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MOX Spent Nuclear Fuel and LaBS Glass for TSPA-LA

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Scientific Analysis/Calculation Signature Page/Change History

Complete only applicable items.

2. Document Title

MOX Spent Nuclear Fuel and LaBS Glass for TSPA-LA

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9. Remarks

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10. Revision No.	11. Description of Change
00	Initial Issue.
01	Changes in analysis due to the changes in the final draft of DCS 2005 [DIRS 173189]. Change in MOX inventory input to avoid using proprietary information from DCS. Due to changing waste package counts, the MOX and LaBS inventory adder is provided as a total grams adder instead of a grams/package adder. Use of TDMS for documentation of the spreadsheets instead of Appendix A. Addition of separate MOX fuel package and LaBS glass package inventories. Add in MOX light elements and traceability information.

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ACRONYMS AND ABBREVIATIONS

CSNF	commercial spent nuclear fuel
DCS	Duke COGEMA Stone & Webster
DOE	Department of Energy
Gd	gadolinium
GWD	gigawatt days
Hf	hafnium
HLW	high-level waste
LA	license application
LaBS	lanthanide borosilicate
MOX	mixed oxide
MT	metric ton
MTHM	metric ton heavy metal
MTPu	metric ton plutonium
O	oxygen
Pu	plutonium
Si	silicon
TSPA	total system performance assessment
U	uranium

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1. PURPOSE

This analysis provides information necessary for total system performance assessment (TSPA) for the license application (LA) to include the excess U.S. Department of Energy (DOE) plutonium in the form of mixed oxide (MOX) spent nuclear fuel and lanthanide borosilicate (LaBS) glass. This information includes the additional radionuclide inventory due to MOX spent nuclear fuel and LaBS glass and the analysis that shows that the TSPA models for commercial spent nuclear fuel (CSNF) and high-level waste (HLW) degradation are appropriate for MOX spent nuclear fuel and LaBS glass, respectively. This document was prepared in accordance with *Technical Work Plan for Waste Form Testing and Modeling* (BSC 2006 [DIRS 177389]) with one deviation. The technical work plan (TWP) (BSC 2006 [DIRS 177389]) specifies that this analysis be prepared in accordance with LP-SIII.9Q-BSC, *Scientific Analyses.* Upon transition to the Lead Laboratory (Sandia National Laboratories) on October 2, LP-SIII.9Q-BSC was replaced by SCI-PRO-005, *Scientific Analyses and Calculations.* This document is intended for use in postclosure dose analysis only and does not discuss postclosure criticality for MOX or LaBS glass.

2. QUALITY ASSURANCE

The Quality Assurance Program applies to the development of this document because this analysis is associated with the characterization of the waste form in support of TSPA-LA (BSC 2006 [DIRS 177389], Section 8). The TWP (BSC 2006 [DIRS 177389], Section 8) states that this activity is subject to *Quality Assurance Requirements and Description* (DOE 2006 [DIRS 176927]). The TWP contains the process control evaluation used to evaluate the control of electronic management of data (BSC 2006 [DIRS 177389], Appendix A) during analysis and documentation activities. This evaluation determined that the methods identified in the implementing procedures are adequate. The TWP identifies the methods to be used (BSC 2006 [DIRS 177389], Section 8). As directed in the TWP (BSC 2006 [DIRS 177389], Section 1.2.2), this document was initially prepared in accordance with LP-SIII.9Q-BSC. Upon transition to the Lead Laboratory on October 2, LP-SIII.9Q-BSC was replaced by SCI-PRO-005.

3. USE OF SOFTWARE

No controlled or baselined software subject to Section 2.0 of IM-PRO-003, *Software Management*, was used in the creation of this document.

The calculations performed in this analysis used commercially available, off-the-shelf software (Microsoft Excel 2000 9.0.6926 SP-3) on a personal computer running Windows 2000 Service Pack 4. This use of Microsoft Excel is exempt in accordance with Section 2, last bullet, of IM-PRO-003 and its use is considered appropriate because it provides: (a) the ability to derive final calculation results using simple mathematical expressions, and (b) built-in graphical charting capabilities. No software routines or macros are developed in this use of Microsoft Excel. Sections 4 and 6 discuss the analysis calculations and use of inputs. Section 6.1 provides details of the workbook and worksheet calculations and corresponding mathematical expressions. Workbooks *MOXLABS_rev01.xls,* and *McClure_rev01.xls* have been submitted to the technical data management system under output DTN: SN0611T0502505.003.

As the above examples indicate, workbook and worksheet names will be denoted in *Courier* font throughout this document.

4. INPUTS

4.1 DIRECT INPUTS

Table 4-1 provides a listing of direct inputs used in this analysis.

Table 4-1. Analysis Direct Inputs

Data/Technical Information	Input #	Value	Units	Source
grams/curie	1	Table 4-2	grams/curie	BSC 2004 [DIRS 170022], Attachment III, *inv_rev01.xls*, worksheet *gpCi*
MOX spent nuclear fuel Ci/assembly	2	DTN: SN0611T0502505.003, *McClure_rev01.xls*, rightmost sheets	curies/assembly and grams/assembly	CRWMS M&O 1998 [DIRS 105977], Attachment file "50-0g-cr.out" (CRWMS M&O 1998 [DIRS 106027])
Number of MOX assemblies	3	1,684	None	DCS 2005 [DIRS 173189], Table 3-2
Year of final MOX discharge	4	2035	year	DCS 2005 [DIRS 173189], Table 3-2
Year of LaBS glass inventory	5	2003	year	Marra and Ebert 2003 [DIRS 172949], Table 9
LaBS glass grams/canister	6	Table 4-3	grams/canister	Marra and Ebert 2003 [DIRS 172949], Table 9
LaBS glass total grams	7	Table 4-3	grams	Marra et al. 2005 [DIRS 173215], Table 2

The data from *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022]) are appropriate for use in this analysis because they are the qualified and controlled information produced specifically for Yucca Mountain inventory for use in TSPA-LA. Specifically, the grams per curie for the radionuclides, as calculated in Appendix II, Equation I-1 of that report (BSC 2004 [DIRS 170022]) and listed in the output Excel spreadsheet in Appendix III, are appropriate for use in this analysis. The values are given in Table 4-2.

