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**EFFECT OF INDUCED FISSION
ON PLUTONIUM ASSAY WITH A
NEUTRON WELL COINCIDENCE COUNTER**

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Special Materials and Services Division

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TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT.....	5
INTRODUCTION.....	5
THEORY.....	6
EQUIPMENT.....	6
PROCEDURE.....	7
CONCLUSION.....	10
REFERENCES.....	11

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	BNL Neutron Well Coincidence Counter and Associated Equipment	6
2.	^{240}Pu Calibration Curves for Neutron Well Coincidence Counter	7
3.	Comparison of Combined- to Single-plate Counts.	8
4.	Results of Inventory Measurements.	9
5.	Calibration Using NBS Isotopic Standards.	10

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ABSTRACT

This report describes an investigation of the neutron-multiplication effects in the Brookhaven neutron well coincidence counter designed for plutonium assay. The study was initiated when anomalous results were obtained after calibrating the counter with samples of plutonium having various ^{240}Pu contents. The data with the cadmium-lined counter indicate that multiplication contributes to the coincident count in three ways: (1) Source size variation from 2.5 to 40 g of ^{240}Pu in 10% ^{240}Pu causes 10-25% of the coincident neutrons to be from induced fission. (2) Induced fission is strongly influenced by the relative position of samples in the counter well. (3) Neutron moderation in the source produces a 10% increase in gross count and a 4% increase in net coincident count.

In extreme cases, if these multiplication effects are not taken into account, the apparent (i.e., measured) plutonium content can exceed the gross weight of the sample by more than a factor of two.

INTRODUCTION

This report describes experiments that demonstrate the presence of significant neutron-multiplication effects in the neutron well coincidence counter designed and built by Brookhaven National Laboratory.¹ This counter was supplied to the AEC Chicago Operations Office by the Technical Support Organization at Brookhaven National Laboratory. Argonne was asked to assist in developing counter-calibration and operating procedures for plutonium assay. This assistance was provided to the Chicago Operations Office through Manuel A. Kanter and Dale J. Henderson in the Argonne Safeguards Training Program, with the Special Materials Division providing calibration sources and technical support.

THEORY

Neutron-coincidence measurements are of interest for plutonium-fuel assay because they separate spontaneous-fission events, which produce several neutrons, from α -n events, which in contrast produce a single neutron. Plutonium alpha particles reacting with light elements in the plutonium compounds contribute significantly to the gross neutron count. However, these neutrons are eliminated by the coincidence measurement. The coincident rate therefore should be proportional to the spontaneous-fission isotopes in the sample. However, if the gross count from α -n reactions becomes too high, the dead time in the coincidence logic becomes excessive and prevents proper operation of the counter. Gross count rates of 800,000 counts/min appear to be the maximum for reliable counter operation. Also, increased multiplication effects in the sample may be brought about by induced fissions from α -n neutrons.

EQUIPMENT

The neutron well coincidence counter consists of 24 BF_3 tubes arranged in two concentric rings of 12 tubes each in a polyethylene moderator. The well is 10 in. high and 8 in. in diameter. Polyethylene inserts can be used to increase the moderator thickness and reduce the diameter to $5\frac{1}{2}$ in. Also, the well can be fully lined with cadmium to reduce the die-away time, count rate, and self-multiplication effects. The counter and associated electronic equipment are shown in Fig. 1.

Fig. 1

BNL Neutron Well Coincidence Counter and Associated Equipment. ANL Neg. No. 150-549.

The electronic equipment consists of a 3000-V power supply, a low-noise preamplifier, an amplifier with pole-zeroing and gaussian pulse shaping, three scalers (a gross scaler, a real scaler, and an accidental scaler), and the neutron-coincidence logic circuit. The 24 tubes are connected in parallel to a common preamplifier. The amplified signal is fed to the coincidence logic circuit, where the first received pulse activates this circuit. After a short delay to allow the amplifier to recover, the circuit opens the real gate during which coincident neutrons along with random neutrons are counted. Following an appropriate delay to allow the coincident neutrons to die away, another gate of exactly the same duration is opened to count random neutrons. The difference between the counts from the two gates is the net number of real-coincidence neutrons detected.

This number, however, must be corrected for circuit dead time in order to obtain a true reflection of the net count. The dead-time corrections are summarized by Sher.¹ All the detected neutrons are counted in the gross scaler. The coincident and random neutrons are counted in the real scaler, and the random neutrons are counted in the accidental scaler.

PROCEDURE

Because of the role played by our group in plutonium-fuel assay for the ZPPR program, we have available a large quantity of 3-8-in.-long plutonium-metal fuel plates and 6-in.-long mixed-oxide rods. These were ideal for calibrating the counter. These fuel elements have accurately known plutonium isotopic contents and accurate weights. In addition, many of them had been specially prepared as gamma-assay standards.

Calibration curves representing the net neutron-coincident count corrected for live-time errors were plotted against the effective ^{240}Pu content. These curves are shown in Fig. 2. The effective ^{240}Pu is a weighted sum of the spontaneous-fission isotopes of plutonium.* Based on what we assumed would happen, we were somewhat surprised by the large difference we noted between the coincident-neutron yield per gram of effective ^{240}Pu for the various materials.

