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## ABSTRACT

An advanced passive neutron counter has been designed and fabricated to measure the plutonium content in large remote handled (RH) waste crates. The waste crate assay system (WCAS) was developed under an agreement between Los Alamos National Laboratory, Japan Nuclear Fuel Limited (JNFL), and BNFL Instruments Inc. (BII) to measure the plutonium content in the waste generated in the Rokkasho reprocessing facility. The primary goal of the design was to produce an assay system for large waste containers. The system also includes 200-L drum pallet assay capability. The measurements are based on neutron-time correlation counting of the passive neutron emissions from the $^{240}$Pu, and the plutonium isotopic ratios are used to calculate the total plutonium. The system is designed for both RH waste and low-activity plutonium waste. The system permits the measurement of the singles, doubles, and triples rates and the multiplicity mode analysis is used together with the "add-a-source" method to correct for the matrix materials in the crates. In the multiplicity analysis, the efficiency for counting the neutrons emitted from the crate is directly calculated from the three measured rates. For improved detectability limits, advanced methods have been incorporated in the WCAS-A to reduce the cosmic-ray neutron backgrounds. These methods include statistical filters and truncation of high-multiplicity events. The paper describes the WCAS-A design, performance, and calibration.

## 1. INTRODUCTION

Los Alamos National Laboratory (LANL) was contracted by Japan Nuclear Fuels Limited (JNFL) in 1998 to develop two nondestructive assay systems for the spent fuel reprocessing plant under construction at Rokkasho, Japan. LANL teamed with BNFL Instruments Inc. (BII) to fabricate and deploy these systems, which can accommodate a maximum crate whose volume is about forty 200-L drum equivalents. The WCAS systems (named A and B) are targeted at making both domestic and international safeguards related measurements on dry, reprocessing waste. The WCAS-A system was designed to work in high radiation fields up to 1.1 Gy/h (110 R/h) for RH debris, with an average $^{240}Pu_{eff}$ content of 40%; the WCAS-B system was designed to measure only low-activity non-RH waste. This paper will focus on the WCAS-A system for the RH wastes.

The design of the WCAS-A had the unique requirements of being the largest 4-π neutron detector that has been built worldwide as well as being hardened for RH waste. Active neutron assay systems were not used in this application because the neutron interrogation flux cannot effectively cover the huge volume of ~ 8000-liters, and the active signal has interference from fissions in $^{235}U$. Normally, the wastes from spent fuel will contain significant amounts of $^{235}U$.

The WCAS-A assay system uses passive neutron measurement principles to determine the mass of waste-entrained radioactive materials. A primary consideration in the use of the passive neutron technique is to minimize or eliminate some of the matrix and radioactive material induced bias effects associated with the active neutron technique. The WCAS-A system has been designed with gamma-ray shielding to operate in high gamma-ray fields. The detection system consists of four atmosphere $^{3}He$ proportional counters located in the top, bottom, and four side walls. The $^{3}He$ detectors are embedded in a polyethylene matrix for optimized spacing and thickness. The system is operated in the passive neutron coincidence or multiplicity mode to measure the $^{240}Pu_{eff}$ spontaneous fission rate. The plutonium isotopic data are acquired from the stream average isotopics in the process area.

The passive neutron assay of wastes with a high fission product (FP) inventory will result in a neutron measurement that is dominated by $^{244}Cm$. The ratios of Cm/Pu and Cm/$^{235}U$ are used to calculate the amount of plutonium and $^{235}U$ in the wastes. These isotope ratios are determined by knowledge of the stream average ratios at the waste generation sites.

Corrections for waste matrix effects on the observed counting rates are addressed through the Add-a-Source (AAS) technique.[1] In parallel with the AAS result, the WCAS-A gives the plutonium mass derived from the multiplicity matrix correction.[2,3] This result is independent of the AAS correction factor. If the AAS matrix correction is small, the AAS calibration result is preferred because of the good precision using the doubles rates.