Table 4-2. Grams per Curie

Isotope	g/Ci[a]		Isotope	g/Ci[a]
^{227}Ac	1.38E-02		^{242}Pu	2.54E+02
^{241}Am	2.92E-01		^{226}Ra	1.01E+00
^{243}Am	5.01E+00		^{228}Ra	3.67E-03
^{14}C	2.24E-01		^{79}Se	6.52E+01
^{36}Cl	3.03E+01		^{126}Sn	8.81E+01
^{245}Cm	5.83E+00		^{90}Sr	7.25E-03
^{135}Cs	8.69E+02		^{99}Tc	5.90E+01
^{137}Cs	1.15E-02		^{229}Th	4.68E+00
^{129}I	5.67E+03		^{230}Th	4.85E+01
^{237}Np	1.42E+03		^{232}Th	9.09E+06
^{231}Pa	2.12E+01		^{232}U	4.53E-02
^{210}Pb	1.33E-02		^{233}U	1.04E+02
^{238}Pu	5.84E-02		^{234}U	1.61E+02
^{239}Pu	1.61E+01		^{235}U	4.63E+05
^{240}Pu	4.40E+00		^{236}U	1.55E+04
^{241}Pu	9.71E-03		^{238}U	2.98E+06

[a] BSC 2004 [DIRS 170022], Attachment III, *inv_rev01.xls,* worksheet *gpCi.*

Table 4-3. LaBS Glass Inventory per Package and Total Inventory Year 2003

Isotope	g/LaBS pkg[a]	Total g[b]
^{241}Am	584	9.49E+04
^{237}Np	10	1.63E+03
^{238}Pu	72	1.17E+04
^{239}Pu	73,008	1.19E+07
^{240}Pu	6,528	1.06E+06
^{241}Pu	264	4.29E+04
^{242}Pu	128	2.08E+04
^{232}Th	328	5.33E+04
^{234}U	89	1.45E+04
^{235}U	5,496	8.93E+05
^{238}U	18,024	2.93E+06

[a] Marra and Ebert 2003 [DIRS 172949], Table 9, 13MT Case, five LaBS canisters per waste package.
[b] Marra et al. 2005 [DIRS 173215], Table 2.

The data from *Westinghouse MOX SNF Isotopic Source* (CRWMS M&O 1998 [DIRS 105977]) are appropriate for use in this analysis because they are the qualified calculation output produced specifically for the Yucca Mountain Monitored Geologic Repository (see Section 5.3 for more details). The data presented in *Mixed Oxide Fuel Interface Document for Office of Civilian Radioactive Waste Management* (DCS 2005 [DIRS 173189]) demonstrate the properties of interest and are considered qualified for use in this document per Section 6.2.1(M) of SCI-PRO-005 because of the reliability of the data source. *Mixed Oxide Fuel Interface Document for Office of Civilian Radioactive Waste Management* (DCS 2005 [DIRS 173189]) was generated to provide the latest MOX spent nuclear fuel data for the Office of Civilian Radioactive Waste Management and prepared by Duke Cogema Stone & Webster (DCS) for use in TSPA-LA. DCS, the limited liability company selected by the DOE to implement the MOX Fuel Fabrication and Reactor Irradiation Services program, is composed of three partner companies—Duke Project Services Group; COGEMA, Inc.; and Stone & Webster, Inc.—and various respected subcontractor firms, all world and industry leaders in their respective fields of expertise. In March 1999, the DOE contracted DCS to:

- Design and operate a Mixed Oxide (MOX) Fuel Fabrication Facility
- Design the commercial MOX fuel
- Use MOX fuel in commercial nuclear plants.

Although the inventory, burn-up, and discharge schedule information is still in planning stages, it is the best currently available.

Data presented in *Accounting for a Vitrified Plutonium Waste Form in the Yucca Mountain Repository Total System Performance Assessment (TSPA)* (Marra and Ebert 2003 [DIRS 172949]) provide radionuclide inventories for use in postclosure analysis. These data demonstrate properties of interest and are considered qualified for use in this document per Section 6.2.1(M) of SCI-PRO-005, qualifications of personnel or organizations generating the data. Marra earned a PhD in ceramic and materials engineering and Ebert earned a PhD in chemistry. They have written extensively on borosilicate and LaBS glasses for use as immobilizers of high-level waste in their long association with Savannah River Laboratory and Argonne National Laboratory, respectively. They have national and international reputations and their work appears in peer-reviewed journals. This document reported the best available information when it was written (2003).

Data presented in *Vitrified Plutonium Waste Form Data for Yucca Mountain License Application* (Marra et al. 2005 [DIRS 173215]) provide radionuclide inventories for use in postclosure analysis and are considered qualified for use in this document per Section 6.2.1(M) of SCI-PRO-005, qualifications of personnel or organizations generating the data. Prepared by Westinghouse Savannah River Company, which has been studying the disposal of plutonium during the plutonium immobilization program, the report provides the latest vitrified plutonium waste form data for use in TSPA-LA. As already stated, the lead author, Marra, is highly qualified. This document (Marra et al. 2005 [DIRS 173215]) reports the best available information and updates and cites *Accounting for a Vitrified Plutonium Waste Form in the Yucca Mountain Repository Total System Performance Assessment (TSPA)* (Marra and Ebert 2003 [DIRS 172949]).

4.2 CRITERIA

Criteria addressed in the TWP (BSC 2006 [DIRS 177389], Section 3) and applicable to this analysis are presented in Sections 4.2 and 4.3.

The following acceptance criterion from Section 2.2.1.3.4.3 of *Yucca Mountain Review Plan, Final Report* (NRC 2003 [DIRS 163274]) was identified in the TWP (BSC 2006 [DIRS 177389], Table 3-1) as applicable to this analysis:

Acceptance Criterion 1—System Description and Model Integration Are Adequate.

