RESULTS OF LOW LEVEL WASTE ANALYSIS OF PLUTONIUM WITH A PASSIVE NEUTRON COINCIDENCE COUNTER

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DEVELOPMENT OF THE PASSIVE NEUTRON WASTE ASSAY SYSTEM

The JCC-21 was jointly developed by the Applied Systems Division of Canberra Industries, Inc. and Los Alamos National Laboratory (LANL) for the Westinghouse Savannah River Site (SRS). Their specifications included a sensitivity of 10 g plutonium in three count times of 300 seconds each plus the ability to assay 200 g ± 25% plutonium.

While the resulting system performed much better than the design objective as demonstrated by ^{240}Pu-eff SRS who routinely measures < 100 nCi/g plutonium, and LANL who measured 14 mg with a 1 cps coincidence background. Initial testing indicated that changes could be made which would further improve the detection limit of the system for the low level waste application of PNC/PFPF. These changes included:

(1) Increasing the active length of the ^{3}He tubes in the top and bottom banks by 2.5 cm (1 in.).

(2) Changing the tube material from aluminum to stainless steel to reduce the α decay from the impurities in the aluminum.

(3) Removing the cadmium liners from the counting chambers to minimize spallation reactions from cosmic ray events.

The net result of these changes was to increase the efficiency of the system from 17.9% to 18.6% and to improve the sensitivity by 20%. This improved version is offered commercially as the Canberra Model JCC-21, and was purchased by LANL for PNC/PFPF.

Prior to installation at PNC/PFPF, LANL made two performance enhancements to the system:

(1) An "add-a-source" facility was incorporated into the system to improve accuracy.

(2) Statistical evaluation of the data was added to the software to further improve the sensitivity.

ADVANTAGES OF PASSIVE NEUTRON COINCIDENCE COUNTING WITH THE JCC-21

The resulting passive neutron waste assay system offers several significant benefits when compared to active neutron coincidence counters:

(1) More accurate results because it is less sensitive to matrix effects.

The matrix effect on fast fission neutrons in a passive counter is less than the total effect on the interrogation neutrons plus the fission neutrons in an active counter.

(2) Higher reliability and requires only minimal routine maintenance.

The preamplifier/discriminators (Model JAB-01) are set at the factory, and in 16 years no JAB-01 and only one ^{3}He tube has ever been replaced.

(3) Easier to use.

No adjustments are required once the system is installed. Due to its straightforward modular design and ease of operation, it took only one week for installation, certification, and operator training at PNC/PFPF.

(4) Less expensive to purchase and operate.

Because no interrogation neutron source or D-T generator is required, the JCC-21 is both less expensive to initially purchase and to operate. In addition, operating costs are further reduced because NDA specialists are not required for routine operations and analysis.

THE BASIC JCC-21 SYSTEM

The following describes the major components which make up the basic JCC-21 passive neutron waste assay system.

The Counting Chamber

The counting chamber, shown in Figure 1, has a 71 x 71 x 107 cm (28 x 28 x 42 in.) interior cavity, which will comfortably hold a standard 200 L (55-gal) drum. To facilitate drum loading and unloading, aluminum rollers are built into the loading platform and in the base of the counting chamber. Once the drum is loaded, the motor-driven door is closed and counting can begin. The door control circuitry includes safety interlocks to prevent the door from closing on an object that may be in its path. The control and signal wiring between the counting chamber and the system's electronics can be seen in Figure 2.

All sides of the counting chamber, including the door, have in excess of 10 cm (4 in.) of high density polyethylene shielding to reduce background and improve the detection limit of the system.

The Control Panel

A JMC-69 control and indicator panel is used by the operator to open and close the door of the counting chamber. As a safety precaution, two pushbuttons – Door Enable plus Open Door or Close Door – must be simultaneously depressed to activate the automatic door mechanism. Indicators are provided for both the normal Opened and Closed limit switches as well as the Emergency Stop safety interlock switch.

The Detectors

A total of 60 ^{3}He filled proportional detectors, 10 on each of the six sides of the chamber, make up the detector array. The tubes in the three walls and the door have a 91 cm (36 in.) active length, and those in the top and bottom a 53 cm (21 in.) active length. Each bank of 10 tubes is embedded in a 10 cm thick high density polyethylene moderator.