The detection of milligram quantities of $^{240}Pu$ requires the use of background reduction techniques. To obtain a low detectability limit, a low neutron background rate is desirable. For the WCAS-A, system, this is obtained by overhead shielding (>1.5 m of concrete) and a location near sea level. Furthermore, as the detectability limit is directly related to the background neutron coincidence rate, techniques to reduce the cosmic-ray signal contribution are required. The cosmic-ray spallation neutron background can be reduced by taking advantage of the differences in the multiplicity distribution

between fission and cosmic-ray events. This can be accomplished through truncated multiplicity (TM) techniques.[4]

Because there were no plutonium standards at the Albuquerque fabrication site, the initial calibration of the WCAS-A was accomplished using a $^{252}Cf$ sample of known mass and relating its counting rate to a plutonium sample. Calibration is performed in an empty container where the calibration is in terms of doubles counts per second versus $^{240}Pu_{eff}$ mass. The corrected doubles rate in an actual waste container is the measured coincidence rate multiplied by the AAS-derived correction factor.

## 2. PLUTONIUM-TO-CURIUM RATIO METHOD

For the high gamma-dose samples, we measure the associated $^{244}Cm$ neutrons to calculate the plutonium based on the stream-averaged ratio of curium to plutonium supplied by the operator. The waste generation is from sites that have known Cm/Pu ratios. Table I gives the neutron yields from $^{244}Cm$ and Pu for both spontaneous fission (SF) and ($\alpha$,n) reactions in oxides. Prior to the first separation stage, the ratio of curium to plutonium is about 0.003. However, the SF emission rate from $^{244}Cm$ is about $3.2 \times 10^4$ times that of plutonium (with 24% $^{240}Pu$). Therefore the emission from $^{244}Cm$ dominates that from plutonium by a factor of ~100. This factor increases to about 6000 in the waste after plutonium separation.

**Table I**. Neutron Emission Rates

| Isotope | Half Life (years) | SF Rate (fissions/s•g) | SF Emission Rate (n/s•g) | Oxide ($\alpha$,n) Rate (n/s•g) |
|---|---|---|---|---|
| $^{238}U$ | $4.47 \times 10^9$ | 0.00677 | 0.0136 | $8.26 \times 10^{-5}$ |
| $^{240}Pu$ | $6.55 \times 10^3$ | 472.2 | 1020 | $1.41 \times 10^2$ |
| $^{244}Cm$ | $1.81 \times 10^1$ | $4.02 \times 10^6$ | $1.08 \times 10^7$ | $7.73 \times 10^4$ |

We see from Table I that the ($\alpha$,n) source neutrons from $^{244}Cm$ are only a small fraction (~1%) of the SF neutrons, so both the singles and doubles rates are directly proportional to the $^{244}Cm$ content. The $^{244}Cm$ spontaneous fissions dominate all other sources of neutrons, and the neutron multiplication is negligible so the $^{244}Cm$ mass can be determined from this simple singles (S) equation:

$$S = \varepsilon\, m_{Cm}\, Y\, (1 + \alpha),$$

where

$\varepsilon$ = counting efficiency for the shielded tubes ≈1.35%,

$m_{Cm}$ = mass of $^{244}Cm$ (grams),

Y = SF neutron yield from $^{244}Cm$ (n/s•g-$^{244}Cm$) = $1.08 \times 10^7$ n/s•g-$^{244}Cm$,

$\alpha$ = ratio of ($\alpha$,n) emission to the SF emission rate ≈about 0.03 for spent fuel.

The counting efficiency is measured using a calibrated $^{252}Cf$ source, and $\alpha$ can be calculated for typical oxide mixtures. If the wastes contain significant amounts of boron or fluorine, the $\alpha$ value can increase enough to be no longer negligible.

A container with fission products corresponding to a gram of plutonium would also contain ~2–3 mg of $^{244}Cm$ for the high burn-up spent fuel. The singles rates would be

S (Cm) ~490 counts/s,

S (Pu) ~25 counts/s, and

S (Bkg) ~3 counts/s.

The high $^{244}Cm$ counting rate makes the sensitivity to plutonium very good when the $^{244}Cm$/Pu ratio method can be used. The software uses the isotopic ratios including $^{244}Cm$ to calculate the total plutonium. If the container has impurities such as boron or fluorine, the value of $\alpha$ would increase, depending on the coupling between the plutonium and the impurity, but the $\alpha$ source neutrons would still be only a small fraction of the spontaneous fission neutrons.