(1)–(2) Not applicable.

(3) The abstraction of radionuclide release rates and solubility limits provides sufficient, consistent design information on waste packages and engineered barrier systems. For example, inventory calculations and selected radionuclides are based on the detailed information provided on the distribution (both spatially and by compositional phase) of the radionuclide inventory, within the various types of high-level radioactive waste;

This analysis and that provided in *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022]) are based on the detailed information provided on the distribution (spatially and by compositional phase) of the radionuclide inventory, within the various types of high-level radioactive waste as described in Sections 4 and 6, and in *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022]).

(4)–(8) Not applicable.

The TWP (BSC 2006 [DIRS 177389], Section 3.3) requires that technical products address and document the accuracy, precision, and representativeness of the work performed as part of the uncertainty analyses. In addition, technical products will meet the level of detail and accuracy needed to support the TSPA-LA model.

4.3 CODES, STANDARDS, AND REGULATIONS

The TWP (BSC 2006 [DIRS 177389], Section 3.1) states that this work will conform, as appropriate, to guidance provided in ASTM C 1174-97 [DIRS 105725], *Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste*. This analysis conforms to this guidance in that it conforms "to methods used to aid in the prediction of the long-term behavior of materials, such as 'engineered barrier' system (EBS) materials and waste forms, used in the geologic disposal of high-level nuclear waste in the U.S. Government disposal site."

The TWP (BSC 2006 [DIRS 177389], Section 3.2) also states that parts of 10 CFR Part 63 [DIRS 173273] are applicable to this work. The output from this analysis will be indirectly used in the TSPA-LA postclosure analysis.

5. ASSUMPTIONS

5.1 ASSUMPTION 1

It is assumed that all 34 metric tons plutonium (MTPu) of MOX spent nuclear fuel and 13 MTPu of LaBS glass will be placed in the repository. This assumption is based on the DOE letter (Arthur 2005 [DIRS 173161]) directing Bechtel SAIC Company (BSC) to include the excess plutonium in the TSPA-LA. The technical direction letter (Arthur 2005 [DIRS 173161]) "directs BSC to include the waste inventories representing mixed oxide (MOX) spent nuclear fuel and lanthanide glass (i.e., LaBS glass) high-level radioactive waste into the LA submittal. The inventory will be approximately 34 tonnes of MOX and approximately 13 tonnes of LaBS glass." This assumption is necessary because it is a programmatic decision and does not require further confirmation. The assumption is used in Sections 6.1 and 6.1.2.

5.2 ASSUMPTION 2

For the purposes of this analysis, the number of MOX spent nuclear fuel assemblies is assumed to be 1,684, and the final year of discharge is assumed to be 2,035 (DCS 2005 [DIRS 173189], Table 3-2). These numbers have changed since Revision 00 of this analysis and are still subject to change because the initial enrichment is given as a range from 3% to 6% (DCS 2005 [DIRS 173189], p. 5), and thus the number of assemblies to contain 34 MTPu is uncertain. At 3% to 6% plutonium, 34 MTPu will be 567 metric tons initial heavy metal (MTIHM) to 1,133 MTIHM of MOX assemblies, and thus the MOX inventory will be between 0.9% to 1.8% of the allotment of 63,000 metric tons heavy metal (MTHM) for CSNF. Because the MOX is a small fraction of the CSNF inventory, the CSNF inventory is relatively insensitive to the exact number of MOX assemblies. The CSNF inventory is uncertain from a range of 0.85 to 1.40 times the nominal inventory due to uncertainty in the average burn-up (BSC 2004 [DIRS 170022], Table 7-2). Therefore, this assumption does not require confirmation because the CSNF inventory is insensitive to this assumption. This assumption is used in Section 6.1.1.

5.3 ASSUMPTION 3

It is assumed that the MOX spent nuclear fuel inventory can be represented by the inventory of case 6 of *Westinghouse MOX SNF Isotopic Source* (CRWMS M&O 1998 [DIRS 105977]). The case 6 inventory was calculated assuming an initial fissile plutonium content of 4.0% and a burn-up of 50.09 gigawatt days (GWD) per MTHM (CRWMS M&O 1998 [DIRS 105977], p. 20, Table 5.5-1). With a ^{239}Pu wt % of 93.6 (Table 5.1-3), the initial plutonium content is 4.3%. The actual initial plutonium content will range from 3 to 6% (DCS 2005 [DIRS 173189], p. 5). The actual burn-up is assumed to range from a minimum of 27, and average of 41 GWD/MTHM and a maximum of 45 GWD/MTHM (DCS 2005 [DIRS 173189], p. 5). Maximizing burn-up will maximize dose because the inventory of all isotopes except ^{238}U, ^{235}U, and ^{239}Pu increases with burn-up. ICN 00 of this analysis used inventory from a draft of *Mixed Oxide Fuel Interface Document for Office of Civilian Radioactive Waste Management* (DCS 2005 [DIRS 173189]) that was subsequently classified as proprietary. The DCS proprietary inventory was based on an assumed initial plutonium content of 5% and burn-up of 51,000 megawatt days per MTHM (51 GWD/MTHM) (DCS 2005 [DIRS 173189], p. 10), which are similar but slightly more bounding than the 4.3% and 50.09 GWD/MTHM assumed in

case 6. Given the insensitivity of the CSNF inventory to the MOX inventory, however, 50.09 GWD/MTHM is bounding enough. This assumption does not require confirmation because the CSNF inventory is insensitive to this assumption and it is conservative. This assumption is used in Section 6.1.1.

