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# **Estimate of Actinide Concentration by Radioactive Decay**

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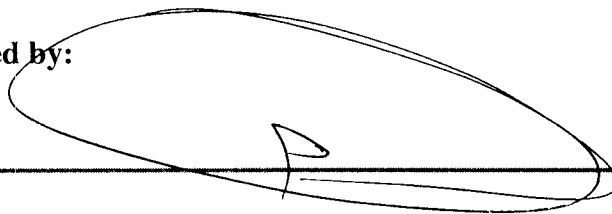
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## Summary of Revisions

3/2005

Revision 0, Initial Issue

# Prediction of Actinide Concentration by Radioactive Decay

## Executive Summary

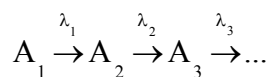
The basic principles of radioactive decay series are presented in order to develop a method for estimating the concentration of selected actinides not currently included in the HLW Waste Characterization System. Example calculations are performed to demonstrate the application and results of this method. Although transient or secular equilibrium will place an upper bound on the concentration of decay products, due to the long half-lives of the parent nuclides, the time scale over which equilibrium is obtained may make the assumption too conservative for current application. However, after evaluation, it may be determined that the degree of conservatism in this assumption is not too extreme. In that case, the calculations can be greatly simplified.

## Introduction

As part of the upgrades planned for the SRS High Level Waste Characterization System (WCS), additional constituents, both stable and radioactive, are being added to the database.<sup>1</sup> The objective here is to develop a method, using radioactive decay series, to estimate the concentration of selected actinides in SRS high level waste sludge.

## Radioactive Decay Series

Consider a parent nuclide with activity,  $A_1(t)$ , that undergoes radioactive decay to a radioactive daughter product with activity,  $A_2(t)$ . The daughter product, in turn, undergoes radioactive decay to a granddaughter product with activity,  $A_3(t)$ , which is itself radioactive. Furthermore, let each radionuclide have a corresponding decay constant,  $\lambda_j$ , where  $j = 1, 2, 3$ . This may be visualized graphically as



Assume initial activities of  $A_1(t=0) = A_{1,0}$ ,  $A_2(t=0) = A_3(t=0) = 0$ . Now the basic balance equation for activity,  $A_j(t)$ , may be written as<sup>2</sup>

$$\frac{dA_1}{dt} = -\lambda_1 A_1(t), \tag{1}$$

$$\frac{dA_2}{dt} = \lambda_2 A_1(t) - \lambda_2 A_2(t), \quad (2)$$

$$\frac{dA_3}{dt} = \lambda_3 A_2(t) - \lambda_3 A_3(t). \quad (3)$$

Equations (1) through (3) may be easily solved either by direct integration using an integrating factor or by Laplace transforms. The resulting solutions are

$$A_1(t) = A_{1,0} \exp[-\lambda_1 t],$$

$$A_2(t) = \left( \frac{\lambda_2 A_1(t)}{\lambda_2 - \lambda_1} \right) (1 - \exp[-(\lambda_2 - \lambda_1)t]),$$

$$A_3(t) = \left( \frac{\lambda_3 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \right) A_1(t) \left\{ 1 - \left( \frac{\lambda_3 - \lambda_1}{\lambda_3 - \lambda_2} \right) \exp[-(\lambda_2 - \lambda_1)t] + \left( \frac{\lambda_2 - \lambda_1}{\lambda_3 - \lambda_2} \right) \exp[-(\lambda_3 - \lambda_1)t] \right\},$$

after applying the assumed initial conditions given above.

In order to keep these equations in a format consistent with those in WCS proceed in the following manner. First, note the relation between the decay constant,  $\lambda_j$ , and the corresponding half-life,  $T_{1/2}$ , namely

$$\lambda_j = \frac{\ln 2}{T_{1/2,j}}.$$

Substitute this relation into the exponential terms to find

$$\exp[-\lambda_j t] = \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,j}}},$$

and

$$\exp[-(\lambda_j - \lambda_k)t] = \left( \frac{1}{2} \right)^{\frac{(T_{1/2,k} - T_{1/2,j})}{T_{1/2,j} T_{1/2,k}} t}$$

