

NUREG/CR-0975  
MLM-2695  
RS  
UC-15

**Nondestructive Assay Confirmatory  
Assessment Experiments: Mixed Oxide**

**John F. Lemming**

**Issued: April 30, 1980**

**MOUND FACILITY**

Miamisburg, Ohio 45342

operated by

**MONSANTO RESEARCH CORPORATION**

a subsidiary of Monsanto Company

for the

**U. S. DEPARTMENT OF ENERGY**

Contract No. DE-AC04-76-DP00053

8005270182

# Contents

	<u>Page</u>
SUMMARY . . . . .	3
INTRODUCTION . . . . .	4
DATA BASE DESCRIPTION . . . . .	4
NDA Measurement Control . . . . .	5
Data Elements . . . . .	7
GENERAL APPROACH . . . . .	8
Sample Sets . . . . .	8
Measurement Systems . . . . .	12
EXPERIMENTAL DATA . . . . .	13
Instrument Sensor Module . . . . .	14
Regression Module . . . . .	14
Correlation Counter . . . . .	19
Coincidence Counter . . . . .	20
Chemical/Calorimetric Calibration Materials . . . . .	20
Dynamic Sensor Module . . . . .	20
Control Module . . . . .	23
CONCLUSIONS . . . . .	25
ACKNOWLEDGEMENTS . . . . .	26
REFERENCES . . . . .	27
APPENDIXES . . . . .	29
DISTRIBUTION . . . . .	44

## Summary

The confirmatory assessment experiments demonstrate traceable NDA measurements of plutonium in mixed oxide powder using commercially available spontaneous-fission assay systems. The experiments illustrate two major concepts: the production of calibration materials using calorimetric assay, and the use of paired measurements for measurement assurance. Two batches of well-characterized mixed oxide powder were used to establish the random and systematic error components. The major components of an NDA measurement assurance technique to establish and maintain traceability are identified and their functions are demonstrated.

# Introduction

Nondestructive assay (NDA) can be an important part of a material control and accounting system. In general, NDA analyses provide more timely determination of material balances for diversion detection than do chemical analyses. NDA instruments have also been adapted to in-line and/or automated applications to minimize material handling and thus the opportunities for material diversion. In addition, NDA provides a means of control and measurement when chemical analysis would destroy a valuable product.

The confirmatory assessment experiments have been designed to provide a data base for evaluation of NDA measurement systems. This data base will supply the Nuclear Regulatory Commission with a technical basis for guides and regulations dealing with measurement and accounting requirements using NDA techniques.

The ability of NDA to meet the precision requirements for many current safeguards applications has been demonstrated [1,2]. However, the needed accuracy has not been demonstrated except in isolated cases [3]. Often the accuracy cannot be related to national standards or nationally recognized measurement systems through an unbroken chain of comparisons. The difficulties associated with providing traceability\* have limited the use of NDA measurements in material control and accounting systems [3,4].

---

\*"Traceability" means the ability to relate individual measurements to national standards or a nationally accepted measurement system through an unbroken chain of comparison [5].

The confirmatory assessment experiments demonstrate traceable nondestructive assay measurements for mixed plutonium-uranium oxide samples. These data fulfill all the requirements of the code of federal regulations part 70.57 (10 CFR 70.57) for measurements to be used in a material control and accounting system for special nuclear material (SNM).

The initial elements of the data base are the random and systematic uncertainties for calorimetric assay and for two spontaneous fission techniques: a neutron correlation counter and a neutron/gamma-ray coincidence counter.

In addition, the major components of a measurement control program, suitable to meet current licensee requirements, have been identified and demonstrated. In particular, dynamic calibration is introduced as a measurement control technique for providing and maintaining traceability for NDA measurement systems.

## Data base description

The data base is composed of random and systematic uncertainties for traceable NDA measurement systems. It is important to realize that traceability and the random and systematic uncertainties are properties of an overall measurement system, not of an instrument. The measurement system, in addition to the instrument, includes the reference materials, the measurement conditions, procedures, techniques, and calculations. For NDA instruments the system also includes a consideration of those sample parameters which might affect the instrument response.

The determination of the error components for NDA systems has been discussed by



Shea [6]. The total error has three contributors: measurement-to-measurement variabilities, item-to-item variabilities, and calibration uncertainties.

The measurement-to-measurement variabilities arise from a combination of factors which cause repeated measurements of the same item to differ.

The item-to-item variabilities arise from differences in the contents or packaging of items from the corresponding characteristics incorporated in the reference calibration standards.

The calibration uncertainties result from fitting imprecise data with approximate relationships and from using a common calibration curve for estimating the contents of a series of similar but not identical assay items.

In order to provide credibility for these error components, it is necessary to demonstrate their relationship to national standards or a nationally accepted measurement system and to demonstrate that the measurement system is in a state of statistical control. This is the purpose of a measurement control program.

## **NDA measurement control**

The measurement control program performs four major functions: qualification, standardization, calibration, and verification; it also provides documentation for each function.

Qualification of an NDA system requires demonstration of the suitability of the measurement for the particular samples being measured. The user must consider,

for example, operating procedures, environmental factors, operator training, and sample characteristics which could affect the NDA response. In particular, the estimator of the SNM content must be chosen and its relation to the SNM content must be established.

The standardization of an NDA instrument is demonstrated by comparing the measured response to a known stimulus. For example, a calorimeter can be standardized by measuring electrical heat standards or certified radioactive heat sources (ANSI N15.22 - "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Material Control") [7].

The calibration of NDA instruments is the subject of the American National Standards Institute N15.20-1975, "Guide to Calibrating Nondestructive Assay Systems" [8]. Calibration includes the establishment of the functional form of the NDA instrument's response to the SNM content of the sample and the determination of the uncertainties resulting from curve-fitting techniques.

Verification demonstrates that no changes have occurred which could invalidate the qualification, standardization, or calibration of the NDA measurement system. It is through verification that statistical control is established and individual measurements can be related to national standards or a nationally recognized measurement system.

Dynamic calibration is a systematic approach for establishing and maintaining traceability of NDA measurement systems. It includes a verification technique which assures the choice of appropriate

calibration materials, and monitors the measurement for changes which would necessitate recalibration.

A schematic which illustrates the dynamic calibration concept is shown in Figure 1. In this example, the inventory or flow is measured by the primary NDA instrument; a portion of the inventory is measured by the control instrument and returned to the inventory; a portion of that measured by the control instrument is assayed using chemical techniques. Chemical assay is used in this system as a part of the measurement control program for the control measurement. The amount of chemical analysis required in this plan is much less than required when using chemical assay as the control measurement.

The implementation of dynamic calibration in a measurement system can be simplified by using a systems approach and separating the functions or operations into four modules. They are the regression, the instrument sensor, the dynamic sensor, and the control modules (Figure 2). The functions of the first two modules are discussed in ANSI N15.20-1975 [8]. The

regression module is used to establish the functional form of the NDA instrument response to the SNM content; the instrument sensor module monitors the instrument performance and stability using working standards and, where necessary, monitors normalization standards to account for electronic instrument drift. In addition to these two functions, dynamic calibration provides a dynamic sensor and a control module. These two modules provide verification of the calibration curve.

The dynamic sensor is used to perform several tests on and between the measured responses from the primary and the control measurements. As a minimum, the primary and control measurements are checked to verify that they are within the range of the calibration curves, and the ratio of the responses is recorded on a control chart. The limits of error on the control chart are determined during calibration. In addition, the sum and the difference of the dynamic sensors for sequential samples are determined. The sums are recorded on a control chart with the control limits determined using the

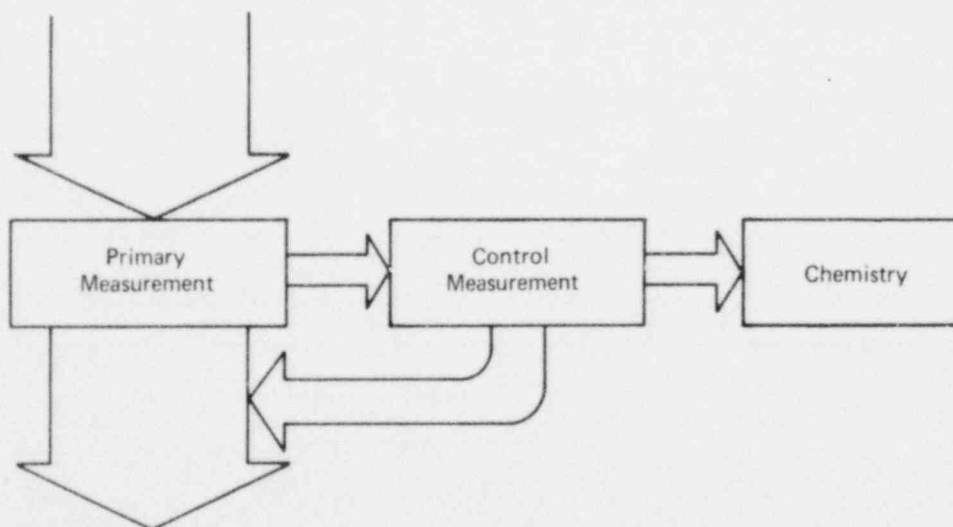


FIGURE 1 - A dynamic calibration system showing the relationship between measurements.

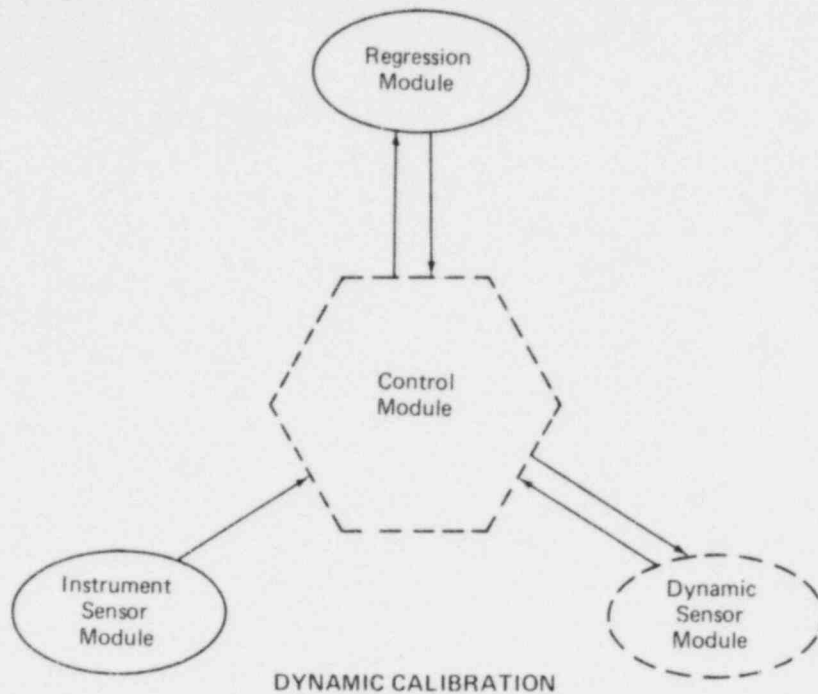


FIGURE 2 - A systems approach separating the four major operations into separate modules.

standard deviation of the difference data. This technique is used because the summed data enhance the systematic error while the difference data tend to nullify the systematic error, making determination of the random error components more apparent [9]. The control limits for these plots are also established during calibration.

The control module provides the necessary interfaces between the instrument operator and the other modules. In this module the calculations are performed to provide the SNM assay values and their uncertainties. This module also performs the calculations necessary to determine the number of paired measurements required for validation of the calibration curve at a stated confidence level.

## Data elements

The tabulated data for the data base are the random and systematic uncertainties of the measurements and the random uncertainties of the instruments. The data base also contains control charts from the instrument sensor module which indicate the instrument is in a state of statistical control, and from the dynamic sensor module which demonstrate that the measurement is in a state of statistical control.

The random error is the measurement-to-measurement variability. The contribution of the instrument to this uncertainty has been measured by making repeated measurements without removing the sample from the instrument. This component is labeled

repeatability. In addition to the instrument uncertainty, the random uncertainty of the measurement includes such things as geometry effects from repositioning the samples, counting statistics, and variations resulting from sample size. These combined components have been identified as the reproducibility, which has been determined by making repeated measurements on several samples throughout the experiment.

In general, the determination of the systematic error of an NDA measurement system is more complicated than determination of the random error. The systematic error is composed of two major components: the error from calibration and the errors from item-to-item variability. The data base contains the systematic errors from calibration. The uncertainty from item-to-item variation and the resulting mismatch between calibration materials and unknown samples must be accounted for to satisfy the requirements of 10 CFR 70.57. In these experiments, dynamic calibration provides the assurance that the calibration materials are appropriate and that the inventory characteristics have not changed in a way which affects the instrument response.

## General approach

Two carefully synthesized mixed oxide blends were prepared with different isotopic compositions and different plutonium concentrations. The chemical analysis techniques used to prepare the samples are traceable to the national measurement system. The blends were weighed into cans containing between 100 and 1000 g using certified balances. Sixteen samples were prepared from one blend and nine from the

second blend. Duplicate samples were prepared to assist in the determination of the error components.

Calorimetric assay and two spontaneous-fission assay systems were used to assay the samples. Each of the samples was measured at least three times with each NDA technique.

The spontaneous-fission assay system used a calorimeter as the control measurement to provide assurance that the item-to-item uncertainty did not change.

The components of the measurement error were evaluated by running the appropriate working standards and analyzing the calibration curves using the techniques outlined by Jaech [10]. The analysis-of-variance technique has been used to ascertain the stability of the measurement systems.

## Sample sets

The sample sets were prepared from blended plutonium-uranium oxide powder. Two isotopic mixtures with different plutonium concentrations were dried and blended in an inert atmosphere. Because of size limitations in the blender, one of the isotopic mixtures had to be split for blending. Six 15-g analytical samples were taken from each blend. The specific power of the mixed oxide (watts per gram of sample) was used to verify the homogeneity of each blend and, for the split sample, the blend-to-blend homogeneity. Each sample was calorimetered at least three times. The within-sample and between-sample variance data showed no inhomogeneity effects. Additional analytical samples which were analyzed for carbon or

total impurities also showed no inhomogeneities (see Appendix B).

After the homogeneity was verified, the mixed oxide was weighed into stainless steel liner cans (6.2 cm in diameter x 15.9 cm high) with quantities between 100 and 1000 g. The balances used are under Mound's Measurement Control Program and are traceable to the national measurement system. The liner cans were welded, removed from the glovebox, leak checked, and decontaminated. A dye penetrant test was used to check the welds. Then each liner can was loaded into a second stainless steel container (6.6 cm in diameter x 16.4 cm high) and welded. The secondary containment was leak checked and the welds were examined using the dye penetrant test. In order to assure safety and to give a base line for future safety checks, all containers were then radiographed. The resulting containers can be handled in radioactively "cold" areas and are compatible with several NDA instruments.