6. SCIENTIFIC ANALYSIS DISCUSSION

6.1 MOX SPENT NUCLEAR FUEL AND LABS GLASS INVENTORY

Initial Radionuclide Inventories (BSC 2004 [DIRS 170022]) provides the initial radionuclide inventory (in grams per waste package) for representative CSNF and codisposal waste packages for use in TSPA-LA. These inventories, however, do not include approximately 47 metric tons (MT) of excess DOE plutonium currently planned to be disposed in the repository (Arthur 2005 [DIRS 173161]). Of the 47 MT, approximately 34 MT are planned to be converted into MOX fuel and eventually disposed of as CSNF, and 13 MT are planned to be immobilized in a lanthanide borosilicate glass (LaBS glass) (Arthur 2005 [DIRS 173161]) (see Assumption 1 in Section 5.1). The calculations to obtain the MOX spent nuclear fuel and LaBS glass inventories were performed in the Excel workbooks *McClure_rev01.xls* and *MOXLABS_rev01.xls* (output DTN: SN0611T0502505.003.)

6.1.1 MOX Spent Nuclear Fuel Radionuclide Inventory

Even though the CSNF inventory will be relatively insensitive to the MOX inventory (see Assumption 2 in Section 5.2), an inventory adder is calculated for the effect of MOX on average CSNF inventory to prevent underestimation of the plutonium activation products. When MOX is burned, it will have higher amounts of plutonium activation products than the equivalent CSNF. Table 6-1 shows a comparison of the inventory of a MOX fuel assembly (CRWMS M&O 1998 [DIRS 105977]), versus a PWR CSNF fuel assembly (CRWMS M&O 2000 [DIRS 138239]), that was chosen to have approximately the same burn-up (50 GWD/MTHM) and age (10 years). The maximum ratio of MOX/CSNF inventory for the isotopes of interest to TSPA dose is 4.94 for ^{245}Cm followed by 2.92 for ^{241}Pu. This MOX has an initial plutonium content of 4.3% (4.0% fissile plutonium), so it represents approximately 790 MTHM, which is only 1.3% of the CSNF inventory. Thus, if 1.3% of the fuel had roughly 5 times as much ^{245}Cm, the total repository ^{245}Cm would be about 1.05 times higher. This is well within the inventory uncertainty range due to average burn-up uncertainty of 0.85 to 1.40 times. The fuel in the average waste package has lower burn-up and greater age than the assumed MOX fuel, so the ratio of short-lived plutonium activation products in the MOX versus the average CSNF is higher. Table 6-2 shows the comparison of the inventory of a 21 MOX fuel assembly waste package at discharge versus the average CSNF waste package (see workbook *MOXLABS_rev01.xls* in sheet *MOXcalc* in output DTN: SN0611T0502505.003; PWR from CRWMS M&O 2000 [DIRS 138239], batch ID 15490 in the file "nuclide_mgr_end.dat," Case_A_63k; MOX from CRWMS M&O 1998 [DIRS 105977], Case 6). The maximum ratio of MOX/CSNF inventory is 16 for ^{241}Pu. If 1.3% of the CSNF had roughly 16 times as much ^{241}Pu, then the total ^{241}Pu would be about 1.2 times higher. This is well within the inventory uncertainty range due to average burn-up uncertainty of 0.85 to 1.40 (BSC 2004 [DIRS 170022], Table 7-2). Thus, the largest increase in inventory (20%) is within the uncertainty in the inventory due to average burn-up uncertainty.

Nonetheless, to avoid an optimistic estimation of the average inventory, an inventory adder is calculated to account for the effect of MOX.

Table 6-1. Comparison of Similar MOX and PWR Fuel Assemblies

Type	PWR	MOX	
	ID 15490	Case 6	
Age (yrs)	11	10	
Enrichment (%)	4.85	4.3	
Burn-up (GWD/MTHM)	50.105	50.09	
	Curies/assembly		MOX/PWR
^{227}Ac	1.09E-05	2.97E-07	0.03
^{241}Am	1.13E+03	2.97E+03	2.63
^{243}Am	1.70E+01	4.70E+01	2.76
^{14}C	2.66E-01	5.50E-01	2.07
^{36}Cl	5.45E-03	3.89E-08	0.00
^{245}Cm	2.31E-01	1.14E+00	4.94
^{135}Cs	3.71E-01	3.94E-01	1.06
^{137}Cs	5.32E+04	5.50E+04	1.03
^{129}I	2.02E-02	2.62E-02	1.30
^{237}Np	2.38E-01	5.61E-02	0.24
^{231}Pa	3.32E-05	1.04E-06	0.03
^{210}Pb	0.00E+00	1.45E-09	—
^{238}Pu	2.40E+03	1.64E+03	0.68
^{239}Pu	1.70E+02	3.07E+02	1.81
^{240}Pu	2.76E+02	7.92E+02	2.87
^{241}Pu	4.45E+04	1.30E+05	2.92
^{242}Pu	1.30E+00	3.45E+00	2.65
^{226}Ra	0.00E+00	1.07E-08	—
^{228}Ra	0.00E+00	1.89E-12	—
^{79}Se	4.33E-02	3.96E-02	0.91
^{126}Sn	3.37E-01	6.26E-01	1.86
^{90}Sr	3.77E+04	1.88E+04	0.50
^{99}Tc	8.51E+00	8.10E+00	0.95
^{229}Th	0.00E+00	4.79E-09	—
^{230}Th	7.72E-05	5.06E-06	0.07
^{232}Th	0.00E+00	4.06E-12	—
^{232}U	2.24E-02	4.49E-03	0.20
^{233}U	2.65E-05	3.43E-06	0.13
^{234}U	6.58E-01	7.40E-02	0.11
^{235}U	9.81E-03	5.81E-04	0.06
^{236}U	1.84E-01	7.10E-03	0.04
^{238}U	1.32E-01	1.31E-01	0.99

Source: Output DTN: SN0611T0502505.003, workbook *McClure_rev01.xls*, sheet *compare MOX CSNF*.

NOTE: The ratio is printed with two decimal points, but the full three significant figures are carried in the workbook.