Figure 1.
The JCC-21 Passive Neutron Waste Assay System

Fast Amptek JAB-01 Amplifier/Discriminator modules are used to process the signals from the detectors. A total of 10 JAB-01s are used: two for each of the four side detector banks and one channel each for the top and bottom detector banks.

The Neutron Coincidence Analyzer

The outputs from the Amplifier/Discriminators are combined and sent to a JSR-12 computer-controlled Neutron Coincidence Analyzer for processing.

Coincidence logic in the JSR-12 is used to discriminate between the neutron multiplets produced by fission events and the randomly generated (α,n) and background neutrons that are detected. At the completion of a count the JSR-12 reports the Total Counts, the Reals+Accidentals Count, the Accidentals Count, and the Reals Count to the PC that supervises the system.

In addition to receiving data from the JSR-12, the PC also starts and stops the data acquisition and sets the JSR-12's counting and analysis parameters. The program-controlled parameters include the counting predelay, the width of the coincidence windows, the gate count time, and the high voltage.

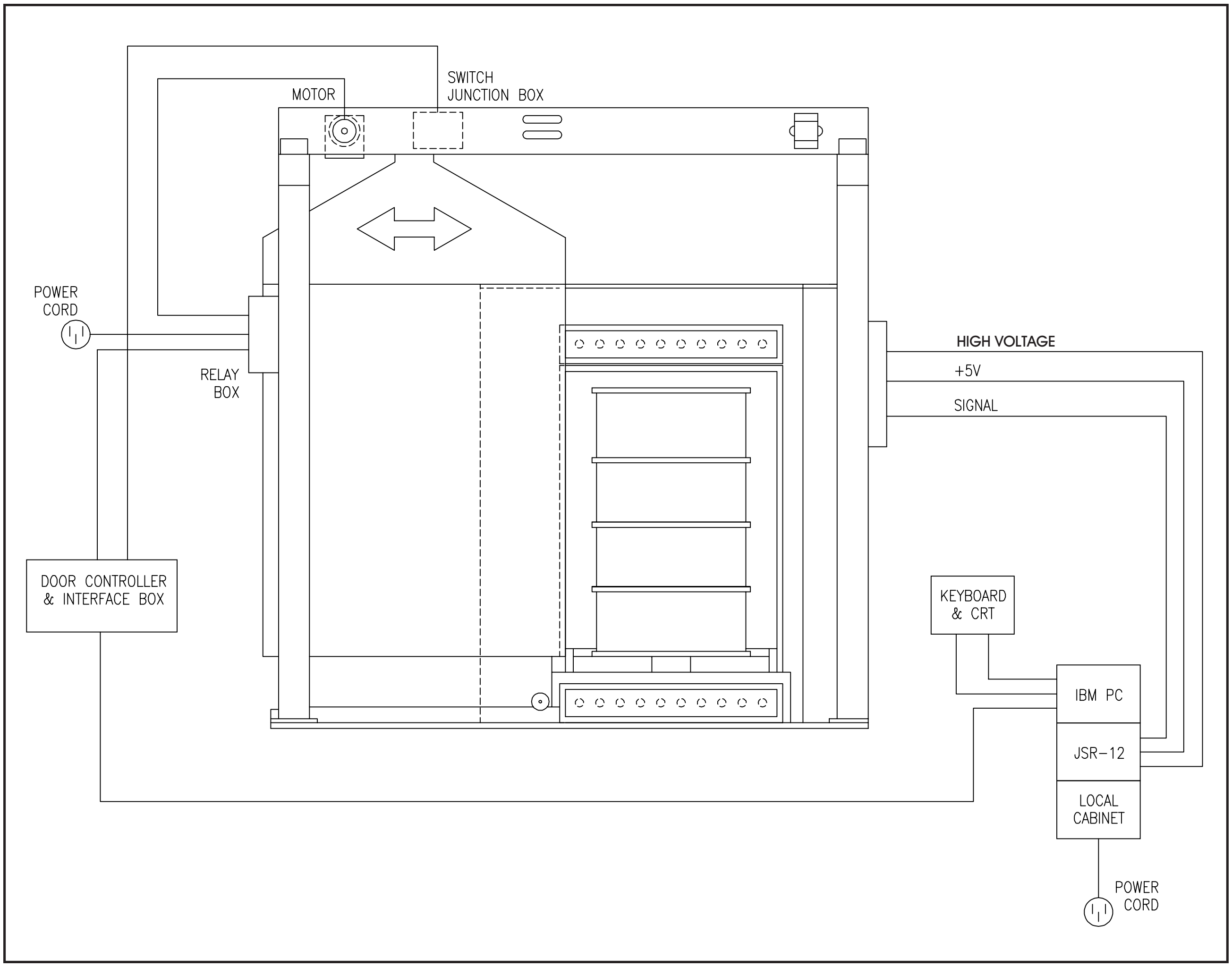

Figure 2.
The JCC-21 Wiring Diagram

System Performance

Measurements made at Los Alamos National Laboratory, who purchased the JCC-21 for PNC/PFPF and added the performance enhancements described in Appendix A, have shown that the JCC-21 provides good sensitivity, but without the high costs and handling concerns associated with neutron generators or large ^{252}Cf sources in active neutron counters.

