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Volatilization of urania under strongly oxidizing conditions

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Abstract

Vapor pressures are presented for UO_3(g) over U_3O_{8-x} in air by the transpiration technique. Measurements are also presented for UO_3 over the two-phase region, $UO_{2.24}$–$UO_{2.60.}$ Measurements made in steam-oxygen led to establishment of the homogeneous equilibrium $UO_3(g) + H_2O(g) = UO_2(OH)_2(g)$. Some thermochemical values are presented. A method is shown for computing UO_3 and $UO_2(OH)_2$ pressure for any temperature, steam concentration, and oxygen potential. Total volatilities and mass loss rates have been computed for some potential loss of pool coolant scenarios.
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1. Introduction

The loss of coolant from a spent-fuel pool is a most unlikely event. Should such an event occur, the very fresh fuel (less than one year out of reactor) could see melting or catastrophic oxidation of its Zircaloy clad through the combined effects of decay heat and rapid surface oxidation of the Zircaloy due to air, and possibly by some steam generated locally by the decay heat in the fuel bundles. There have been no systematic studies of the events likely in a spent-fuel pool loss-of-coolant, but a critique by Alvarez et al. [1] indicates that based on a worst case scenario, a significant release of fission products might occur. An understanding of volatilization under strongly oxidizing conditions will allow for estimates of matrix stripping [2], likely the primary mechanism for fission product release. A distinction needs to be made between *oxidizing* and *strongly* oxidizing. Olander [3] has shown that in H_2/steam, UO_3 and $UO_2(OH)_2$ are the principal vaporizing species over 'UO_2'. For a potential accident within containment, H_2/steam is the proper atmosphere in which to evaluate volatilization. In the spent-fuel pool, however, the atmosphere outside the containment is significantly more oxidizing, i.e., *strongly* oxidizing and volatilization in air or air/steam would be at the oxygen potentials of interest. The species would be the same as Olander observed, but the magnitude would be significantly higher. One could conceivably oxidize the fuel up to the orthorhombic U_3O_{8-x} phase, which would result in significant fuel disruption upon the phase change.

2. The uranium oxide–oxygen system

In contact with air, UO_2 will oxidize further. The structure of UO_2 is fluorite, face centered cubic. UO_2 at temperatures above 1400 K will maintain this structure to a composition of about $UO_{2.24}$. Further oxidation results in a phase change to the orthorhombic structure of which U_3O_8 ($UO_{2.67}$) is the most

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well-known composition. The lower phase limit is approximately U_5O_{13} or $UO_{2.60}$, while $UO_{2.67}$ is the maximum composition realized by air oxidation. Below 1400 K an addition phase, tetragonal U_4O_9 ($UO_{2.25}$) has a narrow homogeneity range. From volatilization considerations, temperatures this low result in negligible volatile losses of fuel or any fission products other than the noble gases, krypton and xenon, and possibly ruthenium as a volatile oxide. The temperature–composition diagram is shown in Fig. 1.

For any specified temperature and pressure there is a unique composition across a homogeneity range. That is at say, $UO_{2.20}$ and temperature, T, there is a unique O_2 pressure, P, associated with this composition. Raising the temperature while holding the O_2 pressure constant will move the composition toward $UO_{2.00}$. Raising the O_2 pressure with temperature, T, fixed will move toward the phase boundary, $UO_{2.24}$. Once this phase boundary is reached, an attempt to continue raising the oxygen pressure will fail because any additional oxygen will result in making more of the orthorhombic $UO_{2.60}$. This is in accord with the Phase Rule:

$$F = C - P + 2. \tag{1}$$

There are two components, C, uranium and oxygen, with $UO_{2.24}$, $UO_{2.60}$, there are two condensed phases plus the gas phase so

$$F = 2 - 3 + 2 = 1. \tag{2}$$

If one is assured that both $UO_{2.24}$ and $UO_{2.60}$ are present in the condensed phase, one can perform transpiration experiments by passing an inert gas such as argon or nitrogen over the two-phase system. Oxygen at the equilibrium pressure would then be supplied by the reaction

