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REDUCTION OF PuO_2 BY CO AND EQUILIBRIUM PARTIAL PRESSURES ABOVE PLUTONIA

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ABSTRACT

The reduction of plutonia in flowing carbon monoxide has been determined over the range 1200–1800°K. The experimental data confirm recently published thermodynamic functions for the plutonium-oxygen system. Equilibrium partial pressures are listed for vapor species above stoichiometric and reduced plutonia in the range 1200–1800°K.

I. INTRODUCTION

Isotopic heat sources, fueled with $^{238}\text{PuO}_2$ (80 at.% ^{238}Pu), have been used in a number of space missions to supply the electrical energy required for onboard instruments. In a typical heat source, the solid $^{238}\text{PuO}_2$ fuel is encapsulated in a noble metal alloy shell that, in turn, is surrounded by graphite that provides an ablation shell during reentry and impact protection following reentry. These heat source components are generally packaged in a noble metal outer container. The heat generated by the $^{238}\text{PuO}_2$ fuel is converted to electrical energy by means of thermoelectric devices mounted on the outer container. Current isotopic heat sources, designed to operate with fuel surface temperatures in the range 1300–1450°C, are contained in iridium. Advanced modular-type heat sources, currently being tested, are designed to operate with fuel surface temperatures in the range 900–1100°C. The container material for these units will be the Pt-3008 alloy (62 wt% Pt, 30 wt% Rh and 8% W).

Because of the relatively large amount of graphite associated with any heat source, significant quantities of carbon monoxide gas can be formed. Possible oxygen sources include oxides such as SiO_2 in the thermoelectric components, oxygen or water vapor adsorbed on the graphite or container surfaces, and oxygen from the fuel. If carbon monoxide gas is formed, it can reduce the $^{238}\text{PuO}_2$ fuel stoichiometry, producing carbon dioxide gas as a by-product. At a given temperature, the plutonium activity

(partial pressure) will increase as the fuel is reduced. This can lead to reaction with the container material to form such compounds as PuIr_2 or PuPt_3 . Formation of such compounds, in turn, can lead to failure of the container during reentry or impact.

In these experiments, $^{238}\text{PuO}_2$ pellets were reduced in flowing CO gas over the range 927-1527°C (1200-1800°K). The experimental results are in good agreement with values calculated from the thermodynamic functions for the Pu-O system, reviewed and listed in a recent Los Alamos Scientific Laboratory report.¹

II. EXPERIMENTAL PROCEDURES AND RESULTS

The experimental technique was to heat weighed plutonia pellets in flowing carbon monoxide gas, weigh the reduced pellets, and reoxidize the pellets to confirm the weight reduction. The plutonia pellets (80 at. % ^{238}Pu) were formed by hot pressing and were then heated at 1350°C in flowing $\text{Ar-H}_2^{16}\text{O}$ to adjust the stoichiometry to $\text{PuO}_{2.00}$. The use of H_2^{16}O limits the (α , n) reactions that occur with ^{17}O and ^{18}O , and thus reduces the neutron emission rate.

The CO/CO₂ partial pressure ratios were "fixed" by passing the CO gas through a horizontal furnace containing graphite powder heated at a constant temperature. Graphite temperatures were varied from 927-1127°C, and CO pressures were varied from 0.08 to 0.80 atm. Standard air pressure at Los Alamos is approximately 80% that at sea level. The lower CO pressures were obtained by blending the CO gas with dry argon before introduction into the furnace containing the graphite.

With the CO/CO₂ ratio fixed, the reactant gas was introduced into a second horizontal furnace containing two or more weighed $^{238}\text{PuO}_2$ pellets. Three sizes of pellets were used, 3.45, 8.85, and 15.90 g. The pellets were heated for a specified time at a constant temperature and then removed from the furnace and weighed. The amount of reduction was calculated from the weight loss, assuming an atomic weight of 238.2 for the plutonium. A typical isotopic analysis for the $^{238}\text{PuO}_2$ fuel material is listed in Table I. The weight losses were confirmed by reoxidizing the pellets to a constant weight by again heating them at 1350°C in flowing $\text{Ar-H}_2^{16}\text{O}$. All experiments were performed in an inert atmosphere glovebox (<10 ppm oxygen).

Reaction times were varied from 6 to 96 h and the temperature of the fuel pellets was varied from 927-1527°C (1200-1800°K). Temperatures were measured with NBS-calibrated Pt-Pt/Rh thermocouples and the furnaces were operated with programmable controllers capable of maintaining the experimental temperatures constant to $\pm 2^\circ\text{C}$.

For the experiments performed with the fuel pellets heated over the range 927-1127°C, the pellets were placed on Pt-3008 sheet located within the furnace. For the experiments with the fuel pellets heated over the range 1227-1527°C, the pellets were placed on iridium sheet. No reactions occurred between the plutonia and the Pt-3008 or iridium during any experiment.

The data for a typical series of experiments are summarized in Table II. For these particular experiments, the CO gas pressure was 0.08 atm, the graphite temperature was 927°C, and the temperature of the fuel pellets was 1127°C. Reduction run times were 6, 24, 32, and 96 h.

For each series of experiments, at least one reduction run was made during which the furnace containing the fuel pellets was operated under a slight vacuum to ensure removal of all reactant and product gases. Each time, the results obtained were essentially the same as when the vacuum conditions were not used, probably because excess CO reactant gas was present during each reduction run.

Because three sizes of pellets were used, the data for each series of experiments were normalized to determine the weight loss for an average size pellet weighing 8.850 g as $\text{PuO}_{2.00}$. The time dependence of the normalized weight loss for each series of experiments was then calculated by the method of least-squares. Weight loss versus time values for the data listed in Table II are summarized in Table III.

The weight loss versus time relationships for each series of experiments were used to calculate maximum reduction values assuming a 10-yr mission for the heat source. For each series of experiments, however, the results indicate that the ultimate weight loss occurred in less than 1000 h and, in many cases, in less than 100 h. The data listed in Table III indicate that a reduction run time of 1000 h, difficult to control in practice, would result in a weight loss only 2 mg greater than that obtained in 96 h.

