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Relative humidity threshold for oxygen generation by high-purity plutonium dioxides in 3013 containers

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Abstract.

High-purity plutonium dioxide with adsorbed water in a sealed container can generate oxygen, but oxygen will not be generated in low water loading conditions. A threshold for oxygen generation has been observed in the hydrogen generation rate normalized by the specific power for high-purity plutonium dioxide. Normalized hydrogen generation rates above this threshold generate oxygen and below the threshold consume oxygen. The threshold rate is determined by the amount of water adsorbed onto the surface of the oxide. The relative humidity of the atmosphere that the oxide is in equilibrium with determines the amount of physisorbed water adsorbed on the surface. We calculate the relative humidity corresponding to the threshold rate as a function of mass and specific surface area of material stored in 3013 containers. Exceeding the calculated relative humidity may result in flammable gas mixtures of hydrogen and oxygen at some time during storage.

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List of Acronyms and Symbols

BET	methodology for describing the number of monolayers physically adsorbed on a surface developed by Brunuaer, Emmet and Teller
MIS	Materials Identification and Surveillance Program
eV	unit of energy, 1.6022e-19 J
ML	unit of surface coverage, monolayer
µmol	unit of amount, micromole = 10^{-6} mole
a_s	specific surface area, m^2 gm^{-1}
f_{H2O}	fraction of material mass that is water, unitless
$G(H2)$	hydrogen G-value, molecule per 100 eV or moles per Joule or µmol per Joule; 1 molecule per 100 eV = 0.1036 µmol J^{-1}
m_{H2O}	mass of water, kg
m_{PuO2}	mass of plutonium dioxide, kg
n_{ML}	number of monolayers, monolayers
P_{mat}	power of material, W
P_s	specific power of material, W kg^{-1}
R	ideal gas constant, 8314.5 kPa cm^3 mol^{-1} K^{-1}
RH	relative humidity, %
R_{ini}	initial rate of hydrogen generation, expressed either as moles per second or in a sealed container can be expressed as kPa per second or kPa per day
S_{rat}	ratio of alpha stopping power, unitless
T	temperature, K
V_g	free gas volume, cm^3
ϑ_{H2O}	mass of water in a monolayer for one square meter of plutonium oxide surface, 0.00022 gm m^{-2} ML^{-1}

Introduction

Radiolysis of water adsorbed on plutonium dioxide can produce oxygen gas.[1-3] Oxygen gas is also consumed by plutonium dioxide. This is evident by the decrease in oxygen in the gas phase over time in sealed systems[1, 3], the absence of oxygen gas in gas samples taken during destructive evaluation of 3013 containers[4], and the negative oxygen G-values observed at low water coverages when air is used as the fill gas[2]. This competition between oxygen generation and consumption complicates evaluation of conditions that can result in a flammable gas mixture in a sealed container. The current inventory of 3013 containers and the conditions at packaging has been evaluated and it was concluded that oxygen generation by high-purity plutonium dioxides would not occur in these containers.[5] The argument that was used, qualitative in nature, was based on the relative humidity at packaging, which, it was believed, was too low to support multi-layers of adsorbed water.

Hydrogen and oxygen gas generation observations from high-purity plutonium oxides in sealed containers have recently been summarized.[3] A broad range of specific power (2.1 to 14.7 W kg^{-1}) and specific surface area (0.8 to 21.5 m^2 gm^{-1}) was studied. It was found that normalizing gas generation rates by the specific power of the material resulted in linearizing several observables such as the maximum hydrogen pressure and maximum oxygen pressure. A threshold for oxygen gas generation was observed in the hydrogen gas generation rate normalized by the specific power, e.g. at normalized hydrogen gas generation rates above the threshold oxygen was generated and below the threshold oxygen was not generated. The initial hydrogen gas generation rate for high-purity plutonium dioxides can be calculated from the hydrogen G-value, for which we have a spread of values, the mass of water associated with the material, the material power and the free gas volume.[6] In addition, the amount of water on high-purity oxides is related to the relative humidity using the BET equation.[7] Using these relationships we can calculate the relative humidity that results in the normalized hydrogen gas generation rate being equal to the threshold for oxygen gas generation. We can quantify what has been a qualitative argument and provide guidance for future packaging of high-purity plutonium dioxide regarding the relative humidity. Relative humidity controls already exist for chloride container materials and the control level of 15% relative humidity would also ensure no oxygen would be generated.[8]

