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Calculated Equilibria Between
Adjacent Valence States of
Plutonium in Simple
Aqueous Solutions

F. H. Sweeton

Environmental Sciences Division
Publication No. 1539

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CALCULATED EQUILIBRIA BETWEEN ADJACENT VALENCE STATES
OF PLUTONIUM IN SIMPLE AQUEOUS SOLUTIONS

F. H. Sweeton*

ENVIRONMENTAL SCIENCES DIVISION

Publication No. 1539

*Presently with the Chemistry Division

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CALCULATED EQUILIBRIA BETWEEN ADJACENT VALENCE STATES OF PLUTONIUM IN SIMPLE AQUEOUS SOLUTIONS

F. H. Sweeton

ABSTRACT

Equilibrium data have been taken from the literature for the oxidation of Pu^{3+} to Pu^{4+} , Pu^{4+} to PuO_2^+ and PuO_2^+ to PuO_2^{2+} , and for the hydrolysis of these four plutonium species in dilute solutions. These data have been used to calculate the oxidation-reduction potentials (expressed as E_h) at which the concentrations of the two species (including the hydrolyzed forms) in each pair are equal. These calculations cover the pH range of 0 to 13. The results indicate that there are conditions under which each species can be predominant. The solubility of PuO_2 under environmental conditions is also discussed.

INTRODUCTION

Plutonium is unusual in having four valence states for its ions in aqueous solution. The different species vary greatly in their properties. For example, the adsorption of Pu(IV) on clay minerals has been found (Dahlman et al., 1976) to be about 2 orders of magnitude greater than that of Pu(VI). Since there is a strong interest in the properties of plutonium under environmental conditions as well as under the highly acid conditions used in chemical processing, a simple method for determining the equilibrium ratios of the different species under a variety

of conditions is of interest.

Plutonium ions in general hydrolyze to an appreciable degree, and therefore the hydrolyzed species need to be included when calculating the amount of plutonium in the different valence states. The purpose of the following discussion is to use the data presently available in the literature to calculate the concentration ratios of the different plutonium valence states under a variety of conditions.

METHOD OF CALCULATING CONDITIONS GIVING EQUAL CONCENTRATIONS OF ADJACENT SPECIES

The following calculations are limited to the data presently available for plutonium species in dilute solutions. Therefore they consider only mononuclear hydrolytic species and neglect polynuclear. If future data show that other mononuclear species exist, these calculations will need to be expanded.

The equilibria used in the calculations are listed in Table 1. The ratio of the concentrations of any two plutonium valence states can be determined by algebraically combining appropriate equations from this list and by substituting in values of the equilibrium constants taken from the literature. The constants used here are listed in Table 2.

For calculating the ratio of Pu(IV) to Pu(III) it is necessary to express their concentrations in terms of the various hydrolysis species. According to Baes and Mesmer (1976) these species are:

$$[\text{Pu(III)}] = [\text{Pu}^{3+}] + [\text{PuOH}^{2+}] \quad (1)$$

and

Table 1. Equilibria Used in the Calculations

<u>Reaction</u>	<u>Equilibrium Constant</u>
<u>Oxidation-Reduction</u>	
1. $\text{Pu}^{3+} + \text{H}^+ = \text{Pu}^{4+} + 1/2 \text{H}_2$	$K_{34} = \frac{[\text{Pu}^{4+}] P_{\text{H}_2}^{1/2}}{[\text{Pu}^{3+}] [\text{H}^+]}$
2. $\text{Pu}^{4+} + 2\text{H}_2\text{O} = \text{PuO}_2^+ + 3\text{H}^+ + 1/2 \text{H}_2$	$K_{45} = \frac{[\text{PuO}_2^+] [\text{H}^+]^3 P_{\text{H}_2}^{1/2}}{[\text{Pu}^{4+}]}$
3. $\text{PuO}_2^+ + \text{H}^+ = \text{PuO}_2^{2+} + 1/2 \text{H}_2$	$K_{56} = \frac{[\text{PuO}_2^{2+}] P_{\text{H}_2}^{1/2}}{[\text{PuO}_2^+] [\text{H}^+]}$
<u>Hydrolysis</u>	
4. $\text{Pu}^{3+} + \text{H}_2\text{O} = \text{PuOH}^{2+} + \text{H}^+$	$K_{3\text{H}1} = \frac{[\text{PuOH}^{2+}] [\text{H}^+]}{[\text{Pu}^{3+}]}$
5. $\text{Pu}^{4+} + \text{H}_2\text{O} = \text{PuOH}^{3+} + \text{H}^+$	$K_{4\text{H}1} = \frac{[\text{PuOH}^{3+}] [\text{H}^+]}{[\text{Pu}^{4+}]}$
6. $\text{Pu}^{4+} + 2\text{H}_2\text{O} = \text{Pu}(\text{OH})_2^{2+} + 2\text{H}^+$	$K_{4\text{H}2} = \frac{[\text{Pu}(\text{OH})_2^{2+}] [\text{H}^+]^2}{[\text{Pu}^{4+}]}$
7. $\text{Pu}^{4+} + 3\text{H}_2\text{O} = \text{Pu}(\text{OH})_3^+ + 3\text{H}^+$	$K_{4\text{H}3} = \frac{[\text{Pu}(\text{OH})_3^+] [\text{H}^+]^3}{[\text{Pu}^{4+}]}$
8. $\text{Pu}^{4+} + 4\text{H}_2\text{O} = \text{Pu}(\text{OH})_4^0 + 4\text{H}^+$	$K_{4\text{H}4} = \frac{[\text{Pu}(\text{OH})_4^0] [\text{H}^+]^4}{[\text{Pu}^{4+}]}$
9. $\text{Pu}^{4+} + 5\text{H}_2\text{O} = \text{Pu}(\text{OH})_5^- + 5\text{H}^+$	$K_{4\text{H}5} = \frac{[\text{Pu}(\text{OH})_5^-] [\text{H}^+]^5}{[\text{Pu}^{4+}]}$

Table 1. Equilibria Used in The Calculations (Cont'd)

<u>Reaction</u>	<u>Equilibrium Constant</u>
<u>Hydrolysis</u> (Cont'd)	
10. $\text{PuO}_2^+ + \text{H}_2\text{O} = \text{PuO}_2\text{OH}^0 + \text{H}^+$	$K_{5\text{H}1} = \frac{[\text{PuO}_2\text{OH}^0] [\text{H}^+]}{[\text{PuO}_2^+]}$
11. $\text{PuO}_2^{2+} + \text{H}_2\text{O} = \text{PuO}_2\text{OH}^+ + \text{H}^+$	$K_{6\text{H}1} = \frac{[\text{PuO}_2\text{OH}^+] [\text{H}^+]}{[\text{PuO}_2^{2+}]}$

Table 2. Values of Equilibrium Constants Used in the Calculations

<u>Constant</u>	<u>Log K</u>	<u>Reference</u>
K_{34}	-16.37	Rai and Serne (1977)
K_{45}	-19.50	"
K_{56}	-15.7	"
K_{3H1}	- 8.0	Baes and Mesmer (1976)
K_{4H1}	- .50	"
K_{4H2}	- 2.3	"
K_{4H3}	- 5.3	"
K_{4H4}	- 9.5	"
K_{4H5}	-15.0	"
K_{5H1}	- 9.7	"
K_{6H1}	- 5.6	"

$$\begin{aligned}
[\text{Pu(IV)}] = & [\text{Pu}^{4+}] + [\text{PuOH}^{3+}] + [\text{Pu(OH)}_2^{2+}] + [\text{Pu(OH)}_3^{+}] \\
& + [\text{Pu(OH)}_4^0] + [\text{Pu(OH)}_5^{-}]
\end{aligned} \quad (2)$$

where brackets indicate concentrations in moles per liter.

The equilibrium constants of lines 4 to 9 in Table 1 can be substituted into the above equations to give the following equations:

$$[\text{Pu(III)}] = [\text{Pu}^{3+}] \left(1 + \frac{K_{3H1}}{[\text{H}^+]} \right) \quad (3)$$

and

$$[\text{Pu(IV)}] = [\text{Pu}^{4+}] \left(1 + \frac{K_{4H1}}{[\text{H}^+]} + \frac{K_{4H2}}{[\text{H}^+]^2} + \frac{K_{4H3}}{[\text{H}^+]^3} + \frac{K_{4H4}}{[\text{H}^+]^4} + \frac{K_{4H5}}{[\text{H}^+]^5} \right) \quad (4)$$

