310
5-27-82

LA-9318-MS
(ISPO-171)

①

I-3418

Lh. 564

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

USA PROGRAM FOR TECHNICAL ASSISTANCE TO IAEA SAFEGUARDS
IAEA Department of Energy Safeguards & Security

MASTER

High-Level Neutron-Coincidence-Counter (HLNCC) Implementation: Assay of the Plutonium Content of Mixed-Oxide Fuel Assemblies

Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

DISCLAIMER

This work was supported by the US Department of Energy, Office of Safeguards and Security, and by the Program for Technical Assistance to IAEA Safeguards.

Edited by Sarah Kreiner, Group Q-1

LA-9318-MS
(ISPO-171)

UC-15
Issued: April 1982

LA--9318-MS

DE82 015083

High-Level Neutron-Coincidence-Counter (HLNCC) Implementation: Assay of the Plutonium Content of Mixed-Oxide Fuel Assemblies

John E. Foley
Gerald E. Bosler

Los Alamos
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

CONTENTS

HIGH-LEVEL NEUTRON COINCIDENCE COUNTER (HLNCC) IMPLEMENTATION: ASSAY OF THE PLUTONIUM CONTENT OF MIXED-OXIDE FUEL ASSEMBLIES

by

John E. Foley and Gerald E. Bosler

ABSTRACT

The portable High-Level Neutron Coincidence Counter is used to assay the ^{240}Pu-effective loading of a reference mixed-oxide fuel assembly by neutron coincidence counting. We have investigated the effects on the coincidence count rate of the total fuel loading (UO_2 + PuO_2), the fissile loading, the fuel rod diameter, and the fuel rod pattern. The coincidence count rate per gram of ^{240}Pu-effective per centimeter is primarily dependent on the total fuel loading of the assembly; the higher the loading, the higher the coincidence count rate. Detailed procedures for the assay of mixed-oxide fuel assemblies are developed.

I. INTRODUCTION

Mixed-oxide (MOX) reactor fuel is being used in the world today in limited applications in thermal reactors. Experimental MOX fuel rods and fuel assemblies are being tested in both light-water and heavy-water reactors.[1] MOX fuel consists of a mixture of UO_2 and PuO_2, generally with the uranium having natural enrichment (0.72% ^{235}U) or low enrichment. The plutonium fraction of the heavy metal (uranium plus plutonium) in the MOX fuel for thermal reactors is typically a few per cent.

Most of the fast breeder reactors being designed or built also use MOX fuel. The plutonium fraction of the heavy metal content is much higher in fast reactor fuel than thermal reactor fuel; typically, it is ∿20%.

Because both plutonium and uranium must be safeguarded by the International Atomic Energy Agency (IAEA) in those countries that have accepted IAEA safeguards, IAEA inspectors need to be able to measure the uranium and plutonium content of these MOX fuels. However, because the uranium in MOX fuel is generally of normal enrichment and thus has little weapons significance, measurement of the plutonium content is the primary concern.

During fabrication, MOX fuel is found in different physical forms: low density (~ 2 g/cm^3) powder; dense (~ 10 g/cm^3) pellets; individual fuel rods; complete fuel assemblies, which contain tens to hundreds of these fuel rods; scrap; and waste. Each of these forms presents different measurement problems and requires different measurement approaches. For example, the IAEA can use sampling and destructive analysis as an assay technique for powders and pellets, but not for fuel rods and fuel assemblies because of their value. Only nondestructive assay (NDA) techniques can be used to assay the plutonium content of these items.

A. Neutron Coincidence Counting

Neutron coincidence counting has been used for many years to assay the plutonium content of plutonium-bearing scrap and waste[2] and cans (~ 1-ℓ volume) of PuO_2 powder.[3] The assay of these types of low-density (~ 3 g/cm^3) samples is well understood. However, the assay of MOX fuel assemblies is not as straightforward. A MOX fuel assembly is composed of many rods containing MOX pellets that have high densities (~ 10 g/cm^3). These high densities, which result in high MOX linear fuel loadings (mass of MOX per unit length), introduce new assay problems that must be investigated.

The specific neutron coincidence counter used in these investigations is the High-Level Neutron Coincidence Counter (HLNCC).[4] The HLNCC was developed by the Los Alamos National Laboratory for the IAEA under the United States Program for Technical Assistance to IAEA Safeguards. Figure 1 shows the major components of the HLNCC: a thermal neutron detector into which the sample to be assayed is placed; an electronics package, which contains the shift-register coincidence logic circuitry; and an HP-97 calculator. The HLNCC assays the plutonium content of the fuel assembly by detecting coincidence neutrons from the spontaneous fission of primarily the ^{240}Pu isotope. A detailed description of the theory and operation of the instrument is given in Ref. 4.

Fig. 1.
HLNCC for plutonium assay.

B. Reference MOX Fuel Assembly

The IAEA is faced with the task of measuring a wide variety of sizes, shapes, densities, etc., of plutonium-bearing materials throughout the world. Part of the task of implementing the HLNCC is the development of detailed assay procedures for specific measurement problems. In this report, we focus on the assay procedures for a reference MOX fuel assembly. The characteristics of this reference fuel assembly are given in Table I; its fuel rod pattern is shown in Fig. 2.

The major goals of the work summarized in this report are to understand the parameters (such as plutonium loading, uranium loading) that affect the assay results and to develop detailed assay procedures for measurement of the reference MOX fuel assembly.

TABLE I

CHARACTERISTICS OF REFERENCE MOX FUEL ASSEMBLY

Linear MOX loading	∿450 g/cm
Linear plutonium loading	∿3.5 g/cm
Outer diameter of assembly	∿11 cm
Number of fuel rods	28
Rod diameter	1.6 cm

II. SIMULATED MOX FUEL ASSEMBLIES

A. Fuel Rods

We simulated the reference MOX fuel assembly by combining both unirradiated MOX fuel rods and unirradiated UO_2 fuel rods into specific geometries. Figure 3 shows an example of an assembly that contains 28 fuel rods and

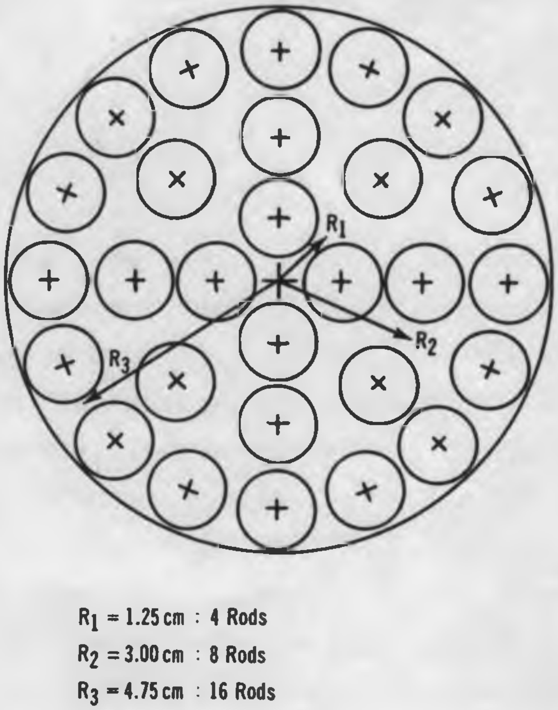

Fig. 2.
Fuel rod pattern of the reference MOX fuel assembly.

Fig. 3.
Simulated 28-rod MOX fuel assembly.

simulates the fuel rod pattern of the reference assembly. Characteristics of the fuel rods used to construct the simulated fuel assemblies are given in Tables II and III. These rods are short sections (1.2 m long) of fuel rods for a boiling-water reactor (BWR).

We are not able to construct the reference MOX assembly exactly because we do not have MOX fuel rods with either the correct diameter or the correct Pu/U loading. However, we can simulate the reference assembly in two ways.

One way is to duplicate the outside diameter, the approximate plutonium loading, and the approximate uranium loading of the reference assembly. We do this by assembling a mixture of MOX rods and UO_2 rods so that (1) the ratio of MOX rods to UO_2 rods gives the same average Pu/U loading as the reference assembly and (2) the total number of rods gives the same linear fuel loading (UO_2 + PuO_2). Because the fuel rods used in making the simulated fuel assembly are smaller in diameter than those of the reference assembly (1.43 cm vs 1.60 cm), we must put more rods into the simulated assembly than there are in the reference assembly to give the correct average linear fuel loading. Thus, we can simulate the correct average linear fuel loading, but not with the proper geometry.

