CALCULATION of Np-237 and Am-241 DETECTOR CALIBRATION CONSTANTS FROM FIRST PRINCIPLES

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Abstract

The Analytical Development Section of Savannah River Technology Center (SRTC) was requested by the Facilities Disposition Projects (FDP) to determine the holdup of enriched uranium in the 321-M facility as part of an overall deactivation project of the facility. The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the production reactors. The results of the holdup assays are essential for determining compliance with the Waste Acceptance Criteria, Material Control & Accountability, and to meet criticality safety controls. This report covers calibration of the detectors in order to support holdup measurements in the C and D out-gassing ovens. These ovens were used to remove gas entrained in billet assembly material prior to the billets being extruded into rods by the extrusion press. A portable high purity germanium detection system was used to determine highly enriched uranium (HEU) holdup and to determine holdup of U-235, Np-237, and Am-241 that were observed in these components. The detector system was run by an EG&G Dart™ system that contains the high voltage power supply and signal processing electronics. A personal computer with Gamma-Vision software was used to control the Dart™ MCA and provide space to store and manipulate multiple 4096-channel γ-ray spectra. The measured Np-237 and Am-241 contents were especially important in these components because their presence is unusual and unexpected in 321-M. It was important to obtain a measured value of these two components to disposition the out-gassing ovens and to determine whether a separate waste stream was necessary for release of these contaminated components to the E-Area Solid Waste Vault. This report presents determination of the calibration constants from first principles for determination of Am-241 and Np-237 using this detection system and compares the values obtained for Np-237 with the calibration factors obtained with a subsequent measurement using a point source of radioactive equilibrium Np-237/Pa-233.

INTRODUCTION

The 321-M facility was used to fabricate enriched uranium fuel assemblies, lithium-aluminum target tubes, neptunium assemblies, and miscellaneous components for the Savannah River production reactors. The facility operated for 25 years. During this time thousands of uranium-aluminum-alloy (U-Al) fuel tubes were produced. After the facility ceased operations in 1995, all of the easily accessible U-Al was removed from the building, and only residual amounts remained. The bulk of this residue is located in the equipment that generated and handled small U-Al particles and in the exhaust systems for this equipment. The Analytical Development Section of Savannah River Technology Center was tasked to perform holdup and material control and accountability assays to determine content of highly enriched uranium (HEU) in the deactivation and decommissioning (D&D) activities of the 321-M facility.[1]

These D&D and assay activities have been on-going from FY00 through FY03. In the course of the FY03 D&D activities Radiation Control Operations (RCO) surveys found Np-237 and Am-241 activity in the C and D outgassing ovens of the facility.[2] Radioactivity from sources other than U-235 was completely unexpected in the facility. In the 25 years of operation the facility performed process and mechanical work on uranium fuel and target rods for the production reactors. While recycled material was run through the facility, and therefore uranium material that contained the isotopes 232, 233, 234, and 236 were observed, at no time did the facility expect to observe plutonium product or trans-plutonium activation products. Observation of Np-237 and Am-241 activity in the RCO surveys of 2003 surprised the Facility Disposition Program (FDP) D&D operations management. Along with radiation control concerns, appearance of Np-237 and Am-241 radioactivity was not consistent with the designated solid waste stream for the facility. Observation of these two species appeared to violate the Waste Acceptance Criteria for the facility.[3] To quickly respond to these concerns, ADS was requested to perform a rapid assessment of the Np-237/Pa-233 equilibrium and Am-241 contents of the C and D ovens.

At the time of the request ADS had no detector efficiency calibrated to perform the Np-237/Pa-233 or Am-241 γ-ray assays. We performed a rapid calculation of the necessary point source and line source calibration constants for the HPGe detector using first principles. Reported Np-237 and Am-241 contents were based on these first principle calculations.[4-7] This report describes the results of the first principles calculation and gives a comparison with the experimental determination of point source and line source efficiency calibration constants for Np-237. The difference between the calculated and experimentally measured line source constant is -10%, and the difference between the calculated and experimentally measured point source constant is -20%. Both agreements are very good and well within the reported 25% uncertainty for the Np-237 and Am-241 measurements.

