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Revised approach to treatment of nitric acid condensation and determination of amount of zirconium. Increased level of detail in calculations throughout.

ANALYSIS OF DEGRADATION DUE TO WATER AND GASES IN MPC

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1. Purpose

This analysis is prepared by the Mined Geologic Disposal System (MGDS) Waste Package Development Department in response to a request received via a QAP-3-12 Design Input Data Request (Ref. 5.16) from WAST Design (formerly MRS/MPC Design). The request is to provide:

- 1) *Dryness requirements for the MPC cavity environment after loading and closure operations at the Purchaser site.*

The objective of this analysis is to provide a response to the foregoing request. The analysis treats nominal loading conditions only, not accidents or accident conditions. In particular, leaky (waterlogged) fuel rods are not considered. The purpose of this analysis is to provide the basis for the response. The response is stated in Section 8 herein.

2. Quality Assurance

The quality assurance (QA) program applies to this analysis. This analysis focuses on compatibility of certain Multi-Purpose Canister (MPC) design features that interface with the MGDS. These features could potentially affect the proper functioning of the MGDS waste package; the waste package has been identified as an MGDS Q-List item that is important to safety and waste isolation (Ref. 5.15). This work is covered by Activity Evaluation *Analyze Material and Performance Information and Data in Support of Waste Package/Engineered Barrier Segment Development* (Ref. 5.2). This QAP-2-0 evaluation determined such activities to be subject to the requirements of the *Quality Assurance Requirements and Description* (Ref. 5.3). Applicable procedural controls are listed in the evaluation.

3. Method

The *Multi-Purpose Canister (MPC) Subsystem Design Procurement Specification* (Ref. 5.1) specifies the amount of water and gases that may be present in an MPC. This design analysis uses the current revision of those specifications to determine limits on internal degradation of an MPC.

The design method is to (1) analyze the wording of the specification to determine limits on the quantity of water and the composition of the gases that will be present in the MPC, and (2) determine limits on amount of degradation that could result from the available gases. The calculation is made for an MPC with a capacity of 21 pressurized-water reactor assemblies (PWRs), and qualitative arguments are used to show that additional, detailed calculations for other types of MPC are unnecessary.

This method will determine whether the specification is adequate but will not provide an upper bound on the allowable amount of water.

4. Design Inputs

4.1 Design Parameters

4.1.1. Design inputs come from the current revision of the *Multi-Purpose Canister (MPC) Subsystem Design Procurement Specification* (Ref. 5.1). Relevant specifications are as follows:

Section 5.1.1.7.1.A: *The MPC shall be capable of being drained, vacuum-dried, and back-filled with an inert gas after SNF loading.* This specification is taken to mean that the MPC may be analyzed as if it is actually vacuum-dried and back-filled with an inert gas.

Section 5.1.1.7.1.C: *Vacuum pumps used to vacuum dry the interior of the MPC shall be of such a design as to achieve an absolute pressure of 3 millibar. Operating procedures for the use of such vacuum pumps shall include a test that this pressure be maintained for a ten minute period. The sensitivity of this test shall be sufficient to demonstrate by calculation that the residual water content of the MPC interior is less than 0.25 volume percent.*

4.1.2. The following physical, chemical, thermal, and geometric data are used as needed in the analysis:

gas constant: 8.31451 J/mol·K (Ref. 5.7)

molar mass of (atomic) hydrogen: 1.00794 g/mol (Ref. 5.7)

molar mass of (atomic) nitrogen: 14.00674 g/mol (Ref. 5.7)

molar mass of (atomic) oxygen: 15.9994 g/mol (Ref. 5.7)

molar mass of chromium: 51.9961 g/mol (Ref. 5.7)

molar mass of iron: 55.847 g/mol (Ref. 5.7)

molar mass of nickel: 58.6934 g/mol (Ref. 5.7)

molar mass of zirconium: 91.224 g/mol (Ref. 5.7)

molar mass of molybdenum: 95.94 g/mol (Ref. 5.7)

vapor pressure of water at 295 K: 2618.4 Pa (Ref. 5.6, p. E-23, calculated by linear interpolation as follows: 295 K = 21.85 °C = 71.33 °F. From Ref. 5.6, the vapor pressure of water is 0.37549 psia at 71 °F and 0.38844 psia at 72 °F. By linear interpolation, the vapor pressure of water at 295 K is $((1 - 0.33) \times 0.37549 + 0.33 \times 0.38844)$ psia = 0.37976 psia = $(0.37976 \times 6894.757)$ Pa = 2618.4 Pa.)

density of saturated liquid water at 295 K: 997.87 kg/m^3 (Ref. 5.6, p. E-23, calculated by linear interpolation as follows: As shown previously, $295 \text{ K} = 71.33 \text{ }^\circ\text{F}$. From Ref. 5.6, the specific volume of saturated liquid water is $0.016052 \text{ lbm/ft}^3$ at $71 \text{ }^\circ\text{F}$ and $0.016054 \text{ lbm/ft}^3$ at $72 \text{ }^\circ\text{F}$. By linear interpolation, the density of saturated liquid water at 295 K is $1 / (((1 - 0.33) \times 0.016052 + 0.33 \times 0.016054) \text{ lbm/ft}^3) = 62.295 \text{ lbm/ft}^3 = (62.295 \times 16.01846) \text{ kg/m}^3 = 997.87 \text{ kg/m}^3$.)

enthalpy of vaporization for water at 295 K: $2,448,300 \text{ J/kg}$ (Ref. 5.6, p. E-23, calculated by linear interpolation as follows: As shown previously, $295 \text{ K} = 71.33 \text{ }^\circ\text{F}$. From Ref. 5.6, the enthalpy of vaporization for water is 1053.5 BTU/lbm at $71 \text{ }^\circ\text{F}$ and 1052.9 BTU/lbm at $72 \text{ }^\circ\text{F}$. By linear interpolation, the enthalpy of vaporization for water at 295 K is $((1 - 0.33) \times 1053.5 + 0.33 \times 1052.9) \text{ BTU/lbm} = 1053.3 \text{ BTU/lbm} = (1053.3 \times 2324.444) \text{ J/kg} = 2,448,300 \text{ J/kg}$.)

