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Plant Instrumentation Program 2nd Quarterly Progress Report

July-Sept. 1970

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Plant Instrumentation Program

Second Quarterly Report
July-September 1970

Project Management

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Introduction

This is the second quarterly progress report covering work performed under the subject contract. A previous report, BHO-69-1, covers work performed during the quarter ending June 1970.

Progress during this quarter was concentrated on three areas of passive techniques (Gamma and Neutron) of the measurements of plutonium in glove box generated waste, hard scrap and exhaust filters.

1.0 Passive Gamma Assay of Plutonium Scrap

The activity in the "384-complex" region⁽¹⁾ from a source containing 76 grams of Pu-239 previously fabricated for the annular-container system was studied. The results, shown in Table 1.1, demonstrate that the density of the material in the annular container, shown in Figure 1.1, makes a considerable difference in the count rate. However, it should be possible to correct for this effect with the attenuation source, shown in Figure 1.2, positioned in the center of the annular container.

Table 1.1 - Attenuation Source Studies

	<u>Activity (I)</u> <u>(counts/100 sec.)</u>	<u>Relative</u> <u>Transmittance</u> <u>(I/I₀)</u>
Attenuation Source without Plexiglas Annular Container	18728	--
Attenuation Source in Empty Plexiglas Annular Container	17649	0.942
Attenuation Source in Annular Container with 1282 grams of UO ₂ , firmly packed (~8.3 cm high)	3678	0.208
Same as above but UO ₂ not firmly packed (~10 cm high)	5080	0.288

FIGURE 1.1
DIMENSIONS OF ANNULAR CONTAINER
FOR USE IN GAMMA ASSAY OF SCRAP.

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FIGURE 1.2
DIMENSIONS OF PLUTONIUM ATTENUATION SOURCE
FOR ANNULAR CONTAINER

The PuO_2 and mixed-oxide standards, prepared for the neutron counter, were counted in one-quart cans in three different conditions: as received from the vault, shaken up, and firmly repacked by tapping the container. The as-received and packed data show a fairly good linearity as can be seen in Figure 1.3. The net count, plotted against the net weight of the entire packed sample, also gave linear plots.

It is apparent from these results that the handling of scrap containers could be sufficient to introduce density variations that will significantly affect the accuracy of nondestructive gamma measurements. Therefore, accurate measurements by this method will not be possible unless attenuation-measurement methods can be developed to correct the results.

Eight mixed-oxide standards were prepared in which the Pu-239 content varied from 20 to 300 grams. In each the net weight was adjusted to 1200 grams with depleted UO_2 . The packed standards were counted in the annular Plexiglas container with and without the attenuation source. The data, without the attenuation source, when plotted as net counts vs weight of Pu-239 on a log-log scale, gave the linear calibration curve shown in Figure 1.4.

Using the packed, pure- UO_2 sample as a "standard" standard, values of the constants A and b in the formula

$$W = AI_u \left[\frac{I_K}{I_s - I_u} \right]^b$$

were determined to be 0.680 gram seconds per count and 0.7206, respectively. In this formula, derived earlier for the solid-waste studies,⁽¹⁾ W is the weight

FIGURE 1.3
THE EFFECT OF THE PACKING DENSITY OF PLUTONIUM IN
ONE QT. CONTAINER

FIGURE 1.4
GAMMA CALIBRATION CURVE
FOR ANNULAR PLUTONIUM SCRAP CONTAINER

Pu-239 in grams, I_u is the net activity of the mixed-oxide standard without the attenuation source, I_s is the net activity with the source, and I_k is the net activity from the source through the packed, pure UO_2 sample. Note that with this technique the attenuation of the unknown is not determined. The technique measures the difference in the attenuation between the unknown and that of a standard (pure UO_2).

Samples of various densities were prepared to simulate the worst possible conditions which could be encountered in scrap material. These samples were corn starch, silicon carbide, and lead shot. Five grams of PuO_2 were mixed with each. The slope of the packed data in Figure 1.4 provided the value for A; it was intended that the slope of a plot of $\log W/I_u$ versus $\log I_k/(I_s - I_u)$ would provide a value for b.