Table 6-2. Comparison of Average CSNF Package with MOX Package at Discharge

Isotope	CSNF (g/pkg)[a] 2033	MOX (g/pkg)[b] 2035	MOX/CSNF[c]
^{227}Ac	2.50E-06	1.13E-08	0.005
^{241}Am	8.28E+03	1.94E+03	0.234
^{243}Am	1.26E+03	4.94E+03	3.916
^{14}C	1.37E+00	2.58E+00	1.879
^{36}Cl	3.27E+00	2.48E-05	0.000
^{245}Cm	1.77E+01	1.39E+02	7.875
^{135}Cs	4.41E+03	7.17E+03	1.626
^{137}Cs	5.97E+03	1.68E+04	2.814
^{129}I	1.75E+03	3.09E+03	1.771
^{237}Np	4.63E+03	1.48E+03	0.319
^{231}Pa	9.28E-03	4.09E-04	0.044
^{210}Pb	0.00E+00	1.16E-10	—
^{238}Pu	1.54E+03	1.50E+03	0.970
^{239}Pu	4.37E+04	1.03E+05	2.355
^{240}Pu	2.08E+04	7.26E+04	3.495
^{241}Pu	2.69E+03	4.30E+04	16.014
^{242}Pu	5.34E+03	1.84E+04	3.442
^{226}Ra	0.00E+00	9.00E-09	—
^{228}Ra	0.00E+00	6.03E-15	—
^{79}Se	4.24E+01	5.42E+01	1.280
^{126}Sn	4.69E+02	1.16E+03	2.468
^{90}Sr	2.52E+03	3.65E+03	1.452
^{99}Tc	7.64E+03	9.98E+03	1.306
^{229}Th	0.00E+00	2.75E-07	—
^{230}Th	1.54E-01	4.39E-04	0.003
^{232}Th	0.00E+00	1.18E-04	—
^{232}U	1.03E-02	1.01E-03	0.098
^{233}U	5.83E-02	2.33E-03	0.040
^{234}U	1.77E+03	9.13E+01	0.052
^{235}U	6.34E+04	5.62E+03	0.089
^{236}U	3.89E+04	2.23E+03	0.057
^{238}U	7.92E+06	8.19E+06	1.034

Source: Output DTN: SN0611T0502505.003, workbook *MOXLABS_rev01.xls*, in columns I-L in sheet *MOXcalc*.

[a] CSNF from BSC 2004 [DIRS 170022], Table 7-1.
[b] MOX converted from CRWMS M&O 1998 [DIRS 105977], Attachment file "50-0g-cr.out."
[c] Three decimal places shown in spreadsheet, but only three figures are significant.

A discharge schedule is included in *Mixed Oxide Fuel Interface Document for Office of Civilian Radioactive Waste Management* (DCS 2005 [DIRS 173189], Table 3-2). Because the discharge schedule is subject to change, and to simplify the analysis, only the final year of discharge, assumed to be 2035 (Section 5), is used in this analysis. Instead of decaying the assemblies discharged each year to 2035 and then adding each up, this analysis treats all the fuel as if it will be discharged in 2035. This is conservative for scenarios with early releases such as volcanic eruption, because it overestimates the short-lived isotopes and makes no difference for scenarios with long transport times, where important daughters such as ^{237}Np will have plenty of time to grow in from short-lived parents.

To further simplify the analysis, the MOX spent nuclear fuel inventory is to be added to the CSNF inventory without displacing any CSNF. The 1,684 MOX spent nuclear fuel assemblies of 4.3% plutonium (Assumption 3 in Section 5.3) will make about 790 MTHM, which comprises only about 1.3% of the total CSNF MTHM. Thus, this simplification results in CSNF inventory that is about 1.3% high, which is a negligible difference compared to the uncertainty due to burn-up (BSC 2004 [DIRS 170022], Table 7-2). Even though this results in greater than 63,000 MT CSNF in the TSPA-LA, it does not imply that more than 63,000 MT CSNF will actually be emplaced.

The inventory per assembly at discharge and up to 10 years after discharge was copied from *Westinghouse MOX SNF Isotopic Source* (CRWMS M&O 1998 [DIRS 105977], Attachment III file "50-0g-cr.out") into the workbook *McClure_rev01.xls* (output DTN: SN0611T0502505.003), sheets *light elements input*, *actinides input,* and *fission products input* (file "50-0g-cr.out" may be obtained by requesting the electronic file from CRWMS M&O 1998 [DIRS 106027] from the Record Information System; the relevant run can be found by searching for the run time "14:17:30"). The text data was converted into columns in *light elements columns, actinides columns,* and *fission products columns*. The extra spaces within the isotope names were deleted and the data was sorted by isotope name in *light elements columns sorted, actinides columns sorted*, and *fission products columns sorted*. The important actinides (actinides that were screened in for TSPA-LA) were copied from the sorted sheet into *32ac*. Likewise, the important fission products and light elements were copied into sheet *32light+fp*. While the actinides and fission products were reported in curies per assembly, the light elements were reported in grams per assembly, so the light element inventory was converted to curies using the grams per curie from *inv_rev1.xls*, worksheet *gpCi* (BSC 2004 [DIRS 170022], Attachment III). The light element curies were added to the fission product curies in columns N and O. The 32 isotopes of importance to TSPA were copied from sheets *32ac* and *32light+fp* into sheet *32isotopes*, and from there into column D of sheet *MOXcalc* of *MOXLABS_rev01.XLS* (output DTN: SN0611T0502505.003). Curies were converted to grams using the grams per curie from *inv_rev1.xls*, worksheet *gpCi* (BSC 2004 [DIRS 170022], Attachment III), and multiplied by the number of assemblies (1,684; Section 5) to obtain the total MOX spent nuclear fuel inventory at the year 2035 as reported in Table 7-1. The inventory in Table 7-1 is to be decayed as appropriate by TSPA, converted to the grams per package using the most recent CSNF waste package count, and added to the CSNF inventory. The uncertainty of the average CSNF inventory reported in *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022], Table 7-2) is not changed by this analysis because the uncertainty in MOX spent nuclear fuel burn-up is similar to the uncertainty in burn-up of the low

enriched uranium fuel (DCS 2005 [DIRS 173189], p. 6). If TSPA models MOX fuel separately instead of averaging the MOX with the CSNF, it can model 80 MOX 21-PWR packages with the year 2035 inventory of column 3 of Table 6-2.