A way to simulate the geometry of the assembly is to use the correct number of fuel rods and the correct rod pattern, but we cannot then simulate the proper linear fuel loading because the diameter of the rods is too small.

We are thus forced to generate a calibration curve and to establish measurement procedures for the reference fuel assembly by making measurements on simulated assemblies ("best available standards") that are not exact duplicates of the reference assembly.

We simulated the correct average linear fuel loading of the reference assembly by using 37 fuel rods in an assembly with an outside diameter of $\sim$11 cm. The proper average linear plutonium loading is achieved using 14 MOX rods and 23 UO_2 rods, which results in an assembly with an average linear fuel loading of 453 g/cm and an average linear plutonium loading of 3.44 g/cm; both of these values are very close to those of the reference assembly (see Table I). The rod pattern of this 37-rod assembly is shown in Fig. 4.

We simulated the correct fuel rod pattern by constructing a 28-rod assembly (Fig. 3). When this assembly is loaded with 14 MOX fuel rods and 14 UO_2 rods, the resulting average linear plutonium loading is 3.44 g/cm, which is

TABLE II

CHARACTERISTICS OF BWR MOX FUEL RODS

Rod diameter	1.43 cm
Clad thickness	0.089 cm
Clad material	Zr-2
Pellet diameter	1.22 cm
Stack length	121.7 cm
Weight MOX/rod	1474.1 g
MOX density	10.29 g/cm^3
Enrichment	2.30 wt% PuO_2
Weight plutonium/rod	29.93 g
Uranium enrichment	natural (0.72%)
Weight ^{235}U/rod	9.06 g
Linear fuel loading (PuO_2 + UO_2)	12.11 g/cm
Linear plutonium loading	0.246 g/cm
Linear uranium loading	10.34 g/cm
Linear ^{240}Pu-effective loading	5.050 x 10^{-2} g/cm

Isotopic Composition of the Plutonium

Isotope	Weight Per Cent
^{238}Pu	0.28
^{239}Pu	75.52
^{240}Pu	18.12
^{241}Pu	5.00
^{242}Pu	1.09

TABLE III

CHARACTERISTICS OF BWR UO_2 FUEL RODS

Rod diameter	1.43 cm
Clad thickness	0.089 cm
Clad material	Zr-2
Pellet diameter	1.22 cm
Stack length	121.9 cm
Weight UO_2/rod	1504 g
UO_2 density	10.48 g/cm^3
Enrichment	2.34 wt% ^{235}U
Linear fuel loading (UO_2)	12.34 g/cm
Linear uranium loading	10.88 g/cm

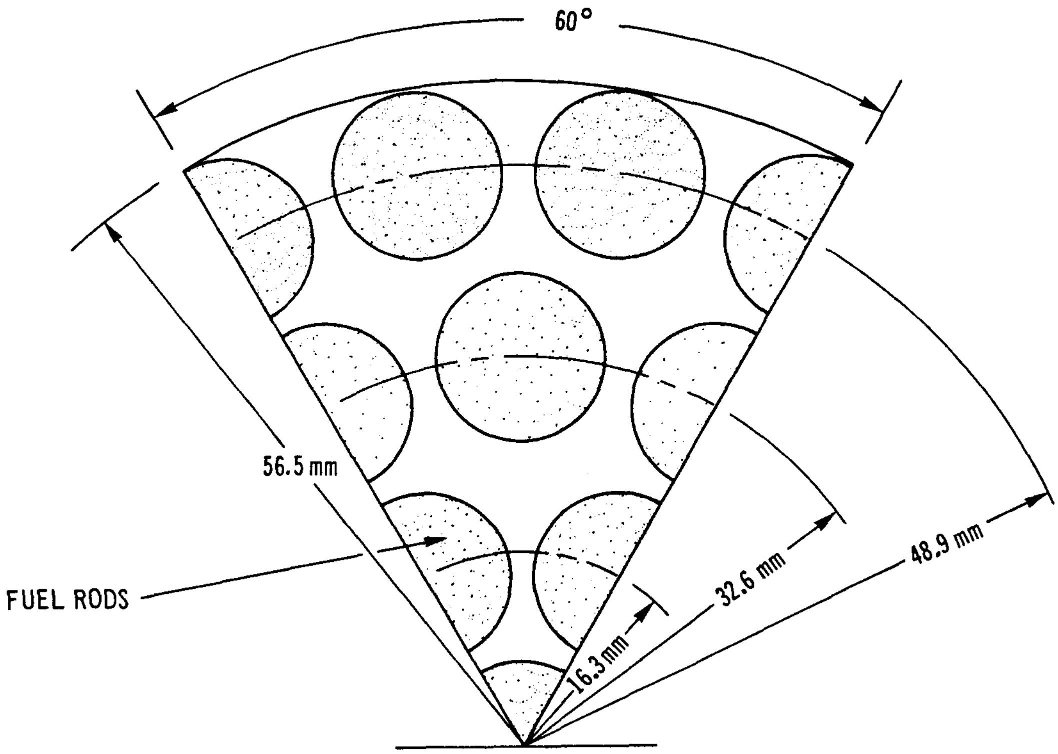

Fig. 4.
Fuel rod pattern of the 37-rod (1/6 section) fuel assembly.

close to that of the reference assembly, but the average linear fuel loading is only ~342 g/cm, which is much lower than that of the reference assembly.

By using various combinations of MOX and UO_2 fuel rods in these 28 and 37-rod assemblies, we are able to vary the average Pu/U ratio over a wide range.

B. Measurement Geometry

The HLNCC was designed primarily to accommodate samples having lengths of <35 cm. However, longer samples can be assayed if they are allowed to extend out of the top and bottom of the neutron detector. Only the portion located inside the detector contributes significantly to the assay.

When assay of the complete fuel assembly is required, the assembly must be scanned; that is, the assembly must be moved through the neutron detector at a constant speed. When it is unnecessary or impossible to scan the complete fuel assembly, assay of a single portion of the fuel assembly will give a determination of the linear plutonium loading.

In the measurements described in this report, we did not scan the fuel assemblies but measured only a single portion. Thus, our results are reported in terms of linear plutonium loading rather than total plutonium content of the assembly.

III. EXPERIMENTAL MEASUREMENTS

A. Analysis Procedures

1. ^{240}Pu-Effective.

The HLNCC measures the spontaneous fission rates of the even-mass plutonium isotopes: ^{238}Pu, ^{240}Pu, and ^{242}Pu. The response of the HLNCC to these isotopes is generally given in terms of the mass of ^{240}Pu-effective, which is the mass of ^{240}Pu that would give the same response as the combination of the three even-mass isotopes. The ^{240}Pu-effective mass m^{240}_{eff} is defined[4] as

$$m^{240}_{eff} = 2.49\ m^{238} + m^{240} + 1.57\ m^{242} \quad , \qquad (1)$$

where m^{238}, m^{240}, and m^{242} are the masses of ^{238}Pu, ^{240}Pu, and ^{242}Pu in the fuel assembly.

The ^{240}Pu isotope is the dominant term of the ^{240}Pu-effective mass of typical reactor plutonium (see Table II).

2. Data Analysis. The following notation is used for data analysis of HLNCC measurements.

t = count time in seconds
T = totals count
$(R+A)$ = reals + accidentals count
(A) = accidentals count.

The observed neutron coincidence count rate $\dot{R}$ is given by

$$\dot{R} = \frac{(R+A) - (A)}{t} \text{ counts/s} \quad . \tag{2}$$

An estimate of the standard deviation $\sigma_{\dot{R}}$ of the observed coincidence count rate is given by

$$\sigma_{\dot{R}} = \frac{\sqrt{(R+A) + (A)}}{t} \text{ counts/s} \quad . \tag{3}$$

However, this observed coincidence count rate $\dot{R}$ decreases as the total count rate increases because of deadtime loss in the electronics instrumentation used with the HLNCC. The amount of decrease is determined experimentally by measuring the $\dot{R}$ of a ^{252}Cf neutron source in the presence of various high background rates from large AmLi neutron sources.

