AUG 1 1996

ENGINEERING DATA TRANSMITTAL

Page 1 of 1

1. EDT 0000

2. To: (Receiving Organization) Distribution	3. From: (Originating Organization) Plutonium Process Support Laboratory	4. Related EDT No.: N/A
5. Proj./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

15. 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-028		0.00	Spent Nuclear Fuel Project (SNFP) Gas Generation From N-Fuel in Multi-Canister Overpacks	S	1-2	1-2	

16										KEY	
Approval Designator (F)		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			1. Approved	4. Reviewed no/comment				
		2. Release	5. Post-Review			2. Approved w/comment	5. Reviewed w/comment				
		3. Information	6. Dist. (Receipt Acknow. Required)			3. Disapproved w/comment	6. Receipt acknowledged				
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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>	7/24/96	T5-12	DR Duncan	<i>DR Duncan</i>	7/24/96	T3-86	1-2	1
1-2	1	Cog. Mgr. PK Shen	<i>PK Shen</i>	7/24/96	H0-40						
		QA									
1-2		Safety SE Nunn	<i>SE Nunn</i>	7/24/96	H0-11						
		Env.									
1-2	1	PA Scott	<i>PA Scott</i>	7/24/96	R3-87						
1-2	1	AM Zaman	<i>AM Zaman</i>	8/1/96	B1-10						

18. Signature of EDT Originator T. D. Cooper Date: 7-24-96	19. Authorized Representative for Receiving Organization <i>PA Scott</i> Date: 7/24/96	20. Cognizant Manager PK Shen Date: 7-24-96	21. DOE APPROVAL (if required) Ctrl. No. - N/A ☐ Approved ☐ Approved w/comments ☐ Disapproved w/comments
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BD-7400-172-2 (04/94) GEF097

BD-7400-172-1

Spent Nuclear Fuel Project (SNFP)

Gas Generation From N-Fuel in Multi-Canister Overpacks

Thurman D. Cooper

Westinghouse Hanford Company, Richland, WA 99352
U.S. Department of Energy Contract DE-AC06-87RL10930

EDT/ECN: 616631

Org Code: 15500

B&R Code: EW3135040

UC: 510

Charge Code: LB040

Total Pages: 75 77 *Aug 1-96*

Key Words: Gas Generation, MCO, N-Fuel

Abstract:

During the conversion from wet pool storage for spent nuclear fuel at Hanford, gases will be generated from both radiolysis and chemical reactions. The gas generation phenomenon needs to be understood as it applies to safety and design issues, specifically over pressurization of sealed storage containers, and detonation/deflagration of flammable gases. This study provides an initial basis to predict the implications of gas generation on the proposed functional processes for spent nuclear fuel conversion from wet to dry storage. These projections are based upon examination of the history of fuel manufacture at Hanford, irradiation in the reactors, corrosion during wet pool storage, available fuel characterization data and available information from literature. Gas generation via radiolysis and metal corrosion are addressed. The study examines gas generation, the boundary conditions for low medium and high levels of sludge in SNF storage/processing containers. The functional areas examined include: flooded and drained Multi Canister Overpacks, cold vacuum drying, shipping and staging, and long term storage.

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SPENT NUCLEAR FUEL PROJECT GAS GENERATION FROM N-FUEL IN MULTI-CANISTER OVERPACKS

July 1996

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

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CONTENTS

1.0 INTRODUCTION	1-1
2.0 FLOODED MULTI-CANISTER OVERPACK CASE	2-1
2.1 UNCORRODED FUEL IN A FLOODED MCO	2-1
2.1.1 Calculation of Radiolytic Gas Generation Rates	2-5
2.2 CORRODED FUEL IN A FLOODED MCO	2-8
2.2.1 Radiolytic Gases	2-8
2.2.2 Chemically Produced Hydrogen	2-8
2.2.3 Hydrogen Generation from Reaction of Hydrides	2-10
2.3 FISSION PRODUCT GAS RELEASE IN A FLOODED MCO	2-15
2.4 HELIUM GAS RELEASE IN A FLOODED MCO	2-16
2.5 WATER VAPOR PRESSURE IN A FLOODED MCO	2-16
2.6 PRESSURIZATION OF A SEALED FLOODED MCO	2-16
2.7 SUMMARY FOR FLOODED CASE	2-24
3.0 DRAINED CASE	3-1
3.1 UNCORRODED FUEL IN A DRAINED MCO	3-1
3.2 CORRODED FUEL IN A DRAINED MCO	3-3
3.2.1 Hydrogen Generated by Water Reaction on Uranium Metal in a Drained MCO	3-3
3.2.2 Hydrogen Generated by Water Reaction on Uranium Hydride in a Drained MCO	3-3
3.2.3 Fission Product Gas Release in a Drained MCO	3-3
3.2.4 Helium Gas Release in a Drained MCO	3-3
3.2.5 Water Vapor Pressure in a Drained MCO	3-3
3.2.6 Pressurization of a Sealed Drained MCO	3-4
4.0 COLD VACUUM DRYING	4-1
4.1 RADIOLYTIC GAS PRODUCTION FOR UNCORRODED FUEL DURING CVD	4-1
4.2 GAS GENERATION FROM CORRODED FUEL DURING CVD	4-1
4.2.1 Hydrogen Generation from Water Reaction with Uranium Metal During CVD	4-1
4.2.2 Hydrogen Generation from Water Reaction with Uranium Hydride During CVD	4-2
4.2.3 Fission Product Gas Release During CVD	4-2
4.2.4 Helium Gas Release During CVD	4-4
4.2.5 Water Vapor Pressure Increase During CVD	4-4
4.2.6 Pressurization of a Sealed MCO During CVD	4-5

CONTENTS cont.

5.0 SHIPPING AND STAGING	5-1
5.1 RADIOLYTIC GAS PRODUCTION	5-1
5.2 CHEMICAL GAS GENERATION DURING SHIPPING AND STAGING	5-2
5.2.1 Estimate of Water Partial Pressure During Shipping	5-2
5.2.2 Hydrogen Generation From the Reaction of Water with Uranium Metal and Uranium Hydride	5-5
5.2.3 Fission Product Gas Release During Shipping and Staging	5-6
5.2.4 Helium Gas Release During Shipping and Staging	5-6
5.2.5 Water Vapor Pressure Increase During Shipping and Staging	5-6
5.2.6 Pressurization of a Sealed MCO During Shipping and Staging	5-6
5.2.7 Reduction of Inventory of Reactants	5-9
6.0 GAS GENERATION DURING HOT VACUUM DRYING AND CONDITIONING ..	6-1
7.0 GAS GENERATION DURING LONG-TERM STORAGE	7-1
7.1 RADIOLYTIC GAS GENERATION DURING LONG-TERM STORAGE	7-1
7.2 CHEMICAL GAS GENERATION	7-6
8.0 REFERENCES	8-1

LIST OF TABLES

2-1. High Sludge Case	2-2
2-2. Low Sludge Case	2-3
2-3. Radiolytic Gas Generation for a Flooded MCO	2-6
2-4. Hydrogen Production From Water on Uranium Metal in Flooded MCO	2-9
2-5. Hydrogen Generation from Uranium Hydride in Sludge in a Flooded MCO	2-14
2-6. Pressure Increase for a Flooded MCO With 2.7 kg Sludge	2-18
2-7. Pressure Increase for a Flooded MCO With 16 kg Sludge	2-20
2-8. Pressure Increase for a Flooded MCO With 277 kg Sludge	2-22
3-1. Radiolytic Gas Generation for Uncorroded Fuel in a Drained MCO	3-1
3-2. Pressure Increase for a Drained MCO With 2.7 kg Sludge	3-5
3-3. Pressure Increase for a Drained MCO With 16 kg Sludge	3-7
3-4. Pressure Increase for a Drained MCO With 277 kg Sludge	3-9
4-1. Hydrogen Generation from Water Reaction with Uranium Metal During CVD	4-3
5-1. Radiolytic Gas Generation During Shipping and Staging	5-1
5-2. Chemical Hydrogen Generation in a Sealed MCO During Shipping and Staging	5-7
5-3. Pressure Increase for a MCO During Shipping and Staging	5-10
5-4. Pressure Increase for a "Worst-Case" MCO Containing 16 kg Sludge During Shipping and Staging	5-12
5-5. Pressure Increase for a "Worst-Case" MCO Containing 277 kg Sludge During Shipping and Staging	5-14
7-1. Water Radiolyzed During Long-term Storage	7-4

LIST OF FIGURES

2-1. Gas Generation for Uncorroded Fuel in a Flooded MCO	2-7
2-2. Hydrogen from Corroded Metal in a Flooded MCO	2-10
2-3. Hydrogen Generation From Uranium Hydride in a Flooded MCO	2-15
2-4. Pressurization of Flooded-Sealed "Worst-Case" MCO With 2.7 kg Sludge	2-19
2-5. Pressurization of Flooded-Sealed "Worst-Case" MCO With 16 kg Sludge	2-21
2-6. Pressurization of Flooded MCO With 277 kg Sludge	2-23
3-1. Radiolytic Gas Generation for Uncorroded Fuel in a Drained MCO	3-2
3-2. Pressurization of a Drained-Sealed "Worst-Case" MCO 2.7 kg Sludge	3-6
3-3. Pressurization of a Drained-Sealed "Worst-Case" MCO 16 kg Sludge	3-8
3-4. Pressurization of a Drained-Sealed "Worst Cast" MCO 277 kg Sludge	3-10
4-1. Hydrogen From Water Reaction with Uranium Metal in MCO during CVD	4-4
5-1. Radiolytic Gas Generation During Shipping and Staging	5-2
5-2. Hydrogen Generation in the MCO During Shipping and Staging	5-8
5-3. Pressure Increase for a "Worst-Case" MCO Containing 2.7 kg of Sludge During Shipping and Staging	5-11
5-4. Pressure Increase for a "Worst-Case" MCO Containing 16 kg of Sludge During Shipping and Staging	5-13
5-5. Pressure for a "Worst-Case" MCO Containing 277 kg of Sludge During Shipping and Staging	5-15
7-1. Water Remaining in MCO During Long-term Storage	7-3
7-2. Water Radiolyzed During Long-term Storage From Sludge Containing 0.5 wt% Water ..	7-5
7-3. Water Radiolyzed During Long-term Storage From Sludge Containing 1.5 wt% Water ..	7-6

SPENT NUCLEAR FUEL PROJECT GAS GENERATION FROM N-FUEL IN MULTI-CANISTER OVERPACKS

1.0 INTRODUCTION

During the conversion from wet pool storage to dry storage for spent nuclear fuel at Hanford, gases will be generated from both radiolysis and chemical reactions. The Spent Nuclear Fuel Project (SNF Project) must investigate this phenomenon to determine if the gases can overpressurize the storage containers through either static or dynamic processes. To understand this phenomenon, it is helpful to understand the history of the fuel during manufacture, irradiation in the reactors, and wet pool storage.

Nuclear fuel from the N-Reactor was manufactured by co-extruding uranium metal and zirconium metal into a zirconium-jacketed tube shape. This fuel was manufactured in two separate diameters. The fuel elements were assembled with an inner element placed concentrically within an outer element to make a single fuel assembly. These fuel assemblies were then loaded into horizontal tube channels within the reactor and exposed to various levels of neutron irradiation, as determined by the operating requirements of the reactor.

At discharge, the fuel elements were forced by pressure out of the rear face of the reactor and would fall onto a trampoline within a water pool. Then the fuel elements would slide down a chute and fall into a metal cart, which was used to transport them underwater to the handling and storage facility.

Some cladding failures occurred within the reactor, and many of the assemblies were damaged during the unloading cycle. Each fuel assembly weighs approximately 50 pounds and fell 30-50 feet upon unloading. The kinetic energy was high enough to cause some of the assemblies to split their cladding, fracture, or chip as they slammed into metal structures within the water pool. This damage was of no great concern during plant operations, because the assemblies were typically held for approximately six months before transfer to the Plutonium Uranium Extraction (PUREX) facility for reprocessing.