heat capacity at constant pressure of liquid water at 295 K: $4179 \text{ J/kg}\cdot\text{K}$ (Ref. 5.6, p. E-24, calculated by linear interpolation as follows: As shown previously, $295 \text{ K} = 71.33 \text{ }^\circ\text{F}$. From Ref. 5.6, the heat capacity at constant pressure of liquid water (atmospheric pressure) is $1.000 \text{ BTU/lbm}\cdot^\circ\text{F}$ at $60 \text{ }^\circ\text{F}$ and $0.998 \text{ BTU/lbm}\cdot^\circ\text{F}$ at $80 \text{ }^\circ\text{F}$. By linear interpolation, the enthalpy of vaporization for water at 295 K is $((80 - 71.33) / (80 - 60)) \times 1.000 + ((71.33 - 60) / (80 - 60)) \times 0.998 \text{ BTU/lbm}\cdot^\circ\text{F} = 0.9989 \text{ BTU/lbm}\cdot^\circ\text{F} = (0.9989 \times 4184) \text{ J/kg}\cdot\text{K} = 4179 \text{ J/kg}\cdot\text{K}$.)

vapor pressure of water at the triple point ($273.16 \text{ K} = 0.01 \text{ }^\circ\text{C} = 32.018 \text{ }^\circ\text{F}$: $0.08865 \text{ psia} = (0.08865 \times 6894.757) \text{ Pa} = 611.22 \text{ Pa}$ (Ref. 5.6, pp. E-23, F-131)

mole fraction of oxygen in dry air: 0.2095 (Ref. 5.6, p. F-208)

mole fraction of nitrogen in dry air: 0.7809 (Ref. 5.6, p. F-208)

density of zirconium: $6.49 \text{ g/cm}^3 = 6490 \text{ kg/m}^3$ (Ref. 5.6, p. B-165)

density of Type 316L stainless steel: $8 \text{ Mg/m}^3 = 8000 \text{ kg/m}^3$ (Ref. 5.12)

nominal composition of Type 316L stainless steel (by mass): 17% Cr, 12% Ni, 2.5% Mo, and balance (68.5%) Fe. Carbon (0.03% maximum) is neglected. (Ref. 5.12)

mass of initial uranium in fuel assembly (Babcock and Wilcox 15×15 / B&W Fuel Company Mark B4): 464.5 kg (output from sec. 6)

inside radius of shell of 21 PWR MPC: $(58.3 / 2) \text{ in.} = 0.74041 \text{ m}$ (Ref. 5.11)

inside length of shell of 21 PWR MPC: $180 \text{ in.} = 4.572 \text{ m}$ (Ref. 5.11)

edge length for square opening in basket fuel channels for 21 PWR MPC: $8.8 \text{ in.} = 0.2235 \text{ m}$ (Ref. 5.11)

length of basket fuel channels for 21 PWR MPC: $160 \text{ in.} = 4.064 \text{ m}$ (Ref. 5.11)

thickness of 21 PWR MPC shell: $1 \text{ in.} = 25.4 \text{ mm}$ (Ref. 5.11)

thickness of inside basket cladding for 21 PWR MPC: $0.25 \text{ in.} = 6.35 \text{ mm}$ (Ref. 5.11)

thickness of outside basket cladding for 21 PWR MPC: $0.094 \text{ in.} = 2.39 \text{ mm}$ (Ref. 5.11)

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outside diameter of fuel rod (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4):
0.43 in. = 10.922 mm (output from sec. 6)

overall length of fuel rod (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4):
153.68 in. = 3903.47 mm (output from sec. 6)

thickness of fuel cladding (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4):
0.0265 in. = 0.673 mm (output from sec. 6)

number of fueled positions (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4):
208 (output from sec. 6)

vapor pressure of HNO_3 and H_2O over solutions of HNO_3 in H_2O :

mass fraction HNO_3	vapor pressure of HNO_3 , mm Hg, at		vapor pressure of H_2O , mm Hg, at	
	20 °C	25 °C	20 °C	25 °C
0.2	--	--	15.2	20.6
0.25	--	--	14.2	19.2
0.3	--	--	13.2	17.8
0.35	--	--	12.0	16.2
0.4	--	0.12	10.8	14.6
0.45	0.15	0.23	9.4	12.7
0.5	0.27	0.39	7.9	10.7
0.55	0.45	0.66	6.7	9.1
0.6	0.84	1.21	5.6	7.7
0.65	1.68	2.32	4.9	6.6
0.7	3.00	4.10	4.1	5.5
0.8	8	10.5	2.4	3.2
0.9	20	27	--	1
1	42	57	--	--

("--" indicates that no datum was given.) (Ref. 5.19)

4.2 Criteria

The analysis addresses the chemical properties of the fill gas and their effects on the function of the waste packages. Such work is a partial response to the requirements of the *Engineered*

Barrier Design Requirements Document (EBDRD) (Ref. 5.13), section 3.7.1.A: Packages for SNF and HLW shall be designed so that the in situ chemical, physical, and nuclear properties of the waste package and its interactions with the emplacement environment do not compromise the function of the waste packages or the performance of the underground facility or the geologic setting.