The equation above was transformed to

$$W = A I_k^b \left[\frac{I_u^{1/b}}{I_s - I_u} \right]^b$$

which yields a straight line plot through the origin, to calculate the values of A and b. Six cases were made of the data, and by computer techniques the best fit for each case was calculated. These six cases were, 1. using only the data from the four varied matrices, 2. using all the data, 3. using only data for top-packed mixed-oxide samples, 4. using only data points for loosely packed mixed-oxide samples, 5. same as case three but excluding data from the 965-gram-Pu-239, PuO_2 sample, and 6. same as case four but excluding the 964-gram-Pu-239, PuO_2 sample. The results can be seen in Table 1.2. A weight factor of $1/W^2$ was used in the reiteration.

Table 1.2 - Evaluation of Constants for Calibration Formula

<u>Case</u>	<u>A</u> <u>(Gram Seconds/Count)</u>	<u>b x 10⁻¹</u> <u>(-)</u>
1	0.61413	7.1995
2	0.57474	6.2845
3	0.60130	4.0462
4	0.58268	2.4882
5	0.61332	2.5130
6	0.57111	6.1127

A plot of case one and two can be seen in Figure 1.5. Only these two cases were plotted as all others fall between these two lines. The data indicate a slightly parabolic curve instead of a straight line. The datum for the 99.96 w/o PuO₂ sample (964 grams of Pu-239) did not fit any curve except a log-log plot of weight vs net counts where a linear relationship exists for all the data (Figure 1.4).

To determine if a difference in the particle-size distributions of the UO₂ and PuO₂ in the standards could be the cause of the deviation at higher PuO₂ contents, the particle-size distributions were measured. The average size for the UO₂ powder was 2.6 microns with a tap density of 3.6 grams per cubic centimeter; the values for PuO₂ were 2.5 and 5.4, respectively. While the particle size is essentially the same, the tap density of the PuO₂ is about 50% greater. If PuO₂ powder of significantly different particle size can be obtained, several standards, identical to the previous ones, will be made and the resultant data compared.

Data is being collected to investigate the possibility of circumventing the present procedure of obtaining attenuation corrections. The method would compare the ratio of the activities in the 129-Kev peak of Pu-239 to the 414-Kev peak of Pu-239. This ratio should change as the attenuating characteristics of the sample change, since the 129-Kev gamma would be attenuated more by the higher density materials. If the

FIGURE 1.5
CALCULATED INTENSITY VERSUS MEASURED WEIGHT OF
 $Pu-239$ FOR CONSTANTS IN TABLE 1.2

technique can be shown to be sufficiently sensitive, with an appropriate calibration curve the attenuation correction could be made simply and quickly with a single measurement rather than the two measurements required with the exterior attenuation source.

2.0 Passive Gamma Assay of Glove Box Filters

A method was developed for the nondestructive gamma assay of possibly-contaminated exhaust filters. Two sizes of filters are used in the Plutonium Plant operation, 12 x 12 x 18 inches and 24 x 24 x 24 inches. Usually, two of the smaller-size filters are placed in a 55-gallon drum for subsequent disposal. Sometimes the method of removal from the glove-box system limits a drum to only one of the smaller-size filters. The large filters are counted "as bagged" out of the system.

Four standard PuO_2 sources were fabricated by encapsulating one and five grams of Pu-239 in small plastic bags. Two such sets were fabricated. By varying the combinations eight different known weights of Pu-239 could be used as standards.

The standards were placed in different positions on the filter media. No attenuation measurement was made. It was assumed that the attenuation of the Pu-239 gamma rays would be essentially constant regardless of filter size since the filter containers are all of the same construction.

Virgin filters with varying standard sources at varying locations were counted at a source-to-detector distance of 54 inches with rotation at 4 RPM. The counting time was 1000 seconds. The resultant data can be seen in Figures 2.1 and 2.2 which are used as standard calibration curves for this method. The precision was determined to be 1.6% on the two-filter drums and 9.1% for a drum with one filter on end or for one large filter.

FIGURE 2.1
 NET COUNT VERSUS Pu-239 CONTENT FOR ONE 12" x 12" x 18"
 FILTER IN A DRUM AND A 24" x 24" x 24" FILTER.