Some of the assemblies were not reprocessed; however, and were held within the storage pools for up to 25 years. Presently, approximately 100,000 fuel assemblies are still in the K-East and K-West Basins. Approximately 45% of the outer assemblies and 30% of the inner assemblies have sustained visible cladding damage and subsequent uranium metal corrosion (WHC 1996a). This corrosion process releases radioactive fission products to the storage pool and could represent an environmental hazard, in the event of a leak from the K Basins to the Columbia River or into the groundwater table.

The Contractor has been tasked by the U.S. Department of Energy to move the spent nuclear fuel from the K Basins to a dry fuel storage building, which is located approximately ten

miles from the Columbia River. The Contractor plans to load the fuel into steel transport and storage containers called Multi-canister Overpacks (MCOs). Each MCO will contain five tiers of fuel. Each tier may contain 54 intact fuel assemblies or a scrap basket filled with irregular-shaped fuel assembly pieces, which range in length from ¼-inch to six inches long. The minimal ¼-inch dimension comes from the requirement that only the scrap retained on a ¼-inch screen will be stored in an MCO.

Current plans are to load the MCO underwater, transport the MCO to a Cold Vacuum Drying (CVD) Station, weld the lid onto the MCO, drain the bulk water from the MCO, cold vacuum dry the fuel with a starting temperature of 50 - 75 °C, transport the MCO to the Canister Storage Building (CSB), hot vacuum dry the fuel, and place the MCO in long-term interim storage.

In removing, drying, transporting, and storing this fuel, the Contractor needs to determine the potential of this fuel for generating gases that could pressurize the MCO. Hydrogen and oxygen are generated by the radiolysis of water. Hydrogen can also be generated through the reaction of water with uranium metal or uranium hydride in a deoxygenated system. Helium gas evolves from alpha particles. Volatile fission products are released from the oxidation of the fuel. Water vapor exists in equilibrium with other water phases. The higher the temperature, the higher the water vapor partial pressure. At each process step, the gas partial pressures must be summed to calculate the gas pressure in the MCO.

Several independent parameters strongly affect the gas generation rates for corroding uranium. Examples of such parameters are the reactive surface area of the uranium metal, the effect of waters of hydration upon the total water inventory for sludges, and the value chosen for the G factor of oxide surfaces.

The surface area used in this evaluation is 127,233 cm² and is based upon a worst-case analysis (WHC 1996b). For this worst case, it is assumed that every fuel assembly position is occupied by a split-end assembly, and that the MCO contained at least one scrap basket filled with 1-inch slices of the fuel assemblies. The surface area may be reduced by administrative measures, and in such cases, the chemical hydrogen production rates will scale proportionately to the exposed area.

It is assumed that the only significant oxides within the MCO are uranium oxides, which can contain strongly bound surface hydroxides and a monolayer of physically bound water. No waters of hydration are assumed within the oxide interiors. Justification for this assumption is found in Section 5.2.1.

The amount of oxide in the MCO cannot be precisely controlled. Initial estimates ranged from 2.7 to 16 kg. At the time this report was written, a larger estimate of 277 kg of oxides was recommended. This 277 kg estimate was intended as an upper bound for purposes of estimating the potential release of particulates.

This report has consistently included each of these three oxide estimates at each process step. This is intended to provide gas generation rates over a wide oxide range.

In evaluating the in-situ production of gases within the MCO, the discussion is organized by looking at specific process steps.

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2.0 FLOODED MULTI-CANISTER OVERPACK CASE

2.1 UNCORRODED FUEL IN A FLOODED MCO

Uncorroded fuel will generate both hydrogen and oxygen through radiolysis, but will not generate gases chemically. In the absence of getters, these gas mixtures are potentially flammable and detonable.

The radiolytic production of hydrogen within an MCO is dependent on many variables; such as, the disintegration rate of radionuclides, the disintegration energy, the types of radiation energy, the mass of water, and the self-shielding by the uranium assemblies. Any discussion of a radiolytic hydrogen generation rate is therefore only meaningful when a set of conditions have been specified. J. S. Lan analyzed the radiation energy absorbed by the components of an MCO for the flooded, drained, cold vacuum dried, and hot vacuum dried configurations using the computer code MCNP, Version 3 (WHC 1996c). P. C. Owczarski took Lan's results and modeled the gas generation rates for different G values—for the high sludge cases as represented in Table 2-1 and the low sludge cases as represented in Table 2-2 (Owczarski 1996). Alpha and beta radiation are ignored, as significant levels of these radiation types are not expected to penetrate the Zircaloy cladding. For uncorroded fuel in a flooded MCO, neither the temperature nor the amount of residual sludge significantly influences the amount of hydrogen being generated.

These gases accumulate in the headspace above the water within the MCO. It is estimated that about 44 liters of free headspace is available.

Table 2-1 considers a case where 16 kg of dry sludge are assumed to be typical of a high sludge process condition. This is not necessarily a bounding sludge condition. Table 2-1 then considers four general cases:

- **A Flooded MCO**

For the flooded case, 500 liters is the nominal free volume estimate; but for this particular case, the analyst used 511 liters for the flooded volume. The increase in the hydrogen generation rate is not linear with water volume, and the hydrogen generation rate is not significantly different for the two cases. The sludge is considered to contain 50% v/v water within the sludge. The free water and the sludge are exposed to a common gamma field.

The computer code MCNP, Version 3, was used to calculate the energy deposition within the free water from alpha, beta, and gamma radiation. This calculation shows that gamma radiation deposits 1.1×10^{-5} watts of energy per gram of water. Alpha and beta particles deposit 1.49×10^{-5} and 1.36×10^{-4} watts per gram, respectively, in the water permeating the sludge.

Table 2-1. High Sludge Case.

MCO Condition	Sludge mass	Target H2O	Kg	Radiation	Watts/g *	G value	H2 Mol/Sec	Duration Hrs	Days	Years	Total H2 Mol.
Flooded	Dry	Flooded Volume 135 gallons	511	gamma(high)	1.10E-05	1	5.82E-07	64	2.6667	0.007306	1.34E-01
		Flooded Volume 135 gallons	511	gamma (Med)	1.10E-05	0.3	1.75E-07	64	2.6667	0.007306	4.02E-02
		Flooded Volume 135 gallons	511	gamma (Low)	1.10E-05	0.1	5.82E-08	64	2.6667	0.007306	1.34E-02
		UO2 Sludge 50% H2O v/v	1.46	alpha (s)	1.49E-06	3	6.77E-09	64	2.6667	0.007306	
		UO2 Sludge 50% H2O v/v	1.46	beta (b)	1.36E-04	1.4	2.88E-08	64	2.6667	0.007306	
		UO2 Sludge 50% H2O v/v	1.46	gamma(high)	1.10E-05	20	3.33E-08	64	2.6667	0.007306	1.59E-02
		UO2 Sludge 50% H2O v/v	1.46	gamma (Med)	1.10E-05	1	1.66E-09	64	2.6667	0.007306	8.57E-03
		UO2 Sludge 50% H2O v/v	1.46	gamma (Low)	1.10E-05	0.1	1.66E-10	64	2.6667	0.007306	8.23E-03
						total, high	6.51E-07				1.50E-01
						total, med	2.12E-07				4.88E-02
						total, low	9.40E-08				2.16E-02
Drained	14 Liters	14 Liters	14	gamma(high)	1.20E-05	1	1.74E-08	2	0.083333	0.000228	1.25E-04
		14 Liters	14	gamma (Med)	1.20E-05	0.3	5.22E-09	2	0.083333	0.000228	3.76E-05
		14 Liters	14	gamma (Low)	1.20E-05	0.1	1.74E-09	2	0.083333	0.000228	1.2534E-5
		UO2 Sludge 50% H2O v/v	1.46	alpha (s)	1.49E-05	3	1.14E-09	2	0.083333	0.000228	
		UO2 Sludge 50% H2O v/v	1.46	beta (b)	1.36E-04	1.4	4.86E-09	2	0.083333	0.000228	
		UO2 Sludge 50% H2O v/v	1.46	gamma(high)	1.20E-05	20	4.86E-09	2	0.083333	0.000228	5.17E-04
		UO2 Sludge 50% H2O v/v	1.46	gamma (Med)	1.20E-05	1	6.13E-09	2	0.083333	0.000228	2.69E-04
		UO2 Sludge 50% H2O v/v	1.46	gamma (Low)	1.20E-05	0.1	3.06E-10	2	0.083333	0.000228	2.57E-04
						total, high	8.93E-08				2.12366 E-04
						total, med	4.26E-08				6.3008 E-04
						total, low	3.75E-08				5.8954 E-04
Cold Vacuum Dried & 2 Year Staging	Thin Monomolecular Film	Thin Monomolecular Film	2.83E-05	gamma(high)	6.90E-06	20	4.0471 E-13	17520	730	2	2.22118 E-05
		Thin Monomolecular Film	2.83E-05	gamma (Med)	6.90E-06	1	2.0235 E-14	17520	730	2	1.11811 E-06
		Thin Monomolecular Film	2.83E-05	gamma (Low)	6.90E-06	0.1	2.0235 E-15	17520	730	2	1.11847 E-7
		UO2 Sludge 10% H2O v/v	1.62E-01	alpha (s)	1.81E-05	3	8.13E-10	17520	730	2	
		UO2 Sludge 10% H2O v/v	1.62E-01	beta (b)	1.47E-04	1.4	3.45E-09	17520	730	2	
		UO2 Sludge 10% H2O v/v	1.62E-01	gamma(high)	1.81E-05	20	5.42E-09	17520	730	2	5.28E-01
		UO2 Sludge 10% H2O v/v	1.62E-01	gamma (Med)	1.47E-04	1	2.47E-09	17520	730	2	3.73E-01
		UO2 Sludge 10% H2O v/v	1.62E-01	gamma (Low)	1.47E-04	0.1	2.47E-10	17520	730	2	2.54E-01
						total, high	9.69E-09				5.28E-01
						total, med	6.74E-09				3.73E-01
						total, low	4.51E-09				2.54E-01
Hot Vacuum CSB Storage	Thin Monomolecular Film	Thin Monomolecular Film	2.83 E-5	gamma(high)	6.90E-06	20	4.05E-13	657000	27375	75	9.18E-05
		Thin Monomolecular Film	2.83 E-5	gamma (Med)	6.90E-06	1	2.02E-14	657000	27375	75	4.72E-06
		Thin Monomolecular Film	2.83 E-5	gamma (Low)	6.90E-06	0.1	2.02E-15	657000	27375	75	4.73E-07
		UO2 Sludge 5% H2O v/v	1.80E-01	alpha (s)	1.62E-05	3	4.05E-10	657000	27375	75	
		UO2 Sludge 5% H2O v/v	8.04E-02	beta (b)	1.48E-04	1.4	1.72E-09	657000	27375	75	
		UO2 Sludge 5% H2O v/v	8.04E-02	gamma(high)	6.90E-08	20	1.15E-09	657000	27375	75	2.22E+00
		UO2 Sludge 5% H2O v/v	8.04E-02	gamma (Med)	6.90E-08	1	5.75E-11	657000	27375	75	2.09E+00
		UO2 Sludge 5% H2O v/v	8.04E-02	gamma (Low)	6.90E-08	0.1	5.75E-12	657000	27375	75	2.08E+00
						total, high	3.28E-09				2.22E+00
						total, med	2.18E-09				2.09E+00
						total, low	2.13E-09				2.08E+00
**Energy absorbed by H2O											
**includes alpha + beta in sludge rows											

Table 2-2. Low Sludge Case.