The observed coincidence count rate must be adjusted to correct for this deadtime loss. The corrected coincidence count rate $\dot{R}_0$ is related to the observed coincidence count rate $\dot{R}$ by the equation[4]

$$\dot{R}_0 = \dot{R}\, e^{\delta \dot{T}} \quad , \tag{4}$$

where $\dot{T}$ = the observed totals count rate, T/t, and δ = deadtime constant = 2.4×10^{-6} s. The deadtime-corrected coincidence count rate $\dot{R}_0$ is used for plutonium assay because it is proportional to the ^{240}Pu-effective content of the fuel assembly.

The estimate of the standard deviation of the measurement must also be corrected for deadtime losses. The corrected estimate $\sigma_{\dot{R}_0}$ is

$$\sigma_{\dot{R}_0} = \left[\frac{\sqrt{(R+A) + (A)}}{t} \right] e^{\delta \dot{T}} \quad . \tag{5}$$

B. Preliminary Experimental Measurements

Three calibration curves[5] were generated* using various mixtures of MOX rods and UO_2 rods and two different fuel assembly rod patterns (28-rod and 37-rod assemblies). The fuel assemblies were placed vertically inside the neutron detector, extending out both the top and bottom. The loading of both MOX and UO_2 was changed by varying the number and type of fuel rods in the assembly.

1. MOX Rods Only, 28-Rod Assembly. The lower curve of Fig. 5 was generated by placing various amounts (4 to 28) of MOX fuel rods into the 28-rod assembly (Fig. 2). For example, in the measurement that corresponds to a linear loading of 0.61 g of ^{240}Pu-effective/cm, 12 MOX fuel rods were in the assembly. The linear fuel loading (UO_2 + PuO_2) varies directly with the number of fuel rods in the assembly. For each measurement shown in Fig. 5, the fuel rods were distributed throughout the assembly. The upward curvature observed[6] is typical of that seen when neutron multiplication is present. This observed curvature is due primarily to fast neutron multiplication in the ^{238}U in the assembly (see Sec. III.C).

---

*These calibration curves were generated from data taken by A. Ramalho and J. Womack of the IAEA and T. D. Reilly and J. E. Foley of Los Alamos National Laboratory.

2. Mixtures of MOX Rods and UO_2 Rods, 28-Rod Assembly. The middle curve of Fig. 5 was generated by keeping the 28-rod assembly always fully loaded with mixtures of MOX rods and UO_2 rods. By keeping the assembly fully loaded, we were able to keep the average linear fuel (UO_2 + PuO_2) loading of the assembly nearly constant (∿342 g/cm). The desired values of ^{240}Pu-effective loadings were obtained by using the appropriate number of MOX rods; the remaining fuel rod positions in the assembly were filled with UO_2 rods. For example, in the measurement that corresponds to a linear loading of 0.5 g/cm of ^{240}Pu-effective, 10 MOX rods and 18 UO_2 rods were in the assembly. For the measurement at 1.0 g/cm of ^{240}Pu-effective, 20 MOX rods and 8 UO_2 rods were used. The MOX rods were spaced reasonably uniformly throughout the assembly. Twenty-eight rods were always used. Note that the middle curve of Fig. 5 is linear with ^{240}Pu-effective mass.

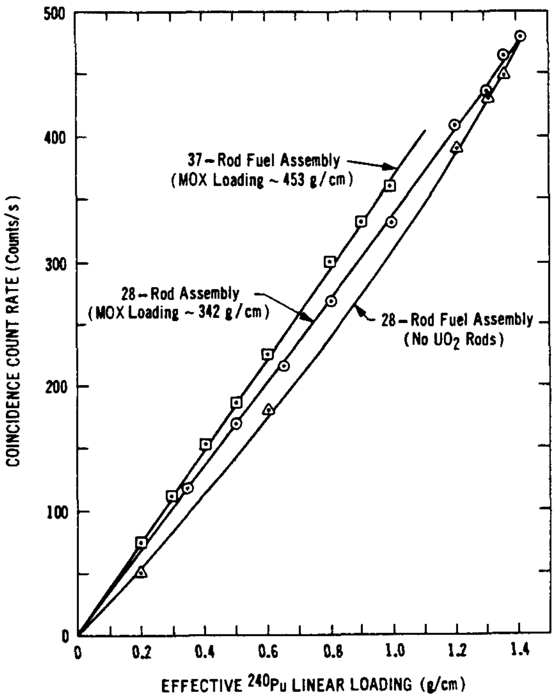

Fig. 5.
Coincidence count rate as a function of ^{240}Pu-effective linear loading in various fuel rod configurations.

3. Mixtures of MOX Rods and UO_2 Rods, 37-Rod Assembly. The upper curve of Fig. 5 was generated by keeping the 37-rod assembly (Fig. 4) always fully loaded with mixtures of MOX rods and UO_2 rods. The linear fuel loading (UO_2 + PuO_2) of the assembly was always ∿453 g/cm. The desired values of ^{240}Pu-effective were again achieved by using the proper numbers of MOX rods; the remaining fuel rod positions were filled with UO_2 rods. For example, in the measurement that corresponds to 1.0 g/cm ^{240}Pu-effective, 20 MOX rods and 17 UO_2 rods were used. As MOX rods were removed to reduce the ^{240}Pu-effective linear loading, they were replaced with UO_2 rods. The upper curve of Fig. 5 is linear with the ^{240}Pu-effective loading; no curvature is observed. The coincidence count rates for all values of ^{240}Pu-effective loadings are higher for the 37-rod assembly (fuel loading of ∿453 g/cm) than for the 28-rod assembly (fuel loading of ∿342 g/cm).

C. Discussion of Experimental Results

We see from Fig. 5 that the coincidence count rate of MOX fuel assemblies is strongly dependent on the linear fuel loading (UO_2 + PuO_2) of the assembly: (1) when the linear fuel loading is constant, the coincidence count rate is linear with plutonium content (at least for low plutonium content); (2) when the linear fuel loading increases ($\sim$453 g/cm vs $\sim$342 g/cm), the coincidence count rate increases.

The coincidence count rate per gram of ^{240}Pu-effective loading of the fuel assembly is shown in Fig. 6 for the three cases given in Fig. 5. We see that the response per gram of ^{240}Pu-effective is constant if the linear fuel loading of the assembly is constant; that is, the coincidence count rate is not a function of the plutonium content, at least for the plutonium loadings studied. Also, the higher the linear fuel loading, the higher the response. The response per gram of ^{240}Pu-effective of the assembly with a linear fuel loading of $\sim$453 g/cm is about 10% higher than that of the assembly with a linear fuel loading of $\sim$342 g/cm. The linear fuel loading, which is primarily ^{238}U, has a large effect on the response.

In Fig. 7, we present the data from Fig. 6 in another way: the coincidence count rate per gram of ^{240}Pu-effective is plotted as a function of the linear fuel loading of the assembly. We see that the response of the HLNCC is a function of the linear fuel loading; the higher the loading, the higher the response. It thus appears that the linear fuel loading (more specifically, the ^{238}U loading) is causing the observed increase in coincidence count rate.

The results presented in Figs. 5, 6, and 7 lead us to the following preliminary conclusions about using the HLNCC to assay MOX fuel assemblies.

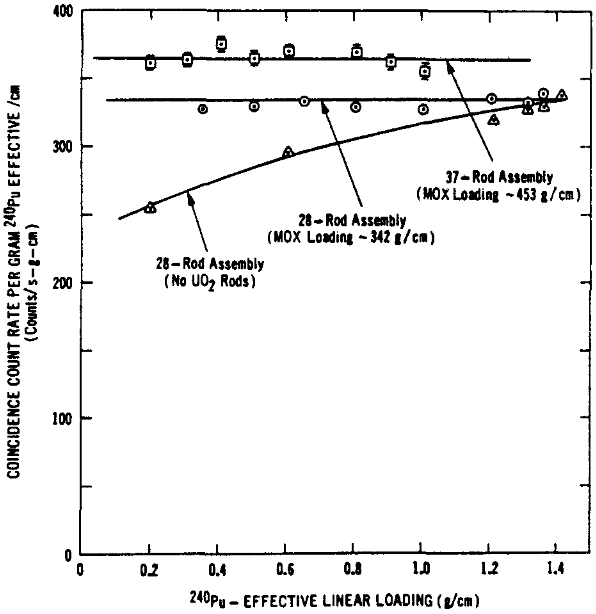

Fig. 6.
Coincidence count rate per gram of ^{240}Pu-effective per centimeter as a function of plutonium loading.