Traceable chemical analysis provided the "true" plutonium content for each bulk sample. When the calorimetric measurements of the analytical samples were completed, the samples were totally dissolved. Mass spectrometry was used to provide plutonium and uranium isotopic concentrations. Duplicate analyses were performed on each sample. Alpha pulse height counting was used to determine the  $^{238}\text{Pu}/^{239}\text{Pu}$  and the  $^{241}\text{Am}/^{239}\text{Pu}$  ratios. The plutonium and uranium concentrations were determined by coulometry. All of the chemical analyses have strict measurement control programs, utilize National Bureau of Standards reference materials, and are linked to a nationally accepted measurement system

through the Metals Exchange and SALE programs\*. A summary of the data with their 95% confidence limits is presented in Tables 1 and 2. March 1, 1978 was chosen as the common date to which all data have been decayed for comparison in this report. The "true" plutonium content and the measured mixed oxide weights are summarized in Table 3.

Duplicate samples at 1000 g and 230 g and a triplicate sample at 100 g were prepared from isotopic mixture A to facilitate random and systematic error determination. Similarly, duplicate samples were prepared at 110, 230, and 900 g from isotopic mixture B.

Calorimetric assay was used to provide an independent verification of the plutonium content of each sample. Power measurements were made on each of the bulk samples at least three times. The samples were measured in a random order. To convert the power measurement from watts to grams of plutonium, it is also necessary to measure the effective specific power (watts per gram of plutonium). In this application, the effective specific power ( $P_{\text{eff}}$ ) is analogous to the gravimetric factor used in chemical analysis.

---

\*The Plutonium Metals Exchange Program serves U.S. Department of Energy weapons laboratories in providing a mechanism for the improvement and standardization of plutonium analysis. The program is managed by Rockwell-Rocky Flats Plant.

The Safeguards Analytical Laboratory Evaluation (SALE) Program serves private and government laboratories in providing a mechanism for the improvement and standardization of safeguards measurements. The program is managed by New Brunswick Laboratory.



Table 1 - CHARACTERISTICS OF BATCH A<sup>a</sup>

Plutonium Concentration (wt %):  
19.226 ± 0.006

Plutonium Isotopics (wt %):

<sup>238</sup> Pu	0.0539
<sup>239</sup> Pu	86.7189
<sup>240</sup> Pu	11.7152
<sup>241</sup> Pu	1.3038
<sup>242</sup> Pu	0.2081
<sup>241</sup> Am	0.1824

Uranium Concentration (wt %):  
66.894 ± 0.017

Uranium Isotopics (wt %):

<sup>234</sup> U	0.007
<sup>235</sup> U	0.783
<sup>236</sup> U	0.002
<sup>238</sup> U	99.208

Carbon Impurity Analysis:

Blend A-1 - 664 ± 22 µg/g sample

Blend A-2 - 664 ± 12 µg/g sample

Total Impurities:

Element	Concentration (ppm)	
	A-1	A-2
Si	25	25
Al	30	30
Ni	150	150
Cr	60	50
Ti	<20	<20
Fe	680	680
Ca	<250	<250
Na	<250	<250
Co	<20	<20
Mn	<20	<20
Pb	<10	<10
Mg	<20	<20
Cu	<20	<20
Ag	<10	<10
B	<10	<10
Ba	<10	<10
Zn	ND <sup>b</sup>	ND

<sup>a</sup>Batch A contains 6 kg of mixed oxides.  
A common decay date of 3/1/78 was used.

<sup>b</sup>ND - Not detected.

Table 2 - CHARACTERISTICS OF BATCH B<sup>a</sup>

Plutonium Concentration (wt %):  
25.712 ± 0.009

Plutonium Isotopics (wt %):

<sup>238</sup> Pu	0.2430
<sup>239</sup> Pu	79.1160
<sup>240</sup> Pu	16.8230
<sup>241</sup> Pu	2.9263
<sup>242</sup> Pu	0.8916
<sup>241</sup> Am	0.6496

Uranium Concentration (wt %):  
59.996 ± 0.021

Uranium Isotopics (wt %):

<sup>234</sup> U	0.012
<sup>235</sup> U	0.876
<sup>236</sup> U	0.005
<sup>238</sup> U	99.107

Carbon Impurity Analysis:

1015 ± 15 µg/g sample

Total Impurities:

Element	Concentration (ppm)
Si	40
Al	35
Ni	35
Cr	30
Ti	20
Fe	100
Ca	<250
Na	<250
Co	<20
Mn	<20
Pb	<20
Mg	<20
Cu	<20
Ag	<20
B	<10
Ba	ND <sup>b</sup>
Zn	ND

<sup>a</sup>Batch B contains 4 kg of mixed oxides.  
A common decay date of 3/1/78 was used.

<sup>b</sup>ND - Not detected.

Table 3 - SUMMARY OF MIXED OXIDE AND PLUTONIUM WEIGHTS<sup>a</sup>

Batch	Can	MOX (g)	Pu (g)
A	31	99.86570 ± 0.00050	19.20018 ± 0.00599
	5	99.96470 ± 0.00050	19.21921 ± 0.00600
	19	100.10440 ± 0.00050	19.24607 ± 0.00601
	6	120.01450 ± 0.00050	23.07399 ± 0.00720
	23	150.02660 ± 0.00050	28.84411 ± 0.00900
	12	190.08000 ± 0.05000	36.54478 ± 0.01492
	11	230.00000 ± 0.05000	44.21980 ± 0.01682
	1	230.11000 ± 0.05000	44.24095 ± 0.01682
	27	280.19000 ± 0.05000	53.86933 ± 0.01937
	30	350.19000 ± 0.05000	67.32753 ± 0.02311
	15	430.17000 ± 0.05000	82.70448 ± 0.05754
	24	530.20000 ± 0.05000	101.93625 ± 0.03323
	35	650.16000 ± 0.05000	124.99976 ± 0.04018
	2	810.18000 ± 0.05000	155.76521 ± 0.04955
	8	1000.07000 ± 0.05000	192.27346 ± 0.06077
	17	1000.07000 ± 0.05000	192.27346 ± 0.06077
	B	20	110.01230 ± 0.00050
29		110.04500 ± 0.00050	28.29477 ± 0.00990
16		145.07160 ± 0.00050	37.30081 ± 0.01306
21		230.08000 ± 0.05000	59.15817 ± 0.02437
22		230.10000 ± 0.05000	59.16331 ± 0.02437
33		400.02000 ± 0.05000	102.85314 ± 0.03823
32		600.06000 ± 0.05000	154.28743 ± 0.05551
4		900.03000 ± 0.05000	231.41571 ± 0.08202
26		900.03000 ± 0.05000	231.41571 ± 0.08202

<sup>a</sup>Common decay date of 3/1/78.

The American National Standards Institute standard N15.22-1975 [7] describes two methods for determining  $P_{eff}$ . The first method, the empirical, involves two measurements: the wattage of a sample and the total plutonium content of the sample. The ratio of these two is  $P_{eff}$ . The second method, which is called the computational method, uses the measured isotopic concentrations and the heat fractions for each isotope determined from nuclear decay constants. The sum of the heat fraction for each radionuclide times its concentration is  $P_{eff}$ . This relationship can be expressed as:

$$P_{eff} = \sum_i R_i(t) P_i$$

where:  $P_i$  = heat fraction for radionuclide  $i$

$R_i(t)$  = weight percent of radionuclide  $i$  relative to total plutonium at time  $t$

The summation is over all the plutonium isotopes and  $^{241}\text{Am}$ .

The chemical analysis performed to determine the gravimetric factor, and the calorimetry measurements to ensure homogeneity, provide the data necessary to determine  $P_{eff}$  by both methods. In the computational method, the constants and procedures of ANSI-N15.22 have been used. Based on these determinations, there is a statistically significant difference between the empirical and the computational methods. At the 95% confidence level, the bias is +0.27% for Batch A and +0.20% for Batch B. These results are summarized in Table 4.



Table 4 - COMPARISON OF EMPIRICAL AND COMPUTATIONAL METHODS OF DETERMINING EFFECTIVE SPECIFIC POWER

Parameters	Empirical Results	Computational Results	
		ANSI-N15.22	ANSI-N15.22 <sup>d</sup>
Batch A (6 samples) <sup>b</sup>			
P <sub>eff</sub>	3.0548 W	3.0634 W	3.0608 W
S <sup>2</sup>	1.42 X 10 <sup>-5</sup>	2.54 X 10 <sup>-6</sup>	
Batch B (3 samples) <sup>b</sup>			
P <sub>eff</sub>	4.9313 W	4.9411 W	4.9388 W
S <sup>2</sup>	5.84 X 10 <sup>-6</sup>	1.73 X 10 <sup>-5</sup>	
Percent Difference = $\frac{\text{Computational}-\text{Empirical}}{\text{Empirical}} \times 100$ (at 95% Confidence Limit)			
Batch A	0.2684 ± 0.1539%		
Batch B	0.1987 ± 0.1298%		

<sup>a</sup>ANSI-N15.22 constants except T<sub>1/2</sub>(<sup>239</sup>Pu) = 24,119 yr.

<sup>b</sup>Duplicate analyses were performed on each sample.

This bias is consistent with the biases observed between the empirical and the computational methods for pure PuO<sub>2</sub>, in the Calibration Alternatives Experiment [11]. The nuclear decay constants recommended in ANSI-N15.22 may be contributors to this apparent bias. This hypothesis is supported by the reduction in the bias which can be obtained by using the revised <sup>239</sup>Pu half-life recommended by the Half-Life Evaluation Committee [12]. The biases using the new half-life (24,119 yr as compared to 24,082 yr) are also summarized in Table 4.

For the remainder of these experiments, the P<sub>eff</sub> determined by the computational method using ANSI-N15.22 [7] nuclear decay constants will be used. Since the plutonium isotopic composition is usually known, the computational method allows calorimetry to be used without additional

analytical measurements. Using the P<sub>eff</sub>, the mean difference (chemical assay minus calorimetric assay) is 0.0009 g for Batch A and 0.0004 g for Batch B.

The sample sets were used to demonstrate traceable spontaneous-fission assay measurement systems.

## Measurement systems

Two spontaneous-fission detection techniques have been evaluated: neutron correlation counting and neutron/gamma-ray coincidence counting. The neutron correlation counter is a neutron well counter manufactured by National Nuclear Corporation.\* This technique employs

\*National Nuclear Corporation, 3150 Spring Street, Redwood City, California 94063.

thermal neutron counters spaced symmetrically around a well cavity. The neutron/gamma-ray coincidence counter is an ISAS(P) (Isotopic Source Assay System operated in the passive mode). The ISAS(P) is manufactured by IRT Corporation.\* The coincidence counter consists of four plastic scintillators optically coupled to photomultiplier tubes.

The NDA measurement systems constructed about these two spontaneous-fission techniques utilized dynamic calibration to ensure the validity of the calibration curves. The control measurement chosen was calorimetric assay.

When considering spontaneous-fission detection, the instrument response is proportional to the effective mass of  $^{240}\text{Pu}$  in the sample ( $M_{240e}$ ). The equation used to determine the effective mass of  $^{240}\text{Pu}$  is

$$M_{240e} = \epsilon_{238} K_{238} M_{238} + M_{240} + \epsilon_{242} K_{242} M_{242} + \epsilon_{U238} K_{U238} M_{U238}$$

where the  $K_i$  are dimensionless constants derived from neutron emission characteristics, the  $\epsilon_i$  are relative coincidence counting efficiencies, and the  $M_i$  are isotopic masses in grams. Several sets of data could be used to calculate the  $K_i$ . For this experiment, the constants found in Regulatory Guide 5.53 [13] have been adopted. A comparison of the  $K_i$  determination from other data sets is given in Table 5, to illustrate possible inconsistencies in the calculation of the effective  $^{240}\text{Pu}$  mass.

Table 5 - COMPARISON OF CONSTANTS FOR  $M_{240e}$  DETERMINATION

$K_{238}$	$K_{242}$	Reference
$2.66 \pm 0.19$	$1.64 \pm 0.07$	13
2.60	1.62	14
2.43	1.69	15
2.41	1.80	16
2.89	1.88	17
2.6	1.7	ISAS(P) Manual
2.6	1.62	NWC Manual

For the ISAS(P) the relative counting efficiencies,  $\epsilon_i$ , are equal to 1. However, when a correlation counter with a shift register is used, the relative counting efficiency is not linearly dependent on the average number of neutrons emitted per spontaneous fission event. The average counting efficiencies relative to the counting efficiency for  $^{240}\text{Pu}$ , calculated by Krick, are 1.067 for  $^{238}\text{Pu}$  and 0.92 for  $^{242}\text{Pu}$  [15].

For samples with substantial quantities of  $^{238}\text{Pu}$  and  $^{242}\text{Pu}$ , the uncertainty in the constants can be the major contributor to the uncertainty in the effective mass of  $^{240}\text{Pu}$ .

## Experimental data

In order to more clearly identify the role of the measurement control program, the data will be discussed in terms of the systems approach to dynamic calibration (see page 6). The four major functions are performed in the instrument sensor, the regression, the dynamic sensor, and the control modules.

\*IRT Corporation, P.O. Box 80817, San Diego, California 92138.

## Instrument sensor module

The purpose of the instrument sensor module is to establish that the instruments are operating in a state of statistical control. This is accomplished by recording on a control chart the measured responses of working standards. As a minimum criterion, if the measured response for a working standard differs from its reference value by more than two standard deviations, the instrument performance should be considered suspect. When the measured responses are within acceptable limits, the instrument is considered to be in statistical control.

The control charts for the calorimeters are shown in Figures 3 and 4. The working standards are four  $^{238}\text{Pu}$  heat sources which span the range of the power output for both sample sets. The working standards were measured while randomly interspersed with the samples. The control limits for these charts are the 95% confidence limits established for each standard during the initial standardization of the calorimeters.

The working standards for the NWC and the ISAS(P) were chosen from the sample sets. Samples from both isotopic compositions were used. They were chosen such that their counting rates spanned the entire range.

In addition to the working standards, the ISAS(P) requires a normalization standard to account for electronic drifts. The sample with the largest counting rate was chosen to perform this function.

The control charts for the NWC and the ISAS(P) are shown in Figures 5 and 6,

respectively. The control limits for each of the working standards are the 95% confidence levels determined during the repeatability measurements. The top graph on the ISAS(P) chart (Figure 6) is the control plot for the normalization standard. The remaining curves have been corrected to account for the instrumental drift.

These graphs are summarized by the repeatability and reproducibility results reported in Table 6. The NWC repeatability varies from 0.7% to 0.4% for the smallest and largest samples, respectively. The ISAS(P) repeatability values vary from 3% to 1%. The reproducibility measurements range from 0.5% to 0.06% for the calorimeter, 1.4% to 0.5% for the NWC, and 5% to 3% for the ISAS(P).

## Regression module

The purpose of the regression module is to determine the calibration parameters which relate the instrument response to the SNM content of the item being measured. A detailed description of calibration functions and least-squares fitting procedures can be found in Appendix C of ANSI-N15.20 [8]. The uncertainties resulting from calibration were determined using the techniques outlined by Jaech [10].

In order to evaluate the effect of using calorimetric assay to produce calibration materials, the calibration curves for both spontaneous-fission assay systems have been generated using assay values for the standards determined both by chemical and calorimetric techniques.