$$UO_{2.60} = UO_{2.24} + 0.18\ O_2(g). \tag{3}$$

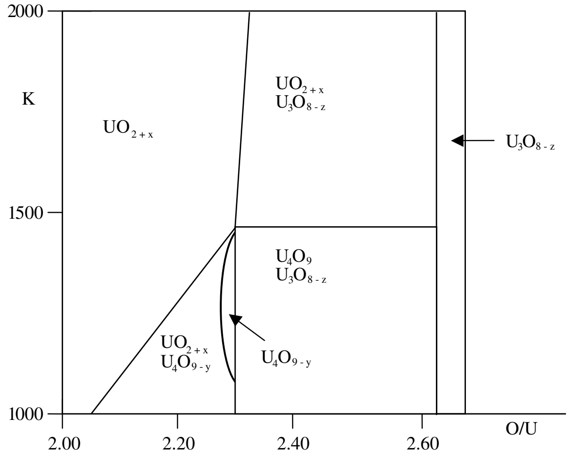

Fig. 1. Uranium–oxygen temperature composition.

It is apparent from the Phase Rule and the discussion above that a different discrete oxygen pressure would exist at each temperature.

When in a single condensed phase region like UO_{2+x} or U_3O_{8-z}, one deals with the condition of constantly changing composition if one selects an oxygen partial pressure and then changes temperature as is generally done in transpiration experiments. If the oxygen partial pressure is held constant and the temperature raised, then the composition will shift to a lower value of the O/U ratio. Since the higher oxidation state at a specific temperature will have a more negative free energy, ΔG, then while the partial pressure of UO_3 will rise, the rise will be less marked because of the slightly more positive change in free energy of the condensed phase. The reaction in general is of the form:

$$UO_{2+x} + \frac{1-x}{2}O_2 = UO_3(g), \tag{4}$$

where x can have any value from 0 to 0.67.

3. Experimental

The transpiration apparatus used for evaluating materials in oxidizing atmospheres has been previously described [4]. The only modification has been to install a ‘reverse flow loop’ so that a reactive gas mixture might enter the condenser pass over and equilibrate the condensed charge prior to the initiation of the experiment. The condenser, boat, and radiation shields were fabricated from platinum-30 wt% rhodium alloy. The condensed charges in all cases were the depleted fuel pellets 8 mm in diameter and 10 mm in length, typically placed in the ends of fuel rods above the fissile material. The pellets were initially crushed then oxidized in a platinum crucible in a muffle furnace to 850 K. They were heated and weighed until a constant weight was obtained.

Transpiration determinations were made for this investigation utilizing three different transpiring gases. These were dry air, dry argon, and pure oxygen containing controlled amounts of water vapor. U_3O_8 was used as starting material in all cases except when argon was employed. In this case, U_3O_8 was mixed with crushed UO_2 and equilibrated at temperature by the reverse flow process. The amount of UO_2 added was sufficient to bring the O/U ratio to a value of about $UO_{2.5}$. This ensured that these determinations would be performed with two condensed phases of uranium oxide present.

This combination was used in the mass spectrometric determinations as well. The mass spectrometer, a Nuclide Corporation 12–90 HT has been described previously [5,6]. The effusion cell selected for this study was fabricated from an iridium bar; otherwise, the spectrometer is as described above.

4. Partial pressures of UO_3

A number of transpiration experiments have been performed in 1 atmosphere oxygen by a number of investigators [7–12] and all gave almost identical results. There are at least two factors favorable to account for the good reproducibility. First are the relatively small differences in Gibbs free energy across the orthorhombic homogeneity range [8]. The second factor is due to the compatibility of urania with platinum and platinum rhodium alloys, coupled with the ease of quantitatively removing the condensate with dilute nitric acid. Earlier measurements [9] in 1 atmosphere oxygen were not repeated herein. The previous equation obtained for the partial pressure of UO_3 over the orthorhombic phase with 1 atmosphere (10^5 Pa) oxygen pressure is

$$\log p\ (\mathrm{Pa}) = \frac{-17\,457}{T} + 11.3211 \tag{5}$$

over the temperature range from 1415 to 1760 K.

This equation can be utilized to establish the free energy of formation of $UO_3(g)$ when used with values for U_3O_8

$$1/3\ U_3O_8 = 1/6\ O_2 = UO_3(g). \tag{6}$$

Utilizing values for U_3O_8 from Ackermann and Chang with Eq. (5) gives

$$\Delta G_f^0 UO_3 = -845\,500 + 86.985 \quad \mathrm{J/mole}. \tag{7}$$

This is as shown in Line 3, Table 5, of Ackermann and Chang.