The analysis addresses the effects of the fill gas on oxidation/reduction reactions, corrosion, and hydriding, and the effects of radiolysis on the fill gas. Such work is a partial response to the requirements of the EBDRD, section 3.7.1.B: *The design of waste packages shall include, but not be limited to, consideration of the following factors: solubility, oxidation/reduction reactions, corrosion, hydriding, gas generation, thermal effects, mechanical strength, mechanical stress, radiolysis, radiation damage, radionuclide retardation, leaching, fire and explosion hazards, thermal loads, and synergistic interactions.*

The analysis addresses the presence of chemically reactive materials in the fill gas. Such work is a partial response to the requirements of the EBDRD, section 3.7.1.C: *The waste packages shall not contain explosive or pyrophoric materials or chemically reactive materials in an amount that could compromise the ability of the underground facility to contribute to waste isolation or the ability of the geologic repository to satisfy the performance objectives.*

The analysis addresses the presence of free liquids. Such work is a partial response to the requirements of the EBDRD, section 3.7.1.D: *The waste package shall not contain free liquids in an amount that could compromise the ability of the waste package to achieve the performance objectives relating to the containment of radioactive waste (because of chemical interactions or formation of pressurized vapor) or that could result in spillage and spread of contamination in the event of waste package perforation during the period through permanent closure.*

The analysis addresses the effect of the fill gas in degrading the ability of the MPC to maintain waste containment during transportation, emplacement, and retrieval. Such work is a partial response to the requirements of the EBDRD, section 3.7.1.E: *Waste packages shall be designed to maintain waste containment during transportation, emplacement, and retrieval.* The more recent *Mined Geologic Disposal System Requirements Document (MGDS-RD)* (Ref. 5.4) clarifies this requirement by changing "transportation" to "transportation within the MGDS".

4.3 Assumptions

To determine the amount of gas in the MPC, it is necessary to know the void volume of the MPC and the pressure/temperature ratio at the time of filling. Rather than making a single assumption about the pressure/temperature ratio, separate assumptions are made about

temperature and pressure. Specific assumptions for these quantities, with values suitable for a 21 PWR burnup credit MPC, as described in Ref. 5.11, are made in the following subsections.

The quantities assumed here do not require verification. The reasoning behind this position is as follows. The amounts of degradation calculated are for complete consumption of any reactive species in the gas, so the amount of degradation is proportional to the quantity of gas. The quantity of reactive gas present will be proportional to volume times pressure divided by temperature. The largest credible pressure and the smallest credible temperature have been chosen. The volume is realistic, but, as discussed in section 4.3.1, within a factor of two of an absolute upper bound. It is not credible, therefore, that changes in volume, pressure, and temperature could cause the amounts of degradation to be more than twice what is predicted here. As is discussed in section 7.5, the predicted amounts of degradation are extremely small, and doubling the amount of degradation does not alter the conclusions of the analysis. There is, therefore, no need to verify the numbers that are assumed here.

4.3.1 Void volume of MPC: The void volume of the MPC (that volume of that portion of the interior of the MPC that is not filled with waste or internal structure) is taken to be 4.555 m³ for an MPC with a capacity of 21 PWRs. This volume was provided by David N. Summers of the Vienna M&O and was supported by a non-QA analysis. The volume for actual MPCs may be slightly different. (For comparison, the void volume of a completely empty MPC is $\pi \cdot (0.74041 \text{ m})^2 \cdot 4.572 \text{ m} = 7.874 \text{ m}^3$. The actual void volume is smaller because of the presence of the fuel, basket, and other structures.) This assumption is used in sections 7.1, 7.3, 7.4, and 7.7.

4.3.2 Fill gas pressure for MPC: The MPC is taken to be filled to an absolute pressure of 152 kPa. This value is the same as that chosen by David N. Summers of the Vienna M&O for a non-QA analysis. It is also the highest fill pressure specified for the four types of dry storage casks studied in PNL-6365 (Ref. 5.17, p. 4). The value for actual MPCs is not expected to be higher than this because a higher initial pressure would increase the operating pressure of the MPC but would not provide any benefits. This assumption is used in sections 7.1, 7.4, and 7.7.

4.3.3 Fill gas temperature for MPC: The MPC is taken to be filled at an average gas temperature of 295 K. This is nominal room temperature. The value for actual MPCs is expected to be higher because of decay heat generated by the fuel in the MPC. Therefore, this value is conservative, since the quantities of gas will be smaller than those calculated here. This assumption is used in sections 7.1, 7.3, 7.4, and 7.7.

4.4 Codes and Standards

The method of analysis is not controlled by codes or standards. Conversion of non-SI quantities to SI is performed with conversion factors given in the American Society for

Testing and Materials standard E 380-91a, "Standard Practice for Use of the International System of Units (SI) (the Modernized Metric System)" (Ref. 5.5).

5. References

- 5.1 *Multi-Purpose Canister (MPC) Subsystem Design Procurement Specification* (DBG000000-01717-6300-00001 REV 04, August 26, 1994)
- 5.2 *Analyze Material and Performance Information and Data in Support of Waste Package/Engineered Barrier Segment Development* (BB0000000-01717-2200-00004 REV 04, September 29, 1995)
- 5.3 *Quality Assurance Requirements and Description* (DOE/RW-0333P, Rev. 4, August 4, 1995)
- 5.4 *Mined Geologic Disposal System Requirements Document* (DOE/RW-0404P, B00000000-00811-1708-00002 REV 01, DCN 01, May 1995)
- 5.5 American Society for Testing and Materials, Standard E 380-91a, "Standard Practice for Use of the International System of Units (SI) (the Modernized Metric System)" (October 3, 1991)
- 5.6 *CRC Handbook of Chemistry and Physics*, 61st ed., Robert C. Weast, editor (CRC Press, Boca Raton, FL, 1980)
- 5.7 *CRC Handbook of Chemistry and Physics*, 74th ed., David R. Lide, editor (CRC Press, Boca Raton, FL, 1993), inside front cover and p. 1-1.
- 5.8 D. T. Reed and R. A. Van Konynenburg, "Effect of Ionizing Radiation on the Waste Package Environment", in *High Level Radioactive Waste Management: Proceedings of the Second International Conference*, pp. 1396-1403 (American Nuclear Society, La Grange Park, IL, and American Society of Civil Engineers, New York, 1991)
- 5.9 American Society for Testing and Materials, Standard B 353-91, "Standard Specification for Wrought Zirconium and Zirconium Alloy Seamless and Welded Tubes for Nuclear Service" (March 26, 1991)
- 5.10 C. Pescatore, M. G. Cowgill, and T. M. Sullivan, *Zircaloy Cladding Performance Under Spent Fuel Disposal Conditions* (BNL 52235).
- 5.11 *MPC Conceptual Design Report* (A20000000-00811-5705-00002 REV 00, September 1994), vol. II.A, table 4.1.6-1, figures 4.2.1.1-1 and 4.2.1.1-3