## 3. WCAS DESIGN

Both RH waste and cold waste assay are performed in the same detector so the shielding was configured to have a combination of shielded and unshielded $^3He$ tubes. The shielded tubes were placed behind 76-mm-thick iron bars, and the unshielded tubes were left exposed to the gamma dose. This reduced the total shield weight compared with a full iron shield by about and order of magnitude. For the cold sample measurements, all tubes are used; whereas, for the hot samples only the shielded tubes are

used. The software is used to make the switch between the two configurations based on the gamma dose and the curium ratio.

The design of the WCAS-A detector body used the Monte Carlo code for neutron and photon transport (MCNP) computer code[5] to meet the following design goals:

- Shielded detectors for counting RH waste.
- Unshielded plus shielded tubes for the cold waste.
- Relative uniform efficiency at all positions inside the sample cavity.
- Insensitivity to a wide range of matrix loadings.
- Capability for accurate plutonium assays for crates loaded with up to 4000 kg of mixed metal waste.
- Detectability limit much less than 1 g of plutonium.

To meet these design goals, $^{3}$He detector tubes containing 4-atm gas pressure were used. A modular design was selected that used shielded and unshielded modules. The detector cavity for the WCAS-A required 98 tubes to give complete 4$\pi$ coverage of the sample.

The design is very insensitive to matrix variations so that the efficiency change is less than 3% as the iron loading changes from zero to 4000 kg. The efficiency is more sensitive to materials containing hydrogen; however, the efficiency change is less than 5% for the first 200 kg of typical combustible materials.

The detector body was designed with a minimum amount of iron or other metals such as cadmium that have a high cosmic-ray interaction probability. The high density polyethylene (HDPE) is held in place by aluminum framing. This material selection reduces the cosmic-ray induced spallation neutron background.

## 4. SYSTEM DESCRIPTION

The detector contains 98 $^{3}$He tubes that have an efficiency for $^{252}$Cf that was measured to be 9.55% for an empty crate. Each of the 98 AMPTEK A111 amplifiers is located in a hermetically sealed high-voltage junction box that services two or three $^{3}$He tubes. The 5-Volt logic signal out of the amplifier-discriminator is transmitted to a light-emitting diode (LED) display board. The status of each amplifier output can be seen on the side of the detector shield.

Figure 1 shows the WCAS-A system at the Albuquerque fabrication site. The large crate on the

right side of the assay system was used to hold surrogate matrix materials to benchmark the MCNP calculations. On top of the detector shield is the AAS shield that is 60-cm cube of HDPE. The AAS $^{252}Cf$ source has a yield of ~ 200,000 n/s (45 micro-Ci) at the time of initial operation.

*Fig. 1. WCAS-A system at the Albuquerque fabrication site with an iron surrogate crate.*

The electronic rack that contains the EG&G Advanced Multiplicity Shift Register (AMSR)[6] and the computer. The AMSR is used to collect the data and to sort the counts into the multiplicity distribution. The computer operates with the Neutron Coincidence Counting (NCC) software that has been customized for the WCAS-A application.

## 5. PERFORMANCE CHARACTERISTICS

The gains of all tubes and amplifiers were measured and matched for consistency. Figure 2 shows the high-voltage plateau for the sum of the amplifier/tube combinations. The WCAS-A is operated at 1680 V that is below the plateau region (1760–1960 V) to be free of gamma-ray pileup pulses.

