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Equations for Plutonium and Americium-241 Decay Corrections

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EQUATIONS FOR PLUTONIUM AND AMERICIUM-241 DECAY CORRECTIONS

by

T. E. Sampson and J. L. Parker

ABSTRACT

We present derivations of equations used in various plutonium accountability measurements to correct for plutonium decay and ^{241}Am ingrowth and decay. The equations are formulated in terms of the most widely available variables and are derived without approximation.

I. INTRODUCTION

In accounting for plutonium one often must correct for the decay of the plutonium isotopes and the ingrowth and decay of the ^{241}Am found in most plutonium.

These corrections involve the solution of a rather simple differential equation, a process taught in all elementary courses on differential equations. The solution to this equation is also found in many textbooks describing radioactive decay.¹

In many cases the solutions found in the literature are not in the best form to be easily used in plutonium accountability problems. For instance, the equations are often solved for the number of atoms, whereas plutonium accounting is done on a mass basis. Equations are often expressed in terms of absolute mass, whereas ^{241}Am is measured relative to plutonium. Also, the textbook solutions usually simplify the initial conditions so that the initial ^{241}Am content and the initial isotopic composition of the plutonium are given on the same day. This is seldom true in practice. Finally, the textbook solutions seldom show how the total plutonium mass varies with time.

It is true that the approximations that are made to obtain these solutions can be adequate for many applications. However, with the widespread availability of computers, there should be no reason for not solving the problem in an exact manner and expressing the solution in terms of useful, observable variables.

In this report we will derive and present solutions for the variation of the plutonium isotopic fractions as a function of time, the ratio of ^{241}Am to plutonium as a function of time, and the variation of the total plutonium mass as a function of time. The solutions will be presented in terms of mass and mass fractions.

It is assumed that three measurements characterize the plutonium sample: (1) its isotopic composition, (2) its americium content, and (3) its plutonium mass. All three measurements are assumed to occur at arbitrary and different times. Using these three initial measurements, the three parameters can then be calculated at any arbitrary time--past, present, or future. All time differences are expressed explicitly in terms of the measurement time or date of the three initial measurements.

To apply these solutions, the following points should be noted: First, "americium," in the context of these deviations, refers only to ^{241}Am . Some plutonium samples, notably those of higher burnup, may contain ^{243}Am . Whether "americium" content refers to total elemental americium or just to ^{241}Am depends on the specific analytical techniques. Also, in the equations ^{241}Am is expressed as a ratio to total plutonium. Analytical results for ^{241}Am are often expressed as ppm or a fraction of the sample mass rather than the plutonium mass. Finally, all plutonium isotopic measurements are expressed here in terms of mass fractions rather than atom fractions.

The user must be sure that the initial analytical results are properly interpreted before using them in the equations.

II. STATEMENT OF THE PROBLEM

Given values of the mass ratio $^{241}\text{Am}/\text{Pu}$ at a known time and values of the plutonium isotopic mass fractions at a known (and generally different) time, compute the $^{241}\text{Am}/\text{Pu}$ mass ratio at an arbitrary time t .

III. NOTATION

Subscripts refer to isotopes:

i = plutonium isotopes, generally $i = {}^{238}\text{Pu}, \dots, {}^{242}\text{Pu}$,

l = ${}^{241}\text{Pu}$, and

A = ${}^{241}\text{Am}$.

Superscripts refer to times (absolute) or dates.

I = values at time of plutonium isotopic determination,

a = values at time of ${}^{241}\text{Am}/\text{Pu}$ ratio determination,

m = values at time of plutonium mass determination,

t^I = time of plutonium isotopic determination,

t^a = time of ${}^{241}\text{Am}/\text{Pu}$ mass ratio determination,

t^m = time of plutonium mass determination,

t = arbitrary time at which new values are to be calculated,

m = total plutonium mass in sample,

m_i = mass of i^{th} plutonium isotope in sample,

$R_i = m_i/m$, mass fraction of i^{th} plutonium isotope relative to total plutonium,

T_i = half-life of i^{th} isotope,

$\lambda_i = \ln 2/T_i$, decay constant of i^{th} isotope,

N_i = number of atoms of isotope i , and

A_i = atomic mass of isotope i .

IV. DERIVATIONS

The amount of ^{241}Am in a plutonium sample arises from its formation in the decay of ^{241}Pu and its destruction from its own decay. The differential equation governing the number of atoms of ^{241}Am in a sample is

$$\frac{dN_A}{dt} = -\lambda_A N_A + K_1 \lambda_1 N_1, \quad (1)$$

where N_A = number of atoms of ^{241}Am ,

N_1 = number of atoms of ^{241}Pu ,

λ_A = decay constant of ^{241}Am ,

λ_1 = decay constant of ^{241}Pu ,

K_1 = 0.9999754, fraction of ^{241}Pu decays that lead to ^{241}Am , accounting for $2.46 \cdot 10^{-3}\%$ that decay to ^{237}U .