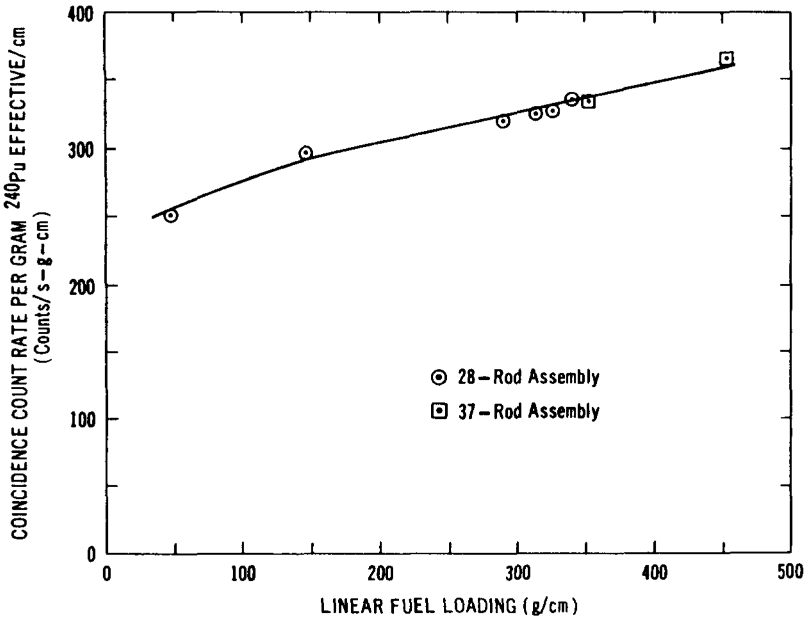

Fig. 7.
Coincidence count rate as a function of linear fuel loading (UO_2 + PuO_2).

1. The linear fuel loading, which is primarily ^{238}U, has a large effect on the response of the HLNCC.
2. Because the response of the HLNCC is strongly dependent on the ^{238}U loading in the assembly, we should view the MOX assembly as a uranium assembly containing a small amount of plutonium, rather than as a plutonium assembly.
3. Possible reasons that the coincidence response of a gram of plutonium in a heavily loaded assembly is higher than it is in a less heavily loaded one are (1) there is more fast multiplication in the heavily loaded assembly (because there is more ^{238}U) and (2) the neutron deection efficiency of the HLNCC is higher in the more heavily loaded assembly because there is less neutron leakage out the ends of the HLNCC.

D. Additional Experimental Measurements

We made additional experimental measurements to understand the effects observed and to demonstrate the sensitivity of the coincidence count rate on various parameters, such as fissile loading, rod diameter, and rod pattern.

We have experimentally established that a MOX fuel assembly can be simulated with UO_2 fuel rods and a neutron source, which provides the spontaneous fission neutrons. Figure 8 shows the results obtained when UO_2 rods were used

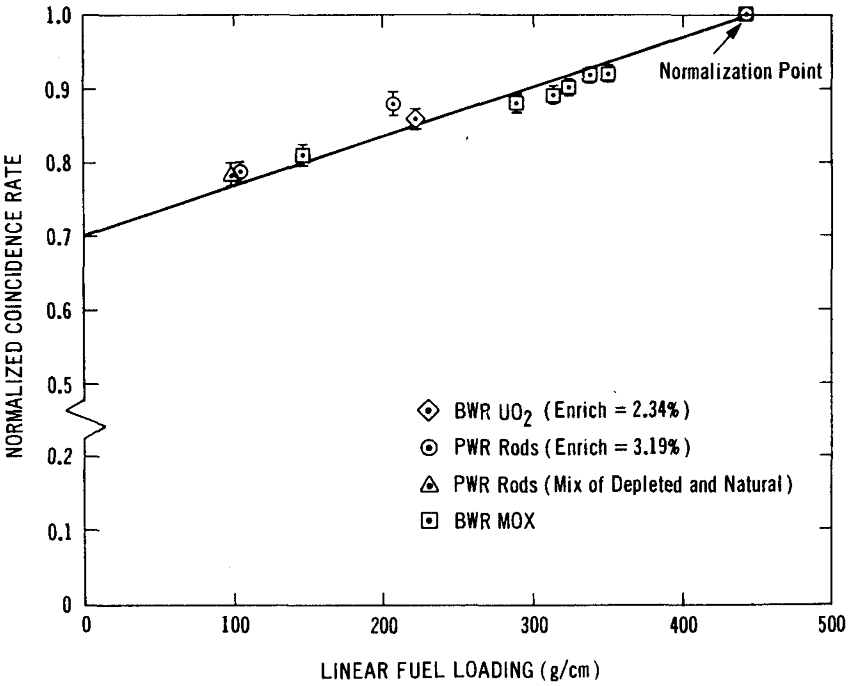

Fig. 8. Normalized coincidence rate as a function of the linear fuel loading for various types and amounts of fuel rods in the 37-rod assembly.

in the 37-rod assembly and a californium neutron source, which was located in the midplane of the neutron counter, provided the source of spontaneous fission neutrons. The californium neutron source was placed in various empty rod positions and the coincidence count rate was volume-averaged to represent a uniform neutron source density in the assembly. The results obtained using UO_2 rods and the californium neutron source are equivalent to those found in Fig. 7. These results are also shown in Fig. 8.

In addition to the BWR MOX rods and UO_2 rods, whose characteristics are shown in Table II and Table III, respectively, we also had available depleted, natural, and 3.19%-enriched pressurized-water reactor (PWR) fuel rods that contained UO_2 only. These rods, which have a smaller diameter than the BWR rods, were used in conjunction with the californium neutron source to investigate the effects on coincidence count rate of fissile loading and rod diameter. The characteristics of these PWR rods are given in Table IV. The results of the measurements made with these PWR rods, which are also shown in Fig. 8, fall on the same curve as that of the larger diameter BWR rods (both the MOX rods and the UO_2 rods).

TABLE IV

CHARACTERISTICS OF PWR FUEL RODS

Rod diameter	1.08 cm
Clad material	zirconium
Weight UO_2/rod	∿704 g
Linear fuel loading (UO_2)	5.77 g/cm
Enrichment	3.19% wt% ^{235}U
	natural (0.72%)
	depleted (0.2%)

1. Effect of Fissile Loading. We conclude from the measurements made with the PWR rods that the fissile loading has little effect on the coincidence count rate, at least for low fissile loadings. The results obtained with 18 PWR rods with enrichments of 3.19% in the 37-rod pattern (giving a linear fuel loading of ∿104 g/cm) are the same as the results obtained using rods with lower enrichment (a mixture of depleted and natural). There is apparently little neutron multiplication in the fissile material in the fuel assembly.

2. Effect of Fuel Rod Diameter. The diameter of the fuel rod has little effect on the coincidence count rate. The small-diameter PWR rods give essentially the same response as the larger diameter BWR rods when the linear fuel loading is the same.

3. Effect of Fuel Rod Pattern. The results in Figs. 5, 6, 7, and 8 for different fuel rod patterns (28-rod and 37-rod) and assemblies filled to varying amounts show that the coincidence count rate is insensitive to the rod pattern. This is probably not true when the number of fuel rods in the assembly is small. When more than a few rods are present, the assembly looks homogeneous.

4. Axial Neutron Efficiency. The axial coincidence response of a californium neutron source in a fuel assembly is shown in Fig. 9. The figure shows that a small, but not insignificant, part of the coincidence count rate comes from those portions of the assembly that are located outside the neutron detector. The ends of the fuel assembly should thus extend at least 30 cm beyond the ends of the detector.

5. Effect of Guide Tube. A 12.7-cm-i.d. aluminum tube with a wall thickness of 0.64 cm was placed around the assembly to simulate a guide tube for inserting the assembly into the neutron detector. This guide tube increased the coincidence count rate by 4%.

