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Report on Intact and Degraded Criticality for Selected Plutonium Waste Forms in a Geologic Repository, Volume I: MOX SNF

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Report on Intact and Degraded Criticality for Selected Plutonium Waste Forms in a Geologic Repository, Volume I: MOX SNF

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Report on Intact and Degraded Criticality for Selected Plutonium Waste Forms in a Geologic Repository, Volume I: MOX SNF

EXECUTIVE SUMMARY

As part of the plutonium waste form development and down-select process, repository analyses have been conducted to evaluate the long-term performance of these forms for repository acceptance. Intact and degraded mode criticality analysis of the mixed oxide (MOX) spent fuel is presented in Volume I, while Volume II presents the evaluations of the waste form containing plutonium immobilized in a ceramic matrix.

Although the ceramic immobilization development program is ongoing, and refinements are still being developed and evaluated, this analysis provides value through quick feed-back to this development process, and as preparation for the analysis that will be conducted starting in fiscal year (FY) 1999 in support of the License Application.

While no MOX fuel has been generated in the United States using weapons-usable plutonium, Oak Ridge National Laboratory (ORNL) has conducted calculations on Westinghouse-type reactors to determine the expected characteristics of such a fuel. These spent nuclear fuel (SNF) characteristics have been used to determine the long-term potential for criticality in a repository environment.

In all instances the methodology and scenarios used in these analyses are compatible with those developed and used for Commercial Spent Nuclear Fuel (CSNF) and Defense High Level Waste (DHLW), as tailored for the particular characteristics of the waste forms. This provides a common basis for comparison of the results.

This analysis utilizes dissolution, solubility, and thermodynamic data that are currently available. Additional data on long-term behavior is being developed, and later analyses (FY 99) to support the License Application will use the very latest information that has been generated. Ranges of parameter values are considered to reflect sensitivity to uncertainty. Most of the analysis is focused on those parameter values that produce the worst case results, so that potential licensing issues can be identified.

MOX (Volume I)

This study is concerned with evaluating the criticality potential of the intact and degraded forms of the MOX SNF in waste packages (WPs). Current WP designs for both the 21 PWR WP and the 12 PWR WP are analyzed. Aluminum thermal shunts were used in both designs to enhance the heat flow rate.

This study also includes an evaluation of the structural, thermal, and shielding impacts of the MOX SNF WP's. Although previous analyses showed these impacts to be within regulatory and safety requirements, a more comprehensive evaluation is appropriate at this time to reflect the current MOX design and to prepare for the License Application analysis phase.

Since the MOX WP's will have criticality performance very similar to the waste packages containing commercial low enriched uranium (LEU) SNF, the criticality evaluations follow the same methodology of initial analysis with the following steps:

1. Criticality evaluation of the intact configuration to demonstrate the effectiveness of the criticality control measures,

2. Criticality evaluation of the same degraded basket configurations that have been used for the commercial LEU SNF,

3. Determination of the configurations having both degraded basket and degraded MOX SNF, using the geochemistry code, EQ6, as has also been done for the commercial LEU SNF, and

4. Criticality evaluation of the combined degraded basket and degraded SNF configurations.

Major Findings, MOX (Volume I)

Based on the current available data and designs for the MOX fuel, we find the following:

1. Assuming that the MOX SNF will be emplaced at least 10 years following discharge, those assemblies having low burnup (≤46 GWd/MTHM) can be loaded into the standard commercial 21 PWR WP, and those assemblies having high burnup (> 46 GWd/MTHM) can be loaded into the standard commercial 12 PWR WP. This strategy will meet the maximum thermal output design criteria of 18 kW per package. With the expected distribution of burnups in the MOX SNF, this strategy will result in approximately half the MOX assemblies being placed in each of the two types of waste package. This emplacement strategy will also have the following performance aspects:

   - The MOX SNF waste packages meet all regulatory requirements.
   - There is no credible intact or degradation scenario leading to an internal criticality in the waste packages.
   - Structural, thermal, and shielding impacts are no greater (and may be less) than those of the corresponding commercial SNF waste packages.

2. The most severe structural hazard to the waste package is modeled by a finite element analysis of a tipover accident. It is found that the peak stress in the waste package, resulting from such an event, will be at least 15% less than the ultimate material tensile strength of the material. This shows that the structural behavior of both the 21 PWR WP and the 12 PWR WP will be within design limits. The MOX SNF WP stress values are very similar to values calculated for commercial SNF WP's, as would be expected, since both fuel types have similar SNF assembly weights.

3. Assuming that the MOX SNF will be emplaced at least 10 years following discharge, the maximum initial heating rates for the MOX SNF were 798 watts/assembly for the 21 PWR WP and 1070 watts/assembly for the 12 PWR WP. These values are less than the 850 watts/assembly and 1500 watts/assembly used as the thermal design basis (maximum thermal output of 18 kW per disposal container) for commercial LEU PWR SNF, indicating that the MOX assemblies are well within the design envelope of the commercial SNF WP. The peak fuel temperature calculated for the 21 MOX PWR WP was approximately 336°C, and that for the 12 MOX PWR was approximately 302°C. These temperatures are well below the established design limit of 350°C.

4. Dose rates from both neutron and gamma radiation were calculated for the 21 PWR WP loaded with the highest burnup MOX SNF and the shortest cooling period after reactor discharge (10 years) to serve as a worst case that would give the highest dose rates. Maximum dose values at the exterior surfaces of the waste package were less than 110 rad/hr. Maximum dose rates from the MOX SNF were much less than from commercial LEU PWR SNF of similar burnup which were calculated to be greater than 150 rad/hr. The 12 MOX PWR WP design has an equivalent amount of shielding with a smaller radiation source, which should result in smaller surface dose rates.

   The design limit of 100 rad/h on the surface rate was specified so that no significant increase could occur in the corrosion rate of the waste package barrier due to any radiolytic compounds synthesized from moist air. For both waste packages, the SNF surface dose rate exceeded the design limit only during the period immediately following emplacement when humidity in the external environment is expected to be low. It is concluded, therefore, that no increase in corrosion rates from radiolysis will occur.

5. Criticality evaluations were performed for the 21 PWR MOX SNF WP and the 12 PWR MOX SNF WP for conditions ranging from intact to fully degraded fuel and basket. The peak k_{eff}'s ranged from 0.55 to 0.90 where the 0.90 resulted from a worst case configuration. The following observations on the criticality potential of the PWR MOX SNF can be made:

   - The 12 PWR WP has a higher k_{eff} than the 21 PWR WP for the flooded conditions with intact fuel and basket because the 12 PWR WP has no neutron absorber plates.
   - The 12 PWR WP has a lower k_{eff} than the 21 PWR WP for the flooded conditions with intact fuel, but with degraded basket, because the iron oxide corrosion products displace moderator compensating, in part, for the absence of absorber plates.
   - The worst case k_{eff} is below the criticality limit of 0.92 for any credible configuration and thus a criticality event internal to the waste package is virtually impossible.

Ceramic (Volume II)

For the ceramic waste form the principal criticality control measure is the incorporation of neutron absorbing material in the waste form itself. The potential for criticality is determined

primarily by the amount of such neutron absorber material remaining in the waste package if, and when, the waste package is breached, and its contents are thereby exposed to aqueous corrosion. Under such conditions the waste form can be corroded; the fissile material in the waste form (either plutonium or its decay product uranium) will remain in the waste package for hundreds of thousands of years, because it is very insoluble under most water chemistry conditions. The neutron absorber hafnium is even less soluble than the fissile material so it will remain in the WP. However, the more neutronically efficient absorber, gadolinium, could become more soluble under some conditions and could eventually be flushed from the waste package.

This study is concerned with evaluating the potential for criticality of the currently defined ceramic waste form. After a few criticality calculations to demonstrate that the intact configuration is safely below the critical limit, the study is focused on identifying those degraded configurations that are most reactive (result in the highest values of the neutron multiplication factor, k_{eff}). The degraded configurations having the greatest potential for criticality are selected out of the range of configurations arising from the set of degradation scenarios analyzed with the geochemistry code, EQ6. The degradation scenarios examined with the geochemistry code are those most likely to lead to a loss of a major fraction of the neutron absorber material, by virtue of an increase in the solubility of that material.

Shielding, thermal, and structural evaluations were not performed explicitly for the immobilized Pu waste package because the comparison cases with the DWPF WP had not yet been completely evaluated. Nevertheless, conservative comparison with previous evaluations of a similar WP concept does support a finding that inclusion of the immobilized plutonium has a negligible repository impact.

Major Findings, Ceramic (Volume II)

Based on the data presently available, and the current canister loading of the current ceramic formulation (28.7 kg of Pu per canister), we find the following:

1. The ceramic plutonium waste form can be emplaced in the repository at a loading of 5 plutonium containing canisters per waste package; this permits the disposal of immobilized plutonium in the same disposal container/waste package as will be used for the disposal of high level waste (HLW) glass.

    - The ceramic plutonium waste package meets all regulatory requirements.
    - There is no credible degradation scenario leading to criticality internal to the waste package.
    - Thermal and shielding impacts are comparable to, or less than, those of the corresponding HLW waste package.

2. The completely intact configuration has virtually no potential for criticality, since the calculated $k_{eff} = 0.12$ for the unbreached wasted package, and $k_{eff} = 0.11$ when all of the void space in the waste package is filled with water.

3. The processes in the expected degradation scenarios will generally have the following sequence:

   - breach of the waste package by aqueous corrosion, and wetting of all interior surfaces,
   - breach of the stainless steel canisters containing the HLW filler glass and the plutonium ceramic waste form,
   - dissolution of the filler glass,
   - breach of the inner cans that actually contain the plutonium ceramic disks,
   - corrosion of the stainless steel of the canisters and cans, and
   - dissolution of the ceramic waste form.

   Many of these processes will overlap in time. In fact, the overlap of the last two processes (corrosion of the stainless steel and dissolution of the ceramic waste form) is what gives rise to the possibility of gadolinium removal.

4. The degraded configurations are divided into two types:

   - intermediate-level degraded, in which the ceramic disks remain intact, while all the other components of the waste package have been degraded or fragmented (and the soluble degradation products are removed from the waste package), and
   - fully collapsed, in which the ceramic disks are also degraded and/or fragmented and all the fragments and insoluble degradation products mixed into a homogeneous layer at the bottom of the waste package.

5. The following are the principal criticality (k_{eff}) results for the worst cases of these two configuration types:

   - For the intermediate degraded configurations there will be no significant loss of the principal neutron absorber, gadolinium, and: $k_{eff} < 0.38$.
   - For the fully collapsed configurations there could be as much as a 13% loss of the neutron absorbing gadolinium, but the more dominating effect is the geometry being less favorable to criticality than the intermediate degraded configurations, so that $k_{eff} < 0.33$, which is less than 0.38 for the intermediate degraded configurations.

ACRONYMS

AML	Areal Mass Loading
AUCF	Advanced Uncanistered Fuel
B&W	Babcock and Wilcox
BOC	Beginning of Cycle
B-SS	Borated Stainless Steel
CDA	Controlled Design Assumptions
DBE	Design Basis Event
DWPF	Defense Waste Processing Facility
FY 99	Fiscal Year 1999
GWd	Gigawatt Days
HFP	Hot Full Power
HEU	Highly Enriched Uranium
HM	Heavy Metal
LEU	Low Enriched Uranium
MGR	Monitored Geologic Repository
MOX	Mixed Oxide Fuel
MTHM	Metric Tons Heavy Metal
PI	Principal Isotopes
PWR	Pressurized Water Reactor
SDD	System Description Document
SNF	Spent Nuclear Fuel
TBV	To Be Verified
UCF	Uncanistered Fuel
USDOE	United States Department of Energy
WP	Waste Package
2-D	Two Dimensional
3-D	Three Dimensional

Report on Intact and Degraded Criticality for Selected Plutonium Waste Forms in a Geologic Repository, Volume I: MOX SNF

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Report on Intact and Degraded Criticality for Selected Plutonium Waste Forms in a Geologic Repository, Volume I: MOX SNF

1. INTRODUCTION AND BACKGROUND

As part of the plutonium waste form development and down-select process, repository analyses have been conducted to evaluate the long-term performance of these forms for repository acceptance. This volume assesses the intact and degraded mode criticality of the mixed oxide (MOX) spent nuclear fuel (SNF). Volume II conducts this assessment for the plutonium waste form immobilized in a ceramic matrix.

Although no MOX fuel has been generated in the United States using weapons-usable plutonium, Oak Ridge National Laboratory (ORNL) has conducted calculations on Westinghouse-type reactors to determine the expected characteristics of such a fuel. This SNF characteristic has been used to determine the long-term potential for criticality in a repository environment.

In all instances, the methodology and scenarios used in these analyses, as tailored for the particular characteristics of the waste forms, are compatible with those developed and used for the Commercial Spent Nuclear Fuel (CSNF) and Defense High Level Waste (DHLW). This provides a common basis for comparison of the results.

This analysis utilizes dissolution, solubility, and thermodynamic data that are currently available. As additional data becomes available, it will be used to support the License Application. Where applicable, ranges of values will be used to bound the results.

The content of this volume is organized as follows:

- Section 2 provides a brief description of the waste packages and the plutonium waste forms and quantities.
- Section 3 discusses the structural analyses conducted on the waste packages.
- Section 4 discusses the thermal analyses conducted on the waste packages.
- Section 5 discusses the radiation shielding analyses conducted on the waste packages.
- Section 6 discusses the criticality evaluations performed on the intact and degraded MOX waste forms. Degradation configurations important for criticality considerations were calculated with the geochemistry code EQ3/6. Results of these calculations were utilized in the criticality evaluations of the degraded configurations and compared to criticality evaluations of low enriched uranium (LEU) SNF.
- Section 7 summarizes the major findings from this study.

This document has been prepared according to Procedure PRO-TS-003, *Development of Technical Documents Not Subject to QARD Requirements*. The specific activities involved with the production and review of this document have been performed according to an approved Technical Document Preparation Plan (Reference 1).

2. WASTE FORM, WASTE PACKAGE, AND WASTE STREAM QUANTITIES

2.1 MOX SNF Characteristics

The potential use of MOX fuel in power reactors has been investigated through the development of conceptual designs for commercial pressurized water reactor (PWR) equilibrium reload cycles fueled with MOX assemblies (Ref. 2 and Ref. 3). The most recent design, documented in Reference 3, utilizes 92 fresh MOX assemblies per reload cycle. Two values of fissile Pu, given as weight percent fissile Pu in the heavy metal (HM), were used in this design (Ref. 3, p. 2-11). The fresh reload batch consisted of 20 assemblies with a 4.5 wt% fissile Pu in HM and 72 assemblies with a 4.0 wt% fissile Pu in HM. The core loading for this design was 81.6 metric tons of heavy metal (Ref. 3, p. 2-9) resulting in an average Pu content of 18.48 kg/assembly. The average burnup for assemblies was targeted at 45 to 50 GWd/MTHM and ranged from a low value of approximately 35 to a high value of approximately 56 GWd/MTHM (Ref. 3, p. 2-39). The steady state discharge distribution consists of 83 assemblies burned for two cycles and 9 assemblies for three cycles (Ref. 3, p. 2-26). All assemblies burned for three cycles were of the 4.0 wt% fissile Pu in HM type.

The conceptual core design documented in Reference 3 utilized the Westinghouse 17x17 Vantage 5 commercial assembly type (Ref. 4, p. 2A-30) and is the reference design for this study. Detailed mechanical parameters for these assemblies are given in Reference 5 (Table 5.1-2) and summarized in Table 2.1-1. Assembly dimensions are given primarily in English units and converted into metric units to maintain consistency between calculations using either set of units. Assembly weights are used in the structural analysis (Section 3).

The initial heavy metal isotopic content of the PWR MOX Westinghouse Vantage 5 assembly fuel important for repository considerations is given in Table 2.1-2.

Table 2.1-1. Mechanical Parameters for Westinghouse 17x17 MOX Fuel Assemblies

	Vantage 5 Assembly		
Parameter	Value Metric Units	Value English Units	References
Fuel Length	365.76 cm	144 in.	3, p. 2-9
Heavy Metal Mass	422.8 kg	932.1 lb	5, p. 12
Assembly Weight	618.8 kg	1364.2 lb	6, p. 6
Weight of Non-fuel Material/Assembly	54.4 kg	120 lb	6, p. 6

Table 2.1-2. Initial Heavy Metal Isotopic Content (wt%) of MOX SNF Assemblies

Isotopes	Vantage 5 Assemblies (4.0 wt% Fissile Pu in HM)[1]	Vantage 5 Assemblies (4.5 wt% Fissile Pu in HM)[1]
^{235}U	0.191	0.190
^{238}U	95.550	95.019
^{239}Pu	3.983	4.481
^{240}Pu	0.251	0.282

[1] Derived from isotopic wt% Ref. 3, p. 2-10. Isotopes comprising < 0.01 wt% not listed.

The characterization of the potential MOX assemblies with respect to the content of those SNF isotopes of greatest abundance or of most neutronic significance was calculated (Ref. 5) with the SAS2H computer code and the ORIGEN-S computer code. The SAS2H and ORIGEN-S codes are part of the SCALE Code System, Version 4.3 (Computer Software Configuration Item [CSCI]: 30011 V4.3) (Ref. 7). A one axial node SAS2H model of the MOX assembly was developed to perform the depletion steps. The multi-cycle burnup histories were derived from the equilibrium MOX core load map (Figure 2-8, Ref. 3, [p. 2-26]). Results from this analysis formed the source data for criticality, thermal, and radiation shielding evaluations of waste package designs for MOX assemblies in the Monitored Geologic Repository (MGR). The particular cases selected under the above criteria are given in Table 2.1-3 together with the controlling criteria. The beginning-of-cycle (BOC) soluble boron concentration (40% ^{10}B, [Ref. 3, p. 2-14]) was 1301 ppm at hot full power (HFP) conditions and tracked approximately through a reload cycle by a series of burnup steps (Ref. 5, Section 5.5).

Table 2.1-3. MOX Assembly Selection Criteria

Case ID	wt% fissile Pu in HM	Discharge Burnup (GWd/MTHM)[1]	Controlling Criteria for Selection
1	4.0	56.5	Heat Generation
2	4.5	46.5	Heat Generation
3	4.0	50.1	Heat Generation; Criticality
4	4.0	35.6	Criticality
5	4.5	39.4	Criticality

[1] gigawatt-days per metric ton heavy metal.

Results from the analysis of Westinghouse MOX SNF relevant to the purpose of this calculation include the thermal power generation and isotopic content of the MOX SNF assemblies as a function of time after discharge from the reactor (Ref. 44). Representative results from the analyses are given in Table 2.1-4 for the thermal power generation in the MOX SNF assemblies.

The total thermal power per assembly generated for each of the heat generation cases as shown in Table 2.1-4 is for a period of 10,000 years beginning 10 years after discharge from the reactor (CDA Key 004 [Ref. 28] specifies that the initial SNF for the repository be at least 10 years old). The total thermal power in the table is the sum of the thermal power generated by radioactive decay of

activated light elements, actinides, and fission products. The heating rate contribution from the different components varies with the assembly burnup value since the SNF isotopic composition is burnup dependent. Over short time periods, heating rates show a direct correlation with burnup due to the short lived isotopes. This correlation does not hold over longer time periods as can be seen by comparing the second and third columns of Table 2.1-4 for times beyond 40 years. This effect is shown graphically in Figure 2.1-1 and also results in a larger source for MOX SNF at times greater than 100-1000 years than is present for LEU SNF (Ref. 24, Figure 5.1). These heating rate values are used as source terms for thermal calculations for the waste packages discussed in Section 4.