A previously unreported set of measurements has been made using air as the transpiring gas. These results are shown in Table 1. A resulting equation for the partial pressure of UO_3 over U_3O_{8-x} in air is given by

$$\log p\ (\mathrm{Pa}) = \frac{-18\,438}{T} + 11.825. \tag{8}$$

Table 1
Partial pressure of UO_3 in dried CO_2-free air at 1 atmosphere (10^5 Pa) pressure

Temperature (K)	Pressure (Pa)
1815	50.0
1802	41.8
1797	37.1
1723	13.1
1666	5.96
1618	2.42
1576	1.52
1555	9.87×10^{-1}
1503	3.83×10^{-1}
1472	1.91×10^{-1}
1441	1.07×10^{-1}
1427	8.86×10^{-2}
1418	6.51×10^{-2}
1410	5.14×10^{-2}

Thermogravimetric [13,14] results would indicate a composition in air at 1410 K of $UO_{2.64}$ and at 1815 K of $UO_{2.61}$, which is near the composition limit for the orthorhombic phase.

Both transpiration and mass spectrometric measurements were made over the two-phase UO_{2+x}–U_3O_{8-x} region.

For the mass spectrometer measurement, the equilibrated uranium oxide was loaded into an iridium lined effusion cell along with a NIST [15] vapor pressure standard silver. The silver was totally evaporated both as a sensitivity check as well as a vapor pressure standard. The ionization cross section for UO_3 was based on values reported by Blackburn and Danielson [16]. But owing for some differences in the electrostatic potentials in the different ion sources the value utilized is slightly different (about 10%) from that given by Blackburn.

These vapor pressures of UO_3 are shown in Table 2 and can be represented by

$$\log p\ (\mathrm{Pa}) = \frac{-20\,370}{T} + 12.7622. \tag{9}$$

One modification made to the transpiration apparatus was the additions at the exit end of the condenser of first a drying tube then a weighing tube filled with copper filings capable of being heated to 450 C. From the weight gain of the copper, the total quantity of oxygen in the transpiring gas could be determined. Initially, it was planned to do this for all runs. After the run at 1594, 1735, and 1830 K the practice was

Table 2
Partial pressures of UO_3 in the two-phase region

Temperature (K)	Pressure (Pa)
Transpiration	
1917	121
1880	76.1
1830	36.2
1803	26.1
1801	25.5
1735	10.0
1594	9.43×10^{-1}
1504	1.48×10^{-1}
1493	1.20×10^{-1}
Mass spectrometry	
1402	1.48×10^{-2}
1383	1.02×10^{-2}
1372	7.51×10^{-3}
1355	5.46×10^{-3}
1330	2.81×10^{-3}
1318	2.02×10^{-3}

stopped. Oxygen pressures calculated from the weight gain were in very good agreement with the measurements of Roberts and Walter [17]. In future calculations, values from Ref. [17] have been employed throughout.

The vapor pressure over the two-phase region can be utilized, along with knowledge of the temperature variation of the equilibrium oxygen pressure to establish values for Gibbs free energies for both $UO_{2.24}$ and $UO_{2.60}$

$$UO_{2.24}(c) + 0.38\ O_2(g) = UO_3(g), \tag{10}$$

$$UO_{2.60}(c) + 0.20\ O_2(g) = UO_3(g). \tag{11}$$

Utilizing from Ref. [17],

$$\log p\ O_2 = \frac{-17460}{T} + 13.78. \tag{12}$$

(their Eq. (1)) with the previously determined free energy of formation of $UO_3(g)$ and Eq. (9) one derives

$$\Delta G_f^0 UO_{2.24} - 1108000 + 171.80T \quad \text{J/mole}, \tag{13}$$

$$\Delta G_f^0 UO_{2.60} - 1170000 + 202.84T \quad \text{J/mole}. \tag{14}$$

Typically, one measures the partial pressure of oxygen at a number of compositions in a single-phase field away from some reference composition such as U_3O_8 or UO_2, then computer values for $\overline{G}O_2$ at a fixed temperature. An analytical relationship is then determined for $\overline{G}O_2$ versus the O/U ratio. One then utilizes the relationship

$$\Delta G = 1/2 \int_{2.00}^{2.24} \Delta\overline{G}O_2 d(\mathrm{O/U}) \tag{15}$$

to determine the integral free energy changes. There is some difficulty in doing so because as $UO_{2.00}$ is approached, the oxygen partial pressure drops precipitously to very low values making measurement most difficult.

