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FAST CRITICAL ASSEMBLY SAFEGUARDS

Summary Report
October 1978—September 1979

by

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PART I

APPLICATIONS OF NDA METHODS TO
VERIFICATION OF FCA PLUTONIUM INVENTORY

by

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ABSTRACT

The effectiveness of a neutron well correlation counter (NWCC) and a random driver (RD) for plutonium-containing item assay and loss detection has been studied. The items were 4" x 2" x 1/4" stainless steel-clad metal plates and 6" x 3/8" stainless steel-clad oxide rods, each in two types of containment. It was found that absorption by dummies increases one's chance of detecting substitution over the chance of detecting simple removal. In all the loss-detection tests, however, there was only one failure to detect a loss. The NWCC did not separate out (α, n) neutrons well enough that one could use a calibration made with plates to assay for rods. The RD was found to have minimal usefulness for the assay of irradiated plates.

PART II

NDA EQUIPMENT FOR FAST CHECKS OF ZPR-6 AND -9 FUEL CANISTERS

by

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ABSTRACT

A neutron-counting/fuel-weighting system has been developed to provide the capability to assay the ZPR-6 and -9 plutonium fuel canisters rapidly and accurately. This system makes feasible the in-line monitoring of fuel transfers from the storage vault to the fuel-loading hoods and vice-versa. To exploit fully the system's potential, the equipment is intended to be used in conjunction with an on-line computer having a fuel and fuel-canister data base.

PART I

APPLICATIONS OF NDA METHODS TO VERIFICATION OF FCA PLUTONIUM INVENTORY

by

G. H. Winslow and F. O. Bellinger

I. INTRODUCTION

The recent development of autoradiographic inventory verification techniques has, to a great extent, lessened the need for verification methods based on electronic radiometric means. The autoradiography method yields more information in less time with far less personnel radiation exposure than do coincidence- or gross-neutron-counting methods. Nevertheless, these latter methods are an important complement to the autoradiography method and should be developed to useful proportions. For example, not all fuel storage practices are amenable to the application of autoradiographic inspection--ZPPR fuel rods and some of the 1/8"-thick fuel pieces fall into this category. It is necessary to have back-up inspection methods for these items. Also, the application of the radiation counting methods offers corroborating evidence of the integrity of the overall fuel inventory when these methods are applied to a statistical sampling of the vault inventory. The objective of this subtask, then, is to explore how and under what circumstances the NDA methods may be applied to the verification of FCA plutonium inventories.

II. SURVEY OF METHODS STUDIED

Three nondestructive measurement devices were studied in this effort. These were: a random driver (RD) coincidence counter used passively, a neutron well correlation counter (NWCC), and a gross neutron counter that had been designed specifically for the measurement of ANL-East ZPPR-fuel storage canisters (see Part II). Of the methods investigated, the gross neutron counter appears to be most suited for the large-scale vault inventory verification task. Here, a rapid gross neutron count is combined with automatic canister weighing and identification via bar-code reading to yield a high throughput measurement system having very little programmatic impact. An on-line computer link provides automatic data reduction and recording, and, by comparison with previously stored information, a judgment regarding fuel content may be made rapidly. Because the neutron detectors are inherently insensitive to gamma radiation, the gross neutron counter is not affected by fission product build-up in FCA fuel having a recent reactor-use history. By contrast, the coincidence counter may be used for the verification of unirradiated fuel only. Some evidence is shown that, by requiring a four-fold coincidence multiplicity, some of the adverse effects of fission-product gamma radiation may be avoided; but this may be done only with a severe loss of counting precision. Attempts to shield the RD detectors from the fission-product gamma radiation were generally unsuccessful.

The neutron well correlation counter may be used to verify FCA

fuel; however, our investigation shows some complications involving the use of a calibration curve for, say, F-rods for the analysis of plate data, and vice versa. The purpose of the NWCC is to separate accurately a measure of the mass of ^{240}Pu or effective ^{240}Pu from a high gross neutron count, varying fractions of which are due to uncorrelated neutrons. Our investigation shows that such a separation was not as complete as desired. In addition, it was found that, in order to obtain a sufficiently small limit of error on the net coincidence count data, rather long counting intervals were required--in some cases, approaching 500 seconds.

III. REMARKS ON STATISTICS

Our general statistical procedures, applicable here mainly to the RD work, are described in some detail in Appendix I. The special calibration and assay procedure that was adopted for the NWCC is described in Appendix II. We repeat here, briefly, a few items for Appendix I that are, principally, matters of definition which are needed to follow the main text.

All the uncertainties to be quoted are at the 95-percent confidence level (CL), and statistical tests are made at this level. The notation LE (Limit of Error) is used for such uncertainties; LE(A) refers to the LE in the assay, A. The principal emphasis in this paper is on loss detection, in which case LE is a one-sided 95-percent CL uncertainty; i.e., the remaining 5 percent is all in one tail of the probability density function. For ordinary assay work we calculate a two-sided LE,

in which case 2.5-percent of the probability is in each of the two tails of the probability density function.

The statistical tests made here are on the null hypotheses that a container contains what it is supposed to. If it is concluded that the null hypothesis is not true when, in fact, it is, that erroneous conclusion is described as a Type I error, which is a false alarm. If it is concluded that the null hypothesis is true when, in fact, it is not, the erroneous conclusion is described as a Type II error. This represents the unwelcome failure to detect a loss.

IV. CANISTER AND ITEM DESCRIPTIONS

A. Plate Canisters

The ANL-East plate storage canister has a circular cross section and 18 radial storage slots. The height will accomodate an 8"-long 2" x 1/4" plate, so that it will hold eighteen such plates or, for these experiments, thirty-six 4" x 2" x 1/4" plates.

The ANL-West plate storage canister is a rectangular box. When the canister is lying flat and the cover is removed, attached to the base are found vertical pins between which plates can be stored. Space is provided for twelve 8" x 2" x 1/4" plates, or twenty-four 4" x 2" x 1/4" plates. When the cover is in place, the container is upended for insertion in either the RD or NWCC counters, so that the plates stand vertically in the counter chamber.

B. Rod Canisters

The ANL-E F-rod canister contains a honey-comb mesh into which rods can be dropped. Thus, the rods stand vertically either when stored or when being counted.

The ANL-W rod canister appears, from the outside, like the ANL-W plate canister. Inside there are no pins, however. Rather, there is a removable test-tube type of rack into which the rods are inserted. There are 16 rows of four rods each, so that up to 64 rods can be stored in one canister. When the canister is upended for insertion into the counter chamber, the rods are lying on their sides, with four rods in one horizontal plane.

C. Test Items

The plates used were 4" x 2" x 1/4" stainless steel-clad ZPPR plates with a Mo-U-Pu core. The rods were 6" x 3/8" stainless steel-clad rods, designated F-rods, with a mixed Pu-U oxide core. The Pu isotopic composition is nearly the same in each. In particular, each had about 11.5 weight percent ^{240}Pu . The uranium is depleted.

V. NEUTRON WELL CORRELATION COUNTER

A. Introduction

The circuitry now in use in this counter came from Brookhaven National Laboratory and has been described by Zucker.¹ We will repeat here only the equations necessary to our discussion. Although earlier exploratory work was done, the investigation reported here should still be considered preliminary, since further work would be required before we would claim to have determined definitively the full range of usefulness of such a counter.

B. Characteristics of the Counter

In the classic coincidence counter, a fission event is recognized if neutron detection occurs in two or more separate detectors within such a short time interval that they can be said to have occurred simultaneously. In a correlation counter the train of pulses from what is, in essence, a single detector is analyzed electronically to separate out those pulses which are correlated in time from those which are randomly distributed in time. Such problems as that correlated neutrons from induced fission are not distinguishable from those due to spontaneous fission, and others, are discussed by Zucker.¹ No attempt to correct for such effects is made here.

