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Plant Instrumentation Program

3rd Quarterly Progress Report

October-December 1970

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Plant Instrumentation Program

Third Quarterly Report
October-December 1970

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Project Management

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Introduction

This is the third quarterly progress report covering work performed under the subject contract. Previous reports of work performed under this contract are:

BHO-69-1 Quarterly Progress Report, April-May-June 1970

BHO-69-2 Quarterly Progress Report, July-August-September 1970

Effort during this quarter was concentrated on the development and demonstration of gamma measurement of the plutonium content of plutonium nitrate solution in polyethylene bottles, coincidence-neutron counting, gamma assay of scrap recovery raffinate in 30-gallon drums, and the gamma assay of scrap.

Summary

The concentration and fissile content of plutonium as plutonium nitrate solution in ten-liter polyethylene storage bottles can be determined by passive counting using a Ge(Li) detector. The relative uncertainty in plutonium concentration is approximately four percent, and the method for determination of plutonium concentration is insensitive to the volume of solution in the bottles. However, the determination of the total plutonium content is sensitive to the level of the liquid. It has been shown that the variation of wall thickness along the length of the bottles partially accounts for the non-linear decrease in specific activity for partially full nitrate bottles.

The coincidence-neutron counter, designed and built by Brookhaven National Laboratory, has been evaluated for counting oxides and oxide scrap. Oxide samples containing up to 100 grams of Pu-240 can be routinely counted with a relative standard deviation in the coincidence counts of approximately five percent for a one-minute counting time.

Raffinates from the recovery of ZPR-III fuel were gamma counted using the same procedure as used to count 55-gallon drums containing glovebox-generated waste. The activity in the "384-Kev complex" of the 30-gallon drums was shown to be a linear function of the amount of plutonium recovered from the drums. This relationship probably results from the fact that the concentration of plutonium in the drums is less than 0.2 grams per liter.

1.0 Nondestructive Gamma Assay of Plutonium Nitrate Solution

It has been shown by J.E. Cline, et.al,⁽¹⁾ that the relative attenuation of the 129-Kev and 413-Kev gamma photons from Pu-239 and the 148-Kev gamma photons from Pu-241 can be used to determine the plutonium concentration (grams of Pu/liter) and the total Pu-239 and Pu-241 contents of plutonium nitrate solution in polyethylene shipping bottles. The nondestructive assay technique developed by Cline was evaluated from the standpoint of adaptability to a typical commercial plutonium recovery plant.

Cline's experimental setup measured a polyethylene bottle by itself and inside a stainless-steel container. Two different stainless-steel containers were used to represent the storage containers for L-10 nitrate shipping bottles used by Atlantic Richfield Hanford Company (ARHCO) at Hanford (#10-gauge stainless steel, 0.134 inches thick) and the Atomic and Space Development Authority (ASDA) in New York (#14-gauge; 0.0747 inches thick). A one-cubic centimeter Ge(Li) detector was used at a container-to-detector distance of 54 inches with the detector located at the mid-longitudinal axis of the container. A counting time of 1000 seconds was used throughout Cline's work.

A plutonium nitrate bottle from the NUMEC plutonium recovery plant, containing 200 grams of plutonium per liter, was counted at a 54-inch container-to-detector distance with a 55-cc Ge(Li) detector. The dead time of the instrument was approximately 50%. At 100 inches from the bottle, the dead time with no absorber in front of the detector was 35%.

The high dead time rate was primarily due to the much higher efficiency of the 55-cc Ge(Li) detector for the 60-Kev gamma photon from americium-241. The use of an absorber in front of the detector reduced the instrument dead time, but it also greatly reduced the activity of the 129-Kev photon, one of the measurements necessary to determine the concentration of the plutonium and then the Pu-239 content of the bottles.

Alternate solutions to this problem were to use a small detector with its inherent lower efficiency, or to use the large detector 100 inches or more from the nitrate bottle. The effects of different absorbers on the dead time and the activities from a bottle containing 232 grams of plutonium per liter counted at 100 inches from the detector are shown in Table 1.1.