Vertical Response Profiles

To determine sample positioning effects, a ^{252}Cf point source was counted at a variety of vertical and radial positions in an empty drum. The vertical profile measurements were made at a radius of 20 cm from the center of the drum, which is approximately the volume-averaged mean radius; that is, the drum volume within the 20 cm radius equals the volume outside that radius.

Figure 3 shows the normalized vertical totals and reals rates for the source (Menlove and Eccleston, 1992). The dips at the top and bottom of the drum (extreme left and right in the figure) are caused by gaps in the detector coverage at the ends of the detector banks.

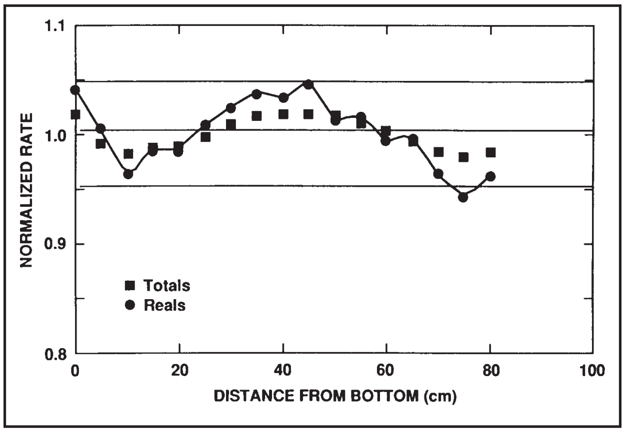

Figure 3.
Totals and reals vertical response profiles measured using ^{252}Cf at a radius of 20 cm in an empty 200 L drum

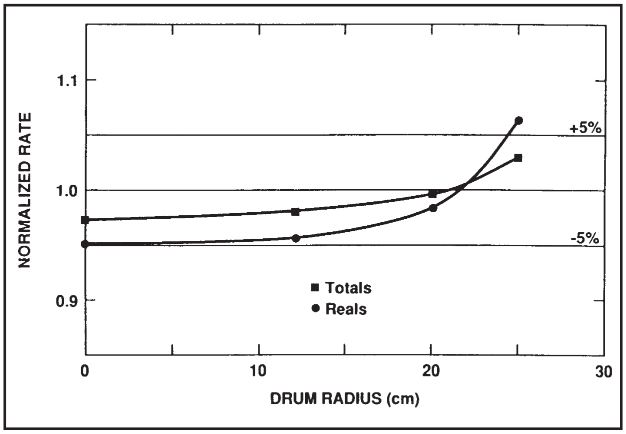

Figure 4.
Totals and reals radial response profiles averaged over the heights of 15, 35, 55 cm in an empty 200 L drum

Radial Profiles

For the radial profiles the source was located at four different radial positions and three different vertical positions. The radial profile for the average of the three vertical positions can be seen in Figure 4 (Menlove and Eccleston, 1992). Figure 5 shows an enlargement of the normalized radial efficiency profiles for the three vertical positions (Menlove and Eccleston, 1992).