If q is the number of spontaneous fission events per sec in the sample, if $\langle \nu \rangle$ is the average of the number of neutrons, ν , per fission,

if ϵ is the absolute detection efficiency, and if the counting interval is t sec, the number of detected fission neutrons is $\epsilon \langle \nu \rangle q t$. If α is the ratio of all other sample-generated neutrons which are detected to the number of detected fission neutrons and if B is the background, the count in the G , or gross scaler, is

$$G = B + \epsilon \langle \nu \rangle q t (1 + \alpha) \quad (1)$$

The circuitry is such that a count

$$R = N + (\tau/t) G^2 \quad (2)$$

where τ is a gate width and

$$N = \frac{1}{2} \epsilon^2 \langle \nu(\nu - 1) \rangle f q t \quad (3)$$

is collected in a second scaler, called the R scaler. In Eq. 3,

$\langle \nu(\nu - 1) \rangle$ is the average value of $\nu(\nu - 1)$ and

$$f = e^{-\tau_1/\tau_0} (1 - e^{-\tau/\tau_0}) \quad (4)$$

where τ_1 is a short delay between the arrival of an initiating pulse and the opening of the gate of duration τ , and τ_0 is the die-away time. The latter is the time in which the neutrons present at a particular moment will decay to e^{-1} of their number due to capture and leakage.

Then the desired quantity, N , called the net count, is

$$N = R - (\tau/t)G^2 \quad (5)$$

Since N , as is seen from Eq. 3, depends only on the spontaneous fission strength, not on α , a measure of q exists independently of the presence of neutrons from (α, n) sources.

If, with calibration measurements on known sources, it is found that N , when plotted against the mass, exhibits positive curvature, one first supposes there are multiplication effects operating, and this might well be the case. On the other hand, if the value of (τ/t) is not known accurately, curvature in either direction could result when N is calculated from Eq. 5.

Various nominal values of τ and of t can be chosen by dial settings. The only time calibration we attempted was a single measurement of τ at a nominal value of 80 μ sec, with the result 79.81 μ -sec. With some τ - and t -dial settings, we have observed negative curvature in plots of N vs mass and, in particular, at the 80- μ sec τ -dial setting, have observed negative curvature with 10-sec counting intervals and, two weeks later, positive curvature with 100-sec counting intervals when the measured value of τ was used for calculation. The measurement of τ was done at the earlier time. It is not yet clear whether there is some long-time instability in the circuitry or there is other than a ratio of 10 between the two t -dial settings. Also pertinent to these comments is the fact that plots of G vs mass showed slight positive curvature for the 100-sec counts but slight negative curvature for the 10-sec counts.

We illustrate the effect of not knowing the true instrumental value of (τ/t) by the calculations shown in Table I. For this example we suppose the only spontaneous fission source in the sample to be assayed is 50 g ^{240}Pu and that $\alpha = 0.50$. We assume the calibrating material has $\alpha = 0.05$. The other parameters of the example are given in the table, where the primed values of $(\tau/t) \times 10^7$ are thought to be correct when N is calculated. We have taken the value of $\langle v \rangle$, as well as those to be mentioned later, and the normal approximation to the probability distribution of v , from Keepin.²

If the counts are as described earlier, we see from Eqs. 1 and 3 that G and N , respectively, should be linear with mass or, more conveniently for this work, with the number, p , of identical items inserted in the counter. As mentioned above, this was not the case for C. We do find, however, that $\bar{R}/p$ is linear with $\bar{G}^2/p$, as shown for 100-sec observations in Figs. 1 and 2 for plates and rods, respectively, in the appropriate ANL-W canisters. Here, the averages are over five separate observations made at each p . The intercept of such a line will be the value of N per item. For these observations, at least, it appears that, whatever the contribution from the G register to the R register, it can be subtracted out accurately to yield a value of N which is linearly related to the amount of fissioning material present, as described by Eq. 3.

C. Calibration and Assay Procedure

Details of the procedure used for calibration and assay with the

Table I. Erroneous assay caused by erroneous (τ/t) .

$(\tau/t) \times 10^7$	N_{calc}	Assay
8.0	18287.00	50.00
7.99	18666.81	50.52
7.97	19426.43	51.49
7.9	22085.10	54.35
7.7	29681.29	59.48

Parameters

ϵ	=	0.08	q	=	451.77 fiss/g sec
$\langle \nu \rangle$	=	2.2575	t	=	100 sec
$\langle \nu(\nu - 1) \rangle$	=	4.003	τ	=	80 μ sec
f	=	0.632

Fig. 1. Demonstration of linearity of $\bar{R}/p$ with $\bar{G}^2/p$ for plates in NWCC.

Fig. 2. Demonstration of linearity of $\bar{R}/p$ with $\bar{G}^2/p$ for rods in NWCC.

NWCC are to be found in Appendix II. Here, we define the numbers a and b , as used in the following text, by the equations

$$N = ap \quad (6)$$

$$\tau/t = b$$

where a , the parameter of particular interest is the net count per item. Then Eq. 2 becomes

$$R = ap + bG^2 \quad (7)$$

the basic equation on which the procedure rests via a modified least squares determination of a and of b .

D. Further Experimental Results

Most of the work reported here was done with ANL-W canisters. First to be mentioned are two preliminary tests.

A single 4" ZPPR plate was systematically moved through all possible canister positions to map the position-effect response. Although the scatter in the count data for a particular position was greater than that expected from counting statistics, there was no well-defined pattern to the collective responses. This means that a large position effect due to geometrical considerations was not evident. Statistically, however, it was more definite that an upper-vs-lower

position effect existed than an edge-to-center position effect.

Background measurements, i.e., with an empty counter chamber, were made with thirty-six 4" ZPPR plates in an ANL-E canister located near the counter. We found essentially no net coincidence count with that collection of 36 ZPPR plates placed alongside, or on top of, the detector assembly.

For measurements during canister loading, plates were loaded three at a time for the results discussed previously, and subsequently. Rods were loaded four at a time, i.e., layer by layer, or eight at a time, i.e., two layers at a time, when the ANL-W canister was used.

All the data described so far were taken with the ANL-W canisters. Five 100-sec counts were made at each value of p , including $p = 0$, for the data of Fig. 1; no background was included for the data of Fig. 2. The measurements on the rods and on the plates were separated by one day. As was mentioned, rod data were taken with 10-sec counts two weeks earlier, and closer to the time the single measurement of τ was made. These data were taken with an ANL-E canister at every four rods from $p = 0$ to $p = 64$.

For comparison of the results, of course, the response per gram is needed. In addition to the information about ν and its distribution, taken from Keepin,² we took spontaneous fission half-lives from the Plutonium Handbook³ to find effective ^{240}Pu masses. The numbers used are collected in Table II; further work would require more up-to-date

Table II. Data to calculate m_{eff}^{240}

Isotope	$\langle \nu \rangle$	$\langle \nu(\nu - 1) \rangle$	$T_{1/2}(\text{years})$
238	2.33	4.265	4.3×10^{10}
240	2.257	4.003	12.2×10^{10}
242	2.18	3.739	6.8×10^{10}

information, but these are sufficiently good for the present purposes.

With these numbers we find

$$m_{\text{eff}} = 2.929m_{238} + m_{240} + 1.733m_{242}$$

and

$$m_{\text{eff}} = 3.023m_{238} + m_{240} + 1.676m_{242}$$

for G and N, respectively. The resultant effective masses per item, calculated from updated compositions, are shown in Table III. Results for τ , a, errors in the latter, responses per gram per second, and so on, are shown in Table IV; the values of τ were found from those for b by accepting the nominal t-values as accurate. We reserve discussion of these, and other, results to the next section.

In Fig. 3 we show plots of $\bar{N}/p$ vs p. The values of N were calculated from the individual (R,G) pairs at each p, using the appropriate value of b, averaged, and divided by p. The error bars are $\pm$ one standard deviation as determined from the variance in $\bar{N}$. The lines drawn across the plots are at the respective values of a.

We made the following estimates to see if we were getting counts about as to be expected. In Eq. 4, for f, we assume that the pre-delay is small compared to the die-away time and that the latter is 80 μ sec for this particular machine. Then f is ~ 0.632 . With this, with the values of a, and with other numbers cited previously, as required, we

Table III. Effective masses of ^{240}Pu (g)

	For G	For N
Plates	14.852	14.845
F-Rods	1.421	1.421

Table IV. Count rate per gram ^{240}Pu .