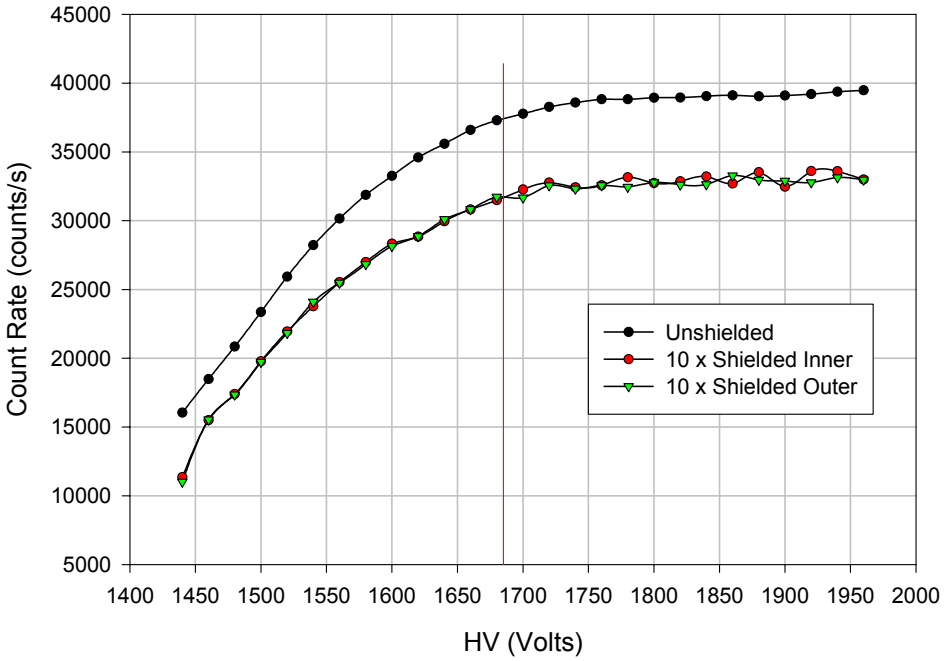


*Fig.2. High-voltage plateau curve for the sum of the amplifiers in WCAS-A.*

The detector was designed to have a flat efficiency profile over the volume of the sample cavity. Figure 3 shows the measured singles rates from a $^{252}$Cf source at different positions in the sample cavity. We see that over the dimensions of the large crate, the efficiency is relatively uniform.

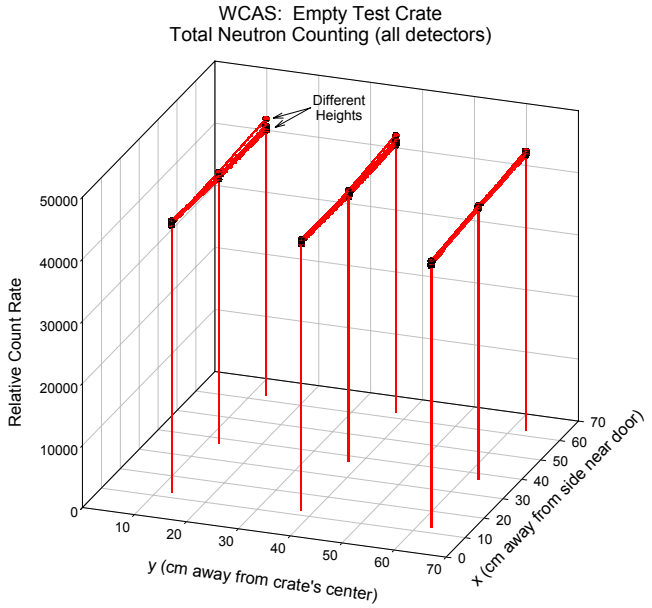


*Fig. 3. The singles efficiency profiles in the WCAS-A sample cavity.*

Table II gives the measured die-away times for different gate intervals. The die-away time is about 125 μs for gate intervals between 100 μs and 400 μs. The relationship between the gate length and the statistical counting error is shown in Fig. 4. The statistical error is at a minimum for a gate setting of ~200 μs. The optimum gate setting is a function of the counting rate, and a shorter gate would be used for kilogram plutonium samples. The WCAS-A detectors and electronics can measure a wide range of plutonium masses from a few mg to tens of kg covering seven orders of magnitude.