The ratio of the two species is then:

$$\frac{[\text{Pu(IV)}]}{[\text{Pu(III)}]} = \frac{[\text{Pu}^{4+}] \left(1 + \frac{K_{4H1}}{[\text{H}^+]} + \frac{K_{4H2}}{[\text{H}^+]^2} + \frac{K_{4H3}}{[\text{H}^+]^3} + \frac{K_{4H4}}{[\text{H}^+]^4} + \frac{K_{4H5}}{[\text{H}^+]^5} \right)}{[\text{Pu}^{3+}] \left(1 + \frac{K_{3H1}}{[\text{H}^+]} \right)} \quad (5)$$

The equilibrium constant expression for K_{34} (line 1 in Table 1) can be substituted for the ratio $[\text{Pu}^{4+}]/[\text{Pu}^{3+}]$ in the above expression to give

$$\frac{[\text{Pu(IV)}]}{[\text{Pu(III)}]} = \frac{K_{34} [\text{H}^+] \left(1 + \frac{K_{4H1}}{[\text{H}^+]} + \frac{K_{4H2}}{[\text{H}^+]^2} + \frac{K_{4H3}}{[\text{H}^+]^3} + \frac{K_{4H4}}{[\text{H}^+]^4} + \frac{K_{4H5}}{[\text{H}^+]^5} \right)}{P_{\text{H}_2}^{1/2} \left(1 + \frac{K_{3H1}}{[\text{H}^+]} \right)} \quad (6)$$

The following relationship can be used to express $P_{\text{H}_2}^{1/2}$ in terms of

the potential of an inert electrode versus a reference electrode:

$$E_h = E - E_R = 59.16 \log \frac{[H^+]}{P_{H_2}^{1/2}} \quad (7)$$

where "E" is the measured potential of the inert electrode relative to the reference electrode, and "E_R" is the potential of the reference electrode relative to a standard H₂ electrode (both measured at 25°C). Both potentials are expressed in millivolts, "[H⁺]" in moles per liter, and "P" in atmospheres. Substitution of equation 7 into equation 6 and changing to logarithms gives

$$\log \frac{[Pu(IV)]}{[Pu(III)]} = \frac{E_h}{59.16} + \log \left(\frac{K_{34} \left(1 + \frac{K_{4H1}}{[H^+]} + \frac{K_{4H2}}{[H^+]^2} + \frac{K_{4H3}}{[H^+]^3} + \frac{K_{4H4}}{[H^+]^4} + \frac{K_{4H5}}{[H^+]^5} \right)}{1 + \frac{K_{3H1}}{[H^+]}} \right) \quad (8)$$

where E_h has been substituted for E - E_R.

When [Pu(III)] = [Pu(IV)]

$$E_h = - 59.16 \log \left(\frac{K_{34} \left(1 + \frac{K_{4H1}}{[H^+]} + \frac{K_{4H2}}{[H^+]^2} + \frac{K_{4H3}}{[H^+]^3} + \frac{K_{4H4}}{[H^+]^4} + \frac{K_{4H5}}{[H^+]^5} \right)}{1 + \frac{K_{3H1}}{[H^+]}} \right) \quad (9)$$

By similar procedures it can be shown that

$$\log \frac{[Pu(V)]}{[Pu(IV)]} = \frac{E_h}{59.16} + \log \left(\frac{K_{45} \left(1 + \frac{K_{5H1}}{[H^+]} \right)}{[H^+]^4 \left(1 + \frac{K_{4H1}}{[H^+]} + \frac{K_{4H2}}{[H^+]^2} + \frac{K_{4H3}}{[H^+]^3} + \frac{K_{4H4}}{[H^+]^4} + \frac{K_{4H5}}{[H^+]^5} \right)} \right) \quad (10)$$

and that when $[\text{Pu(IV)}] = [\text{Pu(V)}]$

$$E_h = -59.16 \log \left(\frac{K_{45} \left(1 + \frac{K_{5H1}}{[H^+]} \right)}{[H^+]^4 \left(1 + \frac{K_{4H1}}{[H^+]} + \frac{K_{4H2}}{[H^+]^2} + \frac{K_{4H3}}{[H^+]^3} + \frac{K_{4H4}}{[H^+]^4} + \frac{K_{4H5}}{[H^+]^5} \right)} \right). \quad (11)$$