Table 1.1 - Effect of Absorber on Instrument Dead Time

Absorber and Thickness (inches)	Dead Time (%)	Total Net Counts in 1000 Seconds			
		129-Kev	413-Kev	129/413	"384 Complex"
None	35	273,500	81,100	3.372	314,500
Hf, 0.009	20	153,300	84,250	1.819	311,000
Hf, 0.018	15	68,200	83,700	0.816	307,800
Pb, 0.125	5	0	48,900	0	168,000

The geometry selected for counting plutonium nitrate solution in polyethylene bottles was a detector-to-bottle centerline distance of 100 inches with the detector located at the mid-axis of the bottle. No absorbers were used over the face of the detector, and the discriminator was set to discriminate below 100 Kev. With these conditions a 230g-per-liter bottle gave an instrument dead time of 17% which is considered an allowable value. A dead time higher than this value would not be likely since the maximum allowable concentration for shipping is 250g/liter.

The L-10 polyethylene bottles used at NUMEC are purchased from Monsanto Corporation of Stonington, Connecticut. The specifications for the manufacture of these bottles were prepared by ARHCO: length, 50.25 ± 0.250 inches; outside diameter, 4.430 inches to 4.570 inches; and a minimum wall thickness of 0.100 inches. The results of destructive tests, performed by the manufacturer, indicate the outside diameter is 4.570 inches and the wall thickness is 0.150 inches at the top, 0.200 inches at the center, and 0.240 inches at the bottom of the polyethylene bottles. The volume is 9.98 liters at the base of the neck, and the density of the polyethylene is 0.955 g/cc.

In order to establish a calibration curve, several different plutonium concentrations covering the range of 100 to 250 grams/liter were required. The normal concentration out of the recovery plant is 200 grams/liter with a maximum allow-

able concentration for the plant of 215 grams/liter. Bottle #409 with an assay of 232 grams/liter was used in the calibration, and a special bottle was prepared at 100 grams/liter concentration. These two containers provided calibration points at the extreme ends of the range of concentrations that might require analysis.

Cline's work has shown that the plutonium concentration can be correlated with the activity ratio of two photons having significantly different energies. The energy selected is not critical, but a high yield and an energy free from interference by other isotopes are desired. For the measurement of the plutonium concentration, the 129-Kev and 413-Kev gamma rays of Pu-239 were selected.

A Nuclear Diodes 55-cm³ Ge(Li) detector was used throughout this work. The number of counts from the 129-Kev gamma ray was summed over six channels, and the subtracted Compton contribution was averaged over the next five channels of higher energy. The 148-Kev gamma ray was summed over six channels and the 413-Kev gamma ray over ten channels with six- and ten-channel averages, respectively, used for the Compton contribution subtraction.

Bottles 409 and 411 were used as the primary calibration points. Table 1.2 gives the analytical results for the two bottles.

Table 1.2 - Analytical Results for Plutonium Nitrate Bottles 409 and 411

Bottle Number	Plutonium Concentration		Plutonium Content (grams)		Isotopic Composition (w/o)				
	(g/l)	(g/g)	Pu-239	Pu-241	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
409	232.11	0.1556	1802.3	21.63	0.028	89.16	9.58	1.07	0.156
411	92.50	0.717	712.4	9.07	0.029	88.74	9.94	1.13	0.159

Table 1.3 gives the mean number of counts corrected for Compton (three repetitive readings) for 1000 and 200 seconds from bottles 409 and 411 for the three energies.

Table 1.3 - Comparison of Data for 1000- and 200-Second Counts

Bottle	Net Counts per 1000 Seconds				Net Counts per 200 Seconds			
	129-Kev	148-Kev	413-Kev	129/413	129-Kev	148-Kev	413-Kev	129/413
409	256,020	173,420	83,220	3.076	52,770	35,240	16,700	3.144
411	227,860	157,050	41,420	5.500	45,510	32,010	8,230	5.530

The standard deviation of the ratio of the activities from the 129-Kev gamma ray to that from the 413-Kev gamma ray is 0.03 for the 1000 second counting and 0.05 for the 200 second counting time for bottles 409 and 411. The above data clearly show that counting time has little effect on the (129/413) ratio for this detector arrangement. Figure 1.1 shows the experimental data and the calculated curve for 1000 seconds of counting time.

