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LA-5202-MS

INFORMAL REPORT

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Thermodynamic Analysis of MHW Space Electric Power Generator

los alamos
scientific laboratory

of the University of California

LOS ALAMOS, NEW MEXICO 87544

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Printed in the United States of America. Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.95

LA-5202-MS
Informal Report
UC-4 & 23

ISSUED: March 1973

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by

R. A. Kent

This work sponsored by the Isotopes Technology Branch,
Space Nuclear Systems, US Atomic Energy Commission.

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THERMODYNAMIC ANALYSIS OF MHW SPACE ELECTRIC POWER GENERATOR

by

R. A. Kent

ABSTRACT

A thermodynamic analysis has been made for the proposed Multihundred Watt (MHW) isotopic electric power heat source for space missions. The heat source consists of plutonium dioxide fuel spheres encapsulated in welded iridium shells that in turn are surrounded by graphite ablation shields. Published data for the component systems Pu-O, Pu-Ir, Pu-C, Ir-O and Ir-C have been reviewed and employed to calculate the thermochemical behavior during mission lifetime.

I. INTRODUCTION

The proposed Multihundred Watt (MHW) helipak heat source assembly (HSA) consists of 24 individual fuel sphere assemblies (FSA) packaged in a 4 x 6 matrix. Each FSA consists of a 100 watt plutonium dioxide fuel sphere with a diameter of 1.465 in. The fuel spheres are encapsulated in welded iridium post-impact containment shells 0.020 in. thick with an o.d. of about 1.60 in. These iridium shells are provided with two small vents (diam = 0.002 in.) to allow for the release of helium gas generated by the fuel. The iridium shells are surrounded by 0.4 - 0.5 in. thick graphite impact shells. Small gaps between the various FSA components are provided to allow for thermal expansion effects.

The 24 FSA's are mounted in a 4 x 6 matrix surrounded by graphite and contained in a graphite ablation shield. The entire 2400 watt assembly is packaged in an iridium outer can provided with a pressure relief device to allow release of helium to outer space.

Various components of the heat source will operate at different temperatures throughout the mission lifetime.

The ambient surface temperature for an unclad fuel sphere is 400 - 500°C. After encapsulation in the iridium the ambient surface temperature rises to 700 - 900°C. The predicted operational temperature at the fuel-iridium interface will be 1350 - 1450°C. Finally the system could be subjected to a 5 min thermal ramp to 1750 - 1900°C during reentry.

The systems of interest from the point of view of chemical and thermodynamic stability are: Pu-O, Pu-Ir, Pu-C, Ir-O and Ir-C. This report consists of three parts. The first is a review of published thermodynamic data for the systems of interest. Thermodynamic data required for the calculations are listed for various temperatures in a series of tables in Appendix A. The second part consists of a series of calculations pertaining to possible reactions between the fuel and the other components in the system. These calculations are summarized in a series of tables in Appendix B. The last part of the report consists of a thermodynamic analysis for various temperatures of interest of the MHW heat source itself.

Throughout the text and appendices the following symbols are employed: < > for solids, () for gases and { } for liquids.

II. SUMMARY OF PUBLISHED DATA

A. Plutonium

1. Thermodynamic Functions. Thermodynamic functions for gaseous Pu are listed in Table A-1. These values, based on ideal gas behavior, were calculated by Feber and Herrick¹ from published data for 293 spectroscopic levels. Unfortunately, accurate heat capacity data for condensed Pu do not exist, especially at high temperatures. Rand² has reviewed available literature data and lists recommended values for the heat capacity as a function of temperature. He also recommends values for S_{298}^0 and for the heat and entropy terms associated with the various phase transitions. Rand's values, which correlate well with published vapor pressure data, were used to calculate the thermodynamic functions for <Pu> and {Pu} listed in Table A-2.

2. Vapor Pressure Measurements. Phipps et al.³, employed Ta Knudsen cells to measure the vapor pressure of Pu over the range 1392 - 1793°K. Mulford⁴ also employed Ta Knudsen cells in his target collection-type experiments over the range 1133 - 1792°K. In addition, he contained liquid Pu in internal cups of TaC, ThO₂ and MgO without finding any significant differences in vapor pressures. Kent⁵ showed that {Pu} vaporizes predominantly to the monomer in a mass spectrometric-Knudsen effusion study over the range 1426 - 1658°K. In the latter study, {Pu} was contained in both W Knudsen cells and ThO₂ internal cups. The vapor pressure data from the three studies are in good agreement and were combined⁵ with the thermodynamic functions listed in Tables A-1 and A-2 to yield the thermodynamic values for the vaporization of Pu listed in Table A-3.

B. Oxygen

Thermodynamics functions for (O) and (O₂) are taken from the JANAF Tables⁶ and are listed in Tables A-4 and A-5 respectively.

C. Plutonium Oxides

1. Thermodynamics Functions for <PuO₂>. The Pu-O system has been studied extensively and excellent review papers pertaining to data published prior to 1966 have been published by Rand², Mulford and Holley⁷, and Markin and Rand⁸.

For this work, the heat of formation for <PuO₂> is taken to be $\Delta H_{f,298}^0 = -252.35 \pm 0.17$ kcal/mole⁹. The heat capacity at 298°K is taken to be 16.4 eu^{10,11}.

The value for the absolute entropy at 298°K is somewhat uncertain. Sandenaw¹⁰ obtained a value of 16.3 eu, which is almost certainly too low. Westrum et al.¹² estimated a higher value based on the measured entropies of <NpO₂>, <ThO₂> and <UO₂>. Both <NpO₂> and <UO₂> show anomalies of magnetic origin near 25°K, while <ThO₂> does not. The authors suggest that the magnetic contribution to the entropy can be approximated by the spin-only contribution computed from the number of unpaired 5f electrons in Np⁴⁺ and U⁴⁺. There are no unpaired electrons for Th⁴⁺, thus it is diamagnetic, shows no magnetic ordering, and the entropy of <ThO₂> is purely lattice entropy. The observed excess entropy of <NpO₂> and <UO₂> over the lattice entropy (assumed the same as for <ThO₂>), is equal to $R \ln (2S + 1) + 0.85$ where S is the spin quantum number for the appropriate number of 5f electrons, and 0.85 is a constant presumed to include any electronic entropy. The value estimated for <PuO₂> is $S_{298}^0 = 19.7$ eu.

If the magnetic ordering does exist and Sandenaw failed to observe it because he did not attain low enough temperatures, one can add $R \ln 5 = 3.2$ to his value of 16.3 eu and obtain $S_{298}^0 = 19.5$ eu, in good agreement with Westrum's estimated value. Accordingly, the value of S_{298}^0 for <PuO₂> is taken to be 19.7 eu.

The heat content of <PuO₂> has been measured in the range 192 - 1400°K by Kruger and Savage¹³ and in the range 1500 - 2400°K by Ogard¹⁴. The agreement between the two sets of data in the overlap region 1400 - 1500°K is excellent. The heat content term derived from Ogard's data is

$$H_T^0 - H_{298}^0 = -6,360 + 18.0T, ^\circ K + 1.95 \times 10^{-3}T, ^\circ K + 2.46 \times 10^5/T, ^\circ K, \text{ (cal/mole)} \quad (1)$$

from which one derives

$$C_p = 18.0 + 3.9 \times 10^{-3} T - 2.46 \times 10^5/T^2, \text{ (eu)} \quad (2)$$

and

$$S_T^0 - S_{298}^0 = 18.0 \ln T/298 + 3.9 \times 10^{-3} (T-298) - 1.23 \times 10^5 (T^2-298^2/298^2 T^2), \text{ (eu)}. \quad (3)$$

Equations (1), (2) and (3), combined with the values

$\Delta H_{f,298}^0 = -252.35 \text{ kcal/mole}$ and $S_{298}^0 = 19.7 \text{ eu}$, were used to calculate the thermodynamic functions for $\langle \text{PuO}_2 \rangle$ listed in Table A-6.

2. Free Energy Values for $\langle \text{PuO}_{2-x} \rangle$ Species and

Partial Molar Free Energies for Oxygen in $\langle \text{PuO}_{2-x} \rangle$

Species. Rand², Markin and Rand⁸ and Ackermann, Faircloth and Rand¹⁵ have reviewed the thermodynamics pertaining to solid Pu-O species ranging from $\langle \text{PuO}_2 \rangle$ to $\langle \text{PuO}_{1.5} \rangle$. In addition, Markin and Rand⁸ measured the oxygen potentials over the single phase oxides between $\langle \text{PuO}_{1.61} \rangle$ and $\langle \text{PuO}_2 \rangle$ in the range 950 - 1350°K using an EMF technique. Finally, Ackermann et al.¹⁵ have measured the vapor pressure above the solid substoichiometric dioxide phase [$\sim 1.6 \leq \text{O/Pu} \leq 2.0$] and combined the vapor pressure data with the oxygen potentials⁸ to yield recommended values for the standard free energies of formation for various $\langle \text{PuO}_{2-x} \rangle$ compositions and for the partial molar free energies of atomic oxygen for these compositions.

Equations for the standard free energies of formation for various solid Pu-O compositions and for the partial molar free energies of atomic oxygen for these compositions are listed in Table A-7.

Values for the free energy of formation for $\langle \text{PuO}_2 \rangle$, listed in Table A-6, and those calculated from the first equation listed in Table A-7, are compared in Table A-8. The agreement is excellent especially in the range 1000-2200°K, the temperature range of interest for $^{238}\text{PuO}_2$ space power generators.

Therefore, the values given by the equations listed in Table A-7 are employed in the calculations that follow. It should be noted that these values lead to a good correlation of the various sets of vapor pressure data to be discussed below.

3. Free Energies of Formation for $\langle \text{PuO} \rangle$ and $\langle \text{PuO}_2 \rangle$.

Ackermann et al.¹⁵ recommend the following expressions for the free energies of formation for $\langle \text{PuO} \rangle$ and $\langle \text{PuO}_2 \rangle$:

For $\langle \text{PuO} \rangle$

$$\Delta G_f = -29,000 - 12.1 T, ^\circ\text{K (cal/mole)}, (1600 - 2200^\circ\text{K}) \quad (4)$$

and for $\langle \text{PuO}_2 \rangle$

$$\Delta G_f = -113,100 + 4.35 T, ^\circ\text{K (cal/mole)}, (1600 - 2200^\circ\text{K}). \quad (5)$$

The values given by Eqs. (4) and (5) lead to a good correlation of the published vapor pressure data. Thus, these values are used in the calculations that follow.

4. Calculated Vapor Pressure Values Above Various

$\langle \text{PuO}_{2-x} \rangle$ Compositions. Solid plutonium dioxide vaporizes in a complex manner to yield $\langle \text{PuO}_{2-x} \rangle$, $\langle \text{Pu} \rangle$, $\langle \text{PuO} \rangle$, $\langle \text{PuO}_2 \rangle$ and $\langle \text{O}_2 \rangle$. This section lists calculated partial pressure values for $\langle \text{Pu} \rangle$, $\langle \text{PuO} \rangle$, $\langle \text{PuO}_2 \rangle$ and $\langle \text{O}_2 \rangle$ above various $\langle \text{PuO}_{2-x} \rangle$ compositions.

a. $\langle \text{O}_2 \rangle$ Partial Pressures. Partial pressure values for $\langle \text{O}_2 \rangle$ were obtained from the relationship

$$\langle \text{O} \rangle = 1/2 \langle \text{O}_2 \rangle. \quad (6)$$

For Eq. (6) the free energy is given by

$$\begin{aligned} \Delta G &= -R T \ln K = -R T \ln P_{\text{O}_2}^{1/2} / P_{\text{O}} \\ &= -(R T/2) \ln P_{\text{O}_2} + \overline{\Delta G}_0. \end{aligned} \quad (7)$$

Finally

$$\ln P_{\text{O}_2} = -(2/R T) \left[-\Delta G_f(\text{O}) - \overline{\Delta G}_0 \right] \quad (8)$$

Values for $\Delta G_f(\text{O})$ are listed in Table A-4. Values for $\overline{\Delta G}_0$ were obtained from the equations listed in Table A-7.

b. $\langle \text{PuO} \rangle$ Partial Pressures. Partial pressure values for $\langle \text{PuO} \rangle$ were obtained from the relationship

$$\langle \text{PuO}_{2-x} \rangle = \langle \text{PuO} \rangle + (1-x) \langle \text{O} \rangle. \quad (9)$$

For Eq. (9) the free energy is given by

$$\begin{aligned} \Delta G &= \Delta G_f(\text{PuO}) + (1-x) \Delta G_f(\text{O}) - \Delta G_f \langle \text{PuO}_{2-x} \rangle \\ &= -R T \ln K = -R T \left[\ln P_{\text{PuO}} + (1-x) \ln P_{\text{O}} \right] \end{aligned} \quad (10)$$

Values for $\Delta G_f(\text{O})$ and $\Delta G_f \langle \text{PuO}_{2-x} \rangle$ are listed in Tables A-4 and A-7, respectively. Values for $\Delta G_f(\text{PuO})$ were calculated using Eq. (4). Values for P_{O} were obtained from a combination of Eqs. (7) and (8).

c. $\langle \text{Pu} \rangle$ Partial Pressures. Partial pressure values for $\langle \text{Pu} \rangle$ were obtained from the relationship

$$\langle \text{PuO}_{2-x} \rangle = \langle \text{Pu} \rangle + (2-x) \langle \text{O} \rangle \quad (11)$$

For Eq. (11) the free energy is given by

$$\Delta G = \Delta G_f(\text{Pu}) + (2-x) \Delta G_f(\text{O}) - \Delta G_f(\text{PuO}_{2-x}) \\ = -R T \ln K = -R T \left[\ln P_{\text{Pu}} + (2-x) \ln P_{\text{O}} \right] \quad (12)$$

Values for $\Delta G_f(\text{Pu})$ are listed in Table A-3.

d. (PuO₂) Partial Pressures. It was noted

above that <PuO_{2-x}> compositions vaporize in a complex manner to yield (Pu), (PuO), (PuO₂) and (O₂). The simplest relationship from which values for the partial pressure of (PuO₂) may be calculated is

$$\langle \text{PuO}_{2-x} \rangle + x(\text{O}) = (\text{PuO}_2). \quad (13)$$

For Eq. (13) the free energy is given by

$$\Delta G = \Delta G_f(\text{PuO}_2) - \Delta G_f(\text{PuO}_{2-x}) - x \Delta G_f(\text{O}) \\ = -R T \ln K = -R T \left[\ln P_{\text{PuO}_2} + x \ln P_{\text{O}} \right] \quad (14)$$

Values for $\Delta G_f(\text{PuO}_2)$ were calculated from Eq. (5).

e. Congruency of Vaporization. A combination of Eqs. (6), (9), (11) and (13) yields for the vaporization of <PuO_{2-x}> the overall relationship

$$\langle \text{PuO}_{2-x} \rangle = 1/3 (\text{PuO}_2) + 1/3 (\text{PuO}) + 1/3 (\text{Pu}) + \\ (1-x/2) (\text{O}_2) \quad (15)$$

Equilibrium partial pressure values of (PuO₂), (PuO), (Pu) and (O₂) calculated at different temperatures for various <PuO_{2-x}> compositions are listed in Tables A-9 to A-19. These values indicate that plutonium dioxide vaporizes congruently, that is, at a given temperature, the overall composition of the condensed and gaseous phases is the same. The exact composition of the congruent phase varies with temperature, the O/Pu ratio ranging from near 2.0 toward 1.61 as the temperature is increased. Values for the congruently vaporizing composition are listed as a function of temperature in Table A-20. Values are listed in Table A-20 both for closed systems (FSA's) and for open systems (Knudsen cells), where the more rapid effusion of lighter species such as (O₂) is taken into account.

Values for the partial pressures above different compositions are shown for 1440 and 1827°C in Figs. 1 and 2.

5. Experimental Vapor Pressures. All of the experimental data indicate that plutonium oxide vaporizes congruently as <PuO_{2-x}> between O/Pu ratios from 2.0 to about 1.6. In the two phase region <PuO_{1.61} - PuO_{1.50}> the vaporization is invariant.

a. <PuO_{2-x}>. A number of workers have measured the vapor pressure above <PuO_{2-x}> compositions and several of the more recent studies will be discussed. Ackermann et al.¹⁵, employed W and Re Knudsen cells over the range 1650 - 2070°K, collecting a known fraction of the effusate on Pt targets. They assumed the vapor to consist primarily of (PuO₂) and obtained for the effective pressure the equation

$$\log P_E (\text{atm}) = 7.50 - 29,260/T, ^\circ\text{K} \quad (16)$$

When <PuO₂> was first heated in the W and Re cells, reactions producing gaseous W and Re oxides were noted. No further W or Re reactions were observed even at higher temperatures. It was concluded that as <PuO₂> is reduced to <PuO_{2-x}> the oxygen activity drops to a level too low to produce significant reaction with the W or Re. The composition of the residue was estimated to be <PuO_{1.92}>.

Ohse and Ciani¹⁶ determined that the congruently vaporizing composition ranges from <PuO_{1.89}> at 1800°K to <PuO_{1.86}> at 2000°K and to <PuO_{1.84}> at 2200°K. These values are in excellent agreement with the calculated values listed in Table A-20. In addition, their data show that the lower phase boundary of the <PuO_{2-x}> phase is at <PuO_{1.61}> up to 2200°K.

Messier¹⁷ employed W Knudsen cells and an automatic-recording semimicro balance to measure the vapor pressure in the range 2070-2380°K. Again, assuming the vapor to be (PuO₂), he obtained for the effective pressure the equation

$$\log P_E (\text{atm}) = 8.38 - 30,970/T, ^\circ\text{K}. \quad (17)$$

The plutonium dioxide was reduced prior to the vapor pressure experiment in order that the congruent composition might be attained more quickly. During the experiment the sample composition changed from <PuO_{1.83}> to <PuO_{1.61}>. He also noticed a reaction between the W cell and <PuO_{2-x}> to produce gaseous W oxides. However, the extent of the reaction was too small to appreciably affect the vapor pressure calculations.

Messier noted that the phase boundary for the <PuO_{2-x}> phase is between <PuO_{1.58}> and <PuO_{1.62}> up to 2380°K. Thus, it is concluded that the lower phase boundary for <PuO_{2-x}> is <PuO_{1.61}> up to the liquidus.

Battles *et al.*¹⁸ employed a mass spectrometer to determine the partial pressures of (PuO) and (PuO₂) above <PuO_{2-x}>. They employed W and Re Knudsen cells and also observed the initial presence of gaseous W and Re oxides. These species disappeared as the plutonium dioxide was reduced. They also employed Ir Knudsen cells and observed no gaseous Ir-O species, indicating little or no reaction between <Ir> and <PuO₂>.

The partial pressure values for (PuO) and (PuO₂) are given by the equations

$$\log P_{\text{PuO}} (\text{atm}) = 6.71 - 27,810/T, ^\circ\text{K} \quad (18)$$

and

$$\log P_{\text{PuO}_2} (\text{atm}) = 7.75 - 30,330/T, ^\circ\text{K} \quad (19)$$

Only trace quantities of (Pu) were observed, in agreement with the calculated values listed in Tables A-9 to A-19.

The residues were found to be approximately <PuO_{1.83}> by analysis.

The mass spectrometric data may be compared to the data of Ackermann *et al.*, and Messier by calculating a P_E value from the mass spectrometric data via the equation

$$P_E (\text{atm}) = P_{\text{PuO}_2} + (M_{\text{PuO}}/M_{\text{PuO}_2})^{1/2} P_{\text{PuO}} \quad (20)$$

The pressures are in atmospheres and M is the molecular weight of the effusing species.

Kent¹⁹ employed W Knudsen cells and a mass spectrometer to measure the partial pressures of (PuO) and (PuO₂) over the range 2000 - 2200°K. Again, small amounts of gaseous W oxides were observed initially. The partial pressure values are given by the equation

$$\log P_{\text{PuO}} (\text{atm}) = 6.81 - 28,257/T, ^\circ\text{K} \quad (21)$$

and

$$\log P_{\text{PuO}_2} (\text{atm}) = 7.69 - 29,928/T, ^\circ\text{K} \quad (22)$$

Values of P_E were calculated using Eq. (20) in order to compare the mass spectrometric results with other data. The residue stoichiometry was <PuO_{1.83}>.

Kent also determined partial pressure values for (PuO) and (PuO₂) above a sample that initially had the stoichiometry <Pu_{0.45} Th_{0.55} O_{2.0}>. The temperature range covered was 2000 - 2230°K. The partial pressure values are given by the equations

$$\log P_{\text{PuO}} (\text{atm}) = 6.96 - 28,476/T, ^\circ\text{K} \quad (23)$$

and

$$\log P_{\text{PuO}_2} (\text{atm}) = 6.64 - 28,229/T, ^\circ\text{K} \quad (24)$$

No gaseous Th-O species were observed. The vapor pressure data suggest that when the solid solution is heated, the ThO₂ remains inert, and all of the sample reduction occurs with the plutonium oxide.

The values of P_E calculated from the data of the various workers are compared in Table A-21. The heat and entropy of vaporization terms obtained by the various workers are in reasonably good agreement except for the results of Messier. The values for P_E are in fair agreement.

One should note that each of these workers operated in a slightly different temperature range and obtained data at different average temperatures. When this is taken into account together with the fact that the stoichiometry of the congruently vaporizing composition changes with temperature, improved agreement is noted between the various sets of data. While the heat terms obtained from the slopes differ, the agreement in the pressure values indicates that the third law heats would be in good agreement.

The calculated partial pressure values, given for various stoichiometries in Tables A-9 to A-19, may be employed to calculate values for P_E according to the equation

$$P_E (\text{atm}) = P_{\text{PuO}_2} + (M_{\text{PuO}}/M_{\text{PuO}_2})^{1/2} P_{\text{PuO}} + (M_{\text{Pu}}/M_{\text{PuO}_2}) P_{\text{Pu}} \quad (25)$$

The experimental P_E values are compared with the calculated P_E values in Table A-22. The stoichiometries listed are those calculated for the experimental temperatures. The agreement for each set of data is reasonably good. The calculated O/Pu ratios are generally in agreement with those determined experimentally. The accuracy estimated for the calculated O/Pu ratios is ± 0.02 . This is the same accuracy that is routinely obtained by chemical analyses of experimental samples.

b. <PuO_{1.61} - PuO_{1.50}>. A number of workers have determined vapor pressures above the two-phase region <PuO_{1.61} - PuO_{1.50}>. Phipps *et al.*²⁰ employed a target-collection technique and heated plutonium dioxide in a Ta Knudsen cell over the range 1600 - 2100°K.

Their intent was to determine the vapor pressure above $\langle \text{PuO}_2 \rangle$, however, the Ta cell reduced the sample and they actually determined the pressure above $\langle \text{PuO}_{1.61} - \text{PuO}_{1.50} \rangle$. Assuming the vapor to consist of (PuO) , the value of P_E calculated from their data is given by the equation

$$\log P_E (\text{atm}) = 8.13 - 27,910/T, ^\circ\text{K}. \quad (26)$$

Ackermann *et al.*¹⁵, deliberately reduced $\langle \text{PuO}_2 \rangle$ to $\langle \text{PuO}_{1.61} - \text{PuO}_{1.50} \rangle$ by using a Ta Knudsen cell over the range 1650–2100^oK. They assumed the vapor to consist of (PuO) and obtained for P_E the equation

$$\log P_E (\text{atm}) = 8.09 - 27,800/T, ^\circ\text{K}. \quad (27)$$

They estimated the residue stoichiometry to be $\langle \text{PuO}_{1.52} \rangle$.

Battles *et al.*¹⁸, employed a mass spectrometer to determine the partial pressures of (Pu) , (PuO) and (PuO_2) over the range 1660–2150^oK. The sample was reduced to $\langle \text{PuO}_{1.54} \rangle$ prior to the vapor pressure measurements. They used W and Re Knudsen cells. The values for the partial pressures are

$$\log P_{\text{PuO}} (\text{atm}) = 7.10 - 25,806/T, ^\circ\text{K} \quad (28)$$

and

$$\log P_{\text{PuO}_2} (\text{atm}) = 5.69 - 27,575/T, ^\circ\text{K} \quad (29)$$

and for (Pu)

$$\log P_{\text{Pu}} (\text{atm}) = 5.27 - 24,090/T, ^\circ\text{K}. \quad (30)$$

The partial pressure data were used to calculate values of P_E , based on (PuO) as the effusing species, from the equation

$$P_E (\text{atm}) = P_{\text{PuO}} + (M_{\text{PuO}_2}/M_{\text{PuO}})^{1/2} P_{\text{PuO}_2} + (M_{\text{Pu}}/M_{\text{PuO}})^{1/2} P_{\text{Pu}}. \quad (31)$$

Kent¹⁹ used a mass spectrometer and Ta Knudsen cells over the range 1870–1960^oK to determine the partial pressures above a sample that had an initial stoichiometry of $\langle \text{Pu}_{0.9} \text{Th}_{0.1} \text{O}_{2.0} \rangle$. No gaseous Th-O species were observed. Values for P_E , based on (PuO) as the effusing species, were calculated using Eq.(31).

The vapor pressure data obtained by the various workers are compared in Table A-23. With the exception of the heat and entropy terms obtained by Battles *et al.*, the agreement is excellent. The partial pressure values calculated for the two-phase region $\langle \text{PuO}_{1.61} - \text{PuO}_{1.50} \rangle$ (Table A-19), were used to calculate P_E values according to Eq. (31).

These calculated P_E values are compared to the experimental values in Table A-24.

6. Summary. It is obvious that more experimental data are required for the Pu-O system: (1) Extended mass spectrometric studies should be performed to determine more exactly the relative partial pressure values above various compositions, (2) The kinetics of the reduction of $\langle \text{PuO}_{2.00} \rangle$ to $\langle \text{PuO}_{2-x} \rangle$ should be determined and (3) The data on oxygen partial molar free energies should be extended above 2000^oK. However, the agreement between the calculated vapor pressure values listed in Tables A-9 to A-19 and the experimental values is reasonably good. Therefore, the calculated values listed in Tables A-9 to A-19 can be used to perform an adequate thermodynamic analysis for proposed space isotopic electric power generator systems.