For the V and VI states, the corresponding expressions are:

$$\log \frac{[\text{Pu(VI)}]}{[\text{Pu(V)}]} = \frac{E_h}{59.16} + \log \left(\frac{K_{56} \left(1 + \frac{K_{6H1}}{[H^+]} \right)}{1 + \frac{K_{5H1}}{[H^+]}} \right) \quad (12)$$

and

$$E_h = -59.16 \log \left(\frac{K_{56} \left(1 + \frac{K_{6H1}}{[H^+]} \right)}{1 + \frac{K_{5H1}}{[H^+]}} \right). \quad (13)$$

With equations 8, 10 and 12 and the data in Table 2, the ratio of IV to III, V to IV and VI to V can be calculated for any combination of E_h and pH. Equations 9, 11 and 13 can be used similarly to calculate the E_h 's which give equal concentrations of III and IV, IV and V, and V and VI.

RESULTS AND DISCUSSION

Values of E_h calculated as described above are listed in Table 3 and are plotted in Figure 1. The values of E_h for both 0.2 atmosphere of O_2 and 1.0 atmosphere of H_2 are also plotted for reference.

Before these curves are discussed, some mention should be made of the limitations in the use of these calculations. For one thing, an inert electrode such as platinum may not give the equilibrium potential that

Table 3. Calculated E_h Giving Equal Concentrations of Adjacent Valence Species of Pultonium at Various pH's (Concentrations include all hydrolyzed species)

pH	E_h (versus Std. H_2 Electrode)		
	III=IV	IV=V	V=VI
0	+ 961.3	+1160.8	+928.8
1	928.8	954.2	928.8
2	853.5	795.3	928.8
3	730.1	682.0	928.7
4	557.7	617.8	928.2
5	337.3	602.2	923.1
6	74.13	628.3	896.5
7	- 213.2	681.1	845.0
8	- 492.8	739.0	787.2
9	- 744.8	794.0	732.3
10	- 983.6	827.7	702.6
11	-1220.5	838.8	687.5
12	-1457.1	839.9	686.4
13	-1693.7	840.1	686.3

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Fig. 1. Conditions for equal concentrations of adjacent valence species of plutonium at equilibrium.

represents the chemical state of the system to which one wants to apply these calculations. The platinum electrode is known to be unreliable in systems containing appreciable concentrations of O_2 . Another questionable aspect might be the assumption that the plutonium species are at equilibrium, for plutonium is often very slow to equilibrate; interconversion between the IV and V states is particularly slow due to the involvement of 2 oxygen atoms in the conversion. Another complicating factor could be the interference of the chemically-active species being generated continuously by the radioactive α decay; such interference may prevent a true chemical equilibrium ever being attained. Another uncertainty in using the calculations stems from the uncertainty in the data presently available in the literature.

Although only the E_h 's giving equal concentrations of different valence states have been plotted, the curves may be used to estimate ratios of valence states at other values of E_h . As an example let us consider a chemical system in which the measured E_h and pH are 0.400 volt and 6.0 respectively. The graph shows that for the Pu(III) and Pu(IV) concentrations to be equal at this pH, the E_h should be 0.070 volt. The measured E_h is thus 0.330 volt higher. According to equation 8 each .05916 volt increase in E_h should raise the IV/III ratio by a factor of 10; thus the excess voltage represents about 5.5 orders of magnitude, or a ratio of IV/III of about 3×10^5 . In the same manner the given value of E_h is about 0.230 volt below that for equal concentrations of IV and V; this voltage difference is equivalent to 7000 times as much IV as V.

The area cut by the curves in Figure 1 can be broken into four main zones, one for each of the Pu valence states. The largest of these is

the Pu(IV) region on the right and between the III-IV and IV-V lines. At these pH's the calculated solubility of PuO_2 is very low, as is shown in Appendix A. Consequently a solution which appears to contain any appreciable concentration of Pu(IV) can be suspected of being a suspension of the polymer; this is commonly observed when the pH of plutonium process solutions is raised above 1 or 2. The Pu(V) region lies in the slightly acidic oxidizing part of the diagram. The Pu(VI) region is associated with oxidizing and highly-acidic conditions, and the Pu(III) region with acidic and reducing conditions.