Several treatments of the data from each spontaneous-fission detector were considered

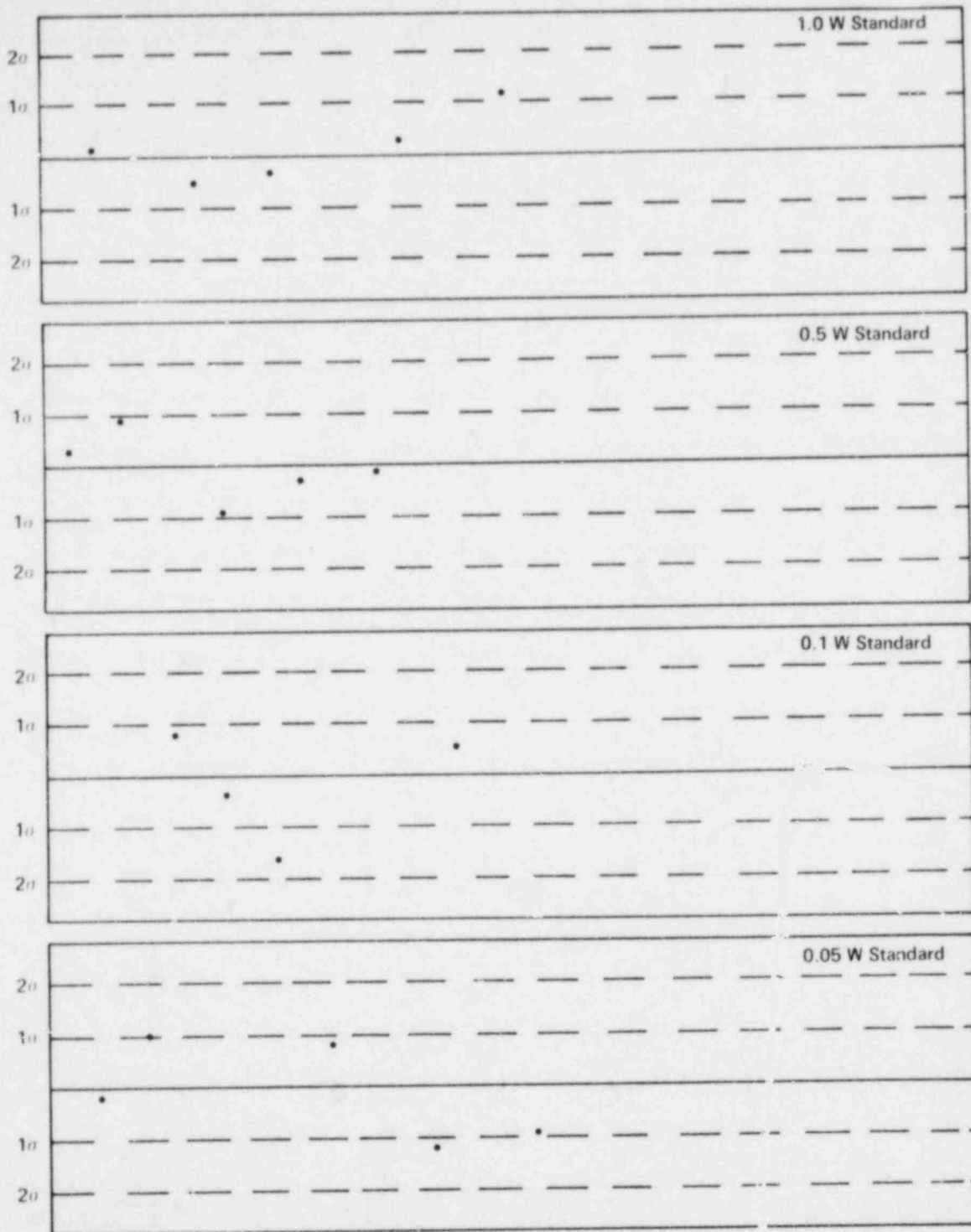


FIGURE 3 - Control charts for heat standards in calorimeter 1.

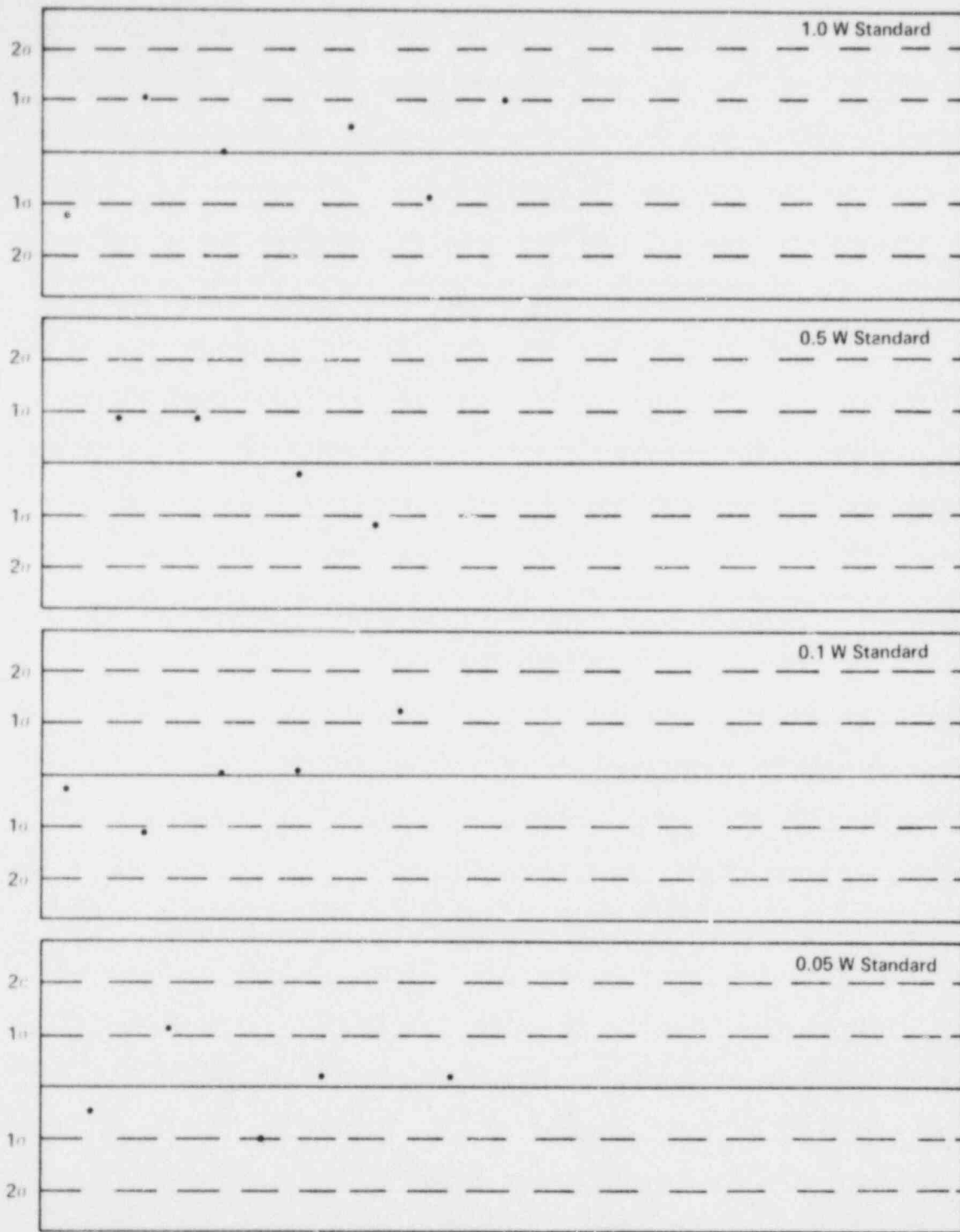


FIGURE 4 - Control charts for heat standards in calorimeter 2.

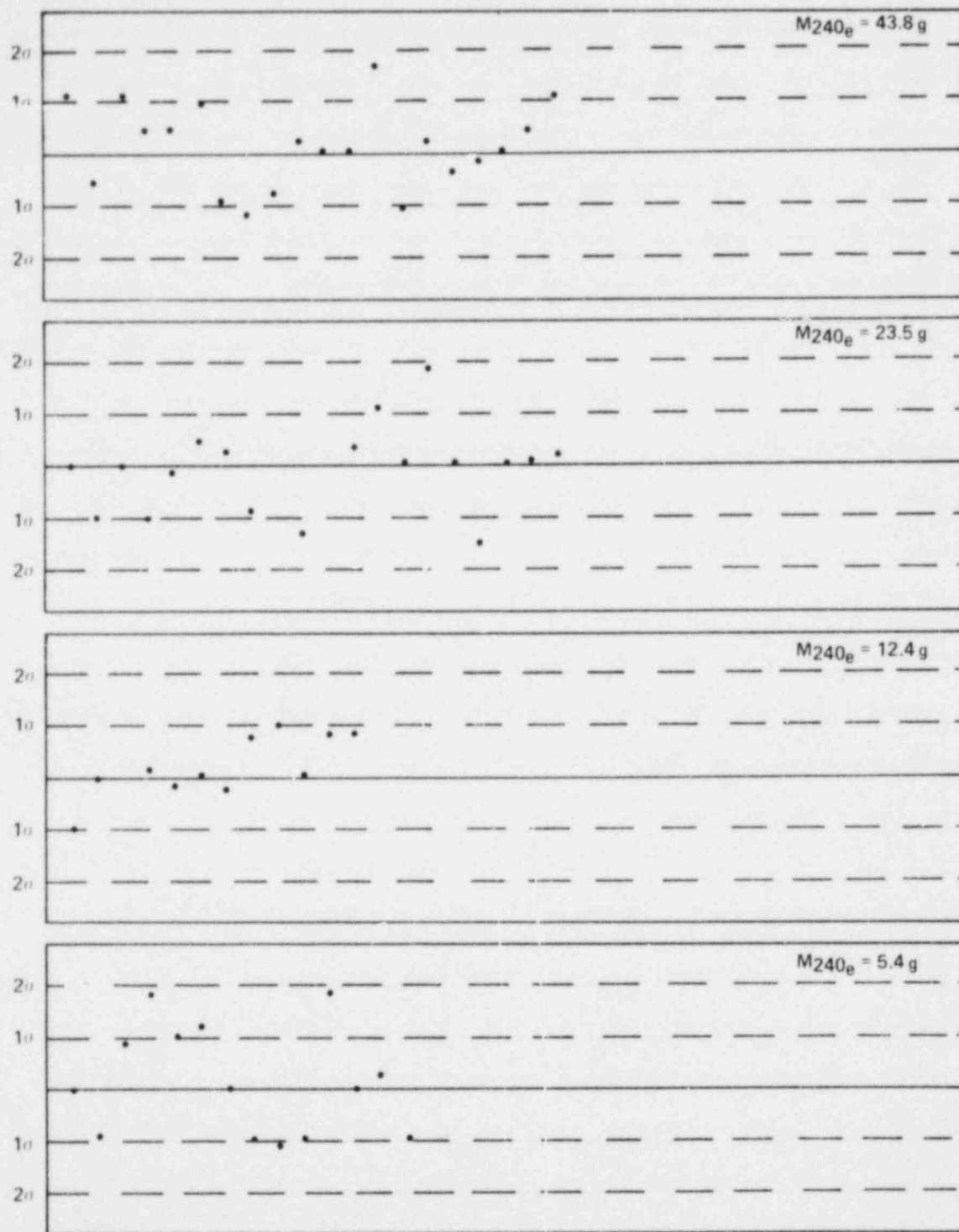


FIGURE 5 - Control charts for working standards in neutron well counter.



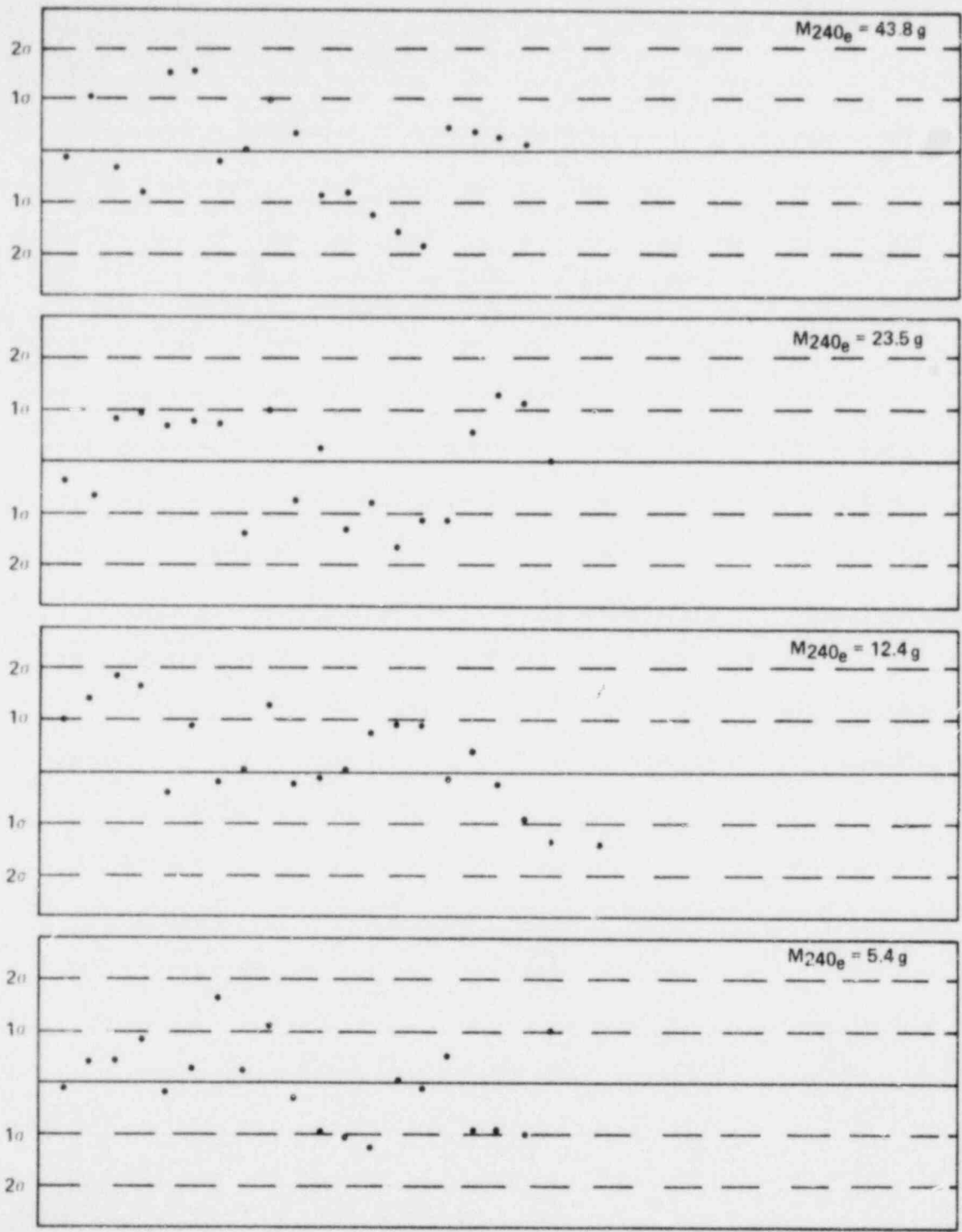


FIGURE 6 - Control charts for working standards in ISAS(P).