Date	Material	t(sec)	$\tau(\mu\text{sec})$	a	s(a)	$a/t\tau_{\text{eff}}^{240}$	$s(a/t\tau_{\text{eff}}^{240})$	$100\frac{s(a)}{a}$
7/18/79	F-Rods	10	79.622	49.322	0.635	3.471	0.045	1.29
8/1/79	F-Rods	100	80.244	385.163	2.703	2.711	0.019	0.070
8/2/79	Plates	100	80.306	4344.513	14.779	2.927	0.010	0.34

Fig. 3. Average count rate per item vs the number of items in NWCC. Curve a is for rods in the ANL-E canister. Curves b and c are for rods and plates, respectively, in ANL-W canisters.

can estimate ϵ . Having this, and using initial slopes of G vs p , we can estimate α . These results are given in Table V.

We have made some count time sensitivity tests and some void and substitution tests with plates. We did not make new calibration curves for each of these, but used the curve made with 100-sec counts on a previous day merely by adjusting a , $s(a)$, and b as required by the various counting intervals used in these tests. For the sensitivity tests, five observations were made for 23 plates at $t = 10, 20$, and 100 sec. For the void and substitution tests, two 500-sec observations were made for a void at each of two positions in the upper layer and for a stainless steel dummy at each of two positions in the lower layer. Thus, there were, again, 23 ZPPR plates present in each of these tests. The results are in Tables VI and VII. The uncertainties are each at the 95% confidence level, but are two-sided, as for assay work, on the time-sensitivity tests and are one-sided, as for loss-detection work, on the void and substitution tests. The principal contribution to these uncertainties is from the counting required to get $\bar{N}$; the contribution from the calibration is minor.

We realize that a more pertinent substitution test for this counter would be made by using a dummy that gave a gross (α, n) count equal to the gross count from a ZPPR plate, but that would be a sizable undertaking.

Table V. Multiplication coefficient.

Date	Material	ϵ	α
7/18/79	F-Rods	0.07793	0.511
8/1/79	F-Rods	0.06887	0.607
8/2/79	Plates	0.07156	0.072

Table VI. Count time sensitivity.

t(sec)	Assay	LE ^(a)
10	23.64	1.73
20	23.20	1.64
100	23.15	0.46

(a) Two-sided 95% CL uncertainty

Table VII. Void and substitution tests.

Type	Assay	LE ^(a)
Void	22.96	1.52
Void	23.14	6.24
Sub.	22.59	0.12
Sub.	23.10	0.96

(a) One-sided 95% CL uncertainty

E. Discussion

The values of τ obtained by the procedure outlined in Section C are, of course, close to the nominal value of 80 μsec . The first one in Table IV, from data taken at about the same time that τ was measured to be 79.81 μsec , is lower than that value by 0.2 percent. The other two, determined on two different days two weeks later, are higher by 0.6 percent, but only differ by 0.08 percent. It should be noted that these latter two were determined with differently formed materials in the counter. It is not certain, of course, whether these differences in τ are real, or, if so, what the correct explanation might be, but it does seem probable that we have shown that the physical form of the sample material will not affect the expected mode of operation of the circuitry.

The most serious problem is the disparity between the values of (a/tm_{eff}) shown in Table IV. The disparity between the two F-rod experiments, done with different container geometries, is greater than between the two types of material, done in a more similar geometry. We interpose that subsequent work, not part of this project, done with F-rods in these two geometries, but in a different correlation counter, and for which multiplication corrections were made, gave a similar difference. That is, the net count rate per rod was higher by more than would be statistically explainable in the ANL-E honeycomb geometry than in the ANL-W test-tube rack geometry. Thus, we feel certain that the differences found here are due, principally, to differences in geometry, though not in a direction we yet understand. Because of the length of

the rods relative to the diameter of the well, and the fact that they were perpendicular to the axis of the well in the test-tube rack geometry, we would have expected the rate for that geometry to be higher.

Before discussing the errors in a , we refer to Fig. 3. From the fact that there is no apparent trend in the size of the error bars, it seems that $s(\bar{N})$ is proportional to p , rather than to $\sqrt{p}$, indicating that the observed $\bar{N}$ is not a Poisson variable. On the other hand, $s(\bar{N})$ does appear to be proportional to $\sqrt{t}$. The average value of $s(\bar{N})/p$ for the 10-sec F-rod data is 2.42, while that value for the 100-sec F-rod data divided by $\sqrt{10}$ is 2.60.

It is also apparent in Fig. 3 that there is a definite sinusoidal behavior in the residuals between a and $\bar{N}/p$ for the rod data, regardless of the geometry. This will increase the variance in a for that data beyond what should be expected. If we multiply each $s(a)$ by the square root of its weight and divide by the corresponding average value of $s(\bar{N})/p$, we find 40.63 and 37.81 for the two sets of rod data, but only 13.74 for the plate data.

In Fig. 3, we would expect a single minimum in the pattern of values of $\bar{N}/p$ if there were multiplication effects detectable beyond the statistical errors. Similarly, by our method of analysis, which extended over 356 effective grams of ^{240}Pu for the plates but only 91 grams for the rods, we would not expect to get the same value of τ for each material. Yet, we found values with these two materials that

differed by only 0.08 percent.

The time-sensitivity tests and the void and substitution tests were not made on the same day that the plate calibration used for them was made. The assay results indicate, then, that this should cause no major problem, comparable to that presented by the nonconstancy of (a/tm_{eff}) between plates and rods.

Although we cannot absolutely rule out the possibility that more sophisticated methods of data treatment which involve multiplication corrections might alleviate this problem, we do not presently believe that such corrections would make possible the direct use, for instance, of a plate calibration for the accurate assay of F-rods. We are faced with at least two problems. One is the possible presence of induced fission, expected to be proportionally greater in material with a high (α, n) background, which is indistinguishable from spontaneous fission. More serious, we believe, in spite of our not finding a large effect, is the fact that ϵ will depend on position within the well. As Zucker¹ has pointed out, this problem can be minimized by locating samples, whose volume is small compared to that of the well, in the center of the well, but, as he also pointed out, this is often impossible. Since the efficiency varies both axially and radially, the optimum location of samples of non-optimum shapes and, probably the development of high-precision correction factors should be of principal importance in further work on such counters.

VI. THE NEUTRON COINCIDENCE COUNTER--PASSIVE MODE

A. Introduction

For this work, a National Nuclear Random Driver (RD) was used in the passive mode, i.e., without the use of the interrogating neutron sources. This is a true coincidence counter. Some models can be set to respond to a count in any one of four detectors, or to (essentially) simultaneous counts in two, or three, or four of the four detectors. With the exception of observations made on an irradiated plate, all of the experimental work was done in the three-out-of-four (3/4) coincidence mode with the lead/boron shields in place.

In the passive mode, the principle response is to the spontaneous fission radiation from the even mass plutonium isotopes; although this instrument is designated as a neutron counter, it also responds, unfortunately, to gamma radiation. Thus, at the outset, problems with gamma radiation from recently irradiated Pu plates were expected, and were found.

Measurement data were collected, and analyses made, for the ANL-East radial-type plate canister and for the ANL-West plate and rod canisters. Unsuccessful efforts were made to characterize and model the response to various accumulations of FCA fuel rods and plates.

B. Experimental Results

B.1 Irradiated Plate Data

Comparative 100-sec counts were taken between an 8" x 2" x 1/4" irradiated plate and a similar unirradiated plate in both the 3/4 and 4/4 coincidence modes in the ANL-W canister. Fission products in the irradiated plate were produced during a 1500 watt-hour foil-irradiation run which lasted about 80 minutes. The plate was allowed to cool for one day after removal from the reactor, and the measurements were made, then, on days 1, 2, 3, 6, 10, 17, and 38. On the first day, the ratio of net counts for the irradiated plate to the unirradiated plate for the 3/4 mode was 2.19, while, on the sixth day, that ratio was still 1.14.

