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# THE GENERAL'S TANKS CHARACTERIZATION ACTIVITIES DOCUMENTED SAFETY ANALYSIS

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## D.1 INTRODUCTION

This Appendix describes the basis and methods to calculate inventories of transuranic radionuclides in the General's Tanks located at TA-21 MDA A. Calculated inventories may be compared to the category 2 nuclear facility thresholds described in DOE-STD-1027-92, *Hazard Categorization and Accident Analysis Techniques for compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports* (DOE, 1992). Similarly, the inventory estimates may be used as the nuclear material at risk (MAR), subject to disturbance and dispersal associated with postulated accident scenarios. The General's Tanks were pumped and sampled several times in the 1970's and early 1980's, and analytical results are available for these events. Similarly, personal interviews have been collected with personnel performing the pumping and sampling in 1981 (Appendix C).

Calculations are performed using basic concepts and equations relating to radioactive decay and ingrowth from the time of the last sampling event, dimensional analysis based on available engineering drawings describing tank features, and reports regarding sludge levels at the last known sampling period. Key assumptions and conclusions related to the following analyses include:

- The use of sampling data associated with the most recent pumping event in 1981 provides the most conservative and current data set,
- No additional nuclear material has been added to the tanks for at least 30 years and all of this was prior to referenced pumping events,
- Analyses using 1970's memoranda describing Pu-239 inventory descriptions yield calculated nuclear material inventories far below that using tank dimensional analysis and sampling data,
- The Pu-241 to Pu-239 mass ratio is assumed to be 0.0025 (0.25%) – no Pu-241 data are available,
- Based on worker interviews and tank design, average depth of sludge/sediment in the tank is 0.458 ft, and
- Individually, neither tank exceeds the category 2 nuclear material threshold. Collectively, the nuclear material inventories exceed the category 2 threshold.

Supporting information regarding these assumptions and conclusions is provided in the following sections of this Appendix.

## D.2 HISTORICAL SAMPLING DATA AND MEMORANDA

The liquids and solids in the General's Tanks were sampled on at least four occasions between 1973 and 1981. Summary data from the sampling events are provided in Table D-1. In some instances reporting units were not legible, thus are not entered in the table.

**Table D-1. Analytical Data for the General's Tanks as Extracted from Original Sampling Reports.**

| Reference | Description of Episode | Am-241 | Pu-238 | Pu-239 |
|---|---|---|---|---|
| (1) | West Tank Sediment | 9.2E5 dpm/g | 2.2E5 dpm/g | 3.9E7 dpm/g |
| (1) | East Tank Sediment | 2.4E5 dpm/g | 6.5E4 dpm/g | 1E7 dpm/g |
| (2) | Sluge (Sludge?) 75415 | 1.6E8 dpm/ml | 1.71E8 dpm/ml | 4.82E9 dpm/ml |
| (2) | 75415 (#2) | <5,000 dpm/ml | 1.62E+05 | 1.46E6 dpm/ml |
| (2) | 75415 (#1 DPE Tank) | 5,000 dpm/ml | 1.80E+05 | 1.69E6 dpm/ml |
| (3) | 75304 | 12,000 dpm/ml | 1.62E+05 | 1.7E6 dpm/ml |
| (4) | Liquid 76253 | 6.9E3 dpm/ml | Not given | 7.68E4 dpm/ml |

(1) Bucholz; 1/27/1981; analysis # 81064

(2) Bucholz; 8/22/1975; analysis # 75415

(3) McGinnis; 6/23/1975; analysis #75304

(4) Valdez; 5/21/1976; analysis #76253

Of these information sources available, the most useful are believed to be the data from the January 1981 sampling episode by J. Bucholz. The Bucholz (1981) analytical results are assumed to be the most representative because they are the most recent, and they are coupled with interview results regarding the remaining sludge levels in 1981. It is also designated as "sediment", indicating that the majority of the liquid phase had been pumped from the tanks. The calculations provided herein use the analytical data from Bucholz (1981) and estimates of the average sludge thickness to obtain the total tank inventory estimates.