MCO Condition	Sludge mass	Target H2O	Kg	Radiation	Watts/g *	G value	H2 Mol/Sec	Duration Hrs	Days	Years	Total H2 Mol.
Flooded	kg H2O	135 gallons	511	gamma(high)	1.10E-05	1	5.82E-07	64	2.6667	0.007306	1.34E-01
		135 gallons	511	gamma (Med)	1.10E-05	0.3	1.17E-07	64	2.6667	0.007306	4.02E-02
		135 gallons	511	gamma (Low)	1.10E-05	0.1	5.82E-08	64	2.6667	0.007306	1.34E-02
	2.7	UO2 Sludge 50% H2O v/v	0.246	alpha (s)	1.49E-06	3	1.14E-09	64	2.6667	0.007306	
	2.7	UO2 Sludge 50% H2O v/v	0.246	beta (b)	1.36E-04	1.4	4.86E-09	64	2.6667	0.007306	
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma(high)	1.10E-05	20	5.62E-09	64	2.6667	0.007306	2.68E-03
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma (Med)	1.10E-05	1	2.81E-10	64	2.6667	0.007306	1.45E-03
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma (Low)	1.10E-05	0.1	2.81E-11	64	2.6667	0.007306	1.39E-03
		Total H2, moles	28402.58			total, high	5.94E-07				1.3682 E-1
						total, med	1.81E-07				4.1859 E-2
Drained						total, low	6.43E-08				1.4803 E-2
	14 Liters	14 Liters	14	gamma(high)	1.20E-05	1	1.74E-08	2	0.083333	0.000228	1.2535 E-4
		14 Liters	14	gamma (Med)	1.20E-05	0.3	5.22E-09	2	0.083333	0.000228	3.76039 E-5
		14 Liters	14	gamma (Low)	1.20E-05	0.1	1.74E-09	2	0.083333	0.000228	1.25346 E-5
	2.7	UO2 Sludge 50% H2O v/v	0.246	alpha (s)	1.49E-05	3	1.14E-09	2	0.083333	0.000228	
	2.7	UO2 Sludge 50% H2O v/v	0.246	beta (b)	1.36E-04	1.4	4.86E-09	2	0.083333	0.000228	
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma(high)	1.20E-05	20	4.86E-09	2	0.083333	0.000228	8.73114 E-5
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma (Med)	1.20E-05	1	6.13E-09	2	0.083333	0.000228	4.5404 E-5
	2.7	UO2 Sludge 50% H2O v/v	0.246	gamma (Low)	1.20E-05	0.1	3.06E-10	2	0.083333	0.000228	4.3419 E-5
		Total H2, moles	791.4639			total, high	2.9536 E-8				2.12366 E-4
						total, med	1.1529 E-8				6.3008 E-5
Cold Vacuum						total, low	7.7714 E-9				5.8954 E-5
	Thin Monomolecular Film	Thin Monomolecular Film	2.83E-05	gamma(high)	6.90E-06	20	4.0471 E-13	17520	730	2	2.22118 E-05
		Thin Monomolecular Film	2.83E-05	gamma (Med)	6.90E-06	1	2.0235 E-14	17520	730	2	1.11811 E-08
		Thin Monomolecular Film	2.83E-05	gamma (Low)	6.90E-06	0.1	2.0235 E-15	17520	730	2	1.11847 E-7
	2.7	UO2 Sludge 10% H2O v/v	2.74E-02	alpha (s)	1.61E-05	3	1.3718 E-10	17520	730	2	
	2.7	UO2 Sludge 10% H2O v/v	2.74E-02	beta (b)	1.47E-04	1.4	5.8299 E-10	17520	730	2	
	2.7	UO2 Sludge 10% H2O v/v	2.74E-02	gamma(high)	1.61E-05	20	9.1454 E-10	17520	730	2	8.9083 E-2
	2.7	UO2 Sludge 10% H2O v/v	2.74E-02	gamma (Med)	1.47E-04	1	4.1642 E-10	17520	730	2	8.29282 E-2
	2.7	UO2 Sludge 10% H2O v/v	2.74E-02	gamma (Low)	1.47E-04	0.1	4.1642 E-11	17520	730	2	4.29318 E-2
		Total H2, moles	1.52225			total, high	1.16351 E-9				8.9105 E-2
Hot Vacuum CSB Storage						total, med	1.1366 E-9				6.2927 E-2
						total, low	7.6182 E-10				4.2932 E-2
	Thin Monomolecular Film	Thin Monomolecular Film	2.83 E-5	gamma(high)	6.90E-06	20	4.05E-13	657000	27375	75	9.18E-05
		Thin Monomolecular Film	2.83 E-5	gamma (Med)	6.90E-06	1	2.02E-14	657000	27375	75	4.72E-06
		Thin Monomolecular Film	2.83 E-5	gamma (Low)	6.90E-06	0.1	2.02E-15	657000	27375	75	4.73E-07
	2.7	UO2 Sludge 10% H2O v/v	1.357 E-2	alpha (s)	1.62E-05	3	6.83E-11	657000	27375	75	
	2.7	UO2 Sludge 10% H2O v/v	1.357 E-2	beta (b)	1.48E-04	1.4	2.90E-10	657000	27375	75	
	2.7	UO2 Sludge 10% H2O v/v	1.357 E-2	gamma(high)	6.90E-08	20	1.94E-10	657000	27375	75	3.75E-01
	2.7	UO2 Sludge 10% H2O v/v	1.357 E-2	gamma (Med)	6.90E-08	1	9.70E-12	657000	27375	75	3.53E-01
	2.7	UO2 Sludge 10% H2O v/v	1.357 E-2	gamma (Low)	6.90E-08	0.1	9.70E-13	657000	27375	75	3.51E-01
		Total H2, moles	0.75534			total, high	5.53E-10				3.75E-01
						total, med	3.69E-10				3.63E-01
						total, low	3.60E-10				3.51E-01
*Energy absorbed by H2O											**includes alpha + beta in sludge rows

G values are defined as the number of product molecules formed by the radiolysis process per 100 electron volts (ev) of radiation energy. G values ranging from 0.3 to 1.0 have been reported for the interaction of gamma rays with free water. To be conservative, the value of 1.0 was chosen for this analysis.

G values ranging from 10 to 40 have been reported for the interaction of gamma radiation with water adsorbed on oxide surfaces (Owczarski 1996). No theoretical explanation can be offered at this time for these rather high G values. Since they were experimentally determined, their authenticity is accepted, and a fairly typical value of 20 was chosen for this analysis.

Using the energy deposition rate calculated by Lan and the G values from Owczarski, the hydrogen generation rate can be calculated. The radiolytic oxygen generation rate is assumed to be one-half of the hydrogen generation rate.

- **A Drained MCO**

For a drained MCO, it is assumed that 14 kg of water represent a reasonable heel for the 16 kg of sludge. The sludge is still permeated with water at the 50% v/v ratio. All other conditions were similar to the flooded case.

- **A Cold Vacuum Dried MCO**

Lacking experimental data, there are differences of opinion as to the amount of sludge within the MCO, and the amount of free water that will be retained within the sludge and upon oxide surfaces. Sixteen kg of sludge was arbitrarily defined as a relatively high, but not necessarily bounding, amount of sludge for a process case.

The amount of water that is retained after CVD will in some measure be proportional to the amount of sludge being dried, because of diffusion and the heat transfer phenomena. No experimental data is available for water retention by UO_2 , so data from PuO_2 will be used as a basis, until characterization data is available (Rasmussen 1969). Rasmussen, quoting data from NUMEC-P-90 (page 17), discusses water absorption rather than water retention; but assuming the phenomenon is reversible, one may calculate a suitable water retention value.

Rasmussen shows that when pure plutonium dioxide is calcined at 350 °C for ½ hour, and subsequently exposed to air at relative humidity (RH) less than 20% at 25 °C, less than 5 grams of water are absorbed per kg. This is ½ % w/w, but by using the appropriate density ratios, a value of 6% v/v can be calculated. This value was increased to 10% v/v for the sake of conservatism.

The gamma energy deposition rate is lower for a thin film of water because of decreased linear energy absorption coefficients within a thin film.

- **A Hot Vacuum Dried MCO**

For a hot vacuum dried MCO, it was assumed the residual moisture is 0.5% w/w. Again, a reduction can be seen in the linear absorption coefficients for gamma energy in the water retained in the sludge.

2.1.1 Calculation of Radiolytic Gas Generation Rates

- **Initial data**

From Table 2-1:

Mass dry sludge = 16 kg

Volume water = 511 liters

Energy deposition rate = $1.1\text{E-}5$ watts/gram water

Conservative G value = 1 molecule H_2 /100 ev energy

- **Example Calculation**

Flooded Volume:

H_2 rate moles/sec =

$(1.10\text{E-}05 \text{ watts/gram } \text{H}_2\text{O}) * (511,000 \text{ grams } \text{H}_2\text{O}) * (6.242 * 10^{18} \text{ ev/watt-sec.}) *$

$(1 \text{ molecule } \text{H}_2/100 \text{ ev}) / (6.023 * 10^{23} \text{ molecules } \text{H}_2/\text{gmole})$

$= 5.8254\text{E-}07 \text{ H}_2 \text{ gmole/sec.}$

- **Sludge**

$16 \text{ kg} / 5.47 \text{ grams/ml} = 2,925 \text{ mls}$

Now, if 50% of that volume is water, there are 1,462.5 grams of water in 16 kg of sludge.

so,

$(1.49\text{E-}05 \text{ watts(alpha)/gram } \text{H}_2\text{O}) * (1462.5 \text{ grams } \text{H}_2\text{O}) * (6.242 * 10^{18} \text{ ev/watt-sec.})$

$* (3 \text{ molecules } \text{H}_2/100 \text{ ev}) / (6.023 * 10^{23} \text{ molecules } \text{H}_2/\text{gmole})$

$= 6.773\text{E-}9 \text{ gmole } \text{H}_2/\text{sec}$ from alpha radiation in sludge from Table 2-1, the beta contribution is $2.878\text{E-}8 \text{ gmole } \text{H}_2/\text{sec}$, and using the conservative G value of 20 for gamma on an oxide surface, the sludge gamma contribution is $3.3282\text{E-}8 \text{ gmole } \text{H}_2/\text{sec}$.

The total H_2 production is the sum of the hydrogen generation in the flooded volume and in the sludge:

$$H_2 = 5.8254E-07 + 6.773E-9 + 2.878E-8 + 3.3282E-8$$

$$= 6.513E-07 \text{ gmoles/sec}$$

$$= 2.35E-3 \text{ gmoles/hr}$$

Similar rates can be calculated for the 2.7 kg sludge case and the 277 kg sludge case.

Table 2-3 and Figure 2-1 were generated by multiplying these rates (for hydrogen), and one half of these rates (for oxygen), by the number of elapsed hours.

Table 2-3. Radiolytic Gas Generation for a Flooded MCO.*

Time Hours	2.7 kg Sludge		16 kg Sludge		277 kg Sludge	
	Oxygen	Hydrogen	Oxygen	Hydrogen	Oxygen	Hydrogen
1	1.069E-3	2.139E-3	1.172E-3	2.345E-3	2.145E-3	4.290E-3
5	5.347E-3	1.069E-2	5.862E-3	1.172E-2	1.073E-2	2.145E-2
10	1.069E-2	2.139E-2	1.172E-2	2.345E-2	2.145E-2	4.290E-2
20	2.139E-2	4.278 E-2	2.345E-2	4.690E-2	4.290E-2	8.580E-2
30	3.208E-2	6.416E-2	3.517E-2	7.034E-2	6.435E-2	1.287E-1
40	4.278E-2	8.555E-2	6.900E-2	9.379E-2	8.580E-2	1.716E-1
50	5.347E-2	1.069E-1	5.862E-2	1.172E-1	1.073E-1	2.145E-1
60	6.416E-2	1.283E-1	7.034E-2	1.407E-1	1.287E-1	2.574E-1
70	7.486E-2	1.497E-1	8.207E-2	1.641E-1	1.501E-1	3.003E-1

* Units are moles of gas.

Figure 2-1. Gas Generation for Uncorroded Fuel in a Flooded MCO.