Sixty reduction runs were made using 118 $^{238}\text{PuO}_2$ sample pellets. The ultimate, or equilibrium, weight loss values are listed and compared to calculated values in the following section.

III. THERMODYNAMIC CALCULATIONS

For the equations and calculations that follow, the symbol $()$ is used to represent gaseous species and $< >$ is used to represent solids.

Markin and Rand,² using an EMF technique, measured the oxygen potentials above the single-phase plutonium oxides between $< \text{PuO}_{1.61} >$ and $< \text{PuO}_2 >$ over the range 600-1100°C. In addition, published vapor pressure data for plutonium^{3,4,5} and for the plutonium-oxygen system^{1,6,7,8,9,10} have been reviewed in a recent Los Alamos Scientific Laboratory report.¹ Previous thermodynamic calculations pertaining to the plutonium-oxygen system have involved an extrapolation of the Markin and Rand² oxygen potential value, $\overline{\Delta G}_O$, to temperatures greater than 1100°C. These experiments were performed not only to measure the reduction of $< \text{PuO}_{2.0} >$ in flowing (CO), but also to confirm the published values² for $\overline{\Delta G}_O$, and to determine values for $\overline{\Delta G}_O$ at temperatures greater than 1100°C.

Free energy of formation values¹ for (Pu) and $< \text{PuO}_2 >$, used in the following calculations, are listed in Table IV. Also listed in Table IV are free energy of formation values for (O), (CO), and (CO₂), taken from the JANAF Tables.¹¹ The free energies of formation for (PuO) and (PuO₂) are given⁶ by

$$\Delta G_f (\text{PuO}) = -29\,060 - 12.1 T, \text{ } ^\circ\text{K (cal/mole)} \quad (1)$$

and

$$\Delta G_f (\text{PuO}_2) = -113\,100 + 4.35 T, \text{ } ^\circ\text{K (cal/mole)}. \quad (2)$$

Equations for the free energy of formation⁶ for $< \text{PuO}_{2-x} >$ and for $\overline{\Delta G}_O$, as a function of composition,^{2,6} are listed in Table V.

The partial pressures of (O₂), above the reduced fuel pellets, were calculated in the following manner. The partial pressure of (CO₂) is related to the experimental (CO) pressure according to the reaction

For Eq. (3)

$$\begin{aligned} \Delta G &= \Delta G_f(\text{CO}_2) - 2 \Delta G_f(\text{CO}) \\ &= -RT \ln K \\ &= -2.3 RT (\log P_{\text{CO}} - 2 \log P_{\text{CO}_2}) \end{aligned} \quad (4)$$

where R is the gas constant, 1.98726 cal/deg mole, and T is the graphite temperature in degrees Kelvin.

The partial pressures of (O₂) above the fuel pellets are determined from the (CO) and (CO₂) partial pressures according to the reaction

For Eq. (5)

$$\begin{aligned} \Delta G &= 2\Delta G_f(\text{CO}) - 2\Delta G_f(\text{CO}_2) \\ &= -2.3 RT (2 \log P_{\text{CO}} + \log P_{\text{O}_2} - 2 \log P_{\text{CO}_2}) \end{aligned} \quad (6)$$

where T is the temperature of the fuel pellet in degrees Kelvin.

The (O₂) partial pressures above the reduced fuel, obtained from Eq. (6), can then be compared to the (O₂) partial pressures calculated, as a function of composition, from the values of $\overline{\Delta G_O}$ listed in Table V. The partial pressure of (O) above <PuO_{2-x}> is given by

$$\overline{\Delta G_O} = 2.3 RT \log P_O. \quad (7)$$

The partial pressure of (O₂) is related to that for (O) according to the reaction

For Eq. (8)

$$\begin{aligned}
 \Delta G &= -\Delta G_f(O) \\
 &= \frac{-2.3 RT}{2} \log P_{O_2} + 2.3 RT \log P_{CO} \\
 &= \frac{-2.3 RT}{2} \log P_{O_2} + \overline{\Delta G}_O.
 \end{aligned} \tag{9}$$

As an example, the equilibrium (10-yr) stoichiometry listed in Table III is $\langle \text{PuO}_{1.960} \rangle$. With the graphite at 927°C and the (CO) pressure at 0.08 atm, the partial pressure of (CO₂), calculated from Eq. (4), is 1.23×10^{-4} atm. The equilibrium pressure of (O₂) above $\langle \text{PuO}_{1.960} \rangle$, at 1127°C, is then calculated from Eq. (6) to be 2.22×10^{-18} atm. The $\overline{\Delta G}_O$ values listed in Table V, combined with Eq. (9), yield an equilibrium (O₂) partial pressure of 2.22×10^{-18} atm above $\langle \text{PuO}_{1.953} \rangle$ at 1127°C, in good agreement with the experimental value of $\langle \text{PuO}_{1.960} \rangle$.

The experimental values for the reduction of $\langle \text{PuO}_2 \rangle$ by (CO) are summarized and compared to the calculated values in Table VI. The experimental values listed are those at equilibrium (end of a 10-yr mission) calculated from the least-squares fit of the weight loss versus time for each series of experiments. The calculated values listed in Table VI were obtained by combining Eqs. (4), (6), and (9) with the $\overline{\Delta G}_O$ values listed in Table V. All calculated and experimental weight losses have been normalized to those for the average sized experimental pellet weighing 8.850 g as $\langle \text{PuO}_{2.00} \rangle$.

The weight losses calculated from the thermodynamic functions are slightly less than the experimental values at low fuel temperatures and slightly greater at the higher fuel temperatures. The overall agreement, however, is very good and is within the standard deviations calculated from the least-squares fits of the experimental data. These results confirm the $\overline{\Delta G}_O$ values determined by Markin and Rand² and indicate that these $\overline{\Delta G}_O$ values can be accurately extrapolated to 1527°C (1800°K). Therefore, the $\overline{\Delta G}_O$ values listed in Table V have been used to calculate the equilibrium $\langle \text{PuO}_{2-x} \rangle$ stoichiometries for a variety of graphite temperatures (827-1527°C), fuel temperatures (927-1527°C), and (CO) pressures (0.008-8.0 atm). These results are summarized in Tables VII-XIII.