REFERENCES

- Charles F. Baes, Jr. and Robert E. Mesmer, "The Hydrolysis of Cations," John Wiley and Sons, New York, 1976.
- Dhanpat Rai and R. J. Serne, "Plutonium Activities in Soil Solution and the Stability and Formation of Selected Plutonium Minerals," J. Environ. Qual. 6, 89-95 (1977).
- F. H. Sweeton, T. Tamura and E. A. Bondietti, unpublished work on dissolution of crystalline PuO_2 .
- Dhanpat Rai and R. J. Serne, "Solution Species of ^{239}Pu in Oxidizing Environments: I. Polymeric Pu(IV)," PNL-SA-6994, Pacific Northwest Laboratory, Richland, Washington, 1978.
- R. C. Dahlman et al. in Environmental Sciences Div. Annual Prog. Report Period Ending Sept. 30, 1976, ORNL-5257, p. 140.

APPENDIX A

THE SOLUBILITY OF PuO_2

Knowledge of the solubility of PuO_2 is important in estimating the environmental transport of Pu(IV) as a soluble species. Baes and Mesmer (1976) have estimated the solubility constant K_{S10} for the dissolution of PuO_2 to give monomeric unhydrolyzed Pu^{4+} :

and

$$K_{S10} = \frac{[\text{Pu}^{4+}]}{[\text{H}^+]^4} .$$

Their estimated value of $\log K_{S10}$ is -6.5, which means that with changing pH

$$\log [\text{Pu}^{4+}] = -6.5 - 4 \cdot \text{pH} .$$

To get the total concentration of monomeric Pu(IV) in solution we can add the concentrations of the monomeric hydrolyzed species as given previously in equation 4. The final equation becomes:

$$\log [\text{Pu(IV)}] = -6.5 - 4 \cdot \text{pH} + \log \left(1 + \frac{K_{4H1}}{[\text{H}^+]} + \frac{K_{4H2}}{[\text{H}^+]^2} + \frac{K_{4H3}}{[\text{H}^+]^3} + \frac{K_{4H4}}{[\text{H}^+]^4} + \frac{K_{4H5}}{[\text{H}^+]^5} \right) .$$

This equation has been used to calculate the curve labeled "Baes and Mesmer" in Figure 2; the values of the constants in the equation were taken from Table 2.

Rai and Serne (1978) have reported data that put an upper limit on the solubility of PuO_2 in the pH range 4 to 8. They give the concentration

Fig. 2. Calculated solubility of PuO_2 as function of pH.

of Pu left in the solution phase after neutralizing acidic solutions of $\text{Pu}(\text{NO}_3)_4$, digesting for 90 days or more, centrifuging for 40 minutes at 6000 rpm and passing through a 0.015 or 0.1 μm filter. Their data also are plotted in Figure 2 and are labeled "Rai and Serne". These values are much higher than predicted by the Baes and Mesmer constants. Part of this difference is likely due to increased activity of the precipitated oxide in its hydrous, non-crystalline form. It is also possible that some of the Pu in the solution was in the form of particles of the oxide too small to filter out or centrifuge out. Another possibility is that some of the activity in the solution was in an oxidized state, for the tests were run in the presence of air.

Sweeton, Tamura and Bondietti have made exploratory studies of the Pu activity picked up by dilute solutions (HClO_4 and NaOH) in contact with crystalline PuO_2 for over four months. Pu(IV) was separated from larger concentrations of Pu(VI) in the final solutions by extracting aliquots with TTA (thenoyltrifluoroacetone) immediately after making them 1 N in HNO_3 . The Pu(IV) concentrations found are plotted in Figure 2. These data are also considerably larger than expected from the Baes and Mesmer data. Autoradiography of these solutions (before extraction) indicated that they contained appreciable amounts of activity in the form of particulates. These particulates are presumed to have been released from the original solid by a spallation process resulting from alpha-recoils. One might postulate that the relatively high apparent solubilities of Pu(IV) in the latter tests might have been the result of TTA solutions extracting very small particles of solid as well as the monomeric Pu(IV) in true solution.

In light of the wide variation of these data for the solubility of PuO_2 , it is clear that additional studies of this difficult system are required in order to have reliable values in this neutral pH range.

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