- 5.12 *Metals Handbook*, 9th ed., vol. 3, *Stainless Steels, Tool Materials and Special-Purpose Metals* (American Society for Metals, Metals Park, Ohio, 1980), pp. 34, 191.
- 5.13 *Engineered Barrier Design Requirements Document* (YMP/CM-0024, REV 0, ICN 1, September 21, 1994)
- 5.14 Gordon M. Barrow, *Physical Chemistry*, 3rd ed. (McGraw-Hill, New York, 1973), pp. 548, 567.
- 5.15 *Q-List* (YMP/90-55Q Rev. 3, December 1994)
- 5.16 IOC VA.DES.NLS.11/94.018, "MPC Design Procurement Specification", from J. B. Stringer to H. A. Benton, November 9, 1994.
- 5.17 R. W. Knoll and E. R. Gilbert, *Evaluation of Cover Gas Impurities and Their Effects on the Dry Storage of LWR Spent Fuel* (PNL-6365, November 1987)
- 5.18 American Society for Testing and Materials, Standard A 480/A 480M-91a, "Standard Specification for General Requirements for Flat-Rolled Stainless and Heat-Resisting Steel Plate, Sheet, and Strip" (August 15, 1991)
- 5.19 *Chemical Engineers' Handbook*, 3rd ed., John H. Perry, editor (McGraw-Hill, New York, 1950), pp. 169-170.

6. Use of Computer Software

The Fuel Assemblies Data Base (FADB) (LWR Assemblies PC Database, CSCI: A00000000-02268-1200-20004 VER 1.1, loaded on a 66 MHz 486 PC, ID: 108141, Serial # 3088159) was used to obtain the following data:

- outside diameter of fuel rod (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4)
- overall length of fuel rod (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4)
- mass of initial uranium in fuel assembly (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4)
- thickness of fuel cladding (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4)
- number of fueled positions (Babcock and Wilcox 15 × 15 / B&W Fuel Company Mark B4).

Because of the interactive nature of FADB, no input file is given here. The description of the results is sufficient to repeat the calculations. Because of the small number of data obtained, no output file is given here. Instead, the results are simply listed in section 4.1.2. The FADB software is appropriate for this application and was used only within the range of validation.

7. Design Analysis

The analysis responds to requirements from the EBD RD that are listed in section 4.2. None of these is listed as TBD, TBV, or TBR.

7.1 Quantity and phases of water allowed by specification

The specification quoted in section 4.1.1 herein has been interpreted by some readers as meaning that up to 0.25 volume percent of the interior void volume of the loaded MPC may be filled with liquid water. Others have interpreted it as meaning that no liquid water will be present and that, by the ideal gas law, the mole fraction of water vapor in the gas inside the backfilled MPC is less than 0.0025. Still others have interpreted it as meaning that the residual water vapor content of the vacuum-dried MPC interior is less than 0.0025 of the interior void volume *while the interior absolute pressure is maintained at 3 millibar*.

Although the specification may, at first glance, appear to be ambiguous about the phases of water present, closer examination shows that the first interpretation, which is discussed in this section, is untenable. The third interpretation is discussed in section 7.7, where it is shown that this interpretation sets the lowest limits on the concentrations of water and potentially reactive gases. For the balance of the analysis, the second interpretation is taken.

Let us examine what the first interpretation implies. Consider a 21 PWR MPC, which has a void volume of 4.555 m^3 (sec. 4.3.1). Using the first interpretation, we may assume that 0.25 percent of this volume, $4.555 \text{ m}^3 \times 0.0025 \times 1000 \text{ L/m}^3 = 11.39 \text{ L}$, is filled with liquid water. Because of the large volume of water and the flat-bottomed shape of an MPC, there will be a large surface area over which water vapor can be produced. The vapor pressure of water at 295 K (sec. 4.3.3) is 2618.4 Pa. If the MPC is evacuated to a pressure of 3 millibar $= (3 \times 100) \text{ Pa} = 300 \text{ Pa}$ as specified (sec. 4.1.1), vigorous vaporization will occur. Continuous, rapid pumping would be necessary to achieve such low pressures while liquid water is present. Without rapid pumping, the specified low pressure would not be achieved until most of the water is evacuated by vacuum pumping.

For reasonable temperatures, evaporative cooling of the water is insufficient to prevent vapor formation; the vapor pressure of water exceeds 600 Pa even at the triple point. In any event, the evaporative cooling effect is small. Evaporating enough water to increase the pressure in the MPC by 300 Pa requires $300 \text{ Pa} \cdot 4.555 \text{ m}^3 / ((8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K}) = 0.557 \text{ mol}$ of water (sec. 4.3.1; sec. 4.3.3). Since the molar mass of water is $(2 \times 1.00794 + 15.9994)$

| g/mol = 18.01528 g/mol = 0.01801528 kg/mol, evaporating that quantity of water at 295 K
| requires an energy of $2,448,300 \text{ J/kg} \cdot 0.01801528 \text{ kg/mol} \cdot 0.557 \text{ mol} = 24.6 \text{ kJ}$. Under
| adiabatic conditions, such evaporation will cool the water by only $24,600 \text{ J} / ((4179 \text{ J/kg}\cdot\text{K}) \cdot$
| $11.39 \text{ L} \cdot 0.99787 \text{ kg/L}) = 0.52 \text{ K}$. An alternative approach to quantifying the cooling effect
| is to determine the heat input necessary to boil water while keeping the temperature constant.
| With a heat input of $24,600 \text{ J} / 600 \text{ s} = 41 \text{ W}$, water can be vaporized, at constant tempera-
| ture, at a rate that increases the pressure in the MPC by 300 Pa over the period of 10 minutes
| = 600 s mentioned in the specification (sec. 4.1.1). The smallness of this input is apparent if
| we compare it to the 14.2 kW spent fuel thermal output that is specified for thermal
| evaluations of an MPC (Ref. 5.1, section 5.1.1.9).