D. Carbon

Graphite vaporizes to (C) , (C_2) , (C_3) , (C_4) and (C_5) . Thermodynamic functions for solid graphite are taken from the JANAF tables⁶ and are listed in Table A-25. Thermodynamic functions for (C) , (C_2) and (C_3) are also taken from the JANAF tables and are listed in Tables A-26, A-27, and A-28, respectively. Functions for (C_4) and (C_5) are not listed as the vapor pressures of these species above graphite below 2400^oK are too low to affect the following calculations.

E. Iridium

The melting point of Ir metal, 2447^oC, is used as a secondary reference point for the International Practical Temperature Scale of 1968²¹. Paule and Margrave²², using a Langmuir Free-Evaporation technique, determined the vapor pressure of Ir metal over the range 2140–2480^oK and obtained

$$\log P_{\text{Ir}} (\text{atm}) = 7.24 - 33,680/T, ^\circ\text{K}. \quad (32)$$

Thermodynamic functions for $\langle \text{Ir} \rangle$ are taken from Stull and Sinke²³ and are listed in Table A-29. They also list values for (Ir) , based on ideal gas calculations. These values for (Ir) , however, when combined with the values for $\langle \text{Ir} \rangle$ yield iridium vapor pressures that are too high when compared to the experimental data of Paule and Margrave. For this work, the vapor pressures of iridium are calculated from Eq. (32).

F. Iridium - Carbon System

A number of workers have investigated the Ir-C system. The results of these studies are in good agreement and indicate that the solubility of C in Ir is low and that Ir and C do not form a compound below 2400°C. However, Ir and C form a eutectic that melts about 150°C below the melting point of <Ir>.

Nadler and Kempter²⁴ report an Ir-C eutectic to be at 2296 ± 16°C. They also report that the alloy Ir-30Rh forms a eutectic with carbon at 1932 ± 17°C.

Lowell *et al.*²⁵ employed x-ray diffraction techniques to determine whether an Ir-C compound formed. They used two types of samples, 0.010 in. diam Ir wire coated with a graphite slurry, and 0.025 in. diam graphite rods coated with a slurry of Ir powder. Both types of samples were heated over the range 800-3000°C and x-ray diffraction patterns were obtained at 100° intervals. In all cases, these patterns indicated the presence of only <Ir> and <C>. Thus, they concluded that no compound was formed.

Harmon²⁶ employed DTA and Pirani techniques and found the Ir-C eutectic composition to be 6.5 at. % C at 2302 ± 15°C. Harmon concluded that the Ir-C phase diagram is a simple eutectic type and that the solubility of C in Ir is low. He further found that the presence of a small amount of Si impurity, 1 wt. %, lowered the eutectic melting temperature to about 2150°C.

Reavis²⁷ employed DTA techniques and determined the Ir-C eutectic point to be at 2296 ± 20°C.

Arnault and McLellan²⁸ have determined the solubility of C in Ir to be low over the range 820-1260°C, in agreement with the data from the other studies. Values for the solubility of C in Ir, calculated from these data, are listed in Table A-30.

G. Iridium - Oxygen System

1. Solid Oxides. The only stable solid iridium oxide appears to be <IrO_{2-x}> where x varies from about 0 to 0.5. Cordfunke and Meyer²⁹ measured the dissociation pressure of (O₂) above <IrO₂> and concluded that, above 800°C, <IrO₂> vaporizes according to the reaction

They claim that the reduced oxide varies from <IrO_{1.97}> at 825°C, to <IrO_{1.60}> at 1050°C. These authors give for the (O₂) pressure above the reduced solid <IrO_{2-x}>

$$\log P_{\text{O}_2} (\text{atm}) = 8.606 - 11,750/T, ^\circ\text{K} \quad (34)$$

and above the stoichiometric oxide <IrO₂>

$$\log P_{\text{O}_2} (\text{atm}) = 9.508 - 12,696/T, ^\circ\text{K}. \quad (35)$$

Equation (34) leads to an oxygen dissociation pressure of 1 atm at 1062°C. From their data, Cordfunke and Meyer obtain $\Delta H_{f,298}^\circ = -64.0$ kcal/mole for <IrO₂>.

Schäfer and Heitland³⁰ measured the (O₂) dissociation pressure above <Ir> + <IrO₂> and obtained

$$\log P_{\text{O}_2} (\text{atm}) = 4.437 - 11,319/T, ^\circ\text{K} + 1.7666 \log T - 1.3965 \times 10^{-2} T - 0.0437 \times 10^5 / T^2. \quad (36)$$

Equation (36) leads to an (O₂) pressure of 1 atm at 1124°C. From their data they obtain $\Delta H_{f,298}^\circ = -52.4$ kcal/mole for <IrO₂>.

More recently, Bell *et al.*³¹ have determined the (O₂) dissociation pressure to be 1 atm at 1105°C. Further, they claim that the composition of <IrO₂> deviates very little from stoichiometry in the range 800-1050°C and that <IrO₂> decomposes according to the equation

From their data, they obtain $\Delta H_{f,298}^\circ = -57.4$ kcal/mole and $\Delta S_{f,298}^\circ = -43.8$ eu. for <IrO₂>.

Obviously, further study is required. The one conclusion that seems certain is that <IrO₂> decomposes when heated and the (O₂) pressure reaches 1 atm at about 1100°C.

2. Gaseous Oxides. A number of workers have studied the formation of gaseous iridium oxides, the most definitive study being that of Norman, *et al.*³² They employed a mass spectrometer, over the range 1830-2020°K, to determine the gases formed when (O₂) was introduced into an Ir Knudsen cell at elevated temperatures. The gases formed were (IrO), (IrO₂) and (IrO₃). At 2033°K, with an (O₂) pressure of 4.3 × 10⁻⁴ atm, they determined the (IrO₂) pressure to be 1.9 × 10⁻⁸ atm and the (IrO₃) pressure to be 3.1 × 10⁻⁹ atm. The (IrO) pressure at 2033°K was about 1/50 that of (IrO₂). They found that (IrO₂) and (IrO₃) formed according to the equations:

and

$$\langle \text{Ir} \rangle + 1.5 (\text{O}_2) = (\text{IrO}_3). \quad (39)$$

The free energy term obtained for Eq. (35) was

$$\Delta G_{f,1900} (\text{IrO}_3) = 48,500 - 3.9 T, ^\circ\text{K} \text{ (cal/mole)} \quad (40)$$

while that for Eq. (39) was

$$\Delta G_{f,1900} (\text{IrO}_3) = 3,500 + 13.1 T, ^\circ\text{K} \text{ (cal/mole)}. \quad (41)$$

Three groups of workers have employed the transpiration technique to measure the formation of (Ir_xO_y) when they passed (O_2) at pressures ranging from 0.05 to 1 atm. over $\langle \text{Ir} \rangle$.

Schäfer and Heitland³⁰ assumed that in the range 1000-1200°C the vapor species was (IrO_3) formed according to Eq. (39). From their data, one obtains

$$\Delta G_f (\text{IrO}_3) = 4,052 + 10.77 T, ^\circ\text{K} \text{ (cal/mole)}. \quad (42)$$

Cordfunke and Meyer³³, working in the range 1170-1450°C, obtained

$$\Delta G_f (\text{IrO}_3) = 4,654 + 10.22 T, ^\circ\text{K} \text{ (cal/mole)}. \quad (43)$$

These authors also measured the formation of (Ir_xO_y) when (O_2) was passed over $\langle \text{IrO}_2 \rangle$. However, they appear to have ignored the variation in $\langle \text{IrO}_2 \rangle$ stoichiometry which they had determined by their own dissociation pressure studies²⁹.

Atcock and Hooper³⁴, working in the range 1200-1400°C, obtained

$$\Delta G_f (\text{IrO}_3) = 4,146 + 10.8 T, ^\circ\text{K} \text{ (cal/mole)}. \quad (44)$$

However, they suggested that the gaseous species might have been (Ir_2O_3) .

Tentative confirmation of this possibility was provided by the mass spectrometric study of Kurlakose, Kent and Margrave³⁵. At 900°C and 1 atm (O_2) pressure, these authors observed extensive transfer of Ir via gaseous oxides. When these species were introduced into the mass spectrometer, the ion Ir_2O_2^+ , presumably a fragment from (Ir_2O_3) , was observed. These authors also noted that the iridium oxide species attacked the SiO_2 molecular leak assembly.

The free energy of formation terms for the formation of (IrO_3) , Eqs. (42), (43) and (44), obtained from the transpiration studies are in good agreement. However, if these data are extrapolated to 1900°K, they yield an average value for the free energy of formation for (IrO_3) of 24.43 kcal/mole in very poor agreement with the value

given by Eq. (41) obtained in the mass spectrometric study,³² i.e., 30.39 kcal/mole. This difference leads to a factor of about 5 in the calculated (IrO_3) pressure at 1900°K. Thus, one must conclude that (1) the mass spectrometric data are wrong, (2) the reaction mechanism at the temperature of the transpiration experiments is more complex than previously realized and perhaps involves the formation of (Ir_2O_3) , or (3) there is a change in the oxidation mechanism in going from the temperature range of the transpiration studies to that of the mass spectrometric studies. The second and third conclusions are believed more likely in view of the results obtained in the kinetic studies described below.

In summary, there are apparently four gaseous oxides of iridium. The major low temperature species apparently is (IrO_2) and (IrO_3) and (IrO) become important at higher temperatures. The species (Ir_2O_3) forms exothermally and is usually a minor species. However, at high oxygen pressures and at low temperatures it may be possible for (Ir_2O_3) to predominate.

3. Kinetic Studies. A number of workers have studied the kinetics of oxidation of $\langle \text{Ir} \rangle$ in (O_2) streams. Krier and Jaffee³⁶ heated $\langle \text{Ir} \rangle$ in a stream of air (oxygen pressure = 0.2 atm) and published rate loss values for three temperatures, 1000°, 1200° and 1400°C. They employed a flow rate of about 2 ft/min.

Kurlakose, Kent and Margrave^{35, 37} heated $\langle \text{Ir} \rangle$ over the range 1200-2220°C in a He-O_2 stream (pressure O_2 = 0.2 atm) moving at about 103 ft/min. They also heated $\langle \text{Ir} \rangle$ over the range 1490-2220°C in a He-O_2 stream (pressure O_2 = 0.02 atm) moving at about 15 ft/min. These data are summarized in Table A-31. They also performed a number of experiments to determine the effects of oxygen pressure, oxygen flow rate, temperature and the presence of water vapor in the flow stream.

Rexer^{38, 39} heated $\langle \text{Ir} \rangle$ in Ar-O_2 streams, employing much higher flow rates. He measured the linear recession rates over the range 1150-1900°C, employing an $\text{Ar-H}_2\text{O}$ stream (pressure O_2 = 0.19 atm) moving at about 12,000 ft/min. He also determined one point at a flow rate of 12,000 ft/min (pressure O_2 = 0.019 atm) and 3 points over the range 1540-1900°C with a flow rate of

35,000 ft/min (pressure $O_2 = 0.93$ atm). The results are listed in Table A-32. In general, Rexer's results are in good agreement with those obtained by Kuriakose, Kent and Margrave.

The data of Krier and Jaffee and of Kuriakose *et al.*, obtained at an (O_2) pressure of 0.2 atm and the data of Rexer obtained at an oxygen pressure of 0.19 atm are plotted in Fig. 3. Also plotted in Fig. 3 are values for the recession rate calculated, assuming the vapor species to be (IrO_3), from the free energy values obtained in the mass spectrometric experiment, Eq. (41), and in the transpiration experiments, Eqs. (42), (43) and (44). In a similar manner the data of Kuriakose *et al.*, obtained at an oxygen pressure of 0.02 atm, and the single point of Rexer, obtained at an oxygen pressure of 0.019 atm, are compared to the calculated recession rates in Fig. 4.

The calculated recession rates were obtained as follows. If the vapor species is indeed (IrO_3), the reaction is given by Eq. (39) and the free energy is

$$\Delta G = -RT \ln K \\ = -RT \left[\ln P_{IrO_3} - 1.5 \ln P_{O_2} \right]. \quad (45)$$

Given a value for ΔG , calculated from the mass spectrometric data in the range 1557-1747°C, or from the transpiration data in the range 1000-1400°C, together with the experimental value for the (O_2) pressure, one can calculate a value for the (IrO_3) pressure. This value can be related to the iridium recession rate by use of the Langmuir equation for free evaporation

$$Z = 44.4 (P) (M/T)^{1/2} \quad (46)$$

where M is the molecular weight of the vaporizing species [here (IrO_3)], T is the temperature in degrees Kelvin, P is the pressure in atmospheres, and Z is the flux in units of g/cm²-sec. Iridium has a density of 22.42 g/cm³, therefore, a piece of $\langle Ir \rangle$ 1 cm long by 1 cm long, weighing 1 g, will be 0.01756 in. thick. Thus, Eq. (46) can be converted to yield the recession rates in units of mil/h.

The curves plotted in Fig. 3 demonstrate that the experimental data of Kuriakose *et al.*, obtained employing a flow rate of 100 ft/min, and the data of Rexer, obtained a flow rate of 12,000 ft/min, are in reasonably good

agreement. Data obtained in both studies indicate that the recession rates vary with the flow rate, increasing quite rapidly at first, and then more slowly, being approximately proportional to the logarithm of the flow rate over a large range. The recession rates obtained by Krier and Jaffee are much lower than those obtained in the other studies. This may simply be because their flow rate, 2 ft/min, was too low. Two other conclusions that may be drawn from the data of Kuriakose *et al.*, and Rexer are that, (1) the recession rates vary nearly linearly with (O_2) pressure and (2) the presence of water vapor increases the recession rates.

The calculated recession rates obtained from the mass spectrometric data agree with the experimental values better than do the rates calculated from the transpiration data. In neither case, however, is the agreement good. This discrepancy in the values for the calculated and experimental recession rates could be due to several factors; (1) There is a kinetic barrier that slows down the recession rate, (2) the vaporization mechanisms are complex and involve species other than (IrO_3) and (3) the data are wrong. The first and second factors are the most likely causes of the differences. The data of Kuriakose *et al.*, imply that (1) at low temperatures, the oxidation rate is low and is surface-reaction controlled, and (2) at higher temperatures, the recession rates are less temperature dependent and are controlled by mass transfer through a boundary layer.

4. Summary. Unfortunately, the phase diagram for the Ir-O system is complex and it has not been fully determined. For many regions of the phase diagram no data exist. In addition, for those regions where more than one study was made, the comparative agreement is often poor. Obviously much work needs to be done before one can confidently predict behavior in a complex system such as Pu-O-Ir-C. For the analysis of the MHW system that follows, it was decided to employ those values that lead to the worst case *i.e.*, maximum calculated interaction between the plutonium oxide fuel and the iridium clad.

H. Plutonium - Iridium System

Olsen⁴⁰ has heated $\langle \text{PuIr}_2 \rangle$ in a tungsten Knudsen cell over the temperature range 1680–2210°K, and determined that $\langle \text{PuIr}_2 \rangle$ vaporizes according to the reaction

The (Pu) pressure is given by

$$\log P_{\text{Pu}} (\text{atm}) = 4.26 - 26,529/T, ^\circ\text{K}. \quad (48)$$

From the vapor pressure data, the free energy for Eq. (47) is calculated:

$$\Delta G_{1900} = 121,391 - 19.49 T, ^\circ\text{K} (\text{cal/mole}). \quad (49)$$

When Eqs. (47) and (49) are combined with the values listed in Table A-3 for the vaporization of {Pu}, one obtains for the reaction

the free energy of formation

$$\Delta G_{1900} = -41,071 - 3.24 T, ^\circ\text{K} (\text{cal/mole}). \quad (51)$$

In the calculations that follow, values for the free energy of formation for $\langle \text{PuIr}_2 \rangle$ are taken from Eq. (51). Values for the dissociation pressure of (Pu) above $\langle \text{PuIr}_2 \rangle$ are taken from Eq. (48).

I. Plutonium - Carbon System

The results of several investigations of the thermodynamic properties of the Pu-C system have recently been published⁽⁴¹⁻⁵⁰⁾. The most recent study by Campbell, Kent and Leary⁵⁰ combined mass spectrometric and emf techniques to determine the free energy of formation values for $\langle \text{PuC}_{0.8} \rangle$, $\langle \text{PuC}_{1.5} \rangle$ and $\langle \text{PuC}_2 \rangle$. The results of this study⁵⁰ may be summarized as follows.

The mass spectrometric data indicate that there are four regions of the Pu-C phase diagram that give rise to invariant but non-congruent vaporization. These are, monocarbide plus sesquicarbide, sesquicarbide plus carbon, sesquicarbide plus dicarbide and dicarbide plus carbon. At a given, constant temperature the vaporization behavior is as follows. The pressure of (Pu) above {Pu} at 1700°K is 4.8×10^{-8} atm. As carbon is added to the system, the (Pu) gradually decreases as the plutonium activity is lowered. The (Pu) pressure is 4.5×10^{-8} atm at a C/Pu ratio of 0.70. As the monocarbide is approached from the low-carbon side the (Pu) pressure decreases rapidly, reaching 4.0×10^{-8} atm at a C/Pu ratio of 0.76.

Between the compositions $\langle \text{PuC}_{0.84} \rangle$ and $\langle \text{PuC}_{1.4} \rangle$ the pressure is constant at 9.8×10^{-7} atm so long as the condensate contains both mono- and sesquicarbide. Between $\langle \text{PuC}_{1.4} \rangle$ and $\langle \text{PuC}_{1.5} \rangle$ the (Pu) pressure again decreases rapidly, becoming 5.3×10^{-7} atm at $\langle \text{PuC}_{1.46} \rangle$. The low-carbon phase boundary for the sesquicarbide curves toward lower C/Pu ratios as the temperature increases, passing through $\langle \text{PuC}_{1.46} \rangle$ at $1300 \pm 50^\circ\text{C}$ and $\langle \text{PuC}_{1.41} \rangle$ at $1500 \pm 50^\circ\text{C}$. Between the compositions $\langle \text{PuC}_{1.5} \rangle$ and $\langle \text{C} \rangle$ the (Pu) pressure at 1700°K remains constant at 2.2×10^{-8} atm so long as the condensate consists of both sesquicarbide and carbon. At $1660 \pm 10^\circ\text{C}$, the sesquicarbide transforms to the dicarbide. Above 1660°C and at C/Pu ratios of 1.5 or greater, the (Pu) pressure is independent of composition so long as the condensate consists of both sesqui- and dicarbide. At the composition $\langle \text{PuC}_{1.84 \pm 0.02} \rangle$, and above 1660°C , the condensate consists of $\langle \text{PuC}_2 \rangle$ plus $\langle \text{C} \rangle$ and the (Pu) pressure is invariant. At temperatures above 2100°K, trace amounts (<1%) of $\langle \text{PuC}_2 \rangle$ were observed.

The following thermodynamic values are taken from the mass spectrometric and emf study of Campbell, Kent and Leary⁵⁰.

Below 1660°C plutonium sesquicarbide decomposes to the elements according to the equation

The (Pu) pressure, over the range 1670–1933°K, is given by

$$\log P_{\text{Pu}} (\text{atm}) = 4.468 - 20,598/T, ^\circ\text{K}. \quad (53)$$

The free energy for the decomposition reaction is given by

$$\Delta G_{1842} = 94,252 - 20.44 T, ^\circ\text{K} (\text{cal/mole}). \quad (54)$$

When Eqs. (52) and (54) are combined with the values listed in Table A-3 for the vaporization of {Pu}, one obtains for the reaction

the free energy expression

$$\Delta G_{1842} = -14,098 - 2.20 T, ^\circ\text{K} (\text{cal/mole}). \quad (56)$$

The corresponding expression obtained in the emf study over the range 920–1060°K, is

$$\Delta G_{1060} = -16,650 + 0.35 T, ^\circ\text{K} (\text{cal/mole}). \quad (57)$$

Equations (56) and (57) yield at 1000°K, $\Delta G_f = -16.3$ kcal/mole, in excellent agreement.

Plutonium monocarbide, when heated, decomposes according to the reaction

The pressure of (Pu), over the range 1450-1850°K, is given by

$$\log P_{\text{Pu}} (\text{atm}) = 5.118 - 18,919/T, ^\circ\text{K} \quad (59)$$

which leads to a free energy value for Eq. (58) of

$$\Delta G_{1662} = 86,570 - 23.42 T, ^\circ\text{K} (\text{cal/mole}). \quad (60)$$

When the free energy of formation expression for <PuC_{0.9}> is combined with those for the formation of <PuC_{1.5}> and the vaporization of {Pu}, one obtains for the reaction

the value

$$\Delta G_{1662} = -11,487 - 0.74 T, ^\circ\text{K}. \quad (62)$$

The corresponding expression obtained in the emf study over the range 920-1060°K is

$$\Delta G_{982} = -14,059 + 1.60 T, ^\circ\text{K}. \quad (63)$$

The average third law value for the heat of formation for <PuC_{0.9}>, obtained in the mass spectrometric and emf studies, Eqs. (62) and (63), is $\Delta H_{298}^0 = -11.1$ kcal/mole. This is in excellent agreement with the value $\Delta H_{298}^0 = -11.4$ kcal/mole, obtained in a recent bomb-calorimeter experiment.⁴⁹

At C/Pu ratios greater than 1.98 and at temperatures above 1660°C, plutonium dicarbide decomposes according to the reaction

The (Pu) pressure over the range 1933-2140°K is given by

$$\log P_{\text{Pu}} (\text{atm}) = 3.787 - 19,288/T, ^\circ\text{K} \quad (65)$$

and the free energy for the decomposition reaction is given by

$$\Delta G_{2056} = 88,258 - 17.33 T, ^\circ\text{K} (\text{cal/mole}). \quad (66)$$

When the free energy expression for the decomposition of <PuC₂> is combined with that for the vaporization of {Pu} one obtains for the reaction

the value

$$\Delta G_{2056} = -7,574 - 5.58 T, ^\circ\text{K} (\text{cal/mole}). \quad (68)$$

J. Plutonium - Oxygen - Carbon System

The Pu-O-C system has not been studied as extensively as the Pu-O or Pu-C systems. Mulford et al.⁵¹ have reported the existence of a single-phase field having a stoichiometry ranging from about <PuO_{0.35}C_{0.64}> to <PuO_{0.65}C_{0.35}>. They present lattice parameters for the various compositions. Potter^{52, 53} reported that a composition near <PuO_{0.5}C_{0.5}> vaporizes congruently over the range 1600-1820°K, according to the reaction

where

$$\Delta G = 150,150 - 54.5 T, ^\circ\text{K} (\text{cal/mole}). \quad (70)$$

From this, one calculates for the free energy of formation for <PuO_{0.5}C_{0.5}>

$$\Delta G_f = -65,500 + 11.5 T, ^\circ\text{K} (\text{cal/mole}). \quad (71)$$

K. Plutonium - Oxygen - Carbon - Iridium System

Very little information has been published for the Pu-O-C-Ir system. However, considerable data pertaining to the MHW system will result from the compatibility tests being conducted at LASL and other laboratories.

Reavis⁵⁴ has conducted a series of experiments in which he heated <PuO₂> in an iridium crucible contained in a graphite susceptor. The results are summarized in Table A-33. The reaction at about 2275°C is believed to be due to the formation of the Ir-C eutectic, allowing the <PuO₂> to react directly with the outer graphite container to produce (CO).

L. Miscellaneous

The thermodynamic functions for (H₂), (H₂O), (CO) and (CO₂) used in the calculations that follow are taken from the JANAF tables.⁶ These functions are listed in Tables A-34 through A-37.

III. CALCULATION OF POSSIBLE REACTIONS

A. Basis for Calculations

The thermodynamic data discussed in section II above and summarized in Appendix A (Tables A-1 to A-37) were employed to calculate equilibrium free energy values for the reactions for <PuO₂> and various components of the MHW system.

B. $\langle \text{PuO}_2 \rangle + \langle \text{C} \rangle$

Under normal operational conditions the $\langle \text{PuO}_2 \rangle$ fuel in the FSA should never come in contact with graphite. Iridium does not form a compound with carbon and the only reported Ir-C eutectic temperature is higher than predicted reentry temperatures. Reactions between the fuel and the graphite could occur if the iridium shell was mechanically breached or if impurities were present in sufficient quantities to significantly lower the Ir-C eutectic temperature.

Reactions between $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ to form $\langle \text{PuO}_{2-x} \rangle$ plus $\langle \text{CO} \rangle$, as well as the various plutonium carbides, are summarized in Appendix B (Tables B-1 to B-17). The calculated free energy values are based on equilibrium conditions for closed systems and indicate that for such systems, no reactions occur below about 1400°K. At higher temperatures, reactions can occur and lower O/Pu ratios become more favored as the temperature increases. For any temperature at which reaction can occur, the reaction can be driven further than predicted by the free energy values if the products can be removed. Such would occur if the breached FSA were vented to vacuum (space). In fact, plutonium carbides are often made by vacuum reduction of $\langle \text{PuO}_2 \rangle$ by graphite.

Temperatures at which the equilibrium ΔG value becomes negative for the reaction between $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ to form a specific reduced oxide are summarized in Table B-78.

C. $\langle \text{PuO}_2 \rangle + \langle \text{CO} \rangle$

Carbon monoxide should not be generated within an FSA. However, $\langle \text{CO} \rangle$ could be produced by reaction between the graphite and an impurity outside an FSA or if the iridium shell were breached allowing the graphite to come in contact with the fuel. Reactions between $\langle \text{PuO}_2 \rangle$ and $\langle \text{CO} \rangle$ at various temperatures are summarized in Appendix B (Tables B-18 to B-34) and indicate no reaction occurs under equilibrium conditions below about 1700°K. At higher temperatures, however, $\langle \text{CO} \rangle$ will reduce $\langle \text{PuO}_2 \rangle$ to $\langle \text{PuO}_{2-x} \rangle$ with the O/Pu ratio decreasing as the temperature increases. An experimental program has been initiated at LASL to determine the exact amount of reduction that occurs.