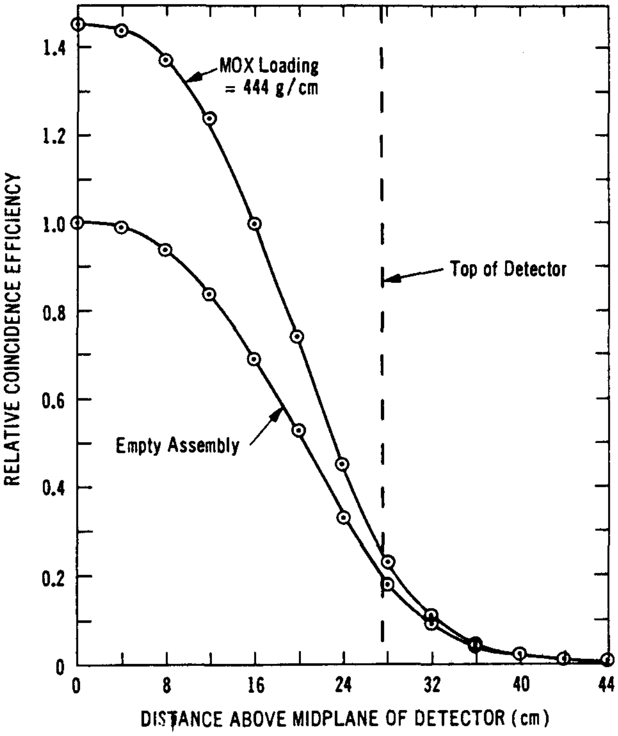

Fig. 9.
Relative coincidence counting efficiency as a function of axial position in the HLNCC for both an empty and a heavily loaded (444 g/cm) assembly.

E. Monte Carlo Calculations

To understand the HLNCC response for MOX rods, calculations were performed with a specially adapted version[7] of the neutron-photon Monte Carlo code MCNP.[8] In this version, the shift-register circuitry of the electronics package[4] is simulated to determine the actual coincidence response for neutrons emitted from either spontaneous or induced fission events.

The measurement geometry was modeled as accurately as possible. The HLNCC portion of the model included the cadmium liners, the polyethylene moderator, and the 4-atm ^{3}He tubes. The MOX and UO_2 rod models included clad, gap, and fuel. Both the 28-rod and 37-rod configurations were investigated. Although the experimental results from the 28-rod model and the 37-rod model differed slightly, the calculational results for the two configurations are within statistical errors. Therefore, only the 37-rod results are discussed.

Calculated results for the 37-rod array are shown in Fig. 10. The two calculational curves are normalized to the result obtained for the fully loaded array. The experimental curve is normalized to an estimated value for a fully loaded array. For the MOX-rods-only calculational case, only the number of rods necessary for a given linear ^{240}Pu-effective loading was used. For the MOX + UO_2 case and the experimental case, the 37-rod assembly

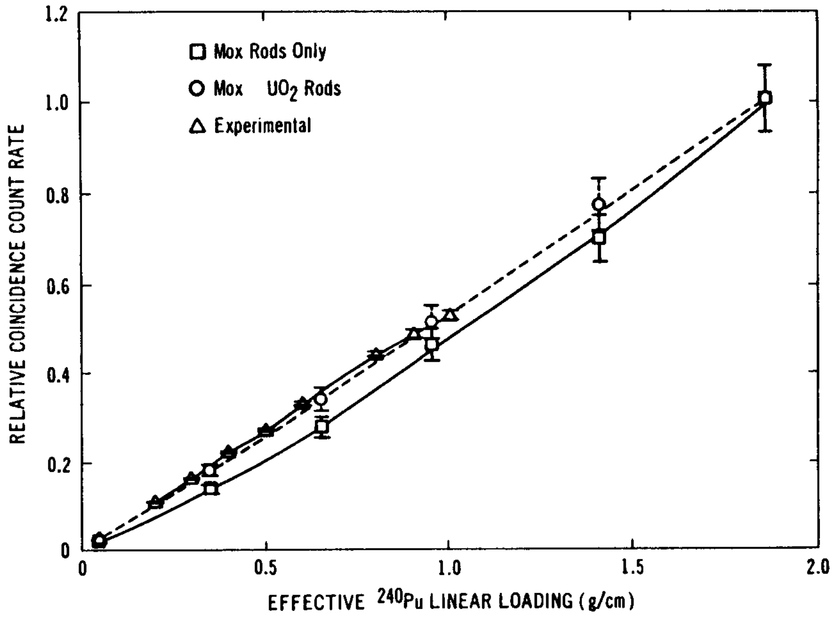

Fig. 10.
Spontaneous fission response: combinations of MOX and UO$_2$ rods.

was always fully loaded with appropriate combinations of MOX and UO$_2$ rods. Error bars shown for the calculated data represent one standard deviation. Errors were typically between 5 and 8%. Reduction of these errors would have required significantly longer computer execution times. Within these errors, experimental and calculational results are in very good agreement.

The calculated results include only neutrons emitted through spontaneous fission events in the plutonium isotopes. Effects of neutrons emitted from other events such as (α,n) emissions are not included. In a coincidence counter, (α,n) neutrons yield a response only when they induce fissions and the resulting fission neutrons are counted. For the 37-rod assembly fully loaded with MOX rods, the (α,n) neutrons contributed no more than 8% to the total coincidence response. Rod configurations with smaller linear plutonium loadings or smaller total fuel loadings would have smaller (α,n) contributions. Because the (α,n) contribution was small (in most cases, less than the statistical errors), the additional computer time needed to determine the (α,n) contribution was deemed unnecessary.

Sample multiplication effects on the coincidence response are shown in Figs. 11 and 12. The multiplication results from induced fission events caused by spontaneous-fission-emitted neutrons. To the HLNCC, a spontaneous fission event and subsequent induced fissions appear to be a single event emitting more

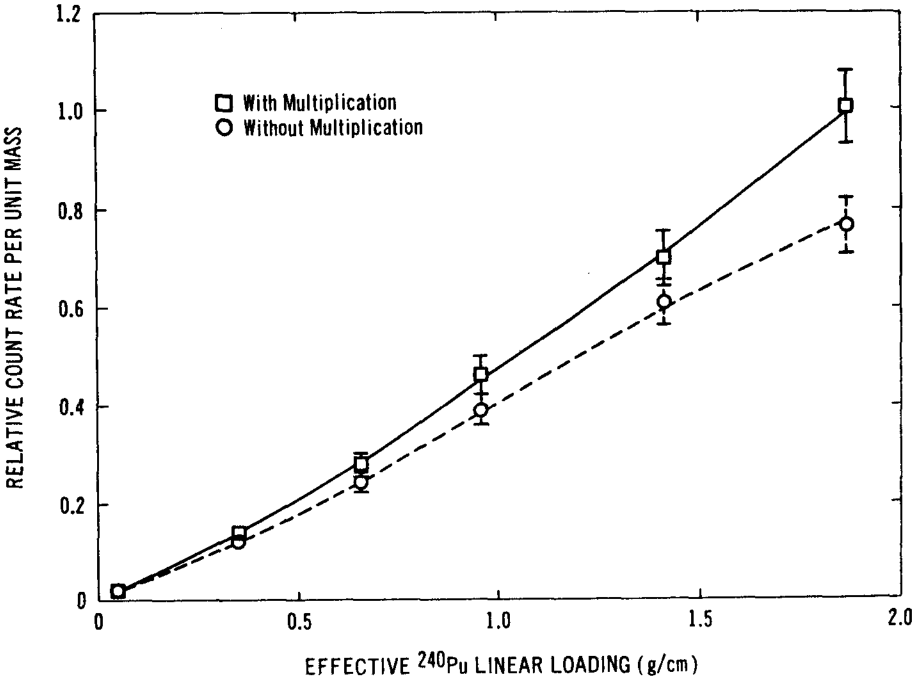

Fig. 11.
Spontaneous fission response: MOX rods only.