Table 2.1-4. Total Thermal Power Generated from Westinghouse MOX SNF

Time (years)	Total Thermal Power Generation/Assembly (watts)		
	56.5 GWd/MTHM	50.1 GWd/MTHM	46.5 GWd/MTHM
0.5	1044.8	871.9	785.3
1.0	1027.6	856.8	772.4
2.0	995.5	831.7	753.5
3.0	968.5	809.9	736.0
4.0	944.9	791.3	721.6
5.0	922.4	773.9	709.4
6.0	903.1	758.7	697.5
7.0	884.0	744.6	687.5
8.0	867.0	731.7	676.7
9.0	850.2	719.9	668.0
10.0	835.4	708.1	659.4
20.0	707.5	613.4	587.2
30.0	613.4	541.4	530.3
40.0	541.1	485.1	485.1
50.0	483.1	440.1	446.0
60.0	438.0	403.4	415.7
70.0	401.0	374.3	390.0
80.0	371.5	349.4	368.2
90.0	347.8	329.9	349.6
100.0	327.9	313.1	334.5
250.0	210.2	211.1	233.0
500.0	144.0	148.0	165.0
750.0	106.0	109.0	122.0
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5000.0	23.4	24.6	28.6
10000.0	16.1	17.0	20.0

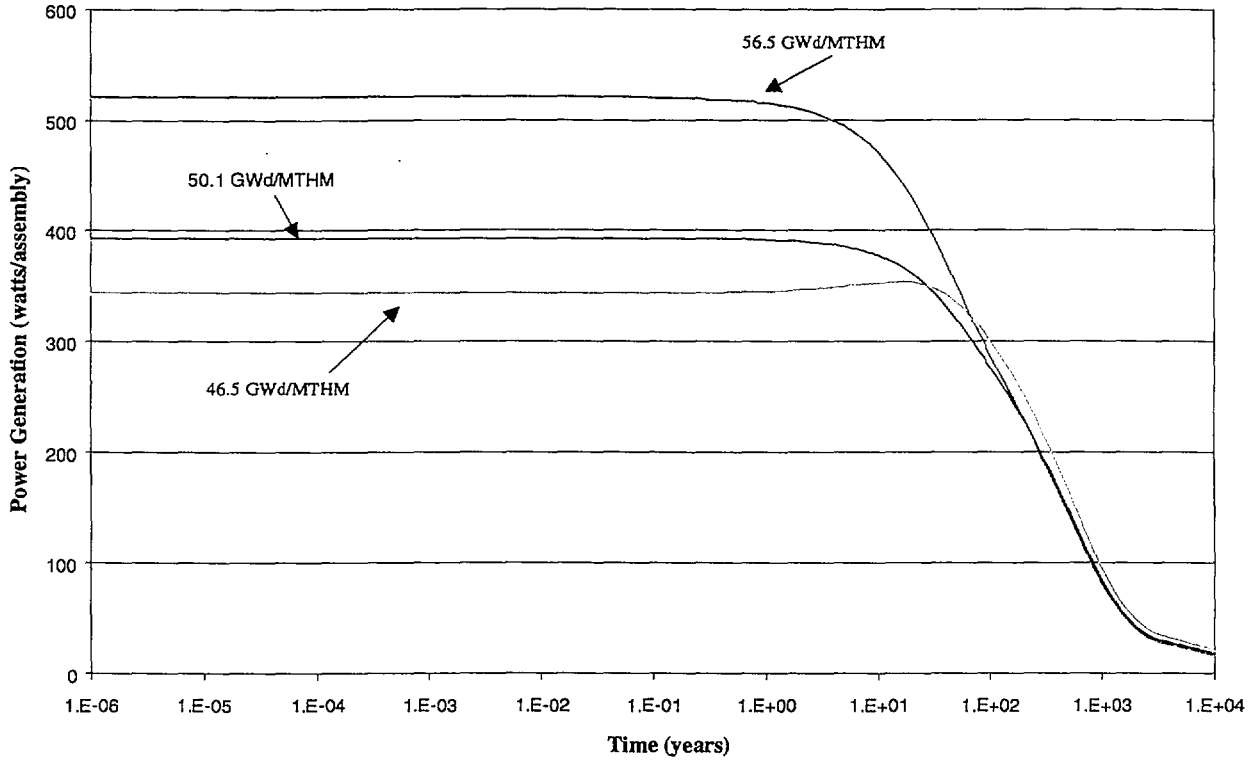

Figure 2.1-1. Thermal Power Generation from MOX SNF Actinide Composition.

Principal Isotope (PI) burnup credit is assumed to be an acceptable method to account for reduced reactivity of SNF in criticality evaluations (CDA Key 009, Ref. 28). A list of 29 "Principal Isotopes" for long-term criticality control in SNF has been previously established (Ref. 8, p. 3-26). The concentrations of these isotopes as a function of time derived from the SAS2H/ORIGEN-S analysis of the MOX SNF is used in the criticality analysis discussed in Section 6.

2.2 Waste Package Description

Waste packages considered for MOX SNF are the 21 PWR Advanced Uncanistered Fuel (AUCF) waste package (WP) and the 12-PWR AUCF waste package which are the same as the current designs for commercial LEU SNF (Ref. 9, Section 8 and Ref. 10, Section 8). These waste packages are illustrated in Figure 2.2-1 and in Figure 2.2-2, respectively. These illustrations depict the waste packages, their internals, and the material specifications. Both designs incorporate techniques to limit the maximum anticipated temperatures in the waste package and fuel cladding materials. The 21 PWR WP design also incorporates borated stainless steel (B-SS) plates in the basket assembly for criticality control. The absorber plates are needed because the MOX assemblies proposed for disposal in this waste package design have the lowest burnup levels and consequently greater fissile Pu content. The nominal 12 PWR WP design does not contain B-SS absorber plates since it is to be used only for high burnup assemblies and the analysis is more conservative by not considering such absorber plates. (Borated stainless steel absorber plates can be used in the 12 PWR WP but are not

required for criticality control. Use of absorber plates in the 12PWR WP would decrease the criticality potential of the 12 PWR WP even further and, thus, were not considered in this analysis). In the uncanistered waste package design, SNF assemblies are placed directly into the steel basket assemblies enclosed within the corrosion resistant and corrosion allowance barriers. The design for the corrosion barrier includes a corrosion allowance outer barrier material and a corrosion resistant inner barrier material.

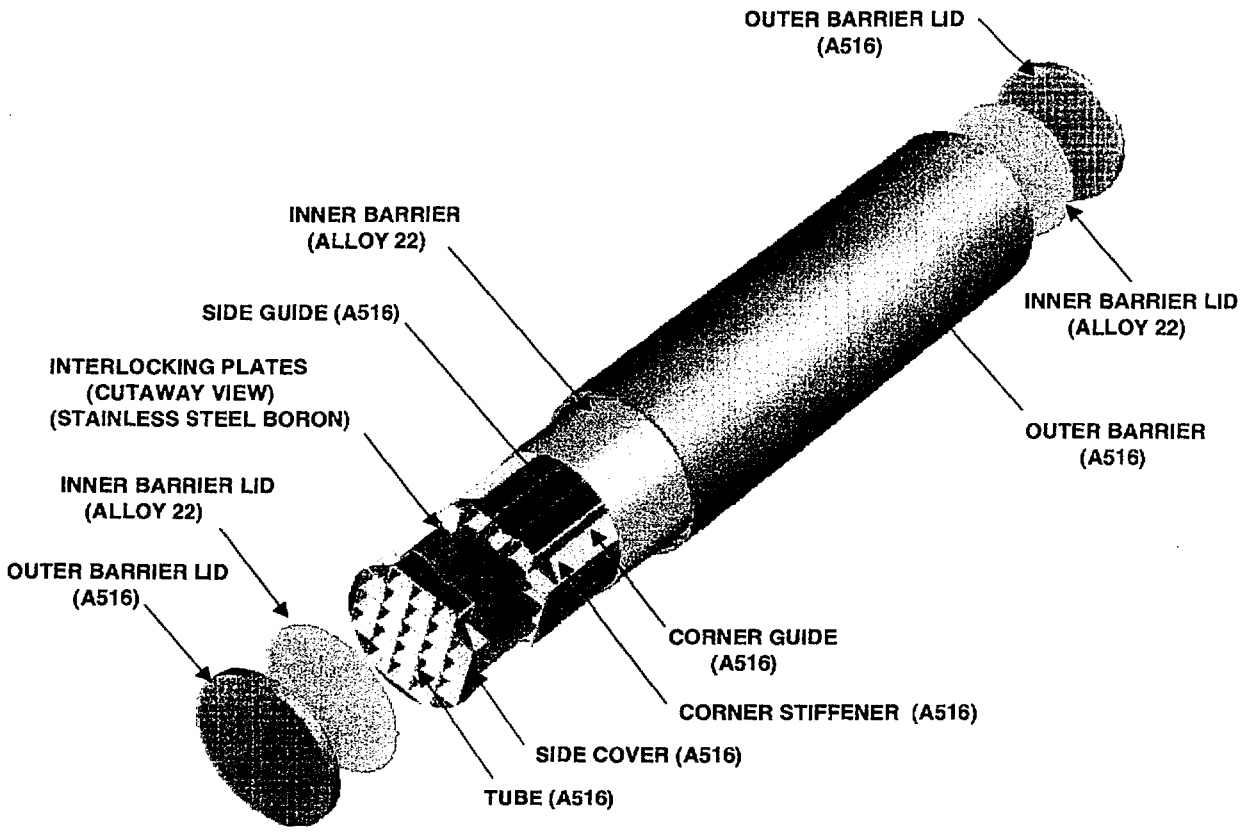

Figure 2.2-1. 21-PWR UCF Waste Package Assembly

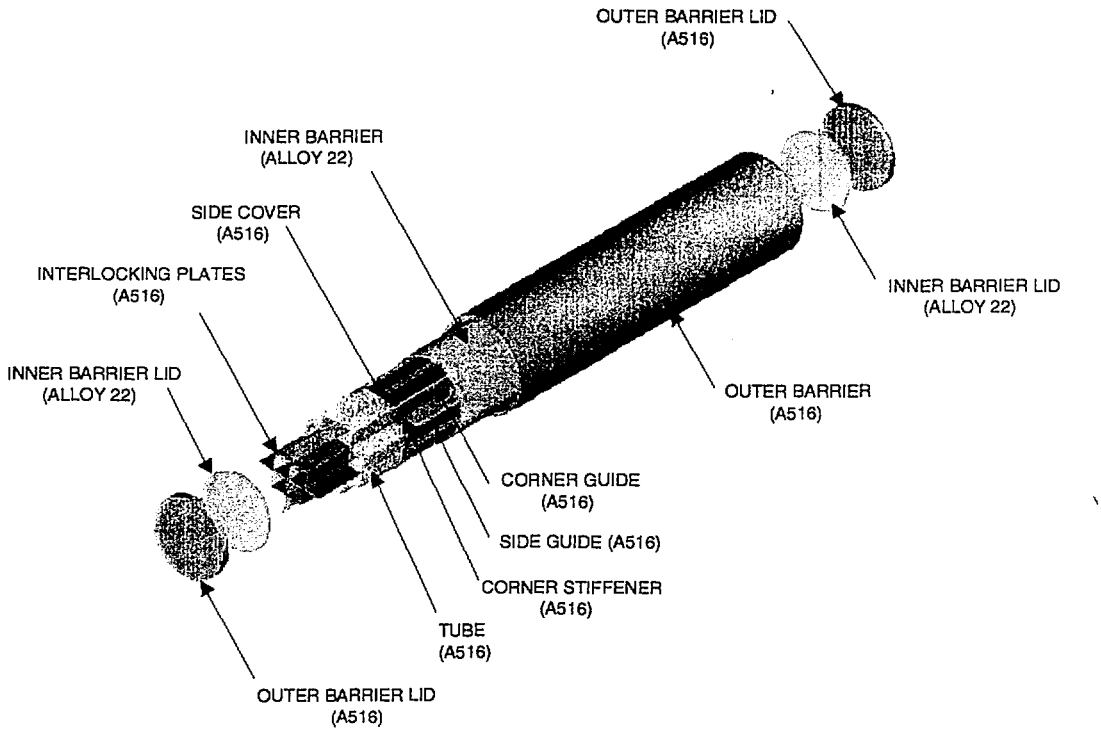

Figure 2.2-2. 12-PWR UCF Waste Package Assembly

All of the analyses were based on these 21 PWR and 12 PWR WP designs but tailored to the particular analysis as appropriate.

The intact waste package geometry parameters used in this analysis are listed in Reference 11, Section 5. The general waste package assembly information was obtained from References 12 and 13. Since the analysis covers both intact and degraded waste forms, modeling the chemical behavior of these systems is necessary which requires the chemical compositions of the waste package materials, their masses, surface areas, and corrosion or degradation rates as input. Corrosion product volume information for the 21 PWR WP was calculated for the geochemistry analysis (Ref. 14) and is summarized in Section 2.2.1 with the material property data. Corrosion product volumes were calculated for the 12 PWR WP assuming only carbon steel in the basket structure with a mass of 4449.7 kg. Calculating the corrosion product volume for the 12 PWR WP from carbon steel only is conservative since aluminum corrosion products (from thermal shunts) will displace a larger moderator volume than the carbon steel products.

2.2.1 Material Properties

Material properties of the 21 PWR MOX SNF WP required for modeling the geochemical behavior of the waste package for the criticality analyses include the masses, surface areas, and average corrosion or degradation rates. Only the average values are listed since variation in the corrosion rate had very minimal effect on the amount of iron or aluminum retained in the WP as solids (Ref. 14, p. 32). These properties are listed in Table 2.2.1-1 (Ref. 14, p. 25). An exception is made, however, for the material of the inner corrosion resistant barrier, which is assumed to react so slowly with the infiltrating water as to have negligible effect on the chemistry (Ref. 14, Assumption 3.7).

Table 2.2.1-1. Material Properties of 21 PWR WP Components

Component Material	Mass (kg)	Surface Area (m^2)	Corrosion Rate (moles/cm^2/sec)
A516 Gr 55Carbon Steel	5443.2	229	1.573e-11
Borated Stainless Steel (SS316B6A)	1882.0	71	1.169e-13
Aluminum (6061 T4)	146.5	43	1.263e-11
SNF	11,054.0	43774	4.419e-14

2.3 Waste Stream Quantities

Approximately 200 metric tons of fissile material (highly enriched uranium (HEU) and plutonium) has been declared surplus and withdrawn from the U. S. nuclear stockpile. The disposition of surplus HEU was addressed in a DOE 1996 Record of Decision (Ref. 15). In a 1997 Record of Decision (Ref. 16), the strategy adopted by the DOE for disposition of surplus weapons grade plutonium consists partly of direct geologic disposal of Pu immobilized in a ceramic matrix and partly of using the Pu as mixed oxide fuel (MOX) in one or more commercial reactors with disposal of the spent nuclear fuel (SNF) according to the Nuclear Waste Policy Act.

There is about 50 metric tons of plutonium in the surplus fissile material. Approximately 18 metric tons of this material contains significant quantities of impurities and is considered unsuitable for reactor fuel as MOX. This material has been designated for direct disposal by immobilization in a ceramic waste form. The remaining 32 metric tons of plutonium are suitable for incorporation into MOX assemblies for commercial reactors. The composition of possible MOX SNF assemblies at discharge from a reactor will be substantially different from standard commercial fuel, and, so, must be analyzed to identify potential impacts on the waste package designs and to provide guidance for potential MOX SNF disposal recommendations.

Approximately 1732 MOX assemblies will be required to consume the 32 metric tons of Pu. This translates into 19 core reloads of 92 assemblies per reload. The standard LEU 21 PWR and 12 PWR waste package design are proposed for disposal of the MOX SNF. The 21 PWR WP design will be used for assemblies with lower burnup values (and consequently more fissile Pu content) and the 12 PWR WP design will be used for assemblies with high burnup values and corresponding high thermal heating rates. The number of highly burned assemblies can be estimated from the discharge

burnup distribution (Ref. 3, p. 2-39). Use of both waste package designs (the 21 PWR and 12 PWR WPs) is necessary to meet the maximum thermal output criteria of 18 kW per waste package (Ref. 22, Vol. 1, p. 19). This analysis shows that a MOX SNF assembly burnup of approximately 46.5 GWd/MTHM will meet the 18 kW thermal output limit for the 21 PWR WP (850 watts per assembly). For the 12 PWR WP, the thermal output criteria limits the maximum output to 1500 watts/assembly, well above the highest output value of 1070 watts/assembly derived from this analysis. This waste package loading criteria results in 43 of the 21 MOX PWR WPs and 72 of the 12 MOX PWR WPs required for the 1732 MOX assemblies.

2.4 Waste Package Criticality Control Measures

The criticality control requirement for emplacement and isolation of radioactive waste is that the system k_{eff} maintains a minimum 5% margin below unity after allowing for biases and uncertainties (Ref. 42, Section 2.1.1). To assure such conditions for long term emplacement of MOX SNF, reactivity control measures are necessary the same as for LEU SNF. Reactivity control in the waste packages while the system is intact is provided by borated stainless steel absorber plates in the assembly basket structure as shown in Figure 2.2-1. Insoluble corrosion products from the A516 carbon steel basket structure (notably hematite [Fe_2O_3]) may provide long term criticality control for breached but structurally intact waste packages because of moderator displacement. This study shows that only the 21 MOX SNF WPs with the larger fissile Pu content will require reactivity control. The 12 MOX PWR WPs remains subcritical under all degradation scenarios because of the smaller initial fissile Pu inventory and subsequent moderator displacement by waste package corrosion products. Thus, no supplemental absorber plates are necessary for reactivity control in this waste package.

3. STRUCTURAL ANALYSIS

The structural design criteria for the waste package is that the waste form be capable of withstanding a two meter drop onto a flat essentially unyielding surface without breaching (EBDRD 3.7.1.1.F, Ref. 28, p. 4-40). The tipover accident produces the highest stresses in the waste package since the upper part of the waste package experiences a drop greater than the two meter criteria. Analyses were performed for the 21 PWR MOX WP (Ref. 17) and 12 PWR MOX WP (Ref. 18) to determine the structural response to a tipover accident design basis event (DBE) dynamic load (Ref. 19, p. 44).

3.1 Structural Analysis Method

A three-dimensional finite-element solution was performed by making use of the ANSYS V5.4 finite-element computer code (CSCI: 30040 V5.4) (Ref. 20). A finite-element model of the waste package was developed to determine the effects of tipover accident DBE loads on the waste package structural components. The basket structure in the 21 PWR MOX WP was modeled with B-SS absorber plates and a combination of A516 carbon steel and aluminum in the basket structure. The aluminum serves as a heat conduit (thermal shunt) in the waste package and is not a structural material. The basket structure in the 12 PWR MOX WP was modeled in a similar manner as the 21 PWR MOX WP except that two calculations were conducted on the waste package, one with and one without B-SS absorber plates. The waste package was modeled with an initial orientation of 30° between the symmetry axis and vertical in order to initiate tipping of the waste package, and gravitational acceleration was then applied to the system. Having the waste package modeled in this configuration, the simulation was continued throughout the impact until the waste package began to rebound, at which time the peak stresses have been obtained. It should be noted that in order to determine the center of gravity offset from vertical, the angle between the axis of symmetry and the waste package diagonal was subtracted from 30 degrees; this offset angle was calculated as 16 degrees for the 12 PWR MOX WP. The percent change in the height of the center of gravity is the complement of the cosine of this angle, which is calculated as 0.039, or 3.9%. Additional calculations (Ref. 18, p. 10) show that the change in the stresses obtained from the finite element solutions is directly proportional to the square root of the change in the height of the center of gravity. Thus, the difference in the initial waste package center of gravity height has been accounted for by using a factor of multiplication of approximately 1.02 (increase of 2%).

PWR SNF assemblies differ in total weight due to the variations in designs. However, the PWR MOX SNF assemblies have the same maximum weight (685.9 kg, allowing for variations about the average) as commercial LEU SNF assemblies of similar design (Ref. 6, p. 6). Weight changes due to burnup are negligible (less than 25 g at the maximum burnup). The structural analyses show that stresses from the tipover accident for both SNF waste forms are of similar magnitude.