The original inventory placed in the tanks may also be estimated from information contained in several other historical memoranda. These memoranda are described briefly below:

> (Gibson 1971). This is a memorandum from W.B.Gibson to W.J. Maraman dated December 6, 1971. It appears to indicate that the amount of alpha activity added to the tanks was equivalent to a mass of 340 g of Pu 239. This memo indicates that the gross alpha equivalent of 180 grams Pu-239 was present in the west tank and 160 grams was present in the east tank. These quantities are not used in subsequent calculations because they do not yield the highest possible inventory estimates.
>
> (McGinnis 1976). This is a memorandum from P.E.McGinnis to Margaret Anne Rogers dated March 12, 1976. It provides insight into the meaning of "total equivalent Pu-239 mass." In section II of the memo, it indicates that values reported as "total equivalent Pu-239 mass" includes the activity contributions from

other alpha emitters. Section IV of this memo indicates that the total equivalent Pu-239 mass was based on gross alpha activity.

Based on this information, it can be concluded that the initial mass of Pu-239 introduced to the tanks was ___ g. This amount of Pu-239 is substantially lower than that calculated using sample results and dimensional analysis as presented in this appendix. Reasons for this discrepancy might include incorrect initial assay as described in the historical memoranda, additional material added to the tank that was not accounted for, or a much lower sludge/sediment level exists in the tank than 1981 observations indicated. Nevertheless, this reference point provides valuable confirmation that the calculations performed in this document are likely quite conservative, and gives some qualitative benchmark for the uncertainty in the analysis. This relationship is described further in the conclusion to this appendix.

## D.3 COMPUTATIONAL METHODS

Several calculation sequences were applied to develop and analyze the nuclear material inventory. In most instances, the Mathematica™ software was used to process equations after a set of input variables was described. Equations were obtained from a variety of reference sources and modified as necessary to support the calculation process.

### D.3.1 Input Parameters

Table D-2 presents a listing and descriptions of each Mathematica term, its associated value and reference source. These terms are the building blocks of equations describing ingrowth and decay, and calculations using tank dimensions and sludge levels.

The mass ratio of Pu-241 to Pu-239, "MassRatioPu241toPu239" is estimated. Pu-241 is of interest because it is fissile and its decay results in ingrowth of Am-241 in the tank inventory. It is clear that the ratio must be very small since, as of 1981, only a small amount of Am-241 was present in the tank even though the available information suggests that no plutonium has been added to the tank in nearly 30 years. The assumed mass ratio value of 0.0025 should overestimate the inventory of both Am-241 and Pu-241 in the tanks.

**Table D-2. Half-lives and Other Parameter Values**

| Term in Mathematica™ Equations | Meaning | Value | Source |
|---|---|---|---|
| HalfLifePu238 | Half-life of Pu-238 | 87.75 years | Radiation Decay software package, version 3.5, distributed by Charles Hacker, October 25, 2000 |
| HalfLifePu239 | Half-life of Pu-239 | 24,131 years | |
| HalfLifePu241 | Half-life of Pu-241 | 14.5 years | |
| HalfLifeAm241 | Half-life of Am-241 | 87.75 years | |
| ActivityFactor | Activity conversion factor | Curie/$2.22 \times 10^{12}$ dpm | -- |
| SpecificAcivityPu239 | Specific Activity of Pu-239 | 0.0617557 Curie/gram | The Health Physics and Radiological Health Handbook, page 170 (Shleien 1984)[1] |
| MassRatioPu241toPu239 | Assumed ratio of Pu-241 to Pu-239 during in 1981 | 0.0025 | A high, but conservative estimate for 1940s vintage plutonium that has aged for more than 30 years. Professional judgment. |
| TankLength | Length of Tank | 62 feet | Engineering drawings |
| Radius | Radius of Tank | 6 feet | Engineering drawings |
| SludgeDensity | Density of Sludge | 1 gram/cm$^3$ | Engineering Estimate |
| SludgeThickness | Thickness of Sludge | 0.455 feet | Interviews with sampling personnel |

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[1] $$\text{SpecificAcivityPu239} = \frac{1.3 \times 10^8}{\text{HalfLifePu239} \times 365 \times 239}$$

It should be noted that the sludge thickness was developed based on the sampling personnel observations of 3-4 inches (0.33 ft), increased by 1.5 inches (0.125 ft) to account for the end to end slope of the tanks (3 inches total).

### D.3.2 Solution of Ingrowth and Decay Equations for Target Radionuclides

The following equations relate the decay of Pu-241 and the ingrowth of Am-241 so that the activity can be corrected from 1981 to 2003. Pu-241 has the following decay scheme:

$$Pu241 \rightarrow Am241 \rightarrow \text{long-lived Np-237}$$

The Pu-241 and Am-241 equations have the following basis:

Let "pu(t)" and "am(t)" be the number of atoms of Pu-241 and Am-241as a function of time. Then pu'(t) and am'(t) represent the rate of change in the number of atoms present as a function of time. Then, Equations (D-1) and (D-2) provide the numbers of atoms present as a function of time:

Equation (D-1):

$$pu'(t) = -\,(0.693/HalfLifePu241) * pu(t),$$

Equation (D-2):

$$am'(t) = (0.693/HalfLifePu241) * pu(t) - ((0.693/HalfLifeAm241) * am(t))$$

Multiply both sides of equation D-1 by "0.693/HalfLifePu241" to obtain it in terms of activity, yielding equation (D-3).