Detectability Limit

The results of the detectability limit measurements, which were performed with a 1000 second counting time, are summarized in Figure 6 (Menlove and Eccleston, 1992). With the 0.006 counts/second ambient background at the test site, a minimum detectable activity of less than 0.002 g ^{240}Pu-eff was achieved. For any location with a similar background level the JCC-21 can routinely yield this level of performance, and it can do it at a fraction of the cost of an active neutron counter.

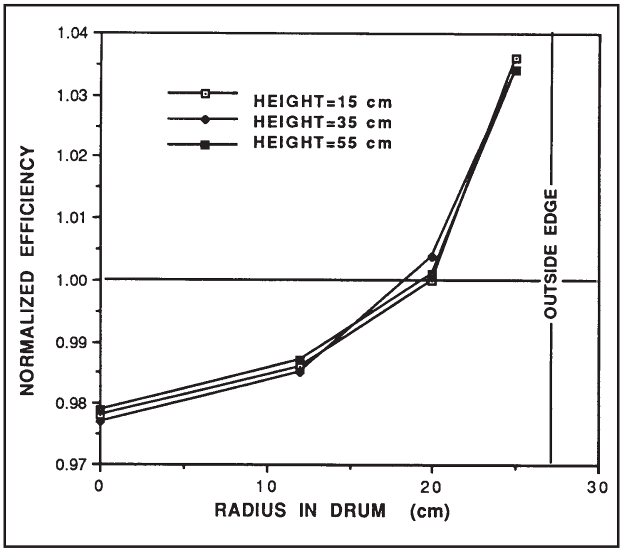

Figure 5.
Counting efficiency vs. radius for various heights in an empty 200 L drum

Summary

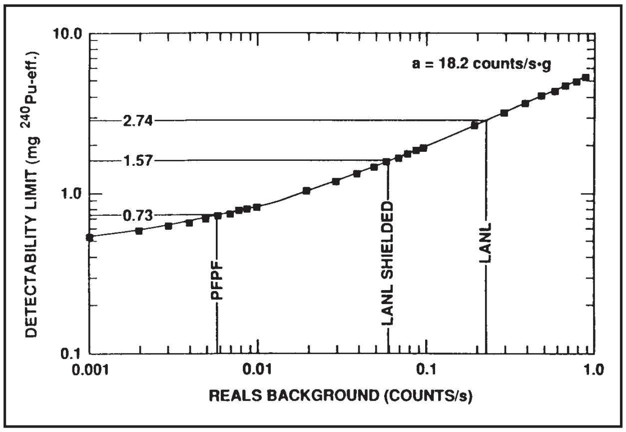

Figure 6.
Calculated detectability limit vs. neutron coincidence background for a 1,000 second measuring time

Plutonium contained in low-level waste is best characterized using passive-neutron assay rather than active-neutron assay systems. A high-efficiency, well-shielded, passive-neutron counter such as the JCC-21 can give a detectability limit of 0.73 mg of ^{240}Pu-eff in a reasonable counting time when operated in an environment with a low and relatively constant rate background. Results to date indicate that additional efforts in the areas of detection efficiency and background reduction, including the incorporation of multiplicity counting, can improve the detectability limit by better than a factor of two.

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Appendix A
The Use of Add-a-Source and Statistical Filtering Techniques to improve the Accuracy and Sensitivity of Passive Neutron Waste Measurements

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Investigations at Los Alamos National Laboratory into methods and techniques for improving the performance of the basic passive counter resulted in two major enhancements. These were incorporated into the design of the JCC-21 which was purchased for and supplied to PNC/PFPF.