3.2 Structural Analysis Results

The structural response of the waste package to tipover accident loads is given as maximum stress values obtained from the finite-element solutions to the problem. These solutions indicate that the maximum stress is located in the region of the inner and outer barrier lids in the vicinity of the impact region between the waste package and the target surface for both waste package designs.

Linearized stress paths were defined in all of the waste package structural components passing through the nodes with the maximum stress intensity in order to determine the maximum membrane and membrane plus bending stresses. The results of the waste package tipover accident structural analysis are provided in Table 3.2-1 for the 21 PWR MOX WP. Maximum stresses from a similar analysis for the 21 PWR WP containing LEU SNF are also included in Table 3.2-1 (non-Westinghouse assembly, 773.4 kg assemblies) (Ref. 21, p. 11). Stress results from both cases (with and without absorber plates) calculated for the 12 PWR MOX WP are given in Table 3.2-2. As shown, maximum stress levels for both waste package designs are at least 15% below the respective ultimate tensile strength values and thus are within design limits (Ref. 22, Vol. 1, p. 13).

Table 3.2-1 shows that for some waste package components, linearized stresses are higher for the 21 PWR MOX WP than the stresses obtained from the 21 PWR WP containing LEU SNF. Considering the fact that structural analyses were performed using a finite element code and the solution is entirely transient dynamic with stress propagation and nonlinear material properties, the stress peaks of the two models do not necessarily occur at the same time step. Therefore, depending on the element and time discretization, a certain fluctuation in stress magnitudes is anticipated. However, the general trend of the results obtained for both MOX and commercial SNF waste package weights show that all stresses are below the ultimate tensile strength of the waste package component materials.

Table 3.2-1. Finite-Element Structural Analysis Results for the 21 PWR WP

WP Component	Ultimate Tensile Strength (MPa)	Maximum Membrane Stress (Mpa)		Maximum Membrane Plus Bending Stress (Mpa)	
		MOX SNF	LEU SNF	MOX SNF	LEU SNF
Outer Barrier and Outer Barrier Lid	483	375	340	390	349
Inner Barrier and Inner Barrier Lid	690	431	407	524	456
Guides	483	401	242	413	340
Tubes	483	283	289	302	294
Criticality Control Plates	550	289	301	319	305

Table 3.2-2. Finite-Element Structural Analysis Results for the 12 PWR MOX WP.

WP Component	Ultimate Tensile Strength (Mpa)	Maximum Membrane Stress (Mpa)		Maximum Membrane Plus Bending Stress (Mpa)	
		With Absorber Plates	Without Absorber Plates	With Absorber Plates	Without Absorber Plates
Outer Barrier and Outer Barrier Lid	483	265	258	360	378
Inner Barrier and Inner Barrier Lid	690	399	418	404	425
Guides	483	220	148	300	285
Tubes	483	326	326	333	347
Carbon Steel Plates	483	N/A[1]	315	N/A	321
Corner Stiffeners	483	307	307	314	317

[1] Not Applicable. Absorber plates replace carbon steel plates. No structural credit taken for absorber plates.

4. THERMAL ANALYSIS

Thermal analyses (Ref. 23) were performed under normal repository disposal conditions on the 21 PWR WP and the 12 PWR WP loaded with MOX SNF using the heating rates from Reference 44 (summarized in Table 2.1-4) to demonstrate that these waste packages can accommodate the entire MOX waste stream. The 46.5 GWd/MTHM heating rates were used in the 21 PWR MOX WP and the 56.5 GWd/MTHM in the 12 PWR MOX WP. In both cases, the SNF assemblies were assumed to have had a 10 year cooling period prior to emplacement in the waste package (CDA Key 004 [Ref. 28] specifies that the initial SNF for the repository be at least 10 years old).

4.1 Thermal Analysis Method

A two-dimensional, time dependent finite-element calculation was performed by making use of the ANSYS V5.1 finite element computer code (Ref. 24). A two-dimensional (2-D) finite-element model was developed (Ref. 23, Section 5.4) for a midpoint cross section of the waste package. This represents the hottest portion of the waste package because of the non-uniform axial heat source distribution. Aluminum thermal shunts were included in the model for both the 21 PWR and 12 PWR WP designs to enhance the heat flow rate.

The SNF assembly, which produces a heat load in the waste package, was modeled as a lumped parameter solid material placed inside each tube in the basket assembly. The time-dependent volumetric heat loads were multiplied by an axial peaking factor of 1.25 (Ref. 25, p. 29) to approximate modeling the axial center of the waste package with a 2-D model. The peaking factor conservatively compensates for the lack of a detailed axially non-uniform assembly power shape. The initial heating rates for the MOX SNF were 798 watts/assembly for the 21 PWR WP and 1070 watts/assembly for the 12 PWR WP. The burnup levels for these assemblies were 46.5 GWd/MTHM and 56.5 GWd/MTHM, which are the hottest assemblies planned for these WP's, respectively. These values compared to 850 watts/assembly and 1500 watts/assembly for commercial PWR thermal design basis fuel assemblies (Ref. 25, p. 67) in the respective waste packages. The burnup level for the design basis commercial PWR assembly was 60 GWd/MTU. The initial heating rate values for the commercial PWR SNF correspond to different cooling periods prior to inclusion in the repository waste stream.

Temperature boundary conditions at the exterior surfaces of the 21 and 12 PWR MOX WPs for the 2-D thermal calculations were derived from the time-dependent temperature boundary conditions resulting from the three-dimensional (3-D) multiple waste package calculation (Ref. 26). The waste package boundary surface temperatures were determined at thermal design basis loading of 85 MTU/acre which gives a constant center-to-center spacing for the 21 PWR WP with absorber plates of 15.4 m and 9.2 m for the 12 PWR WP with no absorber plates (Ref. 27, p. 17). This areal mass loading (AML) is within the AML range (80 to 100 MTU/acre) given in the CDA (Ref. 28, Key 019) as the reference mass loading range. Thus the MOX SNF will pose no additional constraints on the waste package layout. The source for the thermal calculation was derived from the time-dependent radioactive decay heat sources (Ref. 45) documented in Reference 6, Section 6. The 2-D thermal analyses of the waste packages were carried out for a time period of 1000 years following a cooling period of 10 years after discharge from the reactor.

4.2 Thermal Analysis Results

The temperature history containing the peak value in the fuel for 21 PWR MOX SNF WP is shown in Figure 4.2-1 and for the 12 PWR MOX SNF WP in Figure 4.2-2. The location of the peak node was at the center of the innermost assembly in both cases (Note that the fuel assemblies were modeled as a homogenized solid material). The peak values were 336°C for the 21 PWR SNF WP and 302°C for the 12 PWR WP. The outer surface boundary condition temperatures for the respective cases are also shown in the figures. The peak values for the waste package surface temperatures were 234°C for the 21 PWR SNF WP and 218°C for the 12 PWR WP. The time of occurrence of the peak waste package surface temperature was about 20 years after emplacement for both histories.

The fuel temperature (homogenized assembly material) peaks at approximately 336°C about 7 years after emplacement for the 21 PWR MOX SNF WP and at approximately 302°C about 2 years after emplacement for the 12 PWR MOX WP. Both these peak temperatures are well below the maximum permissible waste package temperature of 350°C given in the CDA (Ref. 28, DCWP 001, p. 8-1).

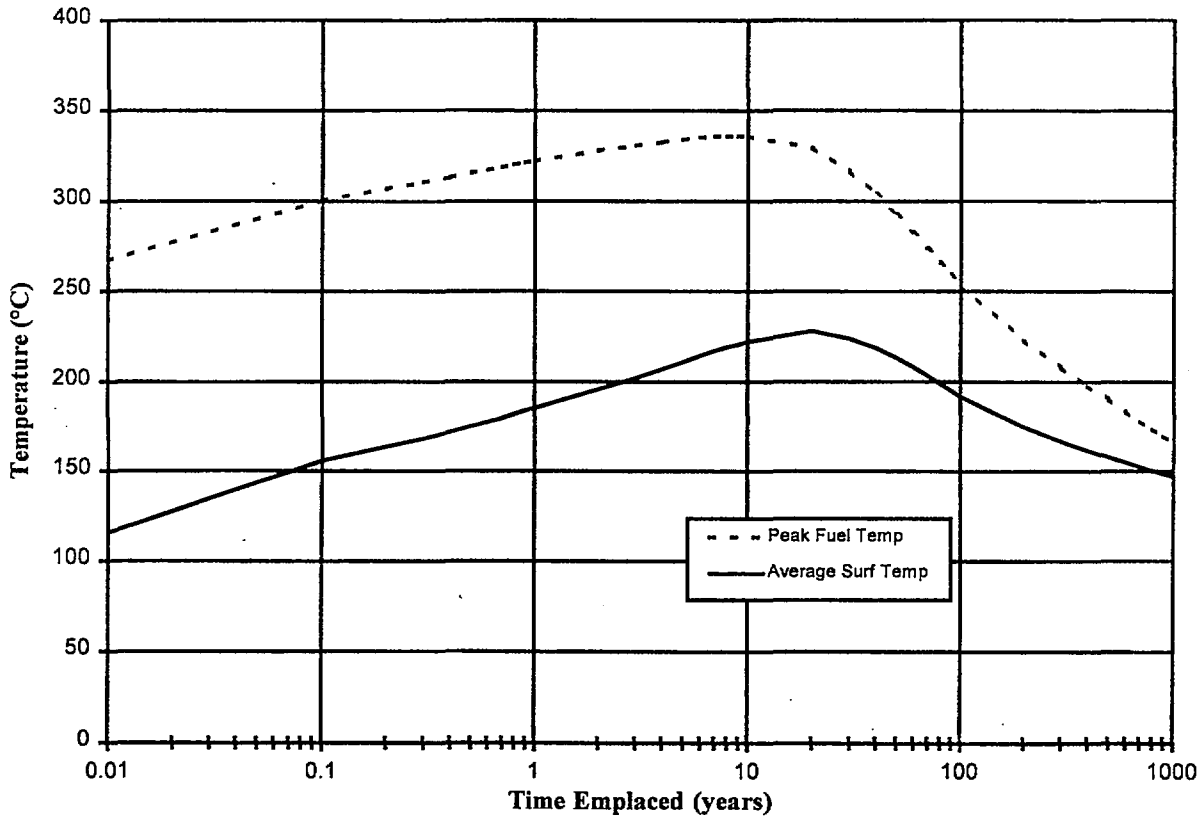

Figure 4.2-1. Temperature Histories for 21 PWR MOX WP

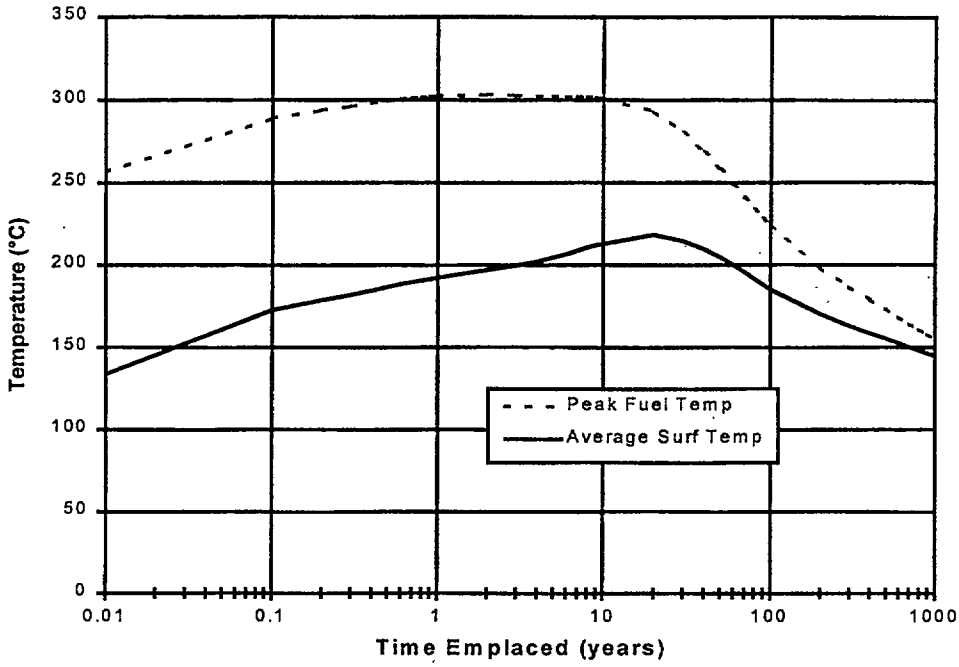

Figure 4.2-2. Temperature Histories for 12 PWR MOX SNF WP

5. SHIELDING ANALYSIS

Shielding analyses were performed for the 21 PWR MOX WPs (Ref. 29) using the MOX SNF assembly producing the highest gamma-heating source following discharge from a hypothetical equilibrium PWR MOX reactor (Ref. 5). These analyses required a two step calculation procedure given by:

1) generation of the appropriate radiation source terms for SNF assemblies (primarily gamma intensity) as a function of time using the SAS2H/ORIGEN-S code sequence from SCALE 4.3 (Ref. 7), and

2) use of the calculated source terms as partial input to the MCNP4B2 code (CSCI: 30033 V4B2LV) (Ref. 33) to calculate time-dependent dose rates in rem/h on various surfaces and external near-field locations around the waste package.

Shielding requirements for AUCF waste packages as given in Key Assumption 031 from the Controlled Design Assumptions Document (CDA) (Ref. 28) states that waste package containment barriers will provide sufficient shielding for protection of waste package materials from radiation enhanced corrosion. Experiments on radiolytic corrosion reported in Reference 30, (Vol. III, p. 8-4) indicate that for iron based materials in an air/steam environment, a 100 rad/h dose rate at 250°C increased the corrosion rate by a factor of 5 but no change in rates were observed at 150°C. Dose rates from the shielding analysis are given in rem/h. The factor for converting dose rates in rad/h to rem/h for gamma radiation is unity and approximately 10 for neutrons. Dose rates in rad/h will always be less than or equal to the dose rate in rem/h.

Methods and results for the shielding calculation are discussed in detail to aid in the interpretation of the time history surface dose rate and to identify the differences between the MOX SNF dose rates and the LEU SNF dose rates.

5.1 Shielding Analysis Method

The source terms for the shielding configuration included activation of assembly hardware and were determined by taking the documented amount of hardware for the assembly (Ref. 31, p.2A-349 through 2A-354) and exposing it to the entire active fuel region via the light element option in SAS2H. To do this, the composition and mass of the hardware in each region of the assembly are determined from Reference 31 above. The masses of the hardware components were estimated from descriptions of the particular assembly, i.e., the bottom end fitting, fuel, plenum, or top end fitting region. The light element masses for these components were then adjusted by a scaling factor (Ref. 33, p. 9) to account for the location of the assembly hardware in various axial positions in the reactor.

The PWR MOX WP source terms are generated from the data files (Ref. 44) developed during the analyses documented in Reference 6, Section 6. The case chosen for the shielding analysis of the 21 PWR MOX WP from the set of analyses reported in this latter reference was the 56.5 GWd/MTHM burnup case with 4.0 wt% initial fissile Pu in HM. Results of the shielding analysis will be conservative since this case produced the largest gamma heating source from the MOX analysis.

Shielding calculations have also been carried out for commercial SNF in the 21 PWR WP using a B&W Mark B assembly having an initial enrichment of 5.05 wt% fissile uranium and a burnup of 75 GWd/MTU (Ref. 32, p. 4). This burnup level was used since it is the worst case situation for which shielding must be designed. The commercial SNF calculation provides a frame of reference for the MOX SNF shielding results.

The SAS2H/ORIGEN-S code is used to simulate the irradiation of the fuel and the light elements and to decay the radiation source. Time dependent gamma and neutron sources are generated for each time step requested in ORIGEN-S. To use this information as a source in MCNP4B2, the spectrum and group structure for the sources are entered and normalized by the code. The source strength is then entered in the form of a tally multiplier. This multiplier is calculated by multiplying the total source determined in SAS2H/ORIGEN-S analysis by the number of assemblies in the package and by an axial peaking factor of 1.25. This factor is based on the axial gamma radiation profile from Reference 40, Figure 3-18. The peaking factor conservatively compensates for the lack of a detailed axially non-uniform assembly source profile.

The major isotopes contributing to the sources for the shielding calculations are given in Table 5.2-1 for the MOX PWR SNF and the commercial PWR SNF. The isotopic inventory in both cases was calculated for a 10 year cooling period following discharge from a reactor (CDA Key 004 [Ref. 28] specifies that the initial SNF for the repository be at least 10 years old). Contributions from the fuel region included actinides, fission products, and the light elements. Contributions from the lower end fittings, representative of the non-fueled regions, included only the light elements, Table 5.2-2. The following observations can be made concerning these radiation sources:

1. The MOX SNF actinide curie source was considerably higher than for the commercial SNF but the actinides decay mainly by alpha emission contributing little to the external dose rate.
2. The fission product isotopic distribution from the MOX SNF results in lower relative source contributions from Sr-90 and Y-90 than from the commercial SNF. This is due partly to the higher burnup in the LEU SNF and due partly to the different fission product inventory (curies) as shown in Table 5.2-1.
3. The major contributors to the radiation source in the non-fuel regions are ^{60}Co and ^{125}Sb, with the commercial PWR SNF source much larger than the MOX SNF source (Table 5.2-2). The differences result from the end fitting composition differences, notably the greater amount of Inconel-718 in the LEU assembly hardware than in the MOX assembly hardware (Ref. 30, p. 10, Ref. 33, p. 8.

The variation in radiation sources between the MOX and LEU PWR assemblies results in higher calculated dose rates from the LEU assemblies compared to the MOX assemblies.

Table 5.2-1. Major Isotopes Contributing to the Fuel Region Radiation Source

Actinides			Fission Products			Light Elements		
Isotope	PWR (curies)	MOX (curies)	Isotope	PWR (curies)	MOX (curies)	Isotope	PWR (curies)	MOX (curies)
Pu238	6080	2720	Kr 85	4080	1810	Fe 55	56.4	28
Pu239	191	267	Sr 90	55800	20600	Co 60	2670	
Pu240	400	757	Y 90	55800	20700	Ni 63	288	
Pu241	63700	124000	Cs134	6930	4520	Nb 93m	28.4	
Am241	1460	2950	Cs137	87900	60900	Sb125	67.4	146
Cm244	9230	8320	Ba137m	83000	57500	Te125m	16.5	36
			Pm147	6240	5700			
			Eu154	3940	3800			
Total	81300	140000	Total	307000	179000	Total	3140	215

Table 5.2-2. Major Isotopes Contributing to the End Fittings Region Radiation Source

Isotope	PWR	MOX
Fe 55	88	43
Co 60	223	109
Ni 63	56	20
Sb125	179	
Te125m	44	
Total	598	177

Fuel assemblies and their hardware compositions are homogenized over the inside dimension of the waste package in the geometric model for the MCNP calculation with no shielding credit taken for the waste package basket and basket guide materials. This is a conservative approach for dose rate calculations since: (1) the internal basket structure would attenuate the neutron and gamma ray flux, and (2), homogenizing the assemblies inside the waste package in effect moves the source closer to the outer surface of the waste package, thereby allowing more particles to reach the outer surface. The corrosion allowance barrier in the waste package was assumed to begin degradation when the repository humidity reaches 75% at approximately 700 years after emplacement, thus gradually reducing the original quantity of shielding material.

A different calculation was made for each of four gamma sources representative of four axial regions in the waste package (bottom end, fuel, upper plenum, and top end) and one fuel region neutron source to isolate the contribution from each. These contributions are then summed to yield a total dose.