6.1.2 LaBS Glass Radionuclide Inventory

For TSPA-LA runs, which only model one representative HLW inventory, the LaBS glass inventory is averaged into the HLW inventory. In packages containing five canisters of LaBS glass, the LaBS glass displaces only 12% of the HLW volume (Marra et al. 2005 [DIRS 173215], p. 13). Therefore, it is a conservative simplification to add the LaBS glass inventory to the existing HLW inventory without displacing the HLW inventory. Accordingly, LaBS glass inventories (total grams) (Marra et al. 2005 [DIRS 173215], Table 2, column 2) are reported in Table 7-2. The total grams should be divided by the number of codisposal packages to yield the total grams to be added to each package. The uncertainty of the average HLW inventory reported in *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022], Table 7-2) is not changed by this analysis because there is no uncertainty associated with the assumed 13 MTPu.

If TSPA models LaBS glass waste separately instead of averaging the LaBS glass inventory with the HLW glass, it will have to make assumptions regarding how many LaBS glass canisters are loaded per waste package. If five LaBS glass canisters were loaded per waste package, and if the 13 MT of plutonium is vitrified (per Assumption 1 in Section 5.1) in 815 vitrified plutonium waste form canisters (Marra et al. 2005 [DIRS 173215], p. 6), there will be 163 LaBS waste packages and the LaBS package will contain up to 200 times the amount of ^{241}Pu and 120 times the amount of ^{239}Pu in an average glass waste package (see output DTN: SN0611T0502505.003, *MOXLABS_rev01.xls*, sheet *labspkg*). This inventory is well beyond the uncertainty of the average HLW inventory of 0.70 to 1.5 times (BSC 2004 [DIRS 170022], Table 7-2).

To capture the uncertainty in how the LaBS canisters will be loaded into a package, a variable "LABSLOAD" is defined that can take the values one through five, for the number of LaBS glass logs loaded into a waste package. The number of waste packages containing LaBS glass is then 815/LABSLOAD, and the inventory of the LaBS waste package is conservatively that of the average HLW package plus LABSLOAD times the "LaBS Adder" inventory in Table 6-3. Once again, the package inventory is not reduced for the 12% of HLW volume that the LaBS glass has displaced.

Table 6-3. LaBS Adder Inventory at 2003

Isotope	LaBS Adder (g/LaBS can)
^{241}Am	1.17E+02
^{237}Np	2.00E+00
^{238}Pu	1.44E+01
^{239}Pu	1.46E+04
^{240}Pu	1.31E+03
^{241}Pu	5.28E+01
^{242}Pu	2.56E+01
^{232}Th	6.56E+01
^{234}U	1.78E+01
^{235}U	1.10E+03
^{238}U	3.60E+03

Source: Output DTN: SN0611T0502505.003, workbook *MOXSLABS_rev01.xls*, worksheet *LaBSadder*, column H. Derived from Marra and Ebert 2003 [DIRS 172949], Table 9, 5th column; or Marra et al. 2005 [DIRS 173215], Table 2, column 2.

6.2 MOX SPENT NUCLEAR FUEL DEGRADATION

6.2.1 Introduction

Mixed oxide fuel (MOX fuel) is a combination of plutonium oxide and uranium oxide. It is similar to CSNF containing only uranium oxide. The fuel pellets and fuel rod cladding are exactly the same in both cases. The pellets are fabricated in a similar manner in that milled powders are blended together, compacted, and sintered, except extra precautions are taken to isolate the workers against exposure to plutonium-containing powders. The cladding will be identical to the current state-of-the-art cladding now utilized by fuel vendors to take advantage of improved process controls and materials. A summary report by Seibold et al. (2000 [DIRS 172859]) compares the performance of the improved claddings for boiling water reactors and pressurized water reactors. Since MOX fuels will utilize improved cladding, they will likely have better performance than the average CSNF cladding, which has a preponderance of older designs and materials.

Instead of relying on the isotope ^{235}U to provide the fission neutrons in the reactor as in CSNF, MOX fuel utilizes ^{239}Pu instead. Once in a reactor, CSNF breeds ^{239}Pu and other transuranic isotopes through absorption rather than fission processes. When spent CSNF is removed from a reactor, the ^{235}U is depleted from an initial content (enrichment) of about 4% down to about 1% and it contains about 1% plutonium and about 3% fission products. The fission yields of the two fuels are similar, but not identical. Some fission products and transuranic isotopes are present in greater numbers while others are lower. In addition, the distribution of these products across the fuel pellets is somewhat different as noted below.

The replacement of MOX fuel in U.S. light water reactors has been recently studied. Anderson et al. (2002 [DIRS 172993]) discussed information relative to pressure vessel integrity and performed transport calculations to examine the impact of the partial replacement of uranium oxide with MOX in McGuire and Catawba reactor cores. They found that although the

calculated neutron fluxes, and expected total fluence, at the wall of the pressure vessel was slightly higher (4% to 6%), the impacts were negligible. Taylor (1965 [DIRS 130762]) made a similar assessment for a partial MOX core for the Saxton reactor. In that report, detailed results of the Westinghouse Electric Corporation critical assembly experiments were provided. Taylor (1965 [DIRS 130762]) found that buckling data, relative power distributions, and relative fluxes were as anticipated (i.e., slightly higher, but similar to the core characteristics with uranium oxide).

6.2.2 In-Reactor Performance

In-reactor experience is available that allows the comparison of MOX spent nuclear fuel and CSNF. The results from three general sources of experiments are available and include those that were prototypic of expected MOX fuels performed in the Advanced Test Reactor, those that come from foreign (mostly French) reactors, and those that come from experiments designed to quickly obtain specimens for postclosure performance, which were conducted by the National Spent Fuel Program Office.

