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MONITORING AND DETECTION OF PLUTONIUM MOVEMENT IN STORAGE VAULTS

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We investigated a method for monitoring a typical large storage vault for unauthorized removal of plutonium. The method is based on the assumption that the neutron field in a vault produced by a particular geometric configuration of bulk plutonium remains constant in time and space as long as the configuration is undisturbed. To observe such a neutron field, we installed an array of 25 ^{3}He proportional counters in the ceiling of a plutonium storage vault at Argonne National Laboratory West. Data collected by each counter were processed to determine whether statistically significant changes had occurred in the neutron field. Continuous observation experiments measured the long-term stability of the system. Removal experiments were performed in which known quantities of plutonium were removed from the vault. Both types of experiments demonstrated that the neutron monitoring system can detect removal or addition of bulk plutonium (11% ^{240}Pu) whose mass is as small as [illegible]% of the total inventory.

INTRODUCTION

Research facilities that maintain large inventories of plutonium need a method for monitoring their storage vaults to detect unauthorized removal of plutonium in a timely fashion. This report describes the vault monitoring system, designed and constructed at Los Alamos National Laboratory and tested at Argonne National Laboratory West, that provides a reliable and inexpensive method for monitoring large storage vaults.

The vault monitoring system provides continuous surveillance to detect changes in a vault's neutron field. These changes reflect whether plutonium has been added to or removed from the vault, pinpoint the location, and indicate how much plutonium is involved.

Bulk plutonium in storage vaults generally is a mixture of several isotopes. The principal source of neutrons emitted by typical metallic plutonium is ^{240}Pu, which fissions spontaneously at a rate of approximately 1000 n/s per gram. Thus, in a plutonium storage vault the strength of the neutron field at any place primarily depends on the mass and spatial distribution of ^{240}Pu. Addition or removal of plutonium causes changes in the neutron field that can be measured using a geometrically appropriate array of neutron detectors. Table I shows the isotopic content for five types of plutonium in storage in a typical facility.

To verify the effect of plutonium removal on a neutron field, the Los Alamos Detection and Verification Group installed a vault monitoring system in the Zero Power Physics Reactor (ZPPR) storage vault of Argonne West at the Idaho National Engineering Laboratory (INEL). The system consisted of an array of 25 neutron detectors and a data acquisition system. The detectors monitored the neutron field in the vault before and after known quantities of plutonium were relocated. The data collected by each detector were processed in an affiliated data acquisition system, which helped us determine whether statistically significant changes had occurred in the neutron field.

Using the vault monitoring system, we performed two types of experiments at ZPPR over an 8-month period to determine the system characteristics for measuring neutron fields associated with the plutonium in storage. In one type of experiment, known quantities of plutonium were removed from the vault, causing observable changes in the neutron field. In the second type of experiment, the vault neutron field was monitored continuously over extended periods (3 days to 7 weeks) when no activity occurred in the vault. The first type of experiment established the minimum quantity of plutonium whose removal the system could detect; the second type demonstrated the effects of long-term parameter drifts in the system components.

The results of these experiments show that the monitoring system can detect the removal from or addition to an inventory of [illegible] kg of 1 kg of plutonium (11% ^{240}Pu) from any location in the vault with reasonable statistical certainty. Consistent results require long-term stability in the detectors and their high-voltage power supplies.

STATISTICAL TESTS FOR DATA ANALYSIS

An experimental setup of 25 neutron detectors and a data acquisition system (Fig. 1) at ZPPR was used to measure statistically significant changes in the neutron field associated with plutonium in storage. The field changed when the spatial distribution of the plutonium was disturbed. The objective of our research was to determine the smallest amount of removed plutonium that we could detect with reasonable confidence, using a manageable number of detectors.

The 25 neutron detectors measured the neutron count rates in the vault. These count rates were compared to a set of previously acquired data. If there was no difference between data sets, we assumed that the field was the same and, hence, the geometric and mass configuration of the plutonium in storage had not changed. We measured differences between data sets using chi-square statistical tests.