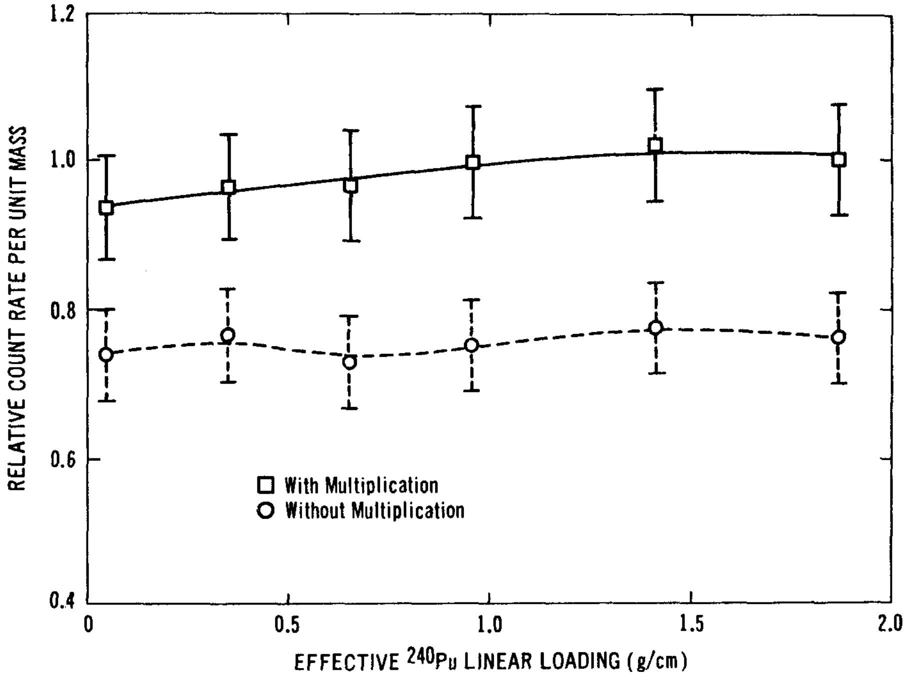

Fig. 12.
Spontaneous fission response: MOX and UO_2 rods.

neutrons per initial event than the average number normally emitted in a spontaneous fission. These additional neutrons per event result in a higher coincidence count rate than that obtained when only the spontaneous fission neutrons are detected. The multiplication effects are nonlinear. Larger samples have more multiplication and the shift-register response is greater for larger multiplication.

In Fig. 11, the coincidence response per unit mass of ^{240}Pu-effective is shown with and without multiplication for the MOX rods only. For a 37-rod array fully loaded with MOX rods, multiplication enhances the signal by approximately 30%. Multiplication effects in arrays with fewer rods are smaller, approaching zero for five rods or less.

Figure 12 shows the multiplication effects for an array fully loaded with combinations of MOX and UO_2 rods. In this configuration the response per unit mass of ^{240}Pu-effective is constant within statistics. Multiplication is a constant 30% effect.

For the partially loaded array, the detector efficiency may increase as more and more MOX rods are added. With additional rods, neutron leakage from the ends of the rods decreases. Less leakage means that more neutrons are detected per initial event. While this effect may be occurring in the HLNCC, it is not determinable within the calculational statistics.

As shown in Table V, 79% of the neutrons that induce fission in the MOX and UO_2 rods have energies above 1.0 MeV. Of the induced fissions, 71% occur in ^{238}U and 18.7% in ^{239}Pu. The remaining fissions occur in ^{235}U and the other plutonium isotopes. Thus, although the neutron spectrum in the sample area of the HLNCC remains fairly hard, high-energy induced fissions provide enough multiplication to increase the response by as much as 30%.

TABLE V

ENERGY OF NEUTRONS INDUCING FISSIONS IN A MOX ARRAY OF 37 RODS

Energy Range	Per Cent Causing Fission
0.0 eV to 1.0 eV	4.06
1.0 eV to 1.0 eV	10.25
1.0 keV to 100.0 keV	1.35
100.0 keV to 1.0 MeV	5.08
Above 1.0 MeV	79.25

IV. SUGGESTED ASSAY PROCEDURES

Two approaches to MOX fuel assembly assay with the HLNCC are discussed. One is used when we do not have a calibration curve for the samples being measured and we must generate it with standards that are available; the other is used when we have a calibration curve from an earlier calibration, but the response of the HLNCC has changed because of drifts in the electronics, slight differences in high-voltage and discriminator settings, etc. Figure 13 illustrates the steps required for each approach.

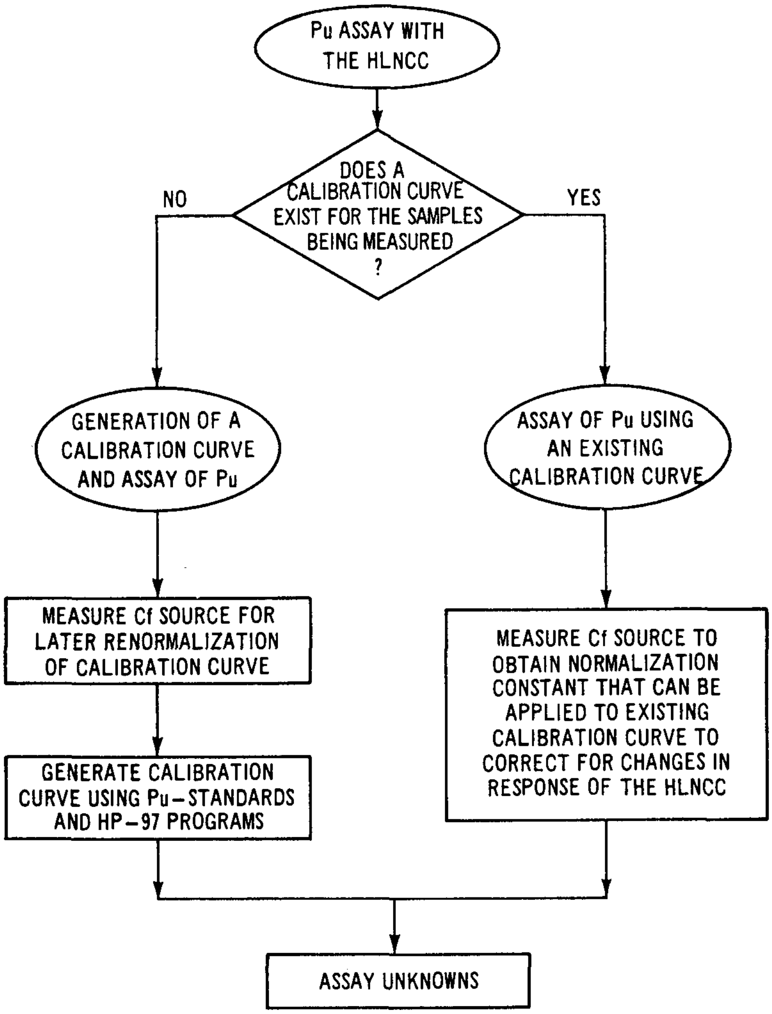

Fig. 13.
Assay procedures used with the HLNCC.

A. Generation of the Calibration Curve for a MOX Fuel Assembly

1. Assemble the HLNCC in standard manner.[9] Use instrument settings:
   a. High voltage = 1500 V,
   b. Discriminator = 1.5 V, and
   c. Gate width = 32 μs.

2. Make a 1000-s count of the ^{252}Cf neutron source and use the DATA-1F HP-97 program with deadtime correction for automatic analysis of the data. The Appendix describes three programs for the HP-97 that can be used for data analysis of MOX fuel assembly measurements. The ^{252}Cf neutron source is stored in the top reflector cap (Fig. 14). The corrected coincidence count rate $\dot{R}_0$ of this source and the calibration date are needed for normalization of the calibration curve at a later date. Record the following information.

   HLNCC Identification: ____________________
   High Voltage: ____________________
   Discriminator: ____________________
   Gate Width: ____________________
   ^{252}Cf Source Identification: ____________________
   Date: ____________________
   $\dot{R}_0$: ____________________ $\sigma_{\dot{R}_0}$: ____________________

3. Collect the data for the construction of the calibration curve using the DATA-1F program for the HP-97 calculator. Load the fuel assembly with the appropriate number of MOX and UO_2 rods to give the correct average fuel and ^{240}Pu-effective loadings. Count the assembly several times for ∿300 s. Change the ^{240}Pu-effective loading, but keep the average fuel loading constant by exchanging MOX rods for UO_2 rods. Repeat the measurements for each new ^{240}Pu-effective loading.