Since it is possible that plutonium nitrate shipping bottles will, at some time in the future, be shipped or stored in polyethylene bottles encased in stainless steel, a series of measurements were made with the bottles shielded with 0.076-inch-wall and 0.134-inch-wall 304-stainless-steel tubing. These wall thicknesses are consistent with the stainless-steel containers used at ASDA and ARHCO.

The stainless-steel-shielded bottles were counted under the same geometrical conditions and with the same detector that was used for the bare polyethylene bottles. A counting time of 1000 seconds was selected due to an expected lower counting rate for the shielded bottles. The experimental data are shown in Table 1.4 and Figure 1.2.

Table 1.4 - Counts for Bottles 409 and 411 when Shielded with Stainless-Steel

Bottle Number	Shielded with 0.076 inches of S.S.				Shielded with 0.134 inches of S.S.			
	Net Counts			Ratio	Net Counts			Ratio
	in 1000 Seconds				in 1000 Seconds			
	129-Kev	148-Kev	413-Kev		129-Kev	148-Kev	413-Kev	
409	207,100	145,200	80,540	2.571	169,590	124,320	72,920	2.326
411	171,460	124,740	37,690	4.550	138,150	106,000	34,790	3.986

To test the accuracy of the above nondestructive gamma-assay technique for the measurement of plutonium concentration, bottle 412 was counted for 1000 seconds. The

FIGURE 1.2

THE EFFECT OF 0.076" AND 0.134" OF STAINLESS STEEL SHIELDS
ON THE RATIO OF 129 KEV TO 413 KEV GAMMA RAYS
FROM Pu-239 AS A FUNCTION OF PLUTONIUM DENSITY

bottle was counted three times, and the mean value of the 129/413 ratio was determined to be 3.237 with a 3σ value of 0.044. This would correspond to a plutonium density (from Figure 1.1) of 206 ± 6 grams/liter. A chemical analysis of a sample from bottle 412 indicated 214.0 ± 0.54 grams/liter. This is a difference of 3.9%.

The ability to detect and assay partially full bottles was evaluated by shielding the lower 20 inches of bottles 409 and 411. The data are shown below in Table 1.5 and in Figure 1.1.

Table 1.5 - Mockup of Half-full Bottles of Plutonium Nitrate Solution

Bottle Number	Counts in 1000 Seconds			Ratio 129/413
	129-Kev	148-Kev	413-Kev	
409	72,250	48,390	24,210	2.984
411	145,670	98,560	27,880	5.223

Each bottle was only counted once; hence, no statistics were calculated. The 129/413 ratio for bottle 409 (232.1 grams/liter) falls within the 3σ value shown in Figure 1.1 and the ratio for bottle 411 (92.5 grams/liter) falls on the calculated curve.

The Pu-239 contents and the Pu-241 contents of the unshielded polyethylene bottles can be obtained from the 129-Kev and 148-Kev counts. Figures 1.3 and 1.4 show the 129-Kev counts/gram of Pu-239 and the 148-Kev counts/gram of Pu-241 versus the plutonium concentration. Similar correlations can be made for polyethylene bottles shielded with stainless steel.

As previously shown, the plutonium concentration determined by gamma counting is rather insensitive to the volume of solution contained in the bottles. However, it is quite obvious from the data in Tables 1.3 and 1.5, which show the number of counts from the 129-Kev, 148-Kev, and 413-Kev gamma protons for simulated half-full and for full plutonium nitrate bottles, that the activities for the gamma ray energies were not reduced in a linear manner.