Briefly, the number of experimental occurrences of an event (such as neutrons detected) observed in a time period T_p can be compared to theoretical predictions of how often the event is expected to occur in time period T_p. Let $U = \{u_1, u_2, \ldots, u_n\}$ be a set of integers denoting how many times n different events occurred and $V = \{v_1, v_2, \ldots, v_n\}$ be a

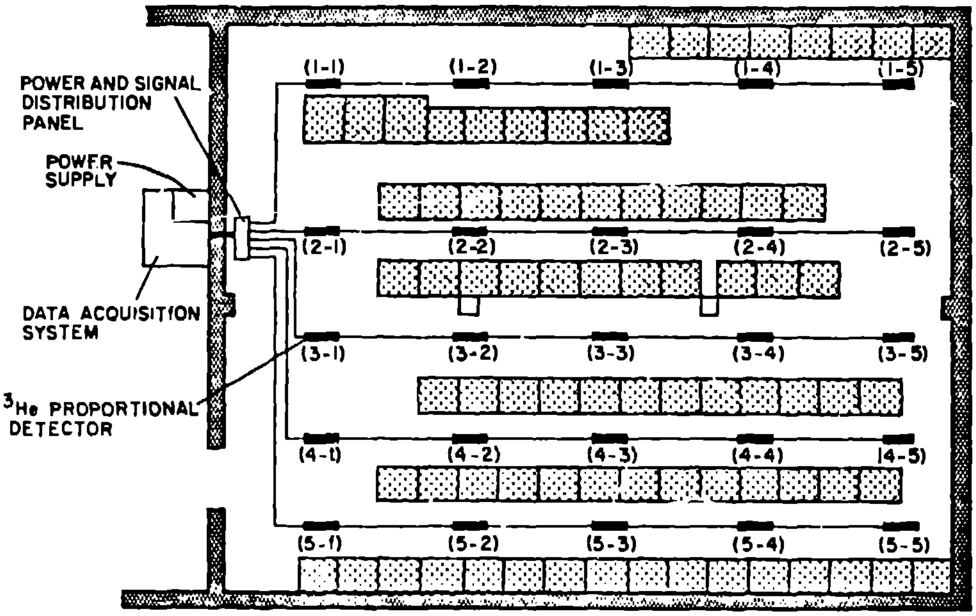

Fig. 1.
Installation of the monitoring system in the ZPPR vault. The neutron detectors, suspended from the ceiling, are identified by aisle and position from the front of the vault. The data analysis equipment is installed outside the vault.

corresponding set of integers denoting how many times each of n events is expected to occur. A figure of merit χ_a^2 can be calculated as

$$\chi_a^2 = \sum_{i=1}^{n} \frac{(o_i - v_i)^2}{v_i} \quad . \tag{1}$$

The probability that $\chi^2 < \chi_a^2$ is described as

$$P(\chi^2 < \chi_a^2) = \int_0^{\chi_a^2} p_{(n-1)}(\chi^2)\, d\chi^2 \tag{2}$$

where

$$p_{(n-1)}(\chi^2) = Y_c \chi^{(n-3)} e^{-\chi^2/2} \quad . \tag{3}$$

The expression $p_{n-1}(\chi^2)$ is also called the probability density function of the χ^2 distribution with (n-1) degrees of freedom. Y_c is a constant that causes the integral of Eq. (2) to be unity when limits of zero and infinity are used. By selecting probability thresholds, one can state that certain small differences are statistically significant, or insignificant, at a certain confidence level based on the value calculated for χ_a^2. Thus, if

$$0.001 < P(\chi^2 < \chi_a^2) < 0.999 \tag{4}$$

is chosen as the probability window required for a valid comparison where changes in frequencies are statistically insignificant, only two comparisons per thousand will produce a false alarm. A low false-alarm rate is an important operational constraint.

Because it was desirable to compare two observed sets of neutron count data that were collected over unequal counting time periods, Eq. (1) can be modified as follows, such that χ_a^2 is calculated in terms of count rates and counting times:

$$\chi_a^2 = \sum_{i=1}^{n} (o_{1i} - o_{2i})^2 \Big/ \left(\frac{o_{1i}}{t_{c_1}} + \frac{o_{2i}}{t_{c_2}}\right) \quad , \tag{5}$$

where $O_1 = (o_{11}, o_{12}, \ldots, o_{1n})$ and $O_2 = (o_{21}, o_{22}, \ldots, o_{2n})$ are two sets of neutron count rates observed over counting times t_{c_1} and t_{c_2}. All data sets of neutron count rates were subjected to χ^2 comparisons with other data sets in the removal experiments we performed.