4. Draw a calibration curve using these calibration data with the corrected coincidence count rate $\dot{R}_0$ on the y-axis and the effective ^{240}Pu mass on the x-axis. An example of a calibration curve is shown as the

Fig. 14.
The californium neutron source is stored in the top reflector cap of the HLNCC.

top curve of Fig. 5. This hand-drawn calibration curve can be used to assay "unknown" MOX assemblies. The mass can be read directly from this calibration curve after the corrected coincidence count rate $\dot{R}_0$ of the unknown assembly is measured. Assaying by reading from a hand drawn calibration curve is an acceptable procedure; however, if a large number of samples are to be assayed, it is easier to let the HP-97 calculate the mass automatically from a least-squares fitted calibration curve.

5. Least-Squares Fitted Calibration Curve

You can obtain an unweighted least-squares fit of the calibration data to the equation

$$\dot{R}_0 = A_0 + A_1M + A_2M^2 \quad ,$$

using an HP-97 calculator FIT-1 program (see Appendix). M is the ^{240}Pu-effective linear loading of the assembly.

a. Load FIT-1 into the HP-97 (two sides of card).
b. Press A to initialize the program.
c. Enter the calibration data in pairs (M, $\dot{R}_0$) as follows.

(1) For the first calibration point:
   - Enter M
   - Press ENTER
   - Enter $\dot{R}_0$
   - Press B.

The data are then printed for reference.

(2) Repeat the above steps for the remaining calibration points.

d. Press D. The least-squares fit parameters are printed in the order A_0, A_1, A_2.

B. Assay of MOX Fuel Assemblies Using the Generated Calibration Curve

Automatic analysis of the data and calculation of the assays in grams per centimeter of ^{240}Pu-effective from the least-squares fitted calibration curve are done with the HP-97 ASSAY-1F program. This program is based on a simplified approach to the error analysis. To use this program, you must input the calibration constants A_0, A_1, and A_2 that were obtained from the least-squares fit (using FIT-1) of the calibration data.

1. Assay Program

a. Load program ASSAY-1F into the HP-97 (two sides of card).

b. Enter the least-squares fitted constants A_0, A_1, and A_2 and a "normalization" constant (see Sec. IV.D) into the HP-97.

(1) Load constant A_0 into Register A.
   - Enter A_0
   - Press STO
   - Press A.

(2) Load constant A_1 into Register B.

(3) Load constant A_2 into Register C.

(4) Load number 1 into Register D.

2. Output

An output of a sample assay problem is shown in Table VI. The program prints the final output as grams per centimeter of ^{240}Pu-effective.

TABLE VI

SAMPLE OUTPUT OF ASSAY 1F

1000	*** ←	count time t in seconds
1212461	*** ←	totals
49825	*** ←	(R+A)
46867	*** ←	(A)
1.0000	*** ←	normalization constant
1.0029	*** ←	coincidence deadtime correction factor
2.9665	*** ←	$\dot{R}_0$ (deadtime corrected)
0.3119	*** ←	$\sigma_{\dot{R}_0}$
10.5123	*** ←	$\%(\sigma_{\dot{R}_0})$
7.0322	*** ←	linear loading of sample, g ^{240}Pu-effective/cm
0.7393	*** ←	standard deviation g ^{240}Pu-effective/cm

C. Information to Record for Future Assays

You have developed a calibration curve for MOX fuel assemblies and have used it to assay unknown samples. To enable another inspector to use this calibration curve at a later time, complete Table VII. Without this information, the calibration curve cannot be used again.

D. Assay of MOX Fuel Assemblies Using an Existing Calibration Curve

The ^{252}Cf neutron source is used to renormalize the response of the instrument so that the existing calibration curve can be used.

1. Make a 1000-s count of the ^{252}Cf neutron source and use the DATA-1F program for collection of the data.
2. Calculate a normalization constant:

$$N = \frac{\dot{R}_0 \text{ (on original calibration date) } \times e^{-7.166 \times 10^{-4} \times d}}{\dot{R}_0 \text{ (present reading)}} , \qquad (6)$$

where d is the time in days since the original calibration. This normalization constant corrects for the change in response of the HLNCC since the original calibration curve was made. The values of both $\dot{R}_0$ (on original calibration date) and the original calibration date are found in the calibration record (Table VII).

3. Assay of unknowns.
   a. Load program ASSAY-1F into the HP-97 (two sides of card).
   b. Enter the least-squares fitted constants A_0, A_1, A_2 that are found in the calibration record (Table VII) and the normalization constant N into the HP-97:
      (1) Load constant A_0 into Register A.
      (2) Load constant A_1 into Register B.
      (3) Load constant A_2 into Register C.
      (4) Load normalization constant N into Register D.
   c. You can now assay unknown MOX fuel assemblies.

TABLE VII

CALIBRATION RECORD

1. INSTRUMENT INFORMATION

HLNCC Identification: ______________________
High Voltage: ______________________
Discriminator Voltage: ______________________
Gate Width: ______________________

2. NORMALIZATION INFORMATION (using DATA-1F program)

^{252}Cf Source Identification: ______________________
Coin. Count Rate $\dot{R}_0$ (deadtime corrected): ______________
Standard Deviation $\sigma_{\dot{R}_0}$: ______________________
Date: ______________________

3. CALIBRATION INFORMATION (using FIT-1 program)

Assembly Geometry: ______________________
____

Least-Squares Fitted Constants:
A_0: ______________
A_1: ______________
A_2: ______________

4. Attach copy of hand-drawn calibration curve.

ACKNOWLEDGMENTS

We wish to thank A. Ramalho, J. Womack, and M. de Carolis of the IAEA and M. S. Krick and T. D. Reilly of Los Alamos for their help in providing much of the information needed for this report.

REFERENCES

1. IAEA Directory of Nuclear Reactors (IAEA, Vienna, 1975), Vol. X, pp. 295-302.

2. G. R. Keepin, Ed., "Nuclear Safeguards Research and Development Program Status Report, September-December 1970," Los Alamos Scientific Laboratory report LA-4605-MS (January 1971), pp. 24-26.

3. M. S. Krick, "Neutron Multiplication Corrections for Passive Thermal Neutron Well Counters," Los Alamos Scientific Laboratory report LA-8460-MS (ISPO-89) (July 1980).

4. M. S. Krick and H. O. Menlove, "The High-Level Neutron Coincidence Counter (HLNCC): Users' Manual," Los Alamos Scientific Laboratory report LA-7779-M (ISPO-53) (June 1979).

5. S. D. Gardner, Ed., "Nuclear Safeguards Research and Development Program Status Report, May-August 1978," Los Alamos Scientific Laboratory report LA-7616-PR (April 1979), pp. 29-31.

6. J. L. Sapir, Comp., "Nuclear Safeguards Research and Development Program Status Report, January-April 1977," Los Alamos Scientific Laboratory report LA-6849-PR (August 1977), pp. 5-6.

7. J. L. MacDonald, "A Monte Carlo Neutronic and Electronic Model for Thermal-Neutron Coincidence Counting," Trans. Am. Nucl. Soc. 32, 635 (June 1979).

8. Los Alamos Group X-6, "MCNP - A General Monte Carlo Code for Neutron and Photon Transport," Los Alamos Scientific Laboratory report LA-7396-M, Revised (November 1979).

9. J. E. Foley, "Field Manual for the High-Level Neutron Coincidence Counter," Los Alamos National Laboratory report (to be issued).

APPENDIX*

BASIC HP-97 PROGRAM SET FOR HLNCC

I. INTRODUCTION

A. Notation

M = ^{240}Pu-effective linear loading (g/cm) of fuel assembly

$\dot{R}_0$ = Corrected coincidence rate (deadtime corrected)

$\dot{T}_0$ = Totals rate (deadtime corrected)

B. Purposes of Programs

1. Program DATA-1F. Used for collection of $\dot{T}_0$ and $\dot{R}_0$ data from a fuel assembly.

2. Program FIT-1. Used to determine a quadratic calibration curve by least-squares fitting of data to the form

$$\dot{R}_0 = A_0 + A_1 M + A_2 M^2 .$$

3. Program ASSAY-1F. Used to assay a fuel assembly from the calibration curve and the measured $\dot{R}_0$ rate.

---

*This appendix is based on programs written by M. S. Krick, Los Alamos National Laboratory.

II. PROGRAM DATA-1F

A. Purpose

Calculates the totals rate and corrected coincidence rate. Correction is made for deadtime.

