with a flanged nozzle on the top to accommodate a grapple-handling mechanism and a special welding process.

Extensive research has been conducted on the suitability of various alloys for contact with molten glass, handling impacts, water storage, and corrosion resistance in geologic repository media (Slate and Maness 1977). Stainless steel (304L) is the primary candidate for HLW glass produced by an electric glass melter. If the canister is to be the melting vessel (as in the in-can

FIGURE 12. Schematic of the Reference Commercial HLW Canister Design (Slate, Ross, and Partain 1981)

melting process), Inconel 601® is the preferred material. Since 304L SS is a common alloy, no physical or mechanical properties are presented here.

Canister diameters studied in the past have ranged from 6 in. (15.2 cm) up to 32 in. (83.8 cm), although 12 to 24 in. (30 to 61 cm) is the more probable range. The diameter of the canister is determined primarily by the thermal conductivity, decay heat of the HLW glass, and transportation limits. Generally, large-diameter canisters are more advantageous from an economical standpoint, although glass centerline temperatures must be kept below the 500°C devitrification limit during storage and shipment.

The wall thickness of a 304L SS canister is usually in the range of 0.25 to 0.375 in. (0.635 to 0.953 cm) for canister diameters up to 24 in. (61 cm). This thickness range, which is larger than needed to contain the glass, was selected primarily to meet handling and impact loads. Canisters designed for HLW from reprocessed short-cooled fuel, such as may occur with fast breeder reactor fuel, may have interior radial fins to reduce the glass centerline temperature below 500°C during the initial decay period.

The canister walls are expected to be decontaminated to smearable limits (49 CFR 173) before they are shipped from the vitrification facility. The oxide layer that forms during filling will be removed during the decontamination process; therefore, no delayed release of contamination during storage is expected.

Decay heat in a canister is another important HLW characteristic for an MRS facility. The canister decay heat depends on the age and concentration of the HLW. Current reprocessing strategies (Anderson, Carr, and Mitchell 1982) predict an average SF age of 16 years if the first commercial fuel reprocessing plant begins operation in 1990. This decay period could become longer as reprocessing is delayed. The reference HLW canister described by Slate, Ross, and Partain (1981) assumed the HLW glass was formed 5 years after reactor discharge. The specific decay heat after a 5-year decay is 19 W/l of glass or

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3.7 kW in a 12-in. (30-cm) diameter, 10-ft (3-m) long canister. The glass centerline temperature for this heat load is ~250°C when the canister is in open air. The canister decay heat as a function of time is shown in Table 12.

The total activity in this canister (Slate, Ross, and Partain 1981) is 1.02×10^6 Ci 5 years after reactor discharge and 6.58×10^5 Ci after a 10-year decay period, as calculated using the ORIGEN-II code (Croff 1980). A self-shielding calculation for this canister produced the dose rate profile shown in Figure 13. The values decrease by approximately a factor of 10 after a 100-year decay period.

The neutron flux from an unshielded HLW canister is very small when compared with the gamma dose rate. However, the ineffectiveness of large steel/lead casks in neutron attenuation should be taken into account in the MRS transporter design since the neutron dose could exceed occupational limits.

Storage Conditions for HLW in MRS

The basic requirements for HLW storage during normal operation of a dry passively cooled MRS facility are straightforward:

- sufficient cooling to maintain the glass centerline temperature below 500°C and the canister temperature below 375°C
- sufficient radiation shielding to protect operating personnel.

TABLE 12. Calculated HLW Canister Decay Heat
(Slate, Ross, and Partain 1981)

<u>Decay Period, yr</u>	<u>Canister Heat Generation Rate, W</u>
5	3690
10	2210
20	1600
50	798
100	292
1000	20

FIGURE 13. Profile of Reference Commercial HLW Canister Dose Rate (mrem/h) After 5-Year Decay (Slate, Ross, and Partain 1981)

3.3.2 HLW Degradations of Concern in MRS

Extensive studies of HLW glasses show that little, if any, changes occur that affect their ability to perform under conditions projected for an MRS facility. Potential degradation mechanisms have been studied and were found to be insignificant in the context of an MRS facility. However, devitrification, volatility, leaching, helium buildup, and stored energy are briefly discussed to provide substantiation for this claim. Degradation of the canister is also discussed because it provides an additional barrier to early release of the radioactive inventory of HLW.

Devitrification. Devitrification is the formation of crystals in the glass. The crystals form because they have a lower free energy than the glass solution. Devitrification is generally only apparent at a microscopic level and usually does not significantly affect glass durability. Devitrification may increase the leach rate of a glass by a factor of from 2 to 5--rates that are still acceptably low for geologic disposal. Measurable devitrification of waste glasses generally occurs from 500 to 950°C (Turcotte and Wald 1978). Above 950°C, the crystals redissolve; and, below 500°C, the viscosity of the glassy matrix is so high that crystallization proceeds at extremely low rates (Mendel et al. 1977; Mendel 1978). A general specification for limiting devitrification is that the glass centerline temperature during normal operations should not exceed 500°C. Temperature excursions between 500°C and 600°C for short periods (<100 h) should not cause significant increases in devitrification, although more testing is needed to determine specific allowances for glass.

Volatility. After initial formation, volatility from waste glass is only a factor in accidents where the temperature is sufficient to remelt the glass (>800°C). Such high temperatures are not anticipated in an MRS facility. Temperature-dependent volatility of elements for molten glass is given by Gray (1976).