Now, substituting these relations into the above equations results in the following

$$A_1(t) = A_{1,0} \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,1}}}, \quad (4)$$

$$A_2(t) = \frac{T_{1/2,1}}{\left( T_{1/2,1} - T_{1/2,2} \right)} A_1(t) \left[ 1 - \left( \frac{1}{2} \right)^{\frac{\left( T_{1/2,1} - T_{1/2,2} \right) t}{T_{1/2,1} T_{1/2,2}}} \right], \quad (5)$$

$$A_3(t) = \frac{T_{1/2,1}^2}{\left( T_{1/2,1} - T_{1/2,2} \right) \left( T_{1/2,1} - T_{1/2,3} \right)} A_1(t) \times \left[ 1 - \frac{T_{1/2,2}}{T_{1/2,1}} \frac{\left( T_{1/2,1} - T_{1/2,3} \right)}{\left( T_{1/2,2} - T_{1/2,3} \right)} \left( \frac{1}{2} \right)^{\frac{\left( T_{1/2,1} - T_{1/2,2} \right) t}{T_{1/2,1} T_{1/2,2}}} + \frac{T_{1/2,3}}{T_{1/2,1}} \frac{\left( T_{1/2,1} - T_{1/2,2} \right)}{\left( T_{1/2,2} - T_{1/2,3} \right)} \left( \frac{1}{2} \right)^{\frac{\left( T_{1/2,1} - T_{1/2,3} \right) t}{T_{1/2,1} T_{1/2,3}}} \right]. \quad (6)$$

It is important to note that Eqs. (5) and (6) may be easily generalized to the case when the initial activities,  $A_2(t=0)$  and  $A_3(t=0)$ , are not zero. In other words,  $A_2(t=0) = A_{2,0}$ , and  $A_3(t=0) = A_{3,0}$ . In this case an additional term in the form

$$A_{j,0} \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,j}}}$$

must be added to each equation for  $j = 2$  and  $j = 3$ , as appropriate. This term accounts for the initial activity of isotope  $j$ .

At this point, note that for decay times large compared to the half-lives of a particular decay chain (more formally, in the limit as  $t \rightarrow \infty$ ), Eqs. (5) and (6) reduce to the following simple forms

$$A_2(t) = \frac{T_{1/2,1}}{\left( T_{1/2,1} - T_{1/2,2} \right)} A_1(t), \quad (7)$$

$$A_3(t) = \frac{T_{1/2,1}^2}{\left( T_{1/2,1} - T_{1/2,2} \right) \left( T_{1/2,1} - T_{1/2,3} \right)} A_1(t). \quad (8)$$



Eqs. (7) and (8) are the equilibrium activities of daughter product,  $A_2(t)$ , and granddaughter product,  $A_3(t)$ . As written, Eqs. (7) and (8) describe the condition of transient equilibrium. In transient equilibrium the daughter and granddaughter product activities increase until each is a constant multiple of the parent activity. After that they remain constant. On the other hand, if the half-life of the parent nuclide is very much greater than the half-lives of either daughter or granddaughter product, then Eqs. (7) and (8) simplify even further to

$$A_2(t) = A_1(t),$$

$$A_3(t) = A_1(t).$$

This condition is known as secular equilibrium.

## Discussion

Equations (4) through (6) will be implemented in the revised version of WCS<sup>1</sup> in order to estimate the concentration of the following actinides: Ra-226, Ra-228, Ac-227, Th-229, Th-230, and Pa-231. However, before these equations can be used to make predictions, the initial conditions assumed above (known parent concentration and zero concentration for daughter and granddaughter products) must be justified.

In order to use uranium as nuclear fuel it must first be refined.<sup>3</sup> The steps in refining uranium from crude ore are classified as *concentration*, *purification*, and *conversion*. In the concentration step, uranium is separated from most of the nonuraniferous diluents that accompany uranium in nature. This step increases the uranium oxide content from a few tenths of a percent in the ore to 85 to 95 percent in the concentrate.<sup>3</sup> The purification step removes the remaining uranium from the nonuraniferous diluents and produces a pure uranium compound.<sup>3</sup> Finally, the conversion step converts the purified uranium compound into the desired chemical form. Since processing raw uranium into a form useful as fuel removes the daughter products and other impurities, the assumption of zero daughter product and granddaughter product activity, used above, is justified.