FIGURE 1.3

THE 129 - Kev COUNTS/GRAM OF Pu-239
AS A FUNCTION OF PLUTONIUM DENSITY

FIGURE 1.4

THE 148 - Kev COUNTS /GRAM OF Pu - 241
AS A FUNCTION OF PLUTONIUM DENSITY

For bottle 409 at 232.1 g/l the number of counts from the 129-Kev gamma ray should have been reduced to $256,000/2 = 128,000$ counts compared to the measured half-bottle value of 72,250 counts. Hence, for the same plutonium concentration, the Pu-239 measurement would have been low by a factor of $128/72.25 = 1.77$. Similarly, for bottle 411 at 92.5 g/l, the number of counts from the 129-Kev gamma ray should have been $227,860/2 = 113,900$ counts compared to the measured half-bottle value of 145,700 counts. Hence, at this lower concentration the Pu-239 content would have been over-estimated by 28%. Similar ratios were measured for the 148-Kev gamma ray from Pu-241. These variations are probably due to the variations in wall thickness of the bottle. The thinner wall at the top of the container would account for the non-linear decrease in counts observed for the simulated half-full bottles. During the next quarter, additional work will be directed towards confirming these findings.

The number I of photons of energy E (Kev), recorded by the detector at a distance A (cm) from the plutonium nitrate/nitric acid/water source inside a polyethylene bottle is

$$I = \frac{K}{4\pi} \int_0^{H/2} dh \int_0^{2\pi} d\phi \int_0^{R_2} r dr \exp[-U_1 R_1 - U_2 R_2] / \rho^2$$

where U_1 and U_2 are the linear attenuation coefficients of the source and polyethylene bottle, respectively; R_2 is the outer radius and R_1 is the inner radius of the polyethylene bottle; and H is the height of the solution. K is a constant determined from the detector efficiency, branching ratio, and geometrical factor. The above equation was solved numerically using the CDC 6600 computer; the code is referenced as "G Cyl" and is written in Fortran IV.

The absorption coefficients in the equation are dependent on the concentration of plutonium in the bottle, the container material, and the photon energy. Table 1.6 gives some mass absorption coefficients (cm^2/gram) for several materials and energies.

Table 1.6 - Mass Absorption Coefficients (cm^2/gram)

Element or Compound	Photon Energy		
	129-Kev	148-Kev	413-Kev
$\text{Pu}(\text{NO}_3)_4$	2.054	1.360	0.1851
HNO_3	0.1432	0.1361	0.0957
Water	0.1566	0.1490	0.1052
CH_2	0.1599	0.1527	0.1075
Stainless Steel	0.2510	0.210	0.0907

The linear attenuation coefficient is the product of the mass absorption coefficient μ and the density ρ ; for the plutonium nitrate solution the linear attenuation coefficient (cm^{-1}) can be written as,

$$U_1 = 2.038 \mu_{\text{Pu}(\text{NO}_3)_4} \cdot \rho_{\text{Pu}} + 0.063N \cdot \mu_{\text{HNO}_3} + \mu_{\text{H}_2\text{O}} \cdot \rho_{\text{H}_2\text{O}}$$

where ρ_{Pu} and $\rho_{\text{H}_2\text{O}}$ are the densities of plutonium and water; N is the normality of nitric acid; and $\mu_{\text{Pu}(\text{NO}_3)_4}$, μ_{HNO_3} , and $\mu_{\text{H}_2\text{O}}$ are the mass absorption coefficients of plutonium nitrate solution, nitric acid, and water, respectively. The acid normality is usually five to six, and the total density of the solution is approximately 1.4 to 1.5 g/cc; hence, the above equation is sensitive only to the plutonium concentration.

As pointed out previously, there is a large variation in the wall thickness of the polyethylene bottles, and this variation is probably the cause of the non-linear decrease in counts for the half-full bottles. The calculations reported above here are for a 0.20-inch average wall thickness, for an outside diameter of 4.570 inches, and for a constant height of solution of 40 inches. Calculations are being performed to evaluate the effect of wall thickness of polyethylene on the 129-Kev, 148-Kev, and 414-Kev gamma rays.