**Table II**. Measurement of Die-Away Time with an Empty WCAS-A

| Gate | Singles | Doubles | Triples | τ(μs) |
|---|---|---|---|---|
| 25 | 4682 | 107.0 | 1.9 | — |
| 50 | 4679 | 196.3 | 10.2 | 92.2 |
| 100 | 4547 | 318.5 | 11.1 | 105.5 |
| 200 | 4543 | 466.1 | 28.5 | 130.0 |
| 400 | 4551 | 562.3 | 42.3 | 126.7 |

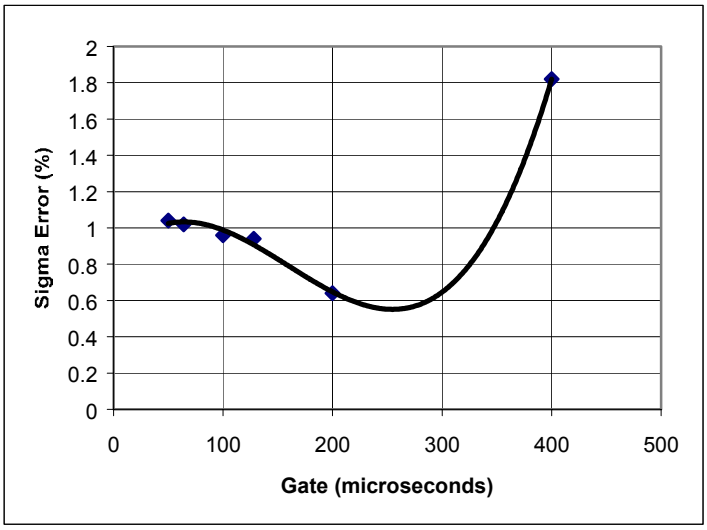


*Fig. 4. The statistical error in the doubles as a function of the coincidence gate length for a small sample with a 450-second measurement.*

The detector efficiency was measured using a NIST calibrated $^{252}$Cf source. The efficiency, die-away time, and coincidence gate fractions are given in Table III. The dead-time coefficients correspond to the $^{252}$Cf normalization source that is counted near the bottom of the sample cavity. For waste type samples, the counting rates are low and the dead-time corrections are negligible.

| **Table III**. Detector Performance Parameters | |
|---|---|
| Parameter | WCAS-A |
| Neutron efficiency for $^{252}$Cf | 9.55 % |
| Neutron die-away time | 125 µs |
| Dead time* (a) | 0.8 µs |
| (b x $10^{-6}$) | 0.26µs$^2$ |
| Multiplicity dead time | 100 ns |
| Coincidence gate | 200 µs |
| Predelay | 1.5 µs |
| Doubles calibration coefficient | 5.46 counts/s•g $^{240}$Pu |
| $^{244}$Cm singles calibration | 150000 counts/s•g Cm |
| Doubles gate fraction | ~0.750 |
| Triples gate fraction | ~0.563 |

* The dead-time coefficients correspond to the $^{252}$Cf AAS that is used as the normalization reference.

The neutron multiplication in waste samples is negligible and the multiplication equals unity. However, the detector system can also be used for cans of plutonium, oxide, or metal components that have a significant multiplication by using a multiplication correction software option.

## 6. CALIBRATION

The WCAS-A was calibrated with an empty sample chamber. Actual measurements with loaded containers are corrected back to the empty container calibration using the AAS method or the truncated multiplicity approach.

Because there is negligible multiplication and no neutron self-shielding in the plutonium over the plutonium mass range of interest, the calibration curve is expected to be a straight line through the origin. There are separate calibration lines for the singles and doubles rates, and we are using the doubles calibration for the primary assay result for plutonium and the singles rate for $^{244}$Cm. Figure 5 shows the doubles calibration for the $^{240}Pu_{eff}$ where

$$^{240}Pu_{eff} = 2.52\ ^{238}Pu + {}^{240}Pu + 1.68\ ^{242}Pu\ .$$

Figure 6 shows the singles calibration for $^{244}$Cm where the detector is in the low-efficiency mode that uses only the shielded $^3$He tubes.