Leaching. Leaching is an important consideration in the geologic disposal of HLW glasses; it is not a concern in an MRS facility, which will be dry. Data on leaching for a wide range of conditions have been compiled by the Materials Characterization Center (MCC) at PNL in support of the DOE HLW programs (Chick and Turcotte 1983).

Helium Buildup. This phenomenon is a result of radioactive decay. The alpha emission by some radionuclides results in the production of helium atoms that diffuse through the glass to the free voids in the canister. The extent of the helium buildup depends on the concentration of alpha emitters in the waste. Pressure buildup in HLW canisters for two cases has been estimated (Turcotte et al. 1976). The helium pressure for the LWR once-through fuel cycle waste is shown in Figure 14a, and the helium pressure for the full plutonium recycle case (with a much larger inventory of alpha-decaying radioisotopes) is shown in Figure 14b. The solid lines represent no helium trapped interstitially in the glass, and the shaded portion below the lines represents the potential effect of trapped helium located at point defects in the glass. A 10 vol% plenum in the canister and 30 wt% waste loading in the glass was assumed. Once-through waste, which is the reference for this study, will not produce significant amounts of helium during the 100-year MRS period. If the scope of the MRS program expands to include high-alpha wastes (for example, breeder waste), additional studies on helium buildup will be needed.

FIGURE 14. Potential Helium Pressure in Waste Glass Containers (Turcotte et al. 1976)

Stored Energy. HLW degradation from stored energy results from the latent energy produced by radiation-induced displacement of atoms from their normal lattice positions. The latent energy is released as heat when the temperature of the glass is raised sufficiently. The maximum stored energies in waste glasses are from 30 to 50 cal/g; thus, under adiabatic conditions, the maximum additional temperature rise that could occur due to an unlikely rapid release of the stored energy is only 150 to 300°C (Weber et al. 1979; Roberts, Jenks, and Bopp 1976). Other aspects of radiation damage to glass have been studied and have been found to be insignificant within the MRS time frame (Gray, Wald, and Turcotte 1981; Turcotte et al. 1981).

Canister Degradation. Degradation of the canister has been the focus of many studies: Simonen and Slate 1979; Solomon et al. 1978; Povich and Rao 1978; McCoy 1983; Angerman and Rankin 1978; Peters and Slate 1981; Smith and Ross 1975; Wu et al. 1978). Under the dry storage conditions of an MRS facility, no significant exterior corrosion of the SS will occur. The solidified glass will not corrode the interior of the canister and only minor corrosion from the air will occur (Slate and Maness 1977; Baxter 1981), but water storage can be a problem. Handling accidents could rupture the canister, but only a relatively small radiological release would occur from a cracked or punctured canister. There will be a small helium buildup in the canister during the MRS time frame (see Figure 14), but the pressure (<15 psi) will be well within the pressurization limits of the canister. Fire or loss of cooling may cause softening of the glass and devitrification, but no release of radioactivity is predicted. No radiation damage of the canister will occur.

Decreases in the glass decay heat will result in changes in the thermal gradient in the canister, which will cause some glass cracking. The amount of cracking will be about the same as observed in canisters that have been cooled in air during nonradioactive tests. The surface area of the glass will be 10 to 15 times the surface area of a glass monolith (Peters and Slate 1981). This effect is expected and is not significant.

Stainless steel is susceptible to SCC, and all SS canisters are assumed to be somewhat sensitized by virtue of being filled with molten glass and cooled slowly through the sensitization temperature envelope. If the canisters were

to be stored in water without the proper water chemistry control, it is possible that canisters could arrive at the MRS facility with incipient cracks and pitting that could eventually lead to loss of integrity. This scenario is very unlikely; it is presented as the only credible degradation event for a SS canister that would produce significant consequences. If cracks occur from SCC, they would be quite small and would probably not allow any significant glass release. Data on releases are presented by Bradley (1977) for a cracked canister stored in water.

HLW canisters will be retrieved from an MRS facility according to normal operations since no degradation of the canister or handling fitting is predicted.

3.4 TRANSURANIC WASTE

Degradation of TRUW during MRS storage has two major impacts: retrievability and safety. Only the safety issue will be reviewed here, although it can be assumed that any degradation that impacts safety will impact recovery. This brief review summarizes current and readily available information on modes of TRUW degradation. As a part of the review, assumptions are made on waste types, form of the stored waste, type of containment, TRUW loading, and temperature. These assumptions will be discussed in the following paragraphs for various degradation modes, and the extent of degradation and the consequences thereof will be estimated.

3.4.1 MRS Operating Parameters for TRUW

Previous work defining TRUW for defense-related waste has been conducted (Molecke 1979). These defense wastes are similar to those defined by Kosiewicz, Barraclough, and Zerwekh (1979); Franklin (1983); and Jensen (1983). A composite of waste types is given in Table 13. Of these, glass, filter media, dirt, and parts of the filters can be omitted from further consideration because they are inert towards the most likely methods of degradation. Others, such as metals, are essentially inert but may degrade when combined with other wastes.