Add-a-Source Option

The primary source of uncertainty in passive neutron measurements comes from the neutron moderation and absorption in matrices with a high hydrogen content (> 0.04 g hydrogen/cm^3 – equivalent to 73 kg of H_2O in a 200 L drum). For significant quantities of plutonium, this uncertainty can be reduced by using the ratio of triples to doubles to measure the matrix correction. This ratio is proportional to the efficiency, including the matrix perturbation to the efficiency. In addition, this ratio of triples to doubles technique also helps to correct for any localization or non-uniform distribution of the plutonium in the matrix. However, the statistical uncertainty in the triples rate is a problem for small plutonium masses (Pedersen, *et al.*, 1992).

A matrix correction technique that has been found to be very effective at all plutonium mass loadings and also has good counting statistics is the "add-a-source" (AS) technique (Menlove and Eccleston, 1992).

The basis of the AS method is to measure the matrix perturbation to the counting rate from a small ^{252}Cf (~3 x 10^4 neutron/s) on the outside of the sample. This information is then used to correct for the matrix perturbation on the inside of the sample. At the present time the AS source is positioned at the bottom center of the 200 L drum, as shown in Figure A-1 (Menlove and Eccleston, 1992).

The sample matrix has two primary effects on the neutrons:

(1) Energy reduction by scattering reactions.

(2) Absorption of the low energy neutrons.

The JCC-21 is designed with a moderator (CH_2) thickness optimized to be insensitive to the neutron energy reduction; however, as the hydrogen density in the drum increases, the absorption process significantly reduces the measured neutron signal.

To correct for the matrix perturbation on the neutron signal, the AS method measures the drum both with and without the ^{252}Cf source at the bottom of the drum. The quantities which are measured are:

T_0, R_0 = Totals and reals rates from ^{252}Cf for an empty drum;

T, R = Totals and reals rates from a sample drum without ^{252}Cf;

$T(Cf)$, $R(Cf)$ = Totals and reals rates from a sample drum with the ^{252}Cf.

The net ^{252}Cf reals rate for the ^{252}Cf and a loaded sample drum is:

$$R(net) = R(Cf) - R \qquad (1)$$

The ratio of the empty drum (after source decay correction) to the net loaded drum is used to make the matrix correction in the following manner:

$$x = \left(\frac{R_0\, e^{-\lambda t}}{R(net)} - 1 \right) \qquad (2)$$

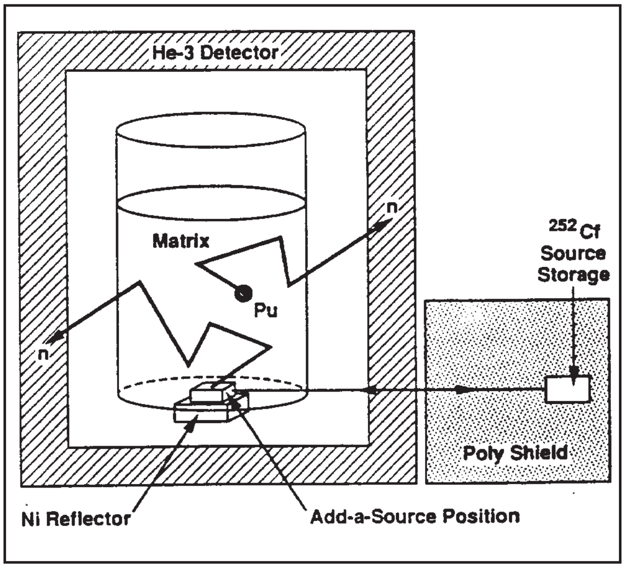

Figure A-1.
Conceptual Diagram of the AS Configuration

where:

λ = ^{252}Cf decay constant (0.2623 y^{-1})

t = time in years from the R_0 measurement.

The correction factor (CF) is defined as

$$CF = 1 + f(x) \qquad (3)$$

where $f(x)$ is a polynomial function of x based upon empirical measurements. The measured R for a drum is corrected to give R(corrected) = R(measured) CF.

The functional relationship between the AS perturbation (x) and the volume-averaged sample perturbation $f(x)$ has been determined empirically at LANL by measuring a wide range of matrix loadings with the “add-a-source” located at the bottom of the drum. A separate (not the AS) neutron source was then counted at nine positions within the drum to give a *volume-averaged* matrix effect. The average of the nine positions was then ratioed to the empty drum to give the volume-averaged perturbation

$$y\ (\text{vol av perturbation}) = \left[\frac{R'_0\ (\text{empty vol})}{R'\ (\text{matrix vol})}\right] - 1 \qquad (4)$$

where:

R'_0 = reals rate averaged over the volume of the empty drum

R' = reals rate averaged over the volume of the drum with matrix material.

