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Nondestructive Measurements in Support of Waste Isolation Pilot Plant at Rockwell Hanford Operations: Problems and Methods

May 1986

George A. Westsik

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Prepared for the U.S. Department of Energy
under Contract DE-AC06-77RL01030

Rockwell International

Rockwell Hanford Operations
Richland, Washington

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Safety and Quality Assurance

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ABSTRACT

The 234-5Z Analytical Laboratory, located in the 200 West Area of the Hanford Site, has been requested to provide waste package measurement capability for both the Plutonium/Uranium Extraction (PUREX) Plant and Plutonium Finishing Plant (PFP) in support of the Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (WAC). The requested WIPP values are sensitive to changes in isotopic distribution. Unfortunately, the determination of the isotopic distribution of an individual waste item or drum is difficult. The problems and uncertainties encountered in providing the values are discussed. Also, examples of calculations for the WIPP-WAC are shown.

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INTRODUCTION

The 234-5Z Analytical Laboratory, located in the 200 West area of the Hanford Site, has been requested to provide waste package measurement capability for both the Plutonium/Uranium Extraction (PUREX) Plant and Plutonium Finishing Plant (PFP) in support of Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (WAC). The requested WIPP values are sensitive to changes in isotopic distribution. Unfortunately, the determination of the isotopic distribution of an individual waste item or drum is difficult. The problems and uncertainties encountered in providing the values are discussed. Also, examples of calculations for the WIPP-WAC are shown.

Several scenarios encountered recently that can affect the WIPP values are listed below. The problems are more pronounced at PFP, but similar problems have the potential of becoming significant at PUREX.

The major obstacle to providing accurate values is the uncertainty in the isotopic distribution of the item being assayed. This uncertainty arises from several sources.

- a. The segmented gamma scan assay system (SGSAS) measures ^{239}Pu only and not the isotopes the WIPP values are most sensitive to.
- b. Several material balance areas (MBAs) generate waste within a processing plant. Waste from one MBA will have a slightly different isotopic distribution than that from another MBA. Sometimes this occurs in the same MBA.
- c. The ^{241}Am content is established as of a certain date. However, there is the ingrowth of ^{241}Am over time. Waste generated a long time after the definition date will potentially have a much greater amount of ^{241}Am . The difference in the WIPP value calculated, based on the definition date, versus the value calculated, based on the true ^{241}Am content, could significantly affect the required calculations.
- d. At the start of a new campaign the isotopic distributions of the material to be processed can change. The waste from the previous campaign may become mixed with waste from the new campaign. An extension of this is the possibility that waste from several years ago is mixed with waste from a recent campaign.

The sources of uncertainty are discussed in the following sections with examples of how the requested values are affected. Each will be treated separately but can occur simultaneously. The isotopic values used are from the Nondestructive Assay Isotopic System (NDAIS) measurements of feed and product items. There is no way to determine which of the scenarios will predominate. Due to characteristics of each processing area there will be a different place where a lump or pile of plutonium will hide until an exhaustive cleanout is performed.

SOURCES OF UNCERTAINTY

THE SEGMENTED GAMMA SCAN/ASSAY SYSTEM DOES NOT DETERMINE ISOTOPICS

In areas where plutonium-bearing items are likely to have differing isotopic distributions, the fact that two items have the same relative ^{239}Pu value (weight percent (wt%)) does not ensure the masses of the other isotopes will be the same. This is important in areas like Miscellaneous Treatment (MT) at PFP and the area where feed is entered into N Cell at PUREX. The isotopic distribution of the feed stock prior to blending is not consistent.

As an example, take the case where 2% and 11% ^{240}Pu material are blended to yield 6% ^{240}Pu product as shown. The isotopic distribution used by the SGSAS to calculate the per gram WIPP values would be significantly different from the actual isotopic distributions. If the cans and plastic waste from the one isotopic group are disposed of at a different time from the waste of the second isotopic group, two or more waste drums would have large errors in the calculated WIPP values. One could not calculate the WIPP-required values based on an assumed stream average in this scenario.

Isotopic distribution (wt%)	A	B	Equal mass mixture of A&B
^{238}Pu	0.005	0.082	0.044
^{239}Pu	97.890	87.240	92.565
^{240}Pu	2.065	11.440	6.752
^{241}Pu	0.026	1.187	0.606
^{242}Pu	0.013	0.051	0.032
^{241}Am	0.506	1.439	0.972

WIPP values for waste drum	A	B	Equal mass mixture of A&B
Ci/g	1.11 E-01	1.37 E + 00	7.40 E-01
alpha-Ci/g	8.36 E-02	1.43 E-01	1.14 E-01
^a PE-Ci/g	7.26 E-02	1.38 E-01	1.05 E-01
^b FGE/g	9.80 E-01	9.02 E-01	9.41 E-01
watts	2.60 E-03	4.60 E-03	3.60 E-03

^aPlutonium equivalent (transuranic activity).