Table 6 - RANGE OF RANDOM ERROR COMPONENTS DETERMINED BY REPEATED MEASUREMENTS ON THE SMALLEST AND LARGEST SAMPLES

Instrument	Repeatability	Reproducibility
Calorimeter	-	0.5% - 0.06%
NWC	0.7% - 0.4%	1.4% - 0.5%
ISAS(P)	3% - 1%	5% - 3%

for constructing the estimators of the SNM content. The estimates were evaluated to determine which would provide the most reliable assay. The selection of the best estimators was based on plots of the residuals and determination of the random and systematic error components resulting from calibration.

Correlation Counter The measured quantities from the NWC are the gross (G) and the coincident (C) neutron counts. From

these measured values two estimators proportional to the fission rate have been calculated: the net neutron count (N) and the gross count squared divided by the net count ( $G^2/N$ ). Theoretically, the response of the correlation counter to both of these estimators should be linear [16,18].

In these experiments the linear fit to the net neutron data showed a systematic trend in the plot of residuals. In addition, the calculated random error components were larger than those determined by the reproducibility measurements. The quadratic model provided the best fit for those data. The departure from linearity is most likely caused in this particular instrument by a nonuniform efficiency over the counting volume. Figure 7 is a plot of the response of the counter to a 15-g MOX analytical sample placed at various heights along the axis.

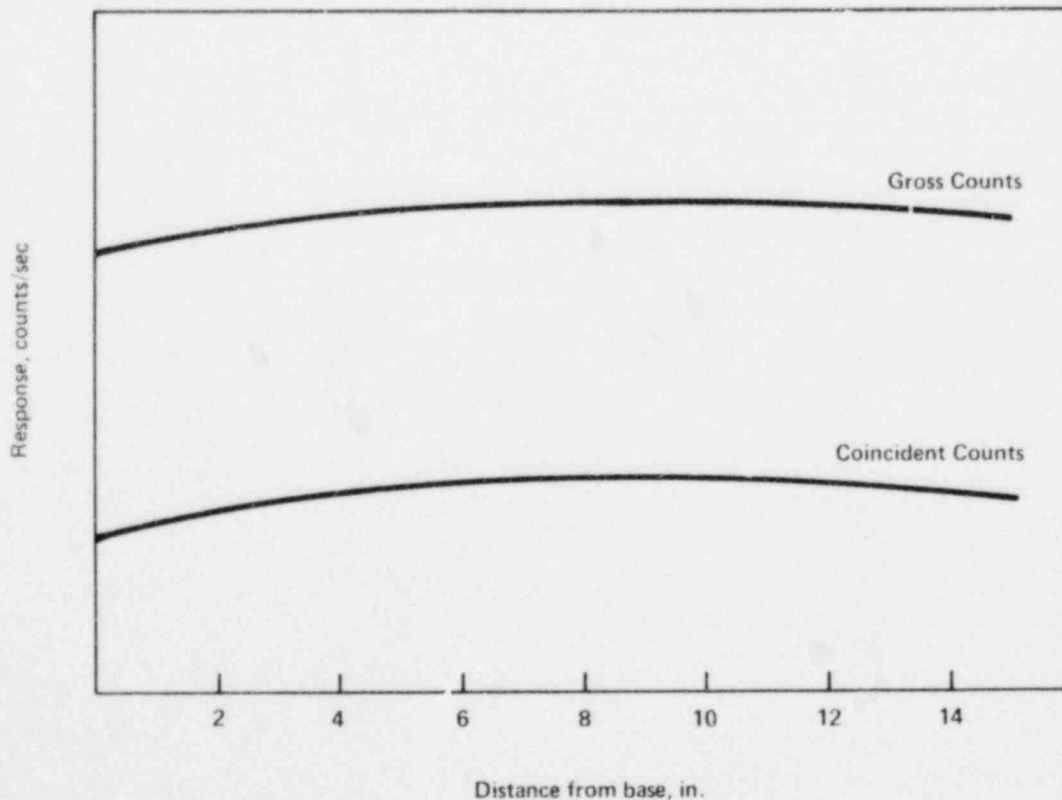


FIGURE 7 - NWC response curves for a 15-g MOX sample.

The  $G^2/N$  estimator, which is independent of efficiency [16], is properly represented by the linear model. Plots of the residuals show no systematic effects.

Coincidence Counter The measured quantities from the ISAS(P) are the number of counts in all four detectors (1/4), the number of coincidences in all sets of two of the four detectors (2/4), and the number of coincidences in all sets of three of the four detectors (3/4). The quantities evaluated as estimators for the SNM content are the (2/4) and (3/4) counts, both corrected and not corrected for accidental coincidences. The uncorrected counts were evaluated because if the accidental terms are related to the amount of SNM they increase the response of the system.

Initially a linear model was used for the calibration curve. In all cases, the residual plots indicate systematic trends. A more appropriate model for the ISAS(P) data is the exponential model. The residuals from this model are random.

#### Chemical/Calorimetric Calibration

Materials There are no statistically significant differences between calibration curves generated from the chemical or calorimetric assay values for the standards. This is illustrated by the data in Table 7 from the  $G^2/N$  estimator and in Table 8 by the data from the 3/4 corrected coincidence estimator.

A comparison of the random and systematic error components for each calibration curve is given in Table 9. These data demonstrate the potential of calorimetric assay as a calibrating technique for plutonium NDA.

## Dynamic sensor module

The purpose of the dynamic sensor module is to establish the continuing validity of the calibration curve. This is achieved through several tests on and between the measured responses from the primary and the control measurements.

For both spontaneous fission detection systems the control measurement is calorimetric assay. Thus, the comparisons are made between the measured quantities (counts and watts). In particular, the following dynamic sensors have been evaluated: for the NWC-CAL system, gross neutrons/watt and coincident neutrons/watt; and for the ISAS(P)-CAL system, singles/watt, double coincidences/watt, and triple coincidences/watt. A comparison was also made between the SNM assay values determined by spontaneous fission assay and by calorimetric assay.

In order to evaluate the dynamic sensors, the samples from Batch A were used to prepare a calibration curve. During a subsequent measurement of the Batch A samples, samples from Batch B were submitted for assay. For these comparisons the data are treated as if the operator did not know the start of a new isotopic batch.

The most sensitive dynamic sensor for both measurement systems is the difference between the primary and the control assay values. For the NWC-CAL system the difference changes from a mean value of  $-0.1 \pm 0.6$  to greater than 3. For the ISAS(P)-CAL system the difference changes from a mean value of  $-0.2 \pm 1.6$  to greater than 32.

Table 7 - CALIBRATION PARAMETERS FOR THE  $G^2/N$  ESTIMATOR

$$R = a + bM_{240}e$$

Batch	Determined by	Calibration Parameter			
		a	S <sub>a</sub>	b	S <sub>b</sub>
A	Chemistry	1080	100	4944	24
A	Calorimetry	1140	100	4956	24
B	Chemistry	1158	328	5606	36
B	Calorimetry	1194	328	5610	36

Table 8 - CALIBRATION PARAMETERS FOR THE 3/4 ESTIMATOR

$$R = a(1 - e^{bM_{240}e})$$

Batch	Determined by	Calibration Parameter			
		a	S <sub>a</sub>	b	S <sub>b</sub>
A	Chemistry	-1.91	0.38	0.011	0.002
A	Calorimetry	-1.97	0.41	0.010	0.002
B	Chemistry	-8.84	2.7	0.0045	0.0013
B	Calorimetry	-8.99	2.8	0.0044	0.0013

Table 9 - COMPARISON OF RANDOM AND SYSTEMATIC ERRORS RESULTING FROM CALIBRATION

Estimator	Batch	Model	Systematic RSD (%)		Random RSD (%)	
			Chem.	Cal.	Chem.	Cal.
$G^2/N$	A	Linear	0.33	0.33	0.77	0.74
$G^2/N$	B	Linear	0.43	0.43	0.62	0.58
3/4	A	Exponential	0.74	0.74	1.68	1.72
3/4	B	Exponential	0.99	0.99	1.32	1.30

The data for each can are presented in Table 10. For the level of sample parameters tested, this sensor requires only one paired measurement to indicate that a change has occurred.

For the ISAS(P)-CAL system, all of the sensors [(1/4)/watt, (2/4)/watt and (3/4)/watt] are affected when the sample parameters change. As an example, the control chart for the summed singles/watt is plotted in Figure 8. The control limits

were determined during the calibration measurements and are derived from the standard deviation of the differences.

The sensors for the NWC-CAL system are not as sensitive as those for the ISAS(P)-CAL system. When the sample parameters are changed, the gross neutron count per watt does not indicate that a change has occurred. The coincident neutron count per watt does show an effect. However, the indication is not as clear as change

Table 10 - ASSAY DIFFERENCES BETWEEN PRIMARY AND CONTROL MEASUREMENT FOR MIXED OXIDE BATCHES A AND B USING BATCH A CALIBRATION PARAMETERS

Can No.	Batch	NWC-CAL	ISAS (P)-CAL	ISAS (P)-NWC
31	A	-0.257	0.095	0.351
5		-0.504	-0.348	0.156
19		-0.248	-0.343	-0.095
6		-0.203	-0.684	-0.481
23		-0.368	-0.613	-0.245
12		-0.116	-2.050	-1.934
11		0.097	-0.350	-0.447
1		0.218	-0.104	-0.321
27		0.529	-0.444	-0.972
30		-0.043	-1.041	-0.998
15		0.274	-0.013	-0.287
24		0.459	0.127	-0.331
35		-0.185	1.598	1.783
2		-0.119	2.225	2.344
8		-0.632	-1.720	-1.089
17		-0.577	-0.183	0.394
20		B	3.727	35.845
29	4.099		35.342	31.242
16	5.435		43.332	37.897
21	8.080		71.636	63.556
22	9.373		70.719	61.346
33	14.408		226.582	102.174
32	24.269		148.421	124.152
4	38.270		184.708	146.438
26	38.172		188.142	149.970

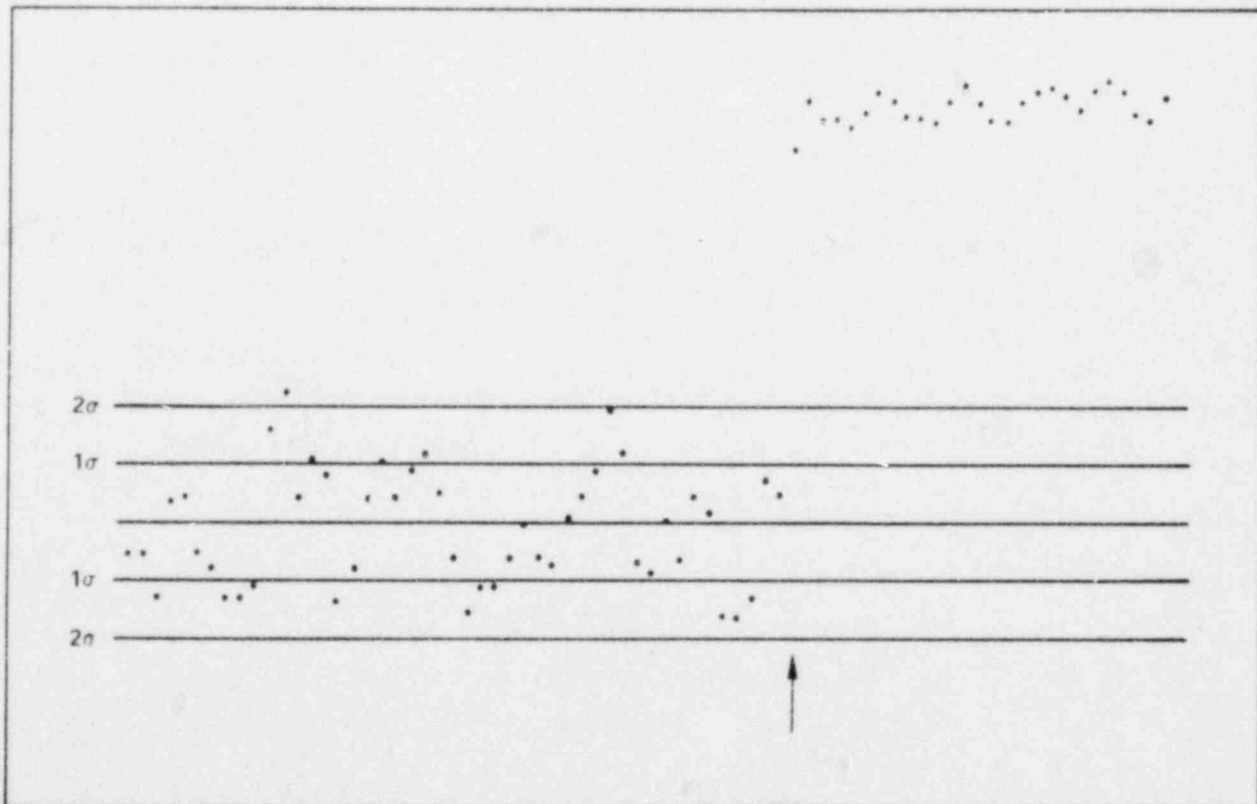


FIGURE 8 - Control chart for summed singles per watt. The arrow indicates change in sample parameters.

in assay differences. The control chart for the summed coincident neutron count per watt is shown in Figure 9. The control limits were derived from the standard deviation of differences during calibration. When the coincident neutron counts are divided by the gross weight of the sample, the resulting sensor (coincident neutron count per gram per watt) is a more sensitive test. This sensor is shown in Figure 10.

For both measurement systems dynamic sensors which indicate the change in sample parameters and thus a need for recalibration have been identified.

## Control module

The control module provides the necessary interfaces between the instrument operators and the other modules. It is in this

module that the SNM assay values and their uncertainties are determined. This module also performs the calculations necessary to determine the number of paired measurements which are required to validate the calibration curve at a stated confidence level.