A plot of the volume-averaged perturbations (y) vs. the AS perturbation (x) is shown in Figure A-2 (Menlove and Eccleston, 1992). The data point with the highest AS perturbation (~300%) corresponds to a drum loaded with CH_2 beads (ρ = 0.60 g/cm^3). This drum has the same neutron shielding as a drum containing H_2O with ρ = 0.72 g/cm^3, which makes it more opaque to neutrons than a drum filled with concrete.

A polynomial was fitted through the y vs. x data to give the predicted volume-averaged matrix perturbation $f(x)$ based on the AS measured perturbations,

$$y = f(x) = a_0 + a_1x + a_2x^2 + a_3x^3. \qquad (5)$$

The AS measurement is accomplished by automatically transferring a small ^{252}Cf source from the shielded location shown in Figure 3 to the bottom of the drum by means of a Teleflex cable and Compumotor drive system (Rinard, 1991). A 5 cm thick nickel reflector at the AS stopping point under the drum reflects additional source neutrons into the drum. Each unknown drum is placed into the counter, and the AS measurement is performed for two to three minutes.

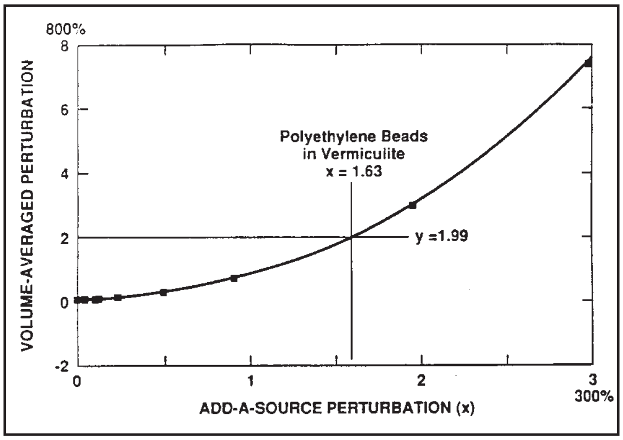

Figure A-2.
The AS perturbation in R vs. the plutonium volume-averaged perturbation in R for a wide range of matrix hydrogen densities

The source is then automatically removed from the counting chamber and an ~10 minute passive neutron measurement completes the assay. All of the time intervals used are contained in the AS program computer parameter files, and may be easily modified as needed.

To evaluate the error in using the AS correction for the matrix materials listed in Table A-1, the fitting function *f*(*x*) was used to obtain *CF*. All of the measured *R* values were normalized to the empty drum case and corrected by *CF*.

The results of these measurements are listed in Table A-1 (Menlove and Eccleston, 1992). The CF-corrected reals deviate from the empty drum with a standard deviation of only ± 1.0%. These same drums (except for the drum containing boron glass) were used to determine *f*(*x*), so the results show the scatter of our *CF* calibration. However, after the *CF* calibration was established, a drum filled with vermiculite and CH_2 beads (ρ_{CH2} = 0.308 g/cm^3) was measured as an unknown and the corrected response ($R * CF$) was within 3% of the empty-drum case. A typical drum of organic waste is expected to have a hydrogen loading that is equivalent to ρ_{CH2} ~ 0.1g /cm^3 (ρ_H ~ 0.014 g/cm^3), and thus the correction factor will be much smaller than the present case with ρ_{CH2} = 0.3 g/cm^3.

Statistical Filtering for Cosmic Ray Rejection

The limit of detection (*d*) of a passive neutron system varies approximately as the square root of the background rate (*B*). Thus, a factor of ten decrease in *B* results in about a factor of three improvement (decrease) in *d*. Once the more traditional background reduction steps have been taken – increased physical shielding, minimization of high mass number materials in the counter, etc. – cosmic ray spallation events become the limiting factor. The background was reduced by a factor of ~1.8 through the use of a statistical filter which eliminates the cosmic ray spallation events which produce high-multiplicity events.