The last sentence in section 5.1.1.7.1.C of Ref. 5.1 is *The sensitivity of this test shall be sufficient to demonstrate by calculation that the residual water content of the MPC interior is less than 0.25 volume percent*. This requirement raises additional difficulties in interpreting the specification as allowing liquid water to be present. It has already been shown that, in the presence of significant volumes of liquid water, continuous and rapid pumping will be necessary to achieve a pressure of 300 Pa. The rate of vapor production will also depend in a complex way on conditions inside the MPC. It is difficult to imagine that the writers of the specification intended the MPC vendors to devise a test that is sufficiently sensitive to demonstrate, under such conditions, that less than 0.25 volume percent of the MPC interior is filled with liquid water.

Interpreting the MPC specification as allowing 0.25 volume percent of liquid water raises numerous difficulties. It is therefore concluded that the correct interpretation of the specification is that the water is in the vapor phase and that the calculated mole fraction of water vapor in the gas inside the MPC must be less than 0.0025. To avoid future confusion, it is suggested that Section 5.1.1.7.1.C of the specification (Ref. 5.1) be revised to change "residual water content of the MPC interior" to "water content of the gas in the filled MPC".

The allowable quantity of water under the second interpretation can be calculated from the ideal gas law. For the assumed temperature, pressure, and void volume, and the maximum allowable water concentration (0.25 volume percent), the amount of water present is $300 \text{ Pa} \cdot 4.555 \text{ m}^3 \cdot 0.0025 / ((8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K}) = 0.706 \text{ mol}$. By using the molar mass of water, it is found that the mass of water is $0.706 \text{ mol} \times 18.01528 \text{ g/mol} = 12.7 \text{ g}$ (sec. 4.3.1; sec. 4.3.2; sec. 4.3.3). Alternatively, if the void volume is filled with water vapor at 300 Pa, the quantity of water present is $300 \text{ Pa} \cdot 4.555 \text{ m}^3 / ((8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K}) = 0.557 \text{ mol}$. The larger value is used here for conservatism and to account for any outgassing that may occur after evacuation.

7.2 Effect of hydrogen from water

Water can be decomposed by radiolysis into hydrogen, hydroxyl (OH) radicals, hydrogen peroxide, oxygen, and other reactive species (Ref. 5.8). The net rate of decomposition under service conditions is not known. It will depend on the strength of the radiation field, the rate at which radiolytic products are consumed (e.g., by oxidation of metals or dissolution of hydrogen in metals), the rate of reverse reactions, and the effectiveness of ionized atoms of inert fill gas in promoting decomposition of water. However, it is conservative to assume that all of the water is decomposed and the products react with the waste or metal components of the MPC.

Because of the affinity of zirconium for hydrogen, we may assume that the radiolytic hydrogen is absorbed by the fuel cladding and determine the effect of such hydrogen on embrittlement of the cladding. The mass of the cladding can be calculated as follows. If end plugs are neglected, the mass of the cladding for one fuel rod can be calculated from the density of zirconium and the formula for the volume of a cylindrical shell to be $\pi \times (10.922 \text{ mm} - 0.673 \text{ mm}) \times 0.673 \text{ mm} \times 3903.47 \text{ mm} \times 10^{-9} \text{ m}^3/\text{mm}^3 \times 6490 \text{ kg}/\text{m}^3 = 0.5490 \text{ kg}$. For an entire 21 PWR MPC, the mass of the cladding is then $208 \times 21 \times 0.5490 \text{ kg} = 2398 \text{ kg}$. Radiolysis of the maximum allowable amount of water (12.7 g) will produce $((2 \times 1.00794) / 18.01528) \times 12.7 \text{ g} = 1.42 \text{ g}$ of hydrogen, so the increase in hydrogen content of the zirconium will be at most $(0.00142) \text{ kg} / 2398 \text{ kg} = 0.59 \times 10^{-6} = 0.59 \text{ parts per million (ppm)}$. In contrast, cladding specifications (Ref. 5.9) allow up to 25 ppm of hydrogen, and fuel designers usually limit hydrogen pickup to 500 to 600 ppm (Ref. 5.10, p. 46) to control hydrogen embrittlement. The additional hydrogen provided by the residual water is negligible.

7.3 Effect of oxygen

To determine the effect of oxygen, we calculate the amount of oxygen present (from water or other sources). We assume (conservatively) that all of the oxygen is present in a reactive form. We calculate the maximum possible depth of oxidation from the amount of oxygen and the area of exposed metal.

Upon evacuation of the MPC to a pressure of 300 Pa (sec. 4.1.1), and if the remaining gas in the MPC is conservatively taken to be dry air, the partial pressure of oxygen will be $300 \text{ Pa} \cdot 0.2095 = 63 \text{ Pa}$. The partial pressure of oxygen will remain after backfilling. The amount of residual oxygen from the dry air is thus $63 \text{ Pa} \cdot 4.555 \text{ m}^3 / ((8.31451 \text{ J}/\text{mol} \cdot \text{K}) \cdot 295 \text{ K}) = 0.117 \text{ mol}$ (sec. 4.3.1; sec. 4.3.3). To this we add the oxygen that will become available if the maximum allowable amount of water is also available. This treatment is appropriate if water outgasses after evacuation of the MPC. The mass of oxygen is $12.7 \text{ g H}_2\text{O} \cdot (15.9994 \text{ g O} / 18.01528 \text{ g H}_2\text{O}) = 11.3 \text{ g}$ from water vapor and $0.117 \text{ mol O}_2 \cdot 31.9988 \text{ g}/\text{mol O}_2 =$

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3.7 g from residual air, for a total of $(11.3 + 3.7) \text{ g} = 15.0 \text{ g} = 15.0 \text{ g} / (2 \times 15.9994 \text{ g/mol}) = 0.470 \text{ mol O}_2$.