Temperatures at which the equilibrium ΔG value becomes negative for the reaction between $\langle \text{PuO}_2 \rangle$ and $\langle \text{CO} \rangle$ to form a specific reduced oxide are summarized in Table B-78.

D. $\langle \text{PuO}_2 \rangle + (\text{H}_2)/(\text{H}_2\text{O})$

Workers at LASL and other laboratories routinely adjust sample stoichiometries by (H_2) reduction of $\langle \text{PuO}_2 \rangle$ above 1300°C. Because moisture can be introduced when FSA's are handled, the reactions between $\langle \text{PuO}_2 \rangle$ and $(\text{H}_2)/(\text{H}_2\text{O})$ are summarized in Tables B-35 to B-51. Again, the temperatures at which the equilibrium ΔG value becomes negative for the reaction between $\langle \text{PuO}_2 \rangle$ and (H_2) to form a specific suboxide are summarized in Table B-78. Obviously, below these temperatures, (H_2O) will oxidize $\langle \text{PuO}_{2-x} \rangle$.

E. $\langle \text{PuO}_2 \rangle + \langle \text{Ir} \rangle$

1. $\langle \text{IrO}_2 \rangle$. Free energy values for the reactions between $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ to form $\langle \text{IrO}_2 \rangle$ are summarized in Tables B-52 to B-58. These values indicate that below the decomposition temperature of $\langle \text{IrO}_2 \rangle$, $\langle \text{PuO}_2 \rangle$ will not react with $\langle \text{Ir} \rangle$ to form $\langle \text{IrO}_2 \rangle$. However, should any $\langle \text{IrO}_2 \rangle$ be initially present as an impurity in the iridium, it will decompose on heating with possibly serious consequences. This will be discussed in section IV below.

2. (IrO_2) . Free energy values for the reactions between $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ to form (IrO_2) are summarized in Tables B-59 to B-67. These values indicate that, under equilibrium conditions in a closed system, no reaction occurs. However, under operating conditions the fuel sphere will release (O_2) that can react with the iridium shell to produce (IrO_2) . These reactions will be discussed below in section IV.

3. (IrO_3) . Free energy values for the possible reactions between $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ to form (IrO_3) are summarized in Tables B-68 to B-77. For this analysis the free energy of formation values for (IrO_3) were taken from the transpiration data, Eqs. (42), (43) and (44), because these data yield the worst case, maximum iridium transport as (IrO_3) . These free energy data predict no reaction between $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ to form (IrO_3) . However, at high temperatures, (O_2) from the dissociation of

the fuel could react with the iridium shell to produce (IrO₃). These reactions will be discussed below in section IV.

4. (Ir₂O₃ and (IrO)). Mass spectrometric data³² indicate that (IrO) is not an important species at any temperature predicted for normal FSA operation including reentry. While (Ir₂O₃) is known to exist, no thermodynamic data are available. Given the conditions under which this species was observed,³⁵ low temperatures and high oxygen pressures, it is not likely that (Ir₂O₃) will be formed under normal FSA operating conditions.

F. <Ir> + <C>

No reaction to form a compound will occur below 3000°C. The reported eutectic temperature, 2296 ± 20°C, is well above predicted reentry temperatures.

IV THERMODYNAMIC ANALYSIS FOR A MHW-FSA

A. Basic Assumptions and Equations

1. Fuel. For the analyses that follow, a number of assumptions have been made about the fuel.

a. The fuel consists of a 100 watt sphere with the initial stoichiometry <PuO_{2.0}>. The ²³⁸Pu isotope contributes 80 at.% of the total plutonium content, thus a 100 watt sphere weighs 251.5g (0.93 moles <PuO_{2.0}>).

b. The sphere diameter at 20°C (68°F) is 1.465 in. Sphere diameters at other temperatures are calculated by correcting for the thermal expansion of plutonium dioxide⁵⁵. Values for the diameter and surface area of a 100 watt sphere are listed for various temperatures in Table C-1.

c. The equilibrium partial pressure values for (O₂), (PuO₂), (PuO) and (Pu) above any <PuO_{2-x}> composition are those given for various temperatures in section II (Tables A-9 to A-19).

d. The rate at which the species (O₂), (PuO₂), (PuO) and (Pu) leave the surface of a fuel sphere maybe calculated from the Langmuir equation

$$Z = (44.36) (P) (M/T)^{1/2} \quad (72)$$

where Z is the loss rate in g/cm² sec, P is the pressure in atmospheres, T is the temperature in degrees Kelvin and M is the molecular weight of the vaporizing species.

For these calculations, instant diffusion of material through the sphere to the surface is assumed. Also, the evaporation coefficient is assumed to be unity. Thus, the calculated rate loss values will be maximum ones.

2. Iridium Shell. Some assumptions have also been made for the iridium shell.

a. The iridium shell is 0.02 in. thick and has an i.d. of 1.56 in. This leads to an inner surface area of about 49.3 cm². For the sake of simplicity, it is assumed that the free volume inside the FSA is constant over a temperature range, i.e., both the fuel sphere and the iridium shell expand as the temperature increases.

b. The iridium shell is provided with two vents, 0.002 in. diam., so that the total vent or orifice area is 4.054x10⁻⁵ cm². Ideal gas behavior is assumed so that the effusion of gases through the vents is given by the Knudsen equation

$$P = \frac{Kw}{AtW} \left[\frac{T}{M} \right]^{1/2} \quad (73)$$

where P is the pressure in atmospheres, T is the temperature in degrees Kelvin and w is the weight, in g, of material of molecular weight M, that effuses through an area of A cm² in a time t sec. The constant K is derived from the kinetic theory of gases and is equal to 0.02256. The ideal Knudsen equation assumes a hole with infinitely thin knife-edges. The vents of the FSA will exert a channeling effect and the number of molecules escaping will be less than predicted. Theoretical corrections for this channel effect have been derived by Clausius and for a long narrow tube the correction is W = 8r/3l⁵⁶. For the FSA vents, r = 0.001 in, l = 0.02 in and W = 0.1333.

B. 1700°K

The predicted maximum surface temperature for the fuel sphere under operating conditions is about 1700°K (1427°C). The calculated equilibrium partial pressures for (O₂), (PuO₂), (PuO) and (Pu) above <PuO_{2.0}> at this temperature are listed in Table A-9. The effects of the vaporization of each of these species will be discussed.

1. Oxygen. The data in Table A-9 indicate that the predominate vapor species is (O₂) with a partial pressure of 2.5x10⁻⁶ atm. This will lead to a reduction of the O/Pu ratio in the fuel. The amount of oxygen given off

as (O_2) can be calculated from the Langmuir equation, Eq. (72), to be

$$Z_{O_2} = (44.36) (2.5 \times 10^{-6}) (32/1700)^{1/2} \\ = 1.52 \times 10^{-6} \text{ g/cm}^2 \text{ sec.} \quad (74)$$

At 1700°K , the sphere surface area is 44.88 cm^2 (Table C-1) so that the (O_2) loss rate becomes

$$Z_{O_2} = 6.8 \times 10^{-4} \text{ g/sec.} \quad (75)$$

a. Fuel Reduction. In order for 0.93 moles of $\langle \text{PuO}_2 \cdot 0 \rangle$ to be reduced to 0.93 moles of $\langle \text{PuO}_1 \cdot 98 \rangle$, a total of $0.93 \times 0.02 \times 16 = 0.3 \text{ g}$ of oxygen must be removed. From Eq. (75) one finds that the minimum time required to move this amount of oxygen is 441 sec or 7.4 min. This is a minimum time because of the assumptions noted above in section IV. A, i.e., instant diffusion and an evaporation coefficient of unity.

Once the fuel is reduced to $\langle \text{PuO}_1 \cdot 98 \rangle$, the (O_2) pressure becomes $4.9 \times 10^{-11} \text{ atm}$ (Table A-10). The oxygen loss rate is then

$$Z_{O_2} = (44.36) (4.9 \times 10^{-11}) (32/1700)^{1/2} \\ = 3.0 \times 10^{-10} \text{ g/cm}^2 \text{ sec,} \quad (76)$$

or for a surface area of 44.88 cm^2

$$Z_{O_2} = 1.3 \times 10^{-8} \text{ g/sec.} \quad (77)$$

For the sphere to be reduced from $\langle \text{PuO}_1 \cdot 98 \rangle$ to $\langle \text{PuO}_1 \cdot 95 \rangle$, a total of $0.93 \times 0.03 \times 16 = 0.45 \text{ g}$ of oxygen must be removed. The minimum time required for this to occur, calculated from Eq. (77) is $3.46 \times 10^7 \text{ sec}$ or 1.1 y. Similar calculations indicate much longer times are required for the fuel to be reduced from $\langle \text{PuO}_1 \cdot 95 \rangle$ to $\langle \text{PuO}_1 \cdot 92 \rangle$, etc. Thus, unless a driving force such as vacuum operation is present, the fuel would not be reduced to the congruently vaporizing composition of $\langle \text{PuO}_1 \cdot 90 \rangle$, (Table A-20), during any expected mission lifetime at 1700°K .

b. Effusion of (O_2) from the FSA. If the (O_2) partial pressure above $\langle \text{PuO}_2 \cdot 0 \rangle$ at 1700°K is $2.5 \times 10^{-6} \text{ atm}$, the amount effusing out the vents, calculated from the Knudsen equation, Eq. (73), is

$$2.5 \times 10^{-6} = \frac{0.02256 w}{(4.054 \times 10^{-9}) (0.1333)} \left(\frac{1700}{32} \right)^{1/2} \\ \text{or} \\ w_{O_2} = 8.2 \times 10^{-11} \text{ g/sec.} \quad (78)$$

This value may be compared to the (O_2) loss rate at the sphere surface, $6.8 \times 10^{-4} \text{ g/sec}$, [Eq. (75)]. For the fuel to be reduced from $\langle \text{PuO}_2 \rangle$ to $\langle \text{PuO}_1 \cdot 98 \rangle$, 0.3 g of

oxygen must be removed. The time required for this amount to effuse out of the FSA, calculated from Eq. (78) is about 116 y. Thus, an (O_2) overpressure in the FSA will prevent further reduction of the fuel and the actual fuel stoichiometry will stabilize somewhere between $\langle \text{PuO}_2 \cdot 0 \rangle$ and $\langle \text{PuO}_1 \cdot 98 \rangle$.

c. Effect of (O_2). The (O_2) pressure at 1700°K , corresponding to an (O_2) pressure of $2.5 \times 10^{-6} \text{ atm}$, is $7.0 \times 10^{-8} \text{ atm}$. The total (O) loss rate from the sphere surface, calculated using the Langmuir equation, is $Z_O = 1.4 \times 10^{-5} \text{ g/sec}$. Thus, the (O) contribution to the oxygen loss rate is about 2% that due to (O_2).

d. Effect of Helium. At 1700°K , the (O_2) loss rate from the surface of a $\langle \text{PuO}_2 \rangle$ sphere is $6.8 \times 10^{-4} \text{ g/sec}$, [Eq. (75)]. This value corresponds to 1.3×10^{19} particles/sec. Current program plans call for a vacuum degassing of the FSA's prior to launch so that they will begin the mission without appreciable helium inventories. Plutonium dioxide (80 at. % ^{238}Pu), generates helium at $0.53 \text{ std cm}^3/\text{y-g}$. This corresponds to $4.2 \times 10^{-6} \text{ cm}^3/\text{sec}$ for 0.93 moles $\langle \text{PuO}_2 \rangle$ or 1.1×10^{14} helium atoms/sec. Thus, $\langle \text{PuO}_2 \rangle$ will generate approximately 10^5 (O_2) molecules per (He) atom, and helium generation should not appreciably affect the (O_2) loss rate. After an extended period of time, the (He) pressure will come to an equilibrium value of about 0.1 atm. This would further suppress the reduction of the fuel by (O_2) evaporation.

e. Reactions with the Iridium Shell. It was noted above in section III and in Tables B-52 to B-77, that the calculated ΔG values predict no reaction between $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$. However, the (O_2) released by the fuel might react with the $\langle \text{Ir} \rangle$ shell, thus removing some of the free oxygen in the system and providing a driving force for further reduction of the fuel.

For the reaction

the free energy at 1700°K , calculated from Eqs. (42), (43) and (44), is

$$\Delta G = -RT \ln K \\ = -RT \left[\ln P_{\text{IrO}_3} - 1.5 \ln P_{\text{O}_2} \right] \\ = 22.31 \text{ kcal/mole.} \quad (80)$$

The ΔG values obtained from the transpiration studies are used for this analyses because they lead to the worst case, i.e., maximum (IrO_3) generation. The equilibrium partial pressure of (O_2) above $\langle \text{PuO}_2 \rangle$ is 2.5×10^{-6} atm at 1700°K . Thus, the (IrO_3) partial pressure is $P_{\text{IrO}_3} = 5.4 \times 10^{-12}$ atm. When this value is introduced into the Langmuir equation, the maximum (IrO_3) loss rate from the shell is calculated to be

$$Z_{\text{IrO}_3} = 9.0 \times 10^{-11} \text{ g/cm}^2 \text{ sec.} \quad (81)$$

If the iridium shell inner surface area is 49.3 cm^2 , this leads to

$$Z_{\text{IrO}_3} = 4.4 \times 10^{-9} \text{ g/sec.} \quad (82)$$

This (IrO_3) generation rate would consume about 8.8×10^{-10} g/sec of oxygen, or in the first 441 sec, 3.9×10^{-7} g of oxygen out of the 0.3 g generated. This reaction of the (O_2) with the $\langle \text{Ir} \rangle$ at 1700°K would not affect the (O_2) generation rate.

The (IrO_3) generation rate, Eq. (81), corresponds to a worst case loss rate of iridium from the inner shell surface of about 0.11 g/y, or about 0.04 mil/y. The rate at which the (IrO_3) could effuse out of the FSA is about 1.5×10^{-8} g/y.

For the reaction

the free energy at 1700°K , calculated from Eq. (40), is

$$\begin{aligned} \Delta G &= -RT \left[\ln P_{\text{IrO}_2} - \ln P_{\text{O}_2} \right] \\ &= 41.87 \text{ kcal/mole.} \end{aligned} \quad (84)$$

If the (O_2) partial pressure is 2.5×10^{-6} atm, the (IrO_2) partial pressure is calculated to be $P_{\text{IrO}_2} = 1.0 \times 10^{-11}$ atm. The maximum (IrO_2) loss rate from the iridium shell, calculated using the Langmuir equation, is

$$Z_{\text{IrO}_2} = 1.7 \times 10^{-10} \text{ g/cm}^2 \text{ sec} \quad (85)$$

or, if the inner shell surface area is 49.3 cm^2 ,

$$Z_{\text{IrO}_2} = 8.4 \times 10^{-9} \text{ g/sec.} \quad (86)$$

This (IrO_2) generation rate would require 1.2×10^{-10} g of oxygen/sec. Thus, in 441 sec, 4.9×10^{-8} g of the 0.3 g of oxygen produced would be taken up. This would not affect the (O_2) generation rate.

The (IrO_2) generation given by Eq. (85) translates to a maximum iridium loss from the inner shell surface of about 0.23 g/y or 0.08 mil/y. The effusion rate of (IrO_2) through the vents is calculated to be about 2.7×10^{-8} g/y.

At 1700°K , reactions between the iridium shell and the (O_2) generated by the fuel should not affect the FSA operation.

2. (PuO_2) . The equilibrium partial pressure of (PuO_2) above $\langle \text{PuO}_2 \rangle$ at 1700°K is 2.1×10^{-10} atm (Table A-9). The maximum loss rate from the sphere surface is calculated to be

$$Z_{\text{PuO}_2} = 3.7 \times 10^{-9} \text{ g/cm}^2 \text{ sec} \quad (87)$$

or for the total sphere

$$Z_{\text{PuO}_2} = 1.7 \times 10^{-7} \text{ g/sec.} \quad (88)$$

The sphere would lose a maximum of 5.2 g (PuO_2) /y.

The amount that could effuse through the vents is 6.3×10^{-7} g/y. The (PuO_2) generation rate, Eq. (87), leads to a calculated value for the deposit of $\langle \text{PuO}_2 \rangle$ on the iridium shell of less than 0.1 mil/y. This should not affect FSA operation.

3. (PuO) . The equilibrium partial pressure of (PuO) above $\langle \text{PuO}_2 \rangle$ at 1700°K is 8.1×10^{-15} atm (Table A-9). This leads to a calculated maximum loss rate of (PuO) from the sphere of 2.0×10^{-4} g/y. The amount that can effuse through the vents is 2.3×10^{-11} g/y. Vaporization of (PuO) from the fuel sphere will not affect FSA operation.

4. (Pu) . The equilibrium partial pressure of (Pu) above $\langle \text{PuO}_2 \rangle$ at 1700°K is 9.7×10^{-24} atm (Table A-9). This value leads to a maximum calculated loss rate of (Pu) from the sphere surface of 2.3×10^{-13} g/y. The rate of (Pu) effusion through the vents is calculated to be 2.7×10^{-20} g/y. These rates will not affect FSA operation.

The (Pu) pressure above $\langle \text{PuIr}_2 \rangle$ at 1700°K is 4.5×10^{-12} atm, [Eq. (48)]. The (Pu) given off by the fuel will not react with the shell to form $\langle \text{PuIr}_2 \rangle$ unless the (Pu) pressure above the fuel is greater than 4.5×10^{-12} atm. For this to occur, the fuel must be reduced to a stoichiometry of about $\langle \text{PuO}_{1.7} \rangle$ (Tables A-16 and A-17). Given the oxygen results discussed above, this is impossible.

5. Iridium and Carbon. The vapor pressure of (Ir) above <Ir> at 1700°K is 2.7×10^{-13} atm, Eq. (32). This corresponds to a loss rate from the iridium shell of about 6.2×10^{-3} g/y or 0.002 mil/y, and will not affect FSA operation. The vapor pressures of the gaseous carbon species (Tables A-26, A-27 and A-28) at 1700°K are all lower than 1.7×10^{-14} atm, and are too low to have any effect. No Ir-C compound forms at 1700°K and the solubility of carbon in <Ir> at this temperature (Table A-30) is too low to have any effect.

6. Impurity Effects. Certain impurities could, if present, affect some of the results calculated above. One of the potentially worst situations could arise if the iridium shell contained significant amounts of <IrO₂> prior to being welded around the fuel sphere. The iridium in an FSA shell weighs about 65g. If the iridium contained 250 ppm of oxygen as <IrO₂> there would be 0.11g or 5.1×10^{-4} moles of <IrO₂> present. One mole of <IrO₂> decomposes at about 1400°K to release 1 atm of (O₂)³¹. If ideal gas behavior is followed, 5.1×10^{-4} moles of <IrO₂> will produce 5.1×10^{-4} atm of (O₂) at 1400°K. As the temperature increases to 1700°K, the (O₂) pressure becomes $(1700/1400) \times (5.1 \times 10^{-4}) = 6.2 \times 10^{-4}$ atm. If this oxygen reacts with the iridium shell, the (IrO₃) loss rate, calculated from Eq. (80), becomes 3.5×10^{-7} g/cm² sec, a value more than 3800 times that caused by the (O₂) from the fuel, Eq (81). In a similar manner, the (IrO₂) loss rate is calculated to be 4.1×10^{-8} g/cm² sec, more than 250 times the rate caused by (O₂) from the fuel. It is obvious that the iridium shell material should be treated to reduce the oxygen content prior to encapsulation.

7. Summary. The impurity levels within the FSA system, especially oxygen in the iridium shell, should be kept as low as possible. If this is accomplished, the thermodynamic stability of a <PuO₂> - <Ir> - <C> FSA system at 1700°K is such that missions could last as long as the half-life of the ²³⁸Pu, 88y. At this temperature the fuel stoichiometry will stabilize between <PuO₂.₀> and <PuO₁.₉₈>.

The maximum rates at which (O₂) and (PuO₂) are given off from the fuel surface are summarized in Table C-2.

These values are calculated assuming the fuel to be <PuO₂.₀>. Also summarized in Table C-2 are maximum generation rates for (IrO₃) and (IrO₂) at the inner surface of the iridium shell. These values are calculated assuming that all of the oxygen comes from <PuO₂.₀> fuel. The presence of any impurities such as <IrO₂> would cause these values to increase.

The maximum rates at which (O₂), (PuO₂), (IrO₃) and (IrO₂) can effuse out the vents are summarized in Table C-3. No vent covers are considered and the fuel was assumed to be <PuO₂.₀>. No impurity effects were considered.

C. 1800°K

If the maximum operating temperature of the fuel sphere is increased to 1800°K the partial pressures within the system, especially that of (O₂), will increase significantly. The following discussion illustrates the effect on FSA operation.

1. Oxygen. The values listed in Table A-9 indicate that at 1800°K, the predominate vapor species above <PuO₂.₀> is (O₂) with a partial pressure of 2.8×10^{-4} atm. This value, combined with the Langmuir equation, yields an (O₂) loss rate of

$$Z_{O_2} = 1.7 \times 10^{-3} \text{ g/cm}^2 \text{ sec.} \quad (89)$$

The total sphere surface area at 1800°K is 45.0 cm² (Table C-1) so that the total (O₂) loss rate from the sphere is

$$Z_{O_2} = 7.5 \times 10^{-2} \text{ g/sec.} \quad (90)$$

This rate is about 110 times that calculated for 1700°K, [Eq. (75)].

a. Fuel Reduction. In order for the fuel to be reduced from <PuO₂.₀> to <PuO₁.₉₈>, a total of 0.3 g of oxygen must be removed. The minimum time for this to occur, based on Eq. (90), is 4 sec.

Once the fuel is reduced to <PuO₁.₉₈>, the (O₂) pressure becomes 2.6×10^{-3} atm (Table A-10) and the loss rate of (O₂) from the sphere becomes

$$Z_{O_2} = 6.9 \times 10^{-7} \text{ g/sec.} \quad (91)$$

The minimum time required to reduce <PuO₁.₉₈> to <PuO₁.₉₅> through evaporation of (O₂) from the sphere, i.e., to remove 0.45g of oxygen is about 7.5d.

Once the fuel is reduced to $\langle \text{PuO}_1.95 \rangle$, the (O_2) pressure becomes 2.5×10^{-10} atm (Table A-11) and the (O_2) loss rate becomes

$$Z_{\text{O}_2} = 6.7 \times 10^{-8} \text{ g/sec.} \quad (92)$$

In order for the sphere to be reduced from 0.93 moles of $\langle \text{PuO}_1.95 \rangle$ to 0.93 moles of $\langle \text{PuO}_1.92 \rangle$, $0.93 \times 0.03 \times 16 = 0.45$ g of oxygen must be removed. The minimum time required for this to occur, based on Eq. (92), is 78d.

In a similar manner, the minimum time required for the sphere to be reduced from $\langle \text{PuO}_1.92 \rangle$ to $\langle \text{PuO}_1.90 \rangle$ is calculated to be 1.8y. Thus, when the operating temperature is increased from 1700°K to 1800°K there is a much greater tendency for the fuel to be reduced. The actual stoichiometry of the fuel will depend primarily on the (O_2) effusion rate through the vents.

b. Effusion of (O_2) from the FSA. If the solid fuel composition is $\langle \text{PuO}_2 \rangle$, the (O_2) partial pressure will be 2.8×10^{-4} atm, and the maximum amount of (O_2) that will effuse out the vents is

$$w_{\text{O}_2} = 9.2 \times 10^{-9} \text{ g/sec.} \quad (93)$$

The time required to remove 0.3g of oxygen so that the fuel is reduced from $\langle \text{PuO}_2 \rangle$ to $\langle \text{PuO}_1.98 \rangle$ is about 1 y. Likewise, the time required to effuse 0.45g of oxygen from the FSA, so that the fuel is reduced from $\langle \text{PuO}_1.98 \rangle$ to $\langle \text{PuO}_1.95 \rangle$ is 1.7×10^5 y. Thus, the fuel stoichiometry will stabilize at approximately $\langle \text{PuO}_1.98 \rangle$.

c. Effect of (O_2) . The partial pressure of (O_2) above $\langle \text{PuO}_2 \rangle$ at 1800°K is 2.0×10^{-6} atm. The maximum loss rate from the sphere surface is

$$Z_{\text{O}} = 3.8 \times 10^{-4} \text{ g/sec.} \quad (94)$$

When this value is compared with the (O_2) loss rate given by Eq (90), the (O) contribution to the total oxygen loss rate is found to be 0.5% that from (O_2) .

d. Effect of Helium. The (He) generation rate was calculated above in section B to be 1.1×10^{14} atoms/sec. The (O_2) loss rate given by Eq. (90) corresponds to 1.4×10^{21} molecules/sec or about 10^7 (O_2) molecules per (He) atom. The (He) generation should have little effect.

e. Reactions with the Iridium Shell. The free energy for the reaction between $\langle \text{Ir} \rangle$ and (O_2) to form (IrO_3) , Eq.(79), is 23.37 kcal/mole at 1800°K . Above $\langle \text{PuO}_2 \rangle$ fuel the (O_2) partial pressure is 2.8×10^{-4} atm and the (IrO_3) partial pressure is calculated to be 6.8×10^{-9} atm. This value leads to an (IrO_3) loss rate of

$$Z_{\text{IrO}_3} = 1.1 \times 10^{-7} \text{ g/cm}^2 \text{ sec,} \quad (95)$$

or from the entire inner surface of the iridium shell

$$Z_{\text{IrO}_3} = 5.4 \times 10^{-6} \text{ g/sec.} \quad (96)$$

This (IrO_3) generation rate will not consume sufficient (O_2) to affect the loss of (O_2) from the fuel surface. However, this rate is about 990 times that caused by reaction with (O_2) from the fuel at 1700°K , [Eq. (82)].