Cosmic ray events can be counted as prompt charged-particle reactions in the detector tubes or as spallations-source neutrons that extend in time over the slowing-down time of the detector body. The predelay (4.5 μs) eliminates the first category because they are short lived and vetoes them from the coincidence gate (Reilly, *et al.*, 1991). The spallation neutrons fall within the coincidence gate, but often with high multiplicity. The data collection software isolates the high multiplicity events and eliminates them from the data averages. Statistical techniques are currently being used to do this, but in the future we will be investigating the use of multiplicity electronics to eliminate these events.

Table A-1.
As Matrix Correction for 200-L Drums

Sample	Vol. Av R_0/R	Relative R	CF	Relative R(corr)
Empty Drum	1.000	1.000	0.996	0.996
Vermiculite (dry)	0.985	1.015	1.005	1.020
Paper (ρ = 0.11)	1.019	0.981	1.023	1.003
Boron Glass	1.031	.970	1.048	1.016
CH2 (ρ = 0.060)	1.039	0.963	1.030	0.992
CH2 (ρ = 0.154)	1.195	0.837	1.217	1.019
CH2 (ρ = 0.159)	1.257	0.796	1.241	0.988
CH2 (ρ = 0.225)	1.685	0.594	1.671	0.993
CH2 (ρ = 0.464)	4.001	0.250	4.020	1.005
CH2 (ρ = 0.604)	8.396	0.119	8.380	0.997
Ch2+Fe+Al Mix	1.071	0.934	1.076	1.005
				1σ = 1.0%

The statistical filter for background reduction consists of a 2.5-3.0 σ rejection threshold from the average of multiple, short data intervals. The normal counting time for a drum is 600 seconds, which is divided into 20 intervals of 30 seconds each. If any interval is more than ± 2.5–3.0 σ out of the average, that interval is rejected from the average. This type of filter does not interfere with the data collection for drums with high- or low-plutonium content, and only ~1–2% of the useful data points are eliminated by this statistical filtering method.

Continuing Investigations

The following areas for improving the performance of the JCC-21 are currently being explored.

Increasing Detection Efficiency

The efficiency of the counter can be increased by the addition of more ^{3}He tubes. Adding more tubes, thereby reducing the spacing between the tubes, increases the response of the counter.

Reducing Background

The detectability limit of the JCC-21 at three standard deviations above background can be calculated using the equation

$$d = \frac{3}{a}\sqrt{\frac{B+ad}{t}} \tag{6}$$

where

d = detectability limit in grams of ^{240}Pu

a = the response of the counter in counts/(s·g ^{240}Pu)

B = counting room background rate

t = counting time

From this it can be seen that, for a given detection efficiency, the detectability limit (d) varies as approximately as the square root of the background rate (B), making background reduction a high priority item for improving the detectability limits of the system. Among the areas being examined are:

(1) Increasing the thickness of the high-density polyethylene shielding. The current 10 cm thickness reduces the room drift neutrons by a factor 10; the added background reduction benefits from increasing its thickness need to be explored.

(2) The use of a concrete bunker to isolate the system from nearby plutonium-containing drums and cosmic ray spallations.

(3) Modifications to the detector signal processing chain to eliminate the high energy pulses from alpha discharges from the trace impurities in the walls of the ^{3}He detector tubes.

(4) The use of multiplicity counting (described separately below) to distinguish plutonium events from cosmic ray spallations and other isotopes.

Multiplicity Counting

Neutron multiplicity counting can be used to separate plutonium events from those caused by interfering radionuclides such as ^{244}Cm because ^{244}Cm has a higher number of neutrons per fission and a lower α value than plutonium. The isotope ^{244}Cm ($T_{½}$ = 18.1y) is a prolific source of neutrons with a specific yield on 1.08 x 10^7 n/s·g from spontaneous fission and 7.73 x 10^4 n/s·g from (α,n) reactions in oxide (Reilly, *et al.*, 1991). This can be compared with the ^{240}Pu yields of 1.02 x 10^3 n/s·g from spontaneous fission and 1.41 x 10^2 n/s·g from (α,n) reactions in oxide.

