AUG 1 1996

Sta 34

22

ENGINEERING DATA TRANSMITTAL

Page 1 of 1

1. EDT 616633

2. To: (Receiving Organization) Distribution	3. From: (Originating Organization) Plutonium Process Support Laboratory	4. Related EDT No.: N/A
5. Prog./Prog./Dept./Div.: SNFP/TAP	6. Cog. Engr.: T. D. Cooper	7. Purchase Order No.: N/A
8. Originator Remarks: This document is being distributed for approval and release.		9. Equip./Component No.: N/A
		10. System/Bldg./Facility: N/A
11. Receiver Remarks:		12. Major Assm. Dwg. No.: N/A
		13. Permit/Permit Application No.: N/A
		14. Required Response Date: N/A

DATA TRANSMITTED					(F)	(G)	(H)	(I)
(A) Item No.	(B) Document/Drawing No.	(C) Sheet No.	(D) Rev. No.	(E) Title or Description of Data Transmitted	Approval Designator	Reason for Transmittal	Originator Disposition	Receiver Disposition
1	WHC-SD-SNF-TI-033		0.00	Spent Nuclear Fuel Project (SNFP) Estimate of Volatile Fission Products Release From Multi-Canister Overpacks	S	1-2	1-2	

16. Approval Designator (F)		KEY Reason for Transmittal (G)				Disposition (H) & (I)	
E, S, Q, D or N/A (See WHC-OM-3-5, Sec. 12.7)		1. Approval	4. Review	Reviewed no/comment		1. Approved	4. Reviewed no/comment
		2. Release	5. Post-Review	Reviewed w/comment		2. Approved w/comment	5. Reviewed w/comment
		3. Information	6. Dist. (Receipt Acknow. Required)	Receipt acknowledged		3. Disapproved w/comment	6. Receipt acknowledged

17. SIGNATURE/DISTRIBUTION (See Approval Designator for required signatures)										(G)	(H)
Reason	Disp.	(J) Name	(K) Signature	(L) Date	(M) MSIN	(J) Name	(K) Signature	(L) Date	(M) MSIN	Reason	Disp.
1-2	1	Cog. Eng. TD Cooper	<i>T.D. Cooper</i>	T5-12		DR Duncan	<i>DR Duncan</i>	7/29/96	R3-86	1-2	2
1-2	1	Cog. Mgr. PK Shen	<i>PK Shen</i>	H0-40							
	1	QA									
1-2		Safety SE Numan	<i>A.M. Zaman</i>	7/31/96	R3-87						
		Env.									
1-2	1	PA Scott	<i>PA Scott</i>	7/31/96	R3-87						
1-2	1	AM Zaman	<i>AM Zaman</i>	8/1/96	B1-10						

18. Signature of EDT Originator <i>T.D. Cooper</i> 7-30-96 Date Originator TD Cooper	19. Authorized Representative <i>PA Scott</i> 7/31/96 Date Authorized Representative PA Scott	20. Cognizant Manager <i>PK Shen</i> Date Cognizant Manager PK Shen	21. DOE APPROVAL (if required) Ctrl. No. - N/A ☐ Approved ☐ Approved w/comments ☐ Disapproved w/comments

BD-7400-172-2 (04/94) GEF097

BD-7400-172-1

Spent Nuclear Fuel Project Estimate of Volatile Fission Products Release From Multi-Canister Overpacks

Thurman D. Cooper

Westinghouse Hanford Company, Richland, WA 99352

U.S. Department of Energy Contract DE-AC06-87RL10930

EDT/ECN: 616633

UC: 510

Org Code: 15500

Charge Code: LB040

B&R Code: EW3135040

Total Pages: ~~22~~ 25 8-1-96

Key Words: MCO, cold vacuum dried, hot vacuum dried, Canister Storage Building, fission

Abstract

Spent N-Reactor fuel will be moved from wet pool storage to dry storage at Hanford Washington. This fuel will be sequentially loaded into a Multiple Container Overpack (MCO), moved to the cold vacuum drying station, drained, cold vacuum dried, shipped to the Canister Storage Building (CSB), staged for up to 2 years, hot vacuum dried at 300 °C, hot conditioned at 150 °C, and finally, sealed and stored for up to 75 years in the CSB.

During each proposed process step, the volatile radioactive fission products released to the atmosphere were estimated. Tritium is the only volatile fission product released in significant amounts during each process step.

For an accident scenario involving interior MCO temperatures of 600 °C for up to 8 hours, it was estimated that many volatile fission products are released.

TRADEMARK DISCLAIMER Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors.

Printed in the United States of America. To obtain copies of this document, contact: WHC/BCS Document Control Services, P.O. Box 1970, Mailstop H6-08, Richland WA 99352, Phone (509) 372-2420, Fax (509) 376-4989.

Chris Stillingham 8-1-96
Release Approval

Date

Release Stamp

Approved for Public Release

SPENT NUCLEAR FUEL PROJECT ESTIMATE OF VOLATILE FISSION PRODUCTS RELEASE FROM MULTI-CANISTER OVERPACKS

July 1996

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and Waste Management

Westinghouse Hanford Company
P.O. Box 1970
Richland, Washington

This page intentionally left blank.