2.0 Coincidence Neutron Counting

The repeatability and performance of the coincidence neutron counter designed and built by Brookhaven National Laboratory has been excellent. During this quarter, PuO_2 and 25 w/o $\text{PuO}_2\text{-UO}_2$ standards were counted in the well counter. The radial and axial variation of the coincidence and gross counts were remeasured, since the data previously reported was in error.⁽³⁾ The profiles for a clean and poisoned detector were measured for a 2 x 32 gate setting. The radial profile of the coincidence counts in a clean detector is shown in Figure 2.1, and the axial profile of coincidence counts is shown in Figure 2.2.

Seven PuO_2 standards and four 25 w/o $\text{PuO}_2\text{-UO}_2$ standards, in one-quart containers, 3-1/2-inches in diameter and 7-inches long, were counted in the coincidence neutron counter at various gate settings. In addition to obtaining calibration curves for a clean detector, the performance of a poisoned detector was also evaluated. The poisoned detector consists of shielding the outside circumference of the twenty-four BF_3 tubes with 0.034-inch cadmium foil.

The plutonium assays and plutonium isotopic compositions of the PuO_2 and 25 w/o $\text{PuO}_2\text{-UO}_2$ standards are shown in Table 2.1, and the inventory value of the plutonium in each one-quart container is shown in Table 2.2. The delays and gate lengths⁽²⁾ for the coincidence counter are shown in Table 2.3.

The gross counts, corrected for clock time and amplifier dead time, and the coincidence counts, corrected for amplifier dead time (corrected as $N \times F^3$), are shown in Table 2.4 and 2.5 for a clean and poisoned detector for several gate settings. Also shown in the tables are the relative standard deviations, expressed as a percent of the coincidence counts.

It has been pointed out by Brookhaven National Laboratory that the coincidence counts of samples, whose corrected gross count is approximately 100,000 counts/minute or greater, are in error.

The coincidence and gross counts for a 2 x 32 gate setting in a clean detector are shown in Figures 2.3 and 2.4. Similar data for a poisoned detector are shown

COINCIDENCE COUNTS IN 5 MINUTES

FIGURE 2.1
RADIAL PROFILE OF COINCIDENCE COUNTS

FIGURE 2.2
AXIAL PROFILE OF COINCIDENCE COUNTS

Table 2.1 - Plutonium Assay and Isotopic
Composition of PuO₂ and 25 w/o PuO₂-UO₂ Standards

Standard Designation	Pu Assay (%)	Plutonium Isotopic Composition (w/o)					Equivalent ⁽¹⁾ Pu-240 (%)
		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	
PuO ₂ - 1 thru 5	86.67	0.104	88.62	9.35	1.74	0.188	9.928
PuO ₂ - 6 and 7	87.40	0.030	90.15	8.55	1.19	0.080	8.759
25 w/o PuO ₂ -UO ₂ (1 thru 4)	21.76	0.090	88.62	9.34	1.75	0.196	9.895

(1) % Pu-240 + 2.6 x % Pu-238 + 1.64 x % Pu-242

Table 2.2 - Inventory Values of PuO₂ and 25% PuO₂-UO₂ Standards
(All Weights in Grams)

Standard Designation	Total Oxide	Pu	Pu-240	Equivalent Pu-240
PuO ₂ - 1	99	85.80	8.02	8.52
- 2	49	42.47	3.97	4.22
- 3	197	170.74	15.96	16.95
- 4	493	427.28	39.95	42.42
- 5	346	299.87	28.04	29.77
PuO ₂ - 6	1341	1172.0	100.2	102.66
- 7	2462	2151.9	184.0	188.48
25% PuO ₂ -UO ₂ - 1	3500	761.6	71.13	75.36
- 2	2243	488.1	45.59	48.30
- 3	1794	390.4	36.46	38.63
- 4	1106	240.7	22.48	23.82

Table 2.3 - Delays and Gate Lengths for Coincidence Neutron Counter⁽²⁾

Gate Length	<u>16</u>		<u>32</u>		<u>64</u>	
	x 1	x 2	x 1	x 2	x 1	x 2
Amplifier, Dead Time D, microseconds	2.125	2.125	4.25	4.25	8.50	8.50
Length of R&A Gates S, microseconds	16.25	32.25	32.5	64.5	65.0	129.0
Delay internal I, microseconds	66.63	130.63	133.25	261.25	266.50	522.50
(D + S)/S	1.131	1.066	1.131	1.066	1.131	1.066

