Operator Experience Measuring Pu-bearing Waste at PNC Using the Waste Drum Assay System*

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Abstract

The Waste Drum Assay System (WDAS) with Add-A-Source matrix correction is a passive neutron coincidence counter designed to accurately measure the mass of plutonium in 200 liter waste drums. Power Reactor and Nuclear Fuel Development Corporation (PNC), Los Alamos National Laboratory and Canberra installed and calibrated six WDASs, built by Canberra, at PNC's production facilities. The WDAS provide input into the site's MC&A program. The materials assayed by the WDAS, results of the calibration tests, and operating experience are reported.

1. Introduction

To safely utilize plutonium for nuclear fuel, it is necessary to develop handling techniques for plutonium and the fuel fabrication processes. Fundamental requirements for the facility handling plutonium include: confinement of the plutonium, criticality control, radiation control and safeguards/[1]/. Passive neutron coincidence counting of the spontaneous fission neutrons emitted by the even isotopes of plutonium provides an accurate and reliable assay result that can be input into a site's material control and accountancy (MC&A) program. The Waste Drum Assay System (WDAS) was jointly developed by Canberra and Los Alamos National Laboratory (LANL) for its improved accuracy and high reliability, and Authorized For Use (AFU) by the IAEA. Six WDAS have been installed at four Power Reactor and Nuclear Fuel Development Corporation (PNC) facilities in Tokai. This is the largest installed quantity of the same waste assay system at a single site worldwide providing operating experience for the same system type in different operating environments. The PNC facilities where the WDAS's are installed, system description, calibration procedures and measurement results will be reported.

2. Facility Description

PNC's Tokai Works has undertaken R&D aimed at identifying the optimum fuel recycling method for Japan. This R&D includes:

- reprocessing of spent fuel from fast breeder reactors (FBR) as well as LWRs
- waste technology development on high-level liquid wastes and transuranic radionuclides
- fabrication of plutonium fuel assemblies for FBRs and Advanced Thermal Reactors (ATR)
- uranium enrichment by means of molecular laser as well as gas centrifuges and
- development of new technology.

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An aerial view of PNC's Tokai Works is shown in Figure 1. PNC has two main facilities for MOX fuel production. The PNC Plutonium Fuel Fabrication Facility (PPFF), which includes PFDF and PFFF, was constructed to confirm the developed fuel production technology on a large scale. PPFF began producing fuel in January 1972 and has been supplying MOX fuel for the DCA reactor, prototype ATR "FUGEN", and the experimental FBR "JOYO". Initial core fuel assemblies were completed for "JOYO" in 1975 and for "FUGEN" in 1978. Production for "FUGEN" continues today. WDAS #2 was installed on the first floor of PPFF.