B. Procedure

1. Load program card DATA-1F (one side).
2. Proceed with measurements.

C. Sample Output

```
      1000.       ***   Time
   1212461.       ***   Totals
     49825.       ***   R+A
     46867.       ***   A

         1.0029   ***   Coincidence deadtime correction factor

         2.9161   ***   Corrected coincidence rate $(\dot{R}_0)$
         0.3066   ***   Standard deviation of real coincidence rate
        10.5123   ***   Standard deviation as per cent.
```

D. Program Listing

Same as for ASSAY-1F, except that mass calculation is not performed.

III. PROGRAM FIT-1

A. Purpose

Performs an unweighted least-squares fit to the equation

$$\dot{R}_0 = A_0 + A_1 M + A_2 M^2 \quad ,$$

where M is the grams of ^{240}Pu-effective linear loading per centimeter of the fuel assembly and $\dot{R}_0$ is the coincidence rate obtained from DATA-1F.

B. Procedure

1. Load program card FIT-1 (two sides).
2. Press A to initialize.
3. Enter data pairs (M, $\dot{R}_0$) as follows.
    (a) Enter M
    (b) Press ENTER
    (c) Enter $\dot{R}_0$
    (d) Press B.
    The data is printed for reference.
4. Press D to calculate parameters. The parameters are printed in the order A_0, A_1, A_2.

C. Extra Feature

After the fitting parameters have been calculated, $\dot{R}_0$ can be calculated for any M as follows.

1. Enter M
2. Press E
3. $\dot{R}_0$ is displayed.

D. Example

The following table provides an example of typical calibration data:

M(^{240}Pu-Effective) (g/cm)	$\dot{R}_0$(Corrected Coincidence Rate) (s^{-1})
0.38	0.223
7.67	5.051
43.4	32.66
54.9	43.80
81.3	69.75

The output of the program lists the input data and the three calculated parameters A_0, A_1, and A_2.

```
 0.380000000     ***
 0.223000000     ***

 7.670000000     ***
 5.051000000     ***
                              calibration data
43.40000000      ***
32.66000000      ***

54.90000000      ***
43.80000000      ***

81.30000000      ***
69.75000000      ***


-0.072252074     ***           A0
 0.650412138     ***           A1     fitting parameters
 0.002576071     ***           A2
```

E. Program Listing

Program FIT-1

```
001    *LBLA
002     DSP9
003     CLRG
004      R/S
005    *LBLB
006     STO0
007       R↓
008     STO1
009     GSBC
010     RCL0
011     X=0?
012     GTOD
013        1
014     ST+9
015      R/S
016     GTOB
017    *LBLC
018     RCL1
019     PRTX
020     RCL0
021     PRTX
022      SPC
023     RCL0
024     ST+3
025     RCL1
026     ST+2
027      X^2
028     ST+4
029     RCL1
030        x
031     ST+5
032     RCL1
033        x
034     ST+6
035     RCL0
036     RCL1
037        x
038     ST+7
039     RCL1
040        x
041     ST+8
042      RTN
043    *LBLD
044     RCL9
045     RLC4
046        x
047     RCL6
048        x
049        2
050     RCL2
051        x
052     RCL5
053        x
054     RCL4
055        x
056        +
057     RCL4
058        3
059      Y^x
060        -
061     RCL5
062      X^2
063     RCL9
064        x
065        -
066     RCL2
067      X^2
068     RCL6
069        x
070        -
071     STOD
072     RCL3
073     RCL4
074        x
075     RCL6
076        x
077     RCL2
078     RCL5
079        x
080     RCL8
081        x
082        +
083     RCL7
084     RCL5
085        x
086     RCL4
087        x
088        +
089     RCL4
090      X^2
091     RCL8
092        x
093        -
094     RCL5
095      X^2
096     RCL3
097        x
098        -
099     RCL7
100     RCL2
```

Program FIT-1 (continued)

```
101      x
102    RCL6
103      x
104      -
105    RCLD
106
107     SPC
108    PRTX
109    STOA
110    RCL9
111    RCL7
112      x
113    RCL6
114      x
115    RCL3
116    RCL5
117      x
118    RCL4
119      x
120      +
121    RCL2
122    RCL8
123      x
124    RCL4
125      x
126      +
127    RCL4
128     X2
129    RCL7
130      x
131      -
132    RCL8
133    RCL5
134      x
135    RCL9
136      x
137      -
138    RCL2
139    RCL3
140      x
141    RCL6
142      x
143      -
144    RCLD
145      ÷
146    PRTX
147    STOB
148    RCL9
149    RCL4
150      x
151    RCL8
152      x
153    RCL2
154    RCL7
155      x
156    RCL4
157      x
158      +
159    RCL2
160    RCL5
161      x
162    RCL3
163      x
164      +
165    RCL4
166     X2
167    RCL3
168      x
169      -
170    RCL7
171    RCL5
172      x
173    RCL9
174      x
175      -
176    RCL2
177     X2
178    RCL8
179      x
180      -
181    RCLD
182
183    STOC
184    PRTX
185     SPC
186     SPC
187     SPC
188     SPC
189     SPC
190     R/S
191   *LBLE
192    STOE
193     X2
194    RCLC
195      x
196    RCLB
197    RCLE
198      x
199      +
200    RCLA
201      +
202     R/S
```

IV. PROGRAM ASSAY-1F

A. Purpose

Determines ^{240}Pu-effective linear loading M from coincidence rate $\dot{R}_0$ and the calibration curve

$$\dot{R}_0 = A_0 + A_1M + A_2M^2 .$$

B. Procedure

1. Load program card ASSAY-1F (two sides).
2. Enter data as follows.

Datum	Register
A_0	A
A_1	B
A_2	C
N (norm. const.)	D

3. Proceed with measurements.

C. Sample Output (Program ASSAY-1F)

```
   1000.       ***
1212461.       ***    Raw data
  49825.       ***
  46867.       ***

     0.9830    ***    Normalization constant
     1.0029    ***    Coincidence deadtime correction factor

     2.9161    ***    Corrected coincidence rate (Ṙ0)
     0.3066    ***    Standard deviation on above
    10.5123    ***    Standard deviation as per cent

     6.9127    ***    240Pu-effective linear loading (g/cm)
     0.7167    ***    Standard deviation on above
```

D. Program Listing

Program ASSAY-1F

```
001    *LBLA
002     DSP4
003     RCL1
004     PRTX
005     RCL2
006     PRTX              raw data
007     RCL3
008     PRTX
009     RCL4
010     PRTX
011      SPC
-----------------------------------------------
012     RCLD
013     X=0?
014     GTOD
015        1              normalization constant
016     STOD
017    *LBLD
018      SPC
-----------------------------------------------
019     RCL2
020     RCL1
021      ÷
022     RCLD
023      √X
024      x
025     STO7
026        .              corrected totals rate
027        6
028      EEX
029      CHS
030        6
031       x
032      e^x
033     RCL7
034       x
035     STO5
-----------------------------------------------
036     RCL3
037     RCL4
038       -
039     STO6
040     RCLD
041       x
042     RCL1
043      ÷
044        2
045        .              corrected coincidence rate
046        4
047      EEX
048      CHS
049        6
050     RCL7
051       x
```

Program ASSAY-1F (continued)

```
052      e^x
053     PRTX
054      SPC
055     RCL5
056     PRTX
057      SPC
058       R↓
059        x
060     PRTX            relative standard deviation
061     ST09
062     RCL3
063     RCL4
064        +
065       √X
066     RCL6
067      ÷
068     ST08
069     RCL9
070        x            standard deviation
071     PRTX
072     RCL8
073        1
074        0
075        0            per cent standard deviation
076       x?
077     PRTX
078      SPC
079     RCLC
080     X=0?            linear or quadratic
081     GTOB
082     RCLB
083      X^2
084     RCLA
085     RCL9
086        -
087     RCLC
088        x
089        4
090        x
091        -            mass calculation (quadratic)
092       √X
093     RCLB
094      CH8
095        +
096        2
097      ÷
098     RCLC
099      ÷
```

Program ASSAY-1F (continued)

```
100     *LBLC
101      PRTX
102      RCL8
103        x
104      PRTX
105       SPC                  print mass and standard deviation
106       SPC
107       SPC
108       SPC
109       SPC
110       R/S
111     *LBLB
112      RCL9
113      RCLA
114        -
115      RCLB                  mass calculation (linear)
116       ÷
117      GTOC
118       R/S
```