TABLE 13. Potential Types of TRUW

<u>Waste Type</u>	<u>Description</u>
Metal	All shapes and sizes, including filings and turnings. Consists mostly of SS or equivalent but also some aluminum and zirconium.
Filters	Process and HEPA, containing some combustible material.
Sludges	From filter systems, oil, dirt, and absorbents.
Combustibles	Wood, paper, rags, plastics, and rubber.
Liquids	Small quantities of free liquid in bottles but mostly absorbed, occasionally on rags; aqueous, organic, and inorganic liquids.
Glass	Laboratory ware and instrument components.
Chemicals	Acids, bases, oxidants, decontamination solutions, and related waste.
Ion-exchange resin	Organic anion and cation resins, inorganic zeolites; may be particulate, powder, or liquid.
Miscellaneous	For example, batteries (flashlight, film packet), soil, compressed gas (mainly propane and Freon from spray cans).

Initial Characteristics

As for SF and HLW, the initial characteristics of TRUW affect degradation during storage in an MRS facility. The following discussion reviews the salient initial characteristics of TRUW, including: 1) waste types; 2) waste content; and 3) waste packages for MRS.

TRUW Types. The stored waste is assumed to be of one of three forms-- loose, compacted, or concreted. These definitions were chosen because Franklin (1983) states the filled drums will weigh 21 to 333 kg (assumed to be net weight). A nominal 100-kg waste loading is assumed for this report. The term "loose" is taken to mean essentially as generated. For the evaluation of consequences, no effect of compaction will be considered other than the mass loading of the container, although at sufficiently high racking densities there may be an enhanced effectiveness of alpha radiation damage.

Waste Content. The TRUW loading is assumed to be normally 0.1 to 1 g per drum, although two orders of magnitude variation may occur in either direction (Kosiewicz, Barraclough, and Zerwekh 1980); the TRUW material is generally

assumed to be weapons-grade Pu, mostly ^{239}Pu . Faust et al. (1977) indicate that commercial waste will be ~55% ^{239}Pu , 25% ^{240}Pu , and 12% ^{241}Pu with the rest being mostly ^{238}Pu and ^{242}Pu . For commercial waste, these values calculate to a dose rate of ~0.1 mR/yr with values up to >10 mR/yr possible.

The concrete form is assumed to be ~30% waste in intimate contact with dry cement. One consequence of using cement is the reduced free volume and the possibility of a more rapid increase in pressure for a given production of gas.

Waste Package. Franklin (1983) describes the four types of containment expected for MRS. Except for the canisters, which are welded shut, all other containers are assumed to be mechanically closed and incapable of being pressurized more than 100 kPa (~15 psi) above ambient. Actual results from SRL suggest that unless special sealing measures are taken, the maximum pressure will be about 70 kPa (Ryan 1982).

The internal coating of the waste containers is expected to be one of four basic types--hot dipped galvanized (zinc-coated) steel, bare steel, pitch (bitumen)-coated steel, or epoxy painted steel. Wood or fiberglass reinforced plastic (FRP) containers are omitted because that type of containment is not mentioned by Franklin (1983), although they are used for defense waste (Jensen 1983).

Storage Conditions for TRUW in MRS

The MRS facility will be operated at essentially atmospheric pressure and controlled to ~25°C. The nominal storage time could be as long as 100 years. Seasonal cycles are expected to have minimal effect inside the facility. The humidity is assumed to be controlled below 60% relative humidity (RH) but will probably never be below 30% RH. The upper humidity limit will prevent catastrophic corrosion of exposed steel on the drums; below 30% RH, existing atmospheric corrosion essentially stops (Rozenfeld 1972).

3.4.2 TRUW Degradations of Concern in MRS

In the past, three degradation modes have been considered to the greatest extent--radiolysis, biodegradation, and thermal degradation (Kosiewicz 1981). Two other modes--corrosion and chemical degradation--are also discussed.

Radiolysis. Radiolysis is the degradation of chemical bonds by the alpha particles from decay of the TRUW contamination. Degradation of combustible organic materials with the release of gas (the mode of most concern in storage) is described by the equation:

$$M = 0.1 AG^\circ$$

where M = moles of gas produced per year per drum

A = number of curies of alpha emitters/drum

G° = total gas molecules evolved per 100 eV; typical values of G° are given in Table 14.

The reaction products of the degradation are usually CO_2 , CO , and H_2 . Helium is produced by neutralization of the alpha particles. Oxygen originally present in air is consumed by reaction with the organic material; it is also produced by radiolysis of water. According to Kosiewicz, Barraclough, and Zerwekh (1979), moist waste produces up to ~25% more gas than dry waste.

The net effect of radiation is not clear-cut. For most materials, increasing the dose rate increases the rate of degradation. As seen from Table 15, the presence of low-level TRUW contamination may be beneficial for some types of waste in preventing other types of degradation. It is reasonably clear that an increase in temperature enhances both radiolytic and thermal modes of degradation. The radiation will also affect container coatings. Pitch (bitumen), for example, is reported to withstand 100 mR before degrading (Perrigo et al. 1979).

TABLE 14. Typical G° Values for Organic Waste (Molecke 1979)

<u>Matrix</u>	<u>G° (gas, total)</u>
Cellulosics	1.3 to 2.9
PVC	3 to 11
Asphalt	0.2 to 1.0
Composite(a)	1.4
Polyethylene	1.9
Pump oil	1.3 to 2.0
Octane	4.5
Concrete/TRU ash	0.0003 to 0.6

(a) See Table 16.