Derivation

The initial rate of hydrogen gas generation is given by[6]

$$R_{ini} = G(H2)\, P_{mat}\, f_{H2O}\, S_{rat}$$

Equation 1

Where R_{ini} is the initial hydrogen gas generation rate with units moles per second, *G(H2)* is the hydrogen G-value in units of moles per Joule (a Joule can be expressed in units of s Watt), P_{mat} is the power of the material in units of W, f_{H2O} is the fraction of the mass that is water adsorbed onto the plutonium dioxide (unitless), and S_{rat} is the ratio of the stopping power of alpha radiation in liquid water to plutonium dioxide (unitless). The hydrogen G-value is widely expressed in units of molecules of hydrogen produced per 100 eV of adsorbed dose, which Spinks and Woods, in their classic text *An Introduction to Radiation Chemistry,* point out 1 molecule $(100\ \mathrm{eV})^{-1}$ is equivalent to 0.1036 $\mu\mathrm{mol\ J}^{-1}$ (page 8).[9] The material power can be expressed as the product of the specific power and mass of PuO_2 and the fraction of water can be expressed as the ratio of the mass of water to the mass of PuO_2 (technically the sum of the mass of PuO_2 and the mass of water which, for small amounts of water, reduces to simply the mass of PuO_2).

$$P_{mat} = P_s\, m_{PuO2}$$

$$f_{H2O} = {}^{m_{H2O}}/_{m_{PuO2}}$$

$$R_{ini} = G(H2)\, P_s\, m_{PuO2}\, {}^{m_{H2O}}/_{m_{PuO2}}\, S_{rat}$$

$$R_{ini} = G(H2)\, P_s\, m_{H2O}\, S_{rat}$$

Equation 2

The mass of the water and plutonium dioxide is given in kg and the specific power has units of W kg^{-1}. The hydrogen gas generation rate normalized by the specific power at which oxygen gas begins to be generated is 0.025 kPa kg day^{-1} W^{-1} which expresses the rate in terms of pressure per time rather than moles per time. The initial rate per unit power within a sealed container can be expressed in units of kPa s^{-1} using the ideal gas law.

$$\frac{R_{ini,P}}{P_s} = G(H2)\, m_{H2O}\, S_{rat}\, \frac{R\,T}{V_g}$$

Equation 3

where R is the gas constant 8314.5 kPa cm^3 mol^{-1} K^{-1}, T is the temperature in K and V_g is the free gas volume in the sealed container. The normalized hydrogen gas generation rate is mostly dependent upon the mass of water within a container because the free gas volume changes little with increasing mass of material, Equation 8. This means that there is an amount of water which will result in oxygen gas generation regardless of the mass of plutonium dioxide loaded into a 3013 container. The 3013 Standard specifies a maximum value for the fraction of water, 0.005, for a package to be certified as compliant with the Standard. The fraction of water, using Equation 2 and Equation 3, is given by

$$f_{H2O} = \frac{\frac{R_{ini,P}}{P_s} V_g}{G(H2)\, m_{PuO2}\, S_{rat} R\, T}$$

Equation 4

In Equation 4, the fraction of water is a function of the mass of material, the free gas volume and the temperature. The free gas volume is also a function of the mass of material. For a Hanford container, the minimum free gas volume within the inner container[10, 11] is given by

$$V_g = 1879\ cm^3 - \frac{m_{PuO2}}{\rho_{PuO2}}$$

Equation 5

where ρ_{PuO2} is the pycnometer density of plutonium dioxide.

The amount of water adsorbed onto plutonium dioxide powder is a function of the specific surface area, the amount of plutonium dioxide and the number of monolayers of water.

$$m_{H2O} = \ a_s\, m_{PuO2}\, n_{ML}\, \theta_{H2O}$$

where a_s is the specific surface area in $m^2\ gm^{-1}$, n_{ML} is the number of monolayers of water in ML and ϑ_{H2O} is the mass of water in a monolayer for one square meter of plutonium oxide surface and is equal to 0.00022 $gm\ m^{-2}\ ML^{-1}$. The value of ϑ_{H2O},which relates the mass of water on a plutonium dioxide surface to the monolayer coverage, was derived from the surface density of plutonium atoms on the common crystallographic faces.[12] Substituting into Equation 3 and rearranging gives

$$\frac{R_{ini,P}}{P_s} = G(H2)\ a_s\, m_{PuO2}\, n_{ML}\, \theta_{H2O}\, S_{rat}\ \frac{R\, T}{V_g}$$

$$n_{ML} = \frac{\frac{R_{ini,P}}{P_s}}{\mathrm{G(H2)}\ a_s\, m_{PuO2}\, \theta_{\mathrm{H2O}}\, \mathrm{S_{rat}} \frac{R\, T}{V_g}}$$