In Fig. 4 we plot the logarithm of the differences in gross 3/4-mode count between the two plates, ΔG , as a function of the number of days through 17 days. By 38 days, the average count for the irradiated plate turned out to be slightly less than that for the unirradiated plate.

From the sixth day on, the excess count decayed with about a 6.5-day half-life. The initial decay is complex, with half-lives less than a day. The average of all the gross counts on the unirradiated plate was 782.0. Using this, we can estimate the number of days after which, with these 100-sec counts, one would accept the hypothesis of no difference, with n observations on each, at a particular confidence level. One would accept that hypothesis if

Fig. 4. Decay of gross count in RD from an irradiated plate. G is the difference between the count from the irradiated plate and that from an unirradiated plate counted at the same time.

$$\Delta G \leq t \sqrt{\frac{1}{n}(2 \times 782 + \Delta G)}$$

where t is chosen according to the desired confidence level, and the other factor on the right is $\sigma(\Delta G)$. With $t = 1$ and 2 , and with $n = 3$ and 5 , we have found ΔG for equality in the above expression and, then, found the corresponding number of days from the decay curve. The results are in Table VIII. For instance, were one using averages of five 100-sec counts, in this RD,⁴ on a plate of this radiation history, without knowing whether it had been recently irradiated, for comparison with a plate known to be unirradiated, we would estimate that one would accept it, at the 2σ confidence level, as being unirradiated if it had been out of the reactor 16.5 days. We put the emphasis back where it belongs by restating the obvious; in the 3/4 mode one cannot assay for Pu content in an irradiated plate, with the usual effort one makes in assay work, until the plate has been out of the reactor for 15 to 25 days.

The fact is, of course, that expenditure of more counting time will increase the required cooling time, because the precision is improved and the criterion for accepting the null hypothesis of no difference between an irradiated plate and an unirradiated plate becomes stricter.

This is illustrated further by the data taken in the 4/4 coincidence mode where the counts per 100 sec are much reduced. Here the average of all gross counts on the unirradiated plate was only 25 instead of 782, and a smooth curve corresponding to that of Fig. 4 did

Table VIII. Days at which $\Delta G = t \sqrt{\frac{1}{n}(2 \times 782 + \Delta G)}$

n	t	1	2
3		20.7	14.1
5		1	16.5

not result. For instance, the average count on the irradiated plate was less than or equal to that for the unirradiated plate on days 10 and 17, nor did the ratios of counts drop smoothly for days 1, 2, 3, and 6. We found for the data of the first day that one would reject the null hypothesis of no difference at the 1 CL and would not reject it at the 2σ level unless $n > 8$.

From the data taken on days 1, 2, and 3, when the excess gamma activity is still fairly high, we find that going to the 4/4 mode does tend to accomplish the desired purpose. When going from the 3/4 mode to the 4/4 mode, the net count from the unirradiated plate is reduced by about 0.03, but the excess count is reduced by about 0.006. Thus, the ratio of the excess count to the unirradiated plate count in the 4/4 mode is 0.2 times that ratio in the 3/4 mode.

B.2 ANL-E Plate Storage Canisters

Again 4" x 2" x 1/4" plates were used, so that up to 36 could be stored in one canister. Reference 4 does not apply to the RD model used here or for work to be described in later sections.

The first measurements, made for use in what turned out to be unsuccessful modelling attempts, were of a sort of semi-integrated differential response curve. That is, a single plate was put in a canister and two 100-sec counts were made at each of eight table heights. If $\bar{G}$ is the average gross count and t is the table height, $\bar{G}$ accurately followed

$$\bar{G} = 1400.9 + 508.00t - 32.637t^2 \quad (9)$$

Four of the positions were below the position of maximum response and three were above; we take the parabolic symmetry to indicate that the aluminum table is not affecting the observation.

Subsequent canister loading and substitution experiments were made with three 10-sec counts at each point. The statistics can always be improved by the expenditure of more time; that expended here was adequate to map out the general effects for which we were looking.

All the canister loading observations were made by adding a single plate at a time. The lower tier was completed before plates were added to the upper tier. Two such loadings were made at a table elevation of 3-7/8" and, between these, one was done at a table elevation of 5-7/8". In every case there was a sharp break to a higher slope in a plot of gross count vs number of plates when plates were added in the second tier. Curve a of Fig. 5, made at a table elevation of 3-7/8", is a typical example; only every other point has been plotted. Some legs of some curves were slightly concave upward. We found no evidence of breaks in gross counts, however; that is, the count for 18 plates in the lower tier always lay on, within statistical variation, the upper tier curve.

From Eq. 9, if there is no multiplication, we would expect the first plate loaded at $t = 3-7/8"$ to give a gross count of 287.9 and, with a background of about 10 counts, that each subsequent plate would

Fig. 5. Loading curves for ANL-E plate storage canister in RD. For curve a, the lower tier is completely loaded before loading of the upper tier is started. For curve b, the upper tier is loaded over 18 SS dummies in the lower tier.

add 277.9 counts. For the second tier we would expect, with $t = 7-7/8"$, an addition of 327.7 counts per plate. In fact, however, the plot of gross count vs number of plates was slightly concave upward in the lower tier with an average slope of 327.3 counts per plate, and the upper tier curve was linear with a slope of 493.7 counts per plate, for the data taken the same day on which Eq. 9 was determined. Four months later, when loading was repeated at this table height, the plot of gross count vs number of plates had the same general shape, but the slopes were 457.6 counts per plate and 676.7 counts per plate for the lower and upper tier legs, respectively. This is the data shown as curve a of Fig. 5. We have no explanation for the considerably higher count rates.

From Eq. 9, one would find that, at $t = 5.78"$, $\bar{G}$ is the same at t as at $t + 4"$. The nearest available table height is $5-7/8"$. We tried this table height, hoping to get rid of the break in the slope; from Eq. 9, one would calculate slopes of 315.9 and 313.5 counts per plate for the lower and upper legs, respectively, although we expected these numbers to be reduced because an additional $3/4"$ of lead shielding was used in this experiment. The result, however, was that, while both legs of the curve were linear, the slopes were 369.1 and 501.9 counts per plate for the lower tier and the upper tier, respectively. Statistically, there is no difference between this upper tier slope and that found one week earlier, 493.7, when the table height was $3-7/8"$ and there was no additional lead shielding. Comparative substitution experiments showed assay results slightly closer to the true number of plates for the earlier work at $t = 3-7/8"$ than for this second test at $t = 5-7/8"$ with additional shielding. Thus, there was no particular

advantage to the latter set-up, and we returned to the former for more thorough analysis. We mention here, however, one specific test made at $t = 5-7/8''$ which is pertinent to later discussion. With four stainless steel (SS) dummies in the upper tier, as widely spaced as possible (at 0° , 80° , 180° , 260°) the gross count was 13069. With those dummies removed, the count went to 13351. These are averages of three observations, and, hence, the hypothesis that they are the same can be rejected at the 95-percent confidence level.

For the final set of observations, at $t = 3-7/8''$, a complete loading curve was run, assays were made for SS dummies in each tier separately and in both tiers, and counts were made during loading of the upper tier when the lower tier held 18 SS dummies. The latter results are shown as curve b of Fig. 5. This curve is slightly concave upward and has an average slope of 539.6 counts per plate. It should be noted that, at 18 plates, the count is about the same as for 18 ZPPR plates in the lower tier plus two in the upper tier. This is one of two cases where we noted an assay greater than the actual number of plates.

For the other substitution experiments we substituted one, two opposing, and four SS plates in each tier separately, and in the upper tier over four dummies in the lower tier. At four dummies, the arrangement was as described perviously. The results are shown in Table IX. Assays were made, of course, with the upper leg of the calibration. The test is made as for loss detection (see Section III). No Type-II errors were made, even at 34 plates with dummies in the upper tier when the normally calculated variance, which barely exceeded acceptable

Table IX. Stainless steel substitution in ANL-E canisters.

