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**PLUTONIUM PASSIVE ASSAY FACILITY
PLANT INSTRUMENTATION PROGRAM**

**First Quarterly Progress Report
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1. INTRODUCTION

As increased amounts of special nuclear materials are processed by private industry, an important problem that emerges is the measurement and accounting of feed materials, fuel, scrap, and waste. This problem has been recognized by the AEC in its timely inauguration of the Plant Instrumentation Program (PIP). The purpose of the PIP is to demonstrate and evaluate the effectiveness of non-destructive techniques for assay of special nuclear material under actual fuel fabrication and processing plant (privately owned) conditions. Use of the instrumentation is intended to increase the effectiveness of the accountability system, and thereby, assure the early detection of an unauthorized diversion or loss.

Under contract to the AEC, United Nuclear Corporation has undertaken to demonstrate and evaluate the relative merits of passive neutron and gamma assay of plutonium fuel rods, scrap packages, and waste barrels using boron trifluoride neutron detectors and a lithium-drifted germanium gamma detector. This work is being performed at the UNC Plutonium Facility at Pawling, New York.

The Plutonium Passive Assay Project consists of assembling the neutron and gamma systems, preparing calibration standards, constructing a counting shield, and routinely operating the entire system. System operating experience, in conjunction with the results of data analyses, will be used to evaluate system performance. The following criteria will form the basis for the performance evaluation:

1. System reliability and availability
2. System operating costs
3. Limits of error on assays
4. Effect of assay program on nuclear material management
5. Benefits of assay program to facility operations.

2. HIGHLIGHTS

The conceptual designs of the neutron detection systems for both 55-gallon drums and small packages have been completed. All of the equipment and materials for both of these systems have been ordered.

The conceptual design for the gamma detection system has been completed. The lithium-drifted germanium gamma detector and the electronics, except the multichannel analyzer, have been ordered. Architectural and engineering services have been contracted for the design and construction of the counting shield. This shield will house the electronic equipment as well as the detection systems, and it will be equipped with the necessary electrical power, lighting, and HVAC facilities.

Samples of mixed UO_2 - PuO_2 powder have been weighed, analyzed, and packaged for use as calibration standards.

3. CURRENT PROGRESS

3.1 NEUTRON SYSTEM

Neutrons are emitted from fissile and fertile materials by two processes: spontaneous fission and (α, n) reactions on low-Z materials associated with the alpha emitter. As a practical matter, only plutonium isotopes, among the ones of interest, emit sufficient neutrons to be important as signatures.

Of the plutonium isotopes, Pu^{238} , Pu^{240} , and Pu^{242} have high spontaneous fission rates (3×10^4 times that of Pu^{239}). The (α, n) neutron production rates vary by about a factor of four between Pu^{239} and Pu^{240} . The Pu^{239} isotope has a high neutron generation rate from (α, n) reaction, but probably will not be present in significant quantities in the material to be assayed.

The neutrons from both the spontaneous fission and the (α, n) reactions are energetic, having energies chiefly in the 1 to 6-Mev range. These neutrons are detected in BF_3 detectors after having been slowed down by polyethylene which surrounds the chambers.

A block diagram of the neutron assay system is shown in Fig. 1. This system is designed only to determine the total number of neutrons emitted from the sample and thus will not be able to distinguish between neutrons from spontaneous fission and those from (α, n) reactions. It is anticipated that Brookhaven will supply circuitry to allow some coincidence measurements to be performed in order

Fig. 1 — Block Diagram of Neutron Assay System

to investigate the feasibility of isotopic ratio determination in the assayed samples.

3.1.1 Small Package Counter

The well counter will be used for the assay of small packages containing plutonium. Fig. 2 is a sketch showing a plan view of this counter. It consists of a 3-in. thick cylindrical annulus of high-density polyethylene 30-in. long, surrounding a 6-in. diameter central well. Ten 2-in. diameter BF_3 chambers having 30-in. active length are arranged with their centers on a 9-in. diameter circle. The inside and outside surfaces of the polyethylene are covered by 0.030-in. thick cadmium sheet.