Equation 6

We now have an expression that relates the water monolayer coverage to the initial hydrogen gas generation rate normalized by the specific power, the specific surface area of the plutonium dioxide and the mass of the plutonium dioxide. The last step is to relate the number of monolayers of water to the relative humidity (RH). Water physically adsorbs onto plutonium dioxide. The most common method to calculate the monolayer coverage from the RH is BET theory.[7]

$$n_{ML} = \frac{c\, RH}{(1 - RH)(1 - (c - 1)RH)}$$

Equation 7

where c is a constant related to the enthalpy of adsorption of water onto plutonium dioxide. The enthalpy of adsorption of water has been measured for water adsorbed onto plutonium dioxide powder resulting in c = 7.[13] The solution to Equation 7is complex:

$$RH = \frac{\sqrt{(c^2 - 4c + 4){n_{ML}}^2 + 2\, c^2\, n_{ML} + c^2} + c\, n_{ML} + c}{(2\, c - 2)n_{ML}}$$

$$\frac{-\sqrt{(c^2 - 4c + 4){n_{ML}}^2 + 2\, c^2\, n_{ML} + c^2} + c\, n_{ML} + c}{(2\, c - 2)n_{ML}}$$

Equation 8

Needless to say, substitution of Equation 6 into Equation 8 would be a visual mess. Nonetheless, Equation 8 can be used to find the RH at which oxygen gas is expected to be generated given a materials mass and specific surface area, which are needed to define n_{ML}, Equation 6.

The values of the variables used in Equation 6, Equation 7 and Equation 8 are given in Table 1. The value for the normalized hydrogen gas generation rate is taken from Figure 4 of Reference 3. The value for G(H2) is taken from the same set of experiments which resulted in the threshold value for in the normalized hydrogen gas generation rate, which results in a self-consistent set of variables. In Figure 5, a limiting value for G(H2) of 0.2 molecules per 100 eV of adsorbed dose (0.0207 µmol J^{-1}) is seen over the range of water adsorption studied.[3]

Table 1. Values of the variables

Variable	Value	Units
$R_{ini,P}/P_s$	0.040	kPa kg day^{-1} W^{-1}
G(H2) maximum value	0.0176 (or 0.17)	µmol s^{-1} W^{-1} (or molecules per 100eV)
G(H2) average value	0.0114 (or 0.11)	µmol s^{-1} W^{-1} (or molecules per 100eV)
G(H2) minimum value	0.0073 (or 0.07)	µmol s^{-1} W^{-1} (or molecules per 100eV)
Θ_{H2O}	0.00022	gm m^{-2} ML^{-1}
S_{rat}	3.6	---
R	8314.5	kPa cm^3 mol^{-1} K^{-1}
T	325	K
c	7	---
ρ_{PuO2}	11.5	gm cm^{-3}

Results

We are interested in the conditions that result in oxygen generation in high-purity plutonium dioxides for certified 3013 packages. If, in order to generate oxygen, a package exceeds the 3013 Standard's criterium of less than 0.005 fraction of water, then it is not a certified package. Equation 4 defines the boundary between packages that generate oxygen and those that do not. We plot the fraction of water as a function of the mass of material in Figure 1. Packages that exceed 0.005 fraction of water, indicated by the gray line, are not 3013 compliant. The hydrogen G-value affects this limit and for the maximum, average and minimum G(H2) values the limit is approximately 0.5 kg, 1.0 kg and 1.4 kg. Materials that are less than this limit are not 3013 compliant. See Appendix 1 for the data.