CONTENTS

1.0 INTRODUCTION	1
2.0 EMPIRICAL MODELS	3
3.0 AIRBORNE RELEASE RATES	9
4.0 FISSION PRODUCT RELEASE CASES	11
4.1 CASE 1	11
4.2 CASE 2	12
4.3 CASE 3	13
4.4 CASE 4	14
4.5 CASE 5	15
5.0 DISCUSSION OF RESULTS	17
6.0 REFERENCES	19

LIST OF TABLES

1. Parameters for Radionuclide Loss from UO_2 Matrix	4
2. Arrhenius Parameters	7
3. High Temperature Fission Product Release from Uranium Metal	11
4. Fission Product Release During Hot Vacuum Drying	12
5. Fission Product Release During Cold Vacuum Drying	13
6. Fission Product Release During Hot Conditioning	14
7. Fission Product Release During Long Term Storage	15
8. Internal and External Radiation Dose Estimates	17

LIST OF FIGURES

1. Diffusion Rate Constants for Strontium/Yttrium-90	6

SPENT NUCLEAR FUEL PROJECT ESTIMATE OF VOLATILE FISSION PRODUCTS RELEASE FROM MULTI-CANISTER OVERPACKS

1.0 INTRODUCTION

Spent nuclear fuel (SNF) has been stored at Hanford in the K Basin pools for up to 30 years. The SNF will be moved from the K Basins to a dry, interim storage facility, the Canister Storage Building.

Spent fuel has been irradiated and contains many fission products. Some of these fission products are volatile and are potential environmental hazards. During the drying, stabilization, and storage of the fuel, the environmental risk must be understood and minimized.

As long as the fuel is intact and non-oxidized, the fission products are contained within the metal matrix. Release of the fission products through solid state diffusion is extremely low at ambient temperature.

Mechanisms for increasing the fission product release rates include heating the fuel and oxidation of the fuel. Heating the fuel increases the solid state diffusion rate. A 50-100 micron layer of oxide on the uranium metal surface is formed upon contact with either water or oxygen. Diffusion of fission products through uranium oxide is much faster than diffusion through the metal, therefore, oxidation is the key mechanism for fission product release.

This page intentionally left blank.

2.0 EMPIRICAL MODELS

It must be clearly stated that no data is available for the release of fission products from either uranium metal or oxide in the temperature range of ambient to 300 °C (the range of interest). Data does exist for the release of fission products from uranium dioxide in the temperature range 1000-2200 °C from the Light Water Reactor (LWR) severe accident studies (Owczarski 1996). Two empirical models have been formulated for this higher temperature range.

The empirical models initially used in the LWR severe accident studies are based on the definition of a fractional release of the radionuclide r per unit time, k_r , time⁻¹. This k_r is defined as the fraction of r released to the atmosphere per unit of time from the mass of r , m_r , in a given amount of UO_2 .

$$dm_r/dt = -k_r m_r \quad (1)$$

This rate constant is a function of temperature alone. Equation (1) was applied to all positions in the LWR during a severe accident scenario (loss of coolant, heat up, and melting), covering a large range of temperatures (NRC 1981). The equation(s) used for calculating the K_s is an empirical fit of data of the form:

$$k_r = A_r \exp(B_r T_c) \quad (2)$$

where T_c is the oxide temperature in degrees Celsius.

Values used for the A_s and B_s are found in Table 1. They are taken from NRC 1981 (see next paragraph) for the low temperature range 1000-2200 °C. Because they are a function of temperature alone, they are independent of exposed surface area and degree of cladding failure. Using this approach in an MCO only requires that the quantities of UO_2 present and the temperature histories of each portion need to be tracked in an MCO model.

Since a number of radionuclides are not specifically identified in NRC 1981, but Table 1 is assigned values of the A_s and B_s for similar elements. All of the transuranic elements were assigned fuel U values. Iron, nickel, and cobalt were assigned the structure (steel) values.

Table 1. Parameters for Radionuclide Loss from UO_2 Matrix (2 Sheets).

Isotope	$k=A\exp(B/T_c)$		$K'=k_0\exp(Q/RT_c)$	
	A, 1/min	$B, 1/T_c$	$K_0, 1/\text{min}$	Q, kJ/mol.
Pu-241	1.4E-14	6.77E-03		
Am-241	1.4E-14	6.77E-03		
Pu-239	1.4E-14	6.77E-03		
Pu-238	1.4E-14	6.77E-03		
Pu-240	1.4E-14	6.77E-03		
Sr/Y-90	7.28E-11	6.77E-03	1.20E+00	200
Cm-244	1.40E-14	6.77E-03		
Ru/Rh m-106	1.38E-08	7.68E-03	1.90E+09	710
Cs/Ba m-137	1.65E-07	6.77E-03	8.50E+05	292
Pm-147			1.90E+09	710
Ce/Pr-144	1.6E-12	6.77E-03	5.20E+11	852
Eu-154	1.00E-08	6.77E-03		
U-234	1.4E-14	6.77E-03		
Cs-134	1.65E-07	6.77E-03	8.50E+05	292
Pu-242	1.4E-14	6.77E-03		
Sb/Te m-125			2.62E+02	207
Cd-113m			1.90E+09	710
Sm-151			5.20E+11	852
Eu-155	1.00E-08	6.77E-03		
Co-60	3.20E-11	5.76E-03		
Eu-152	1.00E-08	6.77E-03		
I-129			1.90E+07	335
Kr-85			3.20E+04	335
Fe-55	3.20E-11	5.76E-03		

Table 1. Parameters for Radionuclide Loss from UO₂ Matrix (2 Sheets).

Isotope	$k=A\exp(B/T_c)$		$K'=k_0\exp(Q/RT_c)$	
	A,1/min	B,1/T _c	K ₀ ,1/min	Q,kJ/mol.
Ni-63	3.20E-11	5.76E-03		
Tc-99			1.90E+09	710
Se-79			1.20E+00	200
H-3				
C-14				

Kelly et al. (1984) introduced the Arrhenius form of temperature dependence as being "more reasonable" and a better fit with the experimental data. In this form

$$k_r = k_{r0}\exp(-Q_r/RT) \quad (3)$$

where R is the universal gas constant and T the absolute temperature in Kelvin degrees.