The ends of the well are plugged with a scattering material leaving a counting volume 6-in. in diameter and 18 to 20 in. in length.

Orders have been placed for the components of this counter. Some of the smaller ones, such as connectors, have been received, and the polyethylene structure and chambers are expected in late August.

3.1.2 Large Package Counter

Waste barrels — 55-gallon drums containing process waste — will be assayed in a barrel counter. The drums will be placed on a rotating platform and turned continuously while counting, in order to reduce the effect of location of material in the drum on the counting rate. The barrel counter consists of twenty-four 2-in. diameter BF_3 chambers having 30-in. active lengths. They are arranged in three groups of eight and placed around the barrel as shown in Fig. 3. Chambers are paired end-to-end making a 5-ft long counter. One-half inch of polyethylene is placed in "front" of the chambers, i.e., toward the drum, and up to 2 in. of polyethylene is placed in the rear of the chambers. The chambers are separated laterally by 3/4-in. thick strips of polyethylene. The counter is surrounded by 0.030-in. thick cadmium sheets.

Fig. 2 — Plan View of Well Counter

Fig. 3 — Plan View of Barrel Counter

Materials for this counter have been ordered. Some structural materials have been received. The polyethylene is expected about the first of August, and the rotating platform and chambers in September.

3.1.3 Electronics

The electronics package is shown on the block diagram of the neutron assay system in Fig. 1. Both the barrel counter and the well counter will be provided with a preamplifier which will convert the charge collected from a neutron interaction in the chambers to a voltage pulse. The high-voltage power supply will be connected to the chambers through these preamplifiers.

It will not be possible to operate both these counters concurrently. The switching between the two systems will be as shown in Fig. 1. It is anticipated that this switching will be accomplished by changing cable connections rather than by a mechanical switch.

The output of the preamplifier will be fed to a linear pulse amplifier where it will be amplified to a level suitable for operation of the output equipment.

The output system is a simple scaler-timer system. The discriminator input to the scaler will be adjusted to yield a high neutron-to-gamma counting rate. The output of the counters then will be collected in the scaler for a fixed period of time and the results recorded.

No provisions are made for automatic printout of the results; the operator will read the scaler and manually record the output.

3.2 GAMMA SYSTEM

Gamma rays are emitted spontaneously by most fissile and fertile materials. The spectrum of gamma rays emitted from plutonium is shown in Fig. 4 (taken from data presented by LASL at the Safeguards Meeting of October 1969).

Fig. 4 — Gamma-Ray Spectra of Two Different Isotopic Compositions of Plutonium
 as Measured with a High Resolution 50 cm³ Ge(Li) Detector

U^{235} has a prominent line at 185 kev and U^{238} has lines in the 700 to 800-kev region. These lines may be used to identify material for assay and to make estimates of their amount in certain cases.

Another item of information which can be obtained from a gamma scan is an estimate of the Pu^{241} content of plutonium. Reference to Fig. 4 shows two lines at 203.5 and 207.9 kev. The line at 203.5 is due to Pu^{239} , while that at 207.9 is due to U^{237} (from alpha decay of Pu^{241}). Increasing amounts of Pu^{241} in the isotopic mixture are reflected in increasing heights of this line (compare the upper and lower curves). Interpretation of the data is complicated by the fact that the amount of U^{237} depends on the lapsed time from separation. However, this information is often known, and permits estimates of relative Pu^{241} content. The amount of Pu^{241} also may be estimated from the fact that the energy of the line is low, and the attenuation of the radiation is a sensitive function of the enclosing matrix of the material. Since the two lines in the neighborhood of 200 kev have very nearly the same attenuation, the intervening material is less important.

The block diagram of the gamma assay system is shown in Fig. 5.