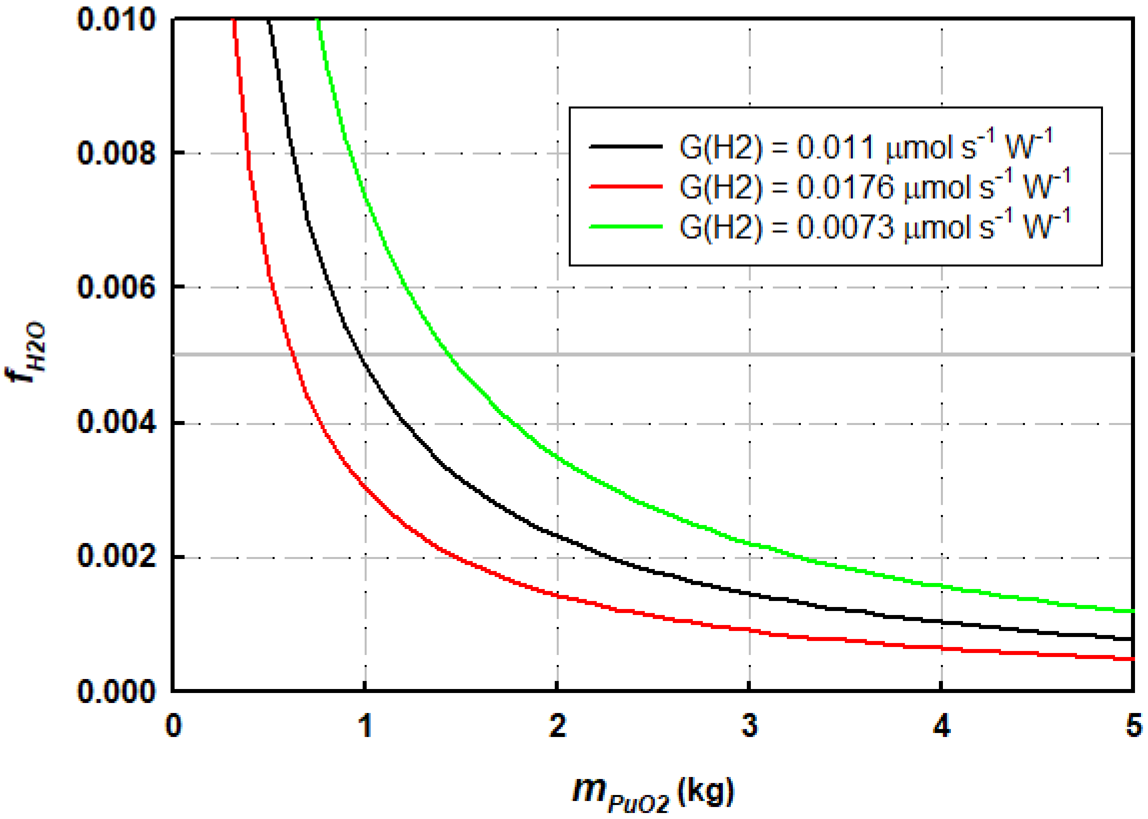

Figure 1. The fraction of water at the transition from no oxygen generated to oxygen generated as a function of the mass of high-purity plutonium dioxide. The 3013 Standard criteria of 0.005 is indicated in gray. A package with less than 0.533 kg of material must exceed the allowed fraction of water to generate oxygen. In considering the relative humidity required to generate oxygen in 3013 containers, masses of less than 0.533 kg are not considered.

We have solved Equation 8 for the range of plutonium dioxide masses from zero to five kgs with specific surface areas of one, two, three, four, and five square meters per gram for the maximum, average and minimum G(H2). The maximum specific surface area was determined by examining the measured values from destructive evaluation and in materials in the MIS inventory of represented materials.[14] For high-purity plutonium dioxides, the highest specific surface area after calcination to 950 °C was near five square meters per gram in both cases.

The result is shown in Figure 2. The data are given in Appendix 2. Hanford did not package unless the RH was less than 60%. The maximum amount of material which avoids oxygen generation is strongly dependent on the specific surface area and G(H2). The maximum amount of material is slightly over four kgs for a specific surface area of one m^2 gm^{-1} and drops to one kg for a specific surface area of five m^2 gm^{-1} for G(H2) = 0.076 µmol s^{-1} W^{-1}. For the average G(H2), specific surfaces areas of three, four and five m^2 gm^{-1} are limited at approximately 4.3, 3.4 and 2.8 kg respectively. For the minimum G(H2), only specific surface areas of four and five m^2 gm^{-1} are limited at approximately 4.8 and 4 kg respectively. A RH limit of 15% is imposed on material with alkaline earth chlorides by the 3013 Standard. If high-purity plutonium dioxide was packaged using this same limit, then oxygen would not be generated for all specific surface areas over the range of G(H2).