For example, consider the difference between the assay values as determined in the dynamic sensor module [19]. If the average difference in assay values was determined during calibration, then the average difference in assay values while data are being taken can be compared to that value to determine if a shift has occurred. The technique for determining changes in average values is a standard statistical test [20]. In order to determine the number of samples necessary to verify the calibration curve at a stated confidence level, the parameters

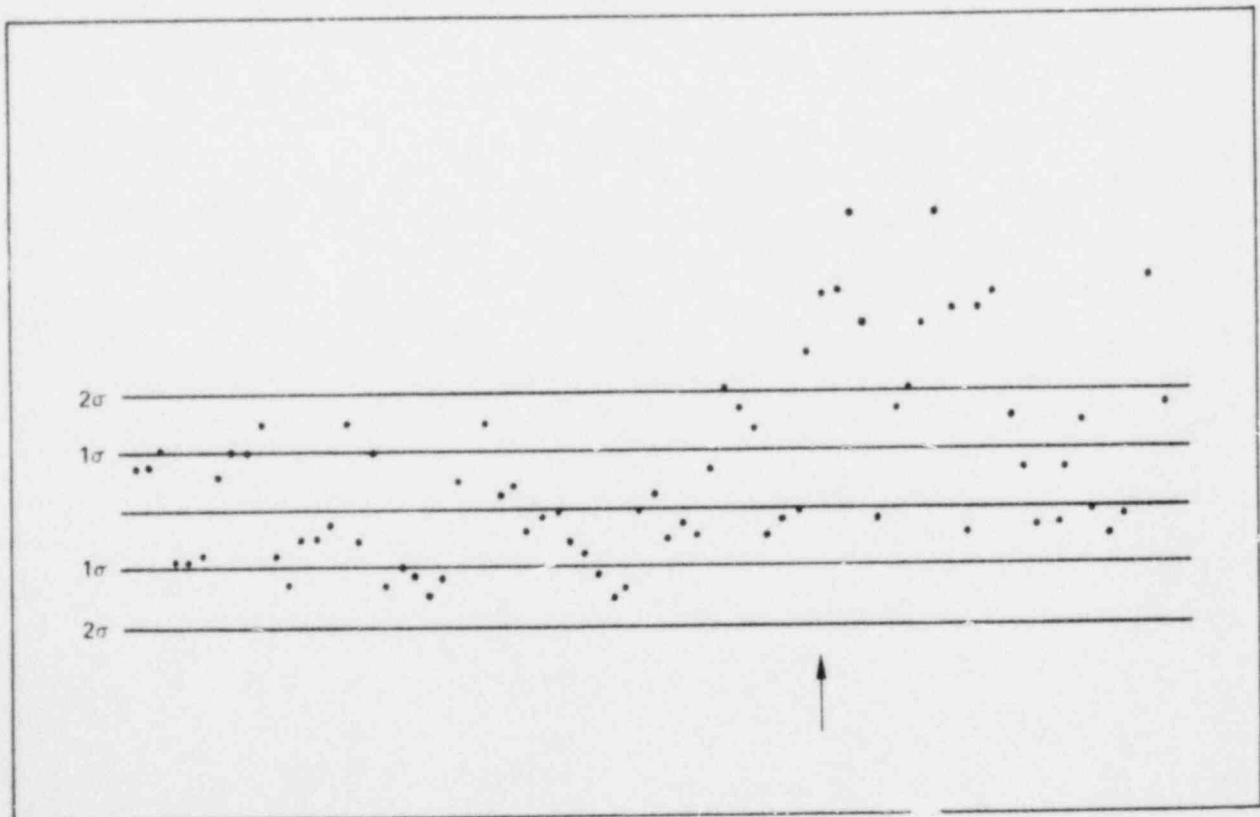


FIGURE 9 - Control chart for summed coincident neutrons per watt. The arrow indicates change in sample parameters.

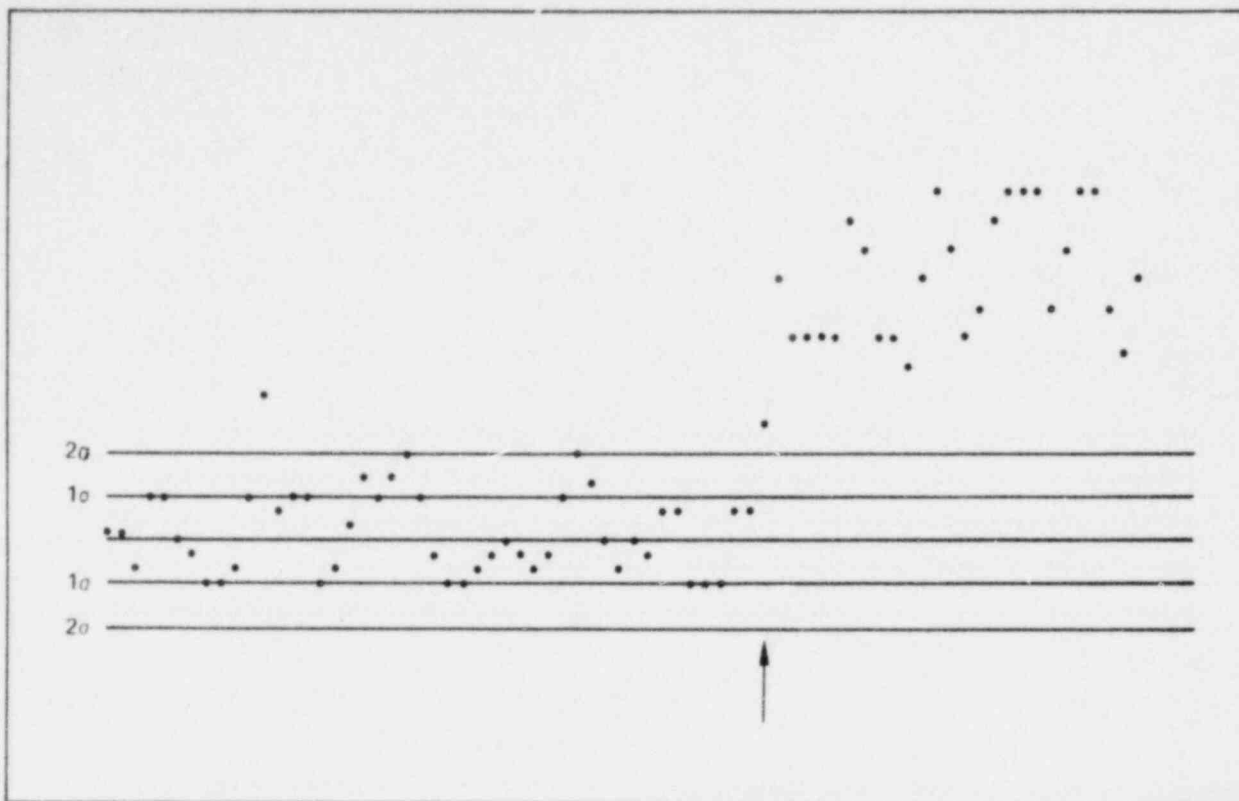


FIGURE 10 - Control chart for summed coincident neutrons per gram per watt. The arrow indicates change in sample parameters.

must be specified by the material control and accounting system. They are: the largest permissible shift ( $\Delta$ ) that can be allowed before the accountability system is affected; the probability ( $\beta$ ) that, although a shift has taken place, the data will indicate that none has occurred; and the probability ( $\alpha$ ) that although no shift in assay differences has occurred, the data will indicate otherwise. A two-sided hypothesis test is used because positive and negative bias shifts are considered equally important. Figure 11 is a plot of the number of paired measurements required to detect differences for two sets of significance parameters ( $\alpha, \beta$ ). The number of measurements is plotted against the standardized difference ( $d$ ) defined by:

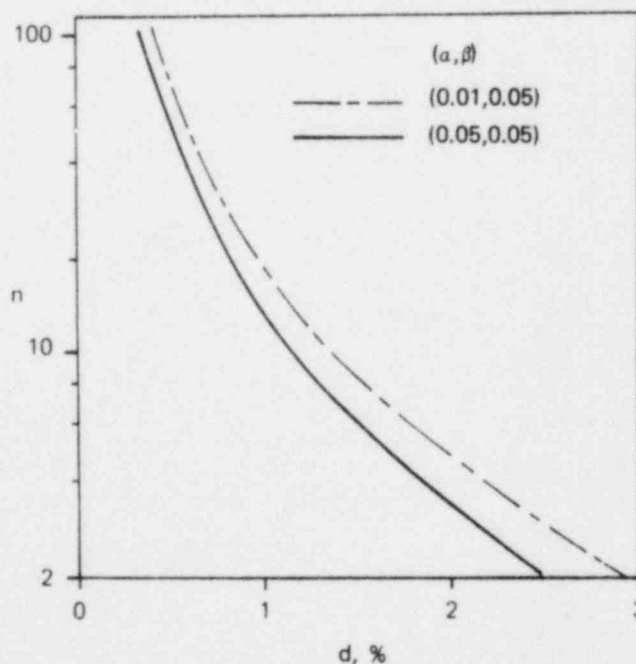


FIGURE 11 - Number of paired measurements ( $n$ ) required to detect a standardized difference ( $d$ ), when the bias shift is expressed in units of the pooled variance.



$$d = \frac{|D_c - D_p|}{\sqrt{(\sigma_{D_c}^2 + \sigma_{D_p}^2)}}$$

where:  $D_c$  is the mean difference determined during calibration

$D_p$  is the mean difference determined during the measurement of the unknowns

$\sigma_{D_c}^2$  and  $\sigma_{D_p}^2$  are the variances associated with their respective means.

On the figure, the standardized difference is expressed in units of pooled sigma

$\left[ \sqrt{(\sigma_{D_c}^2 + \sigma_{D_p}^2)} \right]$ . Thus, to detect a shift of one unit of pooled sigma, approximately 13 paired measurements are required. For the NWC-CAL system one unit of pooled sigma is 1.4%; for the ISAS(P)-CAL system it is 4.3%.

The plutonium concentration and isotopic changes from Batch A to Batch B cause a greater-than  $3\sigma$  change in the response of the instrument. Only one measurement is needed to detect changes of this magnitude.

## Conclusions

The confirmatory assessment experiments demonstrate traceable measurements for plutonium in mixed oxide power using commercially available spontaneous-fission assay systems. The experiments illustrate two major concepts: 1) the production of calibration materials using calorimetric assay, and 2) the use of paired measurements for measurement assurance.

A statistically significant bias between the computational and empirical methods of determining the effective specific power ( $P_{eff}$ ) has been qualified at the 95% confidence level as 0.27% for Batch A and 0.20% for Batch B. Using the computational  $P_{eff}$ , the mean difference between chemical assay and calorimetric assay is 0.0009 g for Batch A and 0.0004 g for Batch B. These differences do not produce statistically significant differences in the calibration curves for the spontaneous-fission assay systems.

A data base which will provide a technical basis for the preparation of guides and regulations has been initiated. The elements of the data base are the random and systematic errors for traceable spontaneous-fission assay systems and for a calorimetric assay system. The data base also includes control charts to demonstrate that the instruments and measurements were in a state of statistical control. Analysis of the calibration curves was used to determine the systematic error for each spontaneous-fission assay system. Repeat measurements were used to determine the random error. The traceability of these measurements has been established and maintained by using the measurement control technique (dynamic calibration). The implementation of dynamic calibration has been clarified by employing a systems approach to separate the four major functions into separate modules: the regression, instrument sensor, dynamic sensor, and control modules. Dynamic sensors have been identified for each system which are capable of detecting changes in the plutonium isotopic composition and in the plutonium concentration which would necessitate recalibration.



Using the method of paired differences, confidence level statements can be derived concerning the current validity of the calibration curve.

The confirmatory assessment experiments demonstrate that the traceability of plutonium NDA can be established and maintained by using calorimetric assay for calibration and measurement assurance.

## Acknowledgements

The author wishes to thank the following people without whose help the experiments could not have been completed: M. J. Harris (data collection and analysis); J. Y. Jarvis (data analysis); R. L. Goss (sample preparation); R. J. Seiler, H. S. Carden, Jr., and W. E. Sheehan (chemical characterization); and B. A. Walters (manuscript preparation).

## References

1. D. C. Wadekamper, D. M. Bishop and L. T. Hagie, Joint-Plant Instrumentation Program and Integrated Safeguards Experiment, GEAP-12114-8, General Electric Co., San Jose, California (June 1972), 26 pp.
2. K. Onishi, H. Akatsu, T. Itaki, K. Miyahara, Y. Tokoro and M. Tsutsumi, "Nondestructive Assay Equipment for Quantitative Determination of Nuclear Material in a Plutonium Fuel Fabrication Facility," in Safeguarding Nuclear Materials, Vol. II, International Atomic Energy Agency, Vienna, Austria, 1976, pp. 551-564.
3. C. D. Bingham, H. T. Yolken and W. P. Reed, "Nondestructive Assay Measurement can be Traceable," Nuclear Materials Management, V:II, 32-35 (Summer 1976).
4. D. M. Bishop, "Nondestructive Assay Measurement Traceability: The Burden of Proof," Nuclear Materials Management, V:I, 16 (Spring 1976).
5. "Considerations for Establishing Traceability of Special Nuclear Material Accounting Measurements," Regulatory Guide 5.58, U. S. Nuclear Regulatory Commission, November 1978.
6. T. E. Shea, "Reduction, Control and Estimation of Nondestructive Assay Errors," Nuclear Materials Management, III:III, 341 (Fall 1974).
7. "American National Standard Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," ANSI-N15.22-1975, American National Standards Institute, Inc., New York, New York, June 5, 1975.
8. "Guide to Calibrating Nondestructive Assay Systems," ANSI-N15.20, American National Standards Institute, Inc., New York, New York, 1975.
9. W. J. Youden, Statistical Techniques for Collaborative Tests, Association of Official Analytical Chemists, Washington, D.C., 1973.
10. J. L. Jaech, Statistical Methods in Nuclear Material Control, TID-26298, Exxon Nuclear Co., Richland, Washington (1973).
11. D. R. Rogers and W. W. Rodenburg, Calibration Alternatives for the Calorimetric Assay of PuO<sub>2</sub>, Mound Facility, to be published.
12. W. W. Strohm, "The Measurement of the Half-Life of Plutonium-239 by the U.S. Half-Life Evaluation Committee," International Journal of Applied Radiation and Isotopes, 29, 481-483 (1978).
13. "Nondestructive Assay for Plutonium in Scrap Material by Spontaneous Fission Detection," Regulatory Guide 5.34, U. S. Atomic Energy Commission, June 1974, 11 pp.
14. "Standard Test Methods for Nondestructive Assay of Special Nuclear Materials Contained in Scrap and Waste," in 1977 Annual Book of ASTM Standards, ANSI/ASTM C853-77, American Society for Testing and Materials, Philadelphia, Pennsylvania, 1977.

15. M. S. Krick, "<sup>240</sup>Pu-Effective Mass Formula for Coincidence Counting with SR Electronics," in Nuclear Safeguards Research and Development Program Status Report, LA-7030-PR, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (March 1978), pp. 16-18.
16. M. S. Zucker, "Neutron Correlation Counting for the Nondestructive Analysis of Nuclear Materials," in Analytical Methods for Safeguards and Accountability Measurements of Special Nuclear Materials, NBS Special Publication 528 (November 1978), pp. 261-283.
17. J. E. Cline and P. G. Voilleque, Quantitative Plutonium and Uranium Analysis Verification Measurements for Gamma-Ray Spectrometry and High-Level Neutron Coincidence Counter Calibrations, ISPO 60, Science Applications, Inc., Rockville, Maryland (February 1979), p. III-2.
18. R. Sher, Operating Characteristics of Neutron Well Coincidence Counters, BNL-50332, Brookhaven National Laboratory, Upton, New York (January 1, 1972), 44 pp.
19. J. F. Lemming and C. R. Rudy, A Sampling Plan for Use with Dynamic Calibration, MLM-2642 (September 14, 1979), 13 pp.
20. M. G. Natrella, Experimental Statistics, National Bureau of Standards Handbook 91, National Bureau of Standards, Washington, D. C., 1966, p. T-16.