Equation (D-3):

$$Pu241activity\,'(t) = -\,(0.693/HalfLifePu241) * Pu241activity(t)$$

In equation (D-2), replace "(0.693/HalfLifePu241) * pu(t)" with "Pu241activity(t)" and replace "(0.693/HalfLifeAm241) * am(t))" with "Am241activity(t)", then multiply both sides of equation (D-2) by "(0.693/HalfLifeAm241)" to obtain equation (D-4):

Equation (D-4):

$$Am241activity\,'(t) = (0.693/HalfLifeAm241) *( Pu241activity(t) - Am241activity(t))$$

Equations (D-3) and (D-4) are solved using Mathematica 5.0 (Wolfram, 2003) for generalized boundary conditions that correspond to the 1981 inventory estimates. The equations that describe the amounts of Pu-238, Pu-239, Pu-241 and Am-241 activity as functions of the 1981 inventory and time are given by equations (D-5) through (D-8):

Equation (D-5):

$$Pu238activityTime[Pu238Act, t] = Pu238Act \times e^{\frac{-t \times 0.693}{HalfLifePu238}}$$

Equation (D-6):

$$Pu239activityTime[Pu239Act, t] = Pu239Act \times e^{\frac{-t \times 0.693}{HalfLifePu239}}$$

Equation (D-7):

$$Pu241activityTime[Pu241Act, t] = Pu241Act \times e^{-0.0477931 \times t}$$

Equation (D-8):

$$Am241activityTime[Am241Act, Pu241Act, t] = Am241Act \times e^{-0.001604166 \times t}$$

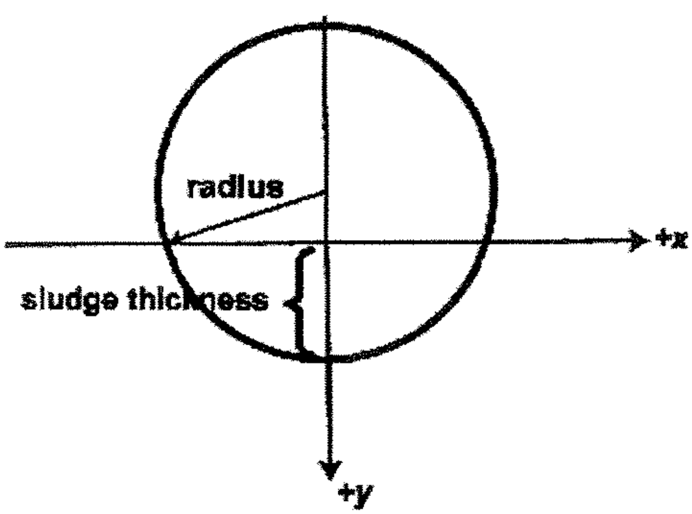
$$+ \; 0.0347305 \times Pu241Act \times \left(e^{-0.00160416 \times t} - e^{-0.04779310 \times t}\right)$$

In Equations D-5 through D-8, "t" represents the time in years with the convention that 1981 is year 0. Pu238Act, Pu239Act, Pu241Act and Am241Act are the 1981 inventories.

### D.3.3 Sludge Volume and Mass

The following calculations derive the cross sectional area of the sludge and the sludge volume as Length x Cross Sectional Area. Parameter inputs have been identified in Table D-2. Figure D-1 is a cross sectional view of one of the General's Tanks.

**Figure D-1. Cross sectional view of one of the General's Tanks**




Equation (D-9) is used to calculate the total tank volume of 1.98559 x$10^8$ cm3.

Equation (D-9):

$$\text{TotalTankVolume} = \text{radius}^2 \times \text{TankLength} \times \text{Pi} \times \left(\frac{2.54\ \text{cm} \times 12\ \text{inch}}{\text{inch}}\right)^3$$

Equation (D-10) is the equation for a circle of radius "radius" centered at x,y coordinates (0, sludge thickness - radius), which has been solved for Y.