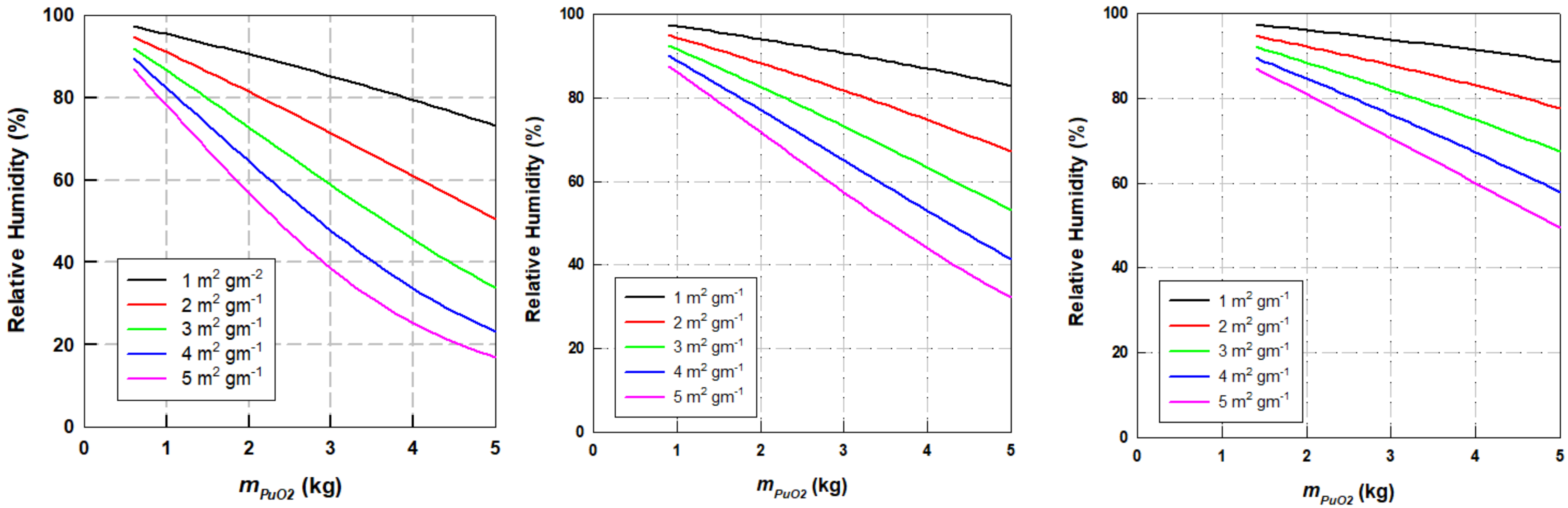

Figure 2. The relative humidity at which oxygen is generated in a Hanford inner container for high-purity plutonium dioxide of different specific surface areas illustrating the effect of G(H2). Left Maximum G(H2) = 0.0176 μmol s^{-1} W^{-1}; center Average G(H2) = 0.0114 μmol s^{-1} W^{-1}; right Minimum G(H2) = 0.0073 μmol s^{-1} W^{-1}. The 3013 Standard's limiting value for the moisture content of 0.5 wt% determines the mass of PuO_2 at which each graph starts, from Figure 1.

Discussion

One way to use the results in Figure 2 is to consider them in terms of probability. If the conditions of a package are below the magenta line for the maximum G(H2) graph, then it is assured that no oxygen will be generated. If the conditions are above the black line for the minimum G(H2), then the package will generate oxygen. Between these two extremes the probability of oxygen being generated will increase going from the maximum G(H2) case to the minimum G(H2) case. If the specific surface area of the material is known, then the conditions can be limited further.

In all cases studied to date, the oxygen that is generated reaches a maximum and then is consumed. For the materials studied in Reference 3, it takes a minimum of eight days to reach a flammable mixture. For high specific activity material, the oxygen is then consumed and reaches a non-flammable concentration in under 200 days. For material with a specific activity of 2.1 W kg^{-1}, the flammable mixture still exists at 849 days, but these were loaded with at least 1 wt% of water. Oxygen is not seen in the gas phase in 3013 containers at destructive evaluation in containers that ranged from five to fifteen years in age. Since all containers started with some oxygen, the initial oxygen was consumed. However, oxygen could have been generated and then consumed in some containers which generated hydrogen. The time for oxygen to be removed in containers in which it was initially generated is less than five years.

The relative humidity at which the threshold for oxygen generation is reached as shown in Figure 2 is calculated using G(H2) values derived from experiments in which the 0.5 wt% limit was exceeded. These conditions favored the generation of oxygen because the hydrogen gas generation rate is considered to be non-linearly dependent upon water loading.[15] As the monolayer coverage decreases the hydrogen G-value decreases. In this respect, the results can be considered conservative. Thus, it would not be surprising if materials at lower water loadings behaved according to the minimum G(H2) plot.