The (IrO_3) generation rate given by Eq. (95) corresponds to an iridium loss of about 49 mil/y. This value is calculated for the worst case, assuming the fuel remains $\langle \text{PuO}_2 \rangle$. Actually, as noted above, the fuel will be rather quickly reduced to about $\langle \text{PuO}_1.98 \rangle$. Thus the corresponding (O_2) and (IrO_3) partial pressures will be lowered. Nevertheless, an FSA operated at 1800°K will have a shorter useful mission lifetime than it would operated at 1700°K .

The free energy of formation for (IrO_2) at 1800°K is 41.87 kcal/mole. Above $\langle \text{PuO}_2 \rangle$ the (O_2) partial pressure is 2.8×10^{-4} atm and the (IrO_2) partial pressure is calculated to be 2.6×10^{-9} atm. This corresponds to a loss rate from the inner shell surface of

$$Z_{\text{IrO}_2} = 2.0 \times 10^{-6} \text{ g/sec} \quad (97)$$

or about 19 mil/y of iridium. Again, this is a value calculated for the worst case and the (IrO_2) generation rate will decrease as the fuel is reduced to $\langle \text{PuO}_1.98 \rangle$. Note however, that the value given in Eq. (97) for the generation rate of (IrO_2) is more than 200 times the value calculated at 1700°K , [Eq. (86)].

2. (PuO_2) . The equilibrium partial pressure of (PuO_2) above $\langle \text{PuO}_2 \rangle$ at 1800°K is 2.0×10^{-9} atm (Table A-9). The maximum loss rate from a fuel sphere, with a surface area of 45.0 cm^2 (Table C-11), is

$$Z_{\text{PuO}_2} = 1.5 \times 10^{-6} \text{ g/sec.} \quad (98)$$

This value is about 9 times greater than that calculated at 1700°K [Eq. (88)].

If the fuel remained $\langle \text{PuO}_2 \rangle$ indefinitely the sphere could lose up to 47g (PuO_2) per y, with a resultant maximum deposit of $\langle \text{PuO}_2 \rangle$ on the iridium shell of 1 mil/y. However, the maximum effusion rate of (PuO_2) through the vents is calculated to be only 6×10^{-6} g/y. In addition, the (PuO_2) partial pressure will decrease as the fuel is reduced to $\langle \text{PuO}_{1.98} \rangle$. It is unlikely that at 1800°K, vaporization of (PuO_2) will seriously affect FSA operation.

3. (PuO). The equilibrium partial pressure of (PuO) above $\langle \text{PuO}_2 \rangle$ at 1800°K is 2.9×10^{-14} atm (Table A-9). This corresponds to a maximum loss rate from the sphere surface of 7.4×10^{-4} g/y and a maximum effusion through the vents of 6×10^{-10} g/y. Neither will affect FSA operation.

4. (Pu). The equilibrium partial pressure of (Pu) above $\langle \text{PuO}_2 \rangle$ at 1800°K is 1.9×10^{-25} atm (Table A-9). This corresponds to a maximum loss rate from the sphere surface of 4×10^{-13} g/y and a maximum effusion through the vents of 5×10^{-20} g/y. FSA operation will not be affected. The partial pressure of (Pu) will be too low for the formation of $\langle \text{PuIr}_2 \rangle$ unless the fuel is reduced to $\langle \text{PuO}_{1.7} \rangle$. The oxygen results discussed above indicate that the fuel should not be reduced below $\langle \text{PuO}_{1.98} \rangle$.

5. Iridium-Carbon. No Ir-C compound will form and the solubility of $\langle \text{C} \rangle$ in $\langle \text{Ir} \rangle$ at 1800°K is very low (Table A-30). The vapor pressures of (Ir), 3.4×10^{-12} atm, and the various (C_x) species, all below 3×10^{-13} atm, are too low to have any appreciable effect.

6. Impurity Effects. The same general comments apply as discussed above for operation at 1700°K, except that the effects of impurities such as $\langle \text{IrO}_2 \rangle$ will be more deleterious at the higher temperature.

7. Summary. Considering only the thermochemical stability within the $\langle \text{PuO}_2 \rangle$ - $\langle \text{Ir} \rangle$ - $\langle \text{C} \rangle$ system, an FSA could probably be operated at 1800°K. However, the vaporization of (O_2) from the fuel and the generation of (IrO_3) and (IrO_2) at the walls of the iridium shell would be greater than at 1700°K. This would lead to a shorter mission lifetime. In addition, if the FSA is to operate at 1800°K, impurities such as $\langle \text{IrO}_2 \rangle$ must be kept at lower

levels than for 1700°K operation. Accordingly, FSA operating temperatures below 1750°K (1480°C) would seem to be recommended.

Maximum generation rates for (O_2), (PuO_2), (IrO_3) and (IrO_2) are summarized in Table C-2. Maximum effusion rates for these species through the FSA vents are summarized in Table C-3. In each case the fuel was assumed to be $\langle \text{PuO}_{2.0} \rangle$ and no impurity effects were considered.

D. 1600°K

The equilibrium partial pressures above $\langle \text{PuO}_{2.0} \rangle$ at 1600°K are listed in Table A-9 as $P_{\text{O}_2} = 1.2 \times 10^{-8}$ atm, $P_{\text{PuO}_2} = 1.7 \times 10^{-11}$ atm, $P_{\text{PuO}} = 2.0 \times 10^{-15}$ atm and $P_{\text{Pu}} = 4.5 \times 10^{-24}$ atm. These values are considerably lower than at 1700°K and useful FSA lifetime would be extended accordingly.

1. Oxygen. An (O_2) partial pressure above $\langle \text{PuO}_{2.0} \rangle$ of 1.2×10^{-8} atm corresponds to a loss rate from the sphere of

$$Z_{\text{O}_2} = 7.5 \times 10^{-8} \text{ g/cm}^2 \text{ sec.} \quad (99)$$

At 1600°K, the sphere surface area is 44.76 cm² (Table C-1) so that the total (O_2) generation rate becomes

$$Z_{\text{O}_2} = 3.4 \times 10^{-6} \text{ g/sec.} \quad (100)$$

This rate is 200 times lower than that given by Eq. (75) for 1700°K.

a. Fuel Reduction. In order for the fuel to be reduced from $\langle \text{PuO}_{2.0} \rangle$ to $\langle \text{PuO}_{1.98} \rangle$, 0.3 g of oxygen must be removed from a sphere containing 0.93 moles $\langle \text{PuO}_{2.0} \rangle$. The minimum time required for this to occur is calculated from Eq. (100) to be 2.6d. The rate at which (O_2) with a partial pressure of 1.2×10^{-8} atm can effuse through the vents is

$$w_{\text{O}_2} = 4.1 \times 10^{-13} \text{ g/sec.} \quad (101)$$

Thus, it would require 2.3×10^5 y to remove 0.3 g of (O_2) from the FSA and the (O_2) overpressure would maintain the fuel at a stoichiometry near $\langle \text{PuO}_{2.0} \rangle$.

b. Effect of (O). The partial pressure of (O) corresponding to an (O_2) partial pressure of 1.2×10^{-8} atm is 1.4×10^{-9} atm. This leads to an (O) loss rate from the sphere surface of

$$Z_{\text{O}} = 3.2 \times 10^{-7} \text{ g/sec.} \quad (102)$$

Thus, the (O) generation rate contributes about 9% of the total oxygen loss from the sphere. The maximum rate at which (O) can effuse through the vents is

$$w_O = 4.8 \times 10^{-14} \text{ g/sec.} \quad (103)$$

It would take 2.5×10^5 y for 0.3 g of oxygen to escape the FSA as (O) and the generation of (O) will have little effect on the fuel stoichiometry.

c. Effect of Helium. The helium generation rate, calculated above in section B, is 1.1×10^{14} atoms/sec. The (O₂) generation rate given by Eq. (100) corresponds to 6.4×10^{16} molecules/sec so that the effect of (He) would be to add about 0.2% to the rate at which oxygen effuses out of the FSA.

d. Reactions with the Iridium Shell. The free energy of formation of (IrO₃) at 1600°K is calculated from Eqs. (42), (43) and (44) to be 21.25 kcal/mole. This value, combined with an (O₂) partial pressure of 1.2×10^{-8} atm, leads to a maximum (IrO₃) partial pressure of 1.6×10^{-15} atm. This value corresponds to an (IrO₃) loss rate of

$$Z_{\text{IrO}_3} = 2.8 \times 10^{-14} \text{ g/cm}^2 \text{ sec} \quad (104)$$

or, for an inner shell area of 49.3 cm²,

$$Z_{\text{IrO}_3} = 1.4 \times 10^{-12} \text{ g/sec.} \quad (105)$$

This (IrO₃) generation rate is more than 3000 times lower than that given by Eq. (82) for 1700°K and will not affect the rate at which (O₂) leaves the sphere surface. The maximum <Ir> recession rate is calculated to be 1.2×10^{-5} mil/y.

The free energy of formation for <IrO₂> at 1600°K, given by Eq. (40) is 42.26 kcal/mole. Given an (O₂) pressure of 1.2×10^{-8} atm, the maximum (IrO₂) partial pressure is calculated to be 2.0×10^{-14} atm. The maximum rate loss is

$$Z_{\text{IrO}_2} = 1.6 \times 10^{-11} \text{ g/sec.} \quad (106)$$

This value corresponds to a maximum <Ir> recession rate of 1.6×10^{-4} mil/y.

2. (PuO₂). The maximum calculated loss rate of (PuO₂) from the sphere surface is

$$Z_{\text{PuO}_2} = 1.4 \times 10^{-8} \text{ g/sec.} \quad (107)$$

This corresponds to only 0.4 g/y. The maximum loss rate of (PuO₂) through the vents is calculated to be 5.3×10^{-8} g/y. Vaporization of (PuO₂) at 1600°K will not affect FSA operation.

3. (PuO). The maximum loss rate for (PuO) from the sphere surface is calculated to be

$$Z_{\text{PuO}} = 1.6 \times 10^{-12} \text{ g/sec.} \quad (108)$$

This corresponds to 5×10^{-5} g/y. The maximum rate at which (PuO) can effuse out the vents is 5.8×10^{-12} g/y. FSA operation will not be affected.

4. (Pu). The maximum (Pu) loss rate from the fuel sphere is

$$Z_{\text{Pu}} = 3.4 \times 10^{-21} \text{ g/sec.} \quad (109)$$

This corresponds to 1.1×10^{-13} g/y. The maximum rate at which (Pu) can effuse through the vents is 3×10^{-22} g/y. The vapor pressure of (Pu) above <PuIr₂> at 1600°K is calculated from Eq. (48) to be 4.8×10^{-13} atm. In order for the (Pu) partial pressure above the fuel to be high enough to overcome this, the fuel would have to be reduced to <PuO_{1.70}> (Table A-17). Vaporization of (Pu) at 1600°K will not affect FSA operation.

5. Iridium-Carbon. At 1600°K, the vapor pressure of (Ir) above <Ir> is given by Eq. (32) as 1.5×10^{-14} atm. The vapor pressures of the various (C_x) species (Tables A-26, A-27 and A-28) are all less than 3×10^{-16} atm. None of these will affect FSA operation.

6. Impurity Effects. Although the effects of impurities will be less deleterious at 1600°K than at 1700°K, it is still necessary to maintain the amounts of impurities such as <IrO₂> at low levels. For example, if the iridium shell contains only 100 ppm oxygen as <IrO₂>, the generation rate of (IrO₃) at 1600°K will be about 10⁶ times greater than that given by Eq. (104).

7. Summary. Provided that the fuel, iridium and graphite are of high purity, an FSA should function satisfactorily at any temperature in the range 1600-1750°K. The maximum rates at which significant vapor species are generated from the surfaces of the sphere or iridium shell are summarized in Table C-2. Maximum rates of effusion through the vents are summarized in Table C-3.

E. Reentry

Current reentry calculations project a 5 min thermal excursion for the FSA. The fuel surface temperature increases from the operational temperature to a maximum value in the range 2000-2200°K. Further, it is predicted that this reentry will produce a temperature gradient of up to 200°C across an FSA.

F. Reentry at 2000°K if the Fuel is <PuO₂·0>

The "worst case" situation at reentry would be for the fuel to consist of <PuO₂·0> as this would lead to a maximum (O₂) partial pressure and hence maximum reaction with the iridium shell.

1. Oxygen. The equilibrium partial pressure of (O₂) above <PuO₂·0> at 2000°K is 0.87 atm (Table A-9). This leads to a maximum loss of (O₂) from the fuel of

$$Z_{O_2} = 4.9 \text{ g/cm}^2 \text{ sec.} \quad (110)$$

At 2000°K, the surface area for a 100 watt sphere is given in Table C-1 to be 45.18 cm² so that the loss rate becomes

$$Z_{O_2} = 220.3 \text{ g/sec.} \quad (111)$$

The time required to remove 0.3 g of oxygen from the sphere and reduce the fuel from <PuO₂·0> to <PuO_{1.98}> is only 1.4×10^{-4} sec. The maximum rate at which (O₂) at a partial pressure of 0.87 atm could effuse through the vents is

$$w_{O_2} = 2.6 \times 10^{-5} \text{ g/sec.} \quad (112)$$

Thus, it would take 3.2 h to remove 0.3 g of oxygen from the FSA and the (O₂) over pressure would prevent further reduction of the fuel. Thus, during the 5 min reentry, 0.3 g of oxygen could be removed from the sphere producing <PuO_{1.98}>, but only 0.008 g of oxygen could come out the vents.

2. Effect of (O). The equilibrium partial pressure of (O), corresponding to an (O₂) partial pressure of 0.87 atm, is 6.2×10^{-4} atm. The maximum calculated rate loss of (O) from the sphere surface is

$$Z_O = 0.11 \text{ g/sec,} \quad (113)$$

a value that is only 0.05% of that given by Eq. (111) for (O₂). In 5 min, 4×10^{-6} g of oxygen in the form of (O) could escape through the vents.

3. Effect of Helium.

The (He) generation rate was calculated above in section B to be 1.1×10^{14} atoms/sec. The (O₂) generation rate given by Eq. (111) corresponds to 4.1×10^{24} molecules/sec, i.e., 10^{10} (O₂) molecules per (He) atom. Generation of (He) will not affect the generation of (O₂). While a large (He) inventory present at the time of reentry would suppress the generation of (O₂) somewhat, experiments conducted at LASL predict little retention of helium at the operating temperatures, 1600-1800°K. If the FSA's are degassed prior to launch, there will be no appreciable helium inventory in the event of an immediate reentry should the capsule not achieve orbit.

4. Reactions with the Iridium Shell. The free energy of formation of (IrO₃) at 2000°K, calculated from Eqs. (42) (43) and (44), is 25.49 kcal/mole. The maximum partial pressure of (IrO₃) is calculated to be 1.3×10^{-3} atm. This leads to a maximum generation rate of

$$Z_{IrO_3} = 0.02 \text{ g/cm}^2 \text{ sec} \quad (114)$$

or, for the entire iridium shell,

$$Z_{IrO_3} = 1 \text{ g/sec.} \quad (115)$$

The (IrO₃) generation rate given by Eq. (114) predicts a maximum iridium loss of 86 mil in 5 min. The maximum rate at which (IrO₃) could effuse through the vents is 3.2×10^{-5} g in 5 min. These, however, are maximum values, based on a 300°C extrapolation of the transpiration data, and undoubtedly are much too large. The (O₂)/(IrO₃) partial pressure ratio is 670 and the (O₂) overpressure would inhibit the loss of (IrO₃) from the iridium shell. If the Rice kinetic data^{35,37} are extrapolated to an (O₂) pressure of 0.87 atm, the predicted iridium loss is 3 mil in 5 min. Further indication that these maximum values are too high comes from compatibility experiments conducted at LASL.⁵⁷ In these experiments, <PuO₂·0> was heated in an iridium capsule for 30 min. at about 2200°K (1927°C) with no deleterious effects. Finally, Battles *et al.*,¹⁸ heated <PuO₂·0> in an iridium Knudsen cell and observed no Ir-O species at 2000°K.

The free energy of formation for (IrO₃) is calculated from Eq. (40) to be 40.70 kcal/mole. From this, the maximum partial pressure is calculated to be 3.1×10^{-5} atm from which the loss rate is calculated to be

$$Z_{IrO_2} = 0.023 \text{ g/sec.} \quad (116)$$

This corresponds to a loss of 2 mil of iridium in 5 min. The maximum amount of (IrO_2) that could effuse through the vents in 5 min. is 7.5×10^{-7} g. As for the case of (IrO_2) these calculated maximum values are too high.

5. (PuO_2) . The maximum rate at which (PuO_2) could leave the surface of a $\langle \text{PuO}_2 \cdot 0 \rangle$ sphere at 2000°K is

$$Z_{\text{PuO}_2} = 7.2 \times 10^{-5} \text{ g/sec.} \quad (117)$$

This value leads to a maximum transport of 0.02 g of (PuO_2) in 5 min. The maximum amount that could effuse out the vents in 5 min is 2.6×10^{-3} g. Vaporization of (PuO_2) will not affect successful reentry of the FSA.

6. (PuO) . The maximum rate at which (PuO) could leave the surface of the fuel sphere is 5.1×10^{-8} g in 5 min. The maximum amount that could effuse through the vents in 5 min is 6.1×10^{-15} g. FSA reentry will not be affected.

7. (Pu) . The maximum amount of (Pu) that could leave the fuel surface in 5 min is 1.3×10^{-17} g. The maximum amount that could escape through the vents is 1.6×10^{-24} g. Reentry of the FSA would not be affected. The fuel would not be sufficiently reduced for the (Pu) partial pressure to become high enough to allow formation of $\langle \text{PuIr}_2 \rangle$.

8. Iridium. The vapor pressure of (Ir) at 2000°K is calculated from Eq. (32) to be 2.5×10^{-10} atm. The maximum loss of iridium due to vaporization during reentry would be 9×10^{-4} mil.

9. Impurity Effects. If the system initially contained oxide impurities such as $\langle \text{IrO}_2 \rangle$, reactions at the operating temperatures discussed previously would lessen the ability of the FSA to survive reentry.

10. Reentry at 2000°K if the Fuel is $\langle \text{PuO}_1 \cdot 98 \rangle$. The calculations performed above for FSA's operating in the range 1600 – 2000°K , indicate that except for the case of immediate reentry due to failure to achieve orbit, the fuel that reenters will be reduced somewhat. If the fuel is reduced to $\langle \text{PuO}_1 \cdot 98 \rangle$, the equilibrium partial pressure of (O_2) at 2000°K becomes 2.2×10^{-6} atm (Table A-10). The maximum loss rate of (O_2) is calculated to be

$$Z_{\text{O}_2} = 5.6 \times 10^{-4} \text{ g/sec} \quad (118)$$

or 0.17 g in 5 min. This would lead to a reduction of the fuel from $\langle \text{PuO}_1 \cdot 98 \rangle$ to $\langle \text{PuO}_1 \cdot 97 \rangle$. The amount of oxygen

as (O_2) , that could escape through the vents in 5 min is only 2×10^{-8} g. Thus, the fuel stoichiometry would remain at $\langle \text{PuO}_1 \cdot 98 \rangle$ through reentry.

The maximum amount of (IrO_2) that could be generated in 5 min is 1.2×10^{-6} g, corresponding to 3×10^{-7} mil. The calculated ratio of (IrO_2) generation if the fuel is $\langle \text{PuO}_1 \cdot 98 \rangle$ to that if the fuel is $\langle \text{PuO}_2 \cdot 00 \rangle$ is 2×10^8 . Thus, even the slight amount of fuel reduction at operating temperatures as low as 1600°K (Section D, above) would greatly increase the probability of the FSA surviving reentry.

Similar calculations indicate that if the fuel reenters as $\langle \text{PuO}_1 \cdot 98 \rangle$, the maximum (IrO_2) generation would be 5.8×10^{-8} g or 5×10^{-6} mil in 5 min. Generation of other species such as (PuO_2) would be little affected if the fuel were $\langle \text{PuO}_1 \cdot 98 \rangle$.

11. Loading Fuel as $\langle \text{PuO}_1 \cdot 98 \rangle$. The above calculations indicate that if the FSA attains orbit and operates at 1600 – 1800°K , the fuel will reenter at a stoichiometry below $\langle \text{PuO}_2 \cdot 0 \rangle$. At first glance, it would seem to be advantageous to employ $\langle \text{PuO}_1 \cdot 98 \rangle$ fuel in order to enhance reentry performance for those situations involving immediate reentry due to failure to achieve orbit.

Immediately following fabrication, a plutonium dioxide fuel sphere may have an O/Pu ratio less than 2.0, perhaps 1.98 to 1.95. However, a 100 watt sphere can have an ambient surface temperature as high as 800°K . The free energy expressions listed in Table A-7 indicate that for the reaction

ΔG at 800°K is -1.76 kcal/mole. For the reaction

ΔG at 800°K is -4.24 kcal/mole. In order to prevent oxidation of $\langle \text{PuO}_{2-x} \rangle$ to $\langle \text{PuO}_2 \cdot 0 \rangle$, the fuel would have to be held in an environment with an oxygen pressure of less than 10^{-48} atm. The normal Ar-flush inert boxes used at LASL and at MRC contain about 100 ppm of oxygen corresponding to an (O_2) pressure of 10^{-4} atm or 10^{44} times the pressure needed to oxidize the fuel to $\langle \text{PuO}_2 \cdot 0 \rangle$.

12. Summary. An FSA operating for a time at 1600 – 1800°K will survive reentry at 2000°K , provided no impurities such as $\langle \text{IrO}_2 \rangle$ were initially present in significant amounts. An FSA would also be expected to survive an

immediate reentry at 2000°K. Loss rates of significant vapor species are summarized in Tables C-2 and C-3.

G. Reentry at 2100°K if Fuel is <PuO₂.₉₈>

If the reentry temperature is increased to 2100°K, the "worst case" situation is as follows.

1. Oxygen. The equilibrium partial pressure of (O₂) at 2100°K is 27 atm (Table A-9). This leads to a maximum (O₂) loss rate of

$$Z_{O_2} = 6.7 \times 10^3 \text{ g/sec.} \quad (121)$$

The maximum rate at which (O₂) could effuse out the vents is

$$w_{O_2} = 8 \times 10^{-4} \text{ g/sec.} \quad (122)$$

Thus, in 5 min, the fuel would be reduced to <PuO_{1.98}>. Once the fuel attained a stoichiometry of <PuO_{1.98}> the (O₂) partial pressure becomes 4.0 x 10⁻⁵ atm and the loss rate from the fuel decreases to 9.9 x 10⁻³ g/sec. The rate at which (O₂) at a partial pressure of 4.0 x 10⁻⁵ atm can effuse through the vents is 1.2 x 10⁻³ g/sec. Thus, during reentry the fuel stoichiometry would go from <PuO_{2.98}> to <PuO_{1.98}>. These values for a closed system (FSA) may be compared to those reported by Messier¹⁷ for an open system (Knudsen cell) at 2090°K. He reports that <PuO₂> goes to <PuO_{1.98}> in 17 min, to <PuO_{1.91}> in 30 min and to <PuO_{1.90}> in 60 min.

2. Effect of (O). The equilibrium partial pressure of (O), corresponding to an (O₂) pressure of 27 atm, is 7.2 x 10⁻³ atm. The maximum generation rate of (O) from the sphere surface is calculated to be

$$Z_O = 1.3 \text{ g/sec.} \quad (123)$$

Thus, the (O) contribution to the total amount of oxygen released is only 0.02% that of (O₂).

3. Effect of Helium. The (O₂) generation rate given by Eq. (121) corresponds to 1.3 x 10²⁷ molecules/sec. This is 10¹³ times greater than the (He) generation rate and the generation of (He) will not affect the generation of (O₂).

4. Reactions with the Iridium Shell. The free energy of formation for (IrO₃), extrapolated to 2100°K, is 26.55 kcal/mole. From this, the partial pressure of (IrO₃) is calculated to be 0.24 atm. This corresponds to a generation rate at the surface of the iridium shell of

$$Z_{IrO_3} = 17.9 \text{ g/sec} \quad (124)$$

or an iridium loss of about 1.1 in. in 5 min. The amount that could effuse out the vents in 5 min is 5.8 x 10⁻³ g.

These are maximum values and undoubtedly are much too large for the reasons discussed above for reentry at 2000°K. However, it should be noted that the generation rate of (IrO₃), calculated for reentry at 2100°K, is 180 times greater than that calculated for reentry at 2000°K.

The free energy of formation for (IrO₂), extrapolated to 2100°K, is 40.3 kcal/mole. This leads to a calculated (IrO₂) partial pressure of 1.7 x 10⁻³ atm and a calculated (IrO₂) generation rate of

$$Z_{IrO_2} = 1.2 \text{ g/sec.} \quad (125)$$

This corresponds to about 113 mil of iridium in 5 min.

Again, this value is too high, but the values calculated at 2100°K are about 50 times those calculated at 2000°K.

5. (PuO₂). The equilibrium partial pressure of (PuO₂) above <PuO₂> at 2100°K is 4.6 x 10⁻⁷ atm. This corresponds to a loss of 0.1 g of (PuO₂) from the sphere in 5 min, and a loss of 1.2 x 10⁻⁸ g through the vents. Vaporization of (PuO₂) will not appreciably affect reentry.

6. (PuO) and (Pu). The equilibrium pressure of (PuO) above <PuO₂> is 6.1 x 10⁻¹³ atm. This corresponds to a loss from the sphere of 1.3 x 10⁻⁷ g in 5 min. During this time, 1.5 x 10⁻¹⁴ g could escape through the vents.

The equilibrium partial pressure of (Pu) is 1.0 x 10⁻²² atm. In 5 min, 2 x 10⁻¹⁷ g could leave the fuel sphere, and 2.4 x 10⁻²⁴ g could escape through the vents. The fuel will not be reduced sufficiently to raise the (Pu) pressure high enough to form <PuIr₂>.

Vaporization of (PuO) and (Pu) will not affect reentry.