IV. CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{2-x} \rangle$

The major uncertainty associated with the thermodynamic functions listed in Tables IV and V has resulted from the necessity to extrapolate the $\overline{\Delta G}_O$ values to temperatures greater than 1100°C. The results discussed in the previous section indicate that the $\overline{\Delta G}_O$ values listed in Table V are indeed accurate over the range 927-1527°C (1200-1800°K). Therefore, it is possible, using the thermodynamic functions listed in Tables IV and V and Eqs. (1) and (2), to calculate the equilibrium partial pressure values for all vapor species above $\langle \text{PuO}_{2-x} \rangle$ over this same temperature range.

The partial pressure of (O) is given by Eq. (7), that for (O₂) by Eq. (9). The partial pressure of (Pu) can be obtained from the reaction

For Eq. (10)

$$\begin{aligned} \Delta G &= \Delta G_f (\text{Pu}) + (2-x) \Delta G_f (\text{O}) - \Delta G_f < \text{PuO}_{2-x} > \\ &= -2.3 RT \left[\log P_{\text{Pu}} + (2-x) \log P_{\text{O}} \right]. \end{aligned} \quad (11)$$

Similarly, the partial pressure of (PuO) can be obtained from the reaction

For Eq. (12)

$$\begin{aligned} \Delta G &= \Delta G_f (\text{PuO}) + (1-x) \Delta G_f (\text{O}) - \Delta G_f < \text{PuO}_{2-x} > \\ &= -2.3 RT \left[\log P_{\text{PuO}} + (1-x) \log P_{\text{O}} \right]. \end{aligned} \quad (13)$$

Finally, the partial pressure of (PuO₂) can be obtained from the reaction

For Eq. (14)

$$\begin{aligned} \Delta G &= \Delta G_f (\text{PuO}_2) - \Delta G_f < \text{PuO}_{2-x} > - x \Delta G_f (\text{O}) \\ &= -2.3 RT \left[\log P_{\text{PuO}_2} - x \log P_{\text{O}} \right]. \end{aligned} \quad (15)$$

Calculated values for the partial pressures of (O), (O₂), (Pu), (PuO), and (PuO₂) above various < PuO_{2-x} > compositions, in the range 927-1527°C (1200-1800°K), are listed in Tables XIV-XX.

V. REACTION OF PLUTONIUM VAPOR WITH IRIIDIUM AND THE Pt-3008 ALLOY

Current isotopic heat sources designed to operate with fuel surface temperatures in the range 1300-1450°C are contained in iridium. Advanced modular-type heat sources, currently being tested, are designed to operate with fuel temperatures in the range 900-1100°C. The container material for these units will be the Pt-3008 alloy (62 wt% Pt, 30 wt% Rh, and 8 wt% W).

The values listed in Tables VII-XIII indicate that if sufficient quantities of (CO) are present the < PuO₂ > fuel can be significantly reduced. The values listed in Tables XIV-XX indicate the increase in the partial pressure of (Pu) as the < PuO₂ > is reduced.

This is of concern because, as the (Pu) pressure increases, it becomes more likely that the (Pu) can react with the noble metal container.

The vapor pressure of (Pu) above $\langle \text{PuIr}_2 \rangle$ is given¹² by the equation

$$\log P_{\text{Pu}} = 4.26 - 26\,529/T, \text{ } ^\circ\text{K}. \quad (16)$$

By combining Eq. (16) with the (Pu) partial pressures listed in Tables XIV-XX, one can determine if the compound $\langle \text{PuIr}_2 \rangle$ will form. So long as the vapor pressure of (Pu) above $\langle \text{PuIr}_2 \rangle$ is greater than that above $\langle \text{PuO}_{2-x} \rangle$, no reaction can occur.

Peterson¹³ has studied the phase relationships and vaporization behavior of plutonium combined with the various components of the Pt-3008 alloy. To date, he has determined the (Pu) pressure above four compounds; $\langle \text{PuPt}_3 \rangle$, $\langle \text{PuPt}_4 \rangle$, $\langle \text{PuPt}_5 \rangle$, and $\langle \text{PuRh}_3 \rangle$. The (Pu) pressures above these compounds are given by the following equations:

$$\langle \text{PuPt}_3 \rangle, \log P_{\text{Pu}} = 5.04 - 26\,940/T, \text{ } ^\circ\text{K} \quad (17)$$

$$\langle \text{PuPt}_4 \rangle, \log P_{\text{Pu}} = 1.11 - 21\,303/T, \text{ } ^\circ\text{K} \quad (18)$$

$$\langle \text{PuPt}_5 \rangle, \log P_{\text{Pu}} = -2.16 - 17\,178/T, \text{ } ^\circ\text{K} \quad (19)$$

and

$$\langle \text{PuRh}_3 \rangle, \log P_{\text{Pu}} = 7.856 - 31\,859/T, \text{ } ^\circ\text{K}. \quad (20)$$

Equations (17), (18), (19), and (20) can be combined with the (Pu) partial pressure values listed in Tables XIV-XX to determine if the various compounds will form.

Compositions of $\langle \text{PuO}_{2-x} \rangle$ that lead to formation of the Pu-Ir, Pu-Pt, and Pu-Rh compounds are listed in Table XXI. These values, in turn, can be combined with those listed in Tables VII-XIII to determine if the (CO), present in the heat source, can reduce the $\langle \text{PuO}_2 \rangle$ fuel to the required stoichiometries.

The results of a recent study¹⁴ indicate that most of the (CO) present in a heat source is probably due to degassing of the graphite components. Furthermore, the quantities and pressures of (CO) likely to be encountered in a vented heat source are such that the (CO) should not reduce the $\langle \text{PuO}_2 \rangle$ sufficiently to cause reaction with the Pt-3008 alloy in the expected operating temperature range (below 1100°C). Similarly, so long as the fuel temperature is higher than the graphite temperature, no reaction should occur with iridium below fuel temperatures of 1450°C . Finally, although higher graphite temperatures can occur during reentry, the exposure times are much too short (5-10 min) to produce significant reduction of the fuel.