Equation (D-10):

$$Y = \sqrt{\text{radius}^2 - x^2} - a\ ,$$

where "a" equals "radius - SudgeThickness". Equation (D-10) yields the limits of integration that are subsequently used in Equation (D-11). To obtain them, Y is set to zero and Equation (D-10) is solved for x.

Mathematica 5.0 is used to solve Equation (D-11) to obtain the cross sectional area of the sludge, 1.40134 $ft^2$. [2]

Equation (D-11):

$$\text{SludgeCrossSectionalArea} = \int_{-(\text{radius}\wedge 2 - a\wedge 2)\wedge 0.5}^{+(\text{radius}\wedge 2 - a\wedge 2)\wedge 0.5} Y\, dx$$

Equation (D-12) is used to calculate the sludge volume as $2.46026 \times 10^6$ $cm^3$. At a density of 1, the estimated sludge mass in a tank is $2.46026 \times 10^6$ $cm^3$:

Equation (D-12):

$$\text{SludgeVolume} = \text{TankLength} \times \text{SludgeCrossSectionalArea} \times (2.54 \times 12)^3$$

**D.3.4 Hazard Category Determination**

Of particular interest is whether the tanks are likely to contain more than the category 2 nuclear facility threshold of Pu and Am. The category 2 threshold is exceeded if the following condition is met:

Equation (D-13)

$$\frac{Pu238}{62\,Ci} + \frac{Pu239}{56\,Ci} + \frac{Pu240}{55\,Ci} + \frac{Pu241}{2900\,Ci} + \frac{Am241}{55\,Ci} > 1$$

Sections D.3.5 and D.3.6 present methods to determine the total quantity of these radionuclides in the East and West Tanks. Equation (D-13) is then applied to these quantities to determine each tanks fraction of the category 2 threshold.

### D.3.5 Inventory Estimates for East Tank

Base parameters for Mathematica calculations have been presented in Table D-2. Several Mathematica radionuclide ratio parameters specific to the inventory of the East Tank are developed, including:

Pu238toPu239ActivityRatio - Ratio of Pu-238 to Pu239 on an activity basis = 0.0065

AmToPu239activityRatio - Ratio of Am-241 to Pu-239 on an activity basis = 0.024

Both of these ratios are derived from sample results (#81064) presented in Bucholz (1981).

Equation (D-14) provides the Pu-239 concentration, in activity units, $4.5045\times10^{-6}$ Curie/gram for reference year 1981:

Equation (D-14):

$$\mathbf{Pu239conc} = 1.0\times 10^{7}\ \frac{\mathbf{dpm}}{\mathbf{gram}} \times \mathbf{ActivityFactor}$$

Equation (D-15) is used to calculate the Pu-239 inventory for reference year 1981, 11.0822 Curie.

Equation (D-15):

$$\text{Pu239Act}=\text{Pu239conc} \times \text{SludgeMass}$$

Equation (D-16) is used to calculate the Am-241 inventory for reference year 1981, 0.265974 Curie.

Equation (D-16):

$$\text{Am241Act}=\text{AmToPu239activityRatio} \times \text{Pu239Act}$$

Equation (D-17) is used to calculate the Pu-241 inventory for reference year 1981, 46.1078 Curie.

[2] This same result can obtained as the difference in the areas of a "pie slice " and the isosceles triangle that results from replacing the arc with a line segment.

Equation (D-17):

$$Pu241Act = \frac{MassRatioPu241toPu239 \times HalfLifePu239 \times Pu239Act}{HalfLifePu241}$$

Equation (D-18) is used to calculate the Pu-238 inventory for reference year 1981, 0.0720345 Curie.

Equation (D-18):

$$Pu238Act = Pu238toPu239ActivityRatio \times Pu239Act$$

The 1981 inventory values for the East Tank are corrected for ingrowth using Equations (D-5) through (D-8) and presented in Table D-3 below. Calculations are performed for theoretical radionuclide inventory in the years 1992, 2003 and 2014. Individual radionuclide inventories are then analyzed with Equation (D-13) to determine the category 2 fraction.