Natural thorium is almost exclusively Th-232. The isotopic distribution is almost 100 atom % Th-232,  $1.35 \times 10^{-8}$  atom % Th-228, with small and variable quantities of Th-227, Th-239, Th-231, and Th-234.<sup>3</sup> Once thorium ore has been processed and purified into fuel, the isotopic content is mainly Th-232 with a small amount of Th-230 which is proportional to the amount of uranium present in the original ore. Once irradiated, Pa-231 is also present in thorium fuel. It is formed by an  $(n,2n)$  reaction in Th-232 and an  $(n,\gamma)$  in Th-230. However, the cross section for the  $(n,2n)$  reaction in Th-232 has an energy threshold of 6.37 MeV.<sup>4</sup> Hence, in a thermal reactor spectrum, very little Pa-231 will be generated by the  $(n,2n)$  reaction in Th-232. And, with a thermal cross section of 23 barns ( $1 \text{ barn} = 1 \times 10^{-24} \text{ cm}^2$ ) and low concentration of Th-230, very little Pa-231 will be generated by the  $(n,\gamma)$  reaction, as well. Hence, the assumption of zero daughter product and granddaughter product activity, used above, is justified.

To estimate the concentration of the above actinides requires knowledge of the parent nuclide concentration and the appropriate half-lives.<sup>5</sup>

**Table I**

**Half-Life of Selected Actinides and Parent Nuclides**

<b>Nuclide</b>	<b>Half-Life (yr)</b>	<b>Parent Nuclide (WCS)</b>	<b>Half-Life (yr)</b>
Ra-226	1599	U-234	$2.46 \times 10^5$
Ra-228	5.76	Th-232	$1.4 \times 10^{10}$
Ac-227	21.774	U-235	$7.04 \times 10^8$
Th-229	$7.3 \times 10^3$	U-233	$1.592 \times 10^5$
Th-230	$7.54 \times 10^4$	U-234	$2.46 \times 10^5$
Pa-231	$3.28 \times 10^4$	U-235	$7.04 \times 10^8$

In particular, the following decay chains are of interest:

U-234 → Th-230 → Ra-226,

Th-232 → Ra-228,

U-233 → Th-229,

U-235 → Pa-231 → Ac-227,

where the short-lived intermediate nuclide, Th-231 (half-life of 1.063 days), has been neglected in the U-235 decay chain. Note that these decay chains begin from a nuclide that has a known concentration and is currently tracked in WCS. Hence, using Eqs. (4) through (6) and data from WCS allows the concentration of the additional actinides to be calculated. In addition, note that since WCS evaluates time in units of years, to maintain that convention, time and half-lives in this report are also in units of years.

### ***U-234 → Th-230 → Ra-226***

Since U-234 is known from WCS, only Eqs. (5) and (6) are required. Note that Eq. (4) is already implemented in WCS so that the decay of U-234 is accounted for. Using data from Table I these equations reduce to

$$A_{\text{Th-230}}(t) = 1.442A_{\text{U-234}}(t) \left[ 1 - \left( \frac{1}{2} \right)^{9.1976 \times 10^{-6} t} \right], \quad (9)$$

$$A_{\text{Ra-226}}(t) = 1.4514A_{\text{U-234}}(t) \left[ 1 - 1.015 \left( \frac{1}{2} \right)^{9.1976 \times 10^{-6} t} + 0.015 \left( \frac{1}{2} \right)^{6.2133 \times 10^{-4} t} \right]. \quad (10)$$

As an example of the application and results of these equations, consider an initial activity of U-234 of 1 Ci, that is,  $A_{\text{U-234}}(t = 0) = 1$  Ci. Then, after 10,000 years, the resulting activity distribution is

$$A_{\text{U-234}} = 0.9722 \text{ Ci},$$

$$A_{\text{Th-230}} = 0.0866 \text{ Ci},$$

$$A_{\text{Ra-226}} = 0.0677 \text{ Ci}.$$

Assuming that transient equilibrium has been reached leads to the limiting activities of

$$A_{\text{Th-230}}(t) = 1.442A_{\text{U-234}}(t),$$

$$A_{\text{Ra-226}}(t) = 1.4514A_{\text{U-234}}(t).$$

### ***Th-232 → Ra-228***

Due to the very short half-life of Ra-228 compared to that of Th-232 and the age of SRS waste (from 15 to more than 50 years), it is clear that Ra-228 will quickly reach secular equilibrium with Th-232, that is the activity of Ra-228 and Th-232 will be equal. Hence, for simplicity it may be assumed that secular equilibrium already exists. In this case, Eq. (5) reduces to