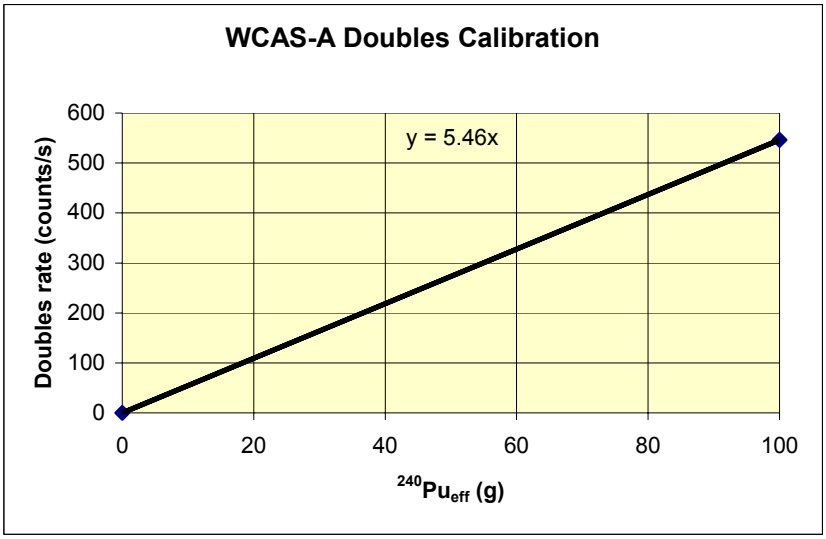


*Fig. 5. The WCAS-A doubles calibration for $^{240}Pu_{eff}$ in the high efficiency mode.*

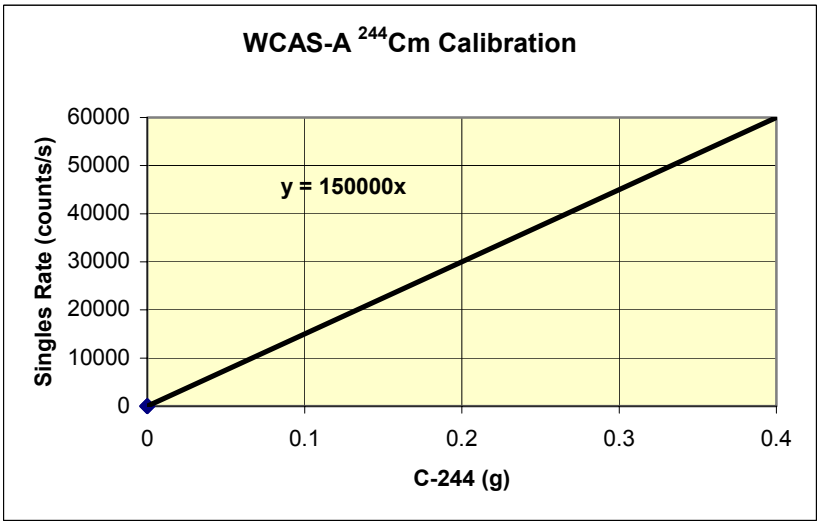


*Fig.6. WCAS-A singles calibration for $^{244}Cm$.*

## 7. SHIELDING AND BACKGROUND REDUCTION

The WCAS-A has two types of shielding: (1) internal iron for the high gamma dose in the RH waste, and (2) external polyethylene for the cosmic-ray source neutrons. Unfortunately, the internal iron is a target for the production of cosmic-ray neutrons, so we designed a shielding configuration with minimum iron by shadow shielding about 40% of the tubes behind iron bars (76 mm thick × 152 mm wide) as shown in Fig. 7. Two $^{3}He$ tubes are positioned behind each bar at different depths into the HDPE to provide a front/back ratio that is sensitive to the neutron moderation in the sample. This can provide an additional check on the hydrogen content in the matrix. Between each iron bar, there are three

unshielded tubes to increase the counting efficiency for the cold samples.