Hodge and Ott (2004 [DIRS 172810]) performed tests in the Advanced Test Reactor in Idaho involving irradiation of capsules placed in the Advanced Test Reactor at heat generation rates slightly higher than those anticipated in a commercial reactor. However, the thermal conditions and total burn-up of the pellets are similar to that expected for MOX fuel. The capsules contained standard-sized pellets of MOX containing 5% weapons-derived plutonium oxide. The capsules were withdrawn periodically and post-irradiation examinations were conducted as detailed by Morris et al. (2004 [DIRS 172811]). Nondestructive examination (dimension and gamma scans) revealed no anomalies. Fission gas release from capsules exposed for 40 GWD/MT was determined. Hodge and Ott (2004 [DIRS 172810], Figure 5) showed fission gas release to be similar to that obtained for CSNF irradiated at similar power conditions. Destructive examination showed that the structure of the fuel was similar to CSNF; however, some agglomerates high in plutonium oxide were clearly visible. The agglomerates were due to the blending process utilized and are not expected with improved blending techniques. Blanpain et al. (2004 [DIRS 172793]) recently summarized the performance of MOX fuel assemblies fabricated by Framatome ANP in commercial reactors. The authors note that at the end of 2003, more than 3,800 MOX fuel assemblies have been delivered to French, Belgian, German, and Swiss light water reactors. Operating experience of burn-ups of about 45 GWD/MT in France and up to 62 GWD/MT in Germany has shown the good performance and reliability of MOX fuel. Examinations of over 100 fuel rods from surveillance programs have shown that there were no significant differences in MOX fuel assembly characteristics relative to those of CSNF. These examinations included dimensional inspection, corrosion, and fission gas release.

The National Spent Fuel Program Office utilized existing MOX fuel rods irradiated previously in the Experimental Breeder Reactor II (EBR-II) as part of a US/Japan cooperative program. The characterization of these rods is described in *Review of DOE Spent Nuclear Fuel Release Rate Test Results* (DOE 2003 [DIRS 166268], pp. 5-1 to 5-5). The fuel pellets contained ~25 wt % plutonium and had a high enrichment of ^{235}U to meet the high-power requirements of the EBR-II. Both solid and annular pellets were irradiated. Characterization included dimensional analysis and gamma scanning. Little change in dimensions occurred, with only small increases

found where cesium peaks in the gamma scans were observed. The gamma scans showed the redistribution of volatile fission products expected in this high-temperature (up to 2,100°C centerline) irradiation. The structure of the fuel consisted of three zones, an inner zone of columnar grains, a central region of equiaxed grains, and an outer region of unrestructured grains. Microprobe analysis confirmed the redistribution of the volatile fission products. While not stated in this report, the characteristics observed are similar to those observed for highly enriched, high-temperature irradiations of uranium oxide (Dehaudt 2001 [DIRS 164019]).

6.2.3 Postclosure Performance

The dissolution rates of MOX spent nuclear fuel was compared to that of CSNF in a series of different tests performed by the National Spent Fuel Program Office. These tests were summarized recently in *Review of DOE Spent Nuclear Fuel Release Rate Test Results* (DOE 2003 [DIRS 166268]). However, as noted above, the MOX spent nuclear fuel chosen for these tests was that obtained from the EBR-II, so the irradiation conditions were significantly hotter than those experienced by average CSNF. The DOE review (DOE 2003 [DIRS 166268]) describes flow-through tests in Section 5.2 and drip tests in Section 5.3. The flow-through tests showed that the dissolution rate of the plutonium-oxide phase was slower than the uranium-oxide phase in MOX spent nuclear fuel and that the uranium-oxide phase in MOX fuel dissolved more slowly than the uranium oxide in CSNF (DOE 2003 [DIRS 166268], p. 5-17.) The drip tests showed that the release rates for all radionuclides, except technetium, were faster for MOX fuel than from CSNF. However, after 2.5 years, the release rates of radionuclides (except ^{129}I and ^{99}Tc) were within an order of magnitude for the two fuel types (DOE 2003 [DIRS 166268], pp. 5-25 through 5-26). These differences may be attributed to the nature of the fuels themselves and the different irradiation conditions noted above.

Recent leach tests by Cobos et al. (2004 [DIRS 172813]) were performed on unirradiated uranium oxide and 10% plutonium oxide MOX fuel in a carbonate groundwater. For these tests, the MOX fuel was prepared utilizing a sol-gel technique to ensure intimate mixing of plutonium in the oxide matrix. The pellets were cut into 1-mm-thick discs and submerged in the test solutions for over 1,000 hours in the deaerated condition. The normalized uranium leach rate after 1,000 hours for MOX fuel was similar to that for the uranium oxide fuel. The normalized release rate for plutonium was two orders of magnitude lower than that of the uranium. While the conditions are not prototypic of those expected at Yucca Mountain, the similarity of leaching performance of the two fuels adds confidence that the fuels will perform similarly postclosure.

6.2.4 Summary of In-Reactor and Postclosure Performance

The in-reactor and post-reactor dissolution properties of the two fuels are very similar, as discussed above. Thus, given the wide variability of the CSNF fuels considered due to enrichment and burn-up differences, the performance of MOX spent nuclear fuel is consistent with the performance of the CSNF fuel. Therefore it is reasonable to use the CSNF models for degradation rate, initial release fractions and carbon-14 hardware inventory for the MOX fuel.