Table 1 also contains the applicable values of the k_{r0} s and Q_r s that were published. Figure 1 displays the k_s calculated from Equations 2 and 3 for strontium/yttrium-90.

Figure 1 is typical in form for the k_s calculated for the other fission products also. Over the experimentally calibrated range of 1000 to 2200 °C, these two functions are very similar. However, as noted above, Kelly states that the Arrhenius form is more reasonable and fits the experimental data more closely.

It is unfortunate that the process temperatures proposed for the spent fuel lies in the uncalibrated range below 1000 °C. These two functions diverge widely below 1000 °C, and one must choose which to use. Since the Arrhenius function appears superior in scientific basis and experimental fit, it will be the function used to estimate the release rate for volatile fission products from the MCO.

Since all of the pre-exponential factors and activation energies were not published for each of the isotopes in Table 1, values were estimated based upon similar ionic charge to radius ratios and are presented in Table 2.

Figure 1. Diffusion Rate Constants for Strontium/Yttrium-90.

Table 2. Arrhenius Parameters (2 Sheets).

Isotope	$K' = k_{\text{exp}}(Q/RT_0)$	
	$K_0, \text{l/min}$	$Q, \text{kJ/mol.}$
Pu-241	1.90E+09	710
Am-241	1.90E+09	710
Pu-239	1.90E+09	710
Pu-238	1.90E+09	710
Pu-240	1.90E+09	710
Sr/Y-90	1.20E+00	200
Cm-244	1.90E+09	710
Ru/Rh m-106	1.90E+09	710
Cs/Ba m-137	8.50E+05	292
Pm-147	1.90E+09	710
Ce/Pr-144	5.20E+11	852
Eu-154	1.90E+09	710
U-234	1.90E+09	710
Cs-134	8.50E+05	292
Pu-242	1.90E+09	710
Sb/Te m-125	2.62E+02	207
Cd-113m	1.90E+09	710
Sm-151	5.20E+11	852
Eu-155	1.90E+09	710
Co-60	1.90E+09	710
Eu-152	1.90E+09	710
I-129	1.90E+07	335
Kr-85	3.20E+04	242
Fe-55	1.90E+09	710
Ni-63	1.90E+09	710
Tc-99	1.90E+09	710
Se-79	1.20E+00	200

Table 2. Arrhenius Parameters (2 Sheets).

Isotope	$K' = k_0 \exp(Q/RT_0)$	
	$K_0, 1/\text{min}$	$Q, \text{kJ/mol.}$
H-3 ¹		
C-14 ²	3.20E+04	242

- ¹ Monatomic tritium forms tritiated water upon reaction with oxygen. Due to the rapid hydrogen isotope exchange occurring upon water and hydroxide ions, it is postulated that the release of tritium to the atmosphere is essentially instantaneous. Although proof of this statement is not included, it is a conservative assumption.
- ² The rate of diffusion of carbon-14 through uranium dioxide is not known or measured. It will probably form carbon monoxide or carbon dioxide in the presence of oxygen or elemental carbon particles in an anoxic atmosphere. To remain conservative, it is assigned the same diffusion rate as the noble gas krypton-85.

3.0 AIRBORNE RELEASE RATES

If m_{ro} is defined as the growth rate of isotope r within the oxide due to oxidation of metal, and $k_r m_r$ is the airborne source term rate, the change in isotope r with respect to time can be given as:

$$dm_r/dt = m_{ro} - k_r m_r \quad (4)$$

This can be rearranged algebraically to give

$$k_r m_r dt = -dm_r + m_{ro} dt \quad (5)$$

and finally:

$$\int k_r m_r dt = A_{st} = \int m_{ro} dt + \int -dm_r$$

allowing time to vary between 0 and time t , and the mass of a particular fission product to vary over that same time interval:

$$A_{st} = m_{ro} * (t_2 - t_1) - (m_{r2} - m_{r1}) \quad (6)$$

Equation 6 can be solved if the change in m_r can be calculated. If Equation 4 is rearranged algebraically:

$$dm_r / (m_{ro} - k_r m_r) = dt \quad (7)$$

the integrated solution is:

$$(m_{ro} - k_r m_{r2}) / (m_{ro} - k_r m_{r1}) = e^{-k_r(t_2 - t_1)}$$

and after appropriate algebraic manipulation:

$$m_{r2} = m_{r1} e^{-k_r(t_2 - t_1)} + (m_{ro}/k_r)(1 - e^{-k_r(t_2 - t_1)}) \quad (8)$$

by substitution of Equation 8 into Equation 6:

$$\begin{aligned} A_{st} &= m_{ro} * (t_2 - t_1) + (m_{r1} - m_{r1} e^{-k_r(t_2 - t_1)}) - (m_{ro}/k_r)(1 - e^{-k_r(t_2 - t_1)}) \\ &= m_{ro} * (t_2 - t_1) + m_{r1} (1 - e^{-k_r(t_2 - t_1)}) - (m_{ro}/k_r)(1 - e^{-k_r(t_2 - t_1)}) \\ &= m_{ro} * (t_2 - t_1) + (m_{r1} - (m_{ro}/k_r))(1 - e^{-k_r(t_2 - t_1)}) \end{aligned} \quad (9)$$

since for most MCO applications, $k_t t \ll 1$, and the spreadsheet function of $\exp(-k_t t)$ is inaccurate.