# Appendixes

## Appendix A

### SAMPLE PREPARATION

The first isotopic mixture, designated Batch A, consists of approximately 6.5 kg of  $\text{PuO}_2$ . It was received in three containers. The contents were dried in the containers for 24 hr at  $110^\circ\text{C}$ . The material was allowed to cool in an inert atmosphere of nominally 99.5% Ar, 0.5%  $\text{N}_2$ , 1 ppm  $\text{H}_2\text{O}$ , and 10 ppm  $\text{O}_2$ . After cooling, the contents of the three containers were mixed by hand and divided into two batches. Each batch was blended separately for 2 hr in a Patterson-Kelly twin-shell blender. One-half from each of these blended batches was poured together to form two new batches. The new batches were mixed by hand and then blended for 2 hr. The materials from these final blends were designated A-1 and A-2. These identifications were placed on all analytical samples and liner cans.

The second isotopic mixture, which consists of approximately 4.5 kg of  $\text{PuO}_2$ , was treated in the same manner except that only one blend was necessary.

Six 15-g analytical samples were chosen from each blend using the ASTM cone and quartering technique to minimize sampling errors. The weights of these samples were determined using a certified balance which has a precision of 0.0005 g. The samples were calorimeted using an analytical calcrimeter which has a precision of 0.1%. Subsequent to the power measurements these samples from each blend were totally dissolved and at least two aliquots were drawn from the solution for each of the following analyses: total Pu (coulometry), total U (coulometry), Pu isotopic concentration (mass spectrometry),  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  concentrations (alpha spectrometry), and U isotopic concentrations (mass spectrometry).

The effective specific power of the mixed oxide in watts per gram of sample was used as a measure of the homogeneity of the blends (see Appendix B).

After the homogeneity was verified, 16 liner containers were loaded from Batch A and 9 liner containers from Batch B. A certified balance was used to weigh the material. Samples were prepared with weights between 100 and 1000 g. The liner container is a welded stainless steel can 6.2 cm in diameter by 15.9 cm high. In order to provide a helium leak check for the welds, the inert atmosphere was purged to approximately 40%  $^4\text{He}$ . After the liner cans were removed from the glovebox they were decontaminated and leak checked, and the welds were examined using a dye penetrant test.

Each liner can was then loaded into a second stainless steel container (6.6 cm in diameter by 16.4 cm high) and welded. The secondary containment was leak checked and the welds were examined using the dye penetrant test. In order to ensure safety and to give a baseline for future safety checks, all containers were radiographed.

Appendix B

HOMOGENEITY VERIFICATION

The effective specific power of the mixed oxide (watts per gram of sample) was used as a measure of the homogeneity of the subsamples generated from each isotopic mixture. Analysis-of-variance techniques were used to determine if the data supported the hypothesis that the samples were not homogeneous. The average values for each aliquot are shown in Tables B-1 and B-2. The data demonstrate that the hypothesis is false.

Table B-1 - ANALYSIS-OF-VARIANCE DATA FOR BATCH A

Blend	Sample No.	Weight (g)	Number of Data Points	P <sub>MOX</sub> (W/g)	S <sub>P</sub> MOX
A-1	L-01	15.1054	3	583.62	0.24
	L-08	15.0275	5	584.90	0.30
	L-09	15.3102	4	583.00	0.64
	L-10	15.2064	4	583.94	0.50
	L-12	14.9676	5	584.70	0.58
	All Data			21	584.28
A-2	L-02	15.0081	4	583.41	0.38
	L-03	15.0655	6	584.17	0.65
	L-04	14.7931	5	584.00	0.70
	L-06	14.9545	5	584.55	0.61
	L-07	14.9353	7	583.93	0.84
	L-11	15.0650	4	583.69	0.64
	All Data		31	584.00	0.70
Total of Blends A-1 and A-2			52	584.11	0.70

Table B-2 - ANALYSIS-OF-VARIANCE DATA FOR BATCH B

Sample No.	Weight (g)	Number of Data Points	P <sub>MOX</sub> (W/g)	S <sub>P</sub> MOX
L-17	14.7517	4	1267.46	2.53
L-18	14.2321	4	1268.07	2.64
L-19	14.2403	4	1267.84	2.78
L-20	15.0474	4	1268.44	1.45
L-21	14.7571	4	1267.97	3.71
L-22	14.4813	4	1267.92	1.97
	All Data	24	1267.95	2.33

## Appendix C

### DESCRIPTION OF CALORIMETER

The calorimeters used for this experiment are model 114A twin resistance-bridge isothermal calorimeters built at Mound. The calorimeters were immersed in a common water bath. The temperature of the bath was held near 29.95°C, varying only 0.01°C throughout the experiment. The sensitivity was also closely monitored and ranged from 0.024814 V/W to 0.024768 V/W during the experiment. A typical value for the  $BP_0$  reading in Calorimeter 1 was 0.0031 V, and 0.0037 V in Calorimeter 2.

A photograph of one of the calorimeters is shown in Figure C-1.

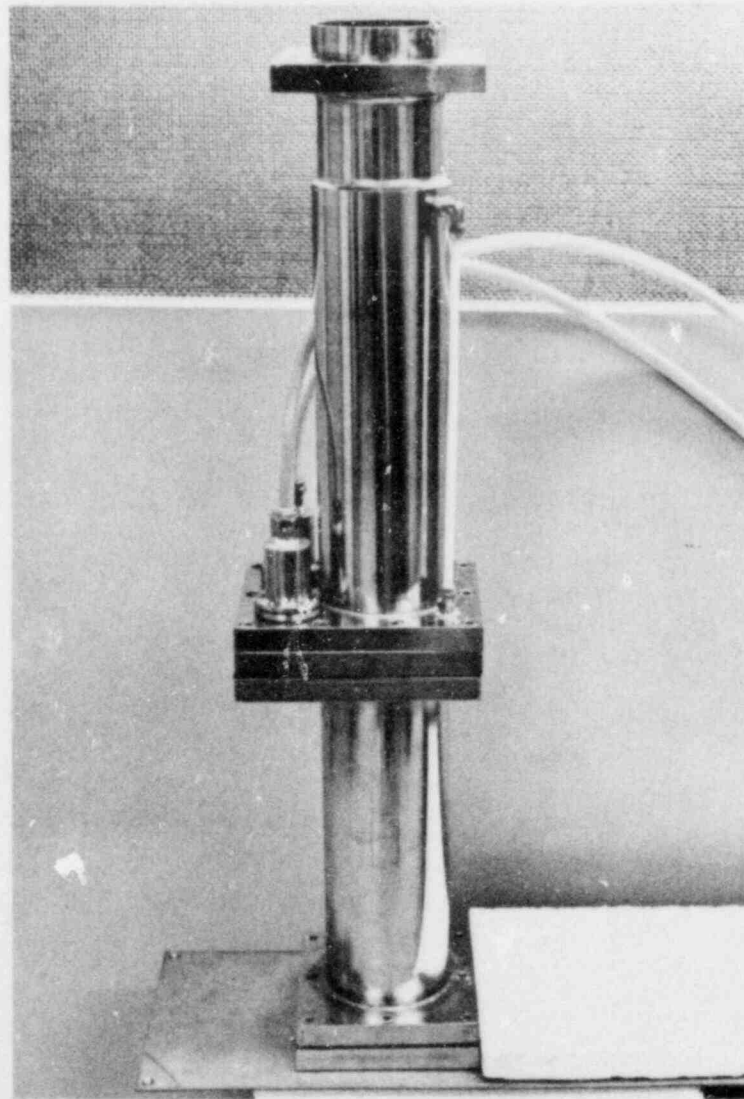


FIGURE C-1 - Twin resistance-bridge isothermal calorimeter.



## Appendix D

### DESCRIPTION OF NEUTRON WELL COUNTER

The neutron well counter uses 24  $\text{BF}_3$  detectors located symmetrically around a well cavity. The detectors are separated into sets of three, each set having its own amplifier. The sample cavity has an 8-in. diameter and is 26 in. high. A spacer was used to center the sample in the well cavity. The response of the well counter as a function of sample position was measured using a 15-g mixed oxide analytical sample placed at various heights along the center axis. The counting period for the well counter was 500 sec.

The electronics package supplied with the well counter does not measure accidental coincidences. If the background is constant during a measurement period, the accidental coincidences can be calculated using the following formula:

$$A = \frac{G^2 \tau}{t}$$

where: A = accidental counts  
G = observed gross counts  
 $\tau$  = gate width in microseconds  
t = counting time in seconds

The gate was set at 64  $\mu\text{sec}$ .

Attachment D-1 is a description from a National Nuclear Corporation brochure describing the well counter.

ATTACHMENT D-1

PLUTONIUM ASSAY

NEUTRON WELL COINCIDENCE COUNTER

DESCRIPTION

The Neutron Well Coincidence Counter is a passive neutron counting instrument for assaying plutonium materials. The Neutron Well Coincidence Counter is used to assay plutonium by measuring the spontaneous-fission neutrons emitted from  $^{240}\text{Pu}$ . The instrument distinguishes between fission neutrons and  $(\alpha, n)$  neutrons by counting only neutrons in coincidence.

The instrument, based on BNL design, can be used to assay powder, liquid, rods, bundles, or waste with minimum size and density effects. The system can be adapted to specific sample sizes. This instrument was recently selected to assay FFTF fuel.

The unknown sample is inserted in the well of the counter and counted for a preset time, usually 500 sec. Comparison with standards provides  $^{240}\text{Pu}$  content. With appropriate corrections and known isotopic ratios, the amount of fissionable plutonium can be determined.

SPECIFICATIONS

- Detector Assembly: 24  $\text{BF}_3$  thermal neutron detectors, 2 in. in diameter X 24 in. high, with matched performance, surrounded by high-density polyethylene.  
8 preamps, low noise, high gain, FET.  
Sample size - 8 in. in diameter X 11 in. high, removable plug in base for longer samples.  
Weight of 400 lb; mounted on casters.
- Electronics: 8 amplifiers, stable, pole-zero cancelled.  
Adjustable high-voltage supply.  
Coincidence circuit to determine real and accidental coincidence rates.  
4 L.E.D. scalars to display gross, real, and accidental counts, and elapsed time.  
Control circuit to provide preset counting times of 50, 100, 250, 500, 1000, and 5000 sec from an internal crystal time base.  
Adjustable gate lengths of 32, 64, and 128  $\mu\text{sec}$ .  
Data presented on scalars or, as an option, interfaced to printer, teletype, or computer.  
All electronics contained in standard 12 wide NIM Bin.
- Typical Performance: 2% uncertainty in 10-min counting interval for 1 g  $^{240}\text{Pu}$ .  
50% uncertainty in 10-min counting interval for 1 mg  $^{240}\text{Pu}$ .

## Appendix E

### DESCRIPTION OF ISAS(P) SYSTEM

The ISAS(P) system consists of a fission multiplicity detector with four plastic scintillators optically coupled to photomultiplier tubes, and a sample handling system. The samples are rotated about a vertical axis as they are scanned. The time to complete one scan is 172 sec.

Because the ISAS(P) electronics are prone to drift, a normalization sample was measured before and after each set of sample runs which consisted of a maximum of six scans. The average value of the before-and-after normalization measurements was used to adjust the data.

Corrections to the count rate for accidentals were made using the following procedure from the operator's manual. The following symbols are used in this procedure:

- $C_{2/4}$  - the observed two-out-of-four count rate
- $C_{3/4}$  - the observed three-out-of-four count rate
- $R_i$  - the count rate of the  $i$ th detector ( $i = 1, 2, 3, 4$ )
- $A_{2/4}$  - the accidental contribution to the two-out-of-four count rate
- $R_{2/4}$  - the true coincidence rate for the two-out-of-four detectors
- $A_{3/4}$  - the accidental contribution between the four detectors for the three-out-of-four count rate
- $a_{3/4}$  - the accidental contribution between a single detector and the true two-out-of-four coincidence rate
- $R_{3/4}$  - the true coincidence rate for the three-out-of-four detectors
- $R_{i,j}$  - the true coincidence rate between detector  $i$  and detector  $j$ .

Since the coincidence circuitry is of the overlay variety, the coincidence resolving time  $\tau$  is equal to two times the discriminator pulse width. The accidental two-out-of-four rate is given as

$$A_{2/4} = \tau(R_1R_2 + R_1R_3 + R_1R_4 + R_2R_3 + R_2R_4 + R_3R_4)$$

From the detector symmetry and by setting the electronic bias the same for all detectors,  $A_{2/4}$  becomes

$$A_{2/4} = 6\tau R_3^2$$

where:  $R_s = R_1 = R_2 = R_3 = R_4$

Then  $R_{2/4}$  is given by the difference between the observed two-out-of-four rate  $C_{2/4}$  and  $A_{2/4}$ , or

$$R_{2/4} = C_{2/4} - A_{2/4}$$

In a similar manner,  $A_{3/4}$  is given by

$$A_{3/4} = \tau^2 (R_1 R_2 R_3 + R_1 R_2 R_4 + R_1 R_3 R_4 + R_2 R_3 R_4)$$

$$A_{3/4} = 4\tau^2 R_s^3$$

and  $a_{3/4}$  is the accidental coincidence between a true coincidence from two detectors with the single counting rate of the other two detectors

$$a_{3/4} = \tau \left( R_{1,2} [R_3 + R_4] + R_{1,3} [R_2 + R_4] + R_{1,4} [R_2 + R_3] \right. \\ \left. + R_{2,3} [R_1 + R_4] + R_{2,4} [R_1 + R_3] + R_{3,4} [R_1 + R_2] \right)$$

When all the single rates are equal,  $a_{3/4}$  reduces to

$$a_{3/4} = 2\tau R_s (R_{1,2} + R_{1,3} + R_{1,4} + R_{2,3} + R_{2,4} + R_{3,4})$$

$$a_{3/4} = 2\tau R_s R_{2/4}$$

The true three-out-of-four rate  $R_{3/4}$  is given by

$$R_{3/4} = C_{3/4} - A_{3/4} - a_{3/4}$$

Attachment E-1 is a description of the ISAS from an IRT Corporation brochure.

## ATTACHMENT E-1

### ISAS

#### FOR ASSAY OF FISSILE OR FERTILE MATERIALS WITHIN CONTAINERS UP TO 8 INCHES IN DIAMETER AND 51 INCHES IN LENGTH

The IRT Isotopic Source Assay System (ISAS) is a commercially available, nondestructive assay system that quickly and accurately measures the fissile material content and, under certain conditions, the fertile material content of materials commonly found throughout the nuclear industry. For most active measurements, ISAS interrogates the sample with a hard-spectrum neutron beam, thereby eliminating the self-shielding problems frequently associated with interrogation by thermal neutrons. ISAS can also be used passively; that is, without the neutron source.

#### APPLICATIONS

ISAS has been used for active assay of the fissile material content of homogeneous or heterogeneous uranium or plutonium-bearing dry oxides, compounds, alloys, residues, calcined ash, and scrap, as well as solutions and materials of known hydrogenous content. Uranium of any enrichment can be measured. Up to four kilograms of  $^{235}\text{U}$  per linear foot of sample container has been measured without approaching the limitations of the ISAS.