5.2 Shielding Analysis Results

Radiation dose rates were calculated at a number of locations both interior and exterior to the 21 PWR MOX. Dose rates were calculated in the radial direction on the inside and outside surfaces of the corrosion allowance and resistant shells, shown in Figure 5.2-1, and at the one and two meter distances from the outside surface of the waste package. Axially, the dose rate was determined on the exterior surface and 2 meters from the waste package. The maximum source strength is in the mid-region of the waste package and maximum dose rates occurred in the radial direction normal to the waste package central axis. The total dose rates, together with the neutron and gamma components, in the radial direction at 10 years following reactor discharge are given in Table 5.2-3 for the surfaces defined in Figure 5.2-1. The dose rates at these surfaces are calculated from contributions from all source regions. The dose rate in rem/h at several radial positions in the waste package mid-region is shown in Figure 5.2-2 as a function of time. At locations interior to the waste package, the dose rate declines monotonically with time as the source decays. Exterior to the waste package, however, the dose rate rises slightly in the period between approximately 700 and 12,000 years but is always lower than at the initial time. This is primarily due to the loss of shielding

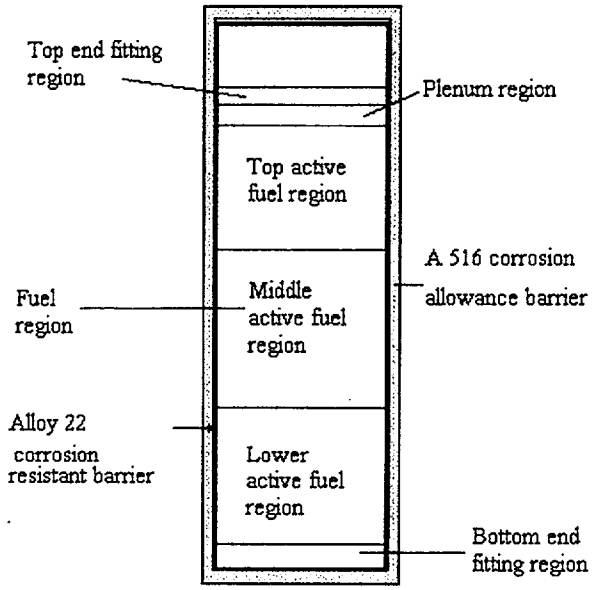

Figure 5.2-1. Waste Package Surfaces for Radial Dose Rate Calculation.

material as the corrosion allowance waste package material (A516 carbon steel) begins to degrade. Axial profiles of the radial dose rates at the waste package at the exterior surface of the waste package are shown in Figure 5.2-3 for various times following emplacement. The gamma dose from the end regions (see Table 5.2-3) results, in part, from ^{60}Co in the Inconel components as shown in Table 5.2-2. The spectrum of the gamma radiation from the fuel and end regions differs, resulting in the modest peaks near the assembly ends. These peaks are short-lived as shown in Figure 5.2-3.

Dose rates on the surface of the 21 PWR WP for commercial PWR SNF at 10 years following reactor discharge are shown in Table 5.2-4 (Ref. 32, Section 6.2). The commercial SNF used in the shielding calculation was a B&W Mark B assembly having an initial enrichment of 5.05 wt% fissile uranium and a burnup of 75 GWd/MTU. This burnup level was used since it is the worst case situation for which shielding must be designed. As shown, the dose rates at the waste package surface from Table 5.2-4 are considerably higher than for the MOX SNF WP consistent with the differences in the source values.

Table 5.2-3. 10 Year Dose Rates from 21 MOX PWR SNF at External Surface of Waste Package

Region	Total (rem/h)	Neutron (rem/h)	Gamma (rem/h)
Top end fitting region	8.23E+01	3.58E+00	7.87E+01
Plenum region	1.12E+02	5.08E+00	1.07E+02
Top active fuel region	8.92E+01	1.13E+01	7.79E+01
Middle active fuel region	9.01E+01	1.33E+01	7.67E+01
Lower active fuel region	9.05E+01	1.16E+01	7.89E+01
Bottom end fitting region	9.53E+01	5.50E+00	8.98E+01

Table 5.2-4. 10 Year Dose Rates at the External Surface from Commercial LEU SNF in the 21 PWR Waste Package

Region	Total (rem/h)	Neutron (rem/h)	Gamma (rem/h)
Top end fitting region	1.96E+02	3.63E+00	1.92E+02
Plenum region	2.93E+02	5.30E+00	2.88E+02
Top active fuel region	1.66E+02	1.25E+01	1.53E+02
Middle active fuel region	1.64E+02	1.49E+01	1.50E+02
Lower active fuel region	1.65E+02	1.29E+01	1.52E+02
Bottom end fitting region	1.76E+02	6.05E+00	1.70E+02

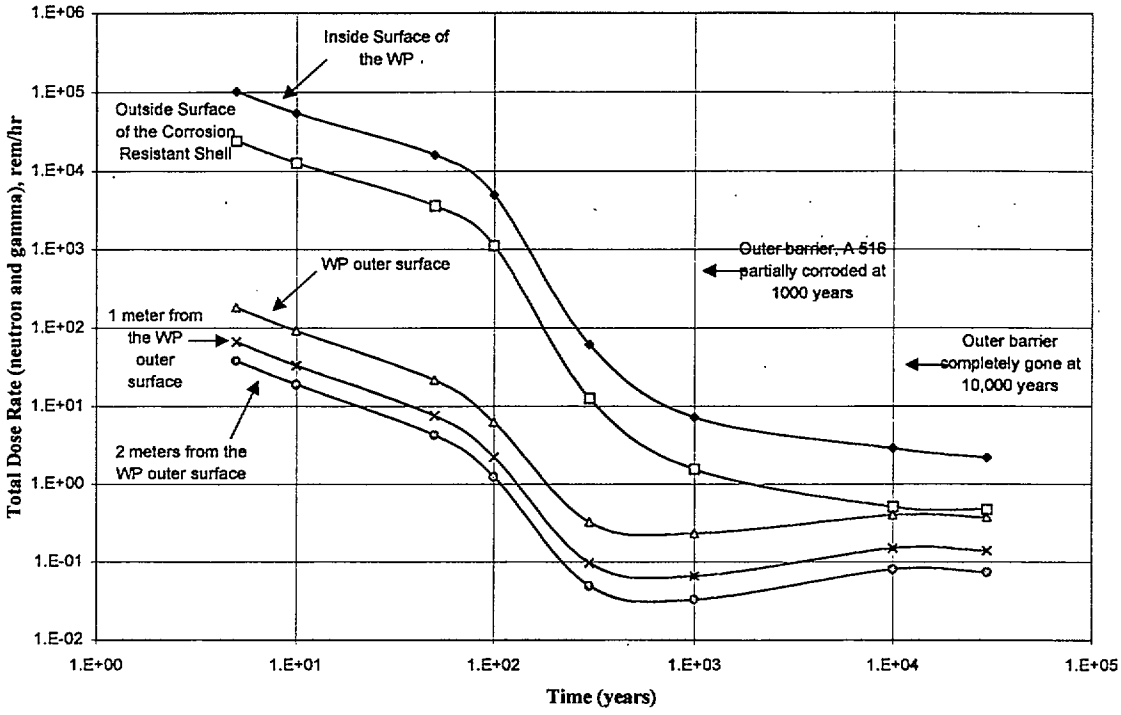

Figure 5.2-2. Radiation Dose Rate Over Time from the 21 MOX PWR WP Mid-Region

For the 21 MOX SNF WP, the dose rates shown in Figure 5.2-2 exceeded 10 rem/h only during the period prior to 100 years when the humidity of the external environment is assumed to be low. High humidity levels were assumed to occur only after approximately 700 years when the waste package surface temperatures are calculated to not exceed approximately 150°C. Thus, it is concluded that there will not be any increase in the waste package barrier corrosion rate due to radiolysis.

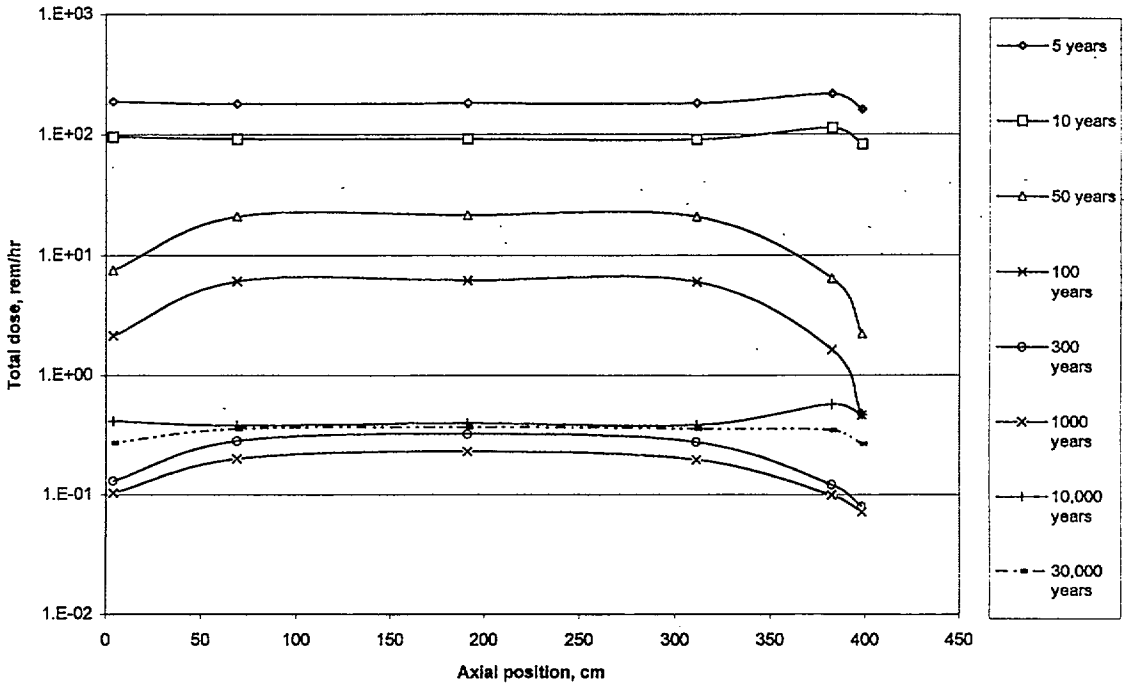

Figure 5.2-3. Axial Profile of Dose Rates at the Outside Surface of the 21 MOX PWR WP

6. DISPOSAL CRITICALITY ANALYSIS

A series of criticality calculations were performed (Ref. 11) for both intact and degraded configurations of the MOX PWR assemblies described in Section 2 to evaluate the criticality potential of the MOX SNF WP configurations. Five wt% fissile Pu in HM/burnup combinations were selected for analysis as given in Table 2.1-3. The cases which will be most important with respect to criticality are those having the lowest burnup for each initial wt% fissile Pu in HM. For the 21 MOX PWR WP, these would be cases 4 and 5 from Table 2.1-3. The calculations for the 12 MOX PWR WP considered a third wt% fissile Pu in HM/burnup pair (case 2 from Table 2.1-3) as this case is more representative of the higher burned fuel for which this package is intended. Isotopic compositions for each fuel in grams per assembly, for decay times from a few days, out to 1 million years are given in Reference 11, Section 6. The compositions from Reference 12 tracked for the criticality calculations were limited to the PI burnup credit list as discussed in Section 2.1.

The criticality analysis methodology for the SNF and waste package basket structure is discussed in Section 6.1 for both intact and degraded configurations. The methodology for the degraded configurations includes a brief discussion of the degradation sequences and composition definition for criticality calculations. The criticality evaluation of the intact SNF and basket structure is given first in Section 6.2. The degraded configurations and results of the criticality analyses of these configurations are discussed in Section 6.3. Results of the criticality analyses are summarized in Section 6.4.

6.1 Disposal Criticality Analysis Methodology for both Intact and Degraded SNF

In this study the methodology for computing k_{eff} values (Ref. 11) for intact and various degraded waste package configurations uses the Monte Carlo N-Particle Code MCNP, Version 4B2 (CSCI: 30033 V4B2LV) (Ref. 33). Fuel region number densities used in this criticality evaluation were calculated simply by homogenizing the isotopic concentrations from Reference 5 for a particular fuel and decay time throughout the volume of the active fuel region.

The degraded configurations to be considered are based on those evaluated for the commercial PWR waste package (Ref. 43, Section 7.1). Figure 6.1-1 shows a schematic view of the degradation sequence for the 21 PWR absorber plate waste package following breach of the package (Ref. 43, Section 7.1.1 provides a description of the degradation process and corrosion product generation). Since the waste package interior was inerted with He prior to time of breach, the initial configuration will be the as-built basket (Fig. 6.1-1A). Within a few hundred years following breach, the carbon steel and aluminum components will degrade to insoluble corrosion products as shown in Figure 6.1-1B (Ref. 14, Section 5.3). While structural calculations show that the absorber plates can support the load of the assemblies (Ref. 43, p. 27), localized corrosion in the crevice regions at the corners of each cell will likely cause collapse shortly after failure of the structural components. However, the majority of the B-SS absorber plates will be only minimally degraded and remain between the assemblies, with corrosion products from the degraded carbon steel tubes (Fig. 6.1-1C). Eventually, after thousands of years, general corrosion will also fully degrade the absorber plates, allowing the soluble boron neutron absorber to be flushed out of the package (Fig. 6.1-1D). The zircaloy cladding and spacers represent the most corrosion resistant material in the waste package, and thus will be the last to degrade. Collapse of the fuel rods at the bottom of the waste package will likely

occur prior to complete cladding degradation (Fig. 6.1-1E), as the spacer grids are typically fabricated from strips of zircaloy that are thinner than the cladding. The final internal configuration (Fig 6.1-1F) is complete degradation of the entire waste package contents, with only the insoluble materials remaining. Similar configurations would also be expected to form during degradation of the 12 PWR WP, with the exception of configurations B and C, which cannot occur because the 12 PWR WP does not contain B-SS absorber plates.

This study summarizes calculations that considered configurations A, D, E, and F for both the 21 and 12 PWR WP. Chemical compositions of the remaining basket and fuel corrosion products were obtained from the geochemistry calculations reported in Reference 14, Section 5.3. As in Reference 11, Section 5.4.3, both settled and uniform corrosion product distributions will be evaluated for configuration D.

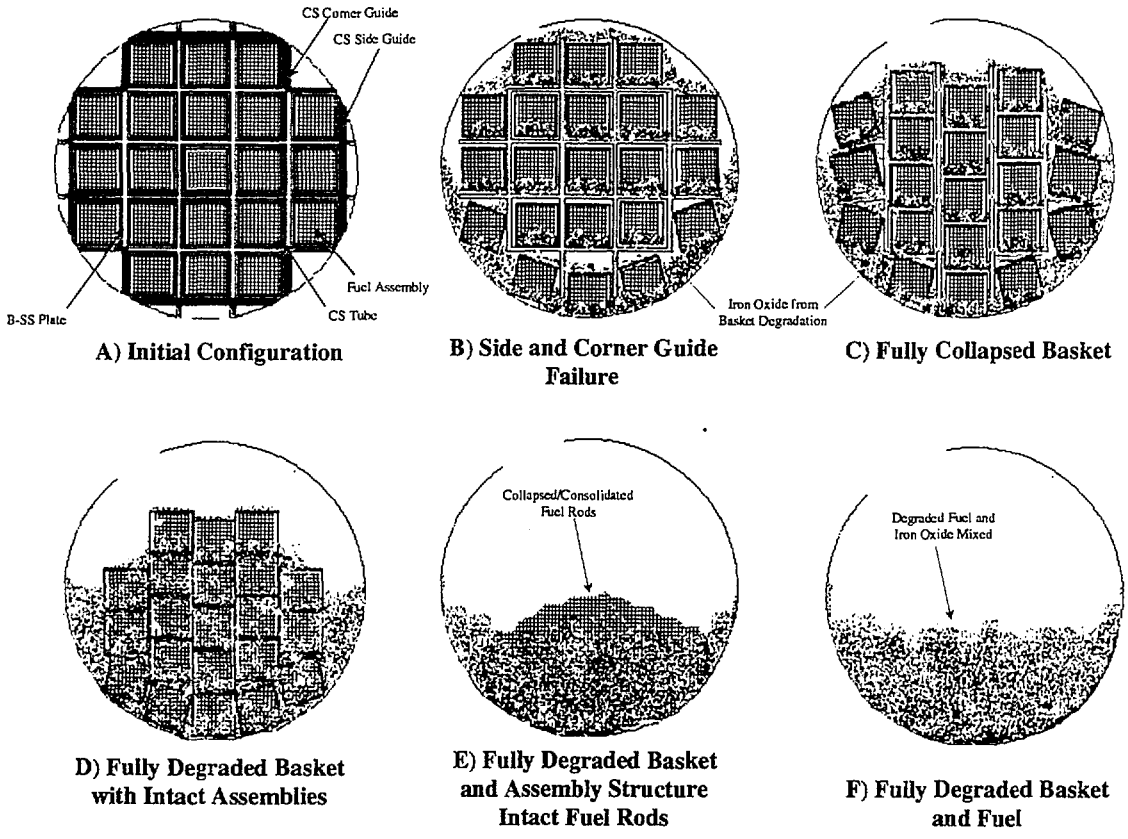

Figure 6.1-1. Degradation Sequence of the 21 PWR Basket Structure Following WP Breach

6.2 Criticality Evaluation of the Intact Configuration

In keeping with the methodology described in Section 6.1, the criticality potential (k_{eff}) of the intact MOX SNF WP configuration (Figure 6.1-1A) was evaluated for both the 21 and 12 PWR WP designs. The criticality control requirement for emplacement and isolation of radioactive waste is that the system k_{eff} maintains a minimum 5% margin below unity after allowing for biases and uncertainties (Ref. 42, Section 2.1.1). Benchmark calculations (Ref. 34, Section 3.1.4) with the MCNP4B2 code showed a maximum difference (or bias) of 3% between the calculated and experimental k_{eff}'s. Statistical uncertainties at the 2σ level in the k_{eff} calculations are normally of the order of 0.2% to 0.3%. Thus the maximum k_{eff} to assure subcriticality is $0.92 = 1.0 - 0.05 - 0.03$. This section describes the MCNP4B2 cases needed to evaluate the k_{eff} of this configuration.

6.2.1 Intact Fuel and Intact Basket Criticality Model

The composition and dimensions of the containment barriers and basket components were modeled explicitly using the information in Section 2.2. Each Westinghouse 17x17 Vantage 5 fuel assembly was treated as a heterogeneous system with the fuel rods and control rod guide tubes modeled explicitly using the information contained in Section 2.1. The fuel rods are conservatively modeled with water in the gap region simulating the effect of penetrated cladding (Zircaloy-4 is highly corrosion resistant but assumed to have sufficient penetration to allow water to fill the gap). Figure 6.2.1-1 shows the details of the MCNP4B2 model for the 21 PWR WP and Figure 6.2.1-2 shows the model details for the 12 PWR WP. In both models, the waste package is filled with water and there is a water reflector on the exterior. In addition to the base design discussed in Section 2.2, an additional case was evaluated with the central basket plates changed from A516 carbon steel to aluminum Alloy 6061. This alternative was evaluated because it was being considered in the thermal analyses of the 12 PWR WP. Each of the intact 21 PWR WP designs was evaluated for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM burnup fuel (fuel #1) and the 4.5 wt% fissile Pu in HM, 39.4 GWd/MTHM burnup fuel (fuel #2), for decay times from 10 years to 250,000 years. In addition, the intact 12 PWR WP designs were evaluated for the 4.0 wt% fissile Pu in HM, 50.1 GWd/MTHM burnup fuel (fuel #3).