The data in Table 2-3 and Figure 2-1 show that the radiolytic gas generation rate is essentially linear with time.

For a flooded MCO, the large amount of liquid water is the main source of radiolytic gases. Small amounts of sludge do not strongly affect the radiolytic gas generation rates.

For the 277 kg case, approximately 42 hours are required to reach a flammable mixture in the headspace in the MCO. This would indicate that a maximum flooded sealed duration would be 21 hours (half the time to reach flammable concentration per packaging and shipping criteria). This is an unacceptably short duration for a sealed package, unless a reliable, positive means to vent and dilute the MCO headspace contents is in place at all times.

2.2 CORRODED FUEL IN A FLOODED MCO

2.2.1 Radiolytic Gases

The radiolytic gas generation rate for corroded fuel in a flooded MCO is approximately the same as for uncorroded fuel. This rate is not significant, when compared to the rate for chemically produced hydrogen from corroded fuel assemblies.

2.2.2 Chemically Produced Hydrogen

Hydrogen is chemically produced from water under strongly reducing conditions; in this case, by contact with uranium metal in the absence of strong oxidizers such as oxygen gas.

The liquid pool water is in direct contact with the atmosphere and is initially oxygenated. Oxygen in liquid water is not quite as effective in reducing the water reaction rate with uranium metal as oxygen in air. It is possible, therefore, that small amounts of hydrogen may be emitted from the reaction of oxygenated liquid water.

For the sake of simplicity, the possibility of a hydrogen production rate from oxygenated water is ignored, and only the hydrogen production from deoxygenated water was tallied.

If the MCO is sealed for shipment to the CVD station, the residual oxygen will react with exposed uranium surfaces, and the system will eventually become deoxygenated. In the absence of oxygen, hydrogen will be produced chemically:

The rate is calculated as follows:

Moles O_2 fixed/hr/cm² =

$$(10) * 10^{(7.634 - 3016/T)} (\text{mg } \text{O}_2/\text{cm}^2/\text{hr}/32000 \text{ mg/mole}) \quad (2)$$

Moles H_2 /hr/cm² =

$$\begin{aligned} & (\text{Moles } \text{O}_2/\text{hr}/\text{cm}^2) * 2 \text{ moles of } \text{H}_2/\text{mole of } \text{O}_2 \\ & = 2 * 10 * 10^{(7.634 - 3016/T)} / 32000 \end{aligned} \quad (3)$$

The hydrogen production rate from the chemical reaction rate of water with uranium metal can now be calculated and is found in Table 2-4 and Figure 2-2.

Table 2-4. Hydrogen Production From Water on Uranium Metal in Flooded MCO.

Time Hours	Temp (°C)				
	10	20	50	75	100
1	6.01E-07	1.39E-06	1.25E-05	5.85E-05	2.23E-04
5	3.00E-06	6.93E-06	6.25E-05	2.93E-04	1.11E-03
10	6.01E-06	1.39E-05	1.25E-04	5.85E-04	2.23E-03
20	1.20E-05	2.77E-05	2.50E-04	1.17E-03	4.45E-03
30	1.80E-05	4.16E-05	3.75E-04	1.76E-03	6.68E-03
40	2.40E-05	5.55E-05	5.00E-04	2.34E-03	8.90E-03
50	3.00E-05	6.93E-05	6.25E-04	2.93E-03	1.11E-02
60	3.60E-05	8.32E-05	7.50E-04	3.51E-03	1.34E-02
70	4.20E-05	9.71E-05	8.75E-04	4.10E-03	1.56E-02

* Units are moles of hydrogen/cm².

Figure 2-2. Hydrogen from Corroded Metal in a Flooded MCO.

2.2.3 Hydrogen Generation from Reaction of Hydrides

According to M. Baker, the aqueous corrosion product on uranium metal consists of a mixture of uranium oxides and uranium hydride (Baker 1966). The percentage of hydride is estimated to vary from 2% to 9% depending on the reaction conditions. The percentage of hydride rises with relative humidity and decreases with oxygen concentration. For corrosion products formed in an open pool, the hydride percentage is conservatively estimated to be 9%.

This finely divided black powder has a surface area of 30 m²/g as measured with the krypton B.E.T. method. X-ray powder crystallography showed a UO₂-type structure and a lattice spacing corresponding to UO_{2-x} where x varied from 0.0 to 0.1. Because of the small crystallite size, a diffuse pattern was obtained. Minor products comprising less than 10-15% of the total would not be distinguishable.

The reaction rate of uranium hydride within the natural corrosion product with water is not known. The very fact that it survives long enough to be measured in the laboratory is an indicator that it is not very reactive at 10 °C.

Pure uranium hydride reacts rapidly with water at room temperature, although the reaction becomes very slow within a few minutes and after 2 weeks, only 15-20% reaction has occurred. At 100 °C, 80% reaction was achieved within 40 hours.

Solid surface reactions are typically first order with respect to surface area. If a constant relationship between the molar-specific surface area and N (the number of moles of uranium hydride) is assumed, then, for first order reactions:

$$dn/dt = -\lambda * N \quad (4)$$

and

$$N = N_0 * \text{Exp}(-\lambda * t) \quad (5)$$

λ can be defined from:

$$\text{Ln}(N/N_0) = -\lambda * t$$

so

$$\lambda_1 = -\text{Ln}(0.8)/336 = 6.64118 * 10^{-4} \quad (6)$$

and

$$\lambda_2 = -\text{Ln}(0.2)/40 = 4.02359 * 10^{-2} \quad (7)$$

Now λ is the fraction that reacts in one hour and, therefore, is a reaction rate. The Arrhenius reaction rate can then be described by:

$$\text{Rate} = A_0 * \text{Exp}(\Delta G/RT) \quad (8)$$

so

$$6.64118 * 10^{-4} = A_0 * \text{Exp}(\Delta G/R * 298) \quad (9)$$

$$4.02359 * 10^{-2} = A_0 * \text{Exp}(\Delta G/R * 373) \quad (10)$$

and solved simultaneously,

$$\lambda = 10^{(5.69034 - 2644.11/T)} \quad (11)$$

where:

λ is the fraction of UH_3 reacted per hour

T is the temperature in $^{\circ}\text{K}$.

Note: The above mathematical treatment is essentially correct for calculating the rate of reaction of uranium hydride with water. When the basic matrix is changed from hydride to oxide, the diffusion rates of water are slower and the resulting reaction rates will slow. There is reason to believe that the hydrogen production from oxides containing small amounts of hydride will be slower than calculated by the above model; however, since we do not have experimental reaction rates of small amounts of hydride embedded in large amounts of oxides, and since these rates are conservative from a safety viewpoint, they will be used.

The reaction of uranium hydride with water is:

Now the hydrogen generation rate from the reaction of uranium hydride with water can be calculated for different temperatures.

First, the case of uranium hydride in the corrosion product attached to uranium metal is examined. This hydride is formed by reaction of uranium metal with nascent hydrogen formed at the metal surface. It exists for some time period and then reacts with either oxygen or water.

If it reacts with oxygen, only water is generated as a product and no hydrogen is emitted. If it reacts with water, hydrogen is emitted as a product and the question is whether this hydrogen should be counted as a gas contributing to the pressurization of the MCO. It should not be counted as a pressurizing gas for the following reason. The initial reaction rate equations describe all of the hydrogen that is being released from water. Part of that hydrogen is trapped temporarily as hydride and is then released at a later time. The hydrogen from hydride in the metal corrosion film is, therefore, already accounted for and is not a new source of hydrogen.

The next question is whether sludge, which is not attached to the metal and is added to the MCO, contains significant quantities of hydrides, and whether hydrogen released from the sludge should be considered a source of pressurizing gas.

To answer this question, the history of the sludge in the canisters must be considered. The sludge in the canisters has formed continuously since the last fuel segregation campaign in 1983-84.

The rate of formation of uranium hydride is the rate of corrosion multiplied by 0.09.

$$+dn/dt = k_1 * \text{Surface Area} * 0.09 = K \quad (13)$$

Where k_1 is the molar corrosion rate constant

The decrease in hydride due to reaction with water

$$-dn/dt = \lambda * n \text{ where } n \text{ is the number of gmoles of hydride} \quad (14)$$

so the net change in n is:

$$\text{net } dn/dt = K - \lambda * n \quad (15)$$

this can be rearranged algebraically to give

$$dn / (K - \lambda * n) = dt \quad (16)$$

upon integrating

$$(K - \lambda * n) / (K - \lambda * n_0) = e^{-\lambda * t} \quad (17)$$

Now since $n_0 = 0$

$$(K - \lambda * n) / K = e^{-\lambda * t} \quad (18)$$

$$1 - \lambda * n / K = e^{-\lambda * t} \quad (19)$$

$$\lambda * n = K(1 - e^{-\lambda * t}) \quad (20)$$

$$\text{or } \lambda * n / K = 1 - e^{-\lambda * t} \quad (21)$$

Now since in this case

$$1 - e^{-\lambda * t} =$$

$$1 - e^{-(10(5.69034-2644.11/283.16) * 13Y * 365.25 * 24}$$

$$= 1 \quad (22)$$

the system is in equilibrium and

$$-dn/dt = \lambda * n = + dn/dt \quad (23)$$

Now the rate of UH_3 reaction with water to produce hydrogen at 10°C is:

$$\lambda \cdot n / \text{kg sludge} =$$

$$(10^{(5.69034-2644.11/283.16)} \cdot 1000/241) \cdot 0.09 \cdot \text{Exp}[(10^{(5.69034-2644.11/283.16)}) \cdot t] \quad (24)$$

$$= 8.408\text{E-}5 \text{ gmoles } \text{UH}_3 \text{ per hour/kg sludge}$$

and since $7/2$ moles of hydrogen are produced per mole of UH_3 reacted:

$$= 2.942 \text{ E-}4 \text{ gmoles } \text{H}_2 \text{ per hour/kg sludge}$$

This calculation is typical of the calculations found in Table 2-5 and Figure 2-3.

The hydrogen generation can never exceed that available, if all of the hydride in the sample were converted to hydrogen. The total hydride in one kg of sludge is assumed to be 90 grams. If all of this hydride were converted to hydrogen, the maximum amount is:

$$90/241 \cdot (7/2) = 1.307 \text{ moles hydrogen/kg sludge.}$$

Table 2-5. Hydrogen Generation from Uranium Hydride in Sludge in a Flooded MCO.

Log Hours	Temp $^\circ\text{C}$				
	10	20	50	75	100
0	2.942E-04	6.125E-04	4.199E-03	1.610E-02	5.052E-02
1	2.939E-03	6.112E-03	4.139E-02	1.523E-01	4.243E-01
2	2.907E-02	5.975E-02	3.555E-01	8.895E-01	1.158E-00
3	2.405E-01	4.048E-01	5.065E-01	5.065E-1	5.065E-1

* Units are in gmoles of hydrogen/cm².

Figure 2-3. Hydrogen Generation From Uranium Hydride in a Flooded MCO.

Figure 2-3 shows that sludge can be a significant source of hydrogen. One kg of sludge at 100 °C can generate a mole of hydrogen in 40 hours. This is five times the flammability limit in a flooded MCO.

2.3 FISSION PRODUCT GAS RELEASE IN A FLOODED MCO

Fission product gases diffuse slowly from the oxide corrosion product. The release rate is too slow to significantly affect the MCO pressure.

2.4 HELIUM GAS RELEASE IN A FLOODED MCO

Helium gas forms from the alpha particles emitted by heavy metals. This helium is not a significant factor in the pressurization of the MCO.

2.5 WATER VAPOR PRESSURE IN A FLOODED MCO

The partial pressure of water increases as the temperature of the MCO is raised. The increase in the number of gmols of water vapor as the MCO heats from 10 °C to any final temperature was added to the other gases to establish the final MCO pressure.