Calculations

Approach 1

For a line source acquisition configuration the HPGe detector has been previously experimentally determined to have a calibration constant of 1.72×10^{-5} g-sec/cm^2 for assay of U-235.[8] The equation for determination of line source content from measured detection rate takes the form[9]

$$[\text{U-235}] = K_l \times \text{cps} \times d \times Cf(T) \times L, \quad (1)$$

where K_l is the calibration constant, d is the distance from the line source to the detector, and Cf(T) is the transmission correction factor for the source (sample). Application of the first four terms of this equation to measured data yields a measure of U-235 content in the line source in units of g/cm of the source. U-235 content in grams is determined by multiplication of the length L of the line source.

The measured counts per second cps in the peak of interest in the γ-ray spectrum is empirically determined by

$$\text{cps} = (\text{dps})(\text{eff})(\text{branch}), \quad (2)$$

where *dps* is the decay rate for the sample, *eff* is the detector efficiency for counting the γ-ray as a photopeak in the detection system, and *branch* is the probability (branching ratio) that a decay of the species will yield a γ-ray of the appropriate energy. For U-235 the branching ratio is 0.53.[10] Substituting and rearranging we obtain an expression for the line source calibration constant

$$K_l = [\text{U-235}]/(\text{dps})(\text{eff})(\text{branch})(d)(Cf(T)). \quad (3)$$

In (3) [U-235] is in units of g/cm, and we have not specified L. We can solve for K_l by assuming a transmission correction factor of 1 if we can determine the detector efficiency. This is the approach we take below to obtain a measure of K_l for Np-237 decay using the 312 keV γ-ray from Np-237/Pa-233 equilibrium decay. That is, we assume we can calculate K_l from first principles if we can obtain a measured value for each of the five factors in the denominator of (3).

To obtain a measure of *eff* for the 312-keV γ-ray from Np-237/Pa-233 decay, we rearrange (3) to determine eff(185) for the 185-keV γ-ray from U-235 decay. From there we will be able to determine eff(312).

$$\text{eff}(185) = [\text{U-235}]/(\text{dps})(K_l)(\text{branch})(d)(Cf(T)). \quad (4)$$

K_l we know already to be 1.72×10^{-5} g-sec/cm^2, and *dps* we obtain from the specific activity of U-235, which is 2.14×10^{-6} Curie/g = 79180 dps/g. Substituting we obtain for counting a line source of 1g/cm content with a self-absorption factor of 1 at a distance of 1 cm

$$\text{eff}(185) = [1 \text{ g/cm}]/(79180/\text{sec})(1.72 \times 10^{-5} \text{ g-sec/cm}^2)(0.53)(1 \text{ cm})(1) = 1.385. \quad (4)$$

This is equivalent to stating that counting a 1-g/cm line source at a distance of 1 cm, we would observe using equation (2)

$$\text{cps} = (1.385)(0.53)(79180) = 58122 \text{ cps} \quad (2)$$

in the 185 keV peak. Note the eff(185) is greater than one in (4) because the detector is able to view much more than one cm of the line source. Counting the same line source at d = 50 cm would yield an efficiency of 0.0277.

We have a defensible approach for determination of K_l from known efficiency. How do we transform this to a line source calibration constant for Np-237/Pa-233 equilibrium decay? Using a planar source that contains Am-241, Eu-152, and Cs-137 activity,[11] we measured the absolute efficiency for counting multiple γ-rays in a fixed geometry. These data are recorded on page 10 of NB-2000-00086[(12)] and are shown in Table 1. Fitting the data of Table 1 with a log-log function we obtain eff(185) = 0.01134 and eff(312) = 0.008717 for the specific configuration and fixed geometry of Table 1. Intrinsically for this detector then eff(312)/eff(185) = 0.7687.

Table 1. Absolute detection efficiency obtained using cylindrical source data of reference 12.