Equation 9 can be expanded for small x as $\exp(-x) = 1 - x + X^2/2!$, which gives:

$$A_{st} = m_p k_t t * (1 - k_t t/2) + m_{ro} k_t t^2/2 \quad (10)$$

Equation 10 is used for spreadsheet calculations if $k_t t < 0.01$. Otherwise, Equation 9 is used.

4.2 CASE 2

Table 4 gives the integrated release of fission products in Bq/cm² for simulated hot vacuum drying conditions (300 °C and at 5 torr water pressure).

Table 4. Fission Product Release During Hot Vacuum Drying.

Isotope	Ci/MTU	koko*Exp(-Q/RT)		Input Temp 300 °C					Mro	Mr	Integrated Doses Bq/cm ²			U metal
		Bq/kgU	kg,1/min	Q,kJ/mol	k,1/min	k,1/s						3000	14400	
Pu-241	9450	3.4865E+11	1.90E+09		710	3.72E-56	2.23E-54	2267.342086	16899908	1.68949E-43	1.07E-42	3.20589E-42		
Am-241	142	5254000000	1.90E+09		710	3.72E-56	2.23E-54	34.37064236	253795.443	2.5387E-45	1.61E-44	4.61731E-44		
Pu-239	139	5143000000	1.90E+09		710	3.72E-56	2.23E-54	33.64450263	248433.567	2.48505E-45	1.58E-44	4.71554E-44		
Pu-238	116	4292000000	1.90E+09		710	3.72E-56	2.23E-54	28.07742666	207325.855	2.07397E-45	1.32E-44	3.93527E-44		
Pu-240	95.2	3522400000	1.90E+09		710	3.72E-56	2.23E-54	23.0426536	170150.184	1.702E-45	1.08E-44	3.22963E-44		
Sr-90	8480	3.1376E+11	1.2		200	7.11E-19	4.26E-17	2052.556708	15156234.9	2.89342E-06	1.84E-05	5.49041E-05		
Cm-244	26.8	9916000000	1.90E+09		710	3.72E-56	2.23E-54	6.486853745	47899.4216	4.79134E-46	3.04E-45	9.09182E-45		
Ru/Rh m-106	2430	8.991E+10	1900000000		710	3.72E-56	2.23E-54	588.1736792	4343118.2	4.34439E-44	2.76E-43	8.24371E-43		
Cs/Ba m-137	10600	3.922E+11	850000		292	2.08E-21	1.25E-19	2565.695984	18945293.6	1.05653E-08	6.71E-08	2.00483E-07		
Pm-147	4440	1.8428E+11	1900000000		710	3.72E-56	2.23E-54	1074.68771	7935575.82	7.9379E-44	5.04E-43	1.50626E-42		
Ce/Pr-144	303	1.1211E+10	5.2E+11		852	1.17E-66	6.99E-65	73.34017481	541549.431	1.6961E-55	1.08E-54	3.21843E-54		
Eu-154	225	8325000000	1.90E+09		710	3.72E-56	2.23E-54	54.46052585	402140.667	4.02258E-45	2.56E-44	7.63306E-44		
U-234	0.395	14615000	1.90E+09		710	3.72E-56	2.23E-54	0.085608479	705.980281	7.06187E-48	4.49E-47	1.34003E-46		
Cs-134	567	2.0979E+10	850000		292	2.08E-21	1.25E-19	137.2405251	1013394.48	5.65146E-10	3.59E-09	1.07239E-08		
Pu-242	0.0566	2094200	1.90E+09		710	3.72E-56	2.23E-54	0.013699848	101.160719	1.0119E-48	6.43E-48	1.92014E-47		
Sb/Te m-125	345.8	1.2795E+10	262		207	3.57E-17	2.14E-15	83.70239378	618064.844	5.92938E-06	3.77E-05	0.000112513		
Cd-113m	4.04	149480000	1900000000		710	3.72E-56	2.23E-54	0.977868997	7220.65908	7.22277E-47	4.59E-46	1.37056E-45		
Sm-151	101	3737000000	5.2E+11		852	1.17E-66	6.99E-65	24.44672494	180516.477	5.65366E-56	3.59E-55	1.07281E-54		
Eu-155	65.4	2419800000	1.90E+09		710	3.72E-56	2.23E-54	15.82985951	116888.887	1.16923E-45	7.43E-45	2.21868E-44		
Co-60	9.74	360380000	1.90E+09		710	3.72E-56	2.23E-54	2.35753652	17408.2226	1.74133E-46	1.11E-45	3.30427E-45		
Eu-152	1.21	44770000	1.90E+09		710	3.72E-56	2.23E-54	0.292876606	2162.62314	2.16326E-47	1.37E-46	4.10496E-46		
I-129	1	37000000	1.90E+07		335	5.99E-24	3.36E-22	0.242046782	1787.29185	2.68527E-15	1.71E-14	5.09544E-14		
Kr-85	772	2.8564E+10	32000		242	2.82E-18	1.69E-16	186.8601154	1379789.31	1.04427E-06	6.63E-06	1.98156E-05		
Fe-55	21.8	8066000000	1.90E+09		710	3.72E-56	2.23E-54	5.276619838	38962.9624	3.89744E-46	2.48E-45	7.39559E-45		
Ni-63	3.11	115070000	1.90E+09		710	3.72E-56	2.23E-54	0.752765491	5558.47766	5.5601E-47	3.53E-46	1.05506E-45		
Tc-99	1.72	63640000	1900000000		710	3.72E-56	2.23E-54	0.416320464	3074.14199	3.07504E-47	1.95E-46	5.83505E-46		
Se-79	1	37000000	1.2		200	7.11E-19	4.26E-17	0.242046782	1787.29185	3.41205E-10	2.17E-09	6.47545E-09		
H-3	45.43	1680900000						10.99611987	39586.03152	158344.1	316688.2522			
C-14	0.443	16391000						0.107226724		386.0162072	1544.065	3088.129658		
HVD at 300 °C				Oxidation Rate										
Temp °K		mg O ₂ /cm ² /s : kg U/cm ² /sec												
50 °C		0.664473684 0.00087957 6.542E-09												

4.3 CASE 3

Table 5 gives the integrated release of fission products in Bq/cm² for simulated cold vacuum drying conditions (50 °C and at 5 torr water pressure).