VI. SUMMARY

The reduction of PuO_2 in flowing (CO) has been determined over the range 927-1527°C (1200-1800°K). The thermodynamic functions listed in Tables IV and V lead to calculated O/Pu ratios that are in good agreement with the experimental data. Thus, these thermodynamic functions can be used to predict accurately the reduction of PuO_2 by (CO) over a wide range of graphite and fuel temperatures and (CO) pressures. Calculated reduction values are listed in Tables VII-XIII. In addition, the thermodynamic functions listed in Tables IV and V can be used to calculate the equilibrium partial pressure values of all vapor species above PuO_{2-x} . These calculated values are listed in Tables XIV-XX. Finally, the quantities of (CO) likely to exist in a typical vented heat source are such that reduction of the fuel, sufficient to promote reaction with iridium or Pt-3008 containers, is unlikely in the expected operational temperature ranges.

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TABLE I
ISOTOPIC ANALYSIS FOR $^{238}\text{PuO}_2$
FUEL FEED MATERIAL

<u>Isotope</u>	<u>Wt%^a</u>
^{238}Pu	80.02
^{239}Pu	16.33
^{240}Pu	2.66
^{241}Pu	0.76
^{242}Pu	0.23

^a Analyses performed at Savannah River.

TABLE II

REDUCTION OF PuO_2 AT 1127°C

BY CO AT 0.08 ATM

(graphite at 927°C)

Time (h)	Pellet No.	Weight (g)		Stoichiometry (O/Pu)
		Oxidized ^a	Reduced	
6	135-2	8.8523	8.8473	1.990
	145-1	3.4465	3.4451	1.993
	135-2	8.8473	8.8454	1.996
	145-2	3.4517	3.4485	<u>1.984</u>
				1.991 ± 0.005
24	135-2	8.8480	8.8365	1.978
	145-2	3.4492	3.4411	<u>1.960</u>
				1.969 ± 0.012
32	134-3	8.8484	8.8365	1.973
	145-3	3.4492	3.4432	<u>1.971</u>
				1.972 ± 0.002
32-V ^b	135-1	8.8467	8.8325	1.973
	145-1	3.4478	3.4410	<u>1.967</u>
				1.965 ± 0.004
96	135-2	8.8520	8.8342	1.966
	145-2	3.4502	3.4429	<u>1.964</u>
				1.965 ± 0.002

^aAverage of initial and reoxidized weights.^bSlight vacuum on furnace.

TABLE III
REDUCTION OF PuO_2 AT 1127°C
VS
TIME

(CO pressure at 0.08 atm, graphite at 927°C)

Time (h)	Stoichiometry (O/Pu)	Weight Loss (g) ^a	
		Calc. ^b	Obs.
6	1.992	0.004	0.005
24	1.973	0.014	0.017
32	1.970	0.015	0.015
96	1.964	0.019	0.019
1000	1.960	0.021	
10 yr	1.960	0.021	

^aBased on average size pellet (8.850 g as $\text{PuO}_{2.00}$).

^bCalculated from least-squares fit of data (8.850 g pellet),

$$\ln(\text{g lost}) = (-9.57 \pm 1.53)/\text{time (h)} - (3.86 \pm 0.15).$$

TABLE IV

FREE ENERGIES OF FORMATION
(kcal/mole)

Temp (°K)	(O)	(CO)	(CO ₂)	(Pu)	< PuO ₂ >
298	55.395	-32.783	-94.265	74.32	-239.54
1200	41.755	-52.049	-94.681	52.73	-200.93
1300	40.186	-54.126	-94.701	50.52	-196.78
1400	38.611	-56.189	-94.716	48.31	-192.68
1500	37.022	-58.241	-94.728	46.10	-188.54
1600	35.448	-60.284	-94.739	43.88	-184.61
1700	33.862	-62.315	-94.746	41.65	-180.65
1800	32.271	-64.337	-94.750	39.38	-176.71

TABLE V

FREE ENERGIES OF FORMATION AND PARTIAL MOLAR FREE
ENERGIES OF ATOMIC OXYGEN FOR SOLID PLUTONIUM OXIDES

Species	$\Delta G_f^0 = A + BT, \text{ } ^\circ K$ (cal/mole)		$\Delta G_o^0 = A + BT, \text{ } ^\circ K$ (cal/mole)	
	A	B	A	B
< PuO ₂ >	See Table IV		-204.750	87.7
< PuO _{1.96} >	-246.700	39.0	-181.750	63.4
< PuO _{1.95} >	-243.100	37.6	-178.000	53.0
< PuO _{1.92} >	-239.700	36.4	-173.750	54.1
< PuO _{1.90} >	-237.500	35.7	-171.150	50.8
< PuO _{1.85} >	-232.100	34.1	-164.250	43.4
< PuO _{1.80} >	-227.100	32.9	-157.500	36.1
< PuO _{1.75} >	-222.500	32.1	-151.500	29.5
< PuO _{1.70} >	-218.000	31.5	-149.250	26.5
< PuO _{1.65} >	-213.600	31.0	-150.250	26.1
< PuO _{1.61} >	-210.000	30.6	153.000	26.9
< PuO _{1.50} >	-206.300	29.6	-153.000	26.9

TABLE VI
REDUCTION OF PuO_2 BY CO
CALCULATED VS OBSERVED VALUES

Temperature ($^{\circ}\text{C}$)		P_{CO} (atm)	Calculated		Observed ^a	
PuO_2	Graphite		O/Pu	Weight ^b Loss (g)	O/Pu	Weight Loss (g)
927	927	0.08	1.999	0.001	1.994	0.003
		0.80	2.000	0.000	1.997	0.002
1027	927	0.08	1.999	0.006	1.979	0.011
		0.80	1.999	0.001	1.995	0.003
1127	927	0.08	1.953	0.025	1.960	0.021
		0.80	1.992	0.004	1.979	0.011
1227	1027	0.08	1.879	0.063	1.851	0.078
		0.80	1.947	0.028	1.952	0.025
1327	1027	0.08	1.852	0.078	1.867	0.069
		0.80	1.920	0.042	1.910	0.047
1427	927	0.08	1.870	0.068	1.878	0.064
	1027	0.08	1.832	0.088	1.846	0.081
	1127	0.80	1.868	0.069	1.868	0.069
1527	1127	0.08	1.755	0.128	1.770	0.121
	1127	0.80	1.823	0.093	1.840	0.084

^aEquilibrium (10-yr) values obtained from least-squares fit of reduction vs time.