TABLE 15. Gas Evolution as a Measure of Degradation (Kosiewicz, Barraclough, and Zerwekh 1980)

Dose Rate ^(a)	20°C		40°C		70°C		100°C			
	Elapsed Time, days	Gas Rate, ^(b) mol/d-g	Elapsed Time, days	Gas Rate, ^(b) mol/d-g	Elapsed Time, days	Gas Rate, ^(b) mol/d-g	Elapsed Time, days	Gas Rate, ^(b) mol/d-g		
Decomposition of Cellulosics (paper)										
1X	484	0.06	427	0	255	0.5	43	2.0		
					352	0.4	127	2.8		
					450	0.3	233	1.6		
							309	1.6		
0.1X	392	0	387	0			402	1.5		
					200	0.3	61	1.3		
					297	0.1	177	1.6		
					395	0.1	293	1.3		
0X	--	--	416	>0			386	1.2		
					241	0.07	--	--		
					338	0.05				
					436	0.08				
Decomposition of Composite ^(c)										
1X	341	0.1	406	0	252	0.2	92	1.4		
	439	0.2			349	0.2	203	1.0		
					447	0.3	300	1.4		
							393	1.5		
0.1X	392	0	278	0.07	296	0.1	105	1.1		
			376	0	394	0	189	1.3		
							281	1.1		
							314	1.3		
0X	--	--	359	0	371	0	379	1.1		
							62	2.9		
							91	2.5		
							166	1.5		
Decomposition of Polyethylene										
1X	150	0.2	406	0	342	0.1	266	0.3		
	248	0.3			440	0.2	363	0.4		
							461	0.4		
							189	0		
0.1X	391	0	387	0	394	0	286	<0.08		
									384	0
0X	--	--	--	--	359	0.1	--	--		

(a) 1X = 4×10^5 nCi/g.

(b) Gas generation rates must be multiplied by 10^{-6} ; for example, 0.2×10^{-6} mol/day-g.

(c) Composite is 35% cellulose, 23% polyethylene, 12% polyvinyl chloride, 15% Neoprene, and 15% Hypalon.

Biodegradation. Biodegradation has been examined from three standpoints: methylation of the heavy elements to form volatile compounds, formation of chelates, and formation of CO₂ (Barnhart et al. 1980). Work had just begun on methylation when it was determined that the compounds are short-lived in the presence of oxygen and of little further concern (Barnhart et al. 1980). The formation of microbial chelates, which will assist the dissolution of PuO₂, is feasible; however, it is not known whether the degradation rate of the chelate by other means will exceed the formation rate. It was noted that the half-life of citric acid due to microbial action is ~3 years and that of EDTA is ~28 years.

Carbon dioxide is produced at the approximate rates shown in Table 16. At increasing temperatures, biological activity will begin to slow and become less important. If protein is present as from biological experiments, some H₂S will be generated. The mean rate of gas production for all tests performed was 6 µg CO₂/g waste/day.

TABLE 16. Average Rates of CO₂ Generation (cross treatments) in 72 CO₂ Simulations with Sterile Background Controls (Barnhart et al. 1980)

<u>Conditions</u>	<u>LASL Composite^(b)</u>	<u>Sawdust</u>	<u>Asphalt</u>
25°C	5.1 (3.9) ^(c)	9.4 (6.3)	2.3 (1.5)
40°C	6.5 (5.8)	8.2 (5.5)	0.88 (0.45)
70°C	8.6 (5.5)	10.4 (5.5)	4.9 (3.2)
Aerobic	4.8 (4.6)	7.3 (5.1)	3.4 (2.9)
Anaerobic	7.4 (5.8)	11.4 (5.6)	2.0 (1.9)
91% H ₂ O	4.45 (3.5)	10.8 (7.4)	1.0 (1.0)
1% H ₂ O	3.8 (4.7)	8.1 (4.7)	2.0 (2.5)
91% brine	2.9 (3.0)	11.4 (5.2)	2.9 (3.0)
Nutrient	8.9 (5.3)	9.0 (4.5)	3.4 (3.2)

(a) Standard deviation given in parentheses.

(b) 7.5% Neoprene, 7.5% Latex, 7.5% Hypalon, and 7.5% butyl rubber.

(c) Rate of CO₂ generation given as µg/day/g.

If the temperature is maintained at ~55°C, optimal microbial growth is expected (Molecke 1979). Other than a difference in gas composition, the extent of decomposition, and resultant residue, the end effect will be basically the same as for radiolysis--easily dispersed contamination when the container fails or is opened.

Thermal Degradation. Thermal degradation is considered to be the result of reactions of each waste material with itself or air. The interaction with chemicals that are present in the waste (such as nitrates) is not included. At or below 70°C, thermal degradation occurs slowly (Kosiewicz, Barraclough, and Zerwekh 1980) and increases rapidly with temperature above that (see Table 15). Threshold decomposition temperatures are given in Table 17, but even below these values sorbed water loss and visible product degradation is noted in cellulosics (Molecke 1979). Kosiewicz, Barraclough, and Zerwekh (1980) also tentatively noted that:

- Sodium chloride may catalyze decomposition of polyethylene at 40°C.
- Sodium chloride may catalyze slight decomposition of cellulosics (paper) at 40°C.