Assume a solution of the form

$$N_A(t) = B \exp(-\lambda_A t) + C \exp(-\lambda_1 t), \text{ and}$$

$$N_1(t) = N_1^a \exp[-\lambda_1 (t - t^a)],$$

with initial conditions that at $t = t^a$,

$$N_A(t^a) = N_A^a \quad \text{and} \quad N_1(t^a) = N_1^a ,$$

where t^a = time that $^{241}\text{Am}/\text{Pu}$ mass ratio was determined,

N_A^a = number of ^{241}Am atoms present at time t^a , and

N_1^a = number of ^{241}Pu atoms present at time t^a .

This gives a solution for the number of atoms of ^{241}Am at time t as

$$N_A(t) = N_A^a \exp[-\lambda_A(t - t^a)] + K_1 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) N_1^a \{ \exp[-\lambda_A(t - t^a)] - \exp[-\lambda_1(t - t^a)] \} . \quad (2)$$

To make this equation more useful, convert atoms to mass noting that

$$N = \frac{m \cdot N_0}{A} ,$$

where N = number of atoms,

m = mass in grams,

A = atomic mass (grams/mole),

$N_0 = 6.022045 \cdot 10^{23}$ atoms/mole,

and denoting

A_A = atomic mass of ^{241}Am , 241.056827,

A_1 = atomic mass of ^{241}Pu , 241.056850,

m_A = mass of ^{241}Am , and

m_1 = mass of ^{241}Pu .

This gives

$$\begin{aligned} \frac{m_A(t)}{A_A} &= \frac{m_A^a}{A_A} \exp[-\lambda_A(t - t^a)] \\ &+ K_1 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) \frac{m_1^a}{A_1} \{ \exp[-\lambda_A(t - t^a)] - \exp[-\lambda_1(t - t^a)] \} . \end{aligned}$$

Define $K_2 \equiv \frac{A_A}{A_1} = 0.999999905$,

thus $K_1 K_2 = 0.9999753$,

and we have for the mass of ^{241}Am

$$\begin{aligned} m_A(t) &= m_A^a \exp[-\lambda_A(t - t^a)] \\ &+ K_1 K_2 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) m_1^a \{ \exp[-\lambda_A(t - t^a)] \\ &- \exp[-\lambda_1(t - t^a)] \} . \end{aligned} \tag{3}$$

Equation (3) is expressed in terms of masses, whereas we desire the results to be expressed in terms of mass ratios to total plutonium because they are the measured observables.

So, let m = mass of plutonium in the sample at the time indicated by its superscript.

Define

$$R_A(t) = \frac{m_A(t)}{m(t)} ,$$

and note that the mass decays just like the number of atoms; that is

$$m_i(t) = m_i^a \exp[-\lambda_i(t - t^a)]$$

if there are m_i^a grams of isotope i present at time $t = t^a$.

The total plutonium mass is the sum of the masses of the individual plutonium isotopes, normally ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . Thus, the total plutonium mass present at time t in terms of the isotopic masses present at time t^a is

$$m(t) = \sum_i m_i(t) = \sum_i m_i^a \exp[-\lambda_i(t - t^a)] , \quad (4)$$

where i = all plutonium isotopes.

Divide Eq. (3) by Eq. (4) and, for compactness, denote the difference of the exponentials in Eq. (3) by $\{ \}$ in

$$R_A(t) = \frac{m_A^a \exp[-\lambda_A(t - t^a)] + K_1 K_2 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) m_1^a \{ \}}{\sum_i m_i^a \exp[-\lambda_i(t - t^a)]} \quad (5)$$

Divide the numerator and denominator by

$$m^a = \sum_i m_i^a$$

and define $R_i \equiv m_i/m$, the mass fractions relative to total plutonium.

We obtain

$$R_A(t) = \frac{R_A^a \exp[-\lambda_A(t - t^a)] + K_1 K_2 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) R_1^a \{ \}}{\sum_i R_i^a \exp[-\lambda_i(t - t^a)]} \quad (6)$$

In Eq. (6) we note the following:

R_A^a , generally known, is $^{241}\text{Am}/\text{Pu}$ at the time of the ^{241}Am concentration measurement.

R_1^a is not generally known. We do not assume that the plutonium isotopic fractions are measured at the same time as the americium.

R_i^a , likewise, is not generally known.

Thus, in Eq. (6) we need to obtain R_i^a in terms of R_i^I .

To do this, let us derive the expression for the mass fractions at time t in terms of those measured at time t^I :

m_i^I = isotopic masses at time t^I ,

$m^I = \sum_i m_i^I$, the total plutonium mass at t^I , and

$R_i^I = \frac{m_i^I}{\sum_i m_i^I}$, the plutonium mass fractions at time t^I .