^bBased on average size pellet (8.850 g as $\text{PuO}_{2.00}$).

TABLE VII
CALCULATED REDUCTION OF PuO_2 BY CO -
 PuO_2 AT 927°C (1200°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.8	8.0
827	2.00	2.00	2.00	2.00
927	1.98	2.00	2.00	2.00
1027	1.93	1.99	2.00	2.00
1127	1.88	1.98	2.00	2.00
1227	1.82	1.94	1.99	2.00
1327	1.77	1.90	1.99	2.00
1427	1.72	1.87	1.97	2.00
1527	1.66	1.83	1.94	2.00

TABLE VIII
CALCULATED REDUCTION OF PuO_2 BY CO -
 PuO_2 AT 1027°C (1300°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.80	8.0
827	1.98	2.00	2.00	2.00
927	1.92	1.99	2.00	2.00
1027	1.87	1.96	1.99	2.00
1127	1.81	1.91	1.99	2.00
1227	1.77	1.88	1.97	1.99
1327	1.72	1.84	1.94	1.99
1427	1.66	1.81	1.91	1.99
1527	1.62	1.78	1.89	1.97

TABLE IX
CALCULATED REDUCTION OF PuO_2 BY CO -
 PuO_2 AT 1127°C (1400°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.80	8.0
827	1.93	1.99	2.00	2.00
927	1.88	1.95	1.99	2.00
1027	1.82	1.91	1.97	2.00
1127	1.78	1.87	1.95	1.99
1227	1.73	1.84	1.92	1.98
1327	1.67	1.81	1.90	1.97
1427	1.62	1.77	1.87	1.94
1527	$1.50 + \ell$	1.74	1.84	1.92

TABLE X
CALCULATED REDUCTION OF PuO_2 BY CO -
 PuO_2 AT 1227°C (1500°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.8	8.0
827	1.90	1.96	1.99	2.00
927	1.84	1.92	1.98	1.99
1027	1.80	1.88	1.95	1.99
1127	1.76	1.84	1.92	1.98
1227	1.70	1.81	1.89	1.95
1327	1.62	1.78	1.86	1.93
1427	$1.50 + \ell$	1.75	1.83	1.91
1527	$1.50 + \ell$	1.70	1.82	1.89

TABLE XI

CALCULATED REDUCTION OF PuO_2 BY CO-
 PuO_2 AT 1327°C (1600°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.8	8.0
827	1.87	1.93	1.99	2.00
927	1.82	1.89	1.95	1.99
1027	1.78	1.85	1.92	1.98
1127	1.73	1.82	1.89	1.95
1227	1.65	1.78	1.86	1.93
1327	1.50 ± 0.02	1.76	1.83	1.90
1427	1.50 ± 0.02	1.72	1.81	1.88
1527	1.50 ± 0.02	1.66	1.79	1.86

TABLE XII

CALCULATED REDUCTION OF PuO_2 BY CO-
 PuO_2 AT 1427°C (1700°K)

Graphite Temperature ($^\circ\text{C}$)	Equilibrium Stoichiometry (O/Pu)			
	CO Pressure (atm)			
	0.008	0.08	0.80	8.0
827	1.85	1.91	1.97	1.99
927	1.80	1.87	1.93	1.98
1027	1.76	1.83	1.90	1.96
1127	1.70	1.79	1.87	1.93
1227	1.61	1.77	1.84	1.90
1327	1.50 ± 0.02	1.73	1.81	1.88
1427	1.50 ± 0.02	1.68	1.79	1.86
1527	1.50 ± 0.02	1.62	1.77	1.84

TABLE XIII
CALCULATED REDUCTION OF PuO_2 BY CO -
 PuO_2 AT 1527°C (1800°K)

Graphite Temperature ($^\circ\text{C}$)	<u>Equilibrium Stoichiometry (O/Pu)</u>			
	<u>CO Pressure (atm)</u>			
	<u>0.008</u>	<u>0.08</u>	<u>0.8</u>	<u>8.0</u>
827	1.83	1.90	1.95	1.99
927	1.79	1.85	1.91	1.97
1027	1.74	1.82	1.88	1.94
1127	$1.50 + \ell$	1.76	1.82	1.89
1227	$1.50 + \ell$	1.75	1.82	1.88
1327	$1.50 + \ell$	1.71	1.80	1.86
1427	$1.50 + \ell$	1.65	1.78	1.84
1527	$1.50 + \ell$	$1.50 + \ell$	1.76	1.83

TABLE XIV
EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{2-x} \rangle$
AT 927°C (1200°K)