Lindemer and Besmann's [18] analysis of 38 previous studies showed a more or less linear relationship between partial molar free energy from $UO_{2.24}$ down to about $UO_{2.05}$, and then the precipitous drop down to $UO_{2.00}$. These authors also were able to equate hyper- and hypo-stoichiometric urania to come up with the oxygen potential for $UO_{2.00}$. This value is

$$\mathrm{RT} \ln p(\mathrm{O})_2 = -897000 - 224.8T \quad \text{J/mole}. \tag{16}$$

(their Eq. (46)).

This then gives one of the boundary conditions for computing the relationship between partial molar free energy and composition. The upper boundary at $UO_{2.24}$ is given by Eq. (12). The linear behavior from 2.05 upward and the sudden drop downward at 2.05 suggest that two trapezoids would be an adequate model for the partial molar free energy variation. Using the literature value for free energy of $UO_{2.00}$ [19] along with the values derived herein for $UO_{2.24}$ gives the integral free energy change.

The temperature, 1673 K, was chosen for the example since it represents the highest temperature (19) for which experimental measurements exist over a significant portion of the UO_2–$UO_{2.24}$ homogeneity range. At 1673 K,

$$\Delta G_f^0 UO_2 = -799324 \quad \text{J/mole} \tag{17}$$

and

$$\Delta G_f^0 UO_{2.24} = -821215 \quad \text{J/mole}. \tag{18}$$

The integral free energy change then is 21 890 J/mole. At the lower boundary, $UO_{2.00}$, from Lindemer and Besmann gives for $\Delta\overline{G}O_2$, −520 900 J/mole. The upper boundary, Eq. (12), then is −53 050 J/mole. Solving for two trapezoids with a common side, '*a*', at $UO_{2.05}$

$$-21890 = 0.025\left[\frac{-520900 + a}{2}\right] + 0.095\left[\frac{a - 53050}{2}\right], \tag{19}$$

$$\Delta\overline{G}O_2 UO_{2.05} = -214300 \quad \text{J/mole}. \tag{20}$$

This computed relationship is compared with the experimental values for $\Delta\overline{G}O_2$ in Fig. 2.

In a similar manner, partial molar free energies were calculated for 1800, 2000, and 2200 K. The rationale for temperature range selection is that 1800 K represents the upper limit of experimentation of $UO_3(g)$ and UO_2-$(OH)_2(g)$, while 2200 K is the observed temperature of fuel-Zircaloy [21] based on Phebus experiments. It was arbitrarily taken that three conditions would be considered at these three temperatures; dry air, air −10% steam, and air −50% steam. O/U values and pressures of $UO_3(g)$ for these conditions are shown in Table 3.

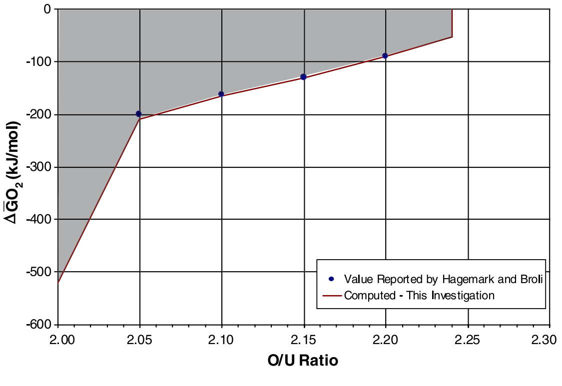

Fig. 2. Partial molar free energy versus composition at 1673 K.

Table 3
Volatility of UO_3 under varying conditions pressure 0.1 MPa

Temperature (K)	Atmosphere	Composition (O/U)	pUO_3 (Pa)
1800	Air	2.24	33.9
1800	Air 10% steam	2.23	32.2
1800	Air 50% steam	2.22	29.1
2000	Air	2.21	193
2000	Air 10% steam	2.20	171
2000	Air 50% steam	2.19	125
2200	Air	2.20	809
2200	Air 10% steam	2.18	718
2200	Air 50% steam	2.17	500