Table 2.4 - Coincidence and Gross Counts of PuO_2 and 25% $\text{PuO}_2\text{-UO}_2$ for a Clean Detector, 1 Minute Count

Standard Designation	Equivalent Pu-240 (grams)	2 x 32 Gate		Coincidence One-Sigma (%)	2 x 64 Gate		Coincidence One-Sigma (%)
		Coincidence	Gross		Coincidence	Gross	
PuO_2 - 1	8.52	7,470	195,820	3.8	10,680	174,850	3.6
- 2	4.22	3,700	87,580	4.0	5,670	87,070	3.5
- 3	16.95	14,680	360,980	3.9	19,940	351,550	3.9
- 4	42.42	34,380	924,430	4.6	42,680	865,600	4.9
- 5	29.77	24,590	638,490	4.3	30,970	608,600	4.6
- 6	102.66	96,180	2,413,200	5.4	161,590	2,109,930	4.0
- 7	188.48	336,110	4,786,000	4.2	724,650	3,910,240	2.1
25% $\text{PuO}_2\text{-UO}_2$ - 1	75.36	61,390	1,730,800	5.5	85,540	1,556,920	5.1
- 2	48.30	39,960	1,085,300	4.8	48,830	1,008,740	5.2
- 3	38.63	31,700	855,820	4.6	38,870	806,740	5.0
- 4	23.82	20,200	526,100	4.2	26,790	506,400	4.3

Table 2.5 - Coincidence and Gross Count of PuO_2 and 25% $\text{PuO}_2\text{-UO}_2$
Standards in a Poisoned Detector (1 minute count)

Standard Designation	Equivalent Pu-240 (grams)	1 x 32		Coincidence One-Sigma (%)	2 x 32		Coincidence One-Sigma (%)	2 x 64		Coincidence One-Sigma (%)
		Coincidence	Gross		Coincidence	Gross		Coincidence	Gross	
PuO_2 - 1	8.52	1,020	66,800	7.7	1,646	66,540	6.5	2,040	66,670	7.2
- 2	4.22	480	33,390	8.7	803	32,770	7.0	890	33,440	8.6
- 3	16.95	1,970	134,600	7.7	3,678	133,990	5.7	4,040	134,100	7.3
- 4	42.42	6,020	342,900	6.4	9,320	343,250	5.8	10,940	342,400	7.3
- 5	29.77	4,070	238,400	6.6	6,467	237,880	5.7	7,170	237,680	7.5
- 6	102.66	15,990	876,900	6.7	23,300	878,170	6.4	31,130	875,840	7.7
- 7	188.48	40,810	1,687,400	5.7	49,030	1,688,460	6.7	99,690	1,707,500	5.9
25% $\text{PuO}_2\text{-UO}_2$ -1	75.36	11,200	627,200	6.5	16,144	628,480	6.4	19,630	625,600	8.1
-2	48.30	6,800	396,300	6.6	10,310	396,500	6.1	11,890	395,600	7.9
-3	38.63	5,070	312,900	6.9	7,540	311,500	6.5	9,310	311,290	7.7
-4	23.82	2,980	193,980	7.3	4,830	192,620	6.2	5,780	193,140	7.4

FIGURE 2.3

GROSS COUNTS OF PuO₂ AND 25% PuO₂-UO₂
STANDARDS FOR A CLEAN DETECTOR

FIGURE 2.4

COINCIDENCE COUNTS OF PuO₂ AND 25% PuO₂-UO₂
STANDARDS FOR A CLEAN DETECTOR

in Figures 2.5 and 2.6. The data in these figures clearly illustrate that no effect of matrix, PuO_2 and 25 w/o $\text{PuO}_2\text{-UO}_2$, on the gross or coincidence count is present, at least within the statistical uncertainty of the data.

Based on the above data, coincidence neutron counting of glove-box-generated scrap, containing up to 100 grams of Pu-240 in an oxide matrix, can routinely be counted in the coincidence neutron counter to determine plutonium content.