Thus the α values for oxide reactions differ with

$$\alpha(^{240}\text{Pu}) = \frac{1.41 \text{ x } 10^2}{1.02 \text{ x } 10^3} = 0.14 \tag{7}$$

$$\alpha(\text{Pu}) \approx 0.06 \tag{8}$$

$$\alpha(^{244}\text{Cm}) = \frac{7.73 \text{ x } 10^4}{1.08 \text{ x } 10^7} = 0.0072 \tag{9}$$

The ratio of α's for plutonium and ^{244}Cm is the discrimination ratio (Menlove, *et al.*, 1971) *DR* and its value is > 20. However, the above α's correspond to oxide (α,n) reaction yields, and impurities can increase the measured value of α resulting in an incorrect assay.

For cases in which impurities cause differences compared to the oxide (α,n) calculation, we can still use the difference in multiplicity between ^{244}Cm and plutonium. The spontaneous fission multiplicity of ^{244}Cm is 2.72, versus 2.16 for ^{240}Pu.

High efficiency passive neutron counters can also make use of the triples to doubles multiplicity ratios to separate ^{244}Cm from ^{240}Pu with independence from α. In this case, the *DR* is small (~1.3) and the statistical precision for counting is marginal. Thus there is a significant error buildup (Menlove, *et al.*, 1971) in the data analysis, and the usefulness of the results has to be evaluated for the particular application.

Preliminary testing has also shown that, because the multiplicity of cosmic events is much greater than it is for the spontaneous fission of plutonium, multiplicity counting can also be effective for separating cosmic ray spallation events from plutonium events. Initial results indicate a potential factor of two improvement in sensitivity.

Because multiplicity counting offers such a great potential for improving the detectability limit of the system, Canberra is commercializing the multiplicity counting hardware which was developed at Los Alamos National Laboratory.

Improving Matrix Corrections

The present AS mechanical system measures primarily the bottom half of the drum, which requires that we assume that the matrix in the bottom half of the drum represents the entire drum. Because this assumption may be too limiting, a mechanism to move the AS source up and down the side of the drum is being explored. This will allow more uniform characterization of, and correction for, effects of the matrix. The plan is to adapt the software and Teleflex cabling of the AS drive system to the more complex scanning of the drum as is done with ^{252}Cf shuffler type systems (Rinard, 1991).

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ABSTRACT

Measurements have been made at the Los Alamos National Laboratory in the United States and the Power Reactor Nuclear Fuel Development Corporation/Plutonium Fuel Production Facility (PNC/PFPF) in Japan with the Passive Neutron Waste Assay System (JCC-21) employing the “add-a-source” technique to quantify the plutonium content in 208 L drums. The JCC-21 was jointly developed by Canberra Nuclear and Los Alamos National Laboratory. High counting efficiency is obtained by using multiple ^{3}He detectors in a 4π geometry, and a carefully-designed high-density polyethylene moderator provides relative insensitivity to the effects of scattering reactions in the matrix. Additional high-density polyethylene also provides shielding to reduce the low-energy ambient neutron background and improves the detection limit.

The “add-a-source” technique developed by Los Alamos National laboratory provides a significant improvement in accounting for matrix effects by actually measuring the matrix absorption of neutrons from a small ^{252}Cf source and correcting the detected flux for the matrix absorption. Matrix effects and corrections using the “add-a-source” technique were evaluated during installation. A detectability limit of 0.73 mg ^{240}Pu-eff was measured for coincidence counting at PNC/PFF, making the JCC-21 a sensitive and accurate NDA system for analysis of plutonium-bearing waste. A detectability limit of 0.4 mg ^{240}Pu-eff was measured for total counts at Los Alamos National Laboratory, demonstrating a lower detection limit for waste where the (α,n) component is known and the background is low.

Enhancements to the counter design are being investigated to improve the sensitivity by reducing the background and increasing the counting efficiency. New counting techniques are also being investigated to improve the accuracy of the assay. The results of these enhancements and new techniques are presented.

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