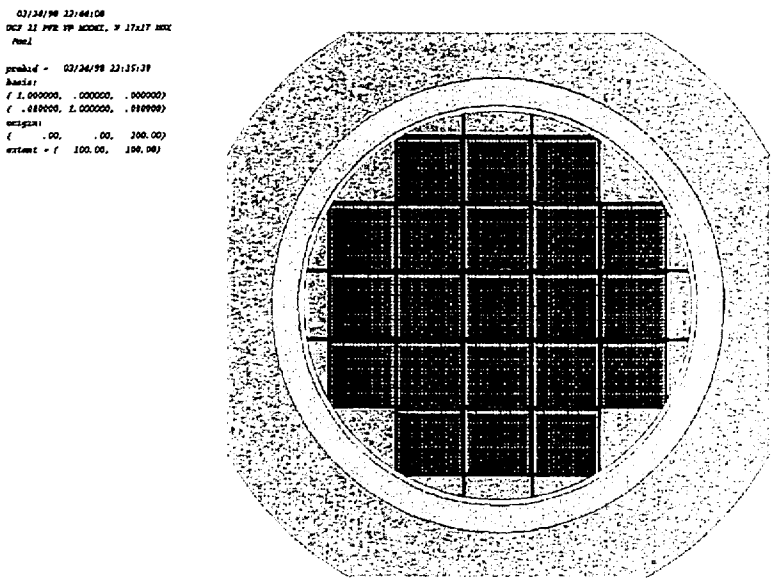

Figure 6.2.1-1. Intact 21 PWR MOX Fuel Waste Package

Principal isotopic compositions for the MOX SNF, summarized in Section 2.2, as obtained from the SAS2H/ORIGEN-S output (Ref. 44) are given in grams per assembly for decay times from a few days out to 1 million years. These were converted to number densities for the criticality calculations with MCNP4B2 using the fuel volume (Ref. 5, p. 10) and isotopic mass (Ref. 34, pp. 29-31).

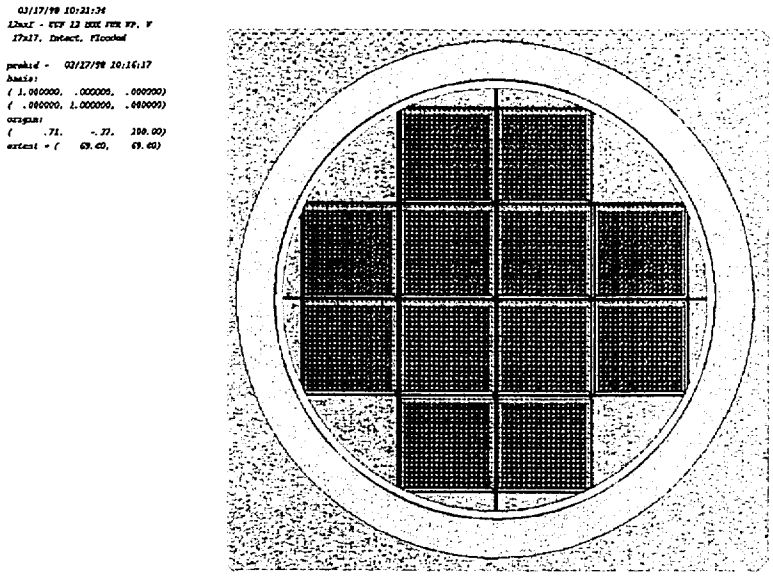

Figure 6.2.1-2. Intact 12 PWR MOX Fuel Waste Package

6.2.2 Intact Fuel and Intact Basket Criticality Evaluation Results

Results of the criticality analyses of the intact 21 PWR WP and 12 PWR WP and basket are discussed below (Table 6.1-1 through Table 6.1-3 of Reference 11, pp. 25-30). The k_{eff} for all cases was well below the critical limit of 0.92. For the first ≈100 years after being discharged, the k_{eff} of the MOX SNF decreases as the Pu-241 (13.2-year half-life) fissile material decays. From ≈100 years out to ≈20,000 years the k_{eff} increases as the quantity of Pu-240 (6580-year half-life) and other intermediate half-life neutron absorbers is reduced through radioactive decay. After the ≈20,000 year local peak, the k_{eff} decreases again as the Pu-239 (24,400-year half-life) fissile material decays into U-235 (still highly fissile material but generally at only 80% the efficiency of Pu-239). These effects on k_{eff} are illustrated in Figures 6.2.2-1, 6.2.2-2, and 6.2.2-3 which show the $k_{eff} \pm 2\sigma$ values as a function of time for the intact 21 PWR WP, 12 PWR WP, and 12 PWR WP with Al thermal shunts, respectively. Assemblies with higher burnup values than shown in the figures will have a lower k_{eff} profile.

The 12 PWR WP designs showed higher k_{eff} values than the 21 PWR WP design for the same fuel type because these waste package designs do not include criticality control plates. The 12 PWR WP with Al thermal shunts showed an ≈1% increase in k_{eff} over the 12 PWR WP with the all carbon steel basket primarily because the Al has a much smaller neutron absorption cross section than the Fe that it replaces. The average energy of the neutron causing fission (defined as the average energy per particle lost to fission divided by the average particle weight lost to fission) was in the range of 0.18 to 0.26 MeV, and generally peaked at ≈100 years. The 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM burnup fuel generally showed higher k_{eff} values than the 4.5 wt% fissile Pu in HM, 39.4 GWd/MTHM fuel for all cases evaluated.

No specific comparisons were made of the intact MOX SNF and intact basket criticality results with LEU criticality results since the MOX SNF k_{eff}'s were well below critical values. Thus, a criticality event for this configuration is virtually impossible.

In comparing the MOX and LEU criticality, it will be noted that for the burnup-enrichment pairs chosen, the k_{eff} increases with increasing burnup and enrichment for the LEU, while it decreases with increasing burnup and enrichment for the MOX. This apparently contradictory behavior is an artifact of the burnup-enrichment pairs that were chosen. In general, for a specific waste package-fuel type combination, a number of points (burnup-enrichment pairs) can be plotted in a burnup-enrichment plane, and the corresponding values of k_{eff} can be calculated. The loci of points having the same value of k_{eff} are curves approximating a family of straight lines with approximately the same slope. The question of which of two burnup-enrichment pairs, for the same waste package-fuel type, has the higher k_{eff} is determined by whether the line connecting those points has a greater or lesser slope than the family of curves of constant k_{eff} for that waste package-fuel type.

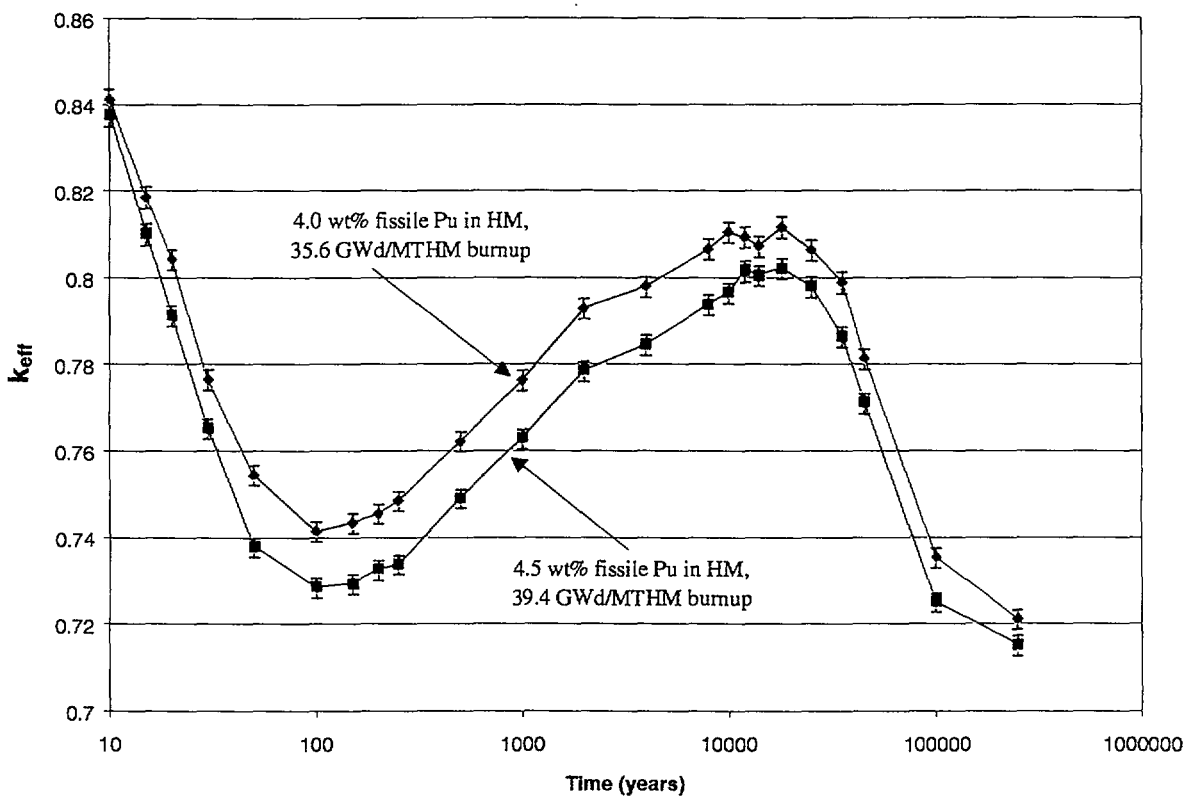

Figure 6.2.2-1. Time Effects on k_{eff} for Intact MOX SNF and Intact Baskets in a 21 PWR Absorber Plate WP

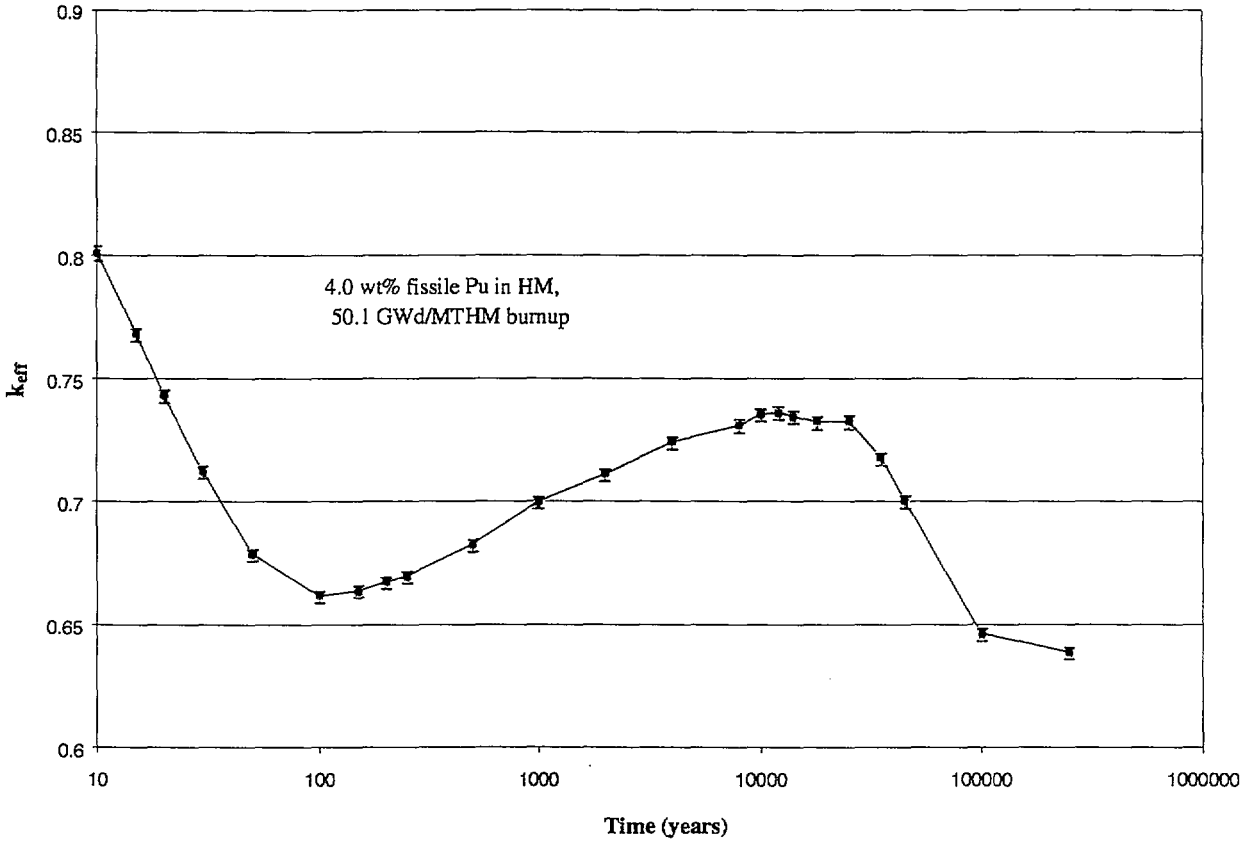

Figure 6.2.2-2. Time Effects on k_{eff} for Intact MOX SNF and Intact Baskets in a 12 PWR WP

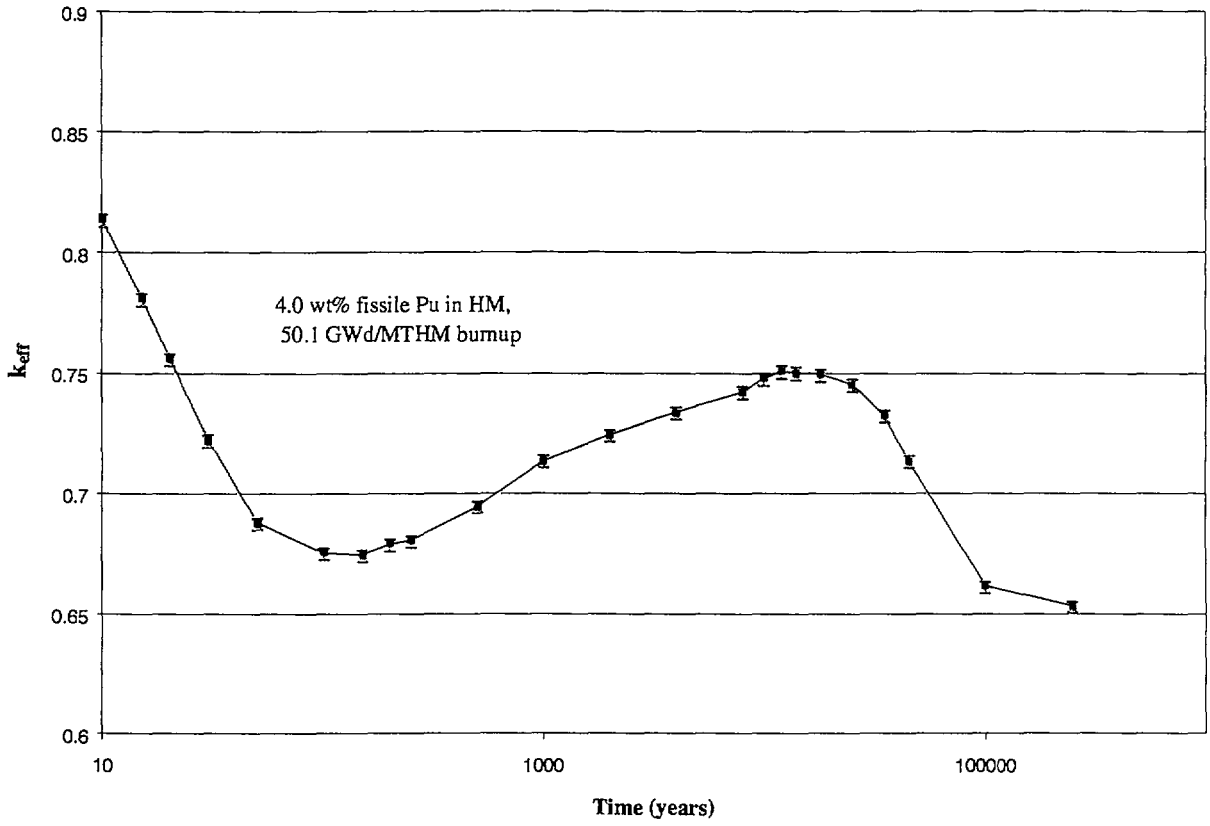

Figure 6.2.2-3. Time Effects on k_{eff} for Intact MOX SNF and Intact Baskets in a 12 PWR WP with Al Shunts

6.3 Degraded Configurations for Criticality Analysis

Analyses of the intact PWR MOX SNF evaluated the criticality potential of the water filled waste packages for the fissile and absorber nuclides in the waste package as a function of time. The analysis included radioactive decay of nuclides, geochemical degradation of materials, and removal of soluble compounds. The objectives of the geochemical analyses (Ref. 14, p. 5) were to determine the geochemical conditions under which:

1) Criticality control material suggested for this design will remain in the degraded waste package after the corrosion/dissolution of its initial form (such that it can be effective in preventing criticality), and

2) Fissile plutonium and uranium will be carried out of the degraded waste package by infiltrating water (such that internal criticality is no longer possible, but the possibility of external criticality may be enhanced).

Configurations of the SNF, as derived from the geochemical analyses, having a reasonable chance of occurring in those waste package's which experience degradation were discussed generally in Section 6.1.

Boron (B) in the form of B-SS was included in the analyses, as were various neutron absorbing fission products, notably Gd and Nd. These elements are important for inclusion in calculations of waste package internal criticality. The results of this analysis were used as input for the criticality evaluations of the degraded waste package configurations to ensure that the type and amount of criticality control material used in the waste package design will prevent criticality. These chemical compositions (and consequent criticality evaluations) were determined for time periods up to 100,000 years.

Geochemistry calculations were not performed for the 12 PWR MOX WP in Reference 14 since the results were expected to be similar to results from the 21 PWR WP adjusted for the differences in waste package volumes. The geochemistry results of the 21 MOX PWR WP indicated that all of the Fe from the carbon and stainless steel components was incorporated into Fe_2O_3, and remained in the package. Since the 12 PWR WP basket is fabricated entirely from carbon steel, it is expected that the corrosion product resulting from degradation of the basket components will also be Fe_2O_3. Since the criticality calculation considered only the initial configuration and fully degraded basket configurations (Figure 6.1-1), compositions for the 12 PWR WP can be calculated from the initial volumes.

6.3.1 Methodology for Determining Degraded Configurations

The following methodology was used for the degradation analysis (Ref. 15) of both basket materials and the waste form (MOX SNF). It is also used for the degradation analysis of the ceramic waste form documented in Volume II of this document. The geochemical analyses used the EQ3/6 V7.2b software package (Ref. 35) whose major components include: EQ3NR, a speciation-solubility code; EQ6, a reaction path code which models water/rock interaction or fluid mixing in either a pure reaction progress mode or a time mode; EQPT, a data file preprocessor; EQLIB, a supporting software library; and several (>5) supporting thermodynamic data files. The software models thermodynamic equilibrium, thermodynamic disequilibrium, and reaction kinetics in chemical systems. EQ3NR is required to initialize an EQ6 calculation and is useful for analyzing groundwater chemistry data, calculating solubility limits, and determining whether certain reactions are in states of partial equilibrium or disequilibrium. EQ6 models the consequences of irreversibly reacting an aqueous solution with a set of reactants. It can also model fluid mixing and the consequences of changes in temperature.

The method used for the geochemical analysis of the PWR MOX SNF involves the following steps:

1) Use of basic EQ3/6 program for tracing the progress of reactions with evolution of the chemistry, including the estimation of the concentrations of minerals remaining in solution and the composition of the precipitated solids.

2) Evaluation of available data on the range of dissolution rates for the materials involved, to be used as material/species input for each time step.

3) Use of the "pseudo flow-through" mode in which:

   a) Water is added continuously to the waste package and builds up in the waste package over a sequence of time steps (typically 200 to 600 steps in the initial sequence, then 15 to 18 steps per sequence in the ensuing ones). The time period per sequence is constant and is determined from the selected drip rate, e.g., 0.15 m^3/yr entering the waste package, and the percentage of added water selected. This percentage is set at 10% at the beginning of a set of runs, and typically increased to 100% to enable modeling of very long times after initial relatively rapid chemical changes have settled down to a quasi-steady state. Individual EQ6 time steps range from 0.01 seconds to 1000 days as determined automatically by the program. EQ3/6 sequences extend over times up to or somewhat greater than 100,000 years.

   b) Flushing action (removal of water added during one EQ6 sequence) is simulated by adjusting the amount of water and solutes for input to the next EQ6 sequence.

   c) Determination of fissile material concentrations in solution as a function of time.

   d) Calculation of the amount of fissile material released from the waste package as a function of time (which thereby reduces the chance of criticality within the waste package).

   e) Determination of concentrations of neutron absorbers (criticality control materials), such as B and Gd, in solution as a function of time.

   f) Calculation of the amount of neutron absorbers retained within the waste package as a function of time.