The above data also shows that the effect of inelastic scattering by the uranium and plutonium and elastic scattering by the oxygen in the oxide standards had little effect on the coincidence and gross counts.

The effect of the cadmium-poisoned detector is to reduce the coincidence counts by a factor of four and the gross counts by approximately 2.6. A cadmium poisoned detector is necessary when the form of the oxide is such as to contain impurities with high (α, n) cross sections. In general, elements whose atomic number is less than 17 will have a high enough cross section for 5.3-Mev alpha particles to produce neutrons by the (α, n) process. Elements such as lithium, beryllium, boron, and fluorine are just a few of the elements that have a high (α, n) yield.

FIGURE 2.5

GROSS COUNTS OF PuO₂ AND 25% PuO₂-UO₂
STANDARDS FOR A POISONED DETECTOR

FIGURE 2,6

COINCIDENCE COUNTS OF PuO₂ AND 25^w% PuO₂-UO₂
STANDARDS FOR A POISONED DETECTOR

3.0 Gamma Counting of Raffinate

During the scrap-recovery operations of ZPR-III fuel, certain raffinates from the NUMEC plutonium recovery plant were stored in 30-gallon stainless-steel drums. These drums were further contained in a 55-gallon drum, and the annulus was filled with Speedidri. The drums were rotated at four RPM and counted with a Ge(Li)-detector multichannel-analyzer system. The same procedure was followed as was used in counting 55-gallon barrels, containing one-and-one-half-gallon waste packages.⁽³⁾

After gamma counting, the amount of plutonium in each drum was determined by measuring the volume and specific gravity of the solution that was removed for further processing and obtaining a plutonium concentration by alpha-counting a representative sample. Following the removal of the liquid the drums were recounted. The net counts in the 384-Kev complex region was less than 500 counts.

The data are shown in Table 3.1 and Figure 3.1. In the figure, the least-square linear-regression equation is shown with the data of Drum 219 excluded from the calculation. The plutonium concentration of the drums is generally less than 0.2 grams of Pu per liter and probably accounts for the linear behavior of the data shown in Figure 3.1.

Table 3.1 - ZPR-III Thirty-Gallon Drum Raffinate Data

<u>Drum No.</u>	<u>Net "384-Complex" Activity (Counts/1000 Seconds)</u>	<u>Volume Removed (Liters)</u>	<u>Pu Removed (grams)</u>	<u>Concentration (grams/liter)</u>
LW-217	4329	90.3	17.99	0.20
LW-148A	2665	102.4	10.4	0.10
LW-219	2320	72.3	17.35	0.24
LW-152	2157	90.5	8.50	0.09
LW-201	1717	92.6	2.51	0.03
LW-096	1373	93.0	1.46	0.02
LW-102	1231	88.5	1.12	0.01
LW-172	887	88.9	1.39	0.02
LW-129	977	102.6	3.15	0.03
LW-202	862	87.8	2.55	0.03
LW-210	391	89.5	1.37	0.02
P-3422	878	97.2	1.54	0.02

FIGURE 3.1
ACTIVITY IN THE 384-KEV COMPLEX FROM
30 GALLON BARRELS OF ZPR III SCRAP RECOVERY RAFFINATE

4.0 Gamma Assay of Scrap

During this quarter, no additional work relative to developing a method to assay plutonium scrap by gamma counting was started, since the remaining funds would be spent on developing a method using the coincidence neutron detector. The data generated to date, and summarized in the previous quarterly report,⁽³⁾ pointed out some interesting facts.

Attenuation by the matrix material and plutonium in the container, is a significant effect and the use of a special container such as the annular source container previously described,⁽³⁾ in place of standard one-quart containers, may be necessary.

An attenuation measurement is necessary; and the method of making this measurement, whether one elects to use an external source, or an insitu type of measurement, is of little consequence. The use of thin absorbers, such as hafnium, on the face of the Ge(Li) detector should be avoided since they greatly reduce the desired sensitivity of the attenuation measurement, if low energy gamma rays are used, for example, the 129-Kev gamma ray from Pu-239.

REFERENCES

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