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Appendix 1. Fraction of H_2O

The calculated fraction of water used in Figure 1.

m_{PuO2} (kg)	f_{H2O} G(H2) = 0.0176 µmol s^{-1} W^{-1}	f_{H2O} G(H2) = 0.0114 µmol s^{-1} W^{-}	f_{H2O} G(H2) = 0.0073 µmol s^{-1} W^{-}
0.1	0.026838	0.050638	0.076680
0.2	0.013357	0.025201	0.038162
0.3	0.008863	0.016722	0.025322
0.4	0.006616	0.012483	0.018903
0.5	0.005268	0.009939	0.015051
0.6	0.004369	0.008243	0.012483
0.7	0.003727	0.007032	0.010649
0.8	0.003246	0.006124	0.009273
0.9	0.002871	0.005417	0.008203
1	0.002572	0.004852	0.007347
1.1	0.002326	0.004389	0.006647
1.2	0.002122	0.004004	0.006063
1.3	0.001949	0.003678	0.005569
1.4	0.001801	0.003398	0.005146
1.5	0.001673	0.003156	0.004779
1.6	0.00156	0.002944	0.004458
1.7	0.001461	0.002757	0.004175
1.8	0.001373	0.002591	0.003923
1.9	0.001294	0.002442	0.003698
2	0.001223	0.002308	0.003495
2.1	0.001159	0.002187	0.003312
2.2	0.001101	0.002077	0.003145
2.3	0.001048	0.001976	0.002993
2.4	0.000999	0.001884	0.002853
2.5	0.000954	0.001799	0.002725
2.6	0.000912	0.001721	0.002606
2.7	0.000874	0.001649	0.002497
2.8	0.000838	0.001581	0.002395
2.9	0.000805	0.001519	0.002300
3	0.000774	0.00146	0.002211
3.1	0.000745	0.001406	0.002129
3.2	0.000718	0.001354	0.002051
3.3	0.000692	0.001306	0.001978
3.4	0.000668	0.001261	0.001909
3.5	0.000646	0.001218	0.001845

3.6	0.000624	0.001178	0.001783
3.7	0.000604	0.00114	0.001726
3.8	0.000585	0.001103	0.001671
3.9	0.000567	0.001069	0.001619
4	0.000549	0.001036	0.001569
4.1	0.000533	0.001005	0.001522
4.2	0.000517	0.000976	0.001478
4.3	0.000502	0.000948	0.001435
4.4	0.000488	0.000921	0.001394
4.5	0.000474	0.000895	0.001355
4.6	0.000461	0.000871	0.001318
4.7	0.000449	0.000847	0.001283
4.8	0.000437	0.000824	0.001248
4.9	0.000425	0.000803	0.001216
5	0.000414	0.000782	0.001184

Appendix 2. Relative humidity.

The relative humidity solutions to Equation 7 for five specific surface areas as a function of mass of PuO_2. There are three tables for G(H2) = 0.0176 µmol s^{-1} W^{-1}, G(H2) = 0.0114 µmol s^{-1} W^{-1} and G(H2) = 0.0074 µmol s^{-1} W^{-1}. These solutions are shown graphically in Figure 2.

mass PuO_2	**Maximum G(H2) = 0.0176 µmol s^{-1} W^{-1} Specific Surface Areas**				
(kg)	**1 m^2 gm^{-1}**	**2 m^2 gm^{-1}**	**3 m^2 gm^{-1}**	**4 m^2 gm^{-1}**	**5 m^2 gm^{-1}**
0.1	99.6	99.1	98.7	98.3	97.8
0.2	99.1	98.3	97.4	96.5	95.7
0.3	98.7	97.4	96.1	94.8	93.5
0.4	98.2	96.5	94.7	93.0	91.3
0.5	97.8	95.6	93.4	91.2	89.1
0.6	97.3	94.7	92.1	89.5	86.9
0.7	96.9	93.8	90.7	87.7	84.7
0.8	96.4	92.9	89.4	85.9	82.5
0.9	96.0	92.0	88.0	84.1	80.3
1	95.5	91.0	86.7	82.4	78.1
1.1	95.0	90.1	85.3	80.6	76.0
1.2	94.5	89.2	83.9	78.8	73.8
1.3	94.1	88.2	82.5	77.0	71.6
1.4	93.6	87.3	81.2	75.2	69.5
1.5	93.1	86.3	79.8	73.4	67.3
1.6	92.6	85.4	78.4	71.6	65.2
1.7	92.1	84.4	77.0	69.9	63.1
1.8	91.6	83.4	75.6	68.1	61.0
1.9	91.1	82.5	74.2	66.3	58.9
2	90.6	81.5	72.8	64.6	56.9
2.1	90.1	80.5	71.4	62.8	54.9
2.2	89.6	79.5	70.0	61.1	52.9
2.3	89.0	78.5	68.6	59.3	51.0
2.4	88.5	77.5	67.2	57.6	49.0
2.5	88.0	76.5	65.8	55.9	47.2
2.6	87.4	75.5	64.4	54.3	45.3
2.7	86.9	74.5	63.0	52.6	43.6
2.8	86.4	73.5	61.6	51.0	41.8
2.9	85.8	72.5	60.2	49.3	40.1
3	85.3	71.4	58.8	47.8	38.5
3.1	84.7	70.4	57.4	46.2	36.9
3.2	84.1	69.4	56.1	44.7	35.4
3.3	83.6	68.3	54.7	43.2	33.9