6.3.2 Degraded Configurations from Geochemistry Analysis

The emphasis in the geochemical analyses was on the composition and composition reactivity, rather than on the physical configurations within different waste packages, although the geometric configurations were used for volume calculations to determine the chemical evolution. As shown in Figure 2.2-1, a 21 PWR MOX SNF WP consists of SNF assemblies held in a basket and placed inside a corrosion barrier. The design for the corrosion barrier itself specifies an outer corrosion allowance and an inner corrosion resistant metal. For modeling of the chemical behavior of this system, the chemical compositions of each of these materials, their masses, their surface areas, and their corrosion or degradation rates are required (Tables 5.2.3-2 and 5.2.3-3 of Reference 14; compositions are given in Reference 14, Section 5.1.1.1). Elemental compositions for the SNF assemblies were obtained from the output files (Ref. 44) from the SAS2H/ORIGEN-S analysis (Ref. 6) in gram-atoms/assembly. The compositions were decayed, following discharge from the reactor, to 10,000 and 25,000 years after emplacement. The resulting isotopic changes were used to adjust the geochemistry results since the EQ3/6 code package does not account for compositional changes due to radioactive decay.

The geochemistry calculations determined the composition of the corrosion product mixture remaining in the 21 PWR waste package following complete basket degradation. Concentrations of insoluble corrosion products that remain after the basket has completely degraded are given in Section 5.3.2 of Reference 14 and Reference 14 indicates that this final composition remains fairly constant over the range of possible B-SS degradation rates and drip rates. The moles/WP column was calculated by multiplying the moles/liter H_2O column by 4550 liters of H_2O, which is the amount of water considered to be in the waste package in Reference 45 (equivalent to the void space available in a loaded waste package with an undegraded basket). The corresponding corrosion product inventory remaining in the 12 PWR WP is derived from the 21 PWR WP values with the appropriate volume ratio.

Table 6.3.2-1. Corrosion Products Remaining Following Basket Degradation in the 21 PWR WP

Basket Corrosion Product	Volume per WP (m^3)	Moles/liter H_2O	moles/WP
Diaspore (AlOOH)	1.8392E-01	2.291	10424.05
Hematite (Fe_2O_3)	1.7707E+00	12.77	58103.5
Pyrolusite (MnO2)	2.7361E-02	0.35	1592.5
Ni_2SiO_4	3.0867E-02	0.1592	724.36
Nontronite-Ca ($Si_{3.7}Ca_{0.33}Al_{0.33}$ $Fe_2H_2O_{12}$)	1.2874E-02	0.0216	98.28
Nontronite-K ($Si_{3.7}K_{0.17}Al_{0.33}$ $Fe_2H_2O_{12}$)	5.6325E-04	0.0009151	4.163705
Nontronite-Mg ($Si_{3.7}Mg_{0.2}Al_{0.33}$ $Fe_2H_2O_{12}$)	8.9323E-03	0.01513	68.8415
Nontronite-Na ($Si_{3.7}Na_{0.33}Al_{0.33}$ $Fe_2H_2O_{12}$)	9.0407E-04	0.001504	6.8432
TOTAL	2.0362E+00		

An important part of the geochemistry results is the effects that fuel degradation will have on the principal isotope inventory since these isotopes are important for burnup credit. Two cases were studied:

1) fuel degradation concurrent with basket degradation, and
2) fuel degradation beginning after basket degradation is completed.

Figure 6.3.2-1 illustrates graphically the simulated history for these elements for the MOX case in which hematite forms showing the quantity in gram-atoms of selected elements of special interest for criticality computations remaining in the MOX PWR WP for the concurrent degradation case. Times are relative to the initial breach of the corrosion barrier. The assumed inflow rate of water into the waste package was 0.15 m^3/yr. Mo and Tc are effectively removed as soluble corrosion products from the waste package as the fuel degrades. Consequently, they will be absent from the waste package, except for very minor amounts of adsorbed species or minute traces left in solution, e.g., as a consequence of incomplete mixing of water within the waste package, soon after the SNF is fully degraded.

Figure 6.3.2-1 shows the rapid total removal of Am, and the early flushing out of Np, Eu, Sm, and Gd. However, these latter elements stabilize to a (approximately) constant fraction of their original

inventory. The solubilities of all the lanthanides (Gd, Nd, Sm, and Eu) are very similar; the different histories reflect differences in their initial inventories in the waste form compositions. Only a small percentage of Nd is removed and nearly all of the U is retained. The inventories for Pu, Rh, and Ru are essentially unchanged.

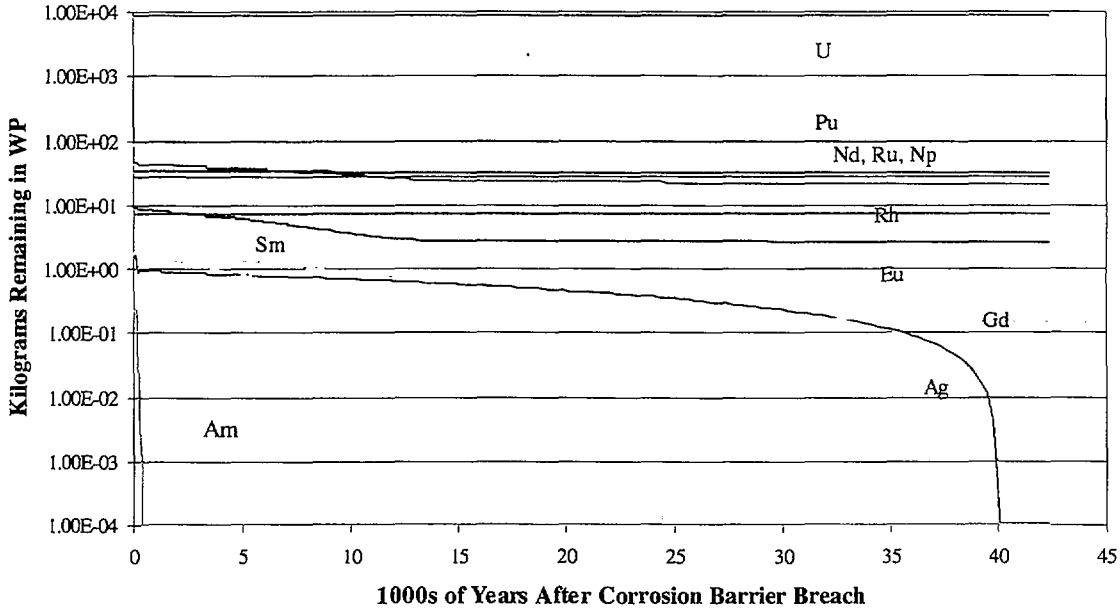

Figure 6.3.2-1. Retention History of Elements of Principal Interest for Criticality Remaining in WP

The case in which the fuel degradation was concurrent with the basket degradation resulted in more of the principal isotopes being lost due to lower pH conditions during degradation of the B-SS. This is the more conservative configuration and the results from this case are used in the degraded criticality analyses.

6.3.3 Criticality Evaluation of Degraded Waste Package Configurations

As discussed in Section 6.3.2, fuel degradation will lead to a reduced inventory of the soluble principal isotopes (summarized in Ref. 11, p. 15). Criticality calculations involving degraded fuel examined cases with the reduced principal isotope inventories, as well as a conservative case including only the U and Pu principal isotopes.

Number densities were calculated for the corrosion product and water mixtures in the waste package by dividing the moles of each element per waste package by the void space they occupied and multiplying by Avogadro's Number (0.602252×10^{24} atoms/mole). Based on the total volume of corrosion products remaining following full basket degradation, and the total volume contained within the inner barrier minus that occupied by the fuel assemblies (volume of the fuel rods), the

corrosion products will occupy 36.8% of the interior void space of a 21 PWR WP. If the corrosion products settle to the bottom of the waste package, the physical geometry of packed solids will occupy 58% of the void space (maximum percent solid content of packed sand [Ref. 12, p.15]). At 58% dense packing, if all of the oxides settle to the bottom, they will completely cover the bottom three rows of the Westinghouse 17x17 Vantage 5 MOX SNF assembly stack, and cover more than 95% of the fourth assembly row. This analysis conservatively assumes 94% or 16 of the 17 fuel pin rows in the assembly are covered.

If all of the Fe from the 12 PWR WP basket were converted to Fe_2O_3, as assumed, it would occupy 37.4% of the interior void space of a loaded waste package. If this material were settled to the bottom of the waste package at a 58% dense packing, it would cover all but the top two assemblies in a 12 PWR WP.

6.3.3.1 Configurations for Intact Fuel with Fully Degraded Basket

The MCNP4B2 cases needed to evaluate the k_{eff} of the 21 PWR MOX SNF and 12 PWR MOX SNF waste package designs with intact fuel and fully degraded basket structures (configuration D from Section 6.1) are described in this section. Both the uniformly distributed corrosion product and the settled corrosion product configurations were evaluated for each waste package. Each Westinghouse 17x17 Vantage 5 fuel assembly was treated as a heterogeneous system with the fuel rods and control rod guide tubes modeled explicitly. The fuel rods are conservatively modeled with water in the gap region and guide tubes, even when surrounded by water/corrosion product mixtures. The fuel rods are assumed to be breached but otherwise intact while the guide tubes are horizontal. There is no physical mechanism for getting basket corrosion products into these locations while the assembly remains intact. Figure 6.3.3.1-1 shows the geometry details of the MCNP4B2 model for the 21 PWR WP with a fully degraded basket and uniformly distributed corrosion products. Figure 6.3.3.1-2 shows the geometry details of the MCNP4B2 model for the 21 PWR WP with a fully degraded basket and settled corrosion products. Figure 6.3.3.1-3 shows the geometry details of the MCNP4B2 model for the base 12 PWR WP with a fully degraded basket and uniformly distributed corrosion products. Figure 6.3.3.1-4 shows the geometry details of the MCNP4B2 model for the base 12 PWR WP with a fully degraded basket and settled corrosion products. Each of the 21 PWR WP configurations was evaluated for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM burnup fuel (fuel #1), and the 4.5 wt% fissile Pu in HM, 39.4 GWd/MTHM burnup fuel (fuel #2), for decay times from 10 years to 250,000 years. In addition, the 12 PWR WP configuration was evaluated for the 4.0 wt% fissile Pu in HM, 50.1 GWd/MTHM burnup fuel (fuel #4).

Similar MCNP4B2 calculations were made to evaluate the k_{eff} for commercial LEU SNF in the 21 PWR WP design (Ref. 36, Section 6) with intact fuel and fully degraded basket structures (configuration D from Section 6.1). The commercial PWR assembly design was based on the Babcock and Wilcox (B&W) 15x15 Mark B assembly (Ref. 37, p. II.6-6). Since this assembly type has one of the largest fuel loading, it is likely to provide a conservative bounding calculation for the degraded mode criticality analysis.

The criticality potential for the LEU SNF waste forms was evaluated over decay times from 10,000 to 45,000 years for a number of enrichment-burnup combinations to identify the time of peak k_{eff} (Ref. 37, Section 6). The MCNP4B2 geometry for the LEU SNF calculations was similar to the MOX SNF geometry shown in Figures 6.3.3.1-1 and 6.3.3.1-2. Both the uniformly distributed corrosion product and the settled corrosion product configurations were evaluated for 21 LEU PWR WP. Results from a subset of the LEU enrichment-burnup combinations which are comparable to the MOX SNF wt% fissile Pu in HM-burnup combinations are included with the MOX SNF results in this study to provide a frame of reference for the MOX SNF results. These cases are as follows: 4.0 wt% U-235, 35.0 GWd/MTU and 4.5 wt% U-235, 40.0 GWd/MTU. These results show that, for most configurations, the MOX SNF has a lower criticality potential (k_{eff}) than B&W LEU SNF waste form.

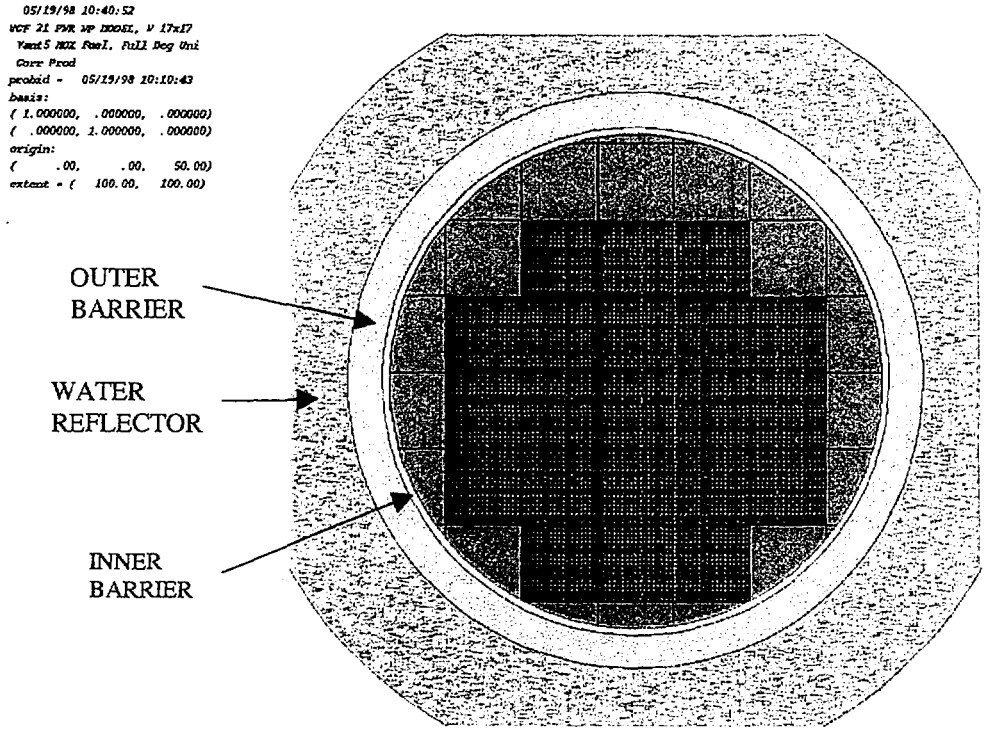

Figure 6.3.3.1-1. Degraded 21 PWR MOX Fuel Waste Package w/ Uniform Corrosion Product Distribution

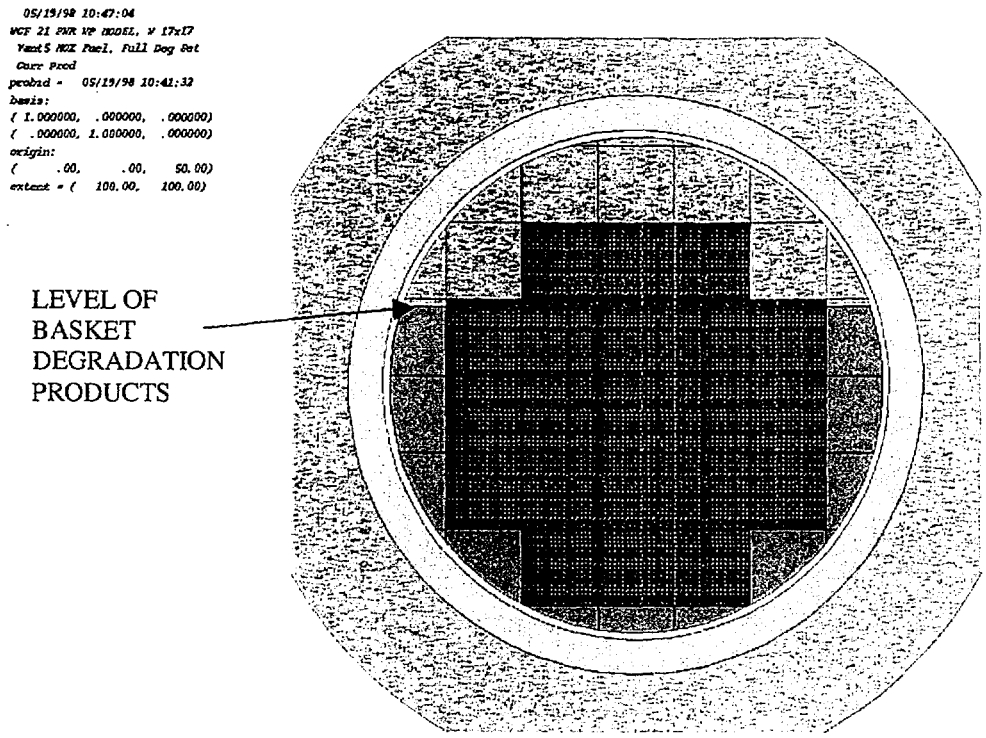

Figure 6.3.3.1-2. Degraded 21 PWR MOX Waste Package With Settled Corrosion Product Distribution (58% solid content)

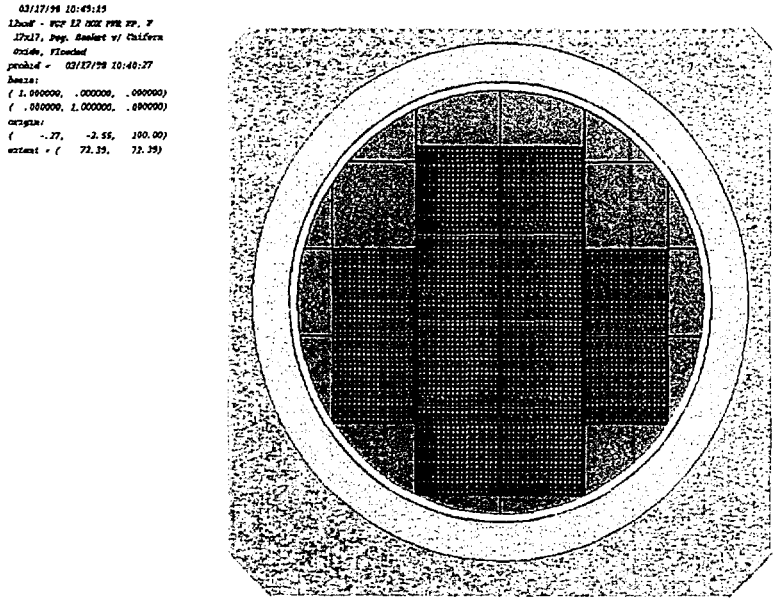

Figure 6.3.3.1-3. Degraded 12 PWR MOX Fuel Waste Package with Uniform Corrosion Product Distribution

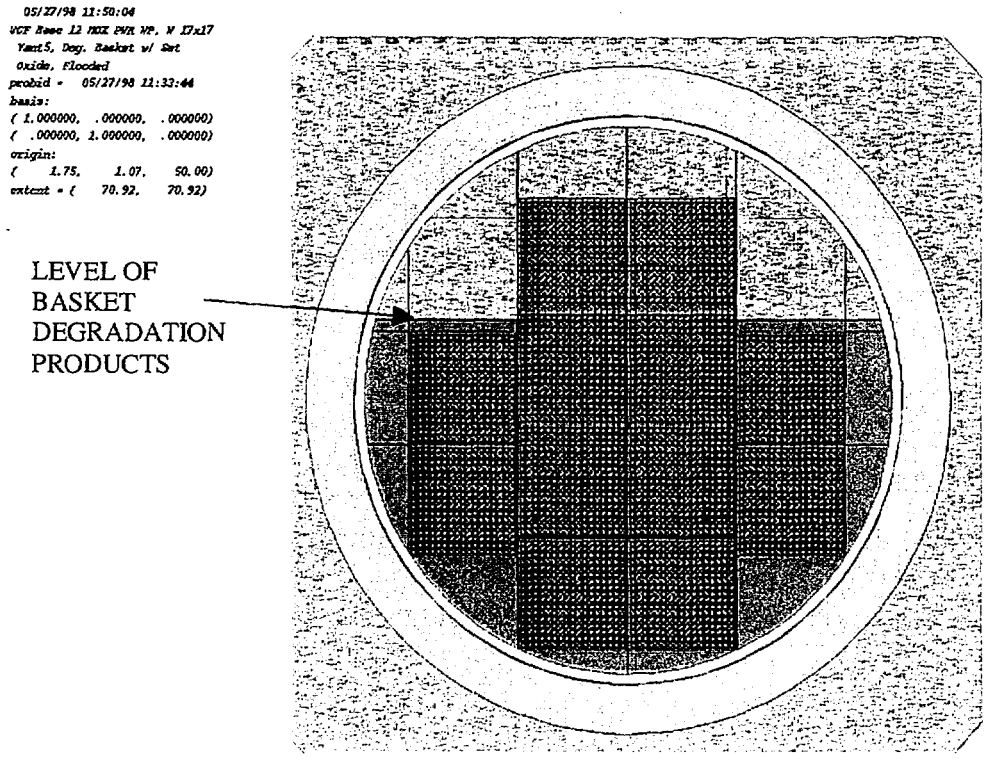

Figure 6.3.3.1-4. Degraded 12 PWR MOX Waste Package With Settled Corrosion Product Distribution (58% solid content)

6.3.3.2 Criticality Results for Intact Fuel with Fully Degraded Basket

Results of the criticality analyses of the intact fuel and degraded basket for MOX SNF in the 21 PWR WP and in the 12 PWR WP are shown (Table 6.2-1 through Table 6.2-4 of Reference 11, pp. 33-39) in Figures 6.3.3.2-1, 6.3.3.2-2, 6.3.3.2-3, and 6.3.3.2-4. The figures show the nominal k_{eff} with a 2σ variance shown as error bars. The time effect behavior is essentially the same as for the intact configurations.