Table 4
Experimental values for $UO_3(g) + H_2O(g) = UO_2(OH)_2(g)$

T (K)	P, Water vapor (Pa)	P, Total uranium (Pa)	P UO_3 (Eq. (5)) (Pa)	P, $UO(OH)_2$ (by difference) (Pa)
1801	1.14^4	6.87×10^1	4.24×10^1	2.62×10^1
1788	1.07×10^4	6.09×10^1	3.61×10^1	2.48×10^1
1718	8.31×10^3	2.71×10^1	1.45×10^1	1.26×10^1
1628	2.19×10^3	5.77×10	3.98	1.79
1584	2.54×10^3	3.94×10	2.00	1.94
1534	2.37×10^3	2.10	8.77×10^{-1}	1.22
1477	2.21×10^3	9.76×10^{-1}	3.19×10^{-1}	6.57×10^{-1}
1443	2.09×10^3	7.36×10^{-1}	1.67×10^{-1}	5.69×10^{-1}

5. Volatility as $UO_2(OH)_2(g)$

This is the third time the volatility as $UO_2(OH)_2(g)$ has been addressed at this laboratory. The first reported transpiration experiment was in 1974 [22] and values found were $pUO_3(g) = 1.31$ pa, $pH_2O = 3290$, $pUO_2(OH)_2 = 1.29$ pa at 1593 K.

Later [23], the Nuclide mass spectrometer was modified by adding an additional level of differential pumping utilizing a Leybold 500 m^3/h Roots Pump. The reaction cell could then be operated at atmospheric pressure and higher. In those experiments a molybdenum apparatus was lined with iridium. Reactant gas was deuterium, D_2O. Deuterium oxide was substituted for light water due to the usual high background at $m/e = 18$ in mass spectrometry. The ion observed was $UO_2(OD)^+$. In that investigation, the ionization cross section for $UO_2(OD)^+$ was estimated to be identical to UO_3^+.

In the current transpiration investigation, low partial pressures of water vapor, as practicable, were employed, on the order of 10^4 Pa or lower in conjunction with oxygen as the carrier gas. There were two reasons to utilize low quantities of water. The first is that with relatively low water addition, the condensed phase stayed very close to $UO_{2.67}$, the nominal composition of U_3O_8. The second reason was that the volatility measurements previously made in pure oxygen showed a quite low probable error of measurement. Since the partial pressure of $UO_3(g)$ would have to be subtracted from the total uranium volatility, by use of these values a more reliable value for $UO_2(OH)_2$ could be attained. To ensure saturation of the carrier gas, a large surface area (15 cm^2) of uranium oxide was exposed to the carrier gas, and gas flow rates on the order of 0.25 cm^3/s were employed. The 'plateau' for saturation of carrier gas for UO_3 volatilization had been established to extend to 3.5 cm^3/s. Results of the transpiration experiments are shown in Table 4.

Utilizing these values, the free energy of formation of $UO_3(g)$ and that for water, the free energy of formation of $UO_2(OH)_2$ over the temperature range from 1440 to 1800 K can be expressed as

$$\Delta G_f^0 UO_2(OH)_2 = -1\,295\,000 + 240.9T \quad \text{J/mol}. \tag{21}$$

6. Free energy functions and the third law

Thermodynamic functions were obtained both by Density Functional Theory (DFT) and Hartree–Fock methods and are listed in Table 5. There is by no means universal agreement on free energy functions. Gorokhov and Sidorova [24] based their computations on $UO_2(F)_2$, CrO_2F_2, MoO_2F_2, and WO_2F_2. They report $S_{298}^0 = 347.45$ Jmol/deg as compared to 368.44 in Table 5 and $S_{2000} = 592.83$ versus 623.03 from Table 5. Their computation lead to a value $\Delta H_{f_{298}}^0 = 1345$ kJ/mol. The situation for gaseous UO_3 appears little better. Cordfunke and Konings compute for the free energy function a value of −322.96 J/mol deg at 298 K versus −309.53 by Glushko et al. [25]. At 2000 K, the values are −407.06 and 380.75, respectively. Third Law heats of vaporization $\Delta H_{f_{298.15}}^0 = -733$ kJ/mol by Cordfunke and Konings versus by $\Delta H_{f_{298.15}}^0 = -799$ kJ/mol by Glushko et al. Because of these variations, the reaction $1/3U_3O_8(c) + 1/8O_2(g) + H_2O(g) = UO_2(OH)_2$ was utilized to compute a Third Law heat from this investigation. Utilizing free energy functions for U_3O_8 [20], water, and oxygen [26], and the values in Table 4 the following was obtained

$$\Delta H_{f_{298.15}}^0 = 1258 \text{ kJ/mol}. \tag{22}$$

Using the same free energy functions and experimental data from Dharwadkar et al., the value is

$$\Delta H_{f_{298.15}}^0 = 1262 \text{ kJ/mol}. \tag{23}$$

No error limits were placed on these values because of the large uncertainties that possibly exist in the free energy computations for the gaseous molecule. It is concluded that the agreement between this investigation