Decay the isotopic masses from time t^I to time t , resulting in

$$m_i(t) = m_i^I \exp[-\lambda_i(t - t^I)] .$$

Then, mass fractions at time t are

$$R_i(t) = \frac{m_i(t)}{m(t)} = \frac{m_i^I \exp[-\lambda_i(t - t^I)]}{\sum_i m_i^I \exp[-\lambda_i(t - t^I)]} .$$

Dividing both numerator and denominator by m^I gives

$$R_i(t) = \frac{R_i^I \exp[-\lambda_i(t - t^I)]}{\sum_i R_i^I \exp[-\lambda_i(t - t^I)]} \quad (7)$$

where i = all plutonium isotopes.

This equation shows how the plutonium mass fractions change with time. The denominator is a normalization factor because all the fractions must sum to unity.

Now one can use the form of Eq. (7) to convert Eq. (6) to a form with observable parameters.

Substitute for R_i^a in the denominator of Eq. (6),

$$\sum_i R_i^a \exp[-\lambda_i(t - t^a)] = \frac{\sum_i R_i^I \exp[-\lambda_i(t^a - t^I)] \exp[-\lambda_i(t - t^a)]}{\sum_i R_i^I \exp[-\lambda_i(t^a - t^I)]}$$

which simplifies to

$$\frac{\sum_i R_i^I \exp[-\lambda_i(t - t^I)]}{\sum_i R_i^I \exp[-\lambda_i(t^a - t^I)]}.$$

Define

$$F(a) \equiv \sum_i R_i^I \exp[-\lambda_i(t^a - t^I)],$$

and likewise

$$F(t) \equiv \sum_i R_i^I \exp[-\lambda_i(t - t^I)].$$

Now Eq. (6) becomes

$$R_A(t) = \frac{R_A^a \exp[-\lambda_A(t - t^a)] + K_1 K_2 \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) K_1^a \{ \}}{F(t)/F(a)}.$$

Now use Eq. (7) again to put R_1^a in terms of R_1^I so that

$$R_1^a = \frac{R_1^I \exp[-\lambda_1(t^a - t^I)]}{\sum_i R_i^I \exp[-\lambda_i(t^a - t^I)]} = \frac{R_1^I \exp[-\lambda_1(t^a - t^I)]}{F(a)} ,$$

giving

$$R_A(t) = \frac{F(a)}{F(t)} R_A^a \exp[-\lambda_A(t - t^a)] \\ + \frac{K_1 K_2}{F(t)} \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) R_1^I \exp[-\lambda_1(t^a - t^I)] \{ \} .$$

Let us summarize this result in Eq. (8).

$$R_A(t) = \frac{F(a)}{F(t)} R_A^a \exp[-\lambda_A(t - t^a)] \\ + \frac{K_1 K_2}{F(t)} \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) R_1^I \exp[-\lambda_1(t^a - t^I)] \\ \cdot \{ \exp[-\lambda_A(t - t^a)] - \exp[-\lambda_1(t - t^a)] \} , \quad (8)$$

where all variables are observables

t = time at which $^{241}\text{Am}/\text{Pu}$ ratio is desired,

t^a = time of known $^{241}\text{Am}/\text{Pu}$ ratio,

t^I = time of known plutonium isotopic values,

λ_A = decay constant of ^{241}Am ,

λ_1 = decay constant of ^{241}Pu ,

λ_i = decay constant of plutonium isotope i , $i = ^{238}\text{Pu}, \dots, ^{242}\text{Pu}$,

R_A = mass fraction of ^{241}Am relative to plutonium, or $^{241}\text{Am}/\text{Pu}$,

R_A^a = mass fraction of ^{241}Am relative to plutonium, or $^{241}\text{Am}/\text{Pu}$ at time t^a ,

R_1^I = ^{241}Pu mass fraction at time t^I ,

$K_1 = 0.9999754$, the ^{241}Pu branching ratio to ^{241}Am ,

$K_2 = 0.999999905$, the ratio of atomic masses $^{241}\text{Am}/^{241}\text{Pu}$,

R_i^I = mass fraction of plutonium isotope i at time t^I ,

$$F(a) = \sum_i R_i^I \exp[-\lambda_i (t^a - t^I)] \quad , \text{ and}$$

$$F(t) = \sum_i R_i^I \exp[-\lambda_i (t - t^I)] \quad .$$

The last item to be derived is defined by the following problem:

Given that the mass of plutonium in a sample is known at time t^m , what is the mass at an arbitrary time t where we assume that we know the isotopic composition at time t^I ?