2.6 PRESSURIZATION OF A SEALED FLOODED MCO

Pressurization of a sealed flooded MCO is caused by the accumulation of all of the gases previously discussed. To illustrate the calculation, for the following set of parameters:

- Reactive metal surface area = 127,233 cm² as described for a “worst-case” MCO (WHC 1996b)
- Sludge mass = 2.7 kg
- Headspace = 44 liters
- Temperature = 10 °C
- Radiation energy absorption as described in Tables 2-1 and 2-2
- Time = 1 hour

The rate of pressurization of a flooded MCO can be modeled in the following manner:

Total gmols of hydrogen = gmols of radiolytic hydrogen + gmols of hydrogen from metal reaction + gmols of hydrogen from hydride reactions with water

$$= ((0.002139)+0+(127233*6.007E-07)+(0.0002942*2.7))$$

Since hydrogen is soluble in water, Henry’s law can be used to estimate the hydrogen fraction remaining in the vapor phase:

$$M_b = k_m P = k_m * N_b * R * T / V$$

Where M_b is the number of gmols of hydrogen in the liquid phase

k_m is henry’s constant in gmols per 1000 g solvent per atm.

N_b is the number of gmols in the gas phase

R is the universal gas constant

T is absolute temperature

V is the free volume in liters

M_h and N_h = total gmols of hydrogen

by solving the above 2 equations simultaneously, one may calculate the number of moles of hydrogen remaining in the gas phase.

Pressure increase in atmospheres =

$$(\text{gmols water vapor}) + (\text{gmols of hydrogen in the vapor phase}) * 0.08205 * 283.16 / 44 \quad (25)$$

$$= (0 + (0.0644073)) * (0.08205 * 283.16 / 44)$$

$$= 0.0340$$

The remaining calculations for the 2.7 kg sludge case, the 16 kg sludge case, and the 277 kg sludge case are found in Tables 2-6, 2-7, and 2-8, and Figures 2-4, 2-5, and 2-6.

Table 2-6. Pressure Increase for a Flooded MCO With 2.7 kg Sludge.*

Time Hours	Temp(°C)				
	10	20	50	75	100
1	0.0340	0.0885	0.7905	3.5410	12.789
5	0.1700	0.4002	3.5683	16.352	60.927
10	0.3401	0.7898	7.0402	32.400	121.05
20	0.6807	1.5690	13.983	64.481	241.20
30	1.0202	2.3481	20.923	96.550	361.20
40	1.3603	3.1272	27.862	128.60	481.14
50	1.7003	3.9063	4.800	160.64	601.04
60	2.0404	4.6853	41.737	192.66	720.92
70	2.3804	5.4643	48.672	224.67	840.78
80	2.7204	6.2433	55.605	256.67	960.62
90	3.0605	7.0222	62.538	288.67	1080.5
100	3.4005	7.8011	69.470	320.65	1200.3

* Units are in atmospheres.

Figure 2-4. Pressurization of Flooded-Sealed "Worst-Case" MCO With 2.7 kg Sludge.

Figure 2-4 shows that for a sealed and flooded MCO containing 2.7 kg of sludge, the nominal 10 atmosphere limit is reached between 100 and 1000 hours at 10 °C and 20 °C, 15 hours at 50 °C, 3 hours at 75 °C, and less than 1 hour at 100 °C.

Table 2-7. Pressure Increase for a Flooded MCO With 16 kg Sludge.*

Time Hours	Temp(°C)				
	10	20	50	75	100
1	0.0358	0.0921	0.8148	3.6022	13.074
5	0.1789	0.4182	3.6889	16.805	62.241
10	0.3577	0.8258	7.2794	33.279	123.44
20	0.7154	1.6409	14.453	66.142	245.15
30	1.0730	2.4556	21.615	98.936	366.04
40	1.4306	3.2702	28.768	131.63	486.57
50	1.7881	4.0846	35.913	164.23	606.87
60	2.1456	4.8988	43.050	196.75	727.01
70	2.5030	5.7128	50.180	229.21	847.05
80	2.8604	6.5266	57.302	261.60	967.01
90	3.2177	7.3402	64.417	293.93	1086.93
100	3.2177	8.1536	71.525	326.22	1206.82

* Units are in atmospheres.

Figure 2-5. Pressurization of Flooded-Sealed “Worst-Case” MCO With 16 kg Sludge.

Figure 2-5 shows that for a sealed and flooded MCO containing 16 kg of sludge, the nominal 10 atmosphere limit is reached between 100 and 1000 hours at 10 °C, approximately 100 hours at 20 °C, 14 hours at 50 °C, 3 hours at 75 °C, and less than 1 hour at 100 °C.

Table 2-8. Pressure Increase for a Flooded MCO With 277 kg Sludge.*

Time Hours	Temp(°C)				
	10	20	50	75	100
1	0.0695	0.1621	1.2901	5.4040	18.652
5	0.3462	0.7677	6.0501	25.681	88.017
10	0.6947	1.5239	11.964	50.517	170.30
20	1.3888	3.0337	23.667	98.712	322.71
30	2.0816	4.5387	35.158	145.73	460.97
40	2.7737	6.0403	46.504	190.97	593.13
50	3.4650	7.5386	57.709	234.66	721.20
60	4.1555	9.0335	68.777	276.96	846.55
70	4.8454	10.525	79.714	318.05	970.06
80	5.5344	12.013	90.522	358.07	1092.3
90	6.2226	13.498	101.20	397.14	1213.8
100	6.9099	14.980	111.77	435.37	1334.8

* Units are in atmospheres.

Figure 2-6. Pressurization of a Flooded “Worst-Case” MCO With 277 kg Sludge.*

Figure 2-6 shows that for a sealed and flooded MCO containing 277 kg of sludge, the nominal 10 atmosphere limit is reached between 100 and 1000 hours at 10 °C, 70 hours at 20 °C, 9 hours at 50 °C, 2 hours at 75 °C, and within 30 minutes at 100 °C.

2.7 SUMMARY FOR FLOODED CASE

Uncorroded Fuel - For uncorroded fuel, the gas production is limited to the radiolytically produced gases. Since the MCO will be vented while in the K Basins area, these radiolytic gases do not pressurize the MCO. They do represent a flammability or detonation hazard unless the MCO is vented and purged.

During shipping to the CVD station, the MCO may be sealed. If this shipment is limited to 8 hours, the amount of hydrogen and oxygen generated radiolytically are 0.021 and 0.010 moles respectively. At ambient temperatures, this corresponds to 1.1 and 0.55 volume %, respectively. If a void volume of 44 liters is assumed within the MCO lid, this increases the static pressure by the ratio of N/N_0 ,

where:

N = number of moles of gas after radiolysis event

N_0 = number of moles of gas before radiolysis event

$$N/N_0 = 1.92 / 1.89 = 1.016.$$

This pressure increase is therefore 1.6% and the hydrogen and oxygen concentrations are below reasonable detonable limits (WHC 1996d).

It has been shown that for non-corroded fuel containing large amounts of sludge, flammable mixtures of hydrogen and oxygen will be reached within two days. Shipping and packaging regulations prevent shipment after one day without a means of a safety class venting and dilution system.

Corroded Fuel - For the worst-case high surface area MCO (in an unsealed, oxygenated MCO environment), oxygen is fixed by the reactive fuel, and hydrogen is limited to that produced radiolytically. The protection of oxygen is a significant concept. As long as oxygen is present, hydrogen is not chemically generated in significant amounts, and the overall corrosion rate that releases heat is 20 times slower than in an oxygen-free environment.

Unless positive measures are introduced to insure the presence of oxygen, it is conservatively assumed the reactions proceed by oxygen-free kinetics with the simultaneous production of hydrogen gas.

Figures 2-4 through 2-6 show that the time to reach the nominal 10 atmosphere limit is exponentially related to temperature and in some positive fashion to the amount of sludge. With large amounts of sludge (277kg) and with high temperatures (100°C), the pressure limit is reached in a flooded, sealed MCO in 26 minutes.

3.0 DRAINED CASE

At the CVD station, the MCO will be heated until the interior temperatures are in the 50 - 75 °C range and will then be drained. Upon draining, the free volume of the MCO is approximately 500 liters. Interior surfaces will be damp, void spaces within the fuel assemblies may still contain liquid water, and a small residual heel of liquid water is expected in the bottom of the MCO. Fourteen kg of water is a reasonable estimate for the residual water. The drained case possesses unique safety concerns since the corrosion reactivity is the same as for the flooded case, but the ability of the system to conduct heat to the environment is much smaller.

3.1 UNCORRODED FUEL IN A DRAINED MCO

From Tables 2-1 and 2-2, the total hydrogen generation rates are derived for the 2.7 kg sludge case, the 16 kg sludge case, and the 277 kg sludge case. From these generation rates, the production of radiolytic gases can be calculated as a function of time. This data for the drained state is presented in Table 3-1 and Figure 3-1.

Table 3-1. Radiolytic Gas Generation for Uncorroded Fuel in a Drained MCO.*

Time Hours	2.7 kg sludge		16 kg sludge		277 kg sludge	
	Hydrogen	Oxygen	Hydrogen	Oxygen	Hydrogen	Oxygen
1	0.000106	0.000053	0.000321	0.000161	0.004479	0.002239
5	0.000532	0.000266	0.003214	0.001607	0.02239	0.01120
10	0.001063	0.000532	0.006427	0.003214	0.04479	0.02239
20	0.002127	0.001063	0.009641	0.004821	0.08958	0.04479
30	0.003190	0.001595	0.012855	0.006427	0.01343	0.06718
40	0.004253	0.002127	0.016069	0.008034	0.01792	0.08958
50	0.005316	0.002658	0.019282	0.009641	0.22394	0.11197
60	0.006380	0.003190	0.020568	0.010284	0.26873	0.13436
70	0.007434	0.003717	0.022304	0.011252	0.31351	0.15676

* Units are moles of gases.

Figure 3-1. Radiolytic Gas Generation for Uncorroded Fuel in a Drained MCO.

These amounts of hydrogen and oxygen are trivial and are not expected to influence the pressure or flammability of the MCO.

3.2 CORRODED FUEL IN A DRAINED MCO

The gas generation rate for corroded fuel must include both radiolysis gases and chemically generated gases. The chemically generated gases are discussed next.

3.2.1 Hydrogen Generated by Water Reaction on Uranium Metal in a Drained MCO

By draining the MCO and replacing the atmosphere with inert gas, the residual oxygen in the MCO is reduced to a very small concentration and ceases to moderate the chemical reactivity of the uranium with water. Since the atmosphere is saturated with water vapor, the exposed uranium will react at its maximum deoxygenated rate. The deoxygenated reaction rate constant for uranium with liquid water is the same as for uranium with 100% RH vapor. Therefore, the same chemical hydrogen generation rates for the drained state are expected for the deoxygenated flooded case. (Refer to Table 2-4 and Figure 2-2 for the hydrogen generation rate per cm² of reactive uranium metal.)

3.2.2 Hydrogen Generated by Water Reaction on Uranium Hydride in a Drained MCO

Since the water vapor remains at 100 % RH, the reaction rate for uranium hydride should remain the same as for the flooded MCO case. The hydrogen generation rates from the reaction of water vapor with uranium hydride can be seen in Table 2-5 and Figure 2-3.

3.2.3 Fission Product Gas Release in a Drained MCO

Fission product gases diffuse slowly from the oxide corrosion product. The release rate is too slow to significantly affect the MCO pressure.

3.2.4 Helium Gas Release in a Drained MCO

Helium gas forms from the alpha particles emitted by heavy metals. This helium is not a significant factor in the pressurization of the MCO.

3.2.5 Water Vapor Pressure in a Drained MCO

The partial pressure of water increases as the temperature of the MCO is raised. The increase in the number of gmols of water vapor, as the MCO heats from 10 °C to any final temperature, was added to the other gases to establish the final MCO pressure.