7. Iridium and Carbon. The vapor pressure of iridium metal at 2100°K is calculated from Eq. (32) to be 1.6 x 10⁻⁹ atm. In 5 min, 3.2 x 10⁻⁴ g or 1 x 10⁻⁴ mil of iridium could be lost due to vaporization. The reported Ir-C eutectic temperature is 2570°K, thus no Ir-C interaction is expected at 2100°K.

8. Impurity Effects. All comments noted above for impurity effects apply, except that these effects increase as the temperature is increased.

9. Reentry at 2100°K if the Fuel is <PuO_{1.98}>. If the FSA has operated for a period of time such that the fuel

has been reduced to $\langle \text{PuO}_{1.98} \rangle$ the oxygen partial pressure becomes 4.0×10^{-5} atm. The partial pressure of (IrO_3) then becomes 4.3×10^{-10} atm and the generation rate becomes

$$Z_{\text{IrO}_3} = 3.2 \times 10^{-7} \text{ g/sec.} \quad (126)$$

This value corresponds to 3×10^{-5} mil in 5 min and is 6×10^8 times lower than that given by Eq. (124) if the fuel is $\langle \text{PuO}_{2.0} \rangle$. Thus, any reduction at all in the O/Pu ratio in the fuel would greatly increase the probability of an FSA surviving reentry.

The partial pressure of (IrO_3) calculated assuming the fuel to be $\langle \text{PuO}_{1.98} \rangle$ is 2.6×10^{-9} atm. The generation rate then becomes 1.8×10^{-6} g/sec, a value 7×10^5 times lower than that given by Eq. (125).

10. Summary. If the FSA has operated long enough for the fuel to be reduced even slightly the FSA should survive reentry. Vaporization results for significant species are summarized in Tables C-2 and C-3.

H. Reentry at 2200°K if the Fuel is $\langle \text{PuO}_{2.0} \rangle$

If the reentry temperature is increased to 2200°K, and the FSA contains $\langle \text{PuO}_{2.0} \rangle$ the pressures within the system become formidable.

1. Oxygen. The equilibrium partial pressure of (O_2) above $\langle \text{PuO}_{2.0} \rangle$ at 2200°K is 620 atm (9100 psi). The (O_2) generation rate at the fuel surface is calculated to be

$$Z_{\text{O}_2} = 1.5 \times 10^5 \text{ g/sec.} \quad (127)$$

The rate at which (O_2) can escape through the vents is

$$w_{\text{O}_2} = 0.018 \text{ g/sec.} \quad (128)$$

It would take 17 sec to remove enough oxygen from the FSA to reduce the fuel to $\langle \text{PuO}_{1.98} \rangle$. Once the fuel is $\langle \text{PuO}_{1.98} \rangle$ the partial pressure of (O_2) decreases to 5.6×10^{-4} atm corresponding to a generation rate at the sphere surface of 0.14 g/sec. It would take 320d for sufficient oxygen to leave the FSA in order for the fuel to be reduced from $\langle \text{PuO}_{1.98} \rangle$ to $\langle \text{PuO}_{1.95} \rangle$, therefore, the fuel will stabilize at a composition near $\langle \text{PuO}_{1.97} \rangle$.

2. Effect of (O) . The partial pressure of (O) corresponding to an (O_2) pressure of 620 atm is 6.7×10^{-2} atm. The generation rate of (O) at the fuel surface is calculated to be 11.4 g/sec so that the (O) contributes only about .008% of the total oxygen loss.

3. Effect of Helium. The generation rate of (O_2) is 10^{13} times greater than that of (He) and helium generation will not affect (O_2) generation.

4. Reactions with the Iridium Shell. The free energy of formation of (IrO_3) , extrapolated to 2200°K, is 27.61 kcal/mole. The partial pressure of (IrO_3) is calculated to be 1.45 atm. This leads to a generation rate of (IrO_3) at the inner surface of the iridium shell of

$$Z_{\text{IrO}_3} = 1044 \text{ g/sec.} \quad (129)$$

This value corresponds to a loss of about 300 mil of iridium per second. The maximum rate at which (IrO_3) could leave the FSA is 0.03 g in 5 min. These values which are 1000 times greater than the values calculated at 2000°K, are extreme values and are undoubtedly too high. However, even the Rice Kinetic data predicts a loss of 3600 mil of iridium in 5 min.

The free energy of formation for (IrO_2) , extrapolated to 2200°K, is 39.9 kcal/mole. The partial pressure of (IrO_2) is calculated to be 0.067 atm and the generation rate at the surface of the iridium shell is calculated to be 47 g/sec, 2000 times greater than at 2000°K.

5. (PuO_2) , (PuO) and (Pu) . The partial pressures above $\langle \text{PuO}_{2.0} \rangle$ at 2200°K are listed in Table A-9 as $P_{\text{PuO}_2} = 2.0 \times 10^{-6}$ atm, $P_{\text{PuO}} = 1.4 \times 10^{-12}$ atm and $P_{\text{Pu}} = 1.7 \times 10^{-22}$ atm. In 5 min, the maximum losses from the sphere surface are calculated to be: 0.42 g (PuO_2) , 2.9×10^{-7} g (PuO) , and 3.4×10^{-17} g (Pu) . The maximum amounts that could effuse through the vents in 5 min are: 5.0×10^{-8} g (PuO_2) , 3.4×10^{-14} g (PuO) , and 4.0×10^{-24} g (Pu) . Thus, it is concluded that vaporization of plutonium-containing gaseous species will not affect satisfactory reentry of the FSA. No $\langle \text{PuIr}_2 \rangle$ will form unless the fuel is reduced to a stoichiometry of $\langle \text{PuO}_{1.75} \rangle$.

6. Iridium and Carbon. The vapor pressure of iridium at 2200°K is 8.5×10^{-9} atm. Thus, the iridium shell could lose 0.03 mil during a 5 min reentry. No Ir-C compound will form and the reported Ir-C eutectic temperature is about 2570°K. However, impurities such as silicon in either the iridium or graphite can reduce this temperature greatly. The Ir-C eutectic is reported to be about 2400°K if 1 wt. % silicon is present.²⁶

7. Reentry at 2200°K if the Fuel is <PuO_{1.98}>. If the FSA has operated in the range 1600–1800°K long enough for the fuel to become <PuO_{1.98}>, the FSA would undoubtedly survive reentry at 2200°K. The equilibrium partial pressure of (O₂) above <PuO_{1.98}> at 2200°K is 5.6×10^{-4} atm. This value is 10^6 times less than that above <PuO_{2.0}>. Thus, even a slight reduction of the O/Pu ratio in the fuel would greatly reduce the (O₂) partial pressure and the subsequent production of (IrO₃) and (IrO₂). It is possible that a 2 h vacuum bakeout of the FSA at 1600°K in order to remove the helium inventory prior to launch will reduce the fuel sufficiently to allow the FSA to survive an immediate reentry of the capsule does not achieve orbit.

If the fuel reenters as <PuO_{1.98}>, the equilibrium (O₂) partial pressure will be 5.6×10^{-4} atm and the maximum loss of (O₂) from the fuel surface is calculated to be 0.14 g/sec. The amount that could escape through the vents is 2.0×10^{-9} g/sec. Thus, during a 5 min reentry the fuel would be reduced from <PuO_{1.98}> to about <PuO_{1.97}>.

The partial pressure of (IrO₃) is calculated to be 2.4×10^{-8} atm. This leads to a calculated generation rate of (IrO₃) of 1.7×10^{-5} g/sec or a loss of 1.5×10^{-3} mil of iridium in 5 min. This value is 6×10^8 times lower than that calculated if the fuel is <PuO_{2.0}>.

The calculated partial pressure of (IrO₂) is 6.1×10^{-8} atm. The calculated (IrO₂) generation rate 4.2×10^{-5} g/sec is 1×10^6 times lower than that calculated if the fuel is <PuO_{2.0}>. This value corresponds to a loss of 4×10^{-3} mil of iridium in 5 min.

8. Summary. It is unlikely that an FSA would survive reentry at 2200°K if the fuel were <PuO_{2.0}>. It was noted above that when a <PuO₂> pellet was heated in an iridium capsule for 30 min. at 2200°K, no deleterious effects were observed.⁵⁷ However, it is likely that some reduction of the fuel occurred during the heat up period so that at 2200°K, the fuel was actually <PuO_{2-x}>. The calculations discussed above indicate that if the FSA has operated long enough for the fuel to be reduced, the FSA should survive reentry at 2200°K. The vaporization of

significant species, assuming <PuO_{2.0}> fuel, is summarized in Tables C-2 and C-3.

1. Miscellaneous Calculations

1. Water. The data listed in Tables B-44 and B-45 indicate that, in the range 1600–1700°K, (H₂O) will oxidize lower oxides to <PuO_{1.95}> or <PuO_{1.92}>, but will not affect <PuO_{2.0}>. In addition the Rice Kinetic data indicate that (H₂O) enhances the reaction of (O₂) with <Ir> to form (IrO₃).^{35,37} Actually an FSA should contain very little water because the fuel, when clad in iridium, has an ambient temperature of about 700°C. In addition, the FSA will receive a 1500°C treatment to anneal the iridium plus a vacuum bakeout to remove helium prior to launch. Accordingly, (H₂O) is not expected to be an important impurity in FSA systems.

2. Carbon and Carbon Monoxide. The data listed in Tables B-8 to B-16 indicate that, above 1600°K, <C> will react with <PuO₂> to produce <PuO_{2-x}> and (CO). In addition, the data listed in Tables B-18 to B-33 indicate that above 1700°K, (CO) will reduce <PuO₂>.

Analyses of fuel spheres fabricated both at MRC and at LASL indicate very little contamination with carbon. The data discussed above in Section II, predict that the fuel will not come in contact with carbon during FSA operation or reentry. Thus, any (CO) generated within an HSA would be due to impurities outside the FSA's or to mechanical failure.

One means by which the fuel could come in contact with carbon to produce (CO) would be for a small meteorite particle to penetrate the HSA and enter an FSA. If this were to happen while an FSA were operating at 1700°K, the data in Table B-11 predict that, under equilibrium conditions, the fuel would be reduced to <PuO_{1.90}> and the (CO) generated would have a partial pressure of about 50 atm. For this (CO) to affect the other FSA's it would have to enter through the vents. The maximum rate at which this could occur is 1.8×10^{-3} g/sec. The data in Table B-28 indicate that, under equilibrium conditions at 1700°K, (CO) will reduce <PuO_{2.0}> to <PuO_{1.98}>. Thus, if the (CO) partial pressure were 50 atm, sufficient (CO) to reduce the fuel to <PuO_{2.0}> could enter an FSA in about 7 min. The discussions above indicate that, at the

operating temperature, the fuel would stabilize at about $\langle \text{PuO}_{1.98} \rangle$ in any case. In the actual event of a meteorite penetrating an FSA in this manner, equilibrium conditions would not be attained, and the (CO) generated would vent to space through the hole in the HSA, leaving the other FSA's unaffected.

J. Summary

1. Comments.

a. The calculations are based on an FSA containing only $\langle \text{PuO}_2 \rangle$, $\langle \text{Ir} \rangle$ and $\langle \text{C} \rangle$. The effect of other species, such as $\langle \text{HfO}_2 \rangle$ on the vent covers, has not been considered.

b. The calculations are based on an FSA having two vents, each with a diam. of 0.002 in. Changes in vent sizes would affect the calculations somewhat. In addition, the calculations were based on open vents. Actual FSA's will be provided with some type of vent cover that will decrease the rates at which gaseous species can leave the FSA's. Reaction rates will be accordingly reduced.

c. The calculations are based on published thermodynamic data. The data available for solid and gaseous plutonium oxides, as well as for iridium and carbon are believed to be reliable. Unfortunately the data published for the gaseous iridium oxides are questionable. The reaction rates between (O_2) and $\langle \text{Ir} \rangle$ to form (IrO_3) and (IrO_2) , predicted from the thermodynamic data, are much higher than the rates obtained in a limited number of kinetics experiments. For each calculation, the thermodynamic data employed were those leading to the worst case, i.e., maximum reaction between the FSA components. In actual life, the thermochemical stability of the FSA system is expected to be better than that predicted by the calculations.

2. Conclusions

a. It is important that impurities such as $\langle \text{IrO}_2 \rangle$ be held to minimum values. The maximum amount of $\langle \text{IrO}_2 \rangle$ that can be present in the iridium without affecting FSA operation is about 1 ppm. The suggested maximum allowable level for routine operations is <20 ppm.

Other impurities such as silicon and iron can, if present in the iridium, lower the Ir-C eutectic temperature and thus affect reentry.

b. If the impurities are kept to low levels, an FSA should function satisfactorily at operating temperatures in the range 1600-1750°C. Slightly higher operating temperatures could be employed with subsequent reduction in mission lifetimes.

c. An FSA that has operated for more than a month or so could probably be reentered at temperatures as high as 2200°K (1927°C). Obviously, the lower the reentry temperature, the better. If an FSA containing $\langle \text{PuO}_{2.0} \rangle$ fuel must reenter immediately due to failure to achieve orbit, the maximum reentry temperature should not exceed 1850°C.

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APPENDIX A

THERMODYNAMIC DATA FOR COMPONENTS OF FSA SYSTEMS

TABLE A-1

THERMODYNAMIC FUNCTIONS FOR GASEOUS PLUTONIUM

<u>Temp., °K</u>	<u>C_p eu</u>	<u>H_T^O - H₂₉₈^O cal/mole</u>	<u>S_T^O - S₂₉₈^O eu</u>	<u>-(F_T^O - H₂₉₈^O/T) eu</u>
298	4.98	4	42.317	42.317
300	4.98 ₅	9	42.347	42.317
400	5.10	512	43.794	42.513
500	5.39	1036	44.961	42.889
600	5.82	1596	45.980	43.321
700	6.32	2203	46.915	43.768
800	6.84	2861	47.794	44.217
900	7.35	3571	48.629	44.661
1000	7.84	4330	49.428	45.098
1100	8.32	5138	50.198	45.527
1200	8.79	5994	50.942	45.948
1300	9.24	6895	51.664	46.360
1400	9.68	7842	52.365	46.764
1500	10.10	8831	53.047	47.160
1600	10.48	9860	53.711	47.549
1700	10.81	10924	54.356	47.930
1800	11.10	12020	54.983	48.305
1900	11.33	13142	55.589	48.672
2000	11.51	14284	56.175	49.033
2100	11.63	15442	56.740	49.386
2200	11.70	16609	57.283	49.733
2300	11.73	17781	57.804	50.073
2400	11.73	18954	58.303	50.405

TABLE A-2

THERMODYNAMIC FUNCTIONS FOR SOLID AND LIQUID PLUTONIUM

<u>Temp., °K</u>	<u>$H_T^O - H_{298}^O$ cal/mole</u>	<u>$S_T^O - S_{298}^O$ eu</u>	<u>$-(F_T^O - H_{298}^O/T)$ eu</u>
298	0	0.00	13.200
300	17	0.057	13.200
400	867	2.502	13.534
500	3257	7.569	14.255
600	4107	9.119	15.474
700	4957	10.429	16.547
800	5807	11.564	17.505
900	6657	12.565	18.368
1000	8177	14.201	19.224
1100	9027	15.011	20.004
1200	9877	15.750	20.720
1300	10727	16.431	21.379
1400	11577	17.061	21.991
1500	12427	17.647	22.562
1600	13277	18.196	23.098
1700	14127	18.711	23.601
1800	14977	19.197	24.076
1900	15827	19.656	24.526
2000	16677	20.092	24.954
2100	17527	20.507	25.361
2200	18377	20.903	25.749
2300	19227	21.280	26.121
2400	20077	21.642	26.477

TABLE A-3

VAPORIZATION OF PLUTONIUM

<u>Temp., °K</u>	<u>ΔH_v kcal/mole</u>	<u>ΔS_v eu</u>	<u>ΔG_T kcal/mole</u>	<u>Pressure, Pu (atm)</u>
298	83.00	29.12	74.32	3.14×10^{-55}
500	80.78	24.20	68.68	9.59×10^{-31}
800	80.05	23.04	61.63	1.46×10^{-17}
900	79.914	22.865	59.336	3.91×10^{-15}
1000	79.15	22.03	57.12	3.28×10^{-13}
1100	79.11	21.99	54.92	1.23×10^{-11}
1200	79.12	21.99	52.73	2.50×10^{-10}
1300	79.11	21.99	54.92	1.23×10^{-11}
1400	79.26	22.11	48.31	2.87×10^{-8}
1500	79.40	22.20	46.10	1.92×10^{-7}
1600	79.58	22.31	43.88	1.01×10^{-6}
1700	79.80	22.45	41.63	4.44×10^{-6}
1800	80.04	22.59	39.38	1.66×10^{-5}
1900	80.32	22.73	37.12	5.37×10^{-5}
2000	80.61	22.89	34.83	1.56×10^{-4}
2100	80.92	23.03	32.55	4.10×10^{-4}
2200	81.23	23.19	30.22	9.95×10^{-4}
2300	81.55	23.33	27.90	2.24×10^{-3}
2400	81.88	23.47	25.55	4.71×10^{-3}

TABLE A-4

THERMODYNAMIC FUNCTIONS FOR GASEOUS O

Temp., °K	$H_T^\circ - H_{298}^\circ$ cal/mole	$S_T^\circ - S_{298}^\circ$ eu	$-(F_T^\circ - H_{298}^\circ/T)$ eu	ΔG_f kcal/mole
298	0	0.00	38.468	55.395
500	1038	2.66	39.055	52.485
800	2550	5.03	40.313	47.960
900	3052	5.62	40.701	46.422
1000	3552	6.15	41.067	44.875
1100	4051	6.63	41.412	43.318
1200	4551	7.06	41.737	41.755
1300	5049	7.46	42.044	40.186
1400	5548	7.83	42.335	38.611
1500	6046	8.17	42.611	37.032
1600	6544	8.50	42.873	35.448
1700	7042	8.80	43.123	33.862
1800	7540	9.08	43.361	32.271
1900	8038	9.35	43.588	30.678
2000	8536	9.61	43.806	29.082
2100	9034	9.85	44.015	27.484
2200	9532	10.08	44.216	25.884
2300	10029	10.30	44.409	24.282
2400	10527	10.51	44.596	22.679

TABLE A-5

THERMODYNAMIC FUNCTIONS FOR GASEOUS O₂

Temp., °K	$H_T^O - H_{298}^O$ cal/mole	$S_T^O - S_{298}^O$ eu	$-(F_T^O - H_{298}^O/T)$ eu
298	0	0.00	49.00
500	1455	3.72	49.81
800	3786	7.36	51.63
900	4600	8.32	52.21
1000	5427	9.19	52.76
1100	6266	9.99	53.30
1200	7114	10.72	53.80
1300	7971	11.41	54.28
1400	8835	12.05	54.74
1500	9706	12.65	55.18
1600	10583	13.22	55.61
1700	11465	13.75	56.01
1800	12354	14.26	56.40
1900	13249	14.74	56.78
2000	14149	15.21	57.14
2100	15054	15.65	56.48
2200	15966	16.07	57.82
2300	16882	16.48	58.14
2400	17804	16.87	58.46

TABLE A-6

SELECTED THERMODYNAMIC FUNCTIONS FOR SOLID PuO_2

Temp., °K	Cp eu	$H_T^O - H_{298}^O$ cal/mole	$S_T^O - S_{298}^O$ eu	$-(F_T^O - H_{298}^O \text{ K})$ eu	$-\Delta G_f^O$ kcal/mole
298	16.39	0	0	19.700	239.54
300	16.44	35	0.043	19.733	239.25
400	18.02	1767	5.080	20.362	235.22
500	18.97	3620	9.210	21.670	231.00
600	19.66	5552	12.732	23.178	226.58
700	20.23	7547	15.807	24.725	222.21
800	20.74	9596	18.552	26.257	217.90
900	21.21	11693	21.011	27.719	213.63
1000	21.65	13836	23.267	29.132	209.35
1100	22.09	16023	25.356	30.490	205.11
1200	22.53	18253	27.300	31.789	200.93
1300	22.95	20524	29.118	33.031	196.78
1400	23.34	22838	30.830	34.217	192.68
1500	23.75	25192	32.400	35.306	188.54
1600	24.17	27586	34.001	36.460	184.61
1700	24.56	30020	35.470	37.511	180.65
1800	24.95	32495	36.889	38.536	176.71
1900	25.36	35009	38.252	39.527	172.83
2000	2575	37563	39.529	40.447	168.92
2100	26.14	40157	40.822	41.399	165.17
2200	26.53	42790	42.049	42.301	161.43
2300	26.94	45462	43.239	43.176	157.70
2400	27.34	48174	44.395	44.023	154.02

TABLE A-7

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

Species	$\Delta G_f^\circ = A + BT, ^\circ K \text{ (cal/mole)}$		$\Delta G_o^\circ = A + BT, ^\circ K \text{ (cal/mole)}$	
	A	B	A	B
$\langle \text{PuO}_2 \cdot 00 \rangle$	-249,500 ^a	40.3	(-204,750) ^b	(87.7)
$\langle \text{PuO}_1 \cdot 98 \rangle$	-246,700	39.0	-181,750	63.4
$\langle \text{PuO}_1 \cdot 96 \rangle$	-243,100	37.6	-178,000	59.0
$\langle \text{PuO}_1 \cdot 92 \rangle$	-239,700	36.4	-173,750	54.1
$\langle \text{PuO}_1 \cdot 90 \rangle$	-237,500	35.7	-171,150	50.8
$\langle \text{PuO}_1 \cdot 85 \rangle$	-232,100	34.1	-164,250	43.4
$\langle \text{PuO}_1 \cdot 80 \rangle$	-227,100	32.9	-157,500	36.1
$\langle \text{PuO}_1 \cdot 75 \rangle$	-222,500	32.1	-151,500	29.5
$\langle \text{PuO}_1 \cdot 70 \rangle$	-218,000	31.5	-149,250	26.5
$\langle \text{PuO}_1 \cdot 65 \rangle$	-213,600	31.0	-150,250	26.1
$\langle \text{PuO}_1 \cdot 61 \rangle$	-210,000	30.6	-153,000	26.9
$\langle \text{PuO}_1 \cdot 50 \rangle$	-200,300	29.6	-153,000	26.9

^a Values believed to be good in range 1600 - 2200°K.

^b Values in parentheses are estimated.