The total count rate accumulated by all the detectors is also an indication of plutonium removal, although this parameter is not as sensitive as the χ^2 criteria because it does not measure the distortion in the slope of the neutron field. Consider a set of observed count rates from n detectors

$$O = (o_1, o_2, o_3, \ldots, o_n) \quad .$$

TABLE I

[illegible] VALUES FOR FIVE TYPES OF [illegible] STORED IN A TYPICAL FACILITY

[illegible]	^{239}Pu (%)	^{240}Pu (%)	^{241}Pu (%)	^{241}Am (%)
[illegible]	95.25	4.50	0.20	0.24
[illegible]	90.00	8.66	0.51	0.46
[illegible]	87.00	11.56	1.20	0.59
Pu-Al	74.20	22.23	2.86	1.80
[illegible]	64.70	26.40	3.39	2.19

Each kilogram of plutonium in storage can be thought of contributing to the total count rate. Thus, changes in the total count rate are indicative of changes in the mass of plutonium in storage. The total count rate C_R can be expressed as

$$C_R = \sum_{i=1}^{n} u_i \quad ,$$

whereas the total number of counts observed is $C_T = C_R t_c$, where t_c is the counting time. An uncertainty in C_T exists and its standard deviation is defined by $(C_T)^{1/2}$. … total count rate has 1-σ brackets as follows:

$$C_R = \sum_{i=1}^{n} u_i \pm \left(t_c \sum_{i=1}^{n} u_i \right)^{1/2} \Big/ t_c \quad .$$

Consider the case of 25 detectors with a total count rate of 50 000 Hz produced by 2500 kg of stored plutonium. Each kilogram of plutonium would account for approximately 20 Hz of the rate, on the average. If the array of 25 detectors were allowed to count for 200 s, the total count rate would be 50 000 ± 15.8 Hz. At the 1-σ level, the statistical uncertainty in the total count rate is the same as that count rate change caused by removal of 1 kg of plutonium. Clearly, extended counting times and highly stable electronics in the detectors are required to detect the removal of 1-kg amounts of plutonium.

DATA ACQUISITION EQUIPMENT AND SOFTWARE

A LeCroy 3500 data acquisition system processes data from the 25 neutron detectors (Fig. 2). The LeCroy interfaces with scientific experiments by means of an 8-slot CAMAC minicrate. The minicrate accommodates standard CAMAC modules and, thus, can both acquire data from a scientific experiment as well as control the experiment. For the ZPPR experiments, we used six Kinetic Systems 3610 hex scalers and one Kinetic Systems 3061 input gate/output register (IGOR) as the interface between the LeCroy and the experiment.

Data acquisition and processing took place as follows. Original $^3He(n,p)$ reaction pulses from each detector passed through a set of separate signal-conditioning electronics that converted them to standard TTL pulses. These logic pulses were fed to the 25 separate counters, housed in five Kinetic Systems 3610 hex scalers, whose counting times were controlled by a single input gate/output register. The LeCroy controlled the scalers via an eight-slot CAMAC minicrate that interfaced with the system bus, the Multibus. A 1.000-kHz crystal-controlled signal was directed to an additional counter to provide accurate timing for processing the neutron signals. Information pertaining to neutron counts and count rates could then be acquired and analyzed by application programs written for the LeCroy.

Two such applications programs were written to acquire and process count data from the 25 neutron detectors. The VAULT program processed data from removal experiments; the VLONG program processed data from stability experiments. Additional application programs were developed to process previously acquired data. Program VAULT acquired data for removal experiments in which certain amounts of plutonium were removed from known locations in the vault. Measurements of the neutron field were made before and after the removals. Program VLONG acquired data for stability experiments, which ran over long periods of time when no activity occurred in the vault. These experiments identified long-term drifts or stability of the equipment used to collect and process the data.