Fig. 2

^{240}Pu Calibration Curves for Neutron Well Coincidence Counter

Plutonium plates containing 11.5% ^{240}Pu gave up to 40% more coincidence neutrons per gram of effective ^{240}Pu than identical-size plates containing 25.8% ^{240}Pu . Mixed-oxide rods containing plutonium with an isotopic

*The effective ^{240}Pu mass is defined as

$$M_{\text{eff}}^{240} = M^{240} + \frac{n^{242}}{n^{240}} M^{242} + \frac{n^{238}}{n^{240}} M^{238},$$

where M^i = actual mass of isotope i in the source ($i = ^{240}\text{Pu}$, ^{242}Pu , or ^{238}Pu), and n^i = number of spontaneous-fission neutrons emitted per second per gram of isotope i .

content identical to that of the 11.5% ^{240}Pu plates gave coincident-neutron counts up to 20% lower for the same quantity of effective ^{240}Pu . These differences could not be explained by absorption, because the total plutonium content of the 11.5% ^{240}Pu metal plates in Fig. 2 was several times that of the 25.8% ^{240}Pu metal plates.

Other laboratories using the counter had reported that self-multiplication was not a problem. However, several simple experiments, as described below, indicated that multiplication could be observed with the counter, even with relatively small samples.

Self-multiplication in the counter has been measured in three different ways:

1. Effect of ratio of fissile to spontaneous-fission isotopes. Two similar metal plates containing plutonium with 5.8% effective ^{240}Pu and 28.5% effective ^{240}Pu were counted individually in the cadmium-lined well. The ratios of fissile to spontaneous-fission isotopes were 16.2 and 2.5, respectively. The number of net coincidence neutrons per gram of effective ^{240}Pu was 28% higher for the plate with the low ^{240}Pu . Therefore, the net count was not proportional to the effective ^{240}Pu content.

Fig. 3. Comparison of Combined- to Single-plate Counts. ANL Neg. No. 150-545 Rev. 1.

in the gross count and 4.2% increase in the net coincident count was observed by inserting eleven 3/8-in.-dia, 6-in.-long Lucite rods into an array of seven 3/8-in.-dia, 6-in.-long mixed-oxide rods. The mixed-oxide rods contained 84 g of plutonium with 10 g of effective ^{240}Pu .

2. Effect of source thickness.

This effect was observed by counting a stack of 2 x 2 x 1/4-in.-thick plates in the counter and also by counting the same plates individually. The count obtained from stacks of two to seven plates was compared to the arithmetic sum of the plates counted individually. The difference was 8% for two plates and 26% for seven plates, as in Fig. 3. This effect was also observed by measuring identical 2 x 4 x 1/4-in. plates side by side and face to face. The total plutonium content of the two plates was 245 g, with 27 g of effective ^{240}Pu . The face-to-face configuration resulted in a 10% increase in net count in the cadmium-lined well.

3. Effect of neutron moderation in the source.

A 10.7% increase

The results of these and other observations indicate that the neutron interactions that take place in the neutron well coincidence counter

are complex. The net coincident neutron count is affected by the ratio of spontaneous fission to fissionable atoms, the geometry of the material in the well, and the amount of moderator in the source. Other problems, not directly demonstrated but associated with the above, are multiplication due to α -n neutrons and difference in fission cross section for ^{235}U , ^{239}Pu , and ^{241}Pu .

Fig. 4. Results of Inventory Measurements.
ANL Neg. No. 150-550.

Figure 4 shows inventory-measurement results for cans of plutonium containing 8.5% ^{240}Pu . The plutonium was in various chemical forms including oxide, metal, and nitrate. The calibration curve was obtained with 8.5% ^{240}Pu in alloy plates of the ZPPR type. As can be seen, the points are widely scattered

about the calibration line and, in some cases, measured values disagree by as much as 50% with the accepted values.

The above examples indicate that the BNL coincidence counter can present some real problems in obtaining reliable count correlations as later experimental measurements and theoretical calculations by Sher² verified. However, the counter has been successfully calibrated for plutonium-scrap samples by limiting the sample total weight to about 1 g. This was done to avoid the above-mentioned problems and take advantage of the relative insensitivity of the counter to sample position and self-absorption. NBS isotopic standards 946, 947, and 948 were used as calibration sources. These standards each contained 0.25 g of plutonium. As shown in Fig. 5, six calibration points were obtained from these three standards by using them in combinations. The standards contain 7.9, 12.1, and 18.3% ^{240}Pu . When the counter was used in this manner, it appeared capable of making unbiased measurements. For most samples, a relative standard deviation of ~5% could be obtained in 30 min. Because of the small sample size, the counter was used without the cadmium liner for maximum efficiency.

Fig. 5

Calibration Using NBS Isotopic Standards

CONCLUSION

A more complex mathematical model is needed for the neutron well coincidence counter. Unless suitable corrections for induced-fission effects can be made,³ many of the advantages that appeared to make this counter attractive for the assay of bulk materials and scrap disappear. Calibration of the counter by plotting net coincidence count against the effective ^{240}Pu will give correct results only if standards and unknowns are very small or are of similar geometry, isotopic content, moderator content, and α -n neutron output. Under these conditions, errors limited only to statistical-counting errors can be approached.⁴ Otherwise, large errors exceeding 40% can be experienced. A combination of the induced-fission effects described above probably explains several measurements made on cans of plutonium-bearing material that gave results indicating the plutonium content exceeded the gross weight of the can by more than a factor of two.

The coincidence counter offers some inherent advantages over current gamma-scanning methods. The radial and axial dependence of the count rate has been shown to vary by $<5\%$ on sources positioned ± 2 in. off-center in the well, whereas for gamma counting the intensity varies by the inverse square of the distance. Self-absorption, also is less of a problem with coincidence counting, in that the self-absorption for a fast neutron is approximately 60 times smaller than that for the 414-keV plutonium gamma ray. When ^{240}Pu sample masses are small and suitable standards are obtained, the neutron coincidence counter can be useful.

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