For the 21 PWR burnup credit MPC, the interior surfaces of the MPC are made of Type 316L stainless steel, which has a density of 8000 kg/m^3 and a nominal composition of 17% Cr, 12% Ni, 2.5% Mo, and 68.5% (balance) Fe. The influence of carbon (0.03% C maximum) is neglected. To oxidize 1 m^3 of this material, we need

$8000 \text{ kg} \times 0.17 = 1360 \text{ kg Cr} = (1360 \text{ kg}) / (0.0519961 \text{ kg/mol}) = 26,200 \text{ mol Cr}$; oxidation to Cr_2O_3 requires $(3/4) \times 26,200 \text{ mol} = 19,600 \text{ mol O}_2$

$8000 \text{ kg} \times 0.12 = 960 \text{ kg Ni} = (960 \text{ kg}) / (0.0586934 \text{ kg/mol}) = 16,400 \text{ mol Ni}$; oxidation to NiO requires $(1/2) \times 16,400 \text{ mol} = 8200 \text{ mol O}_2$

$8000 \text{ kg} \times 0.025 = 200 \text{ kg Mo} = (200 \text{ kg}) / (0.09594 \text{ kg/mol}) = 2100 \text{ mol Mo}$; oxidation to MoO_3 requires $(3/2) \times 2100 \text{ mol} = 3100 \text{ mol O}_2$

$8000 \text{ kg} \times 0.685 = 5480 \text{ kg Fe} = (5480 \text{ kg}) / (0.055847 \text{ kg/mol}) = 98,100 \text{ mol Fe}$; oxidation to Fe_2O_3 requires $(3/4) \times 98,100 \text{ mol} = 73,600 \text{ mol O}_2$.

The total amount of oxygen needed is thus $(19,600 + 8200 + 3100 + 73,600) \text{ mol O}_2 = 104,500 \text{ mol O}_2$ per cubic meter of alloy.

From the formulas for the areas of cylinders and circles, the area of the inner surface of the shell is $2\pi \cdot 0.74041 \text{ m} \cdot 4.572 \text{ m} + 2\pi \cdot (0.74041 \text{ m})^2 = 24.7 \text{ m}^2$; the surface area of the fuel channels is $21 \cdot 4 \cdot 0.2235 \text{ m} \cdot 4.064 \text{ m} = 76.3 \text{ m}^2$; the total exposed surface area inside the MPC is at least 101 m^2 . A substantially larger area might be justified. The fuel basket is composed of an array of square tubes. The tubes are placed so that the outer surfaces face other tubes or support structures and therefore are not fully exposed. The outer surfaces of the tubes are not included in the area calculation above. As a result, the treatment is conservative in that it may overestimate the depth of oxidation. The maximum depth of oxidation for the exposed alloy is thus $0.470 \text{ mol} / (104,500 \text{ mol/m}^3 \cdot 101 \text{ m}^2) = 45 \text{ nm}$. This may be compared to thicknesses of about 25.4 mm for the shell, 6.35 mm for the inside basket cladding, and 2.39 mm for the outside basket cladding. The effect of oxidation is considered to be negligible because the maximum depth of oxidation is smaller than the permissible variations in thickness. Standard specifications for this material allow these members to be up to 0.25 mm, 0.25 mm, and 0.36 mm undersize, respectively (Ref. 5.18, Tables A1.2 and A1.17).

Fuel cladding is zirconium with small amounts of alloying elements (Ref. 5.9). For the purposes of calculating oxidation depths we may use the density of the pure metal.

Zirconium has a density of 6490 kg/m^3 , so to oxidize 1 m^3 of zirconium to ZrO_2 , we need $(6490 \text{ kg/m}^3) / (0.091224 \text{ kg/mol}) = 71,100 \text{ mol O}_2$.

The total number of rods per MPC is 21 assemblies $\cdot$ 208 rods/assembly = 4368 rods. The area of the fuel cladding is $4368 \cdot \pi \cdot 10.922 \text{ mm} \cdot 3903.47 \text{ mm} = 585 \text{ m}^2$. The maximum depth of oxidation for the cladding is thus $0.470 \text{ mol} / (71,100 \text{ mol/m}^3 \cdot 585 \text{ m}^2) = 11 \text{ nm}$. This may be compared to 0.673 mm, the thickness of the fuel cladding (sec. 6). Again, the effect of oxidation is considered to be negligible because the maximum depth of oxidation is smaller than the permissible variations in thickness. Standard specifications for this material allow a variation in wall thickness of $\pm 10\%$ (Ref. 5.9, Table 5).

7.4 Effect of nitrogen

Radiolysis of residual nitrogen in the MPC could lead to formation of gaseous HNO_3 and possibly condensation of nitric acid solutions. Because of the high diffusivity of gases, gaseous HNO_3 would be expected to produce uniform attack, the depth of attack being limited by the amount of available oxygen. Liquid nitric acid solutions could produce localized attack. This section assesses the possibility that such solutions will condense.

Upon evacuation of the MPC to a pressure of 300 Pa, and if the remaining gas in the MPC is conservatively taken to be dry air, the partial pressure of nitrogen will be $300 \text{ Pa} \cdot 0.7809 = 234 \text{ Pa}$. The partial pressure of nitrogen will remain after backfilling. The amount of residual nitrogen is thus $234 \text{ Pa} \cdot 4.555 \text{ m}^3 / ((8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K}) = 0.435 \text{ mol}$ (sec. 4.3.1; sec. 4.3.3). We assume that production of HNO_3 is limited by the availability of constituent elements. This assumption is extremely conservative. Since amount of oxygen available in the gas is equivalent to only 0.470 mol O_2 , the amount of HNO_3 that can be formed is limited by the oxygen supply to $0.470 \text{ mol} \times (2/3) = 0.313 \text{ mol HNO}_3$. Since the molar mass of HNO_3 is $(1.00794 + 14.00674 + 3 \times 15.9994) \text{ g/mol} = 63.01288 \text{ g/mol}$, this is $0.313 \text{ mol} \times 63.01288 \text{ g/mol} = 19.7 \text{ g HNO}_3$. That quantity of gas will have a partial pressure of $0.313 \text{ mol} \cdot (8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K} / 4.555 \text{ m}^3 = 169 \text{ Pa}$ (sec. 4.3.1; sec. 4.3.3).