3.2.1 Detector

The detector for the gamma assay system is a lithium-drifted germanium Ge(Li) coaxial-type crystal. This detector must be stored and operated at liquid nitrogen temperatures, and is thus supplied with a cryostat for cooling. The specifications for the detector are as follows:

Efficiency	5% at 1.33 Mev
Resolution	2.4 kev FWHM at 1.33 Mev 1.5 kev FWHM at 0.123 Mev
Peak to Compton	20 to 1 at 1.33 Mev
Voltage	3000 volts

Fig. 5 — Block Diagram of Gamma Assay System

The efficiency quoted is the percentage of the efficiency of a 3-in. by 3-in. NaI(Tl) detector with a source located at an axial distance of 25 cm from the face. At the energies that the crystal will be used (60 to 700 kev), the efficiency may be greater (7 to 8%) than 5%.

The resolution quoted refers to the full width at half maximum of the line in the output of the preamplifier produced by the total absorption of a gamma ray of the quoted energy in the detector. Stated another way, gamma rays separated in energy by an amount greater than the resolution will be resolved, i.e., appear as separate peaks. By comparison, the resolution of a 3-in. by 3-in. NaI(Tl) detector is of the order of 75 to 100 kev at 1.33 Mev. The high resolution characteristic is the overwhelming reason for the choice of the Ge(Li) detector over the NaI(Tl) detector. The preamplifier is supplied as an integral part of the detector. Delivery is expected in early August.

The power supply will consist of up to ten 300-volt batteries which will be hooked up in series and enclosed in a metal box.

3.2.2 Electronics

This subsystem consists of a linear pulse amplifier, a baseline restorer, a multichannel analyzer, and a printout device. The pulse amplifier receives the pulses from the preamplifier and amplifies them sufficiently for use by the output system. At the same time, they are altered to a Gaussian shape which is suitable for the output system. The baseline restorer circuit is used to correct the "undershoot" at the trailing edge of the pulse from the amplifier. This refinement is necessary only for high pulse rates and following large overloads of the amplifier. High pulse rates are expected in the present system when the plutonium contains fairly large amounts of americium. These pulses normally will not be counted in the output but they will nevertheless traverse the amplifier. The presence of uranium in the samples will produce high-energy pulses which

will overload the amplifier (i.e., produce an output larger than the largest for which the amplifier is designed).

The data collection, a portion of the system, consists of a multichannel analyzer with a teletype page printer. The output pulses from the amplifier are sorted by pulse height (which is linearly dependent on the energy absorbed in the crystal) into as many as 1024 bins or channels. For example, if the gain of the system is adjusted so that each channel represents 0.5 kev, then a "line" at 123 kev would be three bins wide. Provision is made in the multichannel analyzer for "digital offset," i.e., the 1024 channels may be spread over the energy range from 200 to 700 kev. Thus, the interesting parts of the spectrum may be inspected in detail.

The contents of the multichannel analyzer may be printed out channel by channel, the sums of certain groups of channels may be printed out, or a combination of these may be printed. For example, the total counts in the 185-kev line, in the 203.5-kev line, the 207.9-kev line, the lines in the energy range 330 to 420, and the lines in the 700 to 800-kev range may be printed out. Thus, information would be immediately available for estimating the amount of U^{235} , Pu^{239} , Pu^{239}/Pu^{241} ratio, and the amount of U^{238} . The latter number is not of any direct interest, but may be useful in determining a Compton background to be subtracted from lower energy lines.

In addition, the channel-by-channel printout can be used at a later time to investigate details of the spectrum for further analysis.

3.2.3 Traversing Mechanism

The gamma-ray detector will be traversed while the drum is rotating. A collimator will restrict the vertical acceptance angle of the detector so that it may be traversed beyond the end of the drum to a point at which the drum cannot

be seen by the detector. Thus an essentially "infinite" traverse may be accomplished. The conceptual design of the traversing mechanism has been completed.