3.2.6 Pressurization of a Sealed Drained MCO

Pressure in a sealed, drained MCO will be less than for a flooded MCO, at a given temperature, for several reasons. The free volume has increased from 44 liters to 500, and the radiolytic gas generation rate is smaller. As an example, assuming the following parameters:

- Reactive metal surface area = 127233 cm² as described for a “worst-case” MCO. (WHC 1996b)
- Sludge mass = 2.7 kg
- Headspace = 500 liters
- Temperature = 10 °C
- Radiation energy absorption as described in Tables 2-1 and 2-2
- Time = 1 hour

The rate of pressurization of a drained MCO can be modeled by summing in the following manner:

Pressure increase in atmospheres =

$$\begin{aligned} & (\text{gmoles radiolysis gases}) + (\text{gmoles water vapor}) + \\ & (\text{reactive metal surface area} * (\text{gmoles H}_{2\text{ metal}}/\text{cm}^2 + \text{gmoles H}_{2\text{ UH}_3}/\text{cm}^2)) * \\ & 0.08205 * 283.16 / 44 \end{aligned} \quad (26)$$

$$\begin{aligned} & = (0.0000000893 * 3600 * 1) + (0) + (127233 * (6.007\text{E-}07 + 1.6625\text{E-}08)) \\ & * 0.08205 * 283.16 / 44 \end{aligned}$$

$$= 0.041728 \text{ atm.}$$

The remaining calculations are found in Tables 3-2, 3-3, 3-4 and Figures 3-2, 3-3, 3-4.

Table 3-2: Pressure Increase for a Drained MCO With 2.7 kg Sludge.*

Time Hours	Temp (°C)				
	10	20	50	75	100
1	0.0036	0.0092	0.0829	0.3743	1.3884
5	0.0180	0.0423	0.3807	1.7658	6.6759
10	0.0359	0.0837	0.7529	3.5046	13.280
20	0.0719	0.1665	1.4972	6.9807	26.477
30	0.1078	0.2493	2.2413	10.456	39.658
40	0.1437	0.3321	2.9852	13.929	52.832
50	0.1797	0.4149	3.7290	17.400	66.002
60	0.2156	0.4977	4.4727	20.870	79.169
70	0.2515	0.5804	5.2161	24.338	92.334
80	0.2874	0.6632	5.9595	27.806	105.49
90	0.3233	0.7460	6.7028	31.272	118.66
100	0.3593	0.8287	7.4459	34.738	131.82

* Units are in atmospheres pressure.

Figure 3-2. Pressurization of a Drained-Sealed "Worst-Case" MCO 2.7 kg Sludge.

Figure 3-2 shows that for a sealed and flooded MCO containing 2.7 kg of sludge, the nominal 10 atmosphere limit is reached in 2794 hours at 10 °C, 1087 hours at 20 °C, 134.4 hours at 50 °C, 28.7 hours at 75 °C, and within 7.5 hours at 100 °C.

Table 3-3. Pressure Increase for a Drained MCO With 16 kg Sludge.*

Time Hours	Temp (°C)				
	10	20	50	75	100
1	0.0038	0.0096	0.0855	0.3842	1.4197
5	0.0189	0.0443	0.3937	1.8148	6.8202
10	0.0378	0.0876	0.7786	3.6000	13.543
20	0.0757	0.1743	1.5477	7.1607	26.911
30	0.1135	0.2609	2.3156	10.714	40.189
40	0.1514	0.3475	3.0825	14.257	53.429
50	0.1892	0.4341	3.8485	17.789	66.642
60	0.2270	0.5206	4.6137	21.313	79.839
70	0.2648	0.6072	5.3781	24.829	93.024
80	0.3026	0.6937	6.1417	28.339	106.20
90	0.3404	0.7802	6.9045	31.843	119.37
100	0.3782	0.8667	7.6666	35.341	132.54

* Units are in atmospheres pressure.

Figure 3-3. Pressurization of a Drained-Sealed "Worst-Case" MCO 16 kg Sludge.*

Figure 3-3 shows that for a sealed and flooded MCO containing 16 kg of sludge, the nominal 10 atmosphere limit is reached in 2698 hours at 10 °C, 1175 hours at 20 °C, 130.7 hours at 50 °C, in 28 hours at 75 °C, and within 7.4 hours at 100 °C.

Table 3-4. Pressure Increase for a Drained MCO With 277 kg Sludge.*

Time Hours	Temp(°C)				
	10	20	50	75	100
1	0.0075	0.0172	0.1366	0.5796	2.0325
5	0.0377	0.0823	0.6476	2.7774	9.6521
10	0.0754	0.1637	1.2825	5.4692	18.691
20	0.1508	0.3260	2.5388	10.693	35.433
30	0.2260	0.4878	2.7725	15.789	50.621
40	0.3011	0.6493	4.9904	20.693	65.138
50	0.3761	0.8104	6.1933	25.427	79.206
60	0.4511	0.9712	7.3815	30.013	92.975
70	0.5260	1.1315	8.5556	34.466	106.54
80	0.6008	1.2916	9.7159	38.804	119.98
90	0.6755	1.5413	10.863	43.038	133.32
100	0.7501	1.6106	11.997	47.182	146.61

* Units are in atmospheres pressure.

Figure 3-4. Pressurization of a Drained-Sealed "Worst Cast" MCO 277 kg Sludge.*

Figure 3-4 shows that for a sealed and flooded MCO containing 277 kg of sludge, the nominal 10 atmosphere limit is reached in 1486 hours at 10 °C, 695 hours at 20 °C, 82.5 hours at 50 °C, 18.7 hours at 75 °C, and within 5.2 hours at 100 °C.

4.0 COLD VACUUM DRYING

The CVD process is designed to remove the bulk of the water from the MCO, prior to shipping and storage in the CSB. The drained MCO will be evacuated by roughing vacuum pumps.

As the pressure drops, the free water begins to boil when the system pressure reaches the partial pressure of water at the MCO temperature. The free water rapidly cools as it boils until finally, an equilibrium temperature is achieved based upon the heat flow into the sample and the water vapor removal rate of the vacuum system. This process also establishes an equilibrium system pressure.

4.1 RADIOLYTIC GAS PRODUCTION FOR UNCORRODED FUEL DURING CVD

At the beginning of the CVD process, the radiolytic gas generation rate will not be different from the drained MCO case previously considered. By the end of the CVD process, the radiolytic rate will have slowed due to the removal of the free water. These radiolytic rates are trivial and do not significantly affect the MCO during the CVD operation.

4.2 GAS GENERATION FROM CORRODED FUEL DURING CVD

The radiolytic gas production does not vary significantly for corroded fuel from the uncorroded case. The discussion from Section 4.1 can therefore be applied to corroded fuel.

4.2.1 Hydrogen Generation from Water Reaction with Uranium Metal During CVD

To calculate the hydrogen generation rate, it is necessary to know the water vapor pressure. An MCO preheated to 75 °C, possesses a water vapor partial pressure of 283.1 torr or 38.42 kPa. As the water boils under vacuum, the liquid water temperature rapidly cools until the rate at which heat is supplied to the system equals the rate at which heat is removed by the boiling process. It has been estimated that equilibrium will be achieved at approximately 90 torr or 11.96 kPa. This corresponds to an internal temperature of 50 °C.

It is expected that the water vapor partial pressure would be maintained at approximately 90 torr until the free water is almost exhausted. The pressure would then drop precipitously to the lowest pressure (5 torr) maintained by the vacuum pump.

Equation 4 (Ritchie 1981) allows one to calculate the uranium corrosion rate under deoxygenated conditions for liquid water or a 100% RH oxygen-free atmosphere. R. J. Pearce gives an equation (Pearce 1989):

$$\text{Log } K = 4.33 - 2144/T + 0.5 \text{ Log}[P] \quad (27)$$

giving the reaction rate constant as a function of both temperature and water partial pressure in kPa. The oxygen fixation rate during CVD is then:

$$\begin{aligned} \text{wt gain in moles O}_2 / \text{hr/cm}^2 &= 10 \cdot K / 32000 \text{ mg/gmole} \\ &= (10 \cdot 10^{(4.33 - 2144/T + 0.5 \text{ Log}[P])}) / 32000 \end{aligned} \quad (28)$$

The hydrogen generation rate from uranium metal is then twice that predicted from Equation 18.

During CVD, there is typically more interest in the hydrogen generation rate than the total MCO pressure, since the MCO will not normally pressurize. If, however, some off-normal event plugs the lines, it is possible for the system to pressurize. Therefore, the data are presented in the same format as for the rest of this document. The number of gmol/cm² of hydrogen for the first hour give the initial generation rate.

Table 4-1 and Figure 4-1, give the hydrogen generation rates for corroded fuel during CVD.

4.2.2 Hydrogen Generation from Water Reaction with Uranium Hydride During CVD

The equation derived previously for the reaction rate of water with uranium hydride applies to liquid water and to water vapor at 100% RH.

The water partial pressure is not expected to decrease below 100% RH during CVD, until the very end of the process step. For this reason, the same equation will be used to predict hydrogen generation rates from uranium hydride as was used previously. The results of these calculations are the same as found for the flooded MCO and can be found in Table 2-5 and Figure 2-3.

4.2.3 Fission Product Gas Release During CVD

Fission product gases diffuse slowly from the oxide corrosion product. The release rate is too slow to significantly affect the MCO pressure.

Table 4-1. Hydrogen Generation from Water Reaction with Uranium Metal During CVD.*

Time Hours	Temp (°C)				
	10	20	50	75	100
1	3.96E-07	9.91E-07	1.09E-05	5.76E-05	2.41E-04
5	1.98E-06	4.96E-06	5.44E-05	2.87E-04	1.20E-03
10	3.96E-06	9.91E-06	1.09E-04	5.76E-04	2.41E-03
20	7.93E-06	1.98E-05	2.17E-04	1.15E-03	4.83E-03
30	1.19E-05	2.97E-05	3.26E-04	1.73E-03	7.24E-03
40	1.59E-05	3.96E-05	4.35E-04	2.30E-03	9.65E-03
50	1.98E-05	4.96E-05	5.43E-04	2.88E-03	1.21E-02
60	2.38E-05	5.95E-05	6.52E-04	3.45E-03	1.45E-02
70	2.77E-05	6.94E-05	7.61E-04	4.03E-03	1.69E-02
80	3.17E-05	7.93E-05	8.69E-04	4.60E-03	1.93E-02
90	3.57E-05	8.92E-05	9.78E-04	5.18E-03	2.17E-02
100	3.96E-05	9.91E-05	1.09E-03	5.76E-03	2.41E-02

* Units are in atmospheres pressure.

Figure 4-1. Hydrogen From Water Reaction with Uranium Metal in MCO during CVD.

4.2.4 Helium Gas Release During CVD

Helium gas forms from the alpha particles emitted by heavy metals. This helium is not a significant factor in the pressurization of the MCO.

4.2.5 Water Vapor Pressure Increase During CVD

The partial pressure of water increases as the temperature of the MCO is raised. The increase in the number of gmoles of water vapor as the MCO heats from 10 °C to any final temperature was added to the other gases to establish the final MCO pressure.

4.2.6 Pressurization of a Sealed MCO During CVD

During normal operations, the MCO should not be sealed during CVD, but for those off-normal events where sealing could occur, the pressures will be the same as for the drained MCO case.

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5.0 SHIPPING AND STAGING

5.1 RADIOLYTIC GAS PRODUCTION

After completion of CVD, all free water is postulated to be gone and the residual water within oxides is postulated to not exceed 10% by volume, or approximately 2-3 weight %.

Under these conditions, the radiolytic hydrogen generation rate for the 2.7 kg case is $1.366\text{E-}09$ moles per second from Table 2-2 and for the 16 kg case is $9.687\text{E-}09$ moles per second.

The total hydrogen and oxygen accumulating as a function of time are shown in Table 5-1 and Figure 5-1. These radiolytic gas generation rates are only consequential during long-term storage and are not consequential during shipping and staging.