TABLE A-8

VALUES FOR THE FREE ENERGY OF FORMATION FOR $\langle \text{PuO}_2 \rangle$

Temp., °K	$-\Delta G_f$ (kcal/mole)		Δ , Percent
	Table A-6	Equation From Table A-7	
298	239.54	237.49	0.86
500	231.00	229.35	0.72
800	217.90	217.26	0.29
900	213.63	213.23	0.19
1000	209.35	209.20	0.07
1100	205.11	205.17	0.03
1200	200.93	201.14	0.10
1300	196.78	197.11	0.17
1400	192.68	193.08	0.21
1500	188.54	189.05	0.27
1600	184.61	185.02	0.22
1700	180.65	180.99	0.19
1800	176.71	176.96	0.14
1900	172.83	172.93	0.06
2000	168.92	168.90	0.01
2100	165.17	164.87	0.18
2200	161.43	160.84	0.37
2300	157.70	156.81	0.57
2400	154.02	152.78	0.81

TABLE A-9

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_2 \rangle$
(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO_2)
1500	2.9×10^{-11}	1.9×10^{-24}	3.9×10^{-16}	9.6×10^{-13}
1600	1.2×10^{-8}	4.5×10^{-24}	2.0×10^{-15}	1.7×10^{-11}
1700	2.5×10^{-6}	9.7×10^{-24}	8.1×10^{-15}	2.1×10^{-10}
1800	2.8×10^{-4}	1.9×10^{-23}	2.9×10^{-14}	2.0×10^{-9}
1900	1.9×10^{-2}	3.6×10^{-23}	8.8×10^{-14}	1.5×10^{-8}
2000	0.87	6.3×10^{-23}	2.4×10^{-13}	9.0×10^{-8}
2100	2.7×10^{-1}	1.0×10^{-22}	6.1×10^{-13}	4.6×10^{-7}
2200	6.2×10^{-2}	1.7×10^{-22}	1.4×10^{-12}	2.0×10^{-6}
2300	1.1×10^{-1}	2.6×10^{-22}	3.0×10^{-12}	7.9×10^{-6}
2400	1.5×10^{-5}	3.9×10^{-22}	6.1×10^{-12}	2.7×10^{-5}

TABLE A-10

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuC}_{1.98} \rangle$
(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO_2)
1500	3.5×10^{-15}	1.5×10^{-20}	3.4×10^{-14}	9.2×10^{-13}
1600	5.6×10^{-13}	9.2×10^{-20}	2.7×10^{-13}	1.6×10^{-11}
1700	4.9×10^{-11}	4.5×10^{-19}	1.7×10^{-12}	2.0×10^{-10}
1800	2.6×10^{-9}	1.9×10^{-18}	8.8×10^{-12}	1.8×10^{-9}
1900	9.1×10^{-8}	7.1×10^{-18}	3.8×10^{-11}	1.4×10^{-8}
2000	2.2×10^{-6}	2.3×10^{-17}	1.4×10^{-10}	8.3×10^{-8}
2100	4.0×10^{-5}	6.5×10^{-17}	4.6×10^{-10}	4.2×10^{-7}
2200	5.6×10^{-4}	1.7×10^{-16}	1.4×10^{-9}	1.9×10^{-6}
2300	6.1×10^{-3}	4.2×10^{-16}	3.7×10^{-9}	7.2×10^{-6}
2400	5.5×10^{-2}	1.8×10^{-16}	4.1×10^{-9}	2.5×10^{-5}

TABLE A-11

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{1.95} \rangle$
(Pressures in Atmospheres)

Temp., °K	(O ₂)	(Pu)	(PuO)	(PuO ₂)
1500	5.1×10^{-16}	9.8×10^{-20}	8.5×10^{-14}	8.8×10^{-13}
1600	7.0×10^{-14}	7.0×10^{-19}	7.4×10^{-13}	1.5×10^{-11}
1700	5.4×10^{-12}	4.0×10^{-18}	4.9×10^{-12}	1.9×10^{-10}
1800	2.5×10^{-10}	1.9×10^{-17}	2.7×10^{-11}	1.8×10^{-9}
1900	7.9×10^{-9}	7.7×10^{-17}	1.2×10^{-10}	1.3×10^{-8}
2000	1.8×10^{-7}	2.7×10^{-16}	4.7×10^{-10}	7.8×10^{-8}
2100	2.9×10^{-6}	8.6×10^{-16}	1.6×10^{-9}	4.0×10^{-7}
2200	3.7×10^{-5}	2.4×10^{-15}	5.0×10^{-9}	1.8×10^{-6}
2300	3.8×10^{-4}	6.4×10^{-15}	1.4×10^{-8}	6.7×10^{-6}
2400	3.2×10^{-3}	1.6×10^{-14}	3.6×10^{-8}	2.3×10^{-5}

TABLE A-12

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{1.92} \rangle$
(Pressures in Atmospheres)

Temp., °K	(O ₂)	(Pu)	(PuO)	(PuO ₂)
1500	6.4×10^{-17}	7.2×10^{-19}	2.2×10^{-13}	8.2×10^{-13}
1600	7.3×10^{-15}	6.2×10^{-18}	2.1×10^{-12}	1.4×10^{-11}
1700	4.8×10^{-13}	4.1×10^{-17}	1.5×10^{-11}	1.7×10^{-10}
1800	2.0×10^{-11}	2.2×10^{-16}	8.8×10^{-11}	1.6×10^{-9}
1900	5.4×10^{-10}	1.0×10^{-15}	4.2×10^{-10}	1.2×10^{-8}
2000	1.1×10^{-8}	4.1×10^{-15}	1.7×10^{-9}	7.1×10^{-8}
2100	1.6×10^{-7}	1.4×10^{-14}	6.3×10^{-9}	3.6×10^{-7}
2200	1.9×10^{-6}	4.4×10^{-14}	2.0×10^{-8}	1.6×10^{-6}
2300	1.7×10^{-5}	1.2×10^{-13}	5.9×10^{-8}	6.1×10^{-6}
2400	1.4×10^{-4}	3.3×10^{-13}	1.6×10^{-7}	2.1×10^{-5}

TABLE A-13

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{1.90} \rangle$

(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO_2)
1500	1.3×10^{-17}	3.3×10^{-18}	4.6×10^{-13}	7.6×10^{-13}
1600	1.4×10^{-15}	3.1×10^{-17}	4.5×10^{-12}	1.3×10^{-11}
1700	8.1×10^{-14}	2.3×10^{-16}	3.4×10^{-11}	1.6×10^{-10}
1800	3.0×10^{-12}	1.4×10^{-15}	2.1×10^{-10}	1.5×10^{-9}
1900	7.7×10^{-11}	6.6×10^{-15}	1.0×10^{-9}	1.1×10^{-8}
2000	1.4×10^{-9}	2.3×10^{-14}	4.4×10^{-9}	6.6×10^{-8}
2100	2.0×10^{-8}	1.0×10^{-13}	1.6×10^{-8}	3.3×10^{-7}
2200	2.2×10^{-7}	3.4×10^{-13}	5.4×10^{-8}	1.4×10^{-6}
2300	2.0×10^{-6}	1.0×10^{-12}	1.6×10^{-7}	5.6×10^{-6}
2400	1.5×10^{-5}	2.8×10^{-12}	4.3×10^{-7}	1.9×10^{-5}

TABLE A-14

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\langle \text{PuO}_{1.85} \rangle$

(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO_2)
1500	7.9×10^{-19}	4.6×10^{-17}	1.6×10^{-12}	6.4×10^{-13}
1600	6.1×10^{-17}	5.7×10^{-16}	1.8×10^{-11}	1.1×10^{-11}
1700	2.8×10^{-15}	5.3×10^{-15}	1.5×10^{-10}	1.3×10^{-10}
1800	8.4×10^{-14}	3.9×10^{-14}	9.9×10^{-10}	1.2×10^{-9}
1900	1.7×10^{-12}	2.3×10^{-13}	5.4×10^{-9}	8.6×10^{-9}
2000	1.2×10^{-11}	1.2×10^{-12}	2.5×10^{-8}	5.1×10^{-8}
2100	3.2×10^{-10}	4.9×10^{-12}	9.9×10^{-8}	2.5×10^{-7}
2200	3.0×10^{-9}	1.9×10^{-11}	3.5×10^{-7}	1.1×10^{-6}
2300	2.3×10^{-8}	6.4×10^{-11}	1.1×10^{-6}	4.2×10^{-6}
2400	1.5×10^{-7}	2.0×10^{-10}	3.2×10^{-6}	1.4×10^{-5}

TABLE A-15

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\text{PuO}_{1.80}$
(Pressures in Atmospheres)

Temp., °K	(O ₂)	(Pu)	(PuO)	(PuO ₂)
1500	4.7×10^{-20}	6.0×10^{-16}	5.0×10^{-12}	5.0×10^{-13}
1600	2.7×10^{-18}	9.6×10^{-15}	6.3×10^{-11}	8.2×10^{-12}
1700	9.8×10^{-17}	1.1×10^{-13}	5.9×10^{-10}	9.6×10^{-11}
1800	2.3×10^{-15}	1.0×10^{-12}	4.3×10^{-9}	8.6×10^{-10}
1900	4.0×10^{-14}	7.2×10^{-12}	2.6×10^{-8}	6.1×10^{-9}
2000	5.2×10^{-13}	4.2×10^{-11}	1.3×10^{-7}	3.6×10^{-8}
2100	5.2×10^{-12}	2.1×10^{-10}	5.4×10^{-7}	1.8×10^{-7}
2200	4.3×10^{-11}	9.1×10^{-10}	2.0×10^{-6}	7.5×10^{-7}
2300	2.9×10^{-10}	3.5×10^{-9}	6.7×10^{-6}	2.8×10^{-6}
2400	1.7×10^{-9}	1.2×10^{-8}	2.0×10^{-5}	9.6×10^{-6}

TABLE A-16

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\text{PuO}_{1.75}$
(Pressures in Atmospheres)

Temp., °K	(O ₂)	(Pu)	(PuO)	(PuO ₂)
1500	3.5×10^{-21}	6.1×10^{-15}	1.4×10^{-11}	3.7×10^{-13}
1600	1.6×10^{-19}	1.2×10^{-13}	1.9×10^{-10}	5.9×10^{-12}
1700	4.0×10^{-18}	1.8×10^{-12}	2.0×10^{-9}	6.8×10^{-11}
1800	8.8×10^{-17}	1.9×10^{-11}	2.5×10^{-8}	6.0×10^{-10}
1900	1.3×10^{-15}	1.6×10^{-10}	9.8×10^{-8}	4.2×10^{-9}
2000	1.4×10^{-14}	1.1×10^{-9}	5.2×10^{-7}	2.4×10^{-8}
2100	1.2×10^{-13}	6.1×10^{-9}	2.3×10^{-6}	1.2×10^{-7}
2200	8.7×10^{-13}	2.9×10^{-8}	9.2×10^{-6}	4.9×10^{-7}
2300	5.2×10^{-12}	1.2×10^{-7}	3.2×10^{-5}	1.8×10^{-6}
2400	2.7×10^{-11}	4.7×10^{-7}	1.0×10^{-4}	6.1×10^{-6}

TABLE A-17

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\text{PuO}_{1.70}$
(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO ₂)
1500	7.7×10^{-22}	2.3×10^{-14}	2.4×10^{-11}	3.0×10^{-13}
1600	3.1×10^{-20}	4.9×10^{-13}	3.5×10^{-10}	4.8×10^{-12}
1700	8.2×10^{-19}	7.6×10^{-12}	3.6×10^{-9}	5.4×10^{-11}
1800	1.5×10^{-17}	8.6×10^{-11}	2.9×10^{-8}	4.7×10^{-10}
1900	2.0×10^{-16}	7.6×10^{-10}	1.9×10^{-7}	3.3×10^{-9}
2000	5.5×10^{-16}	5.4×10^{-9}	1.0×10^{-6}	1.9×10^{-8}
2100	1.7×10^{-14}	3.2×10^{-8}	4.7×10^{-6}	9.0×10^{-8}
2200	1.2×10^{-13}	1.6×10^{-7}	1.9×10^{-5}	3.7×10^{-7}
2300	6.8×10^{-13}	7.2×10^{-7}	6.7×10^{-5}	1.4×10^{-6}
2400	3.4×10^{-12}	2.8×10^{-6}	2.1×10^{-4}	4.6×10^{-6}

TABLE A-18

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\text{PuO}_{1.55}$
(Pressures in Atmospheres)

Temp., $^{\circ}\text{K}$	(O_2)	(Pu)	(PuO)	(PuO ₂)
1500	2.6×10^{-22}	5.5×10^{-14}	3.5×10^{-11}	2.5×10^{-13}
1600	1.1×10^{-20}	1.2×10^{-12}	4.9×10^{-10}	4.0×10^{-12}
1700	3.0×10^{-19}	1.7×10^{-11}	5.1×10^{-9}	4.6×10^{-11}
1800	5.8×10^{-18}	1.9×10^{-10}	4.1×10^{-8}	4.0×10^{-10}
1900	8.0×10^{-17}	1.7×10^{-9}	2.6×10^{-7}	2.8×10^{-9}
2000	8.5×10^{-16}	1.2×10^{-8}	1.4×10^{-6}	1.6×10^{-8}
2100	7.2×10^{-15}	6.7×10^{-8}	6.4×10^{-6}	7.8×10^{-8}
2200	5.0×10^{-14}	3.4×10^{-7}	2.5×10^{-5}	3.3×10^{-7}
2300	2.9×10^{-13}	1.5×10^{-6}	8.9×10^{-5}	1.2×10^{-6}
2400	1.5×10^{-12}	5.7×10^{-6}	2.8×10^{-4}	4.0×10^{-6}

TABLE A-19

CALCULATED EQUILIBRIUM PARTIAL PRESSURES ABOVE $\text{PuO}_{1.61} - \text{PuO}_{1.50}$
(Pressures in Atmospheres)

<u>Temp., °K</u>	<u>(O₂)</u>	<u>(Pu)</u>	<u>(PuO)</u>	<u>(PuO₂)</u>
1500	9.2×10^{-23}	1.3×10^{-13}	4.8×10^{-11}	2.1×10^{-13}
1600	4.4×10^{-21}	2.5×10^{-12}	6.6×10^{-10}	3.4×10^{-12}
1700	1.3×10^{-19}	3.4×10^{-11}	6.6×10^{-9}	4.0×10^{-11}
1800	2.8×10^{-18}	3.5×10^{-10}	5.1×10^{-8}	3.5×10^{-10}
1900	4.2×10^{-17}	2.8×10^{-9}	3.2×10^{-7}	2.5×10^{-9}
2000	4.7×10^{-16}	1.9×10^{-8}	1.7×10^{-6}	1.4×10^{-8}
2100	4.3×10^{-15}	1.0×10^{-7}	7.5×10^{-6}	7.1×10^{-8}
2200	3.2×10^{-14}	4.9×10^{-7}	2.9×10^{-5}	3.0×10^{-7}
2300	2.0×10^{-13}	2.0×10^{-6}	1.0×10^{-4}	1.1×10^{-6}
2400	1.1×10^{-12}	7.6×10^{-6}	3.2×10^{-4}	3.8×10^{-6}

TABLE A-20

CONGRUENTLY VAPORIZING COMPOSITION OF PuO_{2-x}

<u>Temp., °K</u>	<u>O/Pu Closed System</u>	<u>O/Pu Open System</u>
1600	1.91	1.89
1700	1.90	1.88
1800	1.89	1.87
1900	1.87	1.86
2000	1.86	1.85
2100	1.85	1.83
2200	1.84	1.82

TABLE A-21

EXPERIMENTAL VAPOR PRESSURE DATA ABOVE $\langle \text{PuO}_{2-x} \rangle$

Reference	Temperature Range, $^{\circ}\text{K}$	Av. Temp., $^{\circ}\text{K}$	ΔH_T^a kcal/mole	ΔS_T^a eu
15	1650 - 2070	1900	133.9	34.3
17	2070 - 2380	2243	141.7	38.3
18	1900 - 2300	2219	131.9	34.0
19	2000 - 2200	2110	133.1	34.6
19 ^b	2000 - 2230	2135	129.9	32.7

Pressure Values for P_E in Atmospheres

Reference	2000 $^{\circ}\text{K}$	2200 $^{\circ}\text{K}$
15	7.4×10^{-8}	1.6×10^{-6}
17	7.9×10^{-8}	1.6×10^{-6}
18	1.0×10^{-7}	2.1×10^{-6}
19	9.9×10^{-8}	2.1×10^{-6}
19 ^b	8.5×10^{-8}	1.7×10^{-6}

^a The heat and entropy of vaporization terms are based on P_E values assuming the vapor to be (PuO_2) .

^b Sample initially consisted of $\langle \text{Pu}_{0.45} \text{Th}_{0.55} \text{O}_{2.0} \rangle$.

TABLE A-22

COMPARISON OF EXPERIMENTAL AND CALCULATED P_E VALUESABOVE $\langle \text{PuO}_{2-x} \rangle$

Reference	Av. Temp., $^{\circ}\text{K}$	Experimental P_E (atm)	Calculated Stoichiometry	Calculated P_E (atm)
15	1900	1.3×10^{-8}	$\langle \text{PuO}_{1.86} \rangle$	1.3×10^{-8}
17	2243	3.7×10^{-6}	$\langle \text{PuO}_{1.82} \rangle$	3.5×10^{-6}
18	2219	2.7×10^{-6}	$\langle \text{PuO}_{1.82} \rangle$	2.4×10^{-6}
19	2110	5.7×10^{-7}	$\langle \text{PuO}_{1.83} \rangle$	4.8×10^{-7}

TABLE A-23

EXPERIMENTAL VAPOR PRESSURE DATA ABOVE $\langle \text{PuO}_{1.61} - \text{PuO}_{1.50} \rangle$

<u>Reference</u>	<u>Temperature Range, °K</u>	<u>Av. Temp., °K</u>	<u>ΔH_T^a kcal/mole</u>	<u>ΔS_T^a eu</u>
15	1650 - 2100	1900	127.2	37.0
18	1660 - 2150	2018	117.8	32.6
19 ^b	1870 - 1960	1925	127.4	37.0
20	1600 - 2100	1881	127.7	37.2

Pressure Values for P_E in Atmospheres

<u>Reference</u>	<u>1800°K</u>	<u>2000°K</u>
15	4.4×10^{-8}	1.5×10^{-6}
18	6.6×10^{-8}	1.8×10^{-6}
19 ^b	4.2×10^{-8}	1.5×10^{-6}
20	4.2×10^{-8}	1.5×10^{-6}

^a The heat and entropy terms are based on P_E values assuming the vapor to be (PuO).

^b Sample initially consisted of $\langle \text{Pu}_{0.9} \text{Th}_{0.1} \text{O}_{2.0} \rangle$.

TABLE A-24

COMPARISON OF EXPERIMENTAL AND CALCULATED P_E VALUES
ABOVE $\langle \text{PuO}_{1.61} - \text{PuO}_{1.50} \rangle$

<u>Reference</u>	<u>Av. Temp., °K</u>	<u>Experimental P_E (atm)</u>	<u>Calculated P_E (atm)</u>
15	1900	2.9×10^{-7}	3.3×10^{-7}
18	2018	2.3×10^{-6}	2.3×10^{-6}
20	1881	4.3×10^{-7}	5.0×10^{-7}

TABLE A-25

THERMODYNAMIC FUNCTIONS FOR SOLID CARBON (GRAPHITE)

<u>Temp., °K</u>	<u>$H_T^0 - H_{298}$ cal/mole</u>	<u>$S_T^0 - S_{298}^0$ eu</u>	<u>$-(F_T^0 - H_{298}/T)$ eu</u>
298	0	0.00	1.359
500	569	1.42	1.646
800	1831	3.38	2.449
1000	2824	4.48	3.020
1100	3347	4.98	3.300
1200	3883	5.45	3.573
1300	4432	5.89	3.839
1400	4988	6.30	4.098
1500	5552	6.69	4.348
1600	6122	7.06	4.591
1700	6696	7.41	4.827
1800	7275	7.74	5.055
1900	7857	8.05	5.276
2000	8442	8.35	5.490
2100	9029	8.64	5.698
2200	9620	8.91	5.900
2300	10212	9.18	6.095
2400	10807	9.43	6.286

TABLE A-26

THERMODYNAMIC FUNCTIONS FOR GASEOUS C

Temp., °K	$H_T^0 - H_{298}^0$ cal/mole	$S_T^0 - S_{298}^0$ eu	$-(F_T - H_{298}/T)$ eu	ΔG_f kcal/mole
298	0	0.00	37.761	160.03
500	1004	2.57	38.325	152.55
800	2496	4.91	39.551	141.21
1000	3490	6.02	40.290	133.62
1100	3986	6.49	40.629	129.82
1200	4483	6.92	40.949	126.04
1300	4980	7.32	41.252	122.25
1400	5478	7.69	41.519	118.47
1500	5975	8.03	41.811	114.69
1600	6473	8.36	42.070	110.92
1700	6971	8.66	42.317	107.15
1800	7469	8.94	42.553	103.39
1900	7969	9.21	42.779	99.63
2000	8469	9.47	42.995	95.88
2100	8970	9.71	43.202	92.13
2200	9473	9.95	43.402	88.38
2300	9977	10.17	43.594	84.64
2400	10482	10.39	43.779	80.90

TABLE A-27

THERMODYNAMIC FUNCTIONS FOR GASEOUS C₂

Temp., °K	$H_T^0 - H_{298}^0$ cal/mole	$S_T^0 - S_{298}^0$ eu	$-(F_T^0 - H_{298}^0/T)$ eu	ΔG_f kcal/mole
298	0	0.00	47.627	186.84
500	1924	4.96	48.740	177.50
800	4501	9.00	51.005	163.34
1000	6210	10.91	52.328	153.94
1100	7073	11.73	52.931	149.26
1200	7941	12.49	53.498	144.60
1300	8817	13.19	54.035	139.96
1400	9698	13.84	54.543	135.34
1500	10587	14.46	55.025	130.73
1600	11482	15.03	55.485	126.14
1700	12385	15.58	55.923	121.56
1800	13294	16.10	56.342	117.00
1900	14210	16.60	56.744	112.46
2000	15132	17.07	57.130	107.92
2100	16061	17.52	57.501	103.40
2200	16996	17.96	57.859	98.89
2300	17937	18.38	58.204	94.40
2400	18884	18.78	58.537	89.91

TABLE A-28

THERMODYNAMIC FUNCTIONS FOR GASEOUS C_3

<u>Temp., °K</u>	<u>$H_T^0 - H_{298}^0$ cal/mole</u>	<u>$S_T^0 - S_{298}^0$ eu</u>	<u>$-(F_T^0 - H_{298}^0/T)$ eu</u>	<u>ΔG_f kcal/mole</u>
298	0	0.00	56.677	180.32
500	1818	4.65	57.694	169.62
800	4716	9.18	56.957	153.92
1000	6804	11.90	61.375	143.68
1100	7887	12.53	62.041	138.64
1200	8992	13.50	62.679	133.65
1300	10116	14.40	63.291	128.69
1400	11258	15.24	63.877	123.78
1500	12415	16.04	64.440	118.91
1600	13586	16.80	64.981	114.07
1700	14769	17.50	65.502	109.26
1800	15964	18.20	66.004	104.49
1900	17170	18.85	66.488	99.74
2000	18386	19.47	66.955	95.03
2100	19610	20.07	67.407	90.34
2200	20844	20.64	67.845	85.68
2300	22085	21.19	68.269	81.04
2400	23333	21.72	68.680	76.43

TABLE A-29

THERMODYNAMIC FUNCTIONS FOR SOLID IRIIDIUM

Temp. °K	Cp eu	$H_T^\circ - H_{298}^\circ$ cal/mole	$S_T^\circ - S_{298}^\circ$ eu	$-(F_T^\circ - H_{298}^\circ/T)$ eu
298	6.00	0	0.00	8.70
500	6.27	1235	3.16	9.39
800	6.70	3190	6.22	10.94
1000	6.98	4545	7.73	11.89
1100	7.12	5250	8.40	12.33
1200	7.26	5970	9.03	12.76
1300	7.41	6700	9.61	13.16
1400	7.55	7440	10.16	13.55
1500	7.69	8200	10.68	13.92
1600	7.83	8980	11.19	14.28
1700	7.97	9780	11.67	14.62
1800	8.12	10590	12.13	14.95
1900	8.26	11410	12.57	15.27
2000	8.40	12240	13.00	15.58
2100	8.54	13090	13.41	15.88
2200	8.68	13950	13.81	16.17
2300	8.83	14820	14.20	16.46
2400	8.97	15710	14.58	16.74

TABLE A-30

SOLUBILITY OF CARBON IN IRIIDIUM^a

Temperature, °C	Solubility, C atoms per Ir atom
700	(0.0002) ^b
800	0.0003
900	0.0005
1000	0.0007
1100	0.001
1200	0.002
1300	(0.003)
1400	(0.004)
1500	(0.007)
1600	(0.011)
1700	(0.017)
1800	(0.027)
1900	(0.043)

^a Values calculated from the data from
Ref. 28.

^b Values in parentheses are extrapolated.

TABLE A-31

OXIDATION OF IRIIDIUM-RECESSION RATES FROM
THE DATA OF KURIAKOSE, KENT AND MARGRAVE

<u>Temp., °C</u>	<u>Oxygen Pressure, Torr</u>	<u>Gas Velocity ft/min</u>	<u>Recession Rate mil/h</u>
1198	152	103	0.74
1252	152	103	0.93
1359	152	103	1.07
1487	152	103	1.61
	15	14.7	0.07
1602	152	103	1.86
	15	14.7	0.15
1717	152	103	3.40
	15	14.7	0.18
1830	152	103	5.42
	15	14.7	0.40
1968	152	103	6.87
	15	14.7	0.58
2097	152	103	10.98
	15	14.7	0.84
2217	152	103	13.88
	15	14.7	1.15

TABLE A-32

OXIDATION OF IRIIDIUM - RECESSION RATES FROM THE DATA OF REXER

<u>Temp., °C</u>	<u>Oxygen Pressure, Torr</u>	<u>Gas Velocity ft/min</u>	<u>Recession Rate mil/h</u>
1900	610	25,000	116
	142	12,000	10.50
	14.2	12,000	0.60
1730	142	12,000	6.42
1615	142	12,000	4.80
1500	142	12,000	3.12
1450	142	12,000	3.96
1400	142	12,000	3.42
1330	142	12,000	2.40
1270	142	12,000	1.86
1150	142	12,000	0.96
1500	710	25,000	47.4
1270	710	25,000	17.4

TABLE A-33

RESULTS FOR HEATING PuO₂-Ir-C ASSEMBLY

<u>Run No.</u>	<u>Temp., °C</u>	<u>Time at Temp., min.</u>	<u>Results</u>
1	1260	120	Slight sintering of PuO ₂ particles. No visible reaction between PuO ₂ and Ir or between Ir and graphite. Weight loss for assembly was 0.3 mg.
2	1370	120	Same as for Run No. 1 except further sintering of PuO ₂ particles. Weight loss for assembly was 0.5 mg.
3	1750	5	Same as for Run No. 1. Weight loss for assembly was 0.3 mg.
4	2100	5	Further sintering of PuO ₂ particles. Slight sticking of PuO ₂ to Ir. Iridium stuck tightly to graphite. Weight loss for assembly was 0.7 mg.
5	2275	5	Extensive reaction with evolution of gas, presumably CO. Nearly all of the PuO ₂ and Ir had reacted with the graphite. Weight loss for the assembly was 140 mg.