Stoichiometry (O/Pu)	-log P (at.m)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	18.12	21.04	25.20	18.19	16.95
1.99	18.94	22.67	23.56	17.37	16.98
1.98	19.24	23.28	22.96	17.07	16.99
1.97	19.33	23.45	22.79	16.99	17.00
1.96	19.43	23.65	22.59	16.89	17.00
1.95	19.52	23.84	22.42	16.81	17.00
1.94	19.62	24.04	22.22	16.71	17.01
1.93	19.71	24.22	22.05	16.63	17.02
1.92	19.82	24.43	21.85	16.53	17.03
1.91	19.91	24.61	21.68	16.46	17.04
1.90	20.07	24.93	21.37	16.31	17.05
1.89	20.14	25.07	21.24	16.24	17.06
1.88	20.21	25.21	21.10	16.18	17.06
1.87	20.28	25.36	20.96	16.11	17.07
1.86	20.36	25.50	20.83	16.05	17.08
1.85	20.43	25.65	20.70	15.99	17.09
1.84	20.50	25.79	20.56	15.93	17.10
1.83	20.57	25.94	20.43	15.87	17.12
1.82	20.65	26.09	20.29	15.81	17.13
1.81	20.72	26.23	20.16	15.75	17.14
1.80	20.79	26.38	20.03	15.69	17.16
1.79	20.86	26.52	19.90	15.64	17.17
1.78	20.93	26.66	19.78	15.58	17.19
1.77	21.00	26.80	19.66	15.53	17.21
1.76	21.07	26.94	19.54	15.48	17.22
1.75	21.14	27.08	19.41	15.43	17.24
1.74	21.19	27.18	19.33	15.39	17.25
1.73	21.24	27.28	19.24	15.35	17.27
1.72	21.29	27.37	19.15	15.31	17.28
1.71	21.34	27.47	19.07	15.28	17.29
1.70	21.39	27.57	18.99	15.24	17.31
1.69	21.44	27.68	18.89	15.20	17.32
1.68	21.50	27.79	18.80	15.17	17.34
1.67	21.55	27.89	18.71	15.13	17.36
1.66	21.61	28.00	18.63	15.10	17.38
1.65	21.66	28.11	18.54	15.06	17.40
1.64	21.74	28.27	18.40	15.01	17.43
1.63	21.82	28.44	18.27	14.96	17.46
1.62	21.90	28.60	18.14	14.91	17.49
1.61	21.99	28.76	18.01	14.86	17.52

TABLE XV
EQUILIBRIUM PARTIAL PRESSURES ABOVE PuO_{2-x}
AT 1027°C (1300°K)

Stoichiometry (O/Pu)	<u>-log P (atm)</u>				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	15.25	17.00	24.63	17.12	15.02
1.99	16.35	19.19	22.45	16.03	15.08
1.98	16.70	19.89	21.76	15.69	15.09
1.97	16.80	20.09	21.55	15.59	15.09
1.96	16.92	20.33	21.32	15.47	15.10
1.95	17.03	20.55	21.11	15.37	15.10
1.94	17.15	20.79	20.87	15.26	15.11
1.93	17.26	21.01	20.66	15.15	15.11
1.92	17.33	21.26	20.42	15.04	15.13
1.91	17.50	21.49	20.22	14.95	15.15
1.90	17.67	21.83	19.88	14.78	15.15
1.89	17.76	22.01	19.70	14.70	15.16
1.88	17.85	22.19	19.53	14.62	15.17
1.87	17.94	22.38	19.36	14.54	15.18
1.86	18.04	22.56	19.19	14.46	15.19
1.85	18.13	22.74	19.02	14.38	15.21
1.84	18.22	22.93	18.85	14.30	15.22
1.83	18.31	23.11	18.69	14.22	15.24
1.82	18.40	23.30	18.51	14.15	15.25
1.81	18.50	23.48	18.35	14.07	15.27
1.80	18.59	23.66	18.18	14.00	15.29
1.79	18.67	23.84	18.03	13.93	15.31
1.78	18.76	24.01	17.87	13.86	15.33
1.77	18.85	24.18	17.72	13.80	15.35
1.76	18.93	24.36	17.57	13.73	15.37
1.75	19.02	24.53	17.42	13.67	15.39
1.74	19.08	24.64	17.32	13.63	15.41
1.73	19.13	24.75	17.22	13.58	15.42
1.72	19.19	24.86	17.12	13.54	15.43
1.71	19.24	24.98	17.03	13.50	15.45
1.70	19.30	25.09	16.93	13.46	15.46
1.69	19.35	25.19	16.85	13.43	15.48
1.68	19.40	25.29	16.76	13.39	15.50
1.67	19.45	25.39	16.68	13.36	15.51
1.66	19.50	25.50	16.59	13.33	15.53
1.65	19.55	25.60	16.51	13.29	15.55
1.64	19.58	25.74	16.39	13.25	15.57
1.63	19.70	25.89	16.27	13.20	15.60
1.62	19.77	26.03	16.15	13.16	15.63
1.61	19.84	26.17	16.01	13.11	15.66

TABLE XVI
EQUILIBRIUM PARTIAL PRESSURES ABOVE PuO_{2-x}
AT 1127°C (1400°K)

Stoichiometry (O/Pu)	-log P (atm)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	12.80	13.54	24.14	16.20	13.37
1.99	14.13	16.20	21.49	14.87	13.44
1.98	14.52	16.98	20.72	14.50	13.45
1.97	14.63	17.21	20.49	14.39	13.46
1.96	14.77	17.48	20.23	14.26	13.46
1.95	14.89	17.73	19.99	14.14	13.47
1.94	15.03	18.00	19.72	14.01	13.47
1.93	15.16	18.26	19.47	13.89	13.48
1.92	15.30	18.54	19.20	13.76	13.50
1.91	15.43	18.81	18.96	13.65	13.52
1.90	15.61	19.18	18.60	13.47	13.53
1.89	15.72	19.39	18.39	13.37	13.54
1.88	15.83	19.61	18.19	13.28	13.55
1.87	15.94	19.82	17.98	13.18	13.56
1.86	16.05	20.04	17.78	13.09	13.58
1.85	16.15	20.26	17.58	13.00	13.59
1.84	16.26	20.47	17.38	12.91	13.61
1.83	16.37	20.69	17.18	12.81	13.63
1.82	16.48	20.91	16.99	12.73	13.64
1.81	16.59	21.12	16.79	12.64	13.67
1.80	16.70	21.34	16.60	12.55	13.69
1.79	16.80	21.54	16.41	12.47	13.71
1.78	16.90	21.74	16.23	12.39	13.73
1.77	17.00	21.95	16.05	12.31	13.75
1.76	17.10	22.15	15.88	12.24	13.78
1.75	17.20	22.35	15.70	12.16	13.81
1.74	17.26	22.47	15.59	12.12	13.82
1.73	17.32	22.59	15.49	12.07	13.83
1.72	17.38	22.72	15.38	12.03	13.85
1.71	17.45	22.84	15.28	11.98	13.87
1.70	17.51	22.96	15.17	11.94	13.89
1.69	17.56	23.06	15.09	11.90	13.90
1.68	17.60	23.15	15.01	11.87	13.92
1.67	17.65	23.25	14.93	11.84	13.93
1.66	17.70	23.35	14.85	11.81	13.95
1.65	17.75	23.45	14.77	11.78	13.97
1.64	17.81	23.57	14.66	11.73	13.99
1.63	17.88	23.70	14.56	11.69	14.01
1.62	17.94	23.83	14.45	11.65	14.04
1.61	18.00	23.95	14.35	11.62	14.06