^bFissile gram equivalent.

Although this is an extreme example, it is still credible and has probably happened. Another less extreme example is one where the items originate from the same site but have slightly different isotopes as shown.

Isotope (wt%)	A	B	Equal mass mixture of A&B
²³⁸Pu	0.012	0.013	0.012
²³⁹Pu	93.310	93.580	93.445
²⁴⁰Pu	6.397	6.089	6.243
²⁴¹Pu	0.244	0.293	0.268
²⁴²Pu	0.036	0.026	0.031
²⁴¹Am	0.327	0.017	0.172

WIPP values for waste drum	A	B	Equal mass mixture of A&B
Ci/g	3.38 E-01	3.77 E-01	3.57 E-01
alpha-Ci/g	8.57 E-02	7.46 E-02	8.02 E-02
PE-Ci/g	8.38 E-02	8.09E-02	8.24 E-02
FGE/g	9.40 E-01	9.44 E-01	9.42 E-01
watts	2.70 E-03	2.30 E-03	2.50 E-03

MATERIAL BALANCE AREAS WITHIN A PROCESSING PLANT WITH DIFFERING MATERIAL ISOTOPES

The isotopic distribution may be different for waste generated in the differing MBAs or even potentially in the same MBA. As an example, recently at PFP one MBA was processing freshly

Isotope (wt%)	A	B
²³⁸Pu	0.032	0.082
²³⁹Pu	92.770	87.240
²⁴⁰Pu	6.366	11.440
²⁴¹Pu	0.800	1.187
²⁴²Pu	0.031	0.051
²⁴¹Am	0.006	1.439

WIPP values for waste drum	A	B
Ci/g	9.04 E-01	1.37 E + 00
alpha-Ci/g	7.76 E-02	1.43 E-01
PE-Ci/g	9.59 E-02	1.38 E-01
FGE/g	9.47 E-01	9.02 E-01
watts	2.50 E-03	4.60 E-03

separated 6% ^{240}Pu material while another was processing aged 12% ^{240}Pu material. Large errors in the WIPP values arise if complete isotopic information does not follow the waste drum to the SGSAS.

CHANGE OF ISOTOPES AT CAMPAIGNS

Related to the preceeding section is the situation where the isotopic composition of the material is changed at a change of campaigns. From extensive experience in NDA holdup measurements, it has been shown that even though the surfaces in a glove box have been cleaned, significant quantities of plutonium may remain. The isotopic mixtures shown in the following tables illustrate ideal operations and conservative assumptions about actual operations. The pure (100%) isotopic composition represents an ideal clean-out from the previous campaign. The mixtures (90%-10%) reflect actual operations where the isotopes from the previous campaign are decayed 6 mo from separation.. The actual operations mixture is conservatively given as 90%-10%. A recent case was closer to a 50%-50% mix.

Isotope (wt%)	100% X	Mixed of batch isotopics		100% Y
		90% X 10% Y	10% X 90% Y	
^{238}Pu	0.0320	0.0370	0.0770	0.082
^{239}Pu	92.7700	92.2200	87.7950	87.240
^{240}Pu	6.3660	6.8730	10.9330	11.440
^{241}Pu	0.8004	0.8363	1.1460	1.187
^{242}Pu	0.0320	0.0339	0.0491	0.051
^{241}Am	0.0000	0.0028	0.0019	0.000

WIPP values	100% X	Mixed of batch isotopics		100% Y
		90% X 10% Y	10% X 90% Y	
Ci/g	9.04 E-01	9.43 E-01	1.28 E + 00	1.32 E + 00
alpha-Ci/g	7.44 E-02	7.90 E-02	9.25 E-02	9.40 E-02
PE-Ci/g	9.58 E-02	9.80 E-02	1.19 E-01	1.21 E-01
FGE/g	9.47 E-01	9.43 E-01	9.06 E-01	9.02 E-01
watts	2.40 E-03	2.50 E-03	3.00 E-03	3.00 E-03

INGROWTH OF ^{241}Am

There are many scenarios where the amount of ^{241}Am would be uncertain. Due to the decay of ^{241}Pu to ^{241}Am , the quantity of ^{241}Am is constantly increasing. For a short campaign (3 to 6 mo) the amount of ingrowth will not be significant. However, it can become a problem in a larger campaign. The amount of potential impact depends on the initial ^{241}Pu content, which is higher in ^{240}Pu material.

Any time a quantity of plutonium is not discarded to waste or removed as stable product but remains in the processing area, the ingrowth becomes more significant. Recently, a unit at PFP completed a campaign to remove an old vacuum header. The material in it was several years old. The ^{241}Am content would make a significant impact on the WIPP values. Following are two sets of isotopes, each shown at four stages of decay: freshly separated, then six, twelve, and 60 mo.