TABLE A-34

THERMODYNAMIC FUNCTIONS FOR GASEOUS HYDROGEN

Temp., °K	$H_T^{\circ} - H_{298}^{\circ}$ cal/mole	$S_T^{\circ} - S_{298}^{\circ}$ eu	$-(F_T^{\circ} - H_{298}^{\circ}/T)$ eu
298	0	0.00	31.20
500	1406	3.60	32.00
800	3514	6.90	33.72
1000	4944	8.49	34.76
1100	5670	9.19	35.24
1200	6404	9.82	35.70
1300	7148	10.42	36.13
1400	7902	10.98	36.54
1500	8668	11.51	36.94
1600	9446	12.01	37.31
1700	10233	12.49	37.68
1800	11030	12.94	38.02
1900	11836	13.38	38.36
2000	12651	13.80	38.68
2100	13475	14.20	38.99
2200	14307	14.58	39.29
2300	15146	14.96	39.58
2400	15993	15.32	39.86

TABLE A-35

THERMODYNAMIC FUNCTIONS FOR GASEOUS WATER

<u>Temp., °K</u>	<u>$H_T^O - H_{298}^O$ cal/mole</u>	<u>$S_T^O - S_{298}^O$ eu</u>	<u>$-(F_T^O - H_{298}^O/T)$ eu</u>
298	0	0.00	45.11
500	1654	4.23	46.03
800	4300	8.36	48.02
1000	6209	10.49	49.38
1100	7210	11.44	49.99
1200	8240	12.34	50.58
1300	9298	13.18	51.14
1400	10394	13.99	51.68
1500	11495	14.75	52.20
1600	12630	15.48	52.70
1700	13787	16.19	53.18
1800	14964	16.86	53.65
1900	16160	17.51	54.11
2000	17373	18.13	54.55
2100	18602	18.73	54.98
2200	19846	19.31	55.39
2300	21103	19.86	55.80
2400	22372	20.40	56.19

TABLE A-36

THERMODYNAMIC FUNCTIONS FOR GASEOUS CO

<u>Temp., °K</u>	<u>$H_T^0 - H_{298}^0$ cal/mole</u>	<u>$S_T^0 - S_{298}^0$ eu</u>	<u>$-(F_T^0 - H_{298}^0/T)$ eu</u>	<u>ΔG_f kcal/mole</u>
298	0	0.00	47.21	- 32.78
500	1417	3.68	48.01	- 37.14
800	3627	7.08	49.76	- 43.61
1000	5183	8.81	50.84	- 47.86
1100	5983	9.58	51.35	- 49.96
1200	6794	10.28	51.83	- 52.05
1300	7616	10.94	52.30	- 54.13
1400	8446	11.56	52.74	- 56.19
1500	9285	12.13	53.16	- 58.24
1600	10130	12.68	53.56	- 60.28
1700	10980	13.20	53.95	- 62.32
1800	11836	13.68	54.32	- 64.34
1900	12697	14.15	54.68	- 66.35
2000	13561	14.59	55.03	- 68.35
2100	14430	15.02	55.36	- 70.35
2200	15301	15.42	55.68	- 72.34
2300	16175	15.81	55.99	- 74.31
2400	17052	16.18	56.29	- 76.28

TABLE A-37

THERMODYNAMIC FUNCTIONS FOR GASEOUS CO₂

<u>Temp., °K</u>	<u>H_T^o - H₂₉₈^o cal/mole</u>	<u>S_T^o - S₂₉₈^o eu</u>	<u>-(F_T^o - H₂₉₈^o/T) eu</u>	<u>Δ G_f kcal/mole</u>
298	0	0.00	51.08	- 94.26
500	1987	5.05	52.15	- 94.40
800	5453	10.45	54.71	- 94.56
1000	7984	13.27	56.36	- 94.63
1100	9296	14.52	57.14	- 94.66
1200	10632	15.68	57.90	- 94.68
1300	11988	16.77	58.62	- 94.70
1400	13362	17.79	59.32	- 94.72
1500	14750	18.74	59.98	- 94.73
1600	16152	19.65	60.63	- 94.74
1700	17565	20.51	61.25	- 94.75
1800	18987	21.32	61.84	- 94.75
1900	20418	22.09	62.42	- 94.75
2000	21857	22.83	62.97	- 94.75
2100	23303	23.54	63.51	- 94.75
2200	24755	24.21	64.03	- 94.74
2300	26212	24.86	64.54	- 94.74
2400	27674	25.48	65.02	- 94.72

APPENDIX B

CALCULATED REACTIONS

TABLE B-1

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 298 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	1.75
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	3.95
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	6.02
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	7.35
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	10.63
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	13.63
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	16.36
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	19.05
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	21.66
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	23.83
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	29.62
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	246.2
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	160.2
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	157.2
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	126.2

TABLE B-2

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 500 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	1.41
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	3.19
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	4.88
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	5.99
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	8.73
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	11.27
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	13.62
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	15.96
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	18.25
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	20.17
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	25.28
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	223.8
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	143.2
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	139.9
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	113.9

TABLE B-3

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 800 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	0.89
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	2.06
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	3.19
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	3.96
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	5.90
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	7.76
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	9.54
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	11.38
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	13.20
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	14.73
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	18.84
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	191.7
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	118.0
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	114.2
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	95.5

TABLE B-4

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1000 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	0.54
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	1.31
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	2.07
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	2.61
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	4.02
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	5.43
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	6.84
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	8.34
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	9.85
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	11.14
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	14.57
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	170.6
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	101.2
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	97.2
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	83.4

TABLE B-5

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1100 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	0.37
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	0.93
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	1.51
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	1.94
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	3.09
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	4.27
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	5.49
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	6.83
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	8.18
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	9.35
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	12.45
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	160.2
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	92.9
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	88.7
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	77.3

TABLE B-6

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1200 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	0.20
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	0.56
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	0.96
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	1.28
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	2.15
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	3.11
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	4.15
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	5.32
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	6.52
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	7.56
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	10.34
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	149.8
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	84.7
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	80.3
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	71.4

TABLE B-7

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1300°K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	+	0.03
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	+	0.18
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	+	0.40
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	+	0.61
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	1.22
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	1.95
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	2.81
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	3.82
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	4.86
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	5.78
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	8.22
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	143.8
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	76.4
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	71.9
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	65.4

TABLE B-8

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1400°K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	0.14
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	0.19
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	0.16
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	0.06
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	+	0.29
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	+	0.80
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	1.47
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	2.32
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	3.21
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	4.01
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	6.12
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	129.0
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	68.2
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	63.5
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	59.4

TABLE B-9

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1500 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	0.31
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	0.56
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	0.71
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	0.72
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	0.64
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	0.35
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	+	0.14
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	+	0.83
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	+	1.57
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	2.24
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	4.03
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	118.7
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	30.0
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	55.2
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	53.4

TABLE D-10

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1600 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	0.49
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	0.93
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	1.26
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	1.39
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	1.56
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	1.50
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	-	1.19
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	-	0.66
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	-	0.08
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	+	0.47
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	+	1.94
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	108.3
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	51.8
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	46.8
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	47.5

TABLE B-11

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1700 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	0.66
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	1.31
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	1.82
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	2.05
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	2.49
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	2.64
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	-	2.52
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	-	2.16
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	-	1.72
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	-	1.29
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	-	0.15
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	98.0
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	43.6
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	38.5
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	41.6

TABLE B-12

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1800 °K

Reaction	ΔG kcal/mole		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	0.83
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	1.68
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	2.37
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	2.71
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	3.41
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	3.79
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	-	3.84
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	-	3.64
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	-	3.36
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	-	3.05
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	-	2.23
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	87.7
$\langle \text{PuO}_2 \rangle + 2.9 \langle \text{C} \rangle = \langle \text{PuC}_{0.9} \rangle$	+ 2.0 (CO)	+	35.5
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	30.2
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = \langle \text{PuO}_{0.5}\text{C}_{0.5} \rangle$	+ 1.5 (CO)	+	35.6

TABLE B-13

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 1900 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	1.00
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	2.05
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	2.92
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	3.38
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	4.33
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	4.93
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	-	5.17
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	-	5.12
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	-	4.99
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	-	4.81
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	-	4.30
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	77.4
$\langle \text{PuO}_2 \rangle + 3.5 \langle \text{C} \rangle = \langle \text{PuC}_{1.5} \rangle$	+ 2.0 (CO)	+	22.0

TABLE B-14

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 2000 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO)	-	1.17
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO)	-	2.42
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO)	-	3.47
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO)	-	4.04
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO)	-	5.25
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO)	-	6.07
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO)	-	6.49
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO)	-	6.60
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO)	-	6.62
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO)	-	6.56
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO)	-	6.38
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO)	+	67.0
$\langle \text{PuO}_2 \rangle + 4.0 \langle \text{C} \rangle = \langle \text{PuC}_2 \rangle$	+ 2.0 (CO)	+	13.5

TABLE B-15

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 2100 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO) - 1.34
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO) - 2.79
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO) - 4.02
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO) - 4.70
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO) - 6.17
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO) - 7.21
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO) - 7.81
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO) - 8.08
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO) - 8.25
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO) - 8.31
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO) - 8.44
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO) + 56.7
$\langle \text{PuO}_2 \rangle + 4.0 \langle \text{C} \rangle = \langle \text{PuC}_2 \rangle$	+ 2.0 (CO) + 4.9

TABLE B-16

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 2200 °K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO) - 1.51
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO) - 3.16
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO) - 4.57
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO) - 5.35
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO) - 7.09
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO) - 8.35
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO) - 9.12
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO) - 9.56
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO) - 9.88
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO) - 10.05
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO) - 10.51
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO) + 46.4
$\langle \text{PuO}_2 \rangle + 4.0 \langle \text{C} \rangle = \langle \text{PuC}_2 \rangle$	+ 2.0 (CO) - 3.7

TABLE B-17

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{C} \rangle$ at 2300°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 \langle \text{C} \rangle = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO) - 1.68
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{C} \rangle = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO) - 3.53
$\langle \text{PuO}_2 \rangle + 0.08 \langle \text{C} \rangle = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO) - 5.11
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{C} \rangle = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO) - 6.01
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{C} \rangle = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO) - 8.01
$\langle \text{PuO}_2 \rangle + 0.20 \langle \text{C} \rangle = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO) - 9.48
$\langle \text{PuO}_2 \rangle + 0.25 \langle \text{C} \rangle = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO) - 10.44
$\langle \text{PuO}_2 \rangle + 0.30 \langle \text{C} \rangle = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO) - 11.03
$\langle \text{PuO}_2 \rangle + 0.35 \langle \text{C} \rangle = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO) - 11.50
$\langle \text{PuO}_2 \rangle + 0.39 \langle \text{C} \rangle = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO) - 11.79
$\langle \text{PuO}_2 \rangle + 0.50 \langle \text{C} \rangle = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO) - 12.56
$\langle \text{PuO}_2 \rangle + 2.0 \langle \text{C} \rangle = (\text{Pu})$	+ 2.0 (CO) + 36.1
$\langle \text{PuO}_2 \rangle + 4.0 \langle \text{C} \rangle = \langle \text{PuC}_2 \rangle$	+ 2.0 (CO) - 12.2

TABLE B - 18

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 298°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO ₂) + 1.18
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO ₂) + 2.52
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO ₂) + 3.72
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO ₂) + 4.48
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO ₂) + 6.33
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO ₂) + 7.89
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO ₂) + 9.19
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO ₂) + 10.44
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO ₂) + 11.61
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO ₂) + 12.63
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO ₂) + 15.27

TABLE B - 19

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 500°K

<u>Reaction</u>				ΔG kcal/mole
$\langle \text{PuO}_2 \rangle$	+ 0.02(CO) = $\langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO ₂)	+ 1.00
$\langle \text{PuO}_2 \rangle$	+ 0.05(CO) = $\langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO ₂)	+ 2.19
$\langle \text{PuO}_2 \rangle$	+ 0.08(CO) = $\langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO ₂)	+ 3.27
$\langle \text{PuO}_2 \rangle$	+ 0.10(CO) = $\langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO ₂)	+ 3.97
$\langle \text{PuO}_2 \rangle$	+ 0.15(CO) = $\langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO ₂)	+ 5.71
$\langle \text{PuO}_2 \rangle$	+ 0.20(CO) = $\langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO ₂)	+ 7.25
$\langle \text{PuO}_2 \rangle$	+ 0.25(CO) = $\langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO ₂)	+ 8.58
$\langle \text{PuO}_2 \rangle$	+ 0.30(CO) = $\langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO ₂)	+ 9.92
$\langle \text{PuO}_2 \rangle$	+ 0.35(CO) = $\langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO ₂)	+ 11.21
$\langle \text{PuO}_2 \rangle$	+ 0.39(CO) = $\langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO ₂)	+ 12.32
$\langle \text{PuO}_2 \rangle$	+ 0.50(CO) = $\langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO ₂)	+ 15.22

TABLE B - 20

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 800°K

<u>Reaction</u>				ΔG kcal/mole
$\langle \text{PuO}_2 \rangle$	+ 0.02(CO) = $\langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO ₂)	+ 0.74
$\langle \text{PuO}_2 \rangle$	+ 0.05(CO) = $\langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO ₂)	+ 1.69
$\langle \text{PuO}_2 \rangle$	+ 0.08(CO) = $\langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO ₂)	+ 2.60
$\langle \text{PuO}_2 \rangle$	+ 0.10(CO) = $\langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO ₂)	+ 3.22
$\langle \text{PuO}_2 \rangle$	+ 0.15(CO) = $\langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO ₂)	+ 4.80
$\langle \text{PuO}_2 \rangle$	+ 0.20(CO) = $\langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO ₂)	+ 6.29
$\langle \text{PuO}_2 \rangle$	+ 0.25(CO) = $\langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO ₂)	+ 7.70
$\langle \text{PuO}_2 \rangle$	+ 0.30(CO) = $\langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO ₂)	+ 9.18
$\langle \text{PuO}_2 \rangle$	+ 0.35(CO) = $\langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO ₂)	+ 10.63
$\langle \text{PuO}_2 \rangle$	+ 0.39(CO) = $\langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO ₂)	+ 11.87
$\langle \text{PuO}_2 \rangle$	+ 0.50(CO) = $\langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO ₂)	+ 15.16

TABLE B - 21

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1000°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO_2)	+ 0.56
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO_2)	+ 1.36
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO_2)	+ 2.16
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO_2)	+ 2.72
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO_2)	+ 4.18
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO_2)	+ 5.65
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO_2)	+ 7.11
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO_2)	+ 8.67
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO_2)	+ 10.23
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO_2)	+ 11.56
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO_2)	+ 15.12

TABLE B - 22

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1100°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>		
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO_2)	+ 0.48
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO_2)	+ 1.20
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO_2)	+ 1.93
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO_2)	+ 2.47
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO_2)	+ 3.88
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO_2)	+ 5.32
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO_2)	+ 6.80
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO_2)	+ 8.41
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO_2)	+ 10.02
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO_2)	+ 11.40
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO_2)	+ 15.08

TABLE B - 23

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1200°K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle + 0.02(\text{CO}_2)$	+ 0.39
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle + 0.05(\text{CO}_2)$	+ 1.03
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle + 0.08(\text{CO}_2)$	+ 1.71
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle + 0.10(\text{CO}_2)$	+ 2.22
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle + 0.15(\text{CO}_2)$	+ 3.57
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle + 0.20(\text{CO}_2)$	+ 4.99
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle + 0.25(\text{CO}_2)$	+ 6.50
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle + 0.30(\text{CO}_2)$	+ 8.15
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle + 0.35(\text{CO}_2)$	+ 9.82
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle + 0.39(\text{CO}_2)$	+ 11.23
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle + 0.50(\text{CO}_2)$	+ 15.04

TABLE B - 24

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1300°K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle + 0.02(\text{CO}_2)$	+ 0.30
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle + 0.05(\text{CO}_2)$	+ 0.86
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle + 0.08(\text{CO}_2)$	+ 1.48
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle + 0.10(\text{CO}_2)$	+ 1.96
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle + 0.15(\text{CO}_2)$	+ 3.25
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle + 0.20(\text{CO}_2)$	+ 4.67
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle + 0.25(\text{CO}_2)$	+ 6.20
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle + 0.30(\text{CO}_2)$	+ 7.89
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle + 0.35(\text{CO}_2)$	+ 9.61
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle + 0.39(\text{CO}_2)$	+ 11.07
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle + 0.50(\text{CO}_2)$	+ 15.00

TABLE B - 25

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1400°K

<u>Reaction</u>				ΔG kcal/mole
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$		+ 0.02	(CO_2)	+ 0.21
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$		+ 0.05	(CO_2)	+ 0.69
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$		+ 0.08	(CO_2)	+ 1.26
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$		+ 0.10	(CO_2)	+ 1.71
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$		+ 0.15	(CO_2)	+ 2.94
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$		+ 0.20	(CO_2)	+ 4.33
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$		+ 0.25	(CO_2)	+ 5.89
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$		+ 0.30	(CO_2)	+ 7.62
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$		+ 0.35	(CO_2)	+ 9.39
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$		+ 0.39	(CO_2)	+ 10.89
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$		+ 0.50	(CO_2)	+ 14.96

TABLE B - 26

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1500°K

<u>Reaction</u>				ΔG kcal/mole
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$		+ 0.02	(CO_2)	+ 0.12
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$		+ 0.05	(CO_2)	+ 0.53
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$		+ 0.08	(CO_2)	+ 1.03
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$		+ 0.10	(CO_2)	+ 1.45
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$		+ 0.15	(CO_2)	+ 2.33
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$		+ 0.20	(CO_2)	+ 4.30
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$		+ 0.25	(CO_2)	+ 5.58
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$		+ 0.30	(CO_2)	+ 7.35
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$		+ 0.35	(CO_2)	+ 9.18
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$		+ 0.39	(CO_2)	+ 10.72
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$		+ 0.50	(CO_2)	+ 14.90

TABLE B - 27

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1600°K

<u>Reaction</u>				<u>ΔG</u> <u>kcal/mole</u>	
$\langle \text{PuO}_2 \rangle$	+ 0.02(CO) =	$\langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO ₂)	+	0.03
$\langle \text{PuO}_2 \rangle$	+ 0.05(CO) =	$\langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO ₂)	+	0.36
$\langle \text{PuO}_2 \rangle$	+ 0.08(CO) =	$\langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO ₂)	+	0.80
$\langle \text{PuO}_2 \rangle$	+ 0.10(CO) =	$\langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO ₂)	+	1.19
$\langle \text{PuO}_2 \rangle$	+ 0.15(CO) =	$\langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO ₂)	+	2.31
$\langle \text{PuO}_2 \rangle$	+ 0.20(CO) =	$\langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO ₂)	+	3.67
$\langle \text{PuO}_2 \rangle$	+ 0.25(CO) =	$\langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO ₂)	+	5.26
$\langle \text{PuO}_2 \rangle$	+ 0.30(CO) =	$\langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO ₂)	+	7.08
$\langle \text{PuO}_2 \rangle$	+ 0.35(CO) =	$\langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO ₂)	+	8.96
$\langle \text{PuO}_2 \rangle$	+ 0.39(CO) =	$\langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO ₂)	+	10.54
$\langle \text{PuO}_2 \rangle$	+ 0.50(CO) =	$\langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO ₂)	+	14.85

TABLE B - 28

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1700°K

<u>Reaction</u>				<u>ΔG</u> <u>kcal/mole</u>	
$\langle \text{PuO}_2 \rangle$	+ 0.02(CO) =	$\langle \text{PuO}_{1.98} \rangle$	+ 0.02 (CO ₂)	-	0.06
$\langle \text{PuO}_2 \rangle$	+ 0.05(CO) =	$\langle \text{PuO}_{1.95} \rangle$	+ 0.05 (CO ₂)	+	0.19
$\langle \text{PuO}_2 \rangle$	+ 0.08(CO) =	$\langle \text{PuO}_{1.92} \rangle$	+ 0.08 (CO ₂)	+	0.58
$\langle \text{PuO}_2 \rangle$	+ 0.10(CO) =	$\langle \text{PuO}_{1.90} \rangle$	+ 0.10 (CO ₂)	+	0.94
$\langle \text{PuO}_2 \rangle$	+ 0.15(CO) =	$\langle \text{PuO}_{1.85} \rangle$	+ 0.15 (CO ₂)	+	2.00
$\langle \text{PuO}_2 \rangle$	+ 0.20(CO) =	$\langle \text{PuO}_{1.80} \rangle$	+ 0.20 (CO ₂)	+	3.33
$\langle \text{PuO}_2 \rangle$	+ 0.25(CO) =	$\langle \text{PuO}_{1.75} \rangle$	+ 0.25 (CO ₂)	+	4.95
$\langle \text{PuO}_2 \rangle$	+ 0.30(CO) =	$\langle \text{PuO}_{1.70} \rangle$	+ 0.30 (CO ₂)	+	6.81
$\langle \text{PuO}_2 \rangle$	+ 0.35(CO) =	$\langle \text{PuO}_{1.65} \rangle$	+ 0.35 (CO ₂)	+	8.74
$\langle \text{PuO}_2 \rangle$	+ 0.39(CO) =	$\langle \text{PuO}_{1.61} \rangle$	+ 0.39 (CO ₂)	+	10.36
$\langle \text{PuO}_2 \rangle$	+ 0.50(CO) =	$\langle \text{PuO}_{1.50} \rangle$	+ 0.50 (CO ₂)	+	14.80

TABLE B -29

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1800°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>			
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	$+ 0.02$	(CO_2)	$-$	0.15
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	$+ 0.05$	(CO_2)	$+$	0.02
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	$+ 0.08$	(CO_2)	$+$	0.35
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	$+ 0.10$	(CO_2)	$+$	0.68
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	$+ 0.15$	(CO_2)	$+$	1.68
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	$+ 0.20$	(CO_2)	$+$	3.00
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	$+ 0.25$	(CO_2)	$+$	4.64
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	$+ 0.30$	(CO_2)	$+$	6.54
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	$+ 0.35$	(CO_2)	$+$	8.52
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	$+ 0.39$	(CO_2)	$+$	10.18
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	$+ 0.50$	(CO_2)	$+$	14.74

TABLE B - 30

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 1900°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>			
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	$+ 0.02$	(CO_2)	$-$	0.24
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	$+ 0.05$	(CO_2)	$-$	0.15
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	$+ 0.08$	(CO_2)	$+$	0.12
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	$+ 0.10$	(CO_2)	$+$	0.42
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	$+ 0.15$	(CO_2)	$+$	1.36
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	$+ 0.20$	(CO_2)	$+$	2.66
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	$+ 0.25$	(CO_2)	$+$	4.32
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	$+ 0.30$	(CO_2)	$+$	6.26
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	$+ 0.35$	(CO_2)	$+$	8.29
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	$+ 0.39$	(CO_2)	$+$	9.99
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	$+ 0.50$	(CO_2)	$+$	14.67

TABLE B - 31

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 2000°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>			
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_1 \cdot 98 \rangle$	+	0.02	(CO ₂)	- 0.33
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_1 \cdot 95 \rangle$	+	0.05	(CO ₂)	- 0.32
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_1 \cdot 92 \rangle$	+	0.08	(CO ₂)	- 0.11
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_1 \cdot 90 \rangle$	+	0.10	(CO ₂)	+ 0.16
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_1 \cdot 85 \rangle$	+	0.15	(CO ₂)	+ 1.04
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_1 \cdot 80 \rangle$	+	0.20	(CO ₂)	+ 2.32
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_1 \cdot 75 \rangle$	+	0.25	(CO ₂)	+ 4.00
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_1 \cdot 70 \rangle$	+	0.30	(CO ₂)	+ 5.98
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_1 \cdot 65 \rangle$	+	0.35	(CO ₂)	+ 8.06
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_1 \cdot 61 \rangle$	+	0.39	(CO ₂)	+ 9.80
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_1 \cdot 50 \rangle$	+	0.50	(CO ₂)	+ 14.60

TABLE B - 32

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 2100°K

<u>Reaction</u>	<u>ΔG</u> <u>kcal/mole</u>			
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_1 \cdot 98 \rangle$	+	0.02	(CO ₂)	- 0.42
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_1 \cdot 95 \rangle$	+	0.05	(CO ₂)	- 0.49
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_1 \cdot 92 \rangle$	+	0.08	(CO ₂)	- 0.34
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_1 \cdot 90 \rangle$	+	0.10	(CO ₂)	- 0.10
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_1 \cdot 85 \rangle$	+	0.15	(CO ₂)	+ 0.72
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_1 \cdot 80 \rangle$	+	0.20	(CO ₂)	+ 1.98
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_1 \cdot 75 \rangle$	+	0.25	(CO ₂)	+ 3.68
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_1 \cdot 70 \rangle$	+	0.30	(CO ₂)	+ 5.70
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_1 \cdot 65 \rangle$	+	0.35	(CO ₂)	+ 7.83
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_1 \cdot 61 \rangle$	+	0.39	(CO ₂)	+ 9.61
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_1 \cdot 50 \rangle$	+	0.50	(CO ₂)	+ 14.53

TABLE B - 33

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 2200°K

<u>Reaction</u>			<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO_2)	- 0.51
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO_2)	- 0.66
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO_2)	- 0.57
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO_2)	- 0.36
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO_2)	+ 0.40
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO_2)	+ 1.64
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO_2)	+ 3.36
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO_2)	+ 5.42
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO_2)	+ 7.60
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO_2)	+ 9.42
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO_2)	+ 14.46

TABLE B - 34

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (CO) at 2300°K

<u>Reaction</u>			<u>ΔG</u> <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02(\text{CO}) = \langle \text{PuO}_{1.98} \rangle$	+ 0.02	(CO_2)	- 0.60
$\langle \text{PuO}_2 \rangle + 0.05(\text{CO}) = \langle \text{PuO}_{1.95} \rangle$	+ 0.05	(CO_2)	- 0.83
$\langle \text{PuO}_2 \rangle + 0.08(\text{CO}) = \langle \text{PuO}_{1.92} \rangle$	+ 0.08	(CO_2)	- 0.80
$\langle \text{PuO}_2 \rangle + 0.10(\text{CO}) = \langle \text{PuO}_{1.90} \rangle$	+ 0.10	(CO_2)	- 0.62
$\langle \text{PuO}_2 \rangle + 0.15(\text{CO}) = \langle \text{PuO}_{1.85} \rangle$	+ 0.15	(CO_2)	+ 0.08
$\langle \text{PuO}_2 \rangle + 0.20(\text{CO}) = \langle \text{PuO}_{1.80} \rangle$	+ 0.20	(CO_2)	+ 1.29
$\langle \text{PuO}_2 \rangle + 0.25(\text{CO}) = \langle \text{PuO}_{1.75} \rangle$	+ 0.25	(CO_2)	+ 3.03
$\langle \text{PuO}_2 \rangle + 0.30(\text{CO}) = \langle \text{PuO}_{1.70} \rangle$	+ 0.30	(CO_2)	+ 5.13
$\langle \text{PuO}_2 \rangle + 0.35(\text{CO}) = \langle \text{PuO}_{1.65} \rangle$	+ 0.35	(CO_2)	+ 7.36
$\langle \text{PuO}_2 \rangle + 0.39(\text{CO}) = \langle \text{PuO}_{1.61} \rangle$	+ 0.39	(CO_2)	+ 9.22
$\langle \text{PuO}_2 \rangle + 0.50(\text{CO}) = \langle \text{PuO}_{1.50} \rangle$	+ 0.50	(CO_2)	+ 14.38

TABLE B-35

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 298 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 1.32
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 2.86
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 4.27
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 5.17
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 7.35
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 9.27
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 10.90
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 12.49
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 14.01
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 15.30
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 18.69

TABLE B-36

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 500 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 1.10
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 2.43
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 3.66
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 4.46
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 6.45
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 8.23
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 9.81
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 11.39
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 12.92
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 14.23
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 17.67

TABLE B-37

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 800 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.79
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 1.81
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 2.79
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 3.46
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 5.14
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 6.75
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 8.28
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 9.87
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 11.43
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 12.77
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 16.32

TABLE B-38

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1000 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.58
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 1.40
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 2.22
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 2.80
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 4.29
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 5.79
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 7.29
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 8.89
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 10.49
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 11.84
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 15.48