mass PuO_2 (kg)	**Maximum G(H2) = 0.0176 µmol s^{-1} W^{-1} Specific Surface Areas**				
	1 m^2 gm^{-1}	**2 m^2 gm^{-1}**	**3 m^2 gm^{-1}**	**4 m^2 gm^{-1}**	**5 m^2 gm^{-1}**
3.4	83.0	67.3	53.4	41.7	32.5
3.5	82.4	66.2	52.0	40.3	31.2
3.6	81.9	65.2	50.7	38.9	29.9
3.7	81.3	64.2	49.4	37.5	28.6
3.8	80.7	63.1	48.1	36.2	27.5
3.9	80.1	62.1	46.8	34.9	26.3
4	79.5	61.0	45.5	33.6	25.2
4.1	78.9	60.0	44.2	32.4	24.2
4.2	78.3	58.9	43.0	31.2	23.2
4.3	77.6	57.8	41.8	30.1	22.3
4.4	77.0	56.8	40.6	29.0	21.4
4.5	76.4	55.7	39.4	28.0	20.6
4.6	75.8	54.7	38.2	27.0	19.8
4.7	75.1	53.6	37.1	26.0	19.0
4.8	74.5	52.6	36.0	25.0	18.3
4.9	73.9	51.6	34.9	24.2	17.6
5	73.2	50.5	33.9	23.3	17.0

mass PuO_2 (kg)	**Average G(H2) = 0.0114 µmol s^{-1} W^{-1} Specific Surface Areas**				
	1 m^2 gm^{-1}	**2 m^2 gm^{-1}**	**3 m^2 gm^{-1}**	**4 m^2 gm^{-1}**	**5 m^2 gm^{-1}**
0.1	99.7	99.5	99.2	98.9	98.6
0.2	99.5	98.9	98.4	97.8	97.3
0.3	99.2	98.4	97.5	96.7	95.9
0.4	98.9	97.8	96.7	95.6	94.5
0.5	98.6	97.2	95.9	94.5	93.2
0.6	98.3	96.7	95.0	93.4	91.8
0.7	98.1	96.1	94.2	92.3	90.4
0.8	97.8	95.5	93.3	91.1	89.0
0.9	97.5	95.0	92.5	90.0	87.6
1	97.2	94.4	91.6	88.9	86.1
1.1	96.9	93.8	90.7	87.7	84.7
1.2	96.6	93.2	89.9	86.6	83.3
1.3	96.3	92.6	89.0	85.4	81.9
1.4	96.0	92.0	88.1	84.2	80.4
1.5	95.7	91.4	87.2	83.1	79.0
1.6	95.4	90.8	86.3	81.9	77.6
1.7	95.0	90.2	85.4	80.7	76.1

mass PuO_2 (kg)	Average G(H2) = 0.0114 μmol s^{-1} W^{-1} Specific Surface Areas				
	1 m^2 gm^{-1}	2 m^2 gm^{-1}	3 m^2 gm^{-1}	4 m^2 gm^{-1}	5 m^2 gm^{-1}
1.8	94.7	89.6	84.5	79.5	74.7
1.9	94.4	88.9	83.6	78.3	73.2
2	94.1	88.3	82.6	77.1	71.8
2.1	93.8	87.7	81.7	75.9	70.4
2.2	93.4	87.0	80.8	74.7	68.9
2.3	93.1	86.4	79.9	73.5	67.5
2.4	92.8	85.7	78.9	72.3	66.0
2.5	92.4	85.1	78.0	71.1	64.6
2.6	92.1	84.4	77.0	69.9	63.1
2.7	91.8	83.8	76.1	68.7	61.7
2.8	91.4	83.1	75.1	67.5	60.3
2.9	91.1	82.4	74.1	66.2	58.8
3	90.7	81.8	73.2	65.0	57.4
3.1	90.4	81.1	72.2	63.8	56.0
3.2	90.0	80.4	71.2	62.6	54.6
3.3	89.6	79.7	70.2	61.4	53.2
3.4	89.3	79.0	69.2	60.1	51.9
3.5	88.9	78.3	68.2	58.9	50.5
3.6	88.5	77.6	67.3	57.7	49.1
3.7	88.2	76.9	66.3	56.5	47.8
3.8	87.8	76.1	65.3	55.3	46.5
3.9	87.4	75.4	64.3	54.1	45.2
4	87.0	74.7	63.2	52.9	43.9
4.1	86.6	74.0	62.2	51.7	42.6
4.2	86.2	73.2	61.2	50.5	41.4
4.3	85.8	72.5	60.2	49.4	40.2
4.4	85.4	71.7	59.2	48.2	39.0
4.5	85.0	71.0	58.2	47.1	37.8
4.6	84.6	70.2	57.2	45.9	36.7
4.7	84.2	69.5	56.2	44.8	35.5
4.8	83.8	68.7	55.2	43.7	34.4
4.9	83.3	67.9	54.2	42.6	33.4
5	82.9	67.1	53.2	41.5	32.3