Table 5. Fission Product Release During Cold Vacuum Drying.

Isotope	G/MTU	kmo*Exp(-Q/RT)		Input T=50 °C		X		Integrated Release Bq/Cm ² U metal			
		Bq/kgU	kg,1/min	O ₂ ,l/mol	k,1/min	kg	l/r	3600	14400	28800	
						Bq/cm ² /sec	Bq/cm ²				
Pu-241	9450	3.497E+11	1.90E+09	710	3.25E-106	1.95E-104	29.2087141	16889908	1.15E-93	4.8001E-93	9.73045E-93
Am-241	142	5.254E+08	1.90E+09	710	3.25E-106	1.95E-104	0.43890343	253795.44	1.789E-95	7.2219E-95	1.46214E-94
Pu-239	139	5.143E+08	1.90E+09	710	3.25E-106	1.95E-104	0.42963062	248433.57	1.751E-95	7.0603E-95	1.43125E-94
Pu-238	116	4.282E+09	1.90E+09	710	3.25E-106	1.95E-104	0.35854083	207325.85	1.461E-95	5.8996E-95	1.19443E-94
Pu-240	95.2	3.522E+09	1.90E+09	710	3.25E-106	1.95E-104	0.29425075	170150.18	1.199E-95	4.8417E-95	9.80253E-95
Sr/Y-90	8480	3.138E+11	1.2	200	5.63E-33	3.38E-31	26.2105709	15156235	1.849E-20	7.4658E-20	1.51152E-19
Cm-244	26.8	991600000	1.90E+09	710	3.25E-106	1.95E-104	0.08283529	47599.422	3.376E-96	1.363E-95	2.75953E-95
Ru/Rh m-106	2430	8.991E+10	1.9E+09	710	3.25E-106	1.95E-104	7.51081219	4343119.2	3.061E-94	1.2359E-93	2.50212E-93
Ca/Ba m-137	10600	3.922E+11	850000	292	5.37E-42	3.22E-40	32.7632137	18945294	2.203E-29	8.9935E-29	1.80059E-28
Pm-147	4440	1.643E+11	1.9E+09	710	3.25E-106	1.95E-104	13.724593	7935575.8	5.593E-94	2.2581E-93	4.57177E-93
Ca/P-144	303	1.121E+10	5.2E+11	852	9.91E-127	5.95E-125	0.93653337	541549.43	1.16E-115	4.696E-115	9.5082E-115
Eu-154	225	8.325E+09	1.90E+09	710	3.25E-106	1.95E-104	0.69544557	402140.67	2.834E-95	1.1443E-94	2.31677E-94
U-234	0.395	1.4615000	1.90E+09	710	3.25E-106	1.95E-104	0.00122089	705.98028	4.976E-98	2.0089E-97	4.05722E-97
Cs-134	567	2.098E+10	850000	292	5.37E-42	3.22E-40	1.75252285	1013394.5	1.178E-30	4.7572E-30	9.63143E-30
Pu-242	0.0566	2094200	1.90E+09	710	3.25E-106	1.95E-104	0.00017494	101.16072	7.13E-99	2.8786E-98	5.82979E-98
Sr/Te m-125	345.8	1.28E+10	262	207	9.09E-32	5.45E-30	1.06865599	618064.84	1.216E-20	4.9105E-20	9.94207E-20
Co-113m	4.04	14948000	1.9E+09	710	3.25E-106	1.95E-104	0.01249111	7220.6591	5.089E-97	2.0547E-96	4.1599E-96
Sm-151	101	3.737E+09	5.2E+11	852	9.91E-127	5.95E-125	0.31217778	180516.48	3.88E-116	1.565E-115	3.1694E-115
Eu-155	65.4	2.42E+09	1.90E+09	710	3.25E-106	1.95E-104	0.20214285	116680.89	8.239E-96	3.3261E-95	6.73406E-95
Co-60	9.74	360380000	1.90E+09	710	3.25E-106	1.95E-104	0.03010507	17408.223	1.227E-96	4.9536E-96	1.00291E-95
Eu-152	1.21	44770000	1.90E+09	710	3.25E-106	1.95E-104	0.00373995	2162.6231	1.524E-97	6.1539E-97	1.24591E-96
I-129	1	37000000	1.90E+07	335	1.34E-47	8.06E-46	0.00309087	1787.2919	5.204E-39	2.1012E-38	4.25399E-38
Kr-85	772	2.855E+10	32000	242	2.44E-35	1.46E-33	2.38615103	1379789.3	7.298E-24	2.9468E-23	5.96489E-23
Fe-55	3.178	809600000	1.90E+09	710	3.25E-106	1.95E-104	0.06738995	38862.862	2.746E-96	1.1087E-95	2.2447E-95
Ni-63	3.11	115070000	1.90E+09	710	3.25E-106	1.95E-104	0.0096126	5558.4777	3.918E-97	1.5817E-96	3.2023E-96
Tc-99	1.72	63640000	1.9E+09	710	3.25E-106	1.95E-104	0.0053163	3074.142	2.167E-97	8.7476E-97	1.77104E-96
Se-79	1	37000000	1.2	200	5.63E-33	3.38E-31	0.00309087	1787.2919	2.181E-24	8.804E-24	1.77246E-23
H-3	45.43	1.681E+09					0.14041735	505.50247	2022.00988	4044.018769	
C-14	0.443	16391000	32000	242	2.44E-35	1.46E-33	0.00136826	791.77029	4.189E-27	1.6906E-26	3.42286E-26
Oxidation Rate											
Temp °C	Temp °K	H ₂ O-KPa		mg O ₂ /cm ² /sec		kg U/cm ² /sec					
50	323.16	0.6644737		1.12319E-05		8.3537E-11					