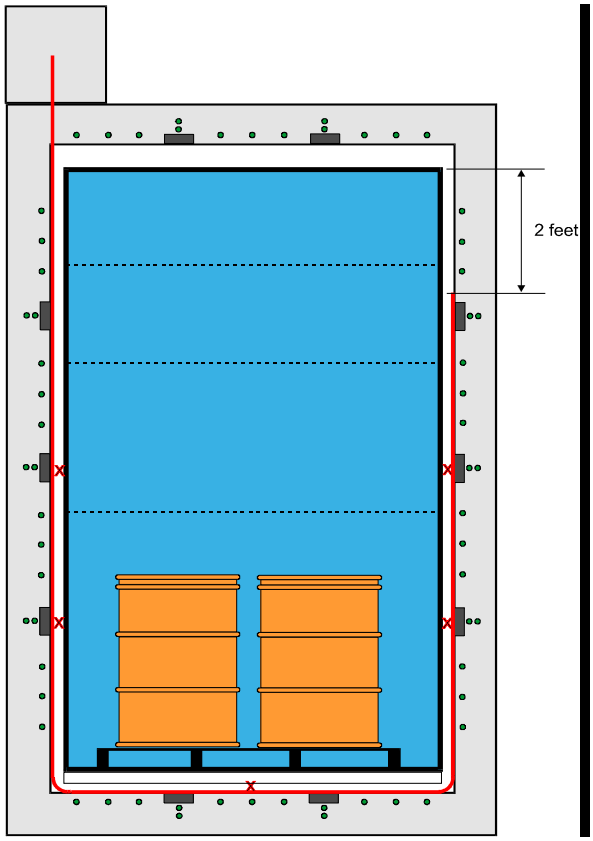


*Fig.7. WCAS-A cross section schematic to show the $^{3}He$ tubes, the iron shielding bars, the AAS shield and guide tube.*

A detailed shielding calculation was done with the Monte Carlo code MCNP using a fission product gamma-ray spectrum from an extended source and a detailed conversion of gamma-ray flux to dose rate as a function of energy. The spectrum was calculated with the ORIGEN2 code for a burnup of 35,000 MWd/tU and 5 years of cooling. A thickness of 3.0 inches (76 mm) has a dose reduction factor of about 33 (transmission of 3%). This thickness of iron reduces the maximum dose at the tube to about 3 R/h, which we can discriminate against. The $^{3}He$ tubes are 1700 mm long and there is a separate AMPTEK amplifier for each tube to limit the gamma-ray pileup.

### 7.1. Cosmic-Ray Neutron Shielding

Cosmic-ray neutron background reduction is achieved by means of the TM method.[4] In the software, the full multiplicity distribution is truncated to remove the higher multiplicities that correspond to cosmic-ray spallation neutrons. The first three real multiplicities [the zeros (singles), ones (truncated

doubles), and twos (truncated triples)] cover more than 97% of the plutonium spontaneous fission neutrons for low counting rates. The ratio of the high and low multiplicities can be used to correct for the iron and other metals in the matrix.

The neutron backgrounds originate from several sources: (1) local area radiation storage (e.g., waste in loaded crates), (2) external-area cosmic-ray neutron sources, (3) cosmic-ray spallation in the detector body, and (4) cosmic-ray spallation in the sample. The first two of the sources create a singles neutron background, and the concrete walls and the 100 mm of polyethylene shielding on the exterior of the detector effectively eliminate this source. The third and fourth neutron sources are significantly reduced by the TM data reduction in the software. The standard statistical filtering method[7] is also used to eliminate cosmic-ray neutron events with high multiplicity.

The cosmic-ray spallation neutron background in the detector body and the sample is difficult to shield because the cosmic rays are very penetrating. One meter of overhead concrete provides a factor of ~ 4 reduction in the neutron background. The 100 mm of HDPE shielding on the detector provides less than a 10% background reduction in the coincidence rate. However, the overhead concrete shielding in the building provides more than an order of magnitude reduction in the neutron background rate. To help reduce the spallation rate, we have used low-Z materials in the detector design such as HDPE and aluminum. The doubles background after installation is estimated to be ~ 0.16 counts/s.

## 8. MATRIX CORRECTIONS

In the consideration of matrix correction on the assay results, there are two major matrix categories to consider:

(1) Primarily metal (iron and mixed metals), and
(2) Hydrogenous matrix materials (combustibles, plastics, etc.).

The WCAS detector was designed so that the metal matrix materials have a negligible effect on the counting efficiency and no matrix efficiency corrections are required. The assay result for metals is independent of the position of the plutonium in the crate.

Corrections for waste matrix effects on the observed counting rates are addressed through the AAS technique. The basis of the AAS method is to measure the matrix perturbation to the counting rate

from a small $^{252}Cf$ source that is extracted from a shield and positioned adjacent to the waste container wall at a select number of locations. The source is then retracted into the shield for the duration of the passive measurement. The observed $^{252}Cf$ AAS perturbation is related to a matrix correction factor through an empirical relationship. This relationship is established through measurements of a set of simulated waste matrices spanning the properties expected in actual waste.