3.3 COUNTING SHIELD

Several locations were considered for possible placement of the passive assay analysis systems. Considerations that went into the choice of a location included:

1. Background radiation
2. Convenience to process lines
3. Cost of construction of shield.

The background radiation in the area must be low in order that small quantities of material may be assayed. It is estimated that one gram of Pu (8 to 10% Pu^{240}) will produce five counts per second in the neutron counter, and one to two counts per second in the gamma counter (if it is moved back to view the whole drum). Backgrounds lower than these amounts by a factor of ten are deemed desirable to obtain valid data.

The location of the assay system should be as close as feasible to the process lines in order to make the system convenient to use. However, location near the process stream will make it necessary to provide additional shielding, thus adding to the cost of the system.

The cost of construction is determined chiefly by the amount of shielding required and the availability of services such as electrical power, heating, ventilating, and air conditioning.

The locations at the Pawling site which were investigated consisted of:

1. The front office area of the Critical Facility building
2. The maintenance area of the Engineering building

3. The front office area of the Plutonium Facility building
4. The office areas immediately to the west and north of the Plutonium Facility vault
5. The storage area next to the south personnel door in the Plutonium Facility
6. The pad to the south of the Plutonium Facility building
7. A remote area approximately 1/2-mile away.

Neutron surveys were made to obtain background information for the various locations. The data were obtained with a small polyethylene-moderated BF_3 counter and converted to the response expected from the large counter to be used in the assay system. The converted data are shown in Table 1.

TABLE 1 — BACKGROUND RADIATION AT VARIOUS LOCATIONS
AT THE PAWLING SITE

Location		Background Rate, counts/second	
Building	Area	Reactor Off	Reactor On
Critical Facility	Front office	20	2000
Engineering	Maintenance	20	130
Plutonium Facility	Front area	20	170
Plutonium Facility	Near vault, west	150	—
	Near vault, north	40	—
Plutonium Facility	South personnel door	20	100
Plutonium Facility	South pad	20	300
—	Remote area 1/2-mile away	10	—

Based on the considerations, the location selected for the system was the area adjacent to the south personnel door of the Plutonium Facility (Fig. 6). In addition to having one of the lowest background rates, this location has the advantage of being in direct communication with the plutonium laboratory, thus reducing the amount of handling required to transport materials between the areas. The remote location, while having a lower background rate, was not considered a logical choice because of the additional expenses incurred in providing services and in material handling.

The counting shield will be made of solid concrete with wall and roof thicknesses ranging from $1\frac{1}{2}$ to 3 ft. The shield will be contained within a concrete block structure approximately 16-ft wide by 16-ft long and 10-ft high attached to the south wall of the Plutonium Facility building. The floor area within the shield will be 36 ft² and will contain a drum station, neutron well counter, three neutron slab counters, and a gamma detector. Readout instrumentation and control equipment will be located in the area adjacent to the south wall of the shield.

All services required to make the system operational will be obtained from the Plutonium Facility building.

3.4 CALIBRATION STANDARDS

Plutonium standards are required for calibration of the passive assay system. These standards were prepared from a batch of mixed uranium-plutonium oxide powder and pellets. This material was crushed, sieved (-60 mesh), sintered at 1425°C for one hour in a helium-6% hydrogen atmosphere, and then pulverized in a ball mill for 20 hr.

Two random 15-gram thief samples were taken, each was divided in four parts and assayed for plutonium. One sample was also taken for analysis of impurities and Am²⁴¹ content.

Fig. 6 — Plutonium Facility Building

The plutonium content is reported in Table 2. The sample means are identical and the difference between their variances is not statistically significant at the 95% confidence level.

TABLE 2 — PLUTONIUM CONTENT OF MIXED-OXIDE STANDARD AS DETERMINED BY CHEMICAL ANALYSIS

<u>Thief Sample</u>	<u>Subsample</u>	<u>Plutonium Content, w/o</u>	<u>Plutonium Content, Avg w/o</u>	<u>Standard Deviation, %</u>
1	a	21.25	21.30	0.07
	b	21.40		
	c	21.31		
	d	21.22		
2	a	21.29	21.30	0.08
	b	21.39		
	c	21.31		
	d	21.21		

The standards were prepared by placing a weighed amount of the mixed oxide into "ointment-type" metal cans which were then sealed in two plastic (20-mil) pouches. Each pouch contained a label showing all the pertinent data. Table 3 lists the standards that have been prepared.