FIGURE 2.2
NET COUNTS VERSUS Pu-239 CONTENT FOR TWO 12"x12"x 18"
FILTERS IN A DRUM

3.0 Nondestructive Gamma Assay of Plutonium-Contaminated Waste

In determining the amount of plutonium in glove-box generated waste contained in 55-gallon drums, it has previously been demonstrated⁽²⁾ that a bias exists between the sum of the results obtained from the separate assay of 1.5-gallon packages and their collective assay in the drum. A procedure, using a Ge(Li) detector, developed by NUMEC for the assay of packages^(1,3) was extended to steel drums containing 21 packages. The experimental setup consisted of counting the drum of packages at a detector-face-to-source centerline distance of 54 inches for 1000 seconds live time. The drums were rotated at 4 RPM.

To minimize the bias noted in the previous quarterly report, five drums, each containing twenty-one 1.5-gallon packages of real plutonium-contaminated waste, were prepared from twenty drums previously assayed. Each of the packages were classified according to attenuation characteristics as measured during the package-assay procedure. Within each drum all the packages were of similar attenuation density, but each drum was of different attenuation density. The data are shown in Table 3.1.

Table 3.1 - Comparison of Drum and Package Assay
When Packages Are Categorized

Package Attenuation ^(a) Range	Plutonium-239 (grams)	
	<u>Sum of Package Assays</u>	<u>Drum Assay</u>
.90-.99	13.32	15.38
1.00-1.05	5.76	5.90
1.06-1.10	12.33	13.67
1.11-1.20	13.71	9.94
1.20-1.30	8.63	9.35
	<hr/> 53.75	<hr/> 54.24

(a) Relative to a standard package containing two PVC bag-out bags.

Of the 53.75 grams of plutonium determined to be present by the package assay technique, 54.24 grams were found by the assay of the five drums, a difference of 1%. The 2σ uncertainty, determined from multiple assays of a single drum, was ± 4.6 grams. It is unfortunate that drums full of waste usually are not homogeneous as to the attenuation characteristics of the waste. Because of the extra expense that will be associated with classifying material to be drummed, errors will result from attempting to assay unsorted waste.

Measurements of the material in twenty-seven drums of packaged waste, assayed both by the package system and by the drum-assay system with no classification regarding package attenuation properties, showed that the sum of the Pu-239 content in the packages was 26% higher than found by assaying the drums (see Table 1 of Reference 2). The possible causes of the low bias were several. Some drums contained one or a few packages which were of high attenuation density and which contained most of the plutonium. While an integral correction was measured for the attenuation of the drum and contents, a single package of high density would not be sufficiently compensated for. The data in Table 3.1 clearly demonstrate that the bias in package versus drum assay of glove box generated waste can be eliminated if the drums contain packages of similar attenuation properties.

Some of the bias may have been caused by a systematic error uncovered in the package-assay technique. About one-half of the packages contained

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considerable void volume. As there was little plutonium contained in the package, the net result was that the plutonium was contained in a matrix of a density higher than if the plutonium and the waste matrix were uniformly distributed throughout the entire volume of the package; the measured package attenuation correction was artificially low because of the large void. Accurate package or drum assay will require that the packages be completely filled with waste.

4.0 Neutron-Coincidence Counting of Plutonium Scrap

Work during this quarter was directed towards trouble shooting several neutron coincidence counters. A chronological description of the events is given below.

The first instrument received from Brookhaven National Laboratory's Technical Support Organization of AEC-OSMM did not function due to a faulty logic circuit, and the system was returned to BNL. This instrument was a well detector with eight BF_3 tubes and a 128-microsecond gate.

A larger detector assembly that can accommodate 1.5-gallon waste packages as well as scrap containers was fabricated by BNL. The new neutron-coincidence counter has a detector well that is nine inches in diameter by ten inches high. It is constructed of polyethylene, lined with cadmium, and surrounded by twenty-four BF_3 tubes. A special Brookhaven designed logic is incorporated into the electronics of the system so that the time of the coincidence gate may be varied.

Paraffin filler plates were used to adjust the height of the scrap container in the detector well in an attempt to obtain a broader maximum in the vertical variation of detector efficiency. The paraffin increased the count rate as expected due to neutron reflection, but no appreciable broadening of the vertical efficiency was discernible.