TABLE XVII
EQUILIBRIUM PARTIAL PRESSURES ABOVE PuO_{2-x}
AT 1227°C (1500°K)

Stoichiometry (O/Pu)	-log P (atm)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	10.66	10.54	23.72	15.40	11.94
1.99	12.20	13.61	20.65	13.87	12.02
1.98	12.62	14.46	19.82	13.47	12.04
1.97	12.75	14.72	19.57	13.34	12.04
1.96	12.90	15.01	19.28	13.20	12.05
1.95	13.04	15.29	19.01	13.07	12.06
1.94	13.19	15.59	18.72	12.92	12.06
1.93	13.33	15.87	18.44	12.79	12.07
1.92	13.49	16.19	18.14	12.65	12.09
1.91	13.64	16.49	17.87	12.52	12.11
1.90	13.83	16.88	17.48	12.34	12.12
1.89	13.96	17.12	17.25	12.22	12.13
1.88	14.08	17.37	17.02	12.12	12.14
1.87	14.20	17.61	16.79	12.01	12.16
1.86	14.32	17.86	16.56	11.90	12.17
1.85	14.45	18.10	16.34	11.80	12.19
1.84	14.57	18.35	16.11	11.70	12.21
1.83	14.69	18.59	15.88	11.59	12.23
1.82	14.81	18.83	15.66	11.49	12.25
1.81	14.94	19.08	15.44	11.39	12.28
1.80	15.06	19.32	15.22	11.30	12.30
1.79	15.17	19.55	15.02	11.21	12.32
1.78	15.28	19.78	14.81	11.12	12.35
1.77	15.40	20.01	14.61	11.03	12.37
1.76	15.51	20.23	14.41	10.94	12.40
1.75	15.63	20.46	14.22	10.86	12.43
1.74	15.69	20.59	14.10	10.81	12.45
1.73	15.76	20.72	13.98	10.76	12.46
1.72	15.82	20.85	13.87	10.71	12.48
1.71	15.89	20.99	13.76	10.66	12.50
1.70	15.95	21.12	13.64	10.62	12.52
1.69	16.00	21.21	13.57	10.58	12.53
1.68	16.05	21.30	13.49	10.55	12.55
1.67	16.09	21.40	13.41	10.22	12.56
1.66	16.14	21.49	13.33	10.49	12.58
1.65	16.19	21.58	13.26	10.46	12.60
1.64	16.24	21.70	13.16	10.42	12.61
1.63	16.30	21.81	13.07	10.39	12.63
1.62	16.36	21.92	12.98	10.35	12.66
1.61	16.41	22.03	12.89	10.32	12.68

TABLE XVIII
EQUILIBRIUM PARTIAL PRESSURES ABOVE PuO_{2-x}
AT 1327°C (1600°K)

Stoichiometry (O/Pu)	-log P (atm)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	8.80	7.92	23.35	14.71	10.72
1.99	10.52	11.35	19.92	13.00	10.78
1.98	10.97	12.26	19.03	12.56	10.80
1.97	11.11	12.54	18.76	12.43	10.81
1.96	11.27	12.85	18.45	12.28	10.81
1.95	11.42	13.15	18.16	12.13	10.82
1.94	11.58	13.48	17.84	11.98	10.82
1.93	11.74	13.79	17.54	11.83	10.84
1.92	11.92	14.16	17.18	11.67	10.85
1.91	12.07	14.46	16.91	11.54	10.87
1.90	12.23	14.87	16.51	11.34	10.88
1.89	12.41	15.14	16.25	11.22	10.90
1.88	12.55	15.41	15.99	11.10	10.91
1.87	12.68	15.68	15.74	10.98	10.93
1.86	12.81	15.95	15.49	10.87	10.95
1.85	12.95	16.22	15.24	10.75	10.97
1.84	13.08	16.49	14.99	10.64	10.99
1.83	13.22	16.75	14.74	10.52	11.01
1.82	13.35	17.02	14.50	10.41	11.03
1.81	13.49	17.29	14.26	10.30	11.06
1.80	13.62	17.56	14.02	10.20	11.09
1.79	13.75	17.81	13.79	10.10	11.11
1.78	13.87	18.06	13.57	10.00	11.14
1.77	14.00	18.31	13.35	9.90	11.17
1.76	14.12	18.56	13.13	9.81	11.20
1.75	14.25	18.81	12.91	9.72	11.23
1.74	14.32	18.95	12.79	9.66	11.24
1.73	14.39	19.09	12.67	9.61	11.26
1.72	14.46	19.23	12.54	9.56	11.28
1.71	14.52	19.37	12.42	9.51	11.30
1.70	14.59	19.51	12.31	9.46	11.32
1.69	14.64	19.59	12.23	9.43	11.33
1.68	14.68	19.68	12.12	9.40	11.35
1.67	14.73	19.77	12.08	9.37	11.36
1.66	14.77	19.86	12.01	9.34	11.38
1.65	14.82	19.95	11.93	9.31	11.39
1.64	14.87	20.05	11.85	9.28	11.41
1.63	14.92	20.15	11.77	9.24	11.43
1.62	14.97	20.25	11.68	9.21	11.45
1.61	15.02	20.35	11.60	9.18	11.47