The fertile material ( $^{238}\text{U}$ ) content of uranium containing up to approximately 10%  $^{235}\text{U}$  can be measured. For homogeneous samples, such as product material, the fertile component can be determined for even higher enrichments.  $^{239}\text{Pu}$  can be measured actively in the same manner as  $^{235}\text{U}$ .  $^{240}\text{Pu}$  is measured passively; that is, without using the neutron source.  $^{232}\text{Th}$  can be measured by ISAS if the age of the unknown lot of thorium is the same as that of the calibration standards.

#### SIMPLICITY OF OPERATION

ISAS is specifically designed to be operated by a technician. The basic measurement operation consists of loading a sample into ISAS, activating the system by pressing a button, reading the resultant data, and comparing the data with a calibration chart. A printer output device, desk-top programmable calculators, or minicomputers can be interfaced with ISAS.

#### ACCURACY

Under average conditions, 50 g or more of fissile material ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ ) or fertile material ( $^{238}\text{U}$ ,  $^{240}\text{Pu}$ ), heterogeneously distributed in up to one gallon of bulk matrix material, can be measured to accuracies of  $\pm 1$  to 2% relative at one standard deviation. Under carefully controlled conditions, accuracies between  $\pm 0.5$  and 1.0% relative can be attained.

When measuring less than 50 g of fissile material with a standard ISAS, the accuracy is more dependent on the quantity and type of matrix material. For example, under optimum conditions, accuracies of  $\pm 0.7\%$  relative at one standard deviation can be attained when measuring 40 g of fissile material,  $\pm 2\%$  when measuring 10 g,  $\pm 10\%$  when measuring 1 g, and  $\pm 100\%$  when measuring 0.1 g. Special-purpose ISAS units having greater accuracies are available.

#### SIZE OF SAMPLE CONTAINERS

ISAS is usually supplied with a vertical sample scanner which will accept sample containers up to 8 in. in diameter and 15 in. in height. Vertical sample scanners are available which will accept sample containers up to 51 in. in height. Standard 11-liter poly bottles are the longest containers which are ordinarily scanned in a vertical position, and ISAS must be mounted on a 3-ft high platform to scan these elongated containers. Special ISAS units are available for samples up to 10 in. in diameter.

The ISAS detector can be rotated to accommodate a horizontal sample drive mechanism. This accessory drive makes it possible to scan elongated pipe-type containers, and fuel elements up to approximately 6 ft in length, in a horizontal position.

#### SENSITIVITY

Using a hard-neutron spectrum, a standard ISAS can routinely detect 0.25 g of fissile material heterogeneously distributed through 5 kg of dense, bulky matrix material. Under optimum conditions, 0.1 g of fissile material can be detected when using soft-spectrum neutrons.

#### RANGE

Without adjustment, the ISAS can measure quantities of fissile material from a tenth of a gram to several kilograms.

#### ELECTRONICS

The ISAS is controlled by simple, extremely reliable modular electronic circuitry. These circuits have proved to be essentially trouble-free under commercial operating conditions. Coincidence circuits have been redesigned to give high-precision measurements never before routinely obtainable with assay systems of this type.

The ISAS electronics are now being supplied in "NIM-Bins". This allows the purchaser the later option of interfacing directly with a programmable desk-top calculator or minicomputer.



## Appendix F

### ADDITIONAL DYNAMIC SENSORS

Additional dynamic sensors are presented in this appendix to show their sensitivity. The arrow indicates the change in sample parameters.

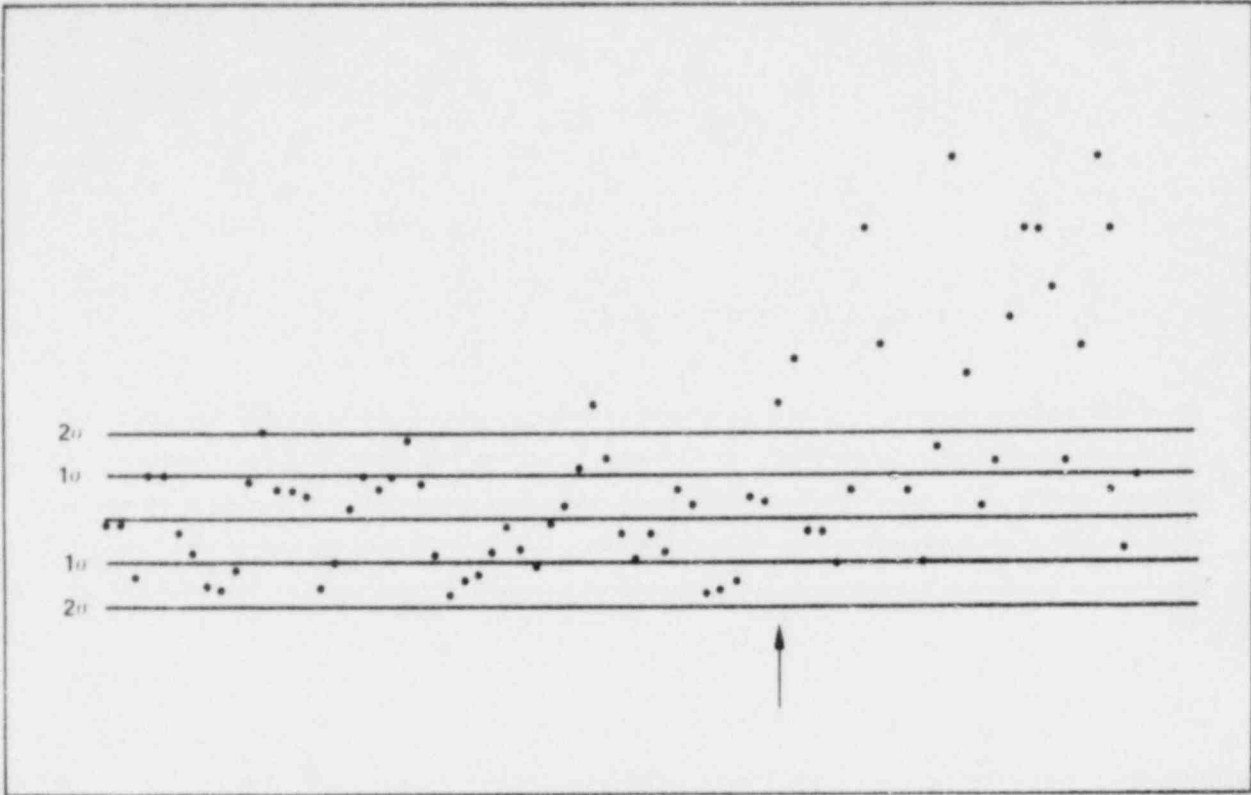


FIGURE F-1 - Control chart for triples per gram per watt.

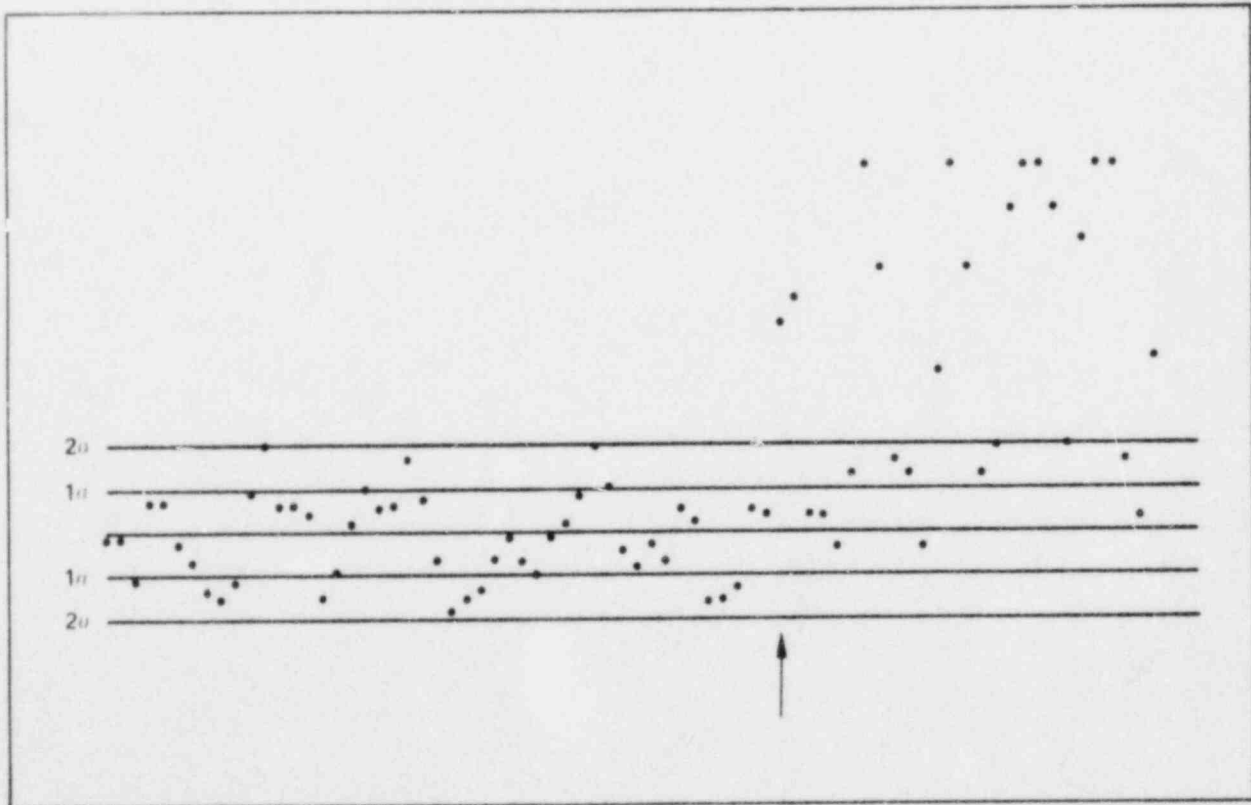


FIGURE F-2 - Control chart for doubles per gram per watt.

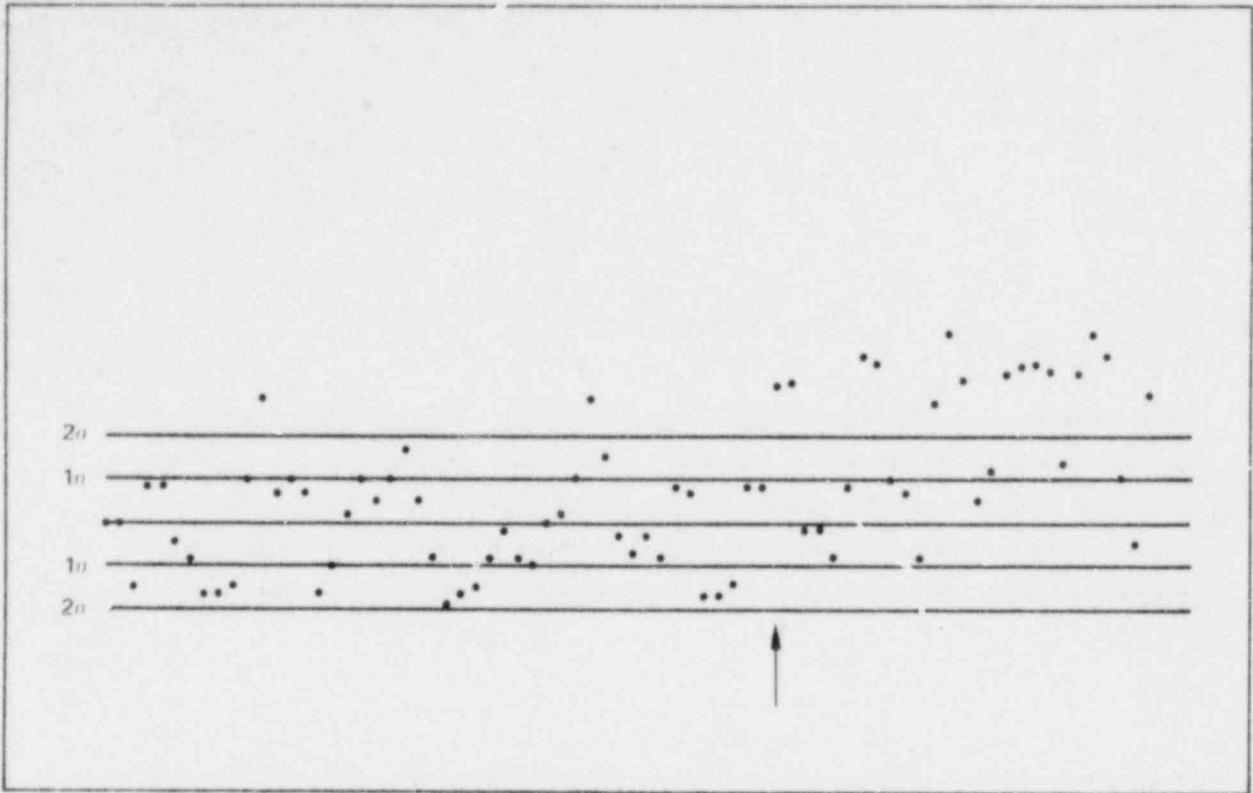


FIGURE F-3 - Control chart for singles per gram per watt.

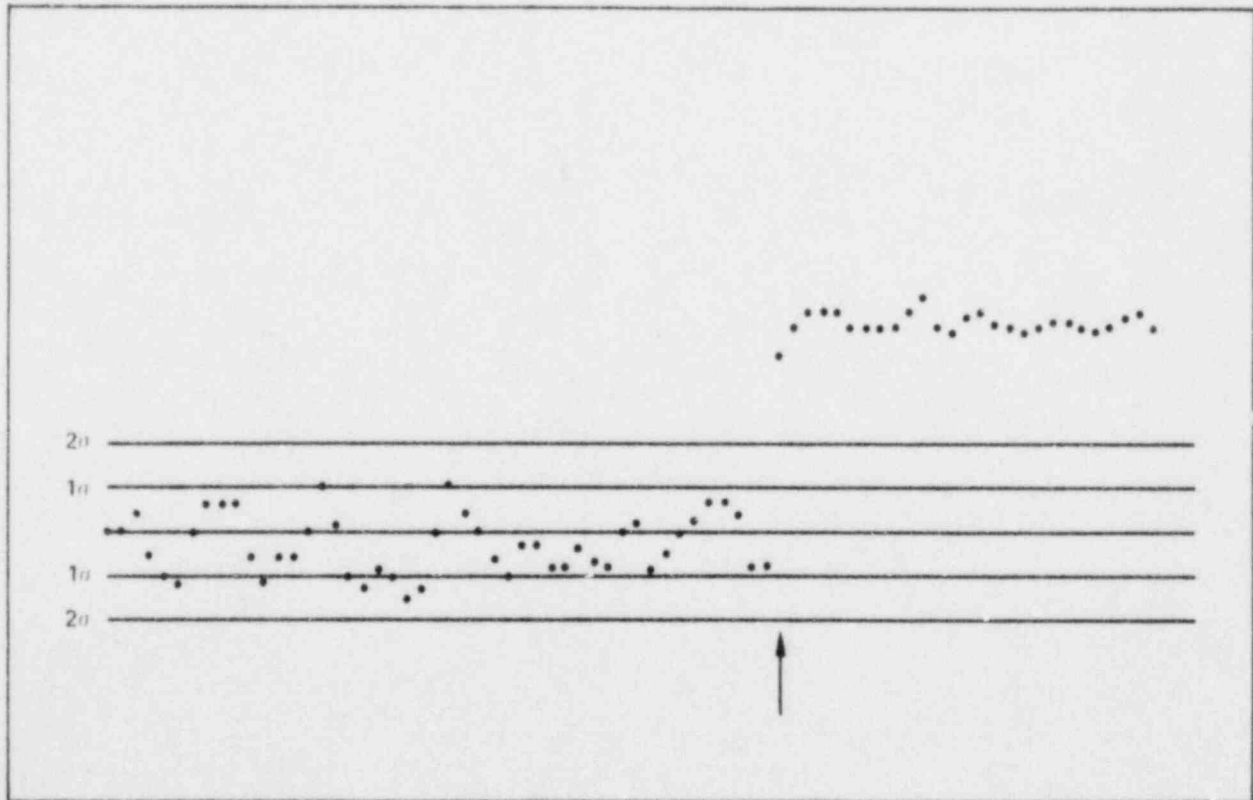


FIGURE F-4 - Control chart for triples per watt.