Isotope (wt%)	Months from plutonium-ameridium separation			
	0	6	12	60
^{238}Pu	0.0320	0.0320	0.0320	0.0310
^{239}Pu	92.7700	92.7900	92.8100	92.9300
^{240}Pu	6.3660	6.3670	6.3680	6.3740
^{241}Pu	0.8004	0.7815	0.7629	0.6298
^{242}Pu	0.0316	0.0320	0.0320	0.0320
^{241}Am	0.0000	0.0190	0.0380	0.1710

WIPP values	Months from plutonium-ameridium separation			
	0	6	12	60
Ci/g	9.04 E-01	8.85 E-01	8.67 E-01	7.34 E-01
alpha-Ci/g	7.76 E-02	7.81 E-02	7.87 E-02	8.32 E-02
PE-Ci/g	9.59 E-02	9.55 E-02	9.53 E-02	9.37 E-02
FGE/g	9.47 E-01	9.47 E-01	9.47 E-01	9.45 E-01
watts	2.50 E-03	2.50 E-03	2.50 E-03	2.60 E-03

Isotope (wt%)	Months from plutonium-ameridium separation			
	0	6	12	60
²³⁸Pu	0.082	0.082	0.081	0.079
²³⁹Pu	87.240	87.270	87.290	87.460
²⁴⁰Pu	11.440	11.440	11.450	11.470
²⁴¹Pu	1.187	1.159	1.132	0.935
²⁴²Pu	0.051	0.051	0.051	0.051
²⁴¹Am	0.000	0.028	0.056	0.254

WIPP values	Months from plutonium-ameridium separation			
	0	6	12	60
Ci/g	1.32 E + 00	1.29 E + 00	1.26 E + 00	1.07 E + 00
alpha-Ci/g	9.40 E-02	9.50 E-02	9.59 E-02	1.03 E-01
PE-Ci/g	1.21 E-01	1.21 E-01	1.21 E-01	1.18 E-01
FGE/g	9.02 E-01	9.01 E-01	9.01 E-01	8.98 E-01
watts	3.00 E-03	3.00 E-03	3.10 E-03	3.30 E-03

MAGNITUDE OF UNCERTAINTIES IN SEGMENTED GAMMA SCAN/ASSAY SYSTEM MEASUREMENTS AND ISOTOPIC-SENSITIVE WASTE ISOLATION PILOT PLANT VALUES

It should be noted that the magnitude of uncertainty in the assay value generated by the SGSAS can be large in comparison to the uncertainties listed previously. Due to the nature of the SGSAS assay, at lower gram levels the uncertainty is a higher percentage of the assay value than at higher gram levels. Background levels also have a large impact on the uncertainty calculations. High background areas such as in room 19 of 236-Z give rise to higher uncertainties at the same gram levels that are found in an item.

At low gram levels (less than 50 to 100 g plutonium) the SGSAS assay uncertainties will predominate. Uncertainties due to isotopic composition will predominate at higher gram levels. Which factor will predominate in any one situation, however, must be evaluated for that specific situation.

CALCULATIONS

CALCULATION OF WASTE ISOLATION PILOT PLANT PER GRAM VALUES

The values and factors used in the previous calculations are noted in the following tables. These are included to ensure consistency among users. Due to slight variations in the half-lives in published literature, compared to those accepted by national standards organizations, the curie per gram of isotope value calculated for some isotopes may change.

HALF-LIVES OF RADIONUCLIDES AND ALPHA BRANCHING RATIOS

Isotope	Half-life (yr) ^a	Alpha branching ratios ^b
²³⁸ Pu	87.79	1.0000000
²³⁹ Pu	24082.00	1.0000000
²⁴⁰ Pu	6537.00	1.0000000
²⁴¹ Pu	14.35	0.0000245
²⁴² Pu	376300.00 ^b	1.0000000
²⁴¹ Am	434.10	1.0000000

^aSource (except for ²⁴²Pu): "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control", ANSI N15.22-1975, p. 21, approved June 5, 1975.

^bSource: Table of Isotopes, Seventh Edition, ed. C. Michael Lederer and Virginia S. Shirley, pp. 1457, 1464, 1469, 1472, 1474, 1477, John Wiley & Sons, Inc., New York, New York (1978).