4.4 CASE 4

Table 6 gives the integrated release of fission products in Bq/cm² for simulated hot conditioning conditions (150 °C and at 2% oxygen in an argon atmosphere).

Table 6. Fission Product Release During Hot Conditioning.

Isotope	CUMTU	k=10 ⁻⁶ Exp(-Q/RT)		Input Temp=150 °C			M _g Bq/cm ² /sec	M _r Bq/cm ²	Integrated Dose Bq no sudge		
		Bq/kgU	kg./l/min	Q.kJ/mol	k ₁ /min	k ₁ /h			3500	14400	28800
Pu-241	9450	3.4965E+11	1.9E+09	710	4.30E-79	2.58E-77	251532.811	16889908	1.5691E-66	6.2765E-66	1.2553E-65
Am-241	142	5254000000	1.9E+09	710	4.30E-79	2.58E-77	3779.64648	253795.44	2.3578E-68	9.4313E-68	1.8862E-67
Pu-239	139	5143000000	1.9E+09	710	4.30E-79	2.58E-77	3689.79479	248433.57	2.308E-68	9.2321E-68	1.84641E-67
Pu-238	118	4292000000	1.9E+09	710	4.30E-79	2.58E-77	3087.59853	207325.85	1.9261E-68	7.7045E-68	1.54089E-67
Pu-240	95.2	3522400000	1.9E+09	710	4.30E-79	2.58E-77	2533.99017	170150.18	1.5807E-68	6.323E-68	1.26459E-67
SrY-90	8480	3.1378E+11	1.2	200	2.48E-25	1.47E-23	225714.1	15156235	8.0435E-13	3.2174E-12	6.43478E-12
Cm-244	28.8	991600000	1.9E+09	710	4.30E-79	2.58E-77	713.34173	47899.422	4.45E-69	1.78E-68	3.55999E-68
RuRh m-106	2430	8981000000	1.9E+09	710	4.30E-79	2.58E-77	64679.8658	4343119.2	4.0349E-67	1.614E-66	3.2279E-66
CaBa m-137	10600	3.922E+11	850000	292	7.65E-31	4.59E-29	282142.624	18945294	3.1315E-18	1.2526E-17	2.50519E-17
Pm-147	4440	1.6428E+11	1.9E+09	710	4.30E-79	2.58E-77	118180.498	7935575.8	7.3724E-67	2.9489E-66	5.89788E-66
CePr-144	303	11211000000	5.2E+11	852	3.48E-94	2.09E-92	8065.0203	541549.43	4.0727E-63	1.6291E-62	3.25815E-62
Eu-154	225	8325000000	1.9E+09	710	4.30E-79	2.58E-77	5988.87646	402140.67	3.736E-68	1.4944E-67	2.9888E-67
U-234	0.395	14615000	1.9E+09	710	4.30E-79	2.58E-77	10.5138053	705.98028	6.5988E-71	2.6235E-70	5.24701E-70
Ca-134	567	20979000000	850000	292	7.65E-31	4.59E-29	15091.9687	1013394.5	1.6751E-19	6.7002E-19	1.34004E-18
Pu-242	0.0566	2094200	1.9E+09	710	4.30E-79	2.58E-77	1.50653515	101.16072	9.3981E-72	3.7593E-71	7.51861E-71
SbTe m-125	345.8	12795000000	262	207	7.34E-24	4.40E-22	9204.52544	618064.84	9.7926E-13	3.917E-12	7.8341E-12
Cd-113m	4.04	149480000	1.9E+09	710	4.30E-79	2.58E-77	107.533604	7220.6591	6.7082E-70	2.6833E-69	5.36655E-69
Sm-151	101	3737000000	5.2E+11	852	3.48E-94	2.09E-92	2688.3401	180516.48	1.3576E-63	5.4302E-63	1.08005E-62
Eu-155	65.4	2418800000	1.9E+09	710	4.30E-79	2.58E-77	1740.78076	116888.88	1.0859E-68	4.3437E-68	8.68744E-68
Co-60	9.74	360380000	1.9E+09	710	4.30E-79	2.58E-77	259.251808	17408.223	1.6173E-69	6.4691E-69	1.29382E-68
Eu-152	1.21	44770000	1.9E+09	710	4.30E-79	2.58E-77	32.2068468	2162.6231	2.0091E-70	8.0365E-70	1.60731E-69
Kr-85	1	37000000	19000000	335	8.41E-35	5.05E-33	26.6172287	1787.2919	3.2486E-26	1.2994E-25	2.59886E-25
Kr-85	772	28564000000	32000	242	4.28E-26	2.57E-24	20548.5006	1379789.3	1.2784E-14	5.1056E-14	1.02113E-13
Fe-55	21.8	8066000000	1.9E+09	710	4.30E-79	2.58E-77	580.255586	38662.962	3.6198E-69	1.4479E-68	2.89581E-68
Ni-63	3.11	115070000	1.9E+09	710	4.30E-79	2.58E-77	82.7795813	5558.4777	5.164E-70	2.0656E-69	4.13118E-69
Tc-99	1.72	63640000	1.9E+09	710	4.30E-79	2.58E-77	45.7816334	3074.142	2.856E-70	1.1424E-69	2.28477E-69
Se-79	1	37000000	1.2	200	2.48E-25	1.47E-23	26.6172287	1787.2919	9.4852E-17	3.7941E-16	7.58819E-16
H-3	45.43	1689000000					196.680983		708051.539	2832206.15	5664412.31
C-14	0.443	16391000	32000	242	4.28E-26	2.57E-24	11.7914323	791.77029	7.3245E-18	2.9298E-17	5.85958E-17
Oxidation Rate											
Temp °C	Temp °K	mg O ₂ /sec. kg U/sec(air)		U/sec(2%O ₂)							
150	423.16	0.096724		7.1938E-07							