Under suitable conditions, gaseous HNO_3 and water vapor can condense to form a solution of nitric acid. At equilibrium, the partial pressures of HNO_3 and H_2O in the gas phase are equal to the respective vapor pressures of these components in the solution. For condensation to occur, therefore, it must be possible to find a liquid composition for which the vapor pressures of HNO_3 and H_2O are both no larger than the respective partial pressures of these gases as calculated above. The vapor pressures of the gases at 295 K may be calculated from the vapor pressure data given in section 4 by interpolating according the Clausius-Clapeyron equation (Ref. 5.14), which states that the logarithm of vapor pressure varies linearly with inverse temperature. To interpolate to 295 K from $20^\circ\text{C} = 293.15 \text{ K}$ and $25^\circ\text{C} = 298.15 \text{ K}$, the appropriate equation is

$$\ln P_{295} = \frac{[(1/298.15 - 1/295) \ln P_{293.15} + (1/295 - 1/293.15) \ln P_{298.15}]}{(1/298.15 - 1/293.15)} \quad (1)$$

Here P is the vapor pressure of the component under consideration and the subscript indicates the temperature, in kelvins, at which the pressure is observed.

From treatments of ideal solutions in physical chemistry (Ref. 5.14), it is expected that vapor pressures will correlate more predictably with the mole fraction of the components than with the mass fraction. For a solution of two components A and B , the mole fractions may be obtained from the equation

$$\chi_A = \frac{m_A/M_A}{m_A/M_A + m_B/M_B}, \quad (2)$$

where m_A and $m_B = 1 - m_A$ are the mass fractions of A and B , respectively, M_A and M_B are the molar masses of A and B , respectively, and χ_A is the mole fraction of A . As was calculated previously, the molar masses of H_2O and HNO_3 are 18.01528 g/mol and 63.01288 g/mol, respectively.

Applying the two equations above gives the following results:

Calculation of vapor pressure of HNO_3 and H_2O at 295 K

1	2	3	4	5	6	7	8	9	10
mole fraction HNO_3	mass fraction HNO_3	$P_{293.15}$ mm Hg HNO_3	$P_{298.15}$ mm Hg HNO_3	P_{295} mm Hg HNO_3	P_{295} Pa HNO_3	$P_{293.15}$ mm Hg H_2O	$P_{298.15}$ mm Hg H_2O	P_{295} mm Hg H_2O	P_{295} Pa H_2O
0.000	0.00				0.0				2618.4
0.067	0.20					15.2	20.6	17.030	2270.5
0.087	0.25					14.2	19.2	15.896	2119.3
0.109	0.30					13.2	17.8	14.761	1968.0
0.133	0.35					12	16.2	13.425	1789.9
0.160	0.40		0.12			10.8	14.6	12.089	1611.7
0.190	0.45	0.15	0.23	0.176	23.5	9.4	12.7	10.519	1402.5
0.222	0.50	0.27	0.39	0.310	41.3	7.9	10.7	8.849	1179.8
0.259	0.55	0.45	0.66	0.519	69.2	6.7	9.1	7.513	1001.6
0.300	0.60	0.84	1.21	0.963	128.4	5.6	7.7	6.308	841.0
0.347	0.65	1.68	2.32	1.896	252.7	4.9	6.6	5.477	730.2
0.400	0.70	3	4.1	3.372	449.5	4.1	5.5	4.576	610.1
0.533	0.80	8	10.5	8.856	1180.7	2.4	3.2	2.673	356.3
0.720	0.90	20	27	22.375	2983.1		1		
1.000	1.00	42	57	47.081	6276.9				0.0

The mass fractions from column 2 are used with Equation 2 to calculate the mole fractions in column 1. The vapor pressures in columns 3 and 4 are used in Equation 1 to calculate the

vapor pressures in column 5. The vapor pressures in columns 7 and 8 are used similarly to calculate the vapor pressures in column 9. Finally, the conversion factor 1 mm Hg = 133.322 Pa is used to convert from the vapor pressures given in columns 5 and 9 to those in columns 6 and 10, respectively. Note that vapor pressure data were sometimes available only at the higher temperature. Where this occurred, interpolation of vapor pressures by Equation 1 was not possible.

In these calculations, the vapor pressure data from the table in section 4 have been supplemented in two ways: First, the vapor pressure of HNO_3 over pure water and the vapor pressure of water over pure nitric acid are both zero. Second, the vapor pressure for pure water has been taken from the independent calculation of this quantity in section 4. To aid visualization, the results in the table have been plotted in the figure below.

With this information, the question of the possibility of condensation can be answered. Because of the requirements of the MPC specification, the partial pressure of water vapor cannot be more than $152,000 \text{ Pa} \times 0.0025 = 380 \text{ Pa}$. It has also been shown above that the partial pressure of HNO_3 cannot exceed 169 Pa. These pressures are indicated in the figure

by dashed lines. Because of the smallness of the partial pressure of water vapor, condensation of solutions with mole fractions of nitric acid less than about 0.521 is impossible. This composition is obtained by linear interpolation on mole fraction from data for H₂O vapor pressure at mole fractions 0.400 and 0.533: A vapor pressure of H₂O of 380 Pa is expected at a mole fraction of HNO₃ of $0.400 + ((380 \text{ Pa} - 610.1 \text{ Pa}) / (356.3 \text{ Pa} - 610.1 \text{ Pa})) \times (0.533 - 0.400) = 0.521$. Similarly, because of the smallness of the partial pressure of HNO₃ vapor, condensation of solutions with mole fractions of nitric acid greater than about 0.315 is impossible. This composition is obtained by linear interpolation on mole fraction from data for HNO₃ vapor pressure at mole fractions 0.300 and 0.347: A vapor pressure of HNO₃ of 169 Pa is expected at a mole fraction of HNO₃ of $0.300 + ((169 \text{ Pa} - 128.4 \text{ Pa}) / (252.7 \text{ Pa} - 128.4 \text{ Pa})) \times (0.347 - 0.300) = 0.315$. Therefore, condensation of nitric acid solutions is impossible, regardless of composition.