Plutonium standards are required for four categories of samples:

1. Fuel rods
2. Feed material (PuO_2)
3. Scrap (powder or pellets)
4. Waste.

TABLE 3 — STANDARDS PREPARED FROM MIXED-OXIDE MATERIAL

Standard Number	UO ₂ -PuO ₂ , g	Plutonium, g	Standard Number	UO ₂ -PuO ₂ , g	Plutonium, g
1	0.5010	0.1066	33	2.0000	0.4258
2	0.5000	0.1065	34	2.0010	0.4260
3	0.5030	0.1071	35	2.0000	0.4258
4	0.5000	0.1065	36	2.0000	0.4258
5	0.5017	0.1068	37	2.0000	0.4258
6	0.5000	0.1065	38	2.0000	0.4258
7	0.5000	0.1068	39	2.0000	0.4258
8	0.5015	0.1071	40	2.0000	0.4258
9	0.5030	0.1065	41	5.0017	1.0649
10	0.5000	0.1065	42	4.9782	1.0599
11	0.4977	0.1060	43	5.0220	1.0692
12	0.5041	0.1073	44	5.0074	1.0661
13	0.5016	0.1068	45	4.9901	1.0624
14	0.5000	0.1065	46	10.0199	2.1332
15	0.5019	0.1069	47	9.9920	2.1273
16	1.0020	0.2133	48	9.9871	2.1263
17	1.0020	0.2133	49	10.0632	2.1425
18	1.0000	0.2129	50	10.0086	2.1308
19	1.0000	0.2129	51	50.000	10.6450
20	1.0015	0.2132	52	50.001	10.6452
21	1.0070	0.2144	53	50.004	10.6459
22	1.0020	0.2133	54	50.022	10.6497
23	1.0000	0.2129	55	50.005	10.6461
24	1.0012	0.2132	56	100.000	21.290
25	1.0000	0.2129	57	100.017	21.294
26	1.0000	0.2129	58	100.018	21.294
27	1.0026	0.2135	59	100.005	21.291
28	1.0000	0.2129	60	200.0	42.6
29	1.0000	0.2129	61	200.0	42.6
30	1.0000	0.2129	62	400.0	85.2
31	2.0000	0.4258	63	400.0	85.2
32	2.0040	0.4267			

No synthetic standard was prepared for fuel rods. Previously fabricated fuel rods having a chemically determined plutonium content are to serve as standards for this category. A homogeneous batch of plutonium oxide, again with a chemically determined plutonium content, is to serve as a standard for feed material.

Since all scrap oxide material is placed in 2.75-in. OD by 2-in. high metal cans (ointment cans), the standards described above are well suited for this category. Normally one or two of these ointment cans are filled, double-pouched, and placed in a 3.75-in. OD by 5-in. high metal can. This size can is used for storage in the vault, and will be used for passive measurements in the neutron well counter.

Since waste materials are pouched and then placed in a 55-gallon drum, it is virtually impossible to synthesize waste calibration standards. For this reason, a modular standard concept has been adopted. In the modular concept, a number of mixed-oxide standards are placed in a wide-mouth plastic bottle or bottles that may or may not contain quantities of the waste matrix that is to be simulated. These bottles are then placed in a 55-gallon drum with other wide-mouth bottles that may or may not contain the waste matrix that is to be simulated. The total drum is the calibration standard.

The major advantage of such a system is that it affords excellent flexibility, using a minimum amount of plutonium. Four types of modular barrels will be prepared:

1. Bottles containing no matrix material
2. Bottles containing low-Z and hydrogenous materials
3. Bottles containing the mixed-oxide standards
4. Combinations of items 2 and 3.

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