Tier	p	A	LE(A)	T	A'
Lower	35	34.83	0.21	0.96	34.47
	34	33.94	0.21	1.85	33.45
	32	31.68	0.20	4.12	30.84
Upper	35	35.14	0.21	0.65	34.48
	34	33.61	0.21 ^(a)	2.18	33.39
	32	31.43	0.19	4.38	30.96
Both	31	30.49	0.19	5.32	
	30	29.52	0.18	6.30	
	28	27.00	0.17	8.83	

Tier	The tier in which dummies were located
p	Actual number of ZPPR plates
A	Assay
LE(A)	One-sided 95-percent uncertainty in A
T	$36 - A - LE(A)$
A'	Assay for $t = 5-7/8"$
(a)	Ratio of normally calculated variance to Poisson variance on the count was 3.02, slightly exceeding the critical value at a 95-percent confidence level of 2.99. If the normally calculated variance is used, $LE(A) = 0.60$ and $T = 1.79$.

limits, is used. Nor would an $LE(A)$ of 0.60, rather than 0.20, have caused such an error at any point in the table. Equipment must be well maintained, however, so that excess variance is rare.

For comparison, assays made at a table height of 5-7/8" are included in the last column of the table. This set-up would be slightly preferable for loss-detection work as opposed to assay work since the smaller is the assay, the less likely is it that T will be less than zero.

In Fig. 6 we show the gross counts vs the actual number of plates for these substitution experiments. In all but one case, the counts lie below the loading, or calibration, curve, so that the assays made on the basis of this curve will be lower than the actual number of plates. Although this means that accurate assays cannot be made, it does enhance one's ability to detect loss.

The explanation of this effect must be that an absorption-modified multiplication process is occurring, even though many of the legs of these loading curves are linear, or only slightly concave upward. The steel dummies absorb, but contribute nothing to multiplication, as was seen also with the experiments at $t = 5-7/8"$ when removal of four SS dummies significantly enhanced the count. It was also seen that the gross counts were considerably greater than would be expected from Eq. 9. When it became apparent that there was this sort of interaction between plates, no further modelling attempts were made for this rather complicated geometry.

Fig. 6. Results of substitution experiments with ANL-E plate canister. The curve is the full canister loading curve that would be used for assay.

B.3 ANL-W Plate Storage Canister

The loading, or calibration, curves with every observed point plotted are shown in Fig. 7. The table height was 4-7/8" and, in fact, the set-up was the same for each of these, except that one inch of additional lead shielding was used for curve b. The latter was the only curve where the top point of the lower tier curve did not lie on the upper tier curve. Again, the lower legs are slightly concave upward and the upper legs are linear.

With substitution of a single SS plate at various positions, we obtained the results shown in Table X. In the position designations, A means the lower tier and B the upper tier. Within a tier, positions are counted across from 1 at one side to 12 at the other. The assay values were uniformly less than 23 plates, and, again, no Type-II errors were committed. (See Section III.)

B.4 F-Rods in ANL-W Canister

The calibration curve is shown in Fig. 8 where all observed points are plotted. The table height was 6-7/8". This curve provides an excellent illustration of the need for care in choosing a mathematical model. The discussion is based on information to be found in Appendix I.

A first glance at a plot of the points suggests a smooth curve, slightly concave downward, to which a second-order curve might be fit.

Fig. 7. Loading curves for ANL-W plate storage canister in RD. Curve a is with the normal set-up; additional lead shielding yielded curve b.

Table X. Stainless steel substitution in ANL-W canister

Position	A	LE	T ^(a)
No additional lead			
A-2	22.83	0.18	0.99
A-6	22.96	0.18	0.86
A-11	22.91	0.18	0.91
Additional lead			
A-2	22.59	0.21	1.20
A-6	22.71	0.21	1.08
A-11	22.87	0.21	0.92
B-2	22.62	0.21	1.17
B-6	22.42	0.21	1.37
B-11	22.81	0.21	0.98

(a) $T = 24 - A - LE$

Fig. 8. Loading curve for F-rods in ANL-W canister. The best fit turned out to be cubic in the number of rods.

With a second-order curve, one finds $(s_\ell/s_e)^2$ to be 1.38. Here, we have 14 degrees of freedom in the numerator and 36 in the denominator. For this, $F_{0.05}(14,36) = 1.89$, so that, by that test, the second-order curve would be acceptable. Yet the error on the coefficient of p^2 , 1.98×10^{-2} , is nearly as great as the absolute value of the coefficient itself, 2.07×10^{-2} , which would tend to indicate that a linear, first-order curve would be adequate. The curve is, however, clearly not linear. Further, the departure of the observed points from the curve is greatest at its upper end, where we need the greatest accuracy for loss-detection work.

The equation

$$G = B + R(1 - e^{-Pp})$$

which again has the background, B , and two other parameters, yields exactly the same value of $(s_\ell/s_e)^2$ as does the second-order curve.

For a third-order curve, however, $(s_\ell/s_e)^2$ drops to 0.62, a considerable improvement. All the coefficients are significant, and close examination shows that the slightly concave upward piece of the curve near the origin fits the data better than would a linear portion.

Loss tests were made by migrating a single hole in the second position from the edge of a layer, designated B, up through the layers. The results are shown in Table XI. Here, a single Type-II error was committed. Again, this work was done with three 10-sec observations for

Table XI. Hole migration in ANL-W rod canister

Position	A	LE	$T^{(a)}$
B-1	62.39	0.95	0.66
B-3	63.47	1.10	-0.57
B-5	62.38	1.05	0.57
B-7	62.17	1.04	0.79
B-9	62.76	1.07	0.17
B-11	62.61	1.06	0.33
B-13	61.62	1.02	1.36
B-16	61.77	1.02	1.21

(a) $T = 64 - A - LE$

each set-up; it is reemphasized that, over the long haul, one would expect LE to be reduced by the expenditure of greater counting effort, and the incidence of Type-II errors, though low throughout this work, to be reduced.

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4. This particular model of the National Nuclear Random Driver has a divide-by-ten circuit between the pulse detection circuitry and the scaler. Anyone checking for excess variance by the calculation of F-ratios should be wary of such a feature.

APPENDIX I

GENERAL STATISTICAL PROCEDURES

The methods described in this section are applicable to RD data, or wherever a Poisson variable is the observation of direct interest. Special procedures were adopted for the NWCC data; they are described in the section on that instrument and in Appendix II.

Recognizing that a Poisson distribution with a large expected value can be replaced by a normal distribution, we standardly make F-ratio tests on the ratio of the normally calculated variance, s^2 , to $\bar{G}$ for an unknown sample, where $\bar{G}$ is the average gross count. For some work we have used the larger of s^2 or $\bar{G}$. Here, however, if there are n observations of G , and if

$$s^2/\bar{G} \leq F_{0.05}(n-1, \infty)$$

where $(n-1)$ is the numerator degrees of freedom and ∞ is the denominator degrees of freedom, we use $\bar{G}/n$ as the variance in $\bar{G}$. Otherwise, we use s^2/n . Whichever is used, we will later call it s_s^2 . It is seen that we work at the 95-percent confidence level (CL) and that we assume that $\bar{G}$ is the true expected variance, with infinite degrees of freedom. Our circuitry and background working conditions are sufficiently good that this condition on the F-ratio is seldom violated.