7.5 Effect of impurities in fill gas

PNL-6365 states that the standard fill gas for dry storage casks is helium with a purity of 99.995% (Ref. 5.17, p. 10), so the maximum impurity content is 0.005%, or $0.00005 \times (152,000 \text{ Pa}) \times (4.555 \text{ m}^3) / ((8.31451 \text{ J/mol}\cdot\text{K}) \times (295 \text{ K})) = 0.014 \text{ mol}$ of impurities. This is much smaller than the amounts of water, oxygen, and nitrogen calculated above and will have a negligible effect on the calculations.

7.6 Other types of MPC

Analysis of other types of MPC will give slightly different results. For the following reasons, however, the results will not differ significantly. (1) The effect of residual water on hydriding is extremely small (sec. 7.2). Credible changes in fuel or MPC design will not give significant amounts of hydriding. (2) The residual oxygen can cause only extremely small depths of oxidation (sec. 7.3). Credible changes in fuel or MPC design will not cause significant amounts of oxidation. (3) The maximum possible vapor pressures of water and HNO₃ are independent of fuel type and void volume, so condensation of nitric acid cannot occur, regardless of fuel or MPC type (sec. 7.4).

7.7 Application of water content limit at 300 Pa pressure

As mentioned previously, Section 5.1.1.7.1.C of the specification (Ref. 5.1) can be interpreted as meaning that the water content is limited to 0.25 volume percent when the MPC interior is at an absolute pressure of 300 Pa. Under this interpretation, the allowable amount of water is $300 \text{ Pa} \cdot 4.555 \text{ m}^3 \cdot 0.0025 / ((8.31451 \text{ J/mol}\cdot\text{K}) \cdot 295 \text{ K}) = 0.00139 \text{ mol H}_2\text{O} = 18.01528 \text{ g/mol} \times 0.00139 \text{ mol H}_2\text{O} = 0.0251 \text{ g H}_2\text{O}$ (sec. 4.3.1; sec. 4.3.3). Upon radiolysis, this water could be dissociated to form $0.00139 \text{ mol H}_2 = 0.00139 \text{ mol} \times 2 \times 1.00794 \text{ g/mol} = 0.00281 \text{ g H}_2 = 2.81 \text{ mg H}_2$ plus $(1/2 \times 0.00139) \text{ mol} = 0.000696 \text{ mol O}_2 = 0.000696 \text{ mol} \times (2 \times 15.9994) \text{ g/mol} = 0.0223 \text{ g O}_2$. Because the available amounts of hydrogen and oxygen

under this interpretation are both smaller than what was calculated in sections 7.2 and 7.3, the amounts of damage will also be smaller.

In contrast to the calculation in section 7.4, the amount of HNO_3 that could form is now limited by the availability of hydrogen. The maximum amount of HNO_3 that could be formed is $(0.00139 \times 2) \text{ mol} = 0.00279 \text{ mol } \text{HNO}_3 = 0.00279 \text{ mol} \times 63.01288 \text{ g/mol} = 0.176 \text{ g } \text{HNO}_3$. Since the amounts of HNO_3 and water vapor are both smaller than the amounts calculated in section 7.4, condensation of nitric acid solutions is of even less concern.

8. Conclusions

Analysis of the effects of water and gases in the MPC shows that the *Multi-Purpose Canister (MPC) Subsystem Design Procurement Specification* (Ref. 5.1) places sufficiently stringent conditions on the dryness requirements for the MPC cavity environment after loading and closure operations at the Purchaser site. The requirements of the specification are found to be inconsistent with the interpretation that up to 0.25 volume percent of the interior void volume of the loaded MPC may be filled with liquid water. To avoid future confusion, it is suggested that Section 5.1.1.7.1.C of the specification (Ref. 5.1) be revised to change "residual water content of the MPC interior" to "water content of the gas in the filled MPC". If high-purity helium is used to fill the MPC, oxygen and nitrogen are also adequately controlled. The MGDS does not impose additional restrictions on water or gases beyond those given in Reference 5.1.

The *Multi-Purpose Canister (MPC) Subsystem Design Procurement Specification* (Ref. 5.1) imposes sufficiently stringent requirements on the quantity of water and composition of gases inside the MPC that any damage to the waste package by the fill gas will be negligible. In view of this, the following criteria are satisfied:

The requirements of the EBD RD, section 3.7.1.A, are met to the extent that the chemical properties of the fill gas do not compromise the function of the waste packages.

The requirements of the EBD RD, section 3.7.1.B, are met to the extent that the effects of oxidation/reduction reactions, corrosion, hydriding, and radiolysis on the fill gas have been analyzed and found not to produce significant harmful effects.

The requirements of the EBD RD, section 3.7.1.C, are met to the extent that the fill gas has been found not to contain chemically reactive materials in an amount that could compromise the ability of the waste package to contribute to waste isolation.

The requirements of the EBD RD, section 3.7.1.D, are met to the extent that the MPC is found not to contain free liquids in an amount that could compromise the ability of

the waste package to achieve the performance objectives relating to the containment of radioactive waste (because of chemical interactions or formation of pressurized vapor) or result in spillage and spread of contamination in the event of waste package perforation during the period through permanent closure.

The requirements of the EBDRD, section 3.7.1.E, are met to the extent that the fill gas will not degrade the ability of the MPC to maintain waste containment during transportation within the MGDS, emplacement, and retrieval.

9. Attachments

Not applicable.