Table 5
Thermodynamic functions for $UO_2(OH)_2(g)$

T (K)	$C_p(T)$ (J/mol K)	$S^0(T)$ (J/mol K)	$H^0(T)-H^0(298)$ (kJ/mol)	$-(G^0(T)-H^0(298))/T$ (J/mol K)
298.15	108.566	368.444	0.00	368.444
300	109.077	369.343	0.201	368.673
400	119.779	402.321	11.690	373.096
500	126.294	429.805	24.020	381.765
600	130.530	453.234	36.874	391.777
700	133.561	473.591	50.087	402.038
800	137.829	491.586	63.564	412.131
900	137.963	507.716	76.132	423.125
1000	139.733	523.347	91.148	432.199
1100	141.323	535.740	105.202	440.102
1200	142.762	548.100	119.407	448.594
1300	144.059	559.581	133.750	456.696
1400	145.235	570.300	148.218	464.430
1500	146.293	580.354	162.795	471.824
1600	147.252	589.827	177.473	478.906
1700	148.109	598.781	192.242	485.698
1800	148.883	607.270	207.091	492.220
1900	149.578	615.337	222.016	498.487
2000	150.209	623.027	237.003	504.465
2100	150.720	629.918	252.048	509.895
2200	151.147	636.319	267.148	514.888
2300	151.490	642.236	282.299	519.497
2400	151.754	647.662	297.491	523.708
2500	151.934	652.604	312.716	527.518

$p^0 = 0.1$ MPa.

and that of Ref. [10] is quite satisfactory, but since nearly identical methods were employed it might be expected that the agreement would be good.

7. Conclusions

It would be expected that upon loss of cladding the fuel bundle would collapse into a rubble bed on the pool base. The surface area for volatilization would be reduced significantly. An effective area for volatilization from the rubble bed might be of the order of 0.5 m^2. Previously [2], from vertically stacked fuel pellets a loss rate of 2.4×10^{-4} kg/m^2 was observed experimentally under conditions of natural convection. With an air flow over the stack, the volatilization rate was higher. A realistic loss rate for the case in air at 1800 K would be 1.2×10^{-4} kg/s and about 30 times higher at 2200 K, accounting for both the temperature effect on volatilization rate and binary diffusivity. Steam would enhance this rate at 1800 and 2000 K but would inhibit volatilization at 2200 K by matrix stripping. As previously mentioned, radiogenic ruthenium would be long gone having readily volatilized as $RuO_3(g)$ or $RuO_4(g)$. Steam would definitely enhance cesium transport. In the fuel, cesium would be in the form of its oxide, Cs_2O. The reaction $Cs_2O(c) + H_2O(g) = 2Cs(OH)$ would readily occur. In laboratory experiments, with Cs_2O, MoO_2, and the $UO_{2.24}$–$UO_{2.60}$ mix $Cs_2MoO_4(g)$ has been observed mass spectrometrically, but the author is unaware of any positive evidence that this species has contributed to cesium transport in any previous study of fission product release.

One must then conclude that a pool fire incident must be considered a very serious accident and can lead to significant fission product release, and steps should be taken to ensure rapid mitigation or, preferably, sufficient hardening to prevent such occurrences (see Table 6).

Table 6
Total volatility of $UO_3(g)$ plus $UO_2(OH)_2(g)$ (Pa)

Temperature (K)	Atmosphere	P_{UO_3}	$P_{UO_2(OH)_2}$	Total volatility (Pa)
1800	Air	33.9		33.9
	Air 10% steam	32.2	16	48.2
	Air 50% steam	29.1	80	109
2000	Air	193		193
	Air 10% steam	171	22.2	193
	Air 50% steam	125	91	216
2200	Air	809		809
	Air 10% steam	718	31	749
	Air 50% steam	500	107	607

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