Energy	Area	Cps	Branch	eff
58.89	117306±628	587±3	0.357	0.00283±1
121.40	422838±1076	2114±5	0.284	0.01339±3
244.48	83674±628	418±3	0.0751	0.01001±7
344.07	227124±689	1136±3	0.266	0.00768±2
661.25	287435±746	1437±4	0.8521	0.00434±1

Returning to (3) for a 1 g/cm line source of Np-237/Pa-233 and counting the 312-keV γ-ray

$$K_l = [\text{Np-237/Pa-233}]/(\text{dps})(\text{eff})(\text{branch})(d)(Cf(T)) \quad (3)$$

$$= [1 \text{ g/cm}]/(2.6085\text{x}10^{7} \text{ sec}^{-1})(1.385\text{x}0.7687)(0.36)(1 \text{ cm})(1) = 1.00\text{x}10^{-7} \text{ g-sec/cm}^2,$$

where $2.6085\text{x}10^{7}$ is the specific activity in decays per second of Np-236, and 0.36 is the branching ratio of the 312-keV γ-ray in the Np-237/Pa-233 equilibrium.[10]

Approach 2

Alternately we can use an approach that uses an absolute measure of the point source efficiency for counting the 185-keV γ-ray from U-235 and attempt to transform this to a line source calibration constant for Np-237/Pa-233 equilibrium. This approach also uses the ratio of efficiencies for eff(185) and eff(312) obtained in Table 1, but might provide the reader with a more comfortable rationalization. Ultimately the two approaches are identical in nature.

From the data on pages 32-39 of Laboratory Notebook NB-95-00180 (reference 13) we have measured a point source detection rate of 46 cps for counting a 1-g source of U-235 with this detector at a distance of 1 foot. That is, rearranging equation (2)

$$\text{eff}(185) = (\text{cps})/(\text{dps})(\text{branch}) \quad (2)$$

$$= 46/(79180)(0.53) = 0.001096.$$

This agrees extremely well with the measured efficiency when we rearrange equation (4) for the point source configuration acquiring at 1 foot (reference 13).

$$\text{eff}(185) = [\text{U-235}]/(\text{dps})(K_p)(\text{branch})(d)^2(Cf(T)). \quad (5)$$

For a 1-g source and a measured point source calibration constant of 2.36×10^{-5} g-sec/cm^2 (reference 8), we obtain

$$\text{eff}(185) = [1\text{ g}]/(79180)(2.36\times10^{-5})(0.53)(12\times2.54)^2(1). \quad (5)$$

$$= 0.001087.$$

Recall from the data of Table 1 that we have a measure of the intrinsic efficiencies for this detector at 185 keV and 312 keV. That is, eff(312)/eff(185) = 0.769. Therefore in an identical point source configuration acquiring from 1 foot we have eff(312) = 0.000843. Hence the ratio of K_p(U-235) to K_p(Np237/Pa-233) is

$$\frac{K_p(\text{U-235})}{K_p(\text{Np-237})} = \frac{(\text{dps})_{Np}(\text{eff})_{312}(\text{branch})_{312}(d)^2}{(\text{dps})_{U}(\text{eff})_{185}(\text{branch})_{185}(d)^2}$$

$$= \frac{(2.6085\times10^{7})(0.000843)(0.36)(d)^2}{(79180)(0.001096)(0.53)(d)^2} = 172. \quad (6)$$

$$K_p(\text{Np-237}) = (2.36\times10^{-5})/172 = 1.37\times10^{-7}\text{ g-sec/cm}^2. \quad (7)$$

Using the identical ratio for the line source calibration constants we obtain

$$K_l(\text{Np-237}) = (1.72\times10^{-5})/172 = 1.00\times10^{-7}\text{ g-sec/cm}^2. \quad (8)$$

Comparison with Experimental Measure

We used the line source and point source calibration constants calculated from the first principles of Approaches 1 and 2 to determine the Np-237/Pa-233 holdup in ovens C and D of 321-M. The measured holdup was reported to the customer in references 5 and 7. A full description of the holdup measurements and calculations using these calibration constants is provided in reference 14. We reported 17±7 mg of Np-237 in oven C and < 5mg Np-237 in oven D. Both of these assay values included transmission correction values that were measured experimentally as reported in reference 14. The oven D measurement includes data acquired with the HPGe detector and with a separate NaI detector. The NaI calibration constants were derived using a first principles calculation very similar to Approach 2 of this paper.

After reporting the measured holdup in March and April of 2003 we were able to find time in June of 2003 to go back and experimentally calibrate the HPGe detector for Np-237/Pa-233 measurements using the accepted techniques of references 9, 15, and 16. This was a very important laboratory exercise to benchmark our reported values. Table 2 is an Excel spread sheet that contains the acquired data for the point, line, and area source configurations using the HPGe detector with a point source of equilibrium Np-237/Pa-233 radioactivity. Table 2 contains the acquired data at an on-axis distance of 20 inches and with off-axis incremental movements of the source by graduations of four inches until the source went out of view of the collimated detector.