CALCULATED CURIE AND ALPHA CURIE PER GRAM FACTORS

Isotope	Curies per gram of isotope	Alpha-curies per gram of isotope
^{238}Pu	17.119320	17.119320
^{239}Pu	0.061964	0.061964
^{240}Pu	0.226640	0.226640
^{241}Pu	103.298100	0.003924
^{242}Pu	0.003924	0.002531
^{241}Am	3.432510	3.432510

The equations used in the above calculations are as follows.

$$\text{Curies per gram of isotope (Ci/gi)} = \lambda N / 3.7 \text{ E} + 10$$

where λ is the decay constant of the isotope

$$\lambda = \frac{\ln 2}{\text{half-life} * 3.1558 \text{ E} + 07}$$

and N is the number of atoms in one gram of the isotope

$$N = \frac{6.02 \text{ E} + 23}{\text{element weight of isotope}}$$

The factor $3.7 \text{ E} + 10$ is the definition of a curie and equals the number of disintegrations per second of an isotope per curie.

As noted earlier the WIPP factors are calculated for a specific isotopic distribution. The isotopic distribution for a recently measured item is the basis for the calculations below.

The curies per gram of plutonium is calculated by:

$$\text{Ci/g Pu} = \text{isotope mass fraction} * \text{curies per gram of isotope}$$

Isotope	Mass fraction of isotope per gram of plutonium	Curies per gram of isotope	Curies per gram of plutonium
²³⁸Pu	0.000322	17.119320	0.005512
²³⁹Pu	0.927700	0.061964	0.057484
²⁴⁰Pu	0.063660	0.226640	0.014427
²⁴¹Pu	0.008004	103.298100	0.826798
²⁴²Pu	0.000314	0.003924	0.000001
²⁴¹Am	0.000060	3.432510	0.000189
TOTAL			0.904411

The number of alpha-curies per gram of isotope is calculated by

$$\text{alpha-Ci/g of isotope} = \text{Ci/g of isotope} * \text{isotope alpha branching ratio}$$

or

$$\text{alpha-ci/g of Pu} = \text{isotope mass fraction} * \text{alpha-Ci/g of isotope}$$

Isotope	Curies per gram of plutonium	Alpha branching ratio	Alpha-Ci/g plutonium
²³⁸Pu	0.0055124	1.0000000	0.005512
²³⁹Pu	0.057484	1.0000000	0.057484
²⁴⁰Pu	0.014427	1.0000000	0.014427
²⁴¹Pu	0.826798	0.0000245	0.000020
²⁴²Pu	0.000001232	1.0000000	0.000001
²⁴¹Am	0.000188788	1.0000000	0.000189
TOTAL			0.077633

The ^{239}Pu Equivalent Transuranic Activity (PE-Ci) per gram of plutonium factor is calculated as follows:

$$\text{PE-Ci/g Pu} = \text{isotope Ci/g Pu} * \text{isotope PE-Ci factor}$$

Isotope	Ci/g plutonium	PE-Ci correction factor*	PE-Ci/g plutonium
^{238}Pu	0.005512	1	0.005512
^{239}Pu	0.057484	1	0.057484
^{240}Pu	0.014427	1	0.014427
^{241}Pu	0.826798	1/45	0.018373
^{242}Pu	0.000001	1	0.000001
^{241}Am	0.000189	1/3	0.000063
TOTAL			0.095860

*Source: RHO-MA-222 Rev. 3, "Hanford Radioactive Solid Waste Packaging, Storage, and Disposal Requirements, Appendix C.

The ^{239}Pu FGE per gram of plutonium factor is calculated as follows:

$$\text{FGE/g Pu} = \text{Isotope Mass Fraction} * \text{Isotope FGE factor}$$

Isotope	Ci/g plutonium	FGE correction factor*	PE-Ci/g plutonium
^{238}Pu	0.000322	9/80	0.000036
^{239}Pu	0.927700	1	0.927700
^{240}Pu	0.063660	9/400	0.001432
^{241}Pu	0.008004	9/4	0.018009
^{242}Pu	0.000314	9/1200	0.000002
^{241}Am	0.000006	9/480	0.000001
TOTAL			0.947181

*Source: RHO-MA-222 Rev. 3, "Hanford Radioactive Solid Waste Packaging, Storage, and Disposal Requirements, Appendix B.

The thermal power of an item is expressed in watts and is calculated using a watts per gram of plutonium (W/g) factor. The calculation of the watt per gram factor is shown below.

$$\text{Watts/g Pu} = \text{Isotope Mass Fraction} * \text{isotope specific wattage}$$

Isotope	Isotope mass fraction	Watts per gram of isotope*	W/g plutonium
²³⁸Pu	0.000322	0.5675700	0.000183
²³⁹Pu	0.927700	0.0019288	0.001789
²⁴⁰Pu	0.063660	0.0070824	0.000451
²⁴¹Pu	0.008004	0.0034120	0.000027
²⁴²Pu	0.000314	0.0001159	0.000000
²⁴¹Am	0.000060	0.1142000	0.000007
TOTAL			0.002457

*Source: "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," ANSI N15.22-1975, p. 21, approved June 5, 1975.

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