After two years staging time, and in the absence of appropriate getters, radiolytic hydrogen and oxygen will accumulate in the following amounts:

- For 2.7 kg of sludge, $\text{H}_2 = 0.1$ and $\text{O}_2 = 0.05$ moles
- For 16 kg of sludge, $\text{H}_2 = 0.6$ and $\text{O}_2 = 0.3$ moles
- For 277 kg of sludge $\text{H}_2 = 6.8$ and $\text{O}_2 = 3.4$ moles.

Only 0.9 moles of O_2 are required to reach the 5% flammability limit.

Table 5-1. Radiolytic Gas Generation During Shipping and Staging.

Time Hours	2.7 kg Sludge		16 kg Sludge		277 kg Sludge	
	Oxygen	Hydrogen	Oxygen	Hydrogen	Oxygen	Hydrogen
0	2.94E-06	5.89E-06	1.74E-05	3.488E-05	0.0001942	0.0003884
1	2.943E-5	5.886E-5	0.000174	0.0003488	0.0019422	0.0038844
2	0.000294	0.000589	0.001743	0.003488	0.019422	0.038844
3	0.002943	0.005886	0.01744	0.03488	0.19422	0.38844
4	0.02943	0.05886	0.1744	0.3488	1.9422	3.8844
5	0.2943	0.5886	1.744	3.488	19.422	38.844
6	2.943	5.886	17.44	34.88	194.22	388.44

Figure 5-1. Radiolytic Gas Generation During Shipping and Staging.

5.2 CHEMICAL GAS GENERATION DURING SHIPPING AND STAGING

5.2.1 Estimate of Water Partial Pressure During Shipping

For the worst-case MCOs with high surface area, the chemical gas generation rate will depend upon the water vapor partial pressure and the temperature. Potential water sources are free water remaining through incomplete CVD; physically absorbed water on surfaces, free water contained in crystal defects within the oxide lattice, chemically bound water as in hydrates, and chemically bound hydroxide ions both on the surface and within the oxide lattice.

If all free water is removed, as should be the case during CVD, the water vapor partial pressure will be determined by the remaining species.

The types and species of hydrates within the MCO are then considered. These are determined by the types of oxides found in the MCO, and the activities of both oxygen and water in equilibrium with these oxides. Three oxides are found in relatively large abundance in the K-East pool: iron oxide, aluminum oxide, and uranium oxide. The origins of these oxides are as follows:

Structural iron reacts with water as in:

The relatively soluble ferrous ions are eventually oxidized to ferric ions by dissolved oxygen:

Ferric hydroxide settles out of solution as an insoluble precipitate covering all objects including fuel assemblies. Ferric hydroxide typically forms a trihydrate ($\text{Fe}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$) at ambient temperature.

Aluminum metal exists in the pool as aluminum cladding upon a few single pass fuel assemblies, and many of the canisters containing fuel assemblies were made of aluminum. This aluminum corrodes in water to form a tightly adherent aluminum oxide $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$ at ambient temperature. Over time, this aluminum oxide can flake off and be carried by water currents to every available surface within the K-East Pool.

Uranium metal corrodes in water to form UO_2 :

Uranium dioxide does not form hydrates; however, a higher oxide UO_3 does. The chemistry of forming the higher oxide is then considered to see if this higher oxide contributes significant amounts of hydrated water to the MCO.

Uranium in UO_2 carries a formal charge of +4. Uranium can also lose two more electrons and go to the +5 and +6 formal oxidation states, as in UO_2^+ and UO_2^{+2} . These higher oxidation states can form oxides as in $(\text{UO}_2)_2\text{O}$ or U_2O_5 (not identified as a pure substance) and $(\text{UO}_2)\text{O}$ or UO_3 . These oxides can be found in a continuum of abundances depending upon the oxygen potential; however, only four pure crystalline phases have been reported: UO_2 , U_4O_9 , U_3O_8 , and UO_3 . Of all of these oxides, only UO_3 has been reported to form a hydrate ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$) at ambient temperature.

The next and most important question is, which of these oxides and in what abundance will it be incorporated into the MCO?

Because only uranium oxides adhere to the uranium metal, it is believed that essentially all of the contaminant aluminum and iron oxides will be removed.

Reasons for this assumption are:

- Exterior coatings of iron and aluminum oxides on the fuel assemblies should be removed by the handling and cleaning processes.
- Both iron and aluminum oxides should be removed from any cavities with large openings by the handling and cleaning processes.
- Aluminum oxides and ferric oxides should not be found behind pinholes and narrow cracks in the cladding. Aluminum oxides are moved as micron-sized particles by water currents from their place of origin, to deposition on or in the fuel assemblies. Narrow cracks or pinholes restrict the size and velocity of water currents carrying aluminum oxide particles. Secondly, the space behind pinholes or narrow cracks are filled with uranium oxides and contain little void space for foreign particles.

Ferrous iron could diffuse through pinholes or narrow cracks, but is not expected to precipitate beneath the cladding due to the reducing environment established by the uranium metal and the slow diffusion of oxygen through the narrow cladding openings.

The MCO is therefore expected to contain principally uranium metal and uranium oxides. It is known that only UO_2 is formed upon reaction of uranium metal with water. Oxygen is required to form the higher oxides. When very small amounts of oxygen are present, UO_{2+x} is formed where x typically varies from 0.01 to 0.1. In water that is in direct contact with the atmosphere, x usually varies from 0.2 to 0.4, and the crystal habit is still that of the lower oxides. It is seen therefore that $\text{UO}_3 \cdot 2\text{H}_2\text{O}$ can only form on the surface of these oxides and is not contained within the crystalline lattice. Hydrated UO_3 is typically yellow in color, is easily spotted, and has been found only in trace quantities on uranium oxides from oxygenated waters. The density of hydrated UO_3 is very low and it is easily lifted and removed from surfaces during the cleaning processes.

The entire thrust of this discussion has been to explain why hydrated oxides exist in the floor sludges, but significant quantities of hydrated oxides in the MCO are not expected.

Physically bound water can be found at the oxide surfaces. The bonding energy of physically bound water is typically 10 kcal/mol or less.

Much of the physically bound water is expected to be removed by the CVD process, but it is not certain that it will be quantitatively removed.

Water will exist at defect sites within the oxide lattice. This water will only migrate out of the oxide readily at higher temperatures than that experienced during CVD. Neither physically bound water nor defect water are expected to be large water sources.

Chemically bound hydroxides are expected to form at the surface of UO_2 . The normal uranium-oxygen bond angles cannot be maintained at the surface, and structures such as highly strained U-O-U, are found. These react readily with water to form:

Once formed, these hydroxides can migrate into the oxide lattice. The rate of migration is greatly enhanced by an electrical field, as long as the oxide is attached to the metal. When the oxide spalls off, the rate of migration is slowed tremendously.

The bonding energy for these hydroxides is significantly higher than for hydrates involving whole water molecules. As a consequence, the temperatures required to completely remove these hydroxide moieties are typically in the range of 700 to 1000 °C. For this reason, the partial pressure of water from chemically bound hydroxide moieties is so low that they are not of concern for the chemical production of gases during shipping and storage.

It is estimated that the water vapor partial pressure will be approximately 5 torr upon removal from the CVD station. This water vapor pressure is being maintained by any free water still remaining, physically absorbed water, interstitial defect water, and any chemically bound water such as waters of hydration. It is estimated that the entire residual water inventory does not exceed 10% by volume or 2% by weight of the entire oxide inventory. For the low sludge case, this would be only 54 grams of water in the MCO. It is obvious that this water inventory will be depleted by the high surface area MCO through chemical reactions with uranium metal until essentially only crystalline defect water and chemically bound hydroxides will remain through shipping and staging.

The calculations for the following tables in this section will use a partial pressure of water vapor of 1 torr. This results from the knowledge that the final water partial pressure at the end of CVD was no more than 5 torr.

After CVD and assuming all free water is removed, the residual water partial pressure will be rapidly depleted to 1 torr or less by continued reaction with the available uranium metal surface.

5.2.2 Hydrogen Generation From the Reaction of Water with Uranium Metal and Uranium Hydride

Equation 18, with a water partial pressure of 1 torr or 0.133 kPa, is used to calculate the hydrogen production rate from the reaction of water with uranium metal. The production of hydrogen from uranium hydride does not appear to change significantly for this amount of water

from the previous case, so it will be carried through unchanged. The results of these calculations are given in Table 5-2 and in Figure 5-2.

5.2.3 Fission Product Gas Release During Shipping and Staging

Fission product gases diffuse slowly from the oxide corrosion product. The release rate is too slow to significantly affect the MCO pressure.

5.2.4 Helium Gas Release During Shipping and Staging

Helium gas forms from the alpha particles emitted by heavy metals. This helium is not a significant factor in the pressurization of the MCO.

5.2.5 Water Vapor Pressure Increase During Shipping and Staging

There is such a small amount of water in the MCO during shipping and staging, that any pressure increase due to water vapor was ignored.

5.2.6 Pressurization of a Sealed MCO During Shipping and Staging

To calculate the pressure in a sealed MCO during shipping and staging, the following parameters are assumed:

- Reactive metal surface area = 127,233 cm² as described for a “worst-case” MCO. (WHC 1996b)
- Sludge mass = 16 kg
- Headspace = 500 liters
- Temperature = 10 °C
- Radiation energy absorption as described in Tables 2-1 and 2-2
- Time = 1 hour

Table 5-2. Chemical Hydrogen Generation in a Sealed MCO
During Shipping and Staging.*

Time Hours	Temp (°C)				
	10	20	50	75	100
1	1.47E-07	2.72E-07	1.37E-06	4.30E-06	1.17E-05
5	7.37E-07	1.36E-06	6.84E-06	2.14E-05	5.73E-05
10	1.47E-06	2.71E-06	1.37E-05	4.25E-05	1.12E-04
20	2.95E-06	5.43E-06	2.72E-05	8.40E-05	2.16E-04
30	4.42E-06	8.14E-06	4.07E-05	1.25E-04	3.15E-04
40	5.89E-06	1.08E-05	5.42E-05	1.65E-04	4.09E-04
50	7.36E-06	1.36E-05	6.75E-05	2.04E-04	5.02E-04
60	8.83E-06	1.62E-05	8.09E-05	2.42E-04	5.93E-04
70	1.03E-05	1.90E-05	9.41E-05	2.80E-04	6.82E-04
80	1.18E-05	2.16E-05	1.07E-04	3.18E-04	7.71E-04
90	1.32E-05	2.44E-05	1.20E-04	3.55E-04	8.60E-04
100	1.47E-05	2.70E-05	1.33E-04	3.91E-04	9.49E-04
1000	1.46E-04	2.65E-04	1.20E-03	3.46E-03	8.83E-03

* Units are in moles H₂/cm³

Figure 5-2. Hydrogen Generation in the MCO During Shipping and Staging.

The rate of pressurization of a MCO during shipping and staging can be modeled by summing in the following manner:

Pressure increase in atmospheres =

$$\begin{aligned}
 & (\text{gmols radiolysis gases}) + (\text{reactive metal surface area} * (\text{gmols H}_2 \text{ metal}/\text{cm}^2 \\
 & \quad + \text{gmols H}_2 \text{ UH}_3/\text{cm}^2)) * 0.08205 * 283.16/44 \quad (33) \\
 & = ((0.0000000096876 * 3600 * 1) + (127233 * (1.307\text{E-}07) + (0.00029422 * 16)) \\
 & \quad * 0.08205 * 283.16/44 \\
 & = 0.00099322 \text{ atm.}
 \end{aligned}$$

The remaining calculations are found in Table 5-3, 5-4, 5-5 and Figure 5-3, 5-4, 5-5.

5.2.7 Reduction of Inventory of Reactants

For the calculations performed in this section, it has been assumed that the water partial pressure remains constant at 1 torr and that the inventory of uranium hydride remains constant in the oxide corrosion product. In fact, either can be depleted. Water has been steadily depleted by draining, CVD, and chemical reaction with the exposed uranium metal. Uranium metal is a more powerful water "getter" than any other material and will eventually react all of the water in the system.