As with the intact results presented in Section 6.2.2, all of the degraded cases for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM fuel consistently showed higher k_{eff} values than those for the 4.5 wt% fissile Pu in HM, 39.4 GWd/MTHM fuel. The degraded basket cases for the 21 PWR WP showed increases in k_{eff} over the intact waste package for the same fuel and decay time due to the loss of boron absorber as the B-SS absorber plates degraded. This effect, together with the fissile material inventory, more than compensated for the increased effectiveness (due to volume increase displacing moderator) of the A516 degradation products in reducing the k_{eff}. The 58 vol% settled corrosion product case showed a 6.8% increase in k_{eff} (measured at the post-closure peak for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM fuel) from the intact configuration, while the uniform corrosion product case showed only a 4.1% increase.

Figures 6.3.3.2-1 and 6.3.3.2-2 also show the k_{eff} results from the LEU SNF calculations of intact fuel and fully degraded baskets for decay times around the time (10,000 years) the k_{eff} generally reaches a secondary peak. This secondary peak is always lower than the 10 year decay values. The k_{eff} values for the LEU SNF cases are up to 4% higher than for the corresponding MOX SNF cases. Therefore, it is concluded that MOX SNF does not pose any greater, and likely less, criticality concerns in the 21 PWR WP than does LEU SNF of similar burnup and fissile content.

The degraded basket cases for the 12 PWR WP actually showed decreases in k_{eff} over the intact waste package for the same fuel and decay time. This is due solely to the increased volume of the carbon steel degradation products displacing moderator. Unlike the 21 MOX PWR WP, there is no boron to be lost in the basket degradation process and, therefore, no compensating increase in k_{eff}. The 58 vol% settled corrosion product case showed a 4.3% decrease in k_{eff} (measured at the post-closure peak for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM fuel) from the intact configuration, while the uniform corrosion product case showed a 6.8% decrease.

The k_{eff}'s were sufficiently far from critical values (0.75 max) that a criticality event is virtually impossible and no comparisons with LEU SNF were necessary.

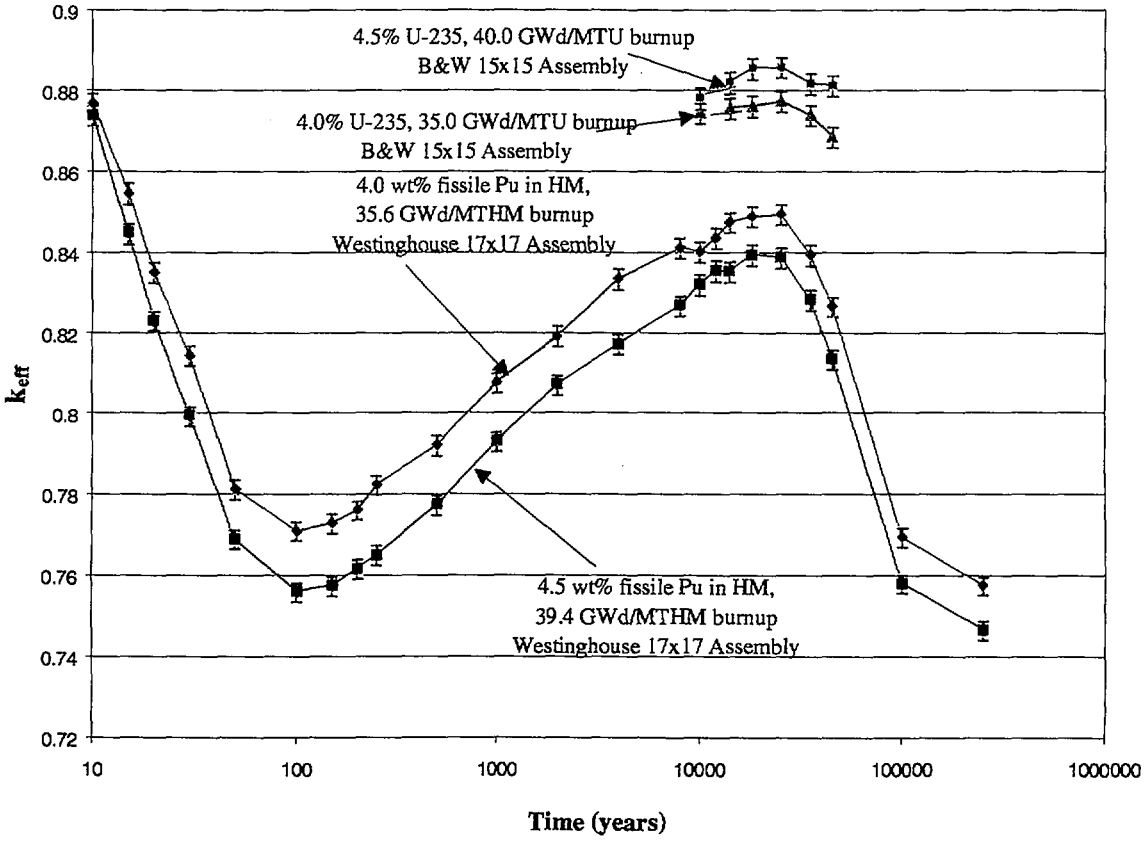

Figure 6.3.3.2-1. Time Effects on k_{eff} for Intact MOX and LEU SNF in a 21 PWR WP with a Fully Degraded Basket (No Boron Remaining) and Uniformly Distributed Corrosion Products

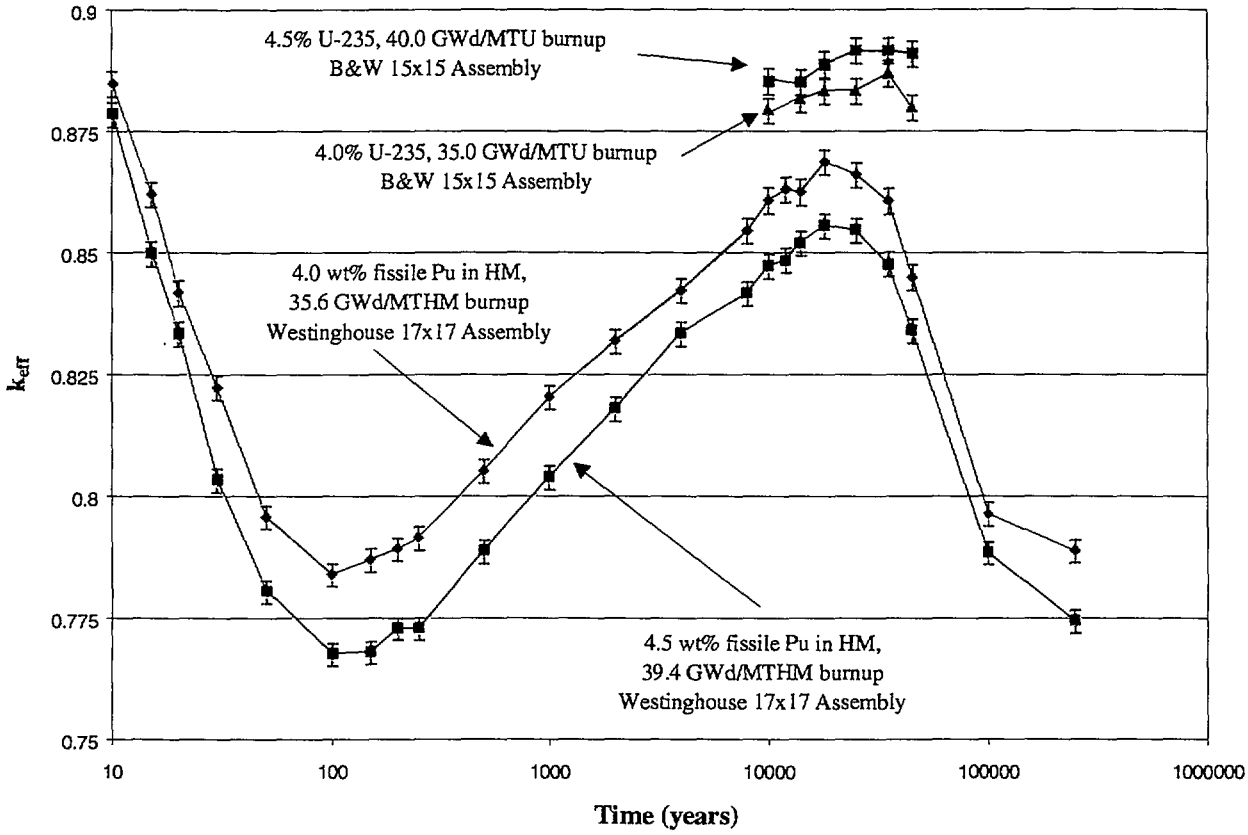

Figure 6.3.3.2-2. Time Effects on k_{eff} for Intact MOX SNF in a 21 PWR WP with a Fully Degraded Basket (No Boron Remaining) and Settled Corrosion Products

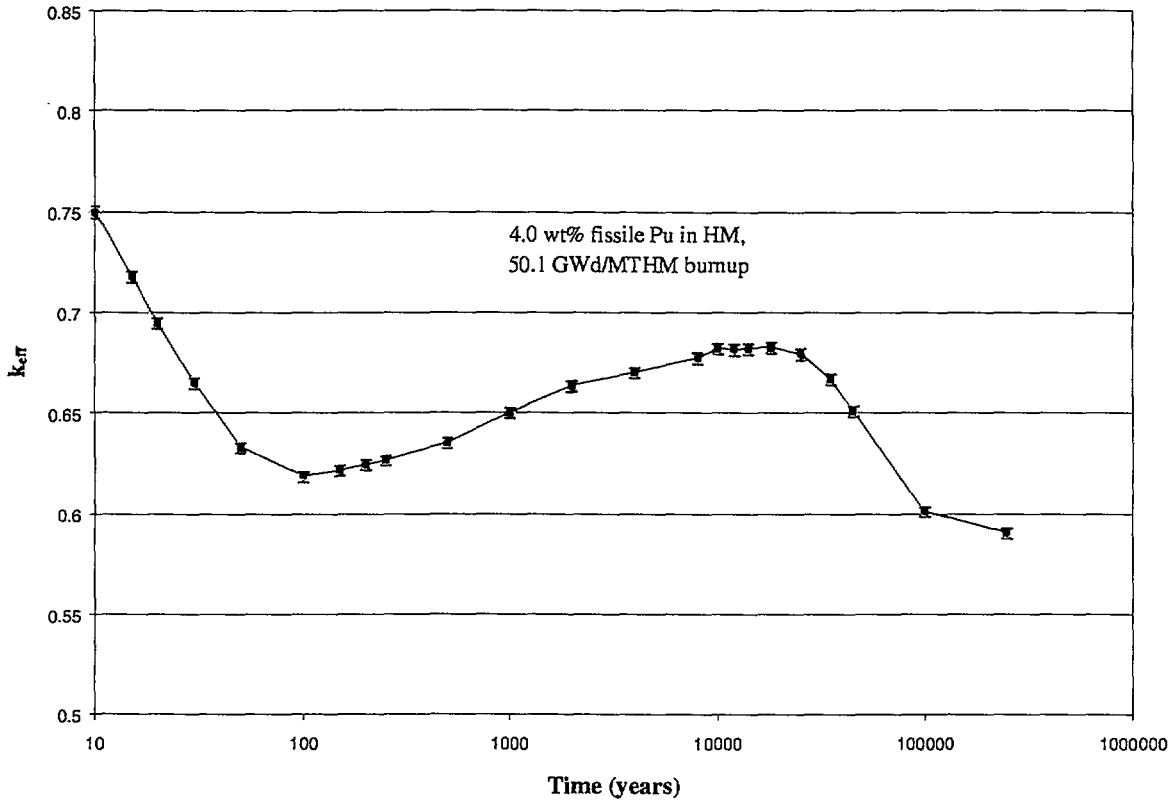

Figure 6.3.3.2-3. Time Effects on k_{eff} for Intact MOX SNF in a 12 PWR WP with a Fully Degraded Basket and Uniformly Distributed Corrosion Products

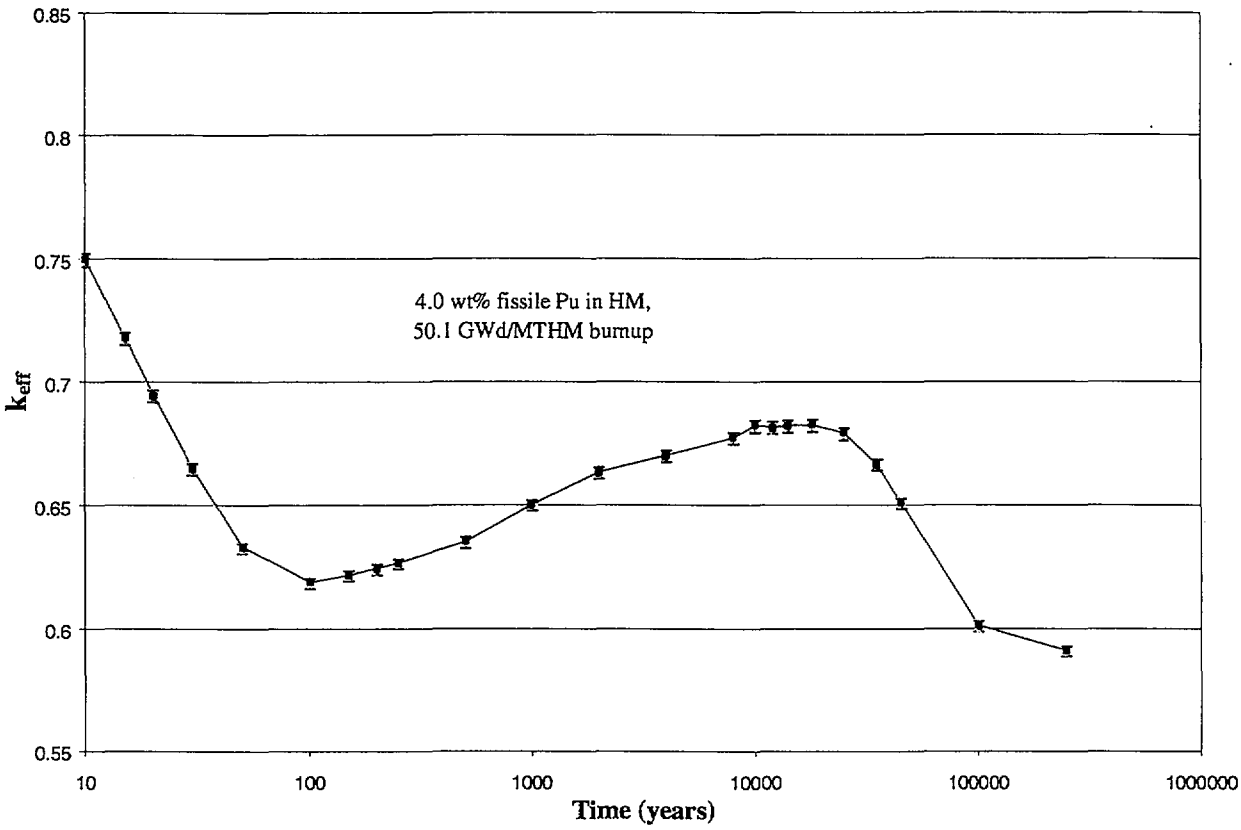

Figure 6.3.3.2-4. Time Effects on k_{eff} for Intact MOX SNF in a 12 PWR WP with a Fully Degraded Basket and Settled Corrosion Products

6.3.3.3 Configurations for Degraded Fuel and Fully Degraded Basket

The MCNP4B2 cases needed to evaluate the k_{eff} of the 21 PWR MOX SNF waste package design with fully degraded basket structures and fuel that is partially (structurally intact but allows water to fill the gap region) or fully degraded (configurations E and F from Section 6.1) are described in this section. Configuration E was modeled by settling fuel rods into a cylinder segment at the bottom of the waste package in a square lattice arrangement, as is shown in Figure 6.3.3.3-1. The square lattice is the most conservative with respect to criticality since space is available for moderator in the lattice. A more likely arrangement such as a close packed one is less conservative because of greater moderator exclusion. The height of the cylinder segment was calculated to be that which would give a volume equal to 5544 fuel rods (264 rods/assembly x 21 assemblies) in a square lattice at a given pitch. Lattice pitches ranging from 0.9144 cm (rods touching) to 1.2598 cm (as-built fuel rod pitch) were evaluated (Ref. 12, Section 6.3) to represent the range of possible separations between collapsed rods which is expected to be less than the original pitch. The k_{eff} in this sensitivity study decreased as the pin pitch decreased.

The fuel rods were modeled explicitly and contained water in the gap region. Only a uniform corrosion product distribution of 36.8 vol% (see Section 6.3.3) was evaluated. Cases were run with full isotope burnup credit, as well as for the reduced principal isotope conditions.

The fully degraded fuel and basket configuration (Configuration F) was modeled by homogenizing the remaining principal isotopes, zircaloy, and basket corrosion products in the waste package interior volume. The volume of degraded fuel material was assumed to be that which would occur if all of the initial UO_2 degraded to soddyite ($[UO_2]_2SiO_4{:}2H_2O$), as is indicated in the geochemistry calculations (Ref. 14, spreadsheet volmas2lc). Additional Si, H, and O were also added to the mixture to account for that which would be present if the fuel degraded to soddyite. The volume of zircaloy was equivalent to that contained in the cladding and guide tubes of 21 Vantage 5 SNF assemblies. All together, the degraded fuel, zircaloy, and basket corrosion products occupied 62.5% of the waste package interior volume. Water was assumed to fill the remaining void space. Figure 6.3.3.3-2 shows the geometry details of the MCNP4B2 model for the 21 PWR WP with fully degraded fuel and basket corrosion products uniformly distributed. Cases were run with reduced principal isotopes resulting from 17,500 years of radioactive decay and geochemical degradation. For comparison purposes only, a worst case was run for U and Pu isotopes only with all absorber isotopes removed.

Each of these configurations were evaluated for the 4.0 wt% fissile Pu in HM, 35.6 GWd/MTHM burnup fuel (fuel #1) for decay times from 1000 years to 250,000 years.