The source was 139 μg of Np-237 made in 1995. With an eight-year-old source we can clearly treat the Pa-233 ($T_{1/2}$ = 27 days) daughter as in radioactive equilibrium so that the 312-keV γ-ray is directly representative of the Np-237 content of the source. The source activity was determined by the Analytical Development Section counting room γ-PHA instrument to be 0.0983(10) μCi in activity.[17] The specific activity for Np-237 is 0.000705 Ci/g,[10] and so that yields a mass of 139 μg.

All of the acquired data and all of the calculations are shown completely in the spreadsheet. Note in the matrix elements K_p and K_l that we obtained a measured point source calibration constant of 1.71×10^{-7} g-sec/cm^2, and a line source constant of 1.11×10^{-7}. These values are in very good agreement with the values derived from first principles, with deviations of +20% and +11% respectively.

Am-241 Contents in Ovens C and D.

Using identical approaches we calculated a point source calibration constant of $K_p = 8.76x10^{-11}$ g-sec/cm^2 and a line source constant of $6.38x10^{-11}$ for Am-241 assays with the HPGe detection system. We used these line source and point source calibration constants to determine the Am-241 holdup in the C and D ovens. The measured holdup was reported to the customer in references 6 and 7. A full description of the holdup measurements and calculations using these calibration constants is provided in reference 14. We reported 1.3 μg of Am-241 in oven C and < 400 ng Am-241 in oven D. The oven D measurement includes data acquired with the HPGe detector and with a separate NaI detector. References 7 and 14 include a short discussion proving that the Am-241 and Np-237 were not in radioactive equilibrium.

Conclusion

In this report we describe determination of the line source and point source γ-ray acquisition efficiency calibration constants for Np-237/Pa-233 using the equilibrium 312-keV photon and for Am-241 using the 59.6 keV photon. The calibration constants were derived from first principles of relative intrinsic detection efficiency versus energy using the published branching ratios of the γ-rays and published specific activities of the Np-237 and Am-241 radioactive species. The values of the calibration constants were established using the empirically measured decay and branching ratio constants and by comparing with the measured point and line source calibration constants for U-235 decay.

The calculations from first principles are then compared with an experimental measure of the two values for Np-237/Pa-233 decay. The point and line source constant were measured for Np-237/Pa-233 using a 139-μg point source of Np-237 in equilibrium with it daughter Pa-233. We obtained agreement within 20% for the point source constant and within 11% for the line source constant. This excellent agreement firmly establishes the method of calculation and supports the reported values of Np-237 and Am-241 holdup in two out-gassing ovens in the Savannah River Site Reactor Fuel Fabrication Facility.

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Table 2. Excel Spreadsheet for calculation of Np-237 detector efficiency

	Kp	=	mass standard/(cps)(Cf)(distance)^2.							
	K_l	=	mass standard/(cps)(Cf)(distance)(L)							
	L	=	$2S\{\Sigma C_i/C_0\}-S$							
	Where S is the increment distance,									
	Ka	=	$m/A(C_0)(Cf)$,							
where	A	=	$\pi S^2/4 + \Sigma 2i\pi S^2 C_i/C_0$							
	$\pi S^2/4$=	81.0732								
	$\Sigma 2i\pi S^2 C_i/C_{0=}$	6478.992								
	A=	6560.066								
1" recess										
Location	Count Time	312 kev	%σ312 kev	cps	% σ	K_p	L	K_l	K_a	$i*C_i$
	sec	Area	Area							
0	400	126	11.6	0.315	0.092063	1.71E-07	78.00085	1.11E-07	6.73E-08	
1	600	178	10.8	0.296667	0.060674					0.296667
2	600	135	14.5	0.225	0.107407					0.45
3	600	83	23	0.138333	0.277108					0.415
4	600	79	22	0.131667	0.278481					0.526667
5	600	61	27	0.101667	0.442623					0.508333
6	600	95	15	0.158333	0.157895					0.95
7	400	0	25.21	0	#DIV/0!					0
1.39E-04	mass of standard									
1	correction factor									
50.8	distance in cm			22	BKG cp 60 sec					
10.16	displacement in cm									