TABLE XIX

EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{2-x} \rangle$
 AT 1427°C (1700°K)

Stoichiometry (O/Pu)	-log P (atm)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	7.16	5.60	23.01	14.09	9.63
1.99	9.03	9.36	19.27	12.23	9.69
1.98	9.51	10.31	18.33	11.77	9.70
1.97	9.66	10.61	18.09	11.67	9.71
1.96	9.83	10.95	17.71	11.46	9.72
1.95	9.99	11.27	17.40	11.31	9.73
1.94	10.16	11.62	17.18	11.20	9.73
1.93	10.33	11.95	16.73	10.99	9.75
1.92	10.51	12.32	16.38	10.82	9.76
1.91	10.69	12.77	16.06	10.67	9.79
1.90	10.90	13.09	15.64	10.46	9.80
1.89	11.05	13.39	15.36	10.33	9.81
1.88	11.19	13.68	15.09	10.20	9.82
1.87	11.34	13.97	14.81	10.07	9.84
1.86	11.48	14.26	14.54	9.95	9.86
1.85	11.63	14.55	14.27	9.83	9.89
1.84	11.78	14.85	14.00	9.70	9.91
1.83	11.92	15.14	13.73	9.58	9.93
1.82	12.07	15.43	13.35	9.46	9.96
1.81	12.21	15.72	13.21	9.34	9.99
1.80	12.36	16.01	12.95	9.23	10.02
1.79	12.49	16.28	12.70	9.12	10.04
1.78	12.63	16.55	12.46	9.01	10.07
1.77	12.76	16.81	12.23	8.91	10.10
1.76	12.89	17.08	11.99	8.81	10.13
1.75	13.03	17.35	11.76	8.71	10.17
1.74	13.10	17.50	11.63	8.65	10.18
1.73	13.18	17.64	11.50	8.60	10.20
1.72	13.25	17.79	11.37	8.54	10.22
1.71	13.32	17.94	11.24	8.49	10.24
1.70	13.40	18.08	11.12	8.44	10.26
1.69	13.44	18.17	11.05	8.41	10.28
1.68	13.48	18.26	10.97	8.38	10.29
1.67	13.52	18.34	10.90	8.35	10.30
1.66	13.57	18.43	10.82	8.31	10.31
1.65	13.61	18.52	10.76	8.29	10.34
1.64	13.66	18.61	10.69	8.26	10.35
1.63	13.70	18.70	10.61	8.24	10.37
1.62	13.75	18.78	10.54	8.21	10.38
1.61	13.79	18.87	10.47	8.18	10.40

TABLE XX
EQUILIBRIUM PARTIAL PRESSURES ABOVE PuO_{2-x}
AT 1527°C (1800°K)

Stoichiometry (O/Pu)	-log P (atm)				
	(O)	(O ₂)	(Pu)	(PuO)	(PuO ₂)
2.00	5.69	3.16	22.72	13.54	8.67
1.99	7.71	7.58	18.69	11.54	8.72
1.98	8.21	8.59	17.71	11.06	8.73
1.97	8.37	8.91	17.40	10.91	8.74
1.96	8.55	9.26	17.06	10.74	8.75
1.95	8.72	9.60	16.72	10.57	8.76
1.94	8.90	9.97	16.36	10.40	8.76
1.93	9.08	10.32	16.02	10.23	8.78
1.92	9.27	10.71	15.65	10.06	8.79
1.91	9.46	11.08	15.30	9.89	8.82
1.90	9.68	11.52	14.87	9.68	8.83
1.89	9.83	11.83	14.57	9.54	8.84
1.88	9.99	12.14	14.28	9.40	8.86
1.87	10.15	12.45	13.99	9.27	8.88
1.86	10.30	12.77	13.70	9.13	8.90
1.85	10.46	13.08	13.41	9.00	8.93
1.84	10.61	13.39	13.12	8.87	8.95
1.83	10.77	13.70	12.84	8.74	8.97
1.82	10.92	14.01	12.55	8.61	9.00
1.81	11.08	14.32	12.27	8.49	9.03
1.80	11.23	14.63	12.00	8.37	9.06
1.79	11.38	14.92	11.74	8.25	9.09
1.78	11.52	15.20	11.48	8.14	9.12
1.77	11.66	15.49	11.23	8.02	9.15
1.76	11.80	15.77	10.96	7.90	9.17
1.75	11.95	16.06	10.73	7.81	9.23
1.74	12.02	16.21	10.59	7.75	9.24
1.73	12.10	16.36	10.46	7.69	9.26
1.72	12.18	16.52	10.33	7.64	9.28
1.71	12.25	16.67	10.20	7.58	9.30
1.70	12.33	16.82	10.07	7.53	9.33
1.69	12.37	16.91	9.99	7.50	9.34
1.68	12.41	16.99	9.92	7.47	9.35
1.67	12.45	17.07	9.85	7.44	9.36
1.66	12.50	17.16	9.78	7.42	9.38
1.65	12.54	17.24	9.72	7.39	9.39
1.64	12.58	17.32	9.65	7.36	9.41
1.63	12.62	17.40	9.58	7.34	9.42
1.62	12.66	17.48	9.52	7.31	9.44
1.61	12.72	17.56	9.46	7.29	9.45

TABLE XXI

<PuO_{2-x}> STOICHIOMETRIES THAT LEAD TO Pu VAPOR
PRESSURES HIGH ENOUGH TO FORM THE LISTED COMPOUNDS

Temp (°C)	<u><PuIr₂></u>	<u><PuPt₃></u>	<u><PuPt₄></u>	<u><PuPt₅></u>	<u><PuRh₃></u>
327	---	---	---	---	1.67
1027	1.62	---	---	---	1.67
1127	1.64	---	---	1.62	1.67
1227	1.67	1.61	1.63	1.69	1.67
1327	1.70	1.63	1.69	1.75	1.67
1427	1.72	1.66	1.72	1.77	1.67
1527	1.73	1.68	1.75	1.79	1.67