TABLE 17. Threshold Decomposition Temperatures (°C) of Organic Waste Matrices (Molecke 1979)

Material	In Air	In Nitrogen with Indicated Catalyst				
		None	NaCl	Fe ₂ O ₃	FeCl ₂	Fe
Cellulosics	185 to 195	185	180	185	170 to 195	195
Cotton	185 to 190	185	195	200	170 to 195	205
Wood	175	--	--	--	--	--
Polyethylene	210	320	290	295	330	335
PVC gloves	150	--	--	--	--	--
Lucite	170 to 175	150	155	170	185	160
Teflon	430 to 435	--	--	--	--	--
Hypalon	165	160	175	160	155	160
Neoprene	175 to 180	--	--	--	--	--
Asphalt	275	290	--	--	--	--

- Ferrous chloride catalyzes decomposition of paper at 40°C.
- Some very slight (thermal or bacterial) decomposition of paper occurs without plutonium or catalysts at 40°C.

As noted in Table 15, thermal degradation appears to be counteracted to some degree by the presence of low levels of radiation. These thermal effects are also expected to affect organic container coatings. Pitch coatings are not too degradable but will slowly flow at the higher temperatures--perhaps enough to bare the container.

Corrosion. Corrosion has not been considered to a significant extent in the past, probably because all waste containers are "coated" for corrosion protection. No data were found in the TRUW literature surveyed for this review on the features of such coatings. However, all coatings contain small holes or "holidays" and are therefore susceptible to attack. More important, however, is the damage done to the coatings during handling. Even well-packaged drums have coating damage during shipment from the manufacturer. In addition, handling at the waste generation site is usually done by fork lift, which often results in severe scraping. The interior of the drum usually escapes handling damage except during the filling process. During filling, metal or glass can scrape the coating; the mixing process for concretization can also break the continuity of the coating. Once the primary packaging material of the waste (probably polyethylene bags) disintegrates, the alpha contamination will be free to contact the drum coating and degrade it.

Corrosion of the interior of the drum will depend on the condition of the interior and its contents. Most waste, if dry, will not affect the container whether it is bare or coated. Moisture, including moisture generated by waste degradation, will accelerate the corrosion of bare steel unless it is present in conjunction with cement; the high pH of the concrete protects the steel. Should the waste also contain chloride from chloride salts, degradation of PVC, or degradation of ion-exchange resin, even the presence of concrete will be insufficient protection; severe corrosion of bare steel can occur in less than 20 to 40 years. The main consideration is that the containers with the largest TRUW loading are the most likely to degrade severely and release dispersible contamination.

Because ion-exchange resin will probably be wet, it must be properly treated. Anion and cation exchange resin, without cement, after a dose of 100 mR, generates acid conditions as low as pH 1 (Gangwer, Goldstein, and Pillay 1977; Pillay 1980). Consequently, many bare metals, whether waste scrap or the container, will rapidly dissolve with the release of hydrogen. The presence of cement is expected to neutralize the acid until the lime is consumed. The extent to which neutralization can occur will depend on the type of cement. For Type II portland cement, the neutralization capacity is ~3.8 meq/g.

External corrosion of the drum depends on the atmosphere and the humidity. Nelson and Divine (1980) reported that abraded TRUW drums stored at Hanford are expected to survive 20 years in waste depositories covered with soil where the humidity can reach saturation. Extrapolating these data suggests penetration of those drums in 30 to 50 years; loss of sufficient handling strength will occur before then. If the humidity is maintained below 60% in the MRS facility, atmospheric corrosion can probably be maintained below 0.1 to 0.5 mpy (3 to 12 $\mu\text{m}/\text{yr}$); these rates would cause penetration of a standard drum after ~100 years. To a very large extent, the corrosion rate will depend on prestorage conditions and treatment; for example, humid storage in an environment with chemicals present can lead to high corrosion rates later on even in controlled storage. Proper prestorage treatment including cleaning, repainting, and/or the use of inhibitors can extend the life of a container (based on external corrosion) to beyond 100 years.

Chemical Degradation. Chemical degradation as discussed here is considered to be the action of various waste materials on other wastes or on container coatings either alone or in conjunction with other agents such as radiation or heat.

Nitrates, including nitric acid, can lead to rapid decomposition of most organics; the presence of radiation or heat will accelerate the effect. As a result of their consumption or an initial low concentration, nitrates or especially nitrites in the presence of water can accelerate the corrosion of steel and result in perforation of the container within weeks.

Other chemicals such as FeCl_3 , as noted above, can catalyze the decomposition of various plastics. In the case of pyrophoric metals, ignition could be initiated, although without the presence of oxidants beyond the limited amount of air in the container combustion will be short-lived.

If the waste is maintained at 25°C , little degradation is expected except from radiolysis, which is expected to destroy most of the plastics and damage organic coatings on the container. Unless special care is taken to seal the container, however, no pressure buildup beyond ~ 70 kPa (10 psig) is expected. Additional examples of the effect of waste materials on other waste products were given in the discussion of thermal effects.

4.0 PREDICTION OF DEGRADATION IN SPENT FUEL

The composition and physical properties of LWR spent fuel make it susceptible to several modes of degradation. Excessive degradation could render stored SF nonretrievable with conventional handling methods or cause it to fail to meet acceptance criteria for subsequent operations such as handling, shipping, repository disposal, and reprocessing. If this were to occur, special and innovative approaches to deal with the recovery and repackaging of the degraded fuel would be required. Excessive degradation can be prevented by using reliable predictive tools for extrapolating the fuel performance during long storage periods, even though the existing data base only extends to a few years. These calculations provide guidance and criteria for design and operation of storage facilities.