A volume-averaged perturbation for each matrix type is obtained by counting a neutron source at multiple positions within an empty waste container and for the simulated waste matrices. This volume-averaged doubles ratio for the empty waste container over the matrix-filled container gives the volume-averaged perturbation over the matrix set. For each waste-matrix configuration, the plutonium-source-empty to matrix-filled-container doubles ratio is also measured or calculated using the MCNP code. These are plotted against each other to obtain an AAS perturbation correction factor curve that addresses a variety of matrices. The actual waste-container-specific AAS perturbation can then be used to compute a corresponding matrix correction.

The AAS correction assumes that the plutonium is not concentrated at the center or edges of the box container because the correction factor corresponds to the volume average for the container. Where practical, a multiplicity-based efficiency correction can be used to make the matrix correction for comparison with the AAS correction. The neutron multiplicity analysis uses the singles, doubles, and triples rates to solve for the $^{240}Pu_{eff}$ mass and the efficiency. This analysis gives the matrix correction relatively independent of plutonium distribution; however, it is only achievable for containers with sufficient plutonium mass loadings, and the statistical precision is not as good as for the AAS method.

## 9. DETECTABILITY LIMITS

The minimum mass detectability limit for WCAS-A depends on the location of the system, the type of matrix, and whether singles or coincidence rates are used for the assay. The detectability limit is determined by the coincidence neutron background generated by cosmic-ray spallation reactions. At sea level, the detectability limit is a factor of ~2 lower than at the higher elevation (~1500 m) at Albuquerque.

In general, the detectability limit increases as the square root of the neutron background rate. Crates loaded with a high iron content have an increase in the coincidence background from the spallation reactions in the iron. The TM method can eliminate part of the coincidence background. The WCAS-A is designed for both drums and crates, and the detectability limit for drums is lower than for crates

because of the high mass of iron in the crates. A typical pallet of four 200-L drums has a tare weight of about 80 kg and an empty iron crate weighs ~660 kg. The background from the detector is determined from long background measurements with an empty crate.

If the detectability limit is defined as three times the standard deviation (3 $\sigma$) in the background, then repeat measurements of the background can be used to predict the minimum detectable mass that gives a signal rate equal to 3 $\sigma$ of the background scatter. The initial measurements (20 minutes) at Albuquerque gave a detectability limit of ~70 mg Pu. The cosmic-ray neutron background rates at the waste storage building are expected to be about 40 times lower than at Albuquerque, and the detectability level is estimated to be ~10 mg Pu for the installed location.

In the singles mode, the detectability limit for plutonium is reduced by an order of magnitude because of the high specific neutron yield from $^{244}$Cm. The $^{244}$Cm singles rate is dominated by spontaneous fission neutrons so the variation in alpha has only a small effect on the measured rate.

## 10. SUMMARY

Innovations in design, analysis, and fabrication have enabled the WCAS-A system to exceed performance expectations, as demonstrated during the factory acceptance test conducted by JNFL in April 2001. The system can operate for both RH waste and cold waste based on the waste generation site. Lower detectability limits for total plutonium are at the 10–20 mg levels for the direct $^{240}$Pu measurement for cold wastes. For the indirect measurements of the RH via the curium ratio, the sensitivity level is ~1-mg of Pu.

The system has demonstrated that very large containers (8000-L) of mixed metals and combustible debris can be measured in short times using both passive neutron singles and coincidence counting. In general, the average density of the waste gets smaller as the container gets larger because of overall weight limits for the handling systems. The large void fraction in the big containers allows for adequate leakage of neutrons from the interior of the crate.

Twenty-minute assay times have met design specifications. The success both in design/fabrication and in physics performance of such enormous assay systems has important implications for domestic applications, such as assay of RH waste for disposal at the Waste Isolation Pilot Plant. The system is designed for dual use by the International Atomic Energy Agency and the facility operator.

## ACKNOWLEDGMENTS

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