6.3 LABS GLASS DEGRADATION

Lanthanide borosilicate glass was derived from the Loffler glass, which was originally designed for submarine periscope lenses. At Savannah River, it was developed for converting a solution containing Am and Cm into a form that could be shipped to Oak Ridge National Laboratory (Bibler et al. 1996 [DIRS 172928]). This glass was adapted in the mid-1990s for use in the disposal of weapons-grade plutonium (Bibler et al. 1996 [DIRS 172928]). Recently, the LaBS glass is being considered for immobilization of weapons-grade plutonium that, for composition or other reasons, will not be processed at the Mixed Oxide Fuel Fabrication Facility (Marra and Ebert 2003 [DIRS 172949]). LaBS glass is a rare earth-bearing glass into which about 15 mass % PuO_2 can be dissolved. It is a high-temperature glass in that the processing temperatures are about 1,500°C. At a 9.5 mass % PuO_2 concentration, the LaBS glass contains a 2.66:1 neutron absorber (Gd + Hf)-to-fissile atom ratio and more than 8 times more moles of boron than fissile isotopes (Marra et al. 2005 [DIRS 173215], p. 16). The neutron absorbers were selected to ensure that their aqueous solubilities bracket those of Pu(IV) and U(VI) (^{235}U is the decay product of ^{239}Pu). Therefore, as the glass dissolves in water, the neutron absorbers remain with or in close proximity to the fissile elements (plutonium or uranium) to decrease the probability of nuclear criticality. This analysis is not concerned with the probability of a nuclear criticality but deals with the behavior of the LaBS glass as it influences postclosure dose.

Strachan et al. (1998 [DIRS 172932]) report results from single-pass flow-through tests that show the tested LaBS glass had similar dissolution rates at 90°C to those of other high-level and low-activity waste glasses, also tested at 90°C, that are being or will be produced in the U.S. and elsewhere in the world. Change in the forward-dissolution rate with changes in solution pH values is similar to these other glasses, as well. Although the temperature dependence of the dissolution rate has not been measured, it is expected that the rate-limiting step will be the dissolution of H_4SiO_4 as is postulated for silica-based waste glasses and silicate minerals. Thus, the activation energy is anticipated to fall within the range of other silica-based glasses (70 kJ/mol to 90 kJ/mol) (Strachan 2001 [DIRS [173684], Sections 3 and 5). Therefore, it is reasonable to use the degradation model in *Defense HLW Glass Degradation Model* (BSC 2004 DIRS 169988]) to represent degradation of LaBS glass in TSPA-LA.

7. CONCLUSIONS

This analysis provides information necessary for TSPA-LA to include the excess DOE plutonium in the form of MOX spent nuclear fuel and LaBS glass. This information includes the additional radionuclide inventory due to MOX spent nuclear fuel and LaBS glass, described in Section 6.1, and the analyses (Sections 6.2 and 6.3) that show that the TSPA models for CSNF and HLW degradation are appropriate for MOX spent nuclear fuel and LaBS glass, respectively.

7.1 DEVELOPED OUTPUT

As discussed in Sections 6.2 and 6.3, MOX spent nuclear fuel is to be treated as CSNF and the LaBS glass is to be treated as HLW glass for degradation rate purposes. An analysis of the excess inventory due to MOX spent nuclear fuel and LaBS glass at the date of the input (2035 and 2003) is provided in output DTN: SN0611T0502505.003 and reported in Tables 7-1 and 7-2. These inventories are to be decayed as appropriate by TSPA, divided by the correct

number of waste packages and added to the existing per package inventories from *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022]). The uncertainty of the average HLW and CSNF inventories reported in *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022], Table 7-2) is not changed by this analysis.

Table 7-1. MOX Spent Nuclear Fuel Total Grams Adder at 2035

Isotope	Average Adder (g)		Isotope	Average Adder (g)
^{227}Ac	9.08E-07		^{242}Pu	1.47E+06
^{241}Am	1.56E+05		^{226}Ra	7.22E-07
^{243}Am	3.96E+05		^{228}Ra	4.84E-13
^{14}C	2.07E+02		^{79}Se	4.35E+03
^{36}Cl	1.99E-03		^{126}Sn	9.29E+04
^{245}Cm	1.12E+04		^{90}Sr	2.93E+05
^{135}Cs	5.75E+05		^{99}Tc	8.01E+05
^{137}Cs	1.35E+06		^{229}Th	2.20E-05
^{129}I	2.48E+05		^{230}Th	3.52E-02
^{237}Np	1.18E+05		^{232}Th	9.44E-03
^{231}Pa	3.28E-02		^{232}U	8.09E-02
^{210}Pb	9.32E-09		^{233}U	1.87E-01
^{238}Pu	1.20E+05		^{234}U	7.32E+03
^{239}Pu	8.25E+06		^{235}U	4.50E+05
^{240}Pu	5.82E+06		^{236}U	1.79E+05
^{241}Pu	3.45E+06		^{238}U	6.56E+08

Source: Output DTN: SN0611T0502505.003, workbook *MOXSLABS_rev01.xls*, worksheet *MOXcalc*.

Table 7-2. LaBS Glass Total Grams Adder at Year 2003

Isotope	LaBS Adder (g)
^{241}Am	9.49E+04
^{237}Np	1.63E+03
^{238}Pu	1.17E+04
^{239}Pu	1.19E+07
^{240}Pu	1.06E+06
^{241}Pu	4.29E+04
^{242}Pu	2.08E+04
^{232}Th	5.33E+04
^{234}U	1.45E+04
^{235}U	8.93E+05
^{238}U	2.93E+06

Source: Output DTN: SN0611T0502505.003, workbook *MOXSLABS_rev01.xls*, worksheet *LaBSadder*, column D.

If TSPA chooses to model LaBS glass and MOX SNF as separate waste package types, then the average inventories are not increased by the adder. Instead, the MOX inventory is given in Table 6-2, column 3, and must be decayed and divided by the correct waste package count. The LaBS inventory is calculated by decaying the per-canister adder from Table 6-3, multiplying by the sampled parameter "LABSLOAD," and adding to the decayed per package HLW inventory from *Initial Radionuclide Inventories* (BSC 2004 [DIRS 170022], Table 7-1). Until there is a project position on the loading of LaBS canisters into waste packages, LABSLOAD should be uniformly sampled on the integers between one and five (inclusive). There are 815/LABSLOAD LaBS codisposal waste packages and 80 MOX waste packages.

This analysis is an indirect feed to TSPA-LA.

This analysis was conducted for TSPA-LA postclosure dose purposes only, and does not discuss postclosure criticality for MOX or LaBS glass.

8. INPUTS AND REFERENCES

8.1 REFERENCES

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