The modes of degradation being considered are those leading to cladding breach or formation of particulate fuel and release through the breached cladding. These modes of degradation, if allowed to occur, lead to release of particulate fuel and dimensional changes in fuel rods. The loose particulate fuel and dimensional changes must be accommodated by storage and handling systems.

4.1 FUEL OXIDATION

Particulate formation from fuel pellets and fragments in SF results if UO_2 oxidizes to low-density U_3O_8 . Oxygen from storage atmospheres can reach the fuel through reactor-induced cladding breaches or through breaches that occur during storage. U_3O_8 formation can be prevented if a nonoxidizing atmosphere is maintained or if the time at temperatures high enough to cause oxygen diffusion into the UO_2 is limited.

U_3O_8 forms when oxygen from an oxidizing atmosphere diffuses into the exposed fuel and the uranium-to-oxygen (U/O) ratio approaches 1.67. As oxygen initially diffuses into the UO_2 , nondamaging, high-density phases such as U_3O_7 form. The oxygen inventory can be monitored by weighing the UO_2 as illustrated in Figure 15. The weight increases uniformly with time until sufficient U_3O_8 product forms in the grain boundaries and sufficient pressure builds up in the grain boundaries to cause grains to be expelled from the UO_2 pellet or

FIGURE 15. Induction Time (t_1) Required for Powder Formation in UO_2 in Air

fragment. As grains are expelled and powder is formed, the surface area of the UO_2 increases (White et al. 1983), which enhances the rate of weight gain because oxygen has a larger surface area for entry into the pellet or fragment. The rate of weight gain increases (Figure 15) as the transition from the preinduction to the postinduction region progresses. Extrapolation of the post-transition curve to zero weight gain defines an approximate boundary between the pre- and postinduction regions and is labeled as t_1 --the induction time for powder formation (Simpson and Wood 1983).

The induction time depends on temperature and neutron irradiation as illustrated by the data in Figure 16. Central Electric Generating Board (CEGB) data (Simpson and Wood 1983) on unirradiated UO_2 show a consistently longer induction time than data for unirradiated pellets generated at PNL (White et al. 1983). PNL data for irradiated LWR spent fuel fragments (White et al. 1983) and CANDU data for irradiated SF fragments (Hastings 1983) show a consistently shorter induction time than for unirradiated pellets. Since the

FIGURE 16. Temperature Dependence of Time for Powder Formation in UO₂ in Air

temperature dependence for both irradiated and unirradiated UO₂ appears similar, the induction time can be described by the following relationship between induction time and temperature:

$$t_I = \alpha A F(T) \quad (1)$$

where t_I = time in years for the start of spalling

α = irradiation factor

A = coefficient (7.1×10^{-16} yr for PNL data on unirradiated UO₂)

$F(T) = \exp (Q/RT)$, where Q is the activation parameter (31,500 cal/g atom); R is 1.987 cal/g atom-K; and T is fuel temperature (K).

Linear oxidation kinetics were used in accordance with the results in White et al. (1983). The value of α derived by comparing irradiated and unirradiated data in Figure 16 is 0.08.

The value of Q is similar to that for diffusion of oxygen in UO_2 . If the temperature decreases, the rate of oxygen diffusion in UO_{2+x} decreases and the rate of approach to U_3O_8 formation is retarded. The temperature of spent LWR fuel decreases with time during dry storage due to a reduction in the concentration of heat-generating fission products associated with radioactive transmutations.

Calculated temperature histories for irradiated fuel in a hypothetical dry storage cask (Johnson, Gilbert, and Guenther 1983) and an FRG cask (Peehs, Kuhnel, and Kaspar 1982) are shown in Figure 17 and empirically described by Equation (2). Decay heat decreases rapidly at first but more slowly as the radioactive isotopes decay. The rate of temperature decrease depends on fuel enrichment, burnup, age of the fuel as it enters dry storage, and thermal properties of the storage system.

FIGURE 17. Temperature History for Spent Fuel in PWR Storage Casks

$$T = (T_1 - T_0)(t + a)^\gamma + T_0 \quad (2)$$

where T = temperature (K) in dry storage at time (years)

T_1 = temperature (K) of the fuel if it was placed in dry storage
1 year after reactor discharge

a = age of the fuel (years) when placed in dry storage

T_0 = ambient external temperature (K)

γ = a constant equal to -0.43 for the hypothetical cask and -0.6 for
Federal Republic of Germany (FRG) PWR-type casks (Peehs, Kuhnel,
and Kaspar 1982).

Substitution of Equation (2) into the differentiated form of Equation (1)
yields the following equation, applicable to predicting the start of powder
formation in unlimited air during dry storage under a decreasing decay heat:

$$\int_0^t \left\{ F \left[(T_1 - T_0)(t + a)^\gamma + T_0 \right] \right\}^{-1} dt \leq \alpha A \quad (3)$$

The cumulative oxidation damage fraction--the value of the integral in Equation (3) divided by A --is plotted versus time in Figure 18 for the temperature/time history of the hypothetical cask shown in Figure 17 and the fuel oxidation properties measured at PNL (White et al. 1983). For an initial storage temperature of 179°C, a cumulative damage fraction just slightly below unity is predicted for unirradiated UO_2 ($\alpha = 1.0$). Therefore, powder formation would not be expected for times less than 100 years.