The mass of each isotope decays as

$$m_i(t) = m_i^m \exp[-\lambda_i (t - t^m)] \quad .$$

The total mass is

$$m(t) = \sum_i m_i(t) = \sum_i m_i^m \exp[-\lambda_i(t - t^m)] \quad .$$

Multiply and divide the right hand side by m^m and note $m_i^m/m^m = R_i^m$ so that

$$m(t) = m^m \sum_i R_i^m \exp[-\lambda_i(t - t^m)] \quad .$$

From Eq. (7)

$$R_i(t^m) = R_i^m = \frac{R_i^I \exp[-\lambda_i(t^m - t^I)]}{\sum_i R_i^I \exp[-\lambda_i(t^m - t^I)]} \quad .$$

Thus,

$$m(t) = m^m \frac{\sum_i R_i^I \exp[-\lambda_i(t^m - t^I)] \exp[-\lambda_i(t - t^m)]}{\sum_i R_i^I \exp[-\lambda_i(t^m - t^I)]}$$

giving

$$m(t) = \frac{m^m \sum_i R_i^I \exp[-\lambda_i(t - t^I)]}{F(m)} \quad , \quad (9)$$

where

$$F(m) = \sum_i R_i^I \exp[-\lambda_i(t^m - t^I)] \quad .$$

V. SUMMARY

We have derived expressions for three time-dependent quantities used in plutonium accountability calculations:

1. The change in the isotopic composition as a function of time,
2. The $^{241}\text{Am}/\text{Pu}$ ratio as a function of time, and
3. The plutonium mass as a function of time.

All three parameters are expressed in terms of the variables that are most commonly available to the user, the time of an initial plutonium isotopic composition measurement, the time of an $^{241}\text{Am}/\text{Pu}$ ratio measurement, and the time of a sample plutonium mass measurement. All these times may be different. Because mass is used in accountability calculations, all ratios are expressed on a mass (not atom) basis.

It is important that the user assure that the input to these equations is in the proper units.

The results are summarized below.

Plutonium mass fraction at time t in terms of those measured at time t^I , from Eq. (7), is

$$R_i(t) = \frac{R_i^I \exp[-\lambda_i(t - t^I)]}{F(t)} \quad .$$

The ^{241}Am mass fraction relative to plutonium at time t , from Eq. (8), is

$$\begin{aligned}
R_A(t) = & \frac{F(a)}{F(t)} R_A^a \exp[-\lambda_A(t - t^a)] \\
& + \frac{K_1 K_2}{F(t)} \left(\frac{\lambda_1}{\lambda_1 - \lambda_A} \right) R_1^I \exp[-\lambda_1(t^a - t^I)] \\
& \cdot \{ \exp[-\lambda_A(t - t^a)] - \exp[-\lambda_1(t - t^a)] \} / \quad .
\end{aligned}$$

The sample plutonium mass at time t in terms of its known value at time t^m , from Eq. (9), is

$$m(t) = \frac{\sum_i^m R_i^I \exp[-\lambda_i(t - t^I)]}{F(m)} \quad .$$

Also summarizing the notation are the definitions

- t = arbitrary time or date at which new values are to be calculated,
- t^I = time or date of initial plutonium isotopic composition determination,
- t^a = time or date of initial $^{241}\text{Am}/\text{Pu}$ mass ratio determination,
- t^m = time or date of initial plutonium sample mass determination,
- m = plutonium mass in sample,
- m_i = mass of i^{th} isotope in sample,
- R_i = m_i/m , or mass fraction of i^{th} isotope relative to plutonium,
- R_i^I = mass fraction of plutonium isotope i at time t^I ,

R_A = mass fraction of ^{241}Am relative to plutonium,

R_A^a = mass fraction of ^{241}Am relative to plutonium at time t^a ,

T_i = half-life of i^{th} isotope,

λ_i = $\ln 2/T_i$, or decay constant of i^{th} isotope,

λ_A = decay constant of ^{241}Am ,

λ_1 = decay constant of ^{241}Pu ,

K_1 = 0.9999754, ^{241}Pu branching ratio to ^{241}Am ,

K_2 = 0.999999905, atomic mass ratio for $^{241}\text{Am}/^{241}\text{Pu}$,

$$F(t) = \sum_i R_i^I \exp[-\lambda_i(t - t^I)] \quad , \text{ in which } i = \text{plutonium isotopes},$$

$$F(a) = \sum_i R_i^I \exp[-\lambda_i(t^a - t^I)] \quad , \text{ in which } i = \text{plutonium isotopes, and}$$

$$F(m) = \sum_i R_i^I \exp[-\lambda_i(t^m - t^I)] \quad , \text{ in which } i = \text{plutonium isotopes}.$$

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1. Robley D. Evans, The Atomic Nucleus (McGraw-Hill Book Company, Inc., New York, 1955), pp. 470-510.