4.5 CASE 5

Table 7 gives the integrated release of fission products in Bq/cm² for simulated long-term storage conditions (50 °C and at 0 torr water and oxygen pressures).

Table 7. Fission Product Release During Long Term Storage.

ln-ko*Exp(-Q/RT)									
Isotope	C/MTU	Bq/kgU	ko,1/min	Q,kJ/mol	k,1/min	K,1/s	Mro	Mr	Integrated Activity Bq/cm ² over 75 years
							Bq/cm ² /sec	Bq/cm ²	2344820000
Pu-241	9450	3.4965E+11	1.90E+09	710	7.86E-70	4.71E-68	0	16889908	1.8453E-51
Am-241	142	5254000000	1.90E+09	710	7.86E-70	4.71E-68	0	253795.443	2.83178E-53
Pu-239	139	5143000000	1.90E+09	710	7.86E-70	4.71E-68	0	248433.567	2.77196E-53
Pu-238	116	4292000000	1.90E+09	710	7.86E-70	4.71E-68	0	207325.855	2.31329E-53
Pu-240	95.2	3522400000	1.90E+09	710	7.86E-70	4.71E-68	0	170150.184	1.89849E-53
StrY-90	8480	3.1376E+11	1.2	200	9.98E-23	5.99E-21	0	15156234.9	0.000214898
Cm-244	26.8	991600000	1.90E+09	710	7.86E-70	4.71E-68	0	47899.4216	5.3445E-54
Ru/Rh m-106	2430	8.991E+10	1900000000	710	7.86E-70	4.71E-68	0	4343119.2	4.84594E-52
Cs/Ba m-137	10600	3.922E+11	850000	292	4.93E-27	2.96E-25	0	18945293.6	1.32631E-08
Pm-147	4440	1.6428E+11	1900000000	710	7.86E-70	4.71E-68	0	7938575.82	8.85431E-52
Ce/Pr-144	303	1.1211E+10	5.2E+11	852	4.53E-83	2.72E-81	0	541549.431	4.48898E-53
Eu-154	225	8325000000	1.90E+09	710	7.86E-70	4.71E-68	0	402140.667	4.48898E-53
U-234	0.395	1.4615000	1.90E+09	710	7.86E-70	4.71E-68	0	705.980281	7.87715E-56
Cs-134	567	2.0979E+10	850000	292	4.93E-27	2.96E-25	0	1013394.48	7.09451E-10
Pu-242	0.0566	2094200	1.90E+09	710	7.86E-70	4.71E-68	0	101.160719	1.12873E-56
Sb/Te m-125	345.8	1.2795E+10	262	207	3.68E-21	2.21E-19	0	618064.844	0.000322849
Cd-113m	4.04	149480000	1900000000	710	7.86E-70	4.71E-68	0	7220.65908	8.05663E-55
Sm-151	101	3737000000	5.2E+11	852	4.53E-83	2.72E-81	0	180516.477	1.1605E-66
Eu-155	65.4	2419800000	1.90E+09	710	7.86E-70	4.71E-68	0	116888.887	1.30422E-53
Co-60	9.74	360380000	1.90E+09	710	7.86E-70	4.7142E-68	0	17408.2226	1.94237E-54
Eu-152	1.21	44770000	1.90E+09	710	7.86E-70	4.71E-68	0	2162.62314	2.413E-55
P-129	1	37000000	1.90E+07	335	1.97E-30	1.18E-28	0	1787.29185	5.00616E-16
Kr-85	772	2.8564E+10	32000	242	6.15E-23	3.69E-21	0	1379789.31	1.20407E-05
Fe-55	21.8	806600000	1.90E+09	710	7.86E-70	4.7142E-68	0	38952.9624	4.34739E-54
Ni-63	3.11	115070000	1.90E+09	710	7.86E-70	4.7142E-68	0	5553.47766	6.20201E-55
Tc-99	1.72	63640000	1900000000	710	7.86E-70	4.71E-68	0	3074.14189	3.43005E-55
Se-79	1	37000000	1.2	200	9.98E-23	5.99E-21	0	1787.29185	2.53417E-08
H-3	45.43	1680900000					0		0
C-14	0.443	16391000	32000	242	6.15E-23	3.69E-21	0	791.77029	6.90837E-09
Oxidation Rate									
Temp °C	Temp °K	H ₂ O-kPa	g O ₂ /cm ² /sec		kg U/cm ² /sec				
200	473.16	0.001	unknown	unknown	assumed 0	for calculational purposes			

This page intentionally left blank.