Mr. M. Zucker of BNL found that the new system was not operating properly. Additional replacement components for the coincidence neutron counter were received from BNL and installed in the system.

Measurements were undertaken to determine the horizontal and vertical efficiency of the well detector. A one-gram Pu-240 sample, obtained from BNL, was used as the source. The isotopic analysis of this source is shown in Table 4.1. The efficiency data are plotted in Figures 4.1, 4.2, and 4.3. From Figure 4.1 the arithmetic mean, taken from one to eight inches above the bottom of the well, produced a relatively flat efficiency curve with a deviation from this mean of only 3.0%. The peak-to-minimum ratio along the seven inches of length is 1.11.

Table 4.1 - Isotopic Composition of 0.997-Gram Plutonium Source

<u>Isotope</u>	<u>Composition (w/o)</u>
Pu-238	0.016
Pu-239	0.955
Pu-240	93.760
Pu-241	0.699
Pu-242	4.560
Pu-244	0.009

Since the NUMEC scrap containers are of such height that their vertical position could be varied in the well only 1-5/8 inches, three efficiency curves were taken with standard samples at different heights. These data are shown in Figure 4.2. When any container was raised from 1/2 to 1-1/8 inches, the counting rate did not change. Consequently, the standard container position was chosen to be 1/2 inch above the bottom of the well.

Figure 4.3 shows the horizontal efficiency as measured with the BNL source. Since the central region that will be occupied by the scrap container is relatively flat, no significant error should be anticipated from an uncertainty in positioning the container.

FIGURE 4.1
AXIAL VARIATION IN COUNT RATE
FOR WELL DETECTOR

FIGURE 4.2
EFFECT OF CENTROID OF ACTIVITY
ON THE COINCIDENCE COUNT RATE

FIGURE 4.3
RADIAL VARIATION OF COUNT RATE

Prepared standards of varying amounts of Pu-240 were counted to obtain calibration curves. Gate-length settings of 64 and 128 microseconds were studied. The results are shown in Figure 4.4. The shorter gate length decreased the real and accidental activity and therefore the dead time of the instrument; but it substantially increased the difference between the real and accidental activities. Subsequent conversations with Dr. G. Weinstock of BNL revealed that the instrument's logic does not function properly with the shorter gate-length setting. A replacement logic unit for the neutron counter was received from Brookhaven National Laboratory. The unit was installed in the existing equipment, and its operating characteristics were determined.

The vertical efficiency of the detector was remeasured at five different locations with three different mixed-oxide standards. The data showed only very slightly decreased efficiencies at the top and bottom. In the worst case the average deviation from the arithmetic mean was only five percent.

All the standards prepared for the gamma-assay investigation were counted with the neutron detector. Because no Pu-240 isotopic analysis was available for this material at this time, the results were plotted against the known weight of Pu-239, assuming a constant ratio of Pu-240 to Pu-239. The plots were not linear. Dr. M. Zucher of BNL indicated that if the pulse width is greater than the dead time of the instrument, a non-linear condition should exist. Replacement components to correct the non-linearity were obtained, and accurate isotopic analysis are currently in progress for the material

FIGURE 4.4
NEUTRON COINCIDENCE ACTIVITY FOR 25 AND 100 %
PuO₂ STANDARDS

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Preliminary testing of the system indicates that all systems are operating satisfactorily and that counting of the calibration standards can begin.

5.0 Proposed Work for Next Quarter

The barrel and package assay of glove-box generated waste and the assay of filters is considered operational, and no additional development work will be required.

Work will continue on developing a method for the gamma assay of plutonium in hard scrap.

Work will also begin on neutron coincident counting standards of 25 w/o PuO_2 in UO_2 and 100% PuO_2 in one-quart cans.

The non-destructive gamma assay of plutonium nitrate solutions will begin during the next quarter. The method was developed by J. E. Cline, et al,⁽⁴⁾ at Idaho Nuclear Corporation.

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 bottles used at ARHCO (#10 gauge S.S. 0.134 inch thick) and the Atomic and Space Development Authority (ASDA) in New York (#14 gauge S.S., 0.0747 inch thick). Similar measurements will be made on plutonium nitrate product from the NUMEC Scrap Recovery Facility.

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