mass PuO_2 (kg)	**Minimum G(H2) = 0.0073 µmol s^{-1} W^{-1} Specific Surface Areas**				
	1 m^2 gm^{-1}	**2 m^2 gm^{-1}**	**3 m^2 gm^{-1}**	**4 m^2 gm^{-1}**	**5 m^2 gm^{-1}**
0.1	99.8	99.6	99.5	99.3	99.1
0.2	99.6	99.3	98.9	98.6	98.2
0.3	99.5	98.9	98.4	97.8	97.3
0.4	99.3	98.5	97.8	97.1	96.4
0.5	99.1	98.2	97.3	96.4	95.5
0.6	98.9	97.8	96.7	95.6	94.5
0.7	98.7	97.4	96.1	94.9	93.6
0.8	98.5	97.0	95.6	94.1	92.7
0.9	98.3	96.7	95.0	93.4	91.7
1	98.1	96.3	94.4	92.6	90.8
1.1	97.9	95.9	93.9	91.8	89.8
1.2	97.7	95.5	93.3	91.1	88.9
1.3	97.5	95.1	92.7	90.3	87.9
1.4	97.3	94.7	92.1	89.5	86.9
1.5	97.1	94.3	91.5	88.7	85.9
1.6	96.9	93.9	90.9	87.9	85.0
1.7	96.7	93.5	90.3	87.1	84.0
1.8	96.5	93.1	89.7	86.3	83.0
1.9	96.3	92.6	89.0	85.5	82.0
2	96.1	92.2	88.4	84.7	81.0
2.1	95.9	91.8	87.8	83.8	80.0
2.2	95.7	91.4	87.2	83.0	78.9
2.3	95.4	90.9	86.5	82.2	77.9
2.4	95.2	90.5	85.9	81.3	76.9
2.5	95.0	90.1	85.2	80.5	75.9
2.6	94.8	89.6	84.6	79.6	74.8
2.7	94.5	89.2	83.9	78.8	73.8
2.8	94.3	88.7	83.3	77.9	72.8
2.9	94.1	88.3	82.6	77.1	71.7
3	93.8	87.8	81.9	76.2	70.7
3.1	93.6	87.3	81.2	75.3	69.6
3.2	93.4	86.9	80.6	74.4	68.5
3.3	93.1	86.4	79.9	73.6	67.5
3.4	92.9	85.9	79.2	72.7	66.4
3.5	92.6	85.4	78.5	71.8	65.4
3.6	92.4	85.0	77.8	70.9	64.3
3.7	92.1	84.5	77.1	70.0	63.2
3.8	91.9	84.0	76.4	69.1	62.1

mass PuO_2 (kg)	**Minimum G(H2) = 0.0073 µmol s^{-1} W^{-1} Specific Surface Areas**				
	1 m^2 gm^{-1}	**2 m^2 gm^{-1}**	**3 m^2 gm^{-1}**	**4 m^2 gm^{-1}**	**5 m^2 gm^{-1}**
3.9	91.6	83.5	75.6	68.2	61.1
4	91.4	83.0	74.9	67.2	60.0
4.1	91.1	82.5	74.2	66.3	58.9
4.2	90.8	82.0	73.5	65.4	57.9
4.3	90.6	81.4	72.7	64.5	56.8
4.4	90.3	80.9	72.0	63.5	55.7
4.5	90.0	80.4	71.2	62.6	54.7
4.6	89.7	79.9	70.5	61.7	53.6
4.7	89.5	79.3	69.7	60.7	52.5
4.8	89.2	78.8	69.0	59.8	51.5
4.9	88.9	78.2	68.2	58.9	50.4
5	88.6	77.7	67.4	57.9	49.4