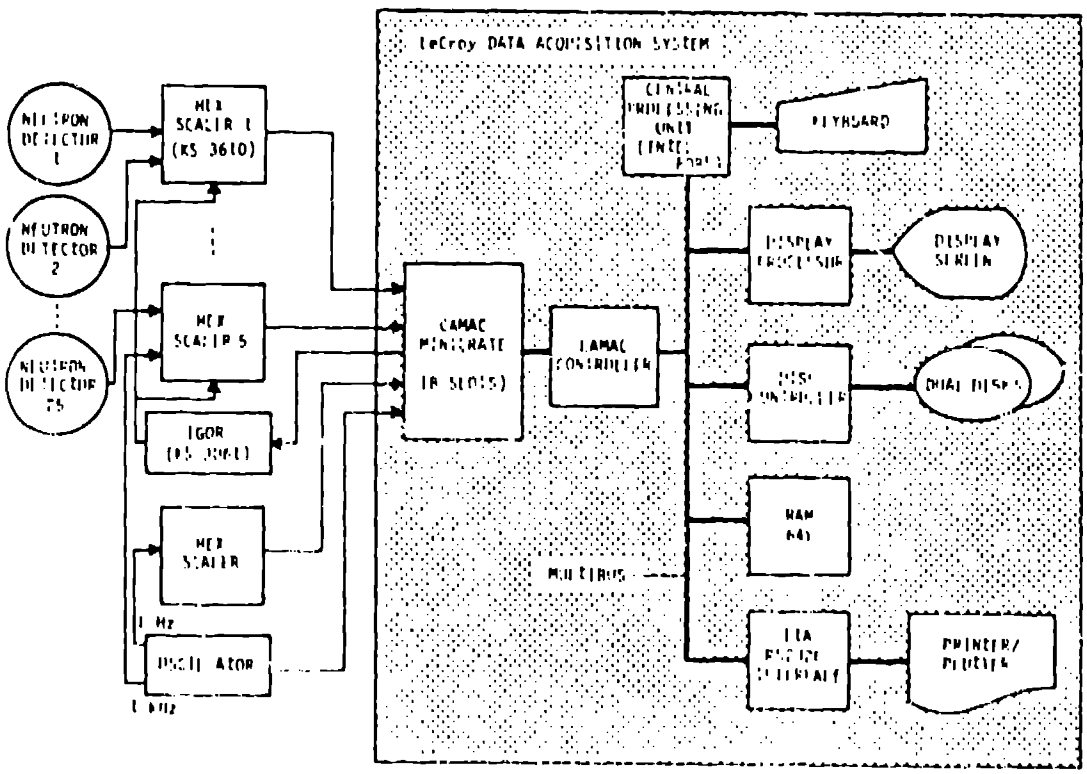

Fig. 2.
Signal processing in the vault monitoring system.

COLLECTION AND ANALYSIS OF NEUTRON COUNT RATE DATA

The ZPPR measurements included 82 stability experiments, a few of which ran for more than a week, and 16 removal experiments.

The results of one such stability experiment, which ran overnight on December 1-2, 1980, are shown in Fig. 3. Total count rate adjusted to remove the effects of one unstable detector, is plotted versus time. Because no activity occurred in the vault during the time of these measurements, the fluctuations in count rate were expected to be statistical in nature if all systematic causes have been eliminated. The figure shows an average count rate of 49 030 counts/s with a fluctuation from average of about ±20 counts/s. This is a count rate stability of ±0.04%, a close approximation to the expected fluctuation pattern for purely statistical fluctuations. The low values at the beginning and end of the figure are caused by neutrons escaping from the vault when the door was open. This stability study began shortly before the vault was closed for the night and ended the next day after it was reopened.

Sixteen direct measurements of plutonium removal were performed during the 8 months the monitoring system was installed in the ZPPR vault. We discovered that small distortions in the neutron field could be detected. Such distortions were caused by persons moving in the vault; the vault door being open; or removed material being too close, less than 3 m, to the vault entrance. Thus, all measurements were made with no one in the vault, the door closed, and all removed material stored about 10 m away and shielded by concrete during measurement of the neutron field.

Results of the December 2 removal experiment appear in Table II. In this experiment, we removed 6 kg of 26% ^{240}Pu from two middle level storage cells and measured the background. Then we inserted these 6 kg of plutonium successively in the top, middle, and bottom cells and made measurements (Table II). The count rate for removing 6 kg of plutonium changes by almost 600 Hz when the plutonium is removed from the top cells, whereas only a 215-Hz difference is noted for material removed from the bottom cells. Count rates per kilogram of 26% ^{240}Pu vary from 98 Hz for the top cell to 35 Hz for the bottom cell.

Measurements of typical canisters of 26% ^{240}Pu and 11% ^{240}Pu indicate that the 26% material emits 2.5 as many neutrons as the 11% material. Thus, the change in count rate ΔC_R per kilogram of 11% ^{240}Pu can be computed from the 26% ^{240}Pu data. A field distortion parameter

$$K = \chi^2/\Delta C_R$$

that indicates a change in the shape of the neutron field is also tabulated in Table II. K appears to be approximately 6 at the top storage cell and less than 3 at other levels.