The reaction rates of water with uranium metal at water pressures below 1 torr cannot be accurately modeled with existing late data, and we have chosen to remain at that level for the sake of conservatism. Because of the depletion of water, the higher pressures predicted in preceding graphs in this section are not actually achievable.

It was estimated at the beginning of this section that no more than 3 wt % of water would survive in the sludge after CVD. For 16 kg of sludge, this limits the pressure increase in a given MCO to 1.65 atmospheres. Larger amounts of sludge could yield proportionately more pressure.

Since water reacts with uranium metal to form uranium hydride, and with uranium hydride to form uranium oxide, uranium hydride exists in secular equilibrium. Once the supply of water is exhausted, it becomes quite possible to oxidize the remaining hydride. The oxidation rate with oxygen gas is extremely fast. Data from S. Robinson shows complete oxidation (at 0-100 °C) of samples of pure uranium hydride in 200 torr oxygen within 6 minutes (Robinson 1996).

Table 5-3. Pressure Increase for a MCO During Shipping and Staging.

Time Hours	Temp (°C)				
	10	20	50	75	100
1	8.10E-04	0.0024	0.0157	0.0484	0.1241
5	4.05E-03	0.0083	0.0445	0.1364	0.3543
10	8.10E-03	0.0157	0.0805	0.2459	0.6373
20	0.0162	0.0305	0.1525	0.4630	1.1903
30	0.0243	0.0453	0.2242	0.6775	1.7281
40	0.0324	0.0600	0.2957	0.8903	2.2594
50	0.0405	0.0748	0.3671	1.1017	1.3117
60	0.0486	0.0896	0.4383	1.3117	3.3104
70	0.0566	0.1043	0.5094	1.5206	3.8324
80	0.0648	0.1191	0.5804	1.7286	4.3531
90	0.0728	0.1338	0.6513	1.9357	4.8730
100	0.0810	0.1486	0.7220	2.1419	5.3923
1000	0.8033	1.4533	6.7617	20.178	52.022

* Units are in atmospheres pressure.

Figure 5-3. Pressure Increase for a "Worst-Case" MCO Containing 2.7 kg of Sludge During Shipping and Staging.

Figure 5-3 shows for a sealed "worst case" MCO containing 2.7 kg of sludge, the nominal 10 atmosphere pressure limit is reached in 12843 hours at 10°C, in 7090 hours at 20°C, in 1485 hours at 50°C, in 492 hours at 75°C, and in 189 hours at 100°C.

Table 5-4. Pressure Increase for a “Worst-Case” MCO Containing
16 kg Sludge During Shipping and Staging.

Time Hours	Temp (°C)				
	10	20	50	75	100
1	9.93E-04	0.0028	0.0183	0.0584	0.1553
5	4.97E-03	0.0103	0.0575	0.1849	0.4985
10	9.93E-03	0.0196	0.1063	0.3399	0.8994
20	0.0199	0.0382	0.2031	0.6395	1.6240
30	0.0298	0.0568	0.2986	0.9232	2.2588
40	0.0397	0.0754	0.3932	1.1970	2.8550
50	0.0496	0.0939	0.4869	1.4623	3.4252
60	0.0595	0.1124	0.5798	1.7200	3.9780
70	0.0693	0.1309	0.6718	1.9708	4.5194
80	0.0793	0.1494	0.7631	2.2159	5.0530
90	0.0892	0.1679	0.8536	2.4556	5.5814
100	0.0991	0.1863	0.9436	2.6907	6.1063
1000	0.9533	1.7117	7.0912	20.718	52.719

Figure 5-4. Pressure Increase for a "Worst-Case" MCO Containing 16 kg of Sludge During Shipping and Staging.

Figure 5-4 shows that for a sealed MCO containing 16 kg of sludge, the nominal 10 atmosphere limit is reached in 12373 hours at 10 °C, 6839 hours at 20 °C, 1436 hours at 50 °C, 465171 hours at 75 °C, and within 175 hours at 100 °C.

Table 5-5. Pressure Increase for a "Worst-Case" MCO Containing 277 kg Sludge During Shipping and Staging.

Time Hours	Temp (°C)				
	10	20	50	75	100
1	0.0046	0.0102	0.0692	0.2536	0.7680
5	0.0228	0.0474	0.3105	1.1371	3.3295
10	0.0457	0.0938	0.6083	2.1867	6.0456
20	0.0914	0.1863	1.1903	4.1065	10.143
30	0.1369	0.2783	1.7497	5.7500	12.685
40	0.1824	0.3698	2.2934	7.2244	14.557
50	0.2277	0.4610	2.8220	8.5495	15.980
60	0.2730	0.5519	3.3360	9.7426	17.104
70	0.3182	0.6424	3.8358	10.820	18.027
80	0.3633	0.7326	4.3219	11.794	18.816
90	0.4083	0.8223	4.7946	12.677	19.515
100	0.4533	0.9118	5.2545	13.484	20.154
1000	3.8869	6.6305	13.233	31.5253	66.784

* Units are in atmospheres pressure.

Figure 5-5. Pressure for a "Worst-Case" MCO Containing 277 kg of Sludge
During Shipping and Staging.

Figure 5-5 shows that for a sealed MCO containing 277 kg of sludge, the nominal 10 atmosphere limit is reached in 3392 hours at 10 °C, 1671 hours at 20 °C, 256 hours at 50 °C, 62.2 hours at 75 °C, and within 19.6 hours at 100 °C.

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6.0 GAS GENERATION DURING HOT VACUUM DRYING (HVD) AND CONDITIONING

Gas generation during HVD and conditioning is not expected to be significant. Although these process steps contain the maximum temperatures, all of the free and physically bound water has been eliminated prior to this step, and a good share of the chemically bound water will have also been eliminated due to radiolysis and chemical reaction during staging.

During hot vacuum drying, most of the remaining chemically bound water will be released as the MCO is heated to 300 °C. This is not expected to pressurize the MCO because the system is being actively vacuum pumped. By the time the MCO reaches Hot Conditioning, the water inventory is so low that no significant pressurization will occur.

Pressurization from radiolysis gases is not expected to be significant during HVD or Hot Conditioning because of the short time interval during these process steps. Calculations substantiating these statements will be released in the near future.

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7.0 GAS GENERATION DURING LONG-TERM STORAGE

7.1 RADIOLYTIC GAS GENERATION DURING LONG-TERM STORAGE

Radiolytic gases are generated during long-term storage. After hot vacuum drying and conditioning, the only water in the MCO should be chemically adsorbed water. This water can either be dissociated into hydroxide ions on the oxide surfaces, or can be molecular water associated with hydrate forming metal ions. Although this water is chemically bound and is very stable, radiolysis is a very high energy event and releases both hydrogen and oxygen gases.

The rate of radiolysis depends upon the radiation fields, the specific surface phenomena occurring, and the quantity of water absorbing energy. The rate equations are:

$$dn/dt = k_1 * G_1 * N + k_2 * G_2 * N + k_3 * G_3 * N \quad (34)$$

$$= (k_1 * G_1 + k_2 * G_2 + k_3 * G_3) * N \quad (35)$$

where:

k_1 = the alpha energy absorbed per gmole of water

k_2 = the beta energy absorbed per gmole of water

k_3 = the gamma energy absorbed per gmole of water

G_1 = # H_2 /100 ev alpha

G_2 = # H_2 /100 ev beta

G_3 = # H_2 /100 ev gamma

all of the preceding constants can be combined mathematically into one large constant K, so with the definition:

N = the number of gmoles of water acting as radiolysis targets

This gives the easily recognizable first order equation whose solution is:

$$N/N_0 = \text{Exp}[-Kt]$$

This equation can be used to calculate the amount of water remaining at any time t .

Calculation of K

From Table 2-1:

$$k_1 * G_1 = (3600 \text{ sec/hour}) * (1.62E-05 \text{ J/g-sec}) * (6.2112E+18 \text{ ev/J}) * (18 \text{ g/m}) * (3 \text{ molecules/100 ev}) * (1 \text{ mole}/6.023E+23 \text{ molecules}) \quad (36)$$

$$= 3.24752\text{E-}07/\text{hour}$$

$$k_2 * G_2 = (3600 \text{ sec/hour}) * (6.9\text{E-}06 \text{ J/g-sec}) * (6.2112\text{E+}18 \text{ ev/J}) * \\ (18 \text{ g/m}) * (20 \text{ molecules/100 ev}) * (1 \text{ mole}/6.023\text{E+}23 \text{ molecules}) \quad (37)$$

$$= 1.38454\text{E-}06/\text{hour}$$

$$k_3 * G_3 = (3600 \text{ sec/hour}) * (6.9\text{E-}06 \text{ J/g-sec}) * (6.2112\text{E+}18 \text{ ev/J}) * \\ (18 \text{ g/m}) * (20 \text{ molecules/100 ev}) * (1 \text{ mole}/6.023\text{E+}23 \text{ molecules}) \quad (38)$$

$$= 9.30155\text{E-}07/\text{hour}$$

so

$$K = 2.63143\text{E-}06/\text{hour} \quad (39)$$

Now the fraction of water remaining after any time t can be defined

$$N/N_0 = \text{Exp}[-2.63143\text{E-}06 * t] \quad (40)$$

This equation is shown graphically in Figure 6-1.

Figure 7-1. Water Remaining in MCO During Long-term Storage.

Now specific cases can be calculated and are included in Table 6-1 and Figures 6-2 and 6-3.

Table 7-1. Water Radiolyzed During Long-term Storage.*

Wt% Water	Log Hours	2.7 kg	16 kg	277 kg
0.5	0	1.9736E-06	1.1695E-05	2.0247E-04
	1	1.9735E-05	1.1695E-04	2.0247E-03
	2	1.9733E-04	1.1694E-04	2.0221E-02
	3	1.9710E-03	1.1680E-02	2.0221E-01
	4	1.9478E-02	1.1680E-01	1.9983E-00
	5	1.7353E-01	1.0283E-00	1.7802E+01
	6	6.9602E-01	4.1245E-00	7.1406E+01
1.5**	0	5.9207E-06	3.5086E-05	6.0742E-04
	1	5.9206E-05	3.5085E-04	6.0741E-03
	2	5.9199E-04	3.5081E-03	6.0734E-02
	3	5.9129E-03	3.5040E-02	6.0662E-01
	4	5.8435E-02	3.4628E-01	5.9950E-00
	5	5.2058E-01	3.0849E-00	5.3407E+01
	6	2.0881E-00	1.2374E+01	2.1422E+02

* Units are in gmoles water radiolyzed, which correspond numerically to gmoles of hydrogen produced. The number of gmoles of oxygen produced are half the number of gmoles of hydrogen.

** Even though only 0.5 weight % is expected after hot vacuum conditioning, a 1.5 weight % water case was added to extend the range of water considered during long-term storage.

Figure 7-2. Water Radiolyzed During Long-term Storage
From Sludge Containing 0.5 wt% Water.

* Initial wt% water was 0.5.

Figure 7-3. Water Radiolyzed During Long-term Storage
From Sludge Containing 1.5 wt% Water.

* 1.5 wt% water.

Pressures from this radiolysis process will depend upon the equilibrium MCO temperature and can be calculated from the number of moles radiolyzed.

7.2 CHEMICAL GAS GENERATION

It is certainly possible for gases to be generated chemically during long-term storage if any water molecules strike uranium metal. It is expected that the rate will be very slow as the partial pressure of water in the MCO is expected to be extremely low. Since we cannot model either the water pressure or the reaction rate accurately, it is conservatively assumed that if exposed uranium metal is available and chemical reactions are possible, all of the available hydrogen on water will be transformed to hydrogen gas. Some fraction of that hydrogen (possibly all) will be converted to uranium hydride by further chemical reactions over the long storage time.

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