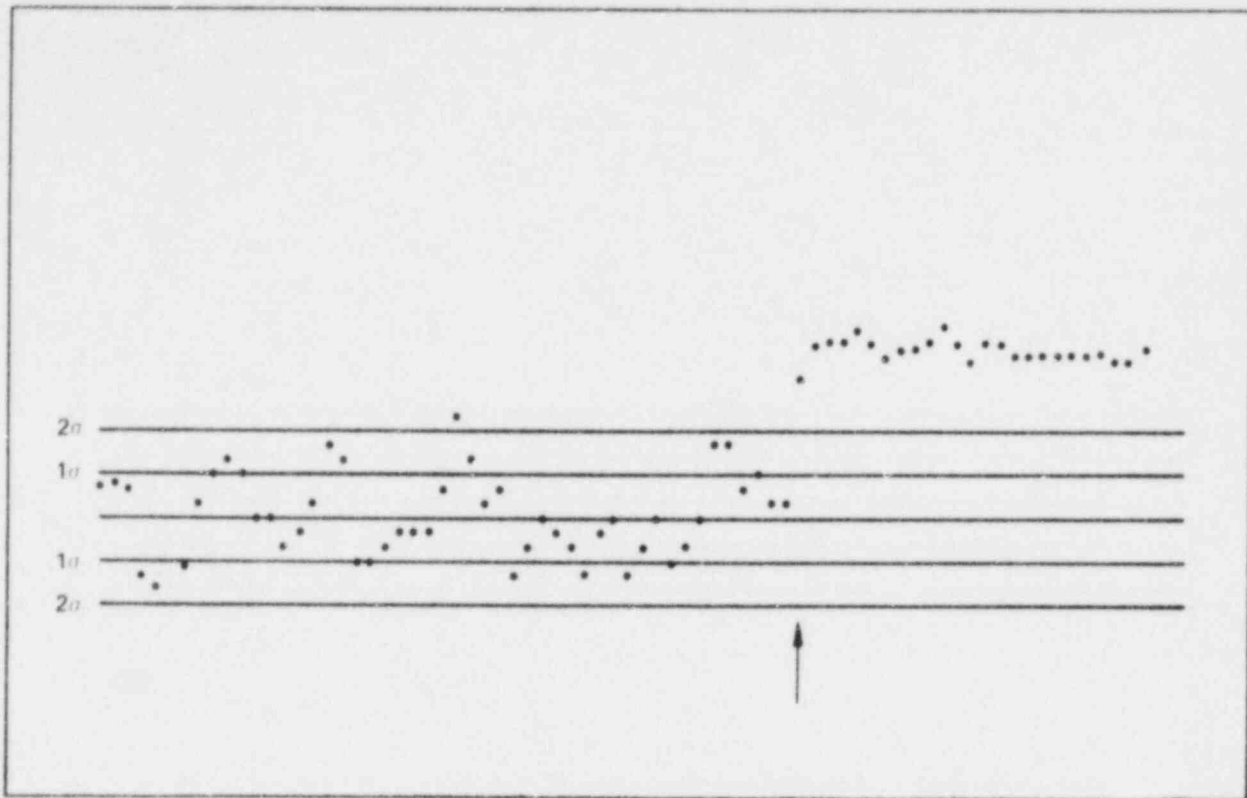


FIGURE F-5 - Control chart for doubles per watt.

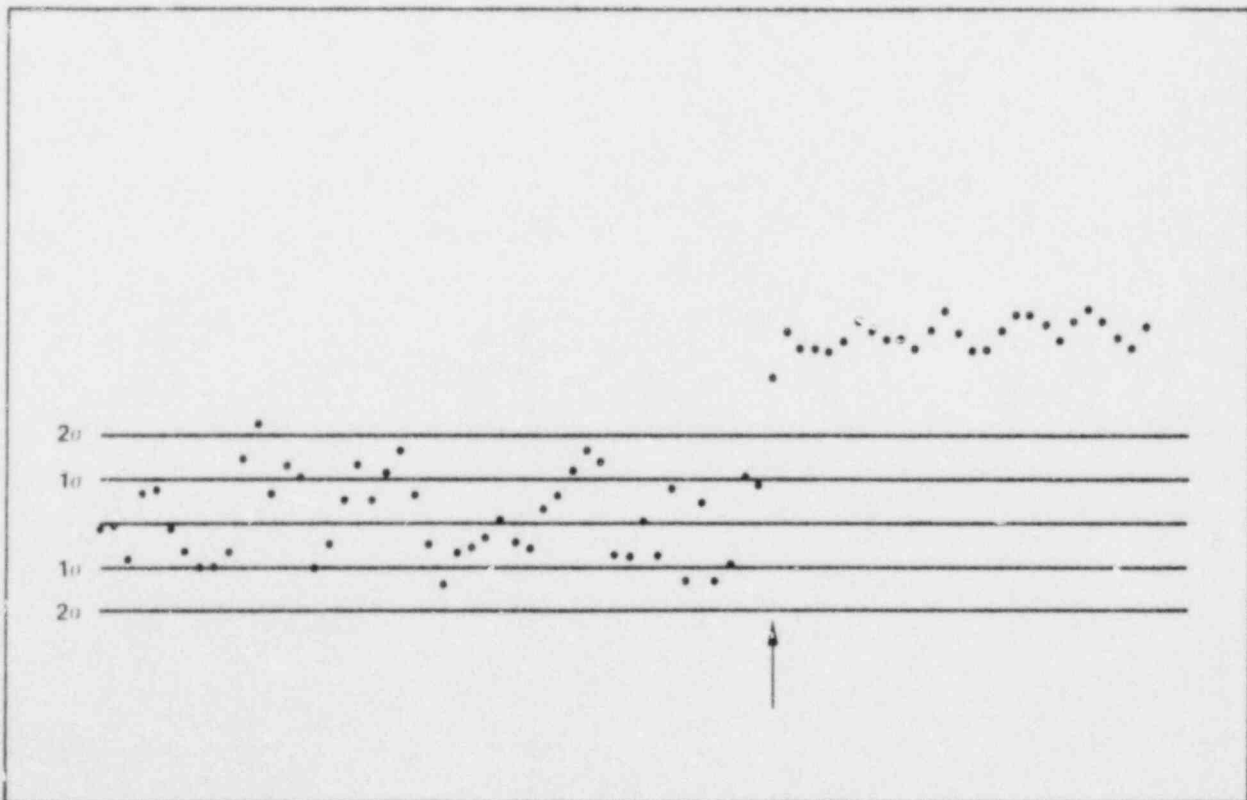


FIGURE F-6 - Control chart for singles per watt.

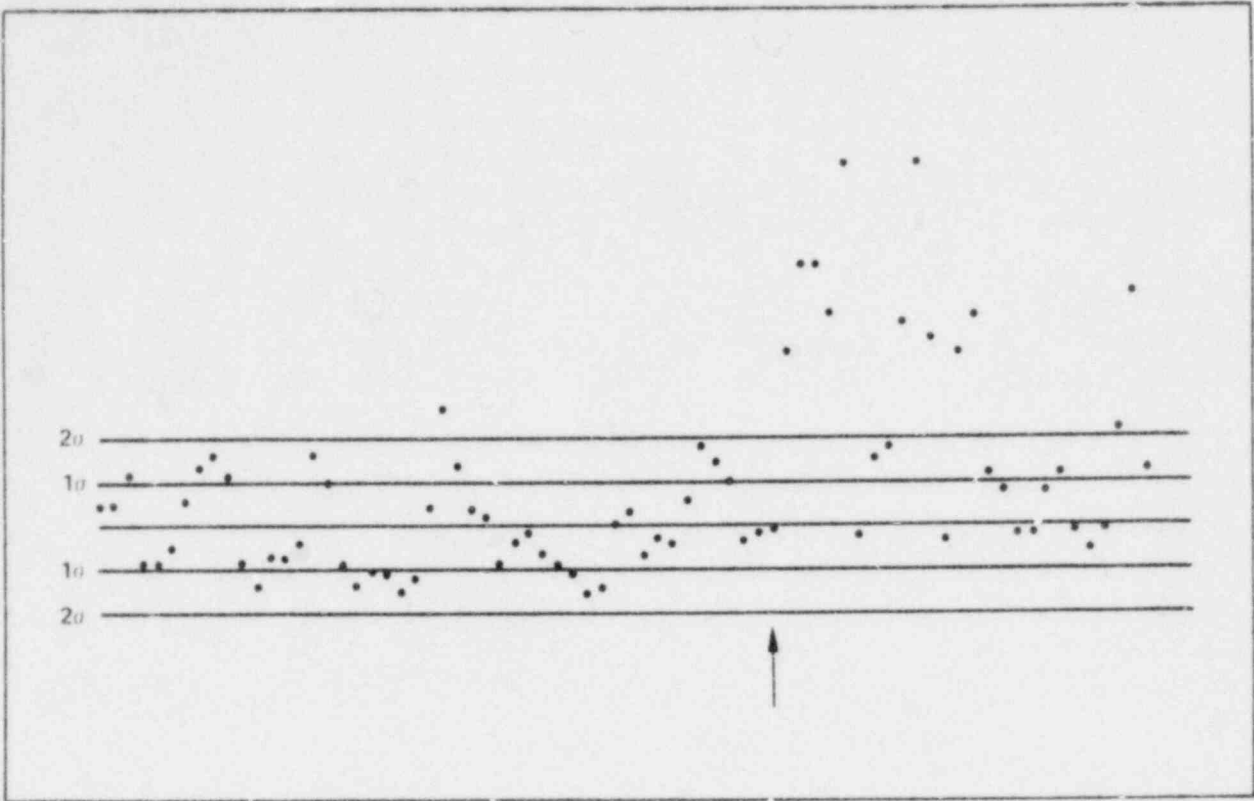


FIGURE F-7 - Control chart for coincident neutrons per watt.

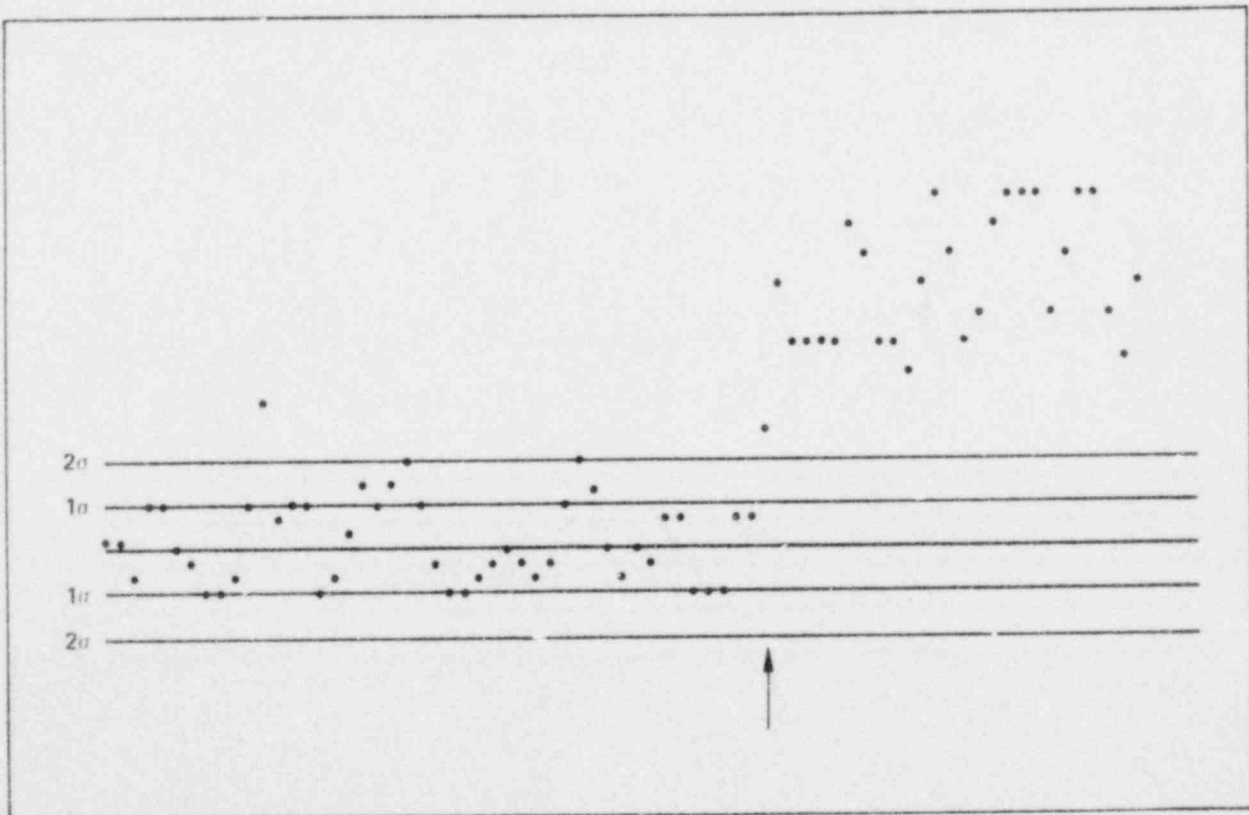


FIGURE F-8 - Control chart for coincident neutrons per gram per watt.

# Distribution

## EXTERNAL

TIC-4500 (336)

Division of Technical Information and Document Control (US NRC) (270)

P. Ting, NRC/Office of Standards Development (30)

H. N. Hill, Dayton Area Office (2)

R. K. Flitcraft, Monsanto Research Corporation  
Monsanto Technical Report Library, St. Louis

## INTERNAL

W. T. Cave

C. L. Fellers

J. G. Fleissner

C. W. Huntington

J. Y. Jarvis

J. F. Lemming (30)

J. R. McClain

D. A. Rakel

W. W. Rodenburg

D. R. Rogers

C. R. Rudy

W. H. Smith

W. W. Strohm

R. E. Vallee

Publications

Library (15)

Published by Information Services:  
Raymond P. Ratay, Editor