CONCLUSIONS AND SUMMARY

The neutron monitoring experiments conducted at ZPPR successfully demonstrated the feasibility of using ^3He proportional counters to measure the neutron field in a plutonium storage vault. During the experiments, count rate stability at the ±0.04 to ±0.05% level was maintained for extended periods of time. A removal sensitivity of about 1 kg of plutonium from a nominal vault inventory of 2500 kg was achieved. This sensitivity applies to removal from any storage cell in the vault and is based on 4000-s counting periods. Statistically, for this situation, we had predicted a false-alarm rate with a 1-kg trip level to result in one false alarm per 2 years of continuous monitoring. The 8-month vault monitoring experience at ZPPR indicates that, with only modest hardware improvements, this level can be achieved.

Our findings indicate that the removal sensitivity for the top cells differs significantly from the removal sensitivity for all other cells. This is to be expected for spatially peaked distributions because the corresponding χ^2 values weight the individual detector excursions as the square of the excursion, which leads to a much larger χ^2 for a given change in total count rate if that change occurs primarily in a few detectors instead of in many.

The practical import of the foregoing observation is that a change in system count rate can be attributed to a removal from a given cell level in the cubicle, using the observed field distortion parameter. With this information, one may then accurately interpret a given change in count rate in terms of a missing plutonium mass.

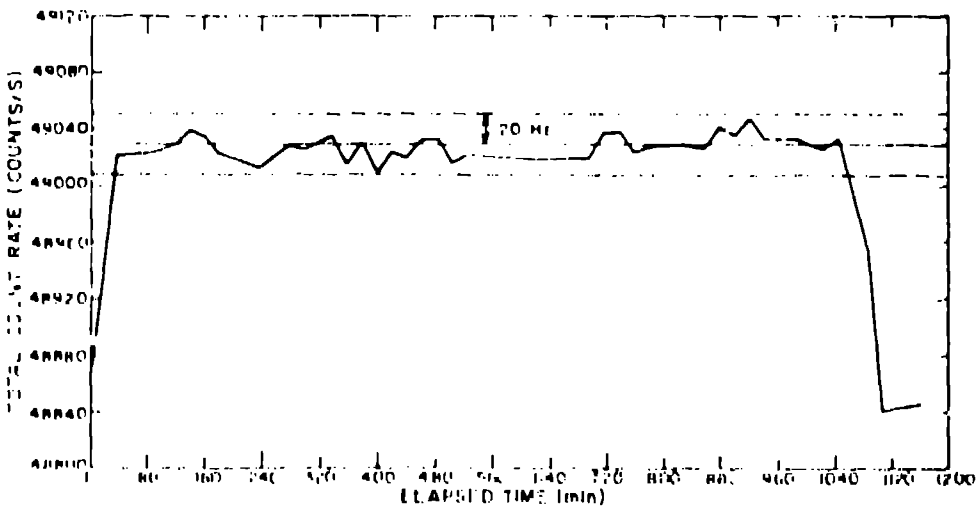

Fig. 3.
Count rate record for stability experiment conducted on December 1-2, with detector [illegible] removed, for 1100 min. Results fall within a 20-Hz error band. Counting times are [illegible] s.

For example, a change in count rate of 40 counts/s corresponds to a removal of 2 kg of plutonium if K is about 3, but corresponds to a removal of 1 kg of plutonium if $K \geq 6$. The ability to interpret count rate changes in terms of the field distortion parameter makes it possible to use neutron monitoring for vault inventory purposes. Based on our measurements, we predict that the vault monitoring system can provide surveillance of a 2500-kg plutonium inventory in real time at a sensitivity level of ±1 kg of plutonium.

TABLE II

REMOVAL EXPERIMENT OF DECEMBER 2[a]

Storage Cell Level	Action	Total Count Rate C_R (Hz)	χ^2	Field Distortion Parameter[b]	Change in Count Rate ΔC_R (Hz)	Total Count Rate Change/kg	
						26% ^{240}Pu	11% ^{240}Pu
middle[c]	removed	48628	---	---	---	---	---
top	inserted	49214	3585	6.12	586	97.7	39.1
top	inserted	49227	3597	6.01	599	99.8	40.0
middle	inserted	48871	490	2.02	243	40.5	16.2
middle	inserted	48922	688	2.34	294	49.0	19.6
bottom	inserted	48843	464	2.25	215	35.8	14.3
bottom	inserted	48853	553	2.45	225	37.5	15.0

[a]Counting for 500 s; each run has a 1-σ uncertainty of ±10 Hz.

[b]Field parameter $K = \chi^2/\Delta C_R$.

[c]Two canisters containing 6 kg of 26% ^{240}Pu were removed from these cells. The removed material was inserted into the other cells, which were empty at the time of the experiment.