When least squares analyses are made to determine the parameters of a calibration curve, the use of equally weighted observations means a tacit assumption that the variance is the same at every point. That is not the case with Poisson variables. Consequently, our regression programs weight an observation of a count inversely proportional to itself; each data point is read in separately rather than the averages of the observations made at each standard; and three estimates of the variance for an observation of unit weight are made. These are the maximum average count, $\bar{G}_{\max}$, the variance deduced from the pooled scatter of the individual observations at each standard about their own averages, s_e^2 (which also is independent of the regression model), and the variance deduced from the scatter of the averages about the regression line, s_ℓ^2 . For this work we have decided to only consider $\bar{G}_{\max}$, which has infinite degrees of freedom, and s_ℓ^2 , which has $f_\ell = \ell - q$ degrees of freedom, where ℓ is the number of standards used and q is the number of parameters being determined. Then if

$$s_\ell^2 / \bar{G}_{\max} \leq F_{0.05}(f_\ell, \infty)$$

we use $\bar{G}_{\max}$ as the calibration variance for an observation of unit weight, on the grounds that the true variance cannot be less than the Poisson variance. If that condition is not satisfied, we use s_ℓ^2 . Whichever is chosen, it will be called s_c^2 in what follows; when a corresponding t-statistic is to be chosen, it will be for ∞ , or $\ell - q$, degrees of freedom. Similarly, for a sample, the corresponding t-statistic will be for $(n - 1)$ or for ∞ degrees of freedom.

If the calibration equation is written as

$$G = G(\text{parameters}, g)$$

where g is a measure of the item that gives rise to the gross count, G , and if $\bar{G}$ is the gross count on an unknown item, g is replaced by the assay value, A , which must be found by solving the calibration equation for g . We choose to write the result as

$$A = g(\text{parameters}, \bar{G})$$

For the first-order curve, for example,

$$G = a + bg$$

$$A = (\bar{G} - a)/b$$

The measure, g , could be mass, or a number of identical items.

We find the variance in A by the usual error propagation formula. In doing so, we recognize that the contribution to that variance due to the calibration cannot be found by partially differentiating the expression for A successively with respect to the parameters, for they are not independent random variables. The successive differentiations must be with respect to each individual, independent observation of G , G_j say, made during the generation of the calibration curve. Then, remembering that the variance in G_j is $s_c^2(G_j/\bar{G}_{\max})$, the contribution

to the variance in A due to the calibration, which we call S_c^2 , is

$$S_c^2 = (s_c^2 / \bar{G}_{\max}) \sum_j \left(\frac{\partial A}{\partial G_j} \right)^2 G_j$$

It turns out that this can be written as

$$S_c^2 = (s_c^2 / L^2) W(A)$$

where L is the slope of the calibration curve at $g = A$, and $W(A)$ is a function of A that depends on the number of parameters.

Similarly, the contribution to the variance in A due to $\bar{G}$, the count on the item being assayed, is found to be

$$S_s^2 = s_s^2 / L^2$$

Then we find the 95-percent CL uncertainty in A, LE, from

$$LE = (t_s^2 S_s^2 + t_c^2 S_c^2)^{1/2}$$

where each Student's-t statistic is chosen for the 95-percent CL and according to the number of degrees of freedom pertinent to s_s^2 or s_c^2 , respectively.

For ordinary assay work we would use two-sided t-values. Here, however, the principal interest is in the detection of loss. That is, we wish to detect the fact, if it is a fact, that one or more items are

missing, possibly via substitution, from a supposedly full canister. For this we use one-sided t-values.

We make the hypothesis that the canister contains the number of items, p_0 , that it is supposed to. Then, if $p_0 - A - LE \leq 0$, it is concluded that p , the number of items in the container, is p_0 . If $p_0 - A - LE > 0$, it is concluded that $p < p_0$. If it is concluded that $p < p_0$ when, in fact, $p = p_0$, a Type-I error has been committed. This is a false alarm. If it is concluded that $p = p_0$ when, in fact, $p < p_0$, a Type-II error has been committed. This is more serious from the safeguards point of view. The Type-I error leads to a waste of time, but the Type-II error means failure to detect a loss.

The use of the one-sided 95-percent CL t-values, and the use of the Poisson variances even if the normally calculated variances are larger, as long as the F-ratio tests permit that, leads to as small values of LE as seems statistically reasonable. While this will tend to increase the false alarm rate, it tends to decrease the chance of making a Type-II error.

APPENDIX II

NWCC CALIBRATION AND ASSAY PROCEDURE

If one knew that the value of b [$= \tau/t$] had long time stability, and if that value were known sufficiently accurately from other sources that it could be treated as a nonrandom variable, it would be used in Eq. 5 to find N , or the several values of N , at each value of p and Eq. 6 used to find a and its variance by usual procedures. With the belief that the conditions for doing this are not well established, we have adopted an alternate procedure.

The slopes and intercepts of Figs. 1 and 2 are b and a , respectively. Their determination by the usual least squares procedures would appear to be illogical, however, since that would imply that G^2 is a nonrandom variable. In fact, both R and G^2 are random variables, but they are not independent. With the parameters used for the example of Table I it can be shown that, even when $\alpha = 0$, the ratio $G^2(\tau/t)/N$ is greater than unity for a sample containing more than about 7 grams of ^{240}Pu . Thus, while there may be small fluctuations introduced by the circuitry, the contribution to R is, principally, $G^2(\tau/t)$ for such a sample. When uncorrelated neutrons are present, that 7-gram limit becomes even lower.

To test the procedure we finally adopted, we wrote the converse of Eq. 7,

$$G^2 = -a'p + b'R$$

We used the data which led to Fig. 1, plus the background data, to find a and b and, conversely, a' and b' , by entering each of the five (R,G) pairs, separately, that were observed for a given value of p . The lowest point in Fig. 1, as mentioned in the text, appears to be an outlier. Nevertheless, we found the correlation coefficient to be very high;

$$\rho = \sqrt{bb'} = 0.999998$$

With these arguments as support, then, we use Eq. 7 to make a least squares determination of a and of b by the usual methods but, for variance determinations, take the resultant value of b to be a nonrandom variable and find the variance in a as though Eq. 6 applied.

We have found the variance in G to be comparable to what would be expected from counting statistics and, by extension, have found the variance in R to be about what we would expect. The variance in N , however, appears to be larger than would be expected. We recognize the possibility that our calculations of what we should expect could be in error, but also believe there could be a variance introduced by the circuitry that is sufficiently small to only show up when the difference between the two large numbers, R and $G^2(\tau/t)$, is found.

Given these facts, then, we have decided to make the regression analyses with equally weighted observations, to use only normally

calculated variances, and to use only that single estimate of the variance which is based on the scatter of the individual values of R about the least squares line modeled on Eq. 7. During the determination of the regression line represented by Eq. 7, all the quantities required to find a and its variance as though Eq. 6 had been used are calculated, so that a regression analysis need only be run once. Then the variance in a predicted assay value, p, due to the calibration, is

$$\frac{p^2}{a^2} s^2(a)$$

where

$$s^2(a) = \frac{v^*}{(n - 1) \sum \mu^2}$$

$$v^* = \sum_{i=1}^n \sum_{j=1}^{n_i} (R_{ij} - a\mu_i - bG_{ij})^2$$

is the sum of squared residuals, and

$$\sum \mu^2 = \sum_{i=1}^n n_i \mu_i^2$$

In these equations, n is the number of standards used, n_i is the number of observations made at the standard, μ_i , and R_{ij} and G_{ij} are the jth values of R and of G, respectively, observed at standard, μ_i .

To get the variance in an assay due to the counts taken on the unknown sample, we use the appropriate value of b to calculate N from each of the individual (R,G) pairs, and average those values of N . Then the contribution to the variance in the assay, A , due to the counting on the sample is $s^2(\bar{N})/a^2$.

If t_c is the value of Student's- t for the desired confidence level and $(n - 1)$ degrees of freedom, and if t_s is the value of Student's- t for the desired confidence level and $(m - 1)$ degrees of freedom, where $m(R,G)$ pairs were used to find $\bar{N}$, the uncertainty in p at that confidence level is

$$\frac{1}{a} [t_s^2 s^2(\bar{N}) + t_c^2 p^2 s^2(a)]^{1/2}$$

The discussion has been made on the basis that p is a number of identical items. If such items are used for calibration, knowledge of the mass of SNM per item can be used to convert it to a mass calibration, or, of course, the masses for individual standards can be used instead of p without altering the procedure.