**Table D-3. Inventory estimates for the East Tank**

| Year | Pu-241 (Ci) | Am-241 (Ci) | Pu-238 (Ci) | Pu-239 (Ci) | Category 2 Fraction |
|---|---|---|---|---|---|
| 1981 | 46.1 | 0.266 | 0.0720 | 11.0822 | 0.220 |
| 1992 | 27.3 | 0.888 | 0.0660 | 11.0787 | 0.224 |
| 2003 | 16.1 | 1.24 | 0.0605 | 11.0752 | 0.227 |
| 2014 | 9.5 | 1.44 | 0.0555 | 11.0717 | 0.228 |

### D.3.6 Inventory Estimates for West Tank

Base parameters for Mathematica calculations have been presented in Table D-2. Several Mathematica radionuclide ratio parameters specific to the inventory of the East Tank are developed, including:

Pu238toPu239ActivityRatio = Ratio of Pu-238 to Pu239 on an activity basis = 0.00564

AmToPu239activityRatio = Ratio of Am-241 to Pu-239 on an activity basis = 0.024

Both of these ratios are derived from sample results (#81064) presented in Bucholz (1981).

Equation (D-19) calculates the Pu-239 concentration, in activity units, $1.75676 \times 10^{-5}$ Curie/gram for reference year 1981:

Equation (D-19):

$$Pu239conc = 3.9 \times 10^{7} \times ActivityFactor$$

Equation (D-20) calculates the Pu-239 inventory for reference year 1981 as 43.2207 Ci.

Equation (D-20):

$$\text{Pu239Act} = \text{Pu239conc} \times \text{SludgeMass}$$

Equation (D-21) calculates the Am-241 inventory for reference year 1981, 1.01957 Ci.

Equation (D-21):

$$\text{Am241Act} = \text{AmToPu239activityRatio} \times \text{Pu239Act}$$

Equation (D-22) calculates the Pu-241 inventory for reference year 1981, 179.821 Ci.

Equation (D-22):

$$\text{Pu241Act} = \frac{\text{MassRatioPu241toPu239} \times \text{HalfLifePu239} \times \text{Pu239Act}}{\text{HalfLifePu241}}$$

Equation (D-23) provides an estimate of the Pu-238 inventory for reference year 1981, 0.243809 Ci.

Equation (D-23):

$$\text{Pu238Act} = \text{Pu238toPu239ActivityRatio} \times \text{Pu239Act}$$

The 1981 inventory values for the West Tank are corrected for ingrowth using Equations (D-5) through (D-8) and presented in Table D-4. Individual radionuclide inventories are then analyzed with Equation (D-13) to determine the category 2 fraction.

**Table D-4. Inventory Estimates for the West Tank.**

| Year | Pu-241 (Ci) | Am-241 (Ci) | Pu-238 (Ci) | Pu-239 (Ci) | Category 2 Fraction |
|---|---|---|---|---|---|
| 1981 | 179.8 | 1.02 | 0.243 | 43.22 | 0.856 |
| 1992 | 106.3 | 3.45 | 0.224 | 43.21 | 0.874 |
| 2003 | 62.8 | 4.83 | 0.205 | 43.19 | 0.884 |
| 2014 | 37.1 | 5.60 | 0.188 | 43.18 | 0.889 |

### D.3.6 Fissile Material Estimates for the East and West Tanks

Fissile material inventory (I) in grams is determined through the following relationship:

Equation (D-24):

I (g) = Activity of fissile radionuclide (Ci) / specific activity of radionuclide (Ci/g)

Table D-5 presents the fissile material mass for the East and West Tanks as calculated for the year 2003. The radionuclides Pu-239 and U-235 are included. It should be noted that isotopic uranium results are unavailable in Bucholz (1981), and it is assumed that U-235 is present at 5% enrichment. Details regarding the calculation of U-235 activities are presented in Appendix E.

**Table D-5. Fissile Nuclear Material Mass Estimates (2003).**

| East Tank | | | | West Tank | | | |
|---|---|---|---|---|---|---|---|
| Isotope | Activity (Ci) | Specific Activity (Ci/g) | Mass (g) | Isotope | Activity (Ci) | Specific Activity (Ci/g) | Mass (g) |
| Pu-239 | 11.07 | 0.0618 | 179 | Pu-239 | 43.19 | 0.0618 | 699 |
| U-235 | 0.0003 | 2.17E-6 | 139 | U-235 | 0.0011 | 2.17E-6 | 511 |

## D.4 SUMMARY OF RESULTS

Based on the results of this evaluation, the Category 2 threshold may be exceeded when the tank inventories are combined. Subsequently, the tanks as a combined unit are a category 2 nuclear facility using DOE-STD-1027-92 "Hazard Categorization and Accident Analysis Techniques for compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports." The west tank has a higher inventory than the east tank. Bounding accident analyses should be based on the west tank's inventory for initiating events other than deflagration, which will conservatively combine the contents of both tanks for accident scenario analyses.

## D.5 REFERENCES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