5.0 DISCUSSION OF RESULTS

From Table 3, at 600 °C, a significant number of fission product isotopes are released within 8 hours. These include Strontium-90, Cesium-137, Cesium-134, Antimony-125, Iodine-125, Krypton-85, Selenium-79, Tritium-3, and Carbon-14. Due to the low diffusion rate constants at or below 300 °C, all significant amounts of the fission products are retained within the uranium oxide, except for tritium.

The tritium release rate is directly related to the oxidation rate of the uranium metal. The highest release rate is observed for Case 1 involving the 600 °C uranium oxidation.

The radiological dose delivered to site workers and the general public from fission product release should be evaluated. The radiological dose from internal absorption and from submersion in a gaseous atmosphere is given in Table 8.

To build an entire dose model, in addition to the data in Table 8, other parameters must be known or postulated. Examples of such parameters are: the oxidation rate of the metal (controlled by temperature, surface area and reactive gases), the release fraction (i.e., the ratio of the amount of isotope that goes airborne versus the amount that is retained on the oxide or other surfaces), the concentration of the isotopes in the atmosphere at given locations, and the breathing rate of exposed personnel.

Factors affecting the isotope concentrations are: the dilution rate due to mixing with non-contaminated gases, the distance from the source, and the diffusion rates of gases and dust particles bearing the isotopes.

The dose model is, therefore, site-specific and cannot be postulated generally. Due to the lack of site-specific information at this time; such as, size of rooms or buildings, ventilation exchange rates, site wind velocities, and diffusion rates of gases and dust particles, a site-specific dose model will be postponed.

Table 8. Internal and External Radiation Dose Estimates (2 Sheets).

Isotope	Effective Dose Internal Sv/Bq	Conversion Factors Submersion Sv-m ³ /Bq-hr
Pu-241	2.23E-06	0.00E+00
Am-241	1.20E-04	0.00E+00
Pu-239	1.16E-04	0.00E+00
Pu-238	1.06E-04	0.00E+00
Pu-240	1.16E-04	0.00E+00

Table 8. Internal and External Radiation Dose Estimates (2 Sheets).

Isotope	Effective Dose Internal Sv/Bq	Conversion Factors Submersion Sv-m ³ /Bq-hr
Sr/Y-90	3.53E-07	0.00E+00
Cm-244	6.70E-05	0.00E+00
Ru/Rh m-106	1.29E-07	0.00E+00
Cs/Ba m-137	8.63E-09	0.00E+00
Pm-147	1.06E-08	0.00E+00
Ce/Pr-144	1.01E-07	0.00E+00
Eu-154	7.73E-08	0.00E+00
U-234	3.58E-05	0.00E+00
Cs-134	1.25E-08	0.00E+00
Pu-242	1.11E-04	0.00E+00
Sb/Te m-125	5.27E-09	0.00E+00
Cd-113m	4.13E-07	0.00E+00
Sm-151	8.10E-09	0.00E+00
Eu-155	1.12E-08	0.00E+00
Co-60	5.91E-08	0.00E+00
Eu-152	5.97E-08	0.00E+00
I-129	4.69E-08	0.00E+00
Kr-85	0.00E+00	4.66E-11
Fe-55	7.26E-10	0.00E+00
Ni-63	1.70E-09	0.00E+00
Tc-99	2.25E-09	0.00E+00
Se-79	2.66E-09	0.00E+00
H-3	1.73E-11	1.19E-15
C-14	5.64E-10	0.00E+00

6.0 REFERENCES

- Kelly, J. L., 1984, *Temperature Dependence of Fission Product Release Rates*, Reynolds, A.B, McGown, M.E., Nuclear Science and Engineering.
- NRC, 1981, *Technical Basis for Estimating Fission Product Behavior During LWR Accidents*, NUREG-0772, U. S. Nuclear Regulatory Commission, Washington, D.C.
- Owczarski, P.C., 1996, *Initial Models for Airborne Radionuclide Releases in an MCO*, Informal Report, Science Applications International Corporation, Richland, Washington.

This page intentionally left blank.

DISTRIBUTION SHEET

To Distribution	From Plutonium Process Support Laboratory/Thurman Cooper	Page 1 of 1
		Date 07/30/96
Project Title/Work Order		EDT No. 616633
SNFP Estimate of Volatile Fission Products Release From Multi-Canister Overpacks, WHC-SD-SNF-TI-033.		ECN No. N/A

Name	MSIN	Text With All Attach.	Text Only	Attach./ Appendix Only	EDT/ECN Only
J Abrefah	P7-34	X			
GS Barney	T5-12	X			
GD Bazinet	B4-55	X			
LD Blackburn	R1-56	X			
TD Cooper	T5-12	X			
RG Cowan	R3-86	X			
ER Cramer	H0-34	X			
DR Duncan	R3-86	X			
FD Fisher	T5-12	X			
JR Frederickson	R3-86	X			
FJ Heard	H0-34	X			
JJ Irwin	H0-34	X			
A Johnson	K8-34	X			
AT Kee	R3-86	X			
CR Miska	R3-86	X			
SD Nunn	S5-11	X			
DM Ogden	H0-34	X			
PA Scott	R3-87	X			
PK Shen	H0-40	X			
DA Smith	R3-11	X			
TA Thornton	P7-18	X			
JC Wiborg	R3-86	X			
AM Zaman	B1-10	X			
Central Files (orig + 1)	A3-88	X			
SNF Project files	R3-11	X			