NDA EQUIPMENT FOR FAST CHECKS OF ZPR-6 AND -9 FUEL CANISTERS

by

R. A. Scharping, G. K. Rusch and E. F. Groh

INTRODUCTION

Critical reactor facilities such as ZPR-6 and ZPR-9^{1,2,3} perform reactor physics experiments to assess reactor loading configurations and changes thereto. Typically, many changes involving fuel are made during relatively short periods of time. It is imperative that these changes be made quickly, with a minimum labor force and with minimum radiation exposure to the personnel involved. Therefore, if NDA of the fuel is required, it must be done in a manner which does not impact adversely on personnel doses or on reactor operations.

The system described herein is capable of providing a high degree of assurance that the material transferred from the vault to the reactor and vice versa is the appropriate fuel. The assessment can be made rapidly with minimum additional radiation exposure and effort.

The assessment is based upon a simultaneous twofold check on the fuel material. Both the neutron emission rate due to spontaneous fissioning of the ^{240}Pu in the fuel and the weight, or in effect the density, of the fuel are checked and compared with standard criteria for the particular type of fuel and with previous values associated with the canister of interest. The complete check requires about 30 to 60 seconds and can be done in parallel with the other operations taking place during loading changes.

BASIS FOR TECHNIQUE

The ZPR-6 and -9 plutonium fuel inventory includes 1/8 and 1/4 inch thick plates ranging in length from 1 to 8 inches. These include SEFOR, DOW, NUMEC and PU/Al alloy plates of various isotopic compositions.

The inventory also includes (Pu-U)O₂ rods, 3/8 inch in diameter and 6 inches long. The DOW and NUMEC fuel plates 4 inches and longer comprise the majority of the inventory and are the pieces used most often. Therefore, as regards NDA, this is the fuel of greatest concern.

In a proposed safeguards system,⁴ the plutonium fuel is stored in a locked vault in sealed canisters. The contents of a canister would be observed visually before sealing by two or more people representing different Divisions within Argonne National Laboratory. (The material flow is typically to or from personnel of the Applied Physics Division which operates the reactors from or to personnel of the Special Materials Division which stores and is responsible for the fuel. Personnel from both Divisions ensure that the proper fuel is placed in a canister.) Thus, reasonable assurance is provided that the material within the storage canisters appears to be fuel-like material.

The plutonium fuel used in the ZPR critical facilities was manufactured to relatively tight specifications as regards isotopic content and mass per plate. The variance in composition is sufficiently small so as to permit the detection of one missing plate, as presently stored in canisters, for all fuel sizes equal to or greater than four inches long; this type of fuel constitutes the vast majority of the inventory.

The neutron measurement is based on the fact that ²⁴⁰Pu undergoes spontaneous fission resulting in about 1000 neutrons per second per gram of ²⁴⁰Pu. This corresponds to a half life for fission of about 1.34×10^{11} years. The plutonium contains ~5% to 25% of the ²⁴⁰Pu isotope, thus ensuring a reasonably strong source for all fuel pieces; the smallest fuel piece contains at least 1 gram of ²⁴⁰Pu and the pieces of primary interest contain at least 6 grams of ²⁴⁰Pu. These quantities are easily measured.

The weight measurement is important because there are few materials available with a density close to that of uranium or plutonium which are neutron emitters; certainly none are available in sufficient quantities to permit counterfeiting of the fuel. Additionally, the uranium isotopes do not have high spontaneous fission rates. Thus the combination of density and neutron emission rate can be used to identify uniquely, the ZPR plutonium fuel.

Visual inspections of the material placed into the canisters coupled with seals on the canisters, and storage of the canisters in a locked vault provide additional backup for the weight/neutron NDA technique.

DESCRIPTION OF EQUIPMENT

Figures 1, 2, 3 and 4 show the weight/neutron assay station. The basic elements comprising the system are (1) the Mettler electronic scale which also supports the canister holder, (2) the benelex moderator block which is machined to house the canister, (3) four Reuter Stokes RS-P1-1613-144 BF_3 proportional counters, (4) a Scan-a-Matic bar code reader to identify the canister number, (5) four sets of precision pulse preamplifiers, amplifiers and discriminators, (6) a pneumatic system to drive the benelex support tables using precision linear bearings, (7) position interlocks and control switches and (8) a heavy, sturdy support table. The system is designed to be used with an on-line computer and an appropriate data base, but scalers and tabulated reference data may be used in place of the computer.

Operation of the system proceeds in the following manner. A sealed fuel canister enroute from the vault to the loading hoods is placed into the canister holder. The personnel-safety button-switches must be depressed in order for the two tables containing the Benelex moderator to close around

the canister. During closure, the bar code identifying the canister is read automatically; closure is effected within 5 seconds. Once the moderator is in place, the weight is obtained and neutron counting begins. The length of the count cycle can be varied, however 10 to 30 seconds is a practical range. If the weight and count data are within specified limits, the tables are separated automatically within 5 seconds. The canister number is read a second time during removal of the Benelex. Because of parity checks a wrong number is not displayed, however a missed reading can occur. The second reading increases the probability that the canister number is obtained to greater than 95%. If the number is not obtained a second closure occurs automatically to obtain the number. If the weight and count data are not within specified limits a second interrogation will take place to confirm the first data. Subsequent non-conformance is signaled via lights and an audible signal.

The Mettler scale is capable of reading up to 30 kg, has a sensitivity of 1 gram and is accurate to within 2 grams. There is a tare option which permits the automatic subtraction of the canister holder weight. The scale is capable of detecting one missing unit of the smallest piece of plutonium fuel. The counting system detects about 60 counts per second per gram of ^{240}Pu and is therefore capable of detecting one missing four inch fuel plate from a nearly full canister (~4.5 kg of Pu).

When the quantity of material contained in a canister is changed, the subsequent neutron emission rate is compared to the inventory average for the type of fuel involved to ensure that no counterfeit fuel has been introduced into the canister. This coupled with a visual inspection and the

proper weight ensures that the material in the canister is the appropriate fuel. Subsequent neutron count interrogations can be compared with the "canister-specific" neutron counting rate to obtain assurance that fuel diversion has not occurred.

SYSTEM PERFORMANCE

System performance is related to equipment performance and to fuel piece uniformity. Equipment performance is relatively easy to characterize. Fuel piece uniformity is more difficult to characterize because of the large quantity of fuel comprising the ZPPR and ZPR-6 and -9 shared inventory and the limited opportunity to assess it.

Several different tests have been run to determine the adequacy of the equipment for performing NDA on canisters containing plutonium fuel plates. Additionally, it has been determined that in a non-multiplying fuel configuration, the counters detect about 60 counts sec^{-1} ($\text{gm of } ^{240}\text{Pu}$) $^{-1}$ and that each of the four counting systems has a dead time of about 3 microseconds. No dead time corrections have been applied to the data.

One test was designed to measure the counter system sensitivity to fuel piece location within the canister. A specified fuel piece or pieces was positioned in one of each of the possible fuel storage locations and the neutron counting rate was determined; this was repeated for each position. The data in Tables I and II show that the counting rate is within $\pm 1\%$ of the average, for all positions. Table III shows the results obtained when a given fuel configuration, in a given canister, was counted nine times over a period of several weeks. The deviation observed is somewhat greater than would be expected from counting statistics alone, ranging to $\pm 0.3\%$ from the average. A one sigma deviation based on counting statistics alone without

accounting for the correlation introduced by the emission of multiple neutrons per fission or the multiplication effect due to the fuel would be about $\pm 0.04\%$.

Another test of equipment performance involved the removal of one fuel piece and the subsequent movement of the resulting hole to each possible canister location. The counting rate was obtained with the hole at each location. The results are shown in Table IV. Included in the table are data when the hole was filled and the canister cover was removed, and data when the hole was filled and the canister cover was in place. All counts obtained with the fuel piece removed were obtained with the canister cover removed. It is seen that removal of one of the thirty-six fuel pieces resulted in a decrease in neutron counts of about 3.5% compared to the 2.8% expected on basis of the proportion of fuel removed without considering multiplication effects.