The potential for oxidation is highest during the first decade of storage. After the first decade, the temperature is predicted to be too low for oxidation to proceed significantly. The corresponding initial storage temperature for irradiated fuel ($\alpha = 0.08$ from Figure 16) to prevent powder formation is 147°C. These calculations were performed for SF that entered dry storage 5 years after reactor discharge.

A set of computations was performed similar to those used to derive Figure 18, except that the cooling curve (Figure 17) for an FRG-type PWR cask was used. The results are plotted in Figure 19 for unirradiated ($\alpha = 1.0$) and

FIGURE 18. Cumulative Oxidation Damage for 5-Year-Old Fuel Placed in a Hypothetical PWR Storage Cask

irradiated ($\alpha = 0.08$) fuel. The computed allowable starting temperatures to prevent powder formation in unirradiated and irradiated fuel are 185°C and 152°C, respectively.

SF storage configurations will have a major impact on temperature during fuel storage. For example, due to higher fuel inventories and slower cooling rates, consolidated older fuels may remain at higher temperatures during storage than younger, unconsolidated fuels. Therefore, the cumulative oxidation fraction must reflect not only the temperature at the start of storage but also the fuel configuration.

The effect of cooling time prior to dry storage on maximum allowable temperatures is illustrated in Figure 20. The maximum initial storage temperatures that could be tolerated before powder formation in irradiated fuel ($\alpha = 0.08$) for the hypothetical (Johnson, Gilbert, and Guenther 1983) and FRG cask

FIGURE 19. Cumulative Oxidation Damage for 5-Year-Old Fuel Placed in an FRG-type Storage Cask

cask cooling curves are plotted versus the SF age. The calculations indicate that older SF requires lower initial storage temperatures, due to the slower cooling rate.

The benefit of using the cumulative oxidation damage method for predicting storage temperature limits is that the predictions are ~35 to 40°C higher than for isothermal methods. This methodology can also be used to predict temperatures during storage in inert gas below which gas monitoring is no longer required. It can also be used to compute the potential oxidation damage incurred during postulated abnormal events.

4.2 CLADDING DEGRADATION

Since SF demonstrations and tests conducted to develop performance and design criteria for dry storage are limited to less than 8 years (Johnson,

FIGURE 20. Maximum Temperature to Prevent U_3O_8 Formation in Irradiated UO_2 Under Cooling Conditions for Two Casks Versus Age of Fuel at Insertion

Gilbert, and Guenther 1983), extrapolations of performance to longer storage times anticipated for MRS are supported by models. Analyses of SF behavior based on stress rupture and SCC degradation mechanisms predict allowable storage temperatures of 150 to 430°C in inert gas, depending on the condition of the fuel. These mechanisms as well as hydride reorientation phenomena are described below.

Nonoxidizing atmospheres will prevent oxidation of UO_2 and formation of particulate fuel in the breached fuel. However, reactor-breached fuel will probably be a source of moisture in the canister or storage system due to the hygroscopic nature of UO_2 and its exposure to high-pressure reactor coolant and water during pool storage. Experience has shown that 5 to 10 g of moisture can be released by heating a reactor-breached BWR fuel rod (Johnson et al. 1983). In a PWR storage cask (Eggers 1982) containing ~5000 SF rods (10,000 in the

consolidated configuration), the number of undetected breached rods corresponding to a 0.1% reactor-induced failure rate and a 90% detection rate (assuming failed rods are separated) is approximately one rod per cask. The introduction of moisture from one reactor-breached, water-logged fuel rod is estimated to be negligible (Johnson and Gilbert 1983), even if the resulting reaction with Zircaloy cladding were concentrated at the hottest 10% of the fuel cladding area.

Analytical studies have been conducted to estimate the susceptibility of LWR spent fuel cladding to breaching during dry storage from SCC and stress rupture (Blackburn et al. 1978; Miller and Tasooji 1983; Chin, Madsen, and Khan 1983). The stress is generated by the gas pressure contained within the fuel cladding. The first analysis of failure by stress rupture (Blackburn et al. 1978) was based on a Larson-Miller parametric evaluation of stress rupture behavior of unirradiated Zircaloy. It used high stress data and did not account for unknown effects of irradiation or for the unknown behavior of Zircaloy at the low stresses encountered during storage, where the deformation and failure mechanisms are different than at high stresses. Allowable storage temperatures on the order of 380 to 400°C were predicted to result in low probability of cladding breach by stress rupture.

Analytical predictions of allowable storage temperatures to prevent cladding breach by SCC (Blackburn et al. 1978; Miller and Tasooji 1983) were found to be very sensitive to assumed cladding defects and assumed presence of reactive fission products such as iodine attacking the inner wall surface of the cladding. The predictions of allowable storage temperatures to prevent cladding breach by SCC during dry storage range from 150 to 400°C, depending on the assumed aggressiveness of the fission products reacting with the cladding and the assumed severity of the cladding defects.

A study was undertaken to determine maximum allowable storage temperatures to prevent cladding breach by stress rupture taking into account the temperature decrease due to thermal decay as well as the effects of irradiation on cladding properties (Chin, Madsen, and Khan 1983). This study was based on the behavior of dominant deformation mechanisms shown on the map in Figure 21. The dependence of deformation mechanisms on temperature, stress, and time led to the prediction of dominant failure mechanisms. A map showing the failure mechanisms is presented in Figure 22.