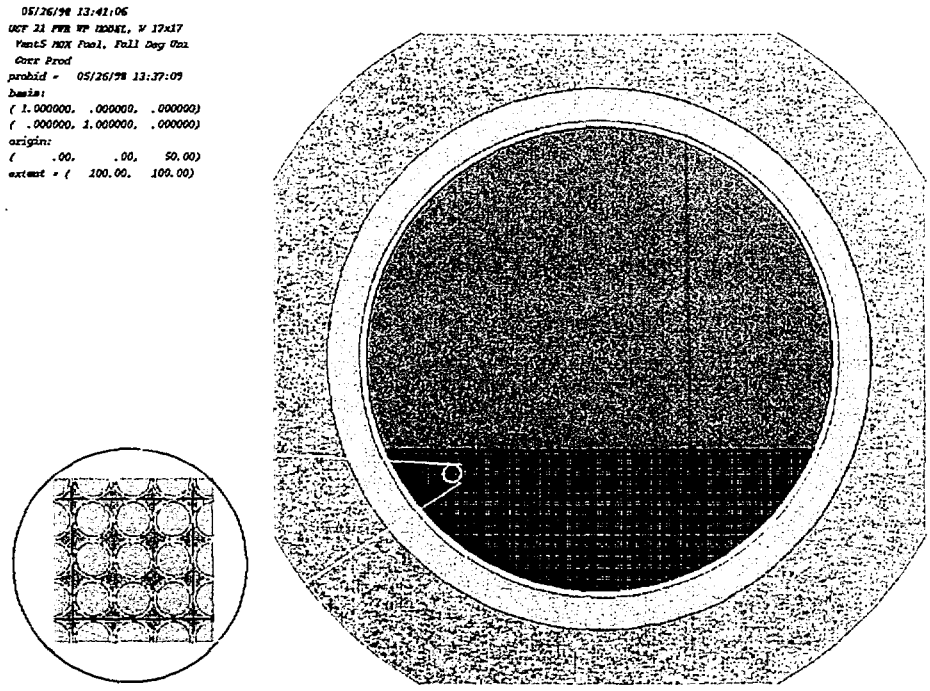

Figure 6.3.3.3-1. Degraded 21 PWR MOX WP with Fuel Rods Collapsed to Bottom of Package Surrounded by Uniformly Distributed Basket Corrosion Products

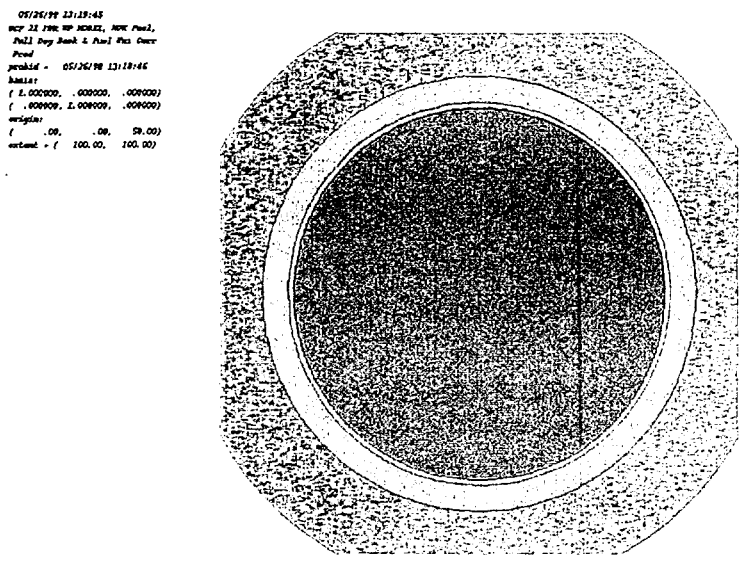

Figure 6.3.3.3-2. Fully Degraded Fuel and Basket Material Uniformly Distributed Throughout Interior Volume of 21 PWR WP

6.3.3.4 Criticality Results for Degraded Fuel and Fully Degraded Basket

Results of the criticality analyses of the 21 MOX PWR WP with a fully degraded basket, minimally spaced collapsed fuel rods (0.9144 cm), and a uniform corrosion product distribution are given in Table 6.3-1 of Reference 11, p. 46. The maximum k_{eff} value was less than 0.675 for these cases, well below the critical limit of 0.92, as shown in Figure 6.3.3.4-1 (MOX labels). All values are for a rod center-to-center spacing of 0.9144 cm (rods touching in square lattice) representative of a nominal configuration. (Note: Nominal configuration; sensitivity to rod spacing is discussed below.) An extreme case in which all principal absorber isotopes are assumed to be lost leaving only the U and Pu isotopes results in a 9% to 10% increase in k_{eff}. The ultra-conservative assumption (because of the low corrosion rate of zircaloy compared to carbon steel) that SNF degradation begins simultaneously with the baskets (see Section 6.3.2) is made in two of the analyses shown in Figure 6.3.3.4-1 (MOX Curves A and C). A more realistic SNF degradation assumption, where loss of the principal isotopes (PI) begins at 10,000 years after the start of basket degradation (MOX Curve B in Figure 6.3.3.4-1), shows a moderate increase in k_{eff} over time relative to the early loss of the PI. Also of interest is the reduced peak-and-valley effect with time, and the movement of the peak k_{eff} out to ≈45,000 years. Both effects result from increased resonance absorption due to the harder spectrum of this configuration. The location of the peak shifts outward in time because the increased absorption in Pu-240 in a harder spectrum is not matched by an equal increase in Pu-239 fission. Thus, longer decay times are required to eliminate the absorption effect from Pu-240.

Results from a similar analysis for the 21 LEU PWR WP (fully degraded baskets, minimally spaced collapsed fuel rods, and uniform corrosion products) (Ref. 37, Section 6) are also shown in Figure 6.3.3.4-1 also with the LEU labels. This case utilized the 4.0 wt% U-235, 35.0 GWd/MTU LEU SNF with a 1.0922 cm square pitch (normal pitch is 1.44272 cm). The MOX SNF k_{eff}'s were less than those from the similar LEU SNF cases; the maximum LEU SNF k_{eff} was approximately 0.7.

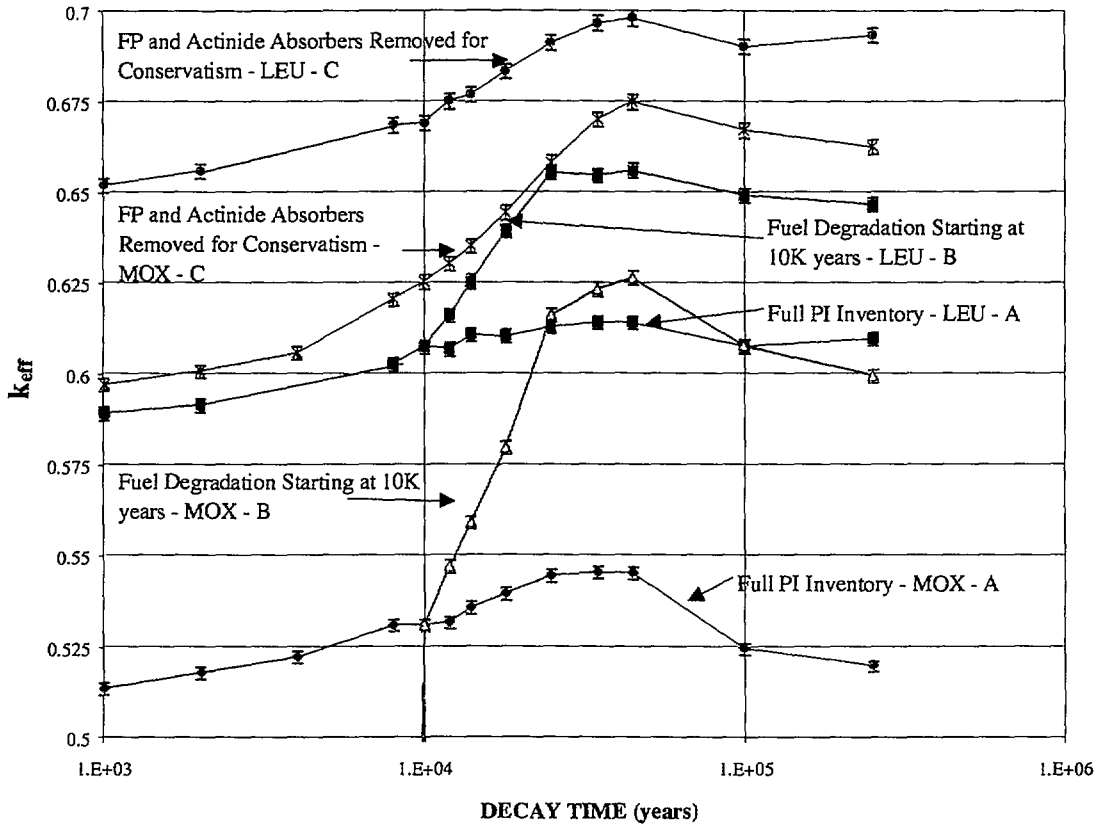

Figure 6.3.3.4-1. Time and Fuel Degradation Effects on k_{eff} for 21 PWR WP with a Fully Degraded Basket, Square Lattice Collapsed Rods (MOX and LEU), and Uniformly Distributed Corrosion Products

Results of the 18,000 and 45,000 year MOX SNF cases run for various fuel rod spacing, up to the original pitch of 1.2598 cm showed that the optimum point of moderation occurs at the original assembly pitch. However, the k_{eff} values only exceeded those of the 21 PWR WP in Configuration D with settled oxide (see Section 6.1) under the combination of extreme fuel degradation (U and Pu principal isotopes only) and rod spacing within ≈1 mm of the original pitch. This is not a likely situation, as the original rod geometry (much less the spacing) would not be expected to be retained at such a degree of fuel degradation.

Figure 6.3.3.4-2 shows the k_{eff} results for the fully degraded fuel and basket configuration (Configuration F, Section 6.1 and Figure 6.3.3.3-2) for both the MOX SNF and the LEU SNF. Note that for this case, the peak k_{eff} for the LEU SNF is approximately 4% less than the MOX SNF value of 0.845. This is well below the critical value of 0.92.

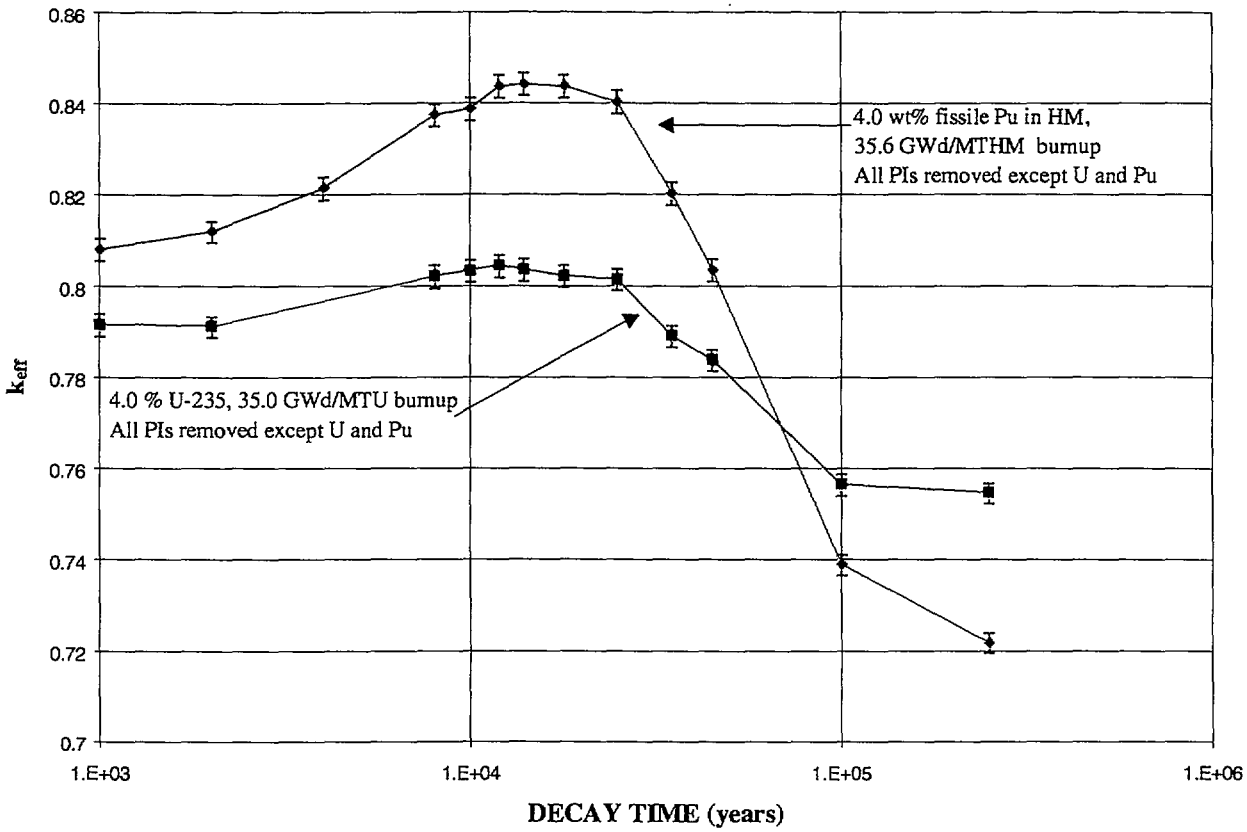

Figure 6.3.3.4-2. Time and Fuel Degradation Effects on k_{eff} for a 21 PWR WP with Fully Degraded Basket and Fuel, and Uniformly Distributed Corrosion Products

6.4 Summary of Evaluation of Potential Critical Configurations

Criticality evaluations were performed for the 21 PWR MOX SNF WP and the 12 PWR MOX SNF WP for conditions ranging from intact to fully degraded fuel and baskets. The peak k_{eff}'s are shown in Table 6.3.3.5-1. The following observations on the criticality potential of the PWR MOX SNF can be made:

1) The worst case k_{eff} is below the criticality limit of 0.92 for all waste package designs examined for any credible waste package internal configuration and thus a criticality event is virtually impossible.

2) The 12 PWR WP has a higher k_{eff} than the 21 PWR WP for the flooded intact fuel and intact basket because this waste package has no neutron absorber plates.

3) The 12 PWR WP has a lower k_{eff} than the 21 PWR WP for the flooded intact fuel and degraded basket because the iron oxide corrosion products displace moderator compensating, in part, for the absence of absorber plates.

4) The MOX PWR SNF has a lower criticality potential (k_{eff}) than the bounding LEU PWR SNF waste form for all configurations examined except for a highly unlikely configuration.

Table 6.3.3.5-1. Peak k_{eff} for Degraded PWR MOX SNF

SNF Configuration	Burnup (GWd/MTHM)		
	35.6 (4.0 wt% fissile Pu in HM)	39.4 (4.5 wt% fissile Pu in HM)	46.5 (4.5 wt% fissile Pu in HM)
	21 PWR WP	21 PWR WP	12 PWR WP
Intact Fuel, Intact Basket	0.84	0.84	0.86
Intact Fuel, Degraded Basket	0.89	0.88	0.81
Partially Collapsed and Degraded SNF – Full PIs	0.55	NA[1]	NA
Partially Collapsed and Degraded SNF – Pu and U PI	0.67	NA	NA
Fully Degraded Basket and SNF	0.84	NA	NA

[1] NA – Not Applicable; Case not run.

7. FINDINGS AND CONCLUSIONS

Structural

The most severe structural design basis event is the waste package tipover accident. The finite element analyses of both the 21 PWR and the 12 PWR WPs for this postulated event show that the peak stress at any location in either of the waste packages will be at least 15% less than the ultimate material tensile strength and thus are within design limits. Calculated stresses in the MOX PWR WPs were of similar magnitude to stresses calculated for a tipover accident in similar waste packages containing commercial PWR SNF.

Thermal

The initial heating rates for MOX SNF were 798 watts/assembly for the 21 PWR WP loaded with the highest heat source MOX SNF to be placed in that package, i.e., assemblies having no greater burnup than 46.5 GWd/MTHM. The initial heating rates were 1070 watts/assembly for the 12 PWR WP loaded with the MOX SNF generating the highest heat source, i.e., assemblies with 56.5 GWd/MTHM burnup. This loading strategy for the MOX SNF meets the maximum thermal output design criteria of 18 kW per waste package. For the 21 PWR waste package, the peak cladding temperature was approximately 234°C. The peak cladding temperature for the 12 PWR waste was approximately 218°C. The fuel temperature (homogenized assemblies) peaks at approximately 336°C for the 21 PWR MOX SNF and at approximately 302°C for the 12 PWR MOX WP. Both of these peak temperatures are well below the maximum permissible waste package fuel temperature of 350°C given in the CDA (Ref. 28, p. 8-1).

The waste package surface temperatures were determined at the thermal design basis loading of 85 MTU/acre which is within the AML range (80 to 100 MTU/acre) given in the CDA (Ref. 28, Key 019) for the repository.

Shielding

Maximum dose rates at the waste package exterior surfaces were less than 110 rad/h for the 21 MOX PWR WP loaded with the highest burnup MOX SNF at 56.5 GWd/MTHM 10 years after reactor discharge. This is conservative since this configuration produces the largest radiation source, thus maximizing the surface dose rate. The 12 MOX PWR WP design has an equivalent amount of shielding with a smaller source which will result in smaller surface dose rates.

No significant increase in the waste package barrier corrosion rate from the radiolytic effects of high surface dose rates is likely since this requires a steam-air environment combined with a greater than 100 rad/h dose rate which is a very unlikely repository condition.

The radiation levels were shown to be much less than the values from commercial LEU PWR SNF of similar burnup which were calculated to be greater than 150 rem/h. This is due primarily to the differences in the isotopic inventory in the two waste forms contributing to the radiation source.

Criticality

The peak k_{eff}'s for the MOX PWR SNF ranged from 0.55 to 0.90 where the 0.90 resulted from a worst case configuration. The k_{eff} for the worst MOX PWR SNF case is below the criticality limit of 0.92 for any credible configuration and thus any criticality event is virtually impossible.

The worst case configuration was for intact MOX SNF fuel assemblies combined with fully degraded baskets. In this configuration, all boron was assumed to be removed from the waste package, so that the only criticality control was provided by the iron oxide left from the corrosion of the carbon steel basket. The iron oxide was assumed to be in the most conservative configuration, i.e., settled). The criticality calculation for the 21 PWR package (35.6 GWd/MTHM burnup and 4% initial fissile Pu loading) resulted in a peak $k_{eff} = 0.87$. For the 12 PWR package, in which the most reactive MOX SNF is nominally 50.1 GWd/MTHM with 4% initial fissile Pu loading, the criticality calculation resulted in a peak $k_{eff} = 0.70$.

A worst case can be defined for the degraded fuel and degraded basket configuration in which the neutron absorber fission products (particularly gadolinium) are leached from the SNF while the assemblies still retain sufficient cladding and spacer grids to maintain the intact SNF geometry. The criticality calculation for this configuration resulted in a peak $k_{eff} = 0.92$. A much more likely configuration would have the SNF completely collapsed by the time the gadolinium had been leached, which leads to the much lower peak $k_{eff} = 0.63$. In both these cases the rate of removal of gadolinium from the waste package is delayed by its low solubility, so that it remains as an effective criticality control material until after the time of peak k_{eff}. These worst cases have significant gadolinium removal because the degradation of the SNF is assumed to occur while the basket steel is not completely degraded so that there is still a possibility of the pH going below 6 where the gadolinium will be moderately soluble. It is more likely that the basket will have completely degraded before the zircaloy cladding degrades sufficiently to permit significant leaching of the SNF.

In summary, analysis of the effects of placing MOX SNF in the 21 and 12 PWR waste packages will not result in the design criteria for the waste packages being exceeded provided that:

1. burnup levels for assemblies placed in the 21 PWR WP be restricted to less than 46.5 GWd/MTHM, and
2. absorber plates be used in the 21 PWR WP basket design.

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