Finally, the effects of neutron multiplication were investigated by adding fuel to a canister in a series of predetermined steps. Counting rates were determined at each loading step. The results are shown in Table V. Neutron multiplication effects increase the counting rate by about 34% for a full canister of 4 in long NUMEC fuel plates. Since the increase is monotonic with increasing numbers of fuel plates, a correction can be applied if necessary.

Fuel piece uniformity was assessed by measuring the neutron emission rate from various canisters prior to shipping the fuel to ANL-W. The equipment shown in Figs. 1-4 was not used for this assessment. Instead a preliminary model of this equipment which used less sensitive counters and less accurate canister positioning capability was utilized. Counting times varied from 10 seconds per canister to 30 seconds per canister. Background

levels varied significantly. An attempt to correct for background variations was made by taking counts before and after each canister count and averaging them. However, because of the large and sudden changes in background, only approximate corrections were possible -- this is evident in the data where inordinately large changes in counting rate occurred for some canisters. Although in these instances it is often known that the corrections were incorrect, the proper corrections are not known.

Even under the adverse conditions existing during these tests only a few of the DOW or NUMEC fuel canisters counts were more than the counts associated with 1/2 of a fuel piece outside of the expected count range, indicating that repeat counts would be practical and would probably have confirmed that no fuel pieces were missing. Better background control and real time background correction capability should improve this situation considerably.

Even so, SEFOR plates, (Pu-U) O_2 pins, and DOW and NUMEC plates less than 4 inches long present more of a problem in detecting one missing piece from an otherwise full canister because of non-uniformity of composition or the larger number of pieces stored in a canister. To obtain a high degree of assurance that no pieces are missing, the allowable number of these pieces stored in a canister would have to be reduced. Other means of performing NDA can also be used since these items comprise a small fraction of the fuel inventory; because these pieces are not used very often, the required NDA frequency is significantly less than for the more often used pieces.

Figures 5 through 20 show the canister count data. One can assume that if the count from a given canister is different by more than about 1/3 of the count from an average piece in the canister, a repeat count may be

necessary to ensure that no pieces are missing. The data indicate that few repeat counts would be necessary except for the canisters containing pins and probably small plates.

It will be necessary to determine the limitations of this system for small fuel pieces and pins. A background counting channel will be added to obtain background counts during the same interval of time that canister counts are obtained; this will improve significantly the background correction. Additionally, control of fuel locations relative to the NDA system location can be exercised when canister counting is in progress. Under achievable conditions, it may be possible to detect one missing pin or small fuel piece from a canister. If this cannot be accomplished, new canister designs may be investigated for pins and small fuel pieces.

TABLE I. Fuel Position Sensitivity

Date: 4/19/79 Fuel Type: 4 In. Num. ^{240}Pu Content: 11.6%
(1 Plate)

Counting Time: 30 sec

Background: Channel 1: 577 Channel 2: 378
Channel 3: 668 Channel 4: 416

Total Counts: 1396616.00
Average Counts: 77589.78

Position	Gross Counts/ 3 Trials	Net Counts/ 3 Trials	Normalized to Average Count
1	83972	77855	1.003
2	83739	77622	1.000
3	84424	78307	1.009
4	84106	77989	1.005
5	84357	78240	1.008
6	83223	77106	0.994
7	83520	77403	0.998
8	83076	76959	0.992
9	83337	77220	0.995
10	83109	76992	0.992
11	84122	78005	1.005
12	83109	76992	0.992
13	83414	77297	0.996
14	83241	77124	0.994
15	84410	78293	1.009
16	83974	77857	1.003
17	83354	77237	0.995
18	84235	78118	1.007

TABLE II. Fuel Position Sensitivity

Fuel Type: 4 In. NUM. ^{240}Pu Content: 11.6% (2 Plates)			
Counting Time: 30 sec			
Background: Channel 1: 545 Channel 2: 357 Channel 3: 631 Channel 4: 393			
Total Counts: 2816414.00 Average Counts: 156467.44			
Position	Gross Counts/ 3 Trials	Net Counts/ 3 Trials	Normalized to Average Count
1	162470	156692	1.001
2	162629	156851	1.002
3	163314	157536	1.007
4	162972	157194	1.005
5	162721	156943	1.003
6	160727	154949	0.990
7	161649	155871	0.996
8	161789	156011	0.997
9	162937	157159	1.004
10	161983	156205	0.998
11	160976	155198	0.992
12	161362	155584	0.994
13	161814	156036	0.997
14	163295	157517	1.007
15	163217	157439	1.006
16	161383	155605	0.994
17	162165	156387	0.999
18	163015	157237	1.005

TABLE III. Safeguards System Reproducibility

Fuel Type: 4 In. Num. ^{240}Pu Content: 11.6%			
Counting Time: 60 sec			
Background: Channel 1: 82 Channel 2: 82			
Channel 3: 78 Channel 4: 64			
Total Counts: 66287037.00			
Average Counts: 7365226.33			

Test Number	Gross Counts/ 3 Trials	Net Counts/ 3 Trials	Normalized to Average Count
1	7374478	7373560	1.001
2	7358546	7357628	0.999
3	7372414	7371496	1.001
4	7349474	7348556	0.998
5	7357832	7356914	0.999
6	7360810	7359892	0.999
7	7346686	7345768	0.997
8	7383957	7383039	1.002
9	7391102	7390184	1.003

TABLE IV. Fuel Removal Position Sensitivity

Fuel Type: 4 In. NUM. ^{240}Pu Content: 11.6% (2 Pieces at each Location Except One Piece at Hole Location)			
Counting Time: 10 sec			
Background: Channel 1: 10 Channel 2: 12 Channel 3: 8 Channel 4: 13			
Total Counts: 24069440.00 Average Counts: 1203472.00			
Position	Gross Counts/ 3 Trials	Net Counts/ 3 Trials	Normalized to Average Count
1	1199484	1199355	0.997
2	1201036	1200907	0.998
3	1200035	1199906	0.997
4	1200138	1200009	0.997
5	1198506	1198377	0.996
6	1199362	1199233	0.996
7	1198813	1198684	0.996
8	1198293	1198164	0.996
9	1200163	1200034	0.997
10	1199939	1199810	0.997
11	1200096	1199967	0.997
12	1200099	1199970	0.997
13	1198595	1198466	0.996
14	1198996	1198867	0.996
15	1197375	1197246	0.995
16	1198297	1198168	0.996
17	1200121	1199992	0.997
18	1199331	1199202	0.996
a	1240623	1240494	1.031
b	1242718	1242589	1.033

^aFull canister - cover removed.^bFull canister - cover in place.

TABLE V. Multiplication Data-Safeguard Assay Station Counter

Fuel Type: 4 In. NUM. ²⁴⁰ Pu Content: 11.6%							
Counting Time: 60 sec							
Initial/Final	Background: Channel 1: 689/0 Channel 2: 445/0						
	Channel 3: 780/0 Channel 4: 514/0						
Total Counts: 2280259.00							
Average Counts: 175404.54							
Test Number	Channel 1	Channel 2	Channel 3	Channel 4	Gross Counts/ 3 Trials	Net Counts/ 3 Trials	Normalized to Average Count Per Fuel Plate
1	47635	31921	47020	31522	158098	151017	0.861
2	90276	61888	96489	65382	314035	307156	0.876
3	142900	96216	139537	94792	473445	466769	0.887
5	242215	167143	235316	164182	808856	802584	0.915
7	330272	242144	336435	238599	1156440	1150573	0.937
9	435820	325853	433725	320801	1516199	1510736	0.957
13	609117	518179	609612	509919	2246827	2242174	0.983
18	808369	794977	814557	791394	3209297	3205655	1.015
22	1060564	969271	1082718	791394	4082473	4079641	1.057
26	1342637	1178657	1306770	1154908	4982972	4980949	1.092
30	1560125	1417103	1543194	1383187	5903609	5902396	1.122
33	1697526	1584952	1710967	1607978	6601423	6600817	1.140
36	1848630	1805944	1856783	1796163	7307520	7307520	1.157

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 11

Figure 13

Figure 14

Figure 15

Figure 16

Figure 17

Figure 19

Figure 20

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