FIGURE 21. Theoretical Deformation Map for Zircaloy (5- μ m grain size)

FIGURE 22. Theoretical Fracture Map for Zircaloy (5-μm grain size)

The failure mechanisms map in Figure 22 shows that at stresses above 300 MPa failure occurs by transgranular cracking. Cracks initiate and grow by formation and linking of small cavities within Zircaloy grains. Large strains are required to propagate the cracks across grains and produce failure. Cladding stresses during dry storage are not predicted to be high enough for failure by this mechanism except at a deep incipient crack. During the final stages of failure, the stress on a small remaining ligament of cladding might approach 300 MPa.

At stresses between 200 and 300 MPa, failure occurs by triple-point cracking at grain boundaries. The triple-point cracking is heterogeneous with cracks initiating on cladding surfaces at grain boundary intersections (triple points). Because of this heterogeneity, the cracking is susceptible to enhancement by corrosive fission products. SCC is associated with the triple-point cracking region. Irradiation, which strengthens the grains with respect to weaker grain boundaries, enhances the occurrence of triple-point cracking and SCC. Consequently, the triple-point cracking region in Figure 22 depicts the conditions under which SCC is most likely to occur. The tendency for irradiation to enlarge this region is illustrated in Figure 23. Failure data on irradiated SF cladding tested in an iodine environment (Yaggee, Mattas, and Neimark 1979) display triple-point cracking outside the domain boundaries identified for unirradiated Zircaloy and thus reveal an expanding effect of irradiation and aggressive chemical environment.

At stresses below 200 MPa, failure occurs by growth and linking of cavities on grain boundaries (grain boundary cavitation). This failure mode accommodates more strain than triple-point cracking and an increase in ductility is encountered as lower stresses are imposed. The cavities form homogeneously on grain boundaries and do not become linked until late stages of deformation. Consequently, failure by grain boundary cavitation has low susceptibility to enhancement by corrosive environments.

The hoop stresses in spent LWR fuel place the failure mode in the grain boundary cavitation region (Figure 24). Only if severe aggressive fission

FIGURE 23. Comparison of Zircaloy Fracture Map Predictions with Irradiated Data Field (5- μm grain size)

FIGURE 24. Comparison of Zircaloy Fracture Map Predictions with Data Field (5- μm grain size)

products or severe stress concentrations were encountered at reactor-induced incipient defects would the stresses be high enough to produce failure by triple-point cracking or SCC.

The deformation and failure mapping methodology was applied to predict the maximum initial storage temperature to prevent failure by stress rupture, assuming that the fuel would cool in accordance with hypothetical curves presented in Johnson, Gilbert, and Guenther (1983) and in Figure 17. For unirradiated and irradiated Zircaloy, the values range from 300 to 430°C depending on the age of the fuel (Figure 25) and the amount of irradiation.

The grain boundary cavitation mode of failure produces minute pinhole leaks ($\sim 10\text{ }\mu\text{m}$ in diameter). These holes vent fission gases from the fuel rod. The holes are not expected to be large enough to allow fuel loss. Further cladding degradation would require the introduction of an oxidizing

FIGURE 25. Modified Predictions of Maximum Allowable Initial Dry Storage Temperatures as a Function of Time in Wet Pool Storage

atmosphere to oxidize the fuel. If sufficient fuel swelling occurs from fuel oxidation, then the cladding could split as illustrated in Figure 26 (Johnson et al. 1983).

Unfortunately, the analyses of SF cladding failure by stress rupture are based primarily on theory and stress rupture data on unirradiated Zircaloy. Data on irradiated SF cladding extend only to less than 100 h (Yaggee, Mattas, and Neimark 1979). The cumulative damage modeling approach indicates that most of the damage is generated in the first 10 years of storage due to the higher temperatures (Figure 27). Stress rupture tests with durations of only a few years could confirm the cumulative damage methodology and provide property data that now require conjecture.

The hydrogen concentration in SF Zircaloy cladding is normally from 20 to 150 wppm. The temperature boundary for solubility of hydrogen in Zircaloy is shown in Figure 28. If the temperature falls below the solubility limit, zirconium hydrides precipitate. The hydrides normally orient circumferentially and axially; however, radial orientation can be induced by sufficiently high stresses. The ductility of Zircaloy cladding with radially oriented hydrides, even with hydrogen contents as low as 40 wppm, is severely reduced (Marshall

FIGURE 26. Example of Split Cladding

FIGURE 27. Accumulated Damage as a Function of Dry Storage for Various Ages of Spent Fuel. Note that over 80% of the damage is accumulated in the first 25% of storage time.

1967). During SF storage, the fuel at peak temperature conditions may be above the solubility limit (Figure 28). As it cools, the cladding will pass through the solubility limit.

If the hoop stresses were sufficiently high to cause radial orientation and if the creep strain generated after hydride formation exceeded the ductility in the hydrided state, cladding breaches could be produced. It is unlikely that very much creep strain would be generated after the fuel had cooled below the solubility limit; however, detailed analyses have not been performed over the range of dry storage conditions.

FIGURE 28. Effect of Spent Fuel Storage Temperature on Susceptibility of Zircaloy Cladding to Hydride Reorientation

5.0 REFERENCES

The references for this report have been separated by subject area: spent fuel, high-level waste, transuranic waste, and degradation modeling. Entries are arranged alphabetically within these sections.

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