TABLE B-39

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1100 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.48
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 1.19
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 1.93
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 2.47
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 3.87
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 5.32
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 6.80
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 8.41
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 10.02
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 11.39
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 15.07

TABLE B-40

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1200 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.37
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 0.99
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 1.65
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 2.14
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 3.45
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 4.85
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 6.32
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 7.93
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 9.56
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 10.95
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 14.67

TABLE B-41

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1300 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.53} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.27
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.63} \rangle + 0.05 (\text{H}_2\text{O})$	+ 0.79
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.72} \rangle + 0.08 (\text{H}_2\text{O})$	+ 1.37
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.83} \rangle + 0.10 (\text{H}_2\text{O})$	+ 1.82
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.15 (\text{H}_2\text{O})$	+ 3.04
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.20 (\text{H}_2\text{O})$	+ 4.38
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 5.83
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 7.45
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 9.10
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 10.50
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.59} \rangle + 0.50 (\text{H}_2\text{O})$	+ 14.28

TABLE B-42

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1400 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.27
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 0.59
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 1.09
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 1.49
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 2.62
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 3.91
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 5.35
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 6.98
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 8.65
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 10.06
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.59} \rangle + 0.50 (\text{H}_2\text{O})$	+ 13.89

TABLE B-43

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1500 °K

Reaction	ΔG kcal/mole
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	+ 0.06
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 0.39
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 0.81
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 1.17
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 2.21
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 3.44
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 4.89
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 6.51
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 8.20
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 9.82
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 13.50

TABLE B-44

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1600 °K

Reaction	ΔG kcal/mole
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.04
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	+ 0.18
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	+ 0.53
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	+ 0.85
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 1.79
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 2.97
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 4.40
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 6.04
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 7.75
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 9.19
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 13.12

TABLE B-45

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1700 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.02} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.14
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.05} \rangle + 0.05 (\text{H}_2\text{O})$	- 0.02
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.08} \rangle + 0.08 (\text{H}_2\text{O})$	+ 0.25
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.10} \rangle + 0.10 (\text{H}_2\text{O})$	+ 0.53
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.15} \rangle + 0.15 (\text{H}_2\text{O})$	+ 1.38
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.20} \rangle + 0.20 (\text{H}_2\text{O})$	+ 2.51
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.25} \rangle + 0.25 (\text{H}_2\text{O})$	+ 3.92
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.30} \rangle + 0.30 (\text{H}_2\text{O})$	+ 5.58
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.35} \rangle + 0.35 (\text{H}_2\text{O})$	+ 7.30
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.39} \rangle + 0.39 (\text{H}_2\text{O})$	+ 8.76
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 12.74

TABLE B-46

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1800 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.02} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.24
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.05} \rangle + 0.05 (\text{H}_2\text{O})$	- 0.22
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.08} \rangle + 0.08 (\text{H}_2\text{O})$	- 0.03
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.10} \rangle + 0.10 (\text{H}_2\text{O})$	+ 0.20
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.15} \rangle + 0.15 (\text{H}_2\text{O})$	+ 0.96
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.20} \rangle + 0.20 (\text{H}_2\text{O})$	+ 2.05
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.25} \rangle + 0.25 (\text{H}_2\text{O})$	+ 3.45
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.30} \rangle + 0.30 (\text{H}_2\text{O})$	+ 5.11
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.35} \rangle + 0.35 (\text{H}_2\text{O})$	+ 6.85
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.39} \rangle + 0.39 (\text{H}_2\text{O})$	+ 8.32
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 12.36

TABLE B-47

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 1900 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.35
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	- 0.42
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	- 0.31
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	- 0.12
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 0.55
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 1.58
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 2.97
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 4.64
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 6.40
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 7.89
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 11.98

TABLE B-48

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 2000 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.45
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	- 0.62
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	- 0.59
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	- 0.44
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	+ 0.14
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 1.12
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 2.50
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 4.18
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 5.96
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 7.46
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 11.60

TABLE B-49

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 2100 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.55
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	- 0.82
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	- 0.87
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	- 0.76
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	- 0.27
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 0.65
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 2.03
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 3.72
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 5.52
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 7.04
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 11.22

TABLE B-50

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 2200 °K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.98} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.65
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	- 1.02
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	- 1.15
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	- 1.08
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	- 0.68
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	+ 0.20
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 1.55
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 3.25
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 5.07
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 6.61
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 10.85

TABLE B-51

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and (H_2) at 2300°K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.02 (\text{H}_2) = \langle \text{PuO}_{1.99} \rangle + 0.02 (\text{H}_2\text{O})$	- 0.75
$\langle \text{PuO}_2 \rangle + 0.05 (\text{H}_2) = \langle \text{PuO}_{1.95} \rangle + 0.05 (\text{H}_2\text{O})$	- 1.22
$\langle \text{PuO}_2 \rangle + 0.08 (\text{H}_2) = \langle \text{PuO}_{1.92} \rangle + 0.08 (\text{H}_2\text{O})$	- 1.43
$\langle \text{PuO}_2 \rangle + 0.10 (\text{H}_2) = \langle \text{PuO}_{1.90} \rangle + 0.10 (\text{H}_2\text{O})$	- 1.40
$\langle \text{PuO}_2 \rangle + 0.15 (\text{H}_2) = \langle \text{PuO}_{1.85} \rangle + 0.15 (\text{H}_2\text{O})$	- 1.09
$\langle \text{PuO}_2 \rangle + 0.20 (\text{H}_2) = \langle \text{PuO}_{1.80} \rangle + 0.20 (\text{H}_2\text{O})$	- 0.27
$\langle \text{PuO}_2 \rangle + 0.25 (\text{H}_2) = \langle \text{PuO}_{1.75} \rangle + 0.25 (\text{H}_2\text{O})$	+ 1.08
$\langle \text{PuO}_2 \rangle + 0.30 (\text{H}_2) = \langle \text{PuO}_{1.70} \rangle + 0.30 (\text{H}_2\text{O})$	+ 2.79
$\langle \text{PuO}_2 \rangle + 0.35 (\text{H}_2) = \langle \text{PuO}_{1.65} \rangle + 0.35 (\text{H}_2\text{O})$	+ 4.63
$\langle \text{PuO}_2 \rangle + 0.39 (\text{H}_2) = \langle \text{PuO}_{1.61} \rangle + 0.39 (\text{H}_2\text{O})$	+ 6.18
$\langle \text{PuO}_2 \rangle + 0.50 (\text{H}_2) = \langle \text{PuO}_{1.50} \rangle + 0.50 (\text{H}_2\text{O})$	+ 10.48

TABLE B - 52

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 298°K

<u>Reaction</u>	<u>ΔG kcal/mole</u>
$\langle \text{PuO}_2 \rangle + 0.61 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.99} \rangle + 0.01 \langle \text{IrO}_2 \rangle$	+ 1.96
$\langle \text{PuO}_2 \rangle + 0.025 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.95} \rangle + 0.025 \langle \text{IrO}_2 \rangle$	+ 4.46
$\langle \text{PuO}_2 \rangle + 0.04 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.92} \rangle + 0.04 \langle \text{IrO}_2 \rangle$	+ 6.83
$\langle \text{PuO}_2 \rangle + 0.05 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.90} \rangle + 0.05 \langle \text{IrO}_2 \rangle$	+ 8.37
$\langle \text{PuO}_2 \rangle + 0.075 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.85} \rangle + 0.075 \langle \text{IrO}_2 \rangle$	+ 12.17
$\langle \text{PuO}_2 \rangle + 0.10 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.80} \rangle + 0.10 \langle \text{IrO}_2 \rangle$	+ 15.68
$\langle \text{PuO}_2 \rangle + 0.125 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.75} \rangle + 0.125 \langle \text{IrO}_2 \rangle$	+ 18.92
$\langle \text{PuO}_2 \rangle + 0.15 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.70} \rangle + 0.15 \langle \text{IrO}_2 \rangle$	+ 22.11
$\langle \text{PuO}_2 \rangle + 0.175 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.65} \rangle + 0.175 \langle \text{IrO}_2 \rangle$	+ 25.23
$\langle \text{PuO}_2 \rangle + 0.195 \langle \text{Ir} \rangle = \langle \text{PuO}_{1.61} \rangle + 0.195 \langle \text{IrO}_2 \rangle$	+ 27.81

TABLE B - 53

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 500°K

<u>Reaction</u>					ΔG kcal/mole
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 99 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.79
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 4.14
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 6.40
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 7.88
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 11.57
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 15.07
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 18.36
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 21.65
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 24.89
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 27.56

TABLE B - 54

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 800°K

<u>Reaction</u>					ΔG kcal/mole
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 99 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.53
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 3.66
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 5.75
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 7.16
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 10.69
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 14.15
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 17.53
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 20.97
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 24.38
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 27.20

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REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 1000°K

<u>Reaction</u>					ΔG <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 98 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.35
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 3.34
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 5.32
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 6.67
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 10.11
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 13.54
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 16.98
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 20.51
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 24.05
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 26.96

TABLE B - 56

REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 1100°K

<u>Reaction</u>					ΔG <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 98 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.27
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 3.17
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 5.10
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 6.43
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 9.81
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 13.24
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 16.70
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 20.28
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 23.88
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 26.83

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REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 1200°K

<u>Reaction</u>					ΔG <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 98 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.18
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 3.01
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 4.88
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 6.19
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 9.52
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 12.93
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 16.42
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 20.06
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 23.71
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 26.71

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REACTIONS BETWEEN $\langle \text{PuO}_2 \rangle$ and $\langle \text{Ir} \rangle$ at 1300°K

<u>Reaction</u>					ΔG <u>kcal/mole</u>
$\langle \text{PuO}_2 \rangle$	+ 0.01	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 98 \rangle$	+ 0.01	$\langle \text{IrO}_2 \rangle$	+ 1.09
$\langle \text{PuO}_2 \rangle$	+ 0.025	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 95 \rangle$	+ 0.025	$\langle \text{IrO}_2 \rangle$	+ 2.85
$\langle \text{PuO}_2 \rangle$	+ 0.04	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 92 \rangle$	+ 0.04	$\langle \text{IrO}_2 \rangle$	+ 4.67
$\langle \text{PuO}_2 \rangle$	+ 0.05	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 90 \rangle$	+ 0.05	$\langle \text{IrO}_2 \rangle$	+ 5.94
$\langle \text{PuO}_2 \rangle$	+ 0.075	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 85 \rangle$	+ 0.075	$\langle \text{IrO}_2 \rangle$	+ 9.22
$\langle \text{PuO}_2 \rangle$	+ 0.10	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 80 \rangle$	+ 0.10	$\langle \text{IrO}_2 \rangle$	+ 12.63
$\langle \text{PuO}_2 \rangle$	+ 0.125	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 75 \rangle$	+ 0.125	$\langle \text{IrO}_2 \rangle$	+ 16.15
$\langle \text{PuO}_2 \rangle$	+ 0.15	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 70 \rangle$	+ 0.15	$\langle \text{IrO}_2 \rangle$	+ 19.83
$\langle \text{PuO}_2 \rangle$	+ 0.175	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 65 \rangle$	+ 0.175	$\langle \text{IrO}_2 \rangle$	+ 23.54
$\langle \text{PuO}_2 \rangle$	+ 0.195	$\langle \text{Ir} \rangle = \langle \text{PuO}_1 \cdot 61 \rangle$	+ 0.195	$\langle \text{IrO}_2 \rangle$	+ 26.59

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REACTIONS TO FORM (IrO₂) at 1000°K

<u>Reactions</u>				<u>ΔG</u> <u>kcal/mole</u>
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01 (IrO ₂)	+ 1.95
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025 (IrO ₂)	+ 4.82
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04 (IrO ₂)	+ 7.68
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05 (IrO ₂)	+ 9.63
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075 (IrO ₂)	+ 14.54
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10 (IrO ₂)	+ 12.46
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125 (IrO ₂)	+ 24.38
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15 (IrO ₂)	+ 29.39
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175 (IrO ₂)	+ 34.40
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195 (IrO ₂)	+ 38.50
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25 (IrO ₂)	+ 60.80

TABLE B - 60

REACTIONS TO FORM (IrO₂) at 1500°K

<u>Reactions</u>				<u>ΔG</u> <u>kcal/mole</u>
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01 (IrO ₂)	+ 1.28
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025 (IrO ₂)	+ 3.42
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04 (IrO ₂)	+ 5.66
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05 (IrO ₂)	+ 7.23
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075 (IrO ₂)	+ 11.30
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10 (IrO ₂)	+ 15.56
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125 (IrO ₂)	+ 20.03
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15 (IrO ₂)	+ 24.70
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175 (IrO ₂)	+ 29.41
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195 (IrO ₂)	+ 33.27
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25 (IrO ₂)	+ 43.81

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REACTIONS TO FORM (IrO₂) at 1600°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01	(IrO ₂)	+ 1.14
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 3.14
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 5.25
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 6.75
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 10.65
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 14.79
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 19.16
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 23.76
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 28.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 32.22
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 42.64

TABLE B - 62

REACTIONS TO FORM (IrO₂) at 1700°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01	(IrO ₂)	+ 1.01
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 2.86
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 4.84
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 6.27
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 10.00
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 14.01
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 18.29
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 22.82
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 27.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 31.17
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 41.48

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REACTIONS TO FORM (IrO₂) at 1800°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.99} >	+ 0.01	(IrO ₂)	+ 0.88
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 2.58
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 4.44
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 5.79
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 9.35
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 13.23
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 17.42
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 21.88
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 26.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 30.13
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 40.31

TABLE B - 64

REACTIONS TO FORM (IrO₂) at 1900°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.99} >	+ 0.01	(IrO ₂)	+ 0.74
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 2.30
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 4.03
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 5.31
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 8.70
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 12.45
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 16.56
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 20.94
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 25.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 29.08
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 39.14

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REACTIONS TO FORM (IrO₂) at 2000°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01	(IrO ₂)	+ 0.61
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 2.02
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 3.63
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 4.84
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 8.05
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 11.67
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 15.69
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 20.00
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 24.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 28.04
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 37.98

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REACTIONS TO FORM (IrO₂) at 2100°K

<u>Reactions</u>					ΔG kcal/mole
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01	(IrO ₂)	+ 0.47
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₂)	+ 1.74
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₂)	+ 3.22
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₂)	+ 4.36
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₂)	+ 7.40
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₂)	+ 10.89
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₂)	+ 14.82
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₂)	+ 19.07
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₂)	+ 23.42
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₂)	+ 26.99
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₂)	+ 36.81

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REACTIONS TO FORM (IrO₃) at 2200°K

<u>Reactions</u>					ΔG <u>kcal/mole</u>
<PuO ₂ >	+ 0.01	<Ir> = <PuO _{1.98} >	+ 0.01	(IrO ₃)	+ 0.34
<PuO ₂ >	+ 0.025	<Ir> = <PuO _{1.95} >	+ 0.025	(IrO ₃)	+ 1.46
<PuO ₂ >	+ 0.04	<Ir> = <PuO _{1.92} >	+ 0.04	(IrO ₃)	+ 2.82
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.90} >	+ 0.05	(IrO ₃)	+ 3.88
<PuO ₂ >	+ 0.075	<Ir> = <PuO _{1.85} >	+ 0.075	(IrO ₃)	+ 6.75
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.80} >	+ 0.10	(IrO ₃)	+ 10.11
<PuO ₂ >	+ 0.125	<Ir> = <PuO _{1.75} >	+ 0.125	(IrO ₃)	+ 13.95
<PuO ₂ >	+ 0.15	<Ir> = <PuO _{1.70} >	+ 0.15	(IrO ₃)	+ 18.13
<PuO ₂ >	+ 0.175	<Ir> = <PuO _{1.65} >	+ 0.175	(IrO ₃)	+ 22.33
<PuO ₂ >	+ 0.195	<Ir> = <PuO _{1.61} >	+ 0.195	(IrO ₃)	+ 25.94
<PuO ₂ >	+ 0.25	<Ir> = <PuO _{1.50} >	+ 0.25	(IrO ₃)	+ 35.64

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REACTIONS TO FORM (IrO₃) at 1000°K

<u>Reactions</u>					ΔG <u>kcal/mole</u>
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067	(IrO ₃)	+ 1.60
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167	(IrO ₃)	+ 3.95
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267	(IrO ₃)	+ 6.30
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333	(IrO ₃)	+ 7.90
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05	(IrO ₃)	+ 11.94
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667	(IrO ₃)	+ 15.99
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833	(IrO ₃)	+ 20.04
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10	(IrO ₃)	+ 24.19
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167	(IrO ₃)	+ 28.34
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13	(IrO ₃)	+ 31.74
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667	(IrO ₃)	+ 40.98

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REACTIONS TO FORM (IrO₃) at 1300°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 1.23
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 3.19
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 4.76
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 6.62
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 10.24
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 13.98
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 17.85
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 21.87
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 25.92
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 29.24
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 38.30

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REACTIONS TO FORM (IrO₃) at 1500°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.99
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 2.69
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 4.49
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 5.77
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 9.11
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 12.65
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 16.38
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 20.32
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 24.31
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 27.57
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 36.52

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REACTIONS TO FORM (IrO₃) at 1600°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.86
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 2.43
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 4.13
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 5.35
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 8.54
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 11.98
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 15.65
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 19.54
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 23.50
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 26.74
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 35.62

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REACTIONS TO FORM (IrO₃) at 1700°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.74
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 2.18
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 3.76
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 4.92
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 7.98
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 11.31
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 14.92
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 18.77
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 22.69
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 25.91
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 34.73

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REACTIONS TO FORM (IrO₃) at 1800°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.62
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 1.93
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 3.40
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 4.50
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 7.41
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 10.64
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 14.19
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 18.00
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 21.89
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 25.08
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 33.84

TABLE B - 74

REACTIONS TO FORM (IrO₃) at 1900°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.49
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 1.68
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 3.04
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 4.07
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 6.84
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 9.97
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 13.46
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 17.22
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 21.08
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 24.25
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 32.94

TABLE B - 75

REACTIONS TO FORM (IrO₃) at 2000°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.37
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 1.42
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 2.68
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 3.65
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 6.27
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 9.30
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 12.72
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 16.45
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 20.27
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 23.41
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 29.92

TABLE B - 76.

REACTIONS TO FORM (IrO₃) at 2100°K

<u>Reactions</u>				ΔG kcal/mole
<PuO ₂ >	+ 0.0067	<Ir> = <PuO _{1.98} >	+ 0.0067 (IrO ₃)	+ 0.25
<PuO ₂ >	+ 0.0167	<Ir> = <PuO _{1.95} >	+ 0.0167 (IrO ₃)	+ 1.17
<PuO ₂ >	+ 0.0267	<Ir> = <PuO _{1.92} >	+ 0.0267 (IrO ₃)	+ 2.32
<PuO ₂ >	+ 0.0333	<Ir> = <PuO _{1.90} >	+ 0.0333 (IrO ₃)	+ 3.22
<PuO ₂ >	+ 0.05	<Ir> = <PuO _{1.85} >	+ 0.05 (IrO ₃)	+ 5.71
<PuO ₂ >	+ 0.0667	<Ir> = <PuO _{1.80} >	+ 0.0667 (IrO ₃)	+ 8.63
<PuO ₂ >	+ 0.0833	<Ir> = <PuO _{1.75} >	+ 0.0833 (IrO ₃)	+ 11.99
<PuO ₂ >	+ 0.10	<Ir> = <PuO _{1.70} >	+ 0.10 (IrO ₃)	+ 15.68
<PuO ₂ >	+ 0.1167	<Ir> = <PuO _{1.65} >	+ 0.1167 (IrO ₃)	+ 19.47
<PuO ₂ >	+ 0.13	<Ir> = <PuO _{1.61} >	+ 0.13 (IrO ₃)	+ 22.58
<PuO ₂ >	+ 0.1667	<Ir> = <PuO _{1.50} >	+ 0.1667 (IrO ₃)	+ 28.94

TABLE B - 77

REACTIONS TO FORM (IrO₃) at 2200°K

Reactions		ΔG kcal/mole	
<PuO ₂ > + 0.0067	<Ir> = <PuO _{1.88} > + 0.0067 (IrO ₃)	+	0.12
<PuO ₂ > + 0.0167	<Ir> = <PuO _{1.95} > + 0.0167 (IrO ₃)	+	0.92
<PuO ₂ > + 0.0267	<Ir> = <PuO _{1.92} > + 0.0267 (IrO ₃)	+	1.96
<PuO ₂ > + 0.0333	<Ir> = <PuO _{1.90} > + 0.0333 (IrO ₃)	+	2.80
<PuO ₂ > + 0.05	<Ir> = <PuO _{1.85} > + 0.05 (IrO ₃)	+	5.14
<PuO ₂ > + 0.0667	<Ir> = <PuO _{1.80} > + 0.0667 (IrO ₃)	+	7.96
<PuO ₂ > + 0.0833	<Ir> = <PuO _{1.75} > + 0.0833 (IrO ₃)	+	11.26
<PuO ₂ > + 0.10	<Ir> = <PuO _{1.70} > + 0.10 (IrO ₃)	+	14.90
<PuO ₂ > + 0.1167	<Ir> = <PuO _{1.65} > + 0.1167 (IrO ₃)	+	18.66
<PuO ₂ > + 0.13	<Ir> = <PuO _{1.61} > + 0.13 (IrO ₃)	+	21.75
<PuO ₂ > + 0.1667	<Ir> = <PuO _{1.50} > + 0.1667 (IrO ₃)	+	27.96

TABLE B - 78

TEMPERATURES AT WHICH EQUILIBRIUM VALUES BECOME NEGATIVE FOR THE REDUCTION OF <PuO₂> TO SPECIFIC <PuO_{2-x}> COMPOSITIONS

Reaction

1. <PuO₂> + x<C> = <PuO_{2-x}> + x (CO)
2. <PuO₂> + x(CO) = <PuO_{2-x}> + x (CO₂)
3. <PuO₂> + x(H₂) = <PuO_{2-x}> + x (H₂O)

<PuO _{2-x} >	Temperature, °C (± 20)		
	Reaction 1	Reaction 2	Reaction 3
<PuO _{1.88} >	1047	1370	1290
<PuO _{1.95} >	1077	1540	1420
<PuO _{1.92} >	1100	1675	1515
<PuO _{1.90} >	1120	1790	1590
<PuO _{1.85} >	1160		1760
<PuO _{1.80} >	1195		1970
<PuO _{1.75} >	1240		
<PuO _{1.70} >	1282		
<PuO _{1.65} >	1322		
<PuO _{1.61} >	1357		
<PuO _{1.50} >	1420		

APPENDIX C
CALCULATIONS PERTAINING TO FSA SYSTEMS

TABLE C-1
DIMENSIONS OF 100 WATT SPHERE AS A FUNCTION OF TEMPERATURE

<u>Temp., °C</u>	<u>Diam, in.</u>	<u>Volume, cm³</u>	<u>Surface Area, cm²</u>
20	1.465	26.98	43.50
727	1.475	27.54	44.10
1027	1.481	27.85	44.46
1127	1.482	27.95	44.52
1227	1.484	28.06	44.64
1327	1.486	28.16	44.76
1427	1.488	28.26	44.88
1527	1.490	28.36	45.00
1627	1.491	28.44	45.06
1727	1.492	28.51	45.12
1827	1.493	28.56	45.18
1927	1.493	28.58	45.18

TABLE C-2

MAXIMUM GENERATION RATE OF IMPORTANT GASEOUS
SPECIES DUE TO FUEL DISSOCIATION
g/cm² sec

Temp. °K	(O ₂)	(PuO ₂)	(IrO ₃)	(IrO ₂)
1600	7.5x10 ⁻⁸	3.1x10 ⁻¹⁰	2.8x10 ⁻¹⁴	3.3x10 ⁻¹³
1700	1.5x10 ⁻⁶	3.7x10 ⁻⁹	9.0x10 ⁻¹¹	1.7x10 ⁻¹⁰
1800	1.7x10 ⁻³	3.4x10 ⁻⁸	1.1x10 ⁻⁷	4.1x10 ⁻⁸
2000	4.9	1.6x10 ⁻⁶	0.02	4.6x10 ⁻⁴
2100	148	7.3x10 ⁻⁶	3.63	2.5x10 ⁻²
2200	3317	3.1x10 ⁻⁵	21.2	0.95

Calculations based on fuel being <PuO₂.₀>. Gaseous Ir-O species are based on oxygen from fuel only, no impurity effects are considered.

TABLE C-3

MAXIMUM EFFUSION RATES THROUGH FSA VENTS
DUE TO DISSOCIATION OF FUEL

Temp. °K	g/y			
	(O ₂)	(PuO ₂)	(IrO ₃)	(IrO ₂)
1600	1.3×10^{-5}	5.3×10^{-8}	4.7×10^{-12}	5.7×10^{-11}
1700	2.6×10^{-3}	6.3×10^{-7}	1.5×10^{-8}	2.7×10^{-8}
1800	0.29	6.6×10^{-6}	1.9×10^{-5}	6.9×10^{-6}
2000	g/5 min			
	(O ₂)	(PuO ₂)	(IrO ₃)	(IrO ₂)
2000	7.8×10^{-3}	2.6×10^{-3}	3.2×10^{-5}	7.5×10^{-7}
2100	0.24	1.2×10^{-8}	5.8×10^{-3}	4.0×10^{-5}
2200	5.4	5.0×10^{-5}	0.03	1.5×10^{-3}

- a. Calculations based on <PuO₂.₀> fuel. Gaseous Ir-O species are due only to oxygen from fuel. No impurity effects are considered.
- b. The values were calculated assuming two vents, 0.002 in. diam. If vent covers are added, the calculated values would decrease.

Fig. 1 Calculated Partial Pressures at 1440°C

Fig. 2 Calculated Partial Pressures at $1827^\circ C$

Fig.3 Recession Rate With 0.2 Atm Oxygen Pressure

Fig.4 Recession Rate With 0.02 Atm Oxygen Pressure