$$A_{\text{Ra-228}}(t) = A_{\text{Th-232}}(t). \quad (11)$$

### ***U-233 → Th-229***

Since U-233 is known from WCS and the daughter product of Th-229 is not of interest, only Eq. (5) is required. Note that Eq. (4) is already implemented in WCS so that the decay of U-233 is accounted for. Using data from Table I, Eq. (5) reduces to

$$A_{\text{Th-229}}(t) = 1.0481A_{\text{U-233}}(t) \left[ 1 - \left( \frac{1}{2} \right)^{1.307 \times 10^{-4} t} \right]. \quad (12)$$

As an example of the application and results of these equations, consider an initial activity of U-233 of 1 Ci, that is,  $A_{U-233}(t = 0) = 1$  Ci. Then, after 10,000 years, the resulting activity distribution is

$$A_{U-233} = 0.9574 \text{ Ci},$$

$$A_{Th-229} = 0.5979 \text{ Ci}.$$

Assuming transient equilibrium has been reached leads to a limiting activity of

$$A_{Th-229}(t) = 1.0481A_{U-233}(t).$$

### **U-235 → Pa-231 → Ac-227**

Since U-235 is known from WCS, only Eqs. (5) and (6) are required. Note that Eq. (4) is already implemented in WCS so that the decay of U-235 is accounted for. Note, however, that due to the very long half-life of U-235 compared to Pa-231 and Ac-227, Eqs. (5) and (6) simplify to the following

$$A_2(t) = A_1(t) \left[ 1 - \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,2}}} \right],$$

$$A_3(t) = A_1(t) \left[ 1 - \frac{T_{1/2,2}}{\left( T_{1/2,2} - T_{1/2,3} \right)} \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,2}}} + \frac{T_{1/2,3}}{\left( T_{1/2,2} - T_{1/2,3} \right)} \left( \frac{1}{2} \right)^{\frac{t}{T_{1/2,3}}} \right].$$

Using data from Table I these equations reduce to

$$A_{Pa-231}(t) = A_{U-235}(t) \left[ 1 - \left( \frac{1}{2} \right)^{3.0488 \times 10^{-5} t} \right], \quad (13)$$

$$A_{Ac-227}(t) = A_{U-235}(t) \left[ 1 - 1.00066 \left( \frac{1}{2} \right)^{3.0488 \times 10^{-5} t} + 6.6 \times 10^{-4} \left( \frac{1}{2} \right)^{4.5926 \times 10^{-2} t} \right]. \quad (14)$$

As an example of the application and results of these equations, consider an initial activity of U-235 of 1 Ci, that is,  $A_{U-235}(t = 0) = 1$  Ci. Then, after 10,000 years, the activity distribution that is obtained is

$$A_{U-235} = 0.99999 \text{ Ci,}$$

$$A_{Pa-231} = 0.19049 \text{ Ci,}$$

$$A_{Ac-227} = 0.18995 \text{ Ci.}$$

Assuming secular equilibrium has been reached leads to the limiting activities of

$$A_{Pa-231}(t) = A_{U-235}(t),$$

$$A_{Ac-227}(t) = A_{U-235}(t).$$

Eqs. (9) through (14) may be implemented in WCS in either of two ways. First, the equilibrium values (transient or secular) may be used. This will place an upper bound on the estimate of the given actinide. Clearly this will provide conservative estimates. Alternatively, the equations, as given, may be implemented to directly calculate the actinide concentrations as functions of time. This will reduce the conservatism in the estimate of actinide concentrations.

## Conclusion

Using the equations of radioactive decay series it is possible to estimate the concentration of selected actinides not currently included in the HLW Waste Characterization System. In order to simplify the calculations, the equations for daughter and granddaughter products are written in terms of the appropriate parent nuclide that is known from WCS. Then, the equations for radioactive decay can be used to determine the time dependent concentration of decay products from the known concentration of parent nuclide.

Finally, although the assumption of transient or secular equilibrium would greatly simplify the calculation, due to the time scale over which equilibrium is obtained, this assumption may prove too conservative for current application. However, after evaluation, it may be determined that the degree of conservatism in the assumption of equilibrium is not too extreme. In that case, the calculations can be greatly simplified.

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