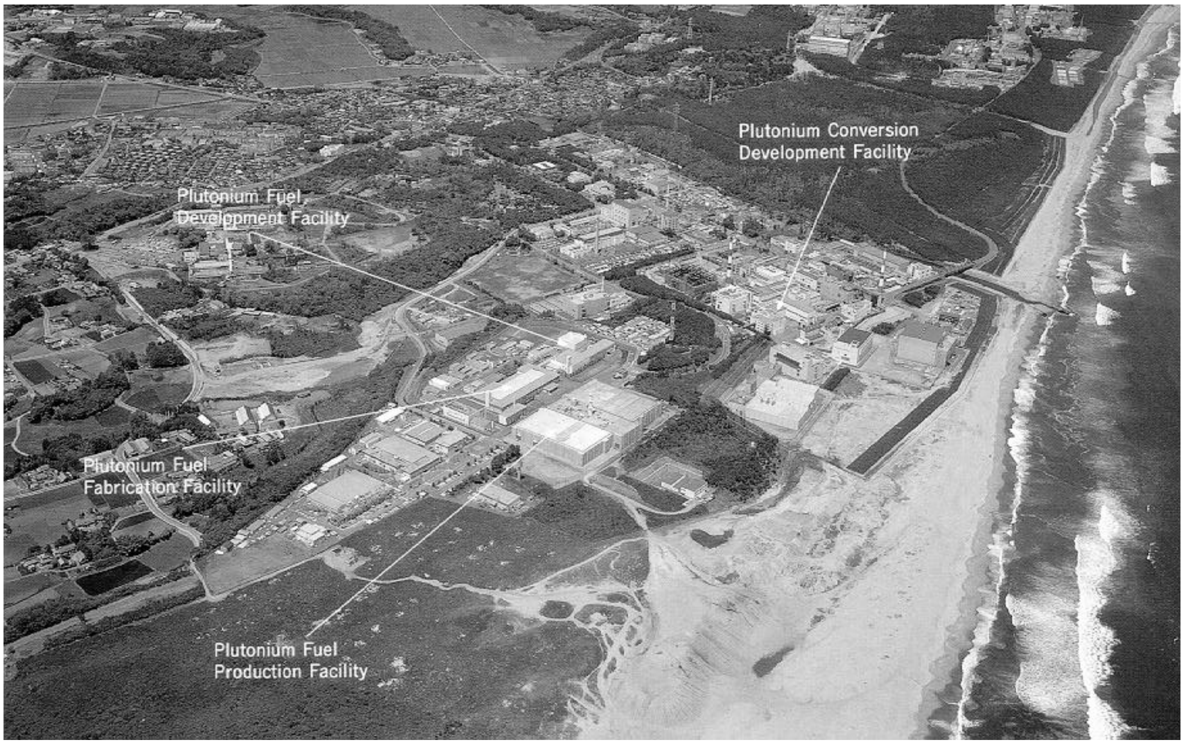

Figure 1. Aerial View of PNC's MOX Fuel Production Facilities

Based on the experience of MOX fuel fabrication technology developed in PPFF, the Plutonium Fuel Production Facility (PFPF) was constructed in October 1987 to demonstrate the mass production technology of MOX fuel, employing world-leading remote and automated technology. PFPF started producing "JOYO" MOX fuel in October 1988. After overcoming a number of technological issues of low-density pellet production by remote and automated systems, the fabrication of the initial fuel for the FBR "MONJU" was completed in January 1994. To accommodate the expected waste flow from this facility three WDAS were installed in PFPF: WDAS #1 and #4 are installed in the same room on the first floor and WDAS #6 in the basement.

In addition to counters installed in the MOX fuel production facilities, WDAS #5 is installed on the first floor of the Tokai Reprocessing Plant (TRP) and WDAS #3 is installed in the Plutonium-contaminated Waste Compact Area (PWCA). TRP began operation in January 1981. With a design process capacity of approximately 0.7 metric ton uranium per day, it is an important step in the establishment of an independent nuclear fuel cycle in Japan. The uranium trioxide (UO_3) and plutonium nitrate [$Pu(NO_3)_4$] product material are transferred to Ningyo Toge for enrichment and PFPF for MOX fuel production, respectively.

PWCA provides volume reduction of the waste generated from the MOX fuel production and provides a basic study for disposal techniques. Depending on the matrix, the plutonium-contaminated waste drum can go through pretreatment and sorting, incineration and melting. Prior to interim storage, the waste drums are assayed in WDAS #3.

3. Waste Description

Five waste types are generated at the MOX facilities and TRP. They are summarized in Table 1. Inflammable, partial combustible and incombustible are placed in cardboard containers measuring 280 mm in diameter by 350 mm tall. Six cardboard containers fit inside a 200 liter drum (see Figure 2) or two HEPA filter boxes fit inside a 200 liter drum. The metal ingots and ceramic-like blocks are also placed in 200 liter drums (see Figure 3).

Table 1. PNC Waste Types

Waste Type	Description
Inflammable	Dry and wet paper, and rags generated at the fuel fabrication process and the chemical analysis process; HEPA filter boxes
Partial Combustible	Gloves and PVC material generated at the fuel fabrication process and the chemical analysis process
Incombustible	metal
Metal Ingot	calcined metal that is melted
Ceramic-like Block	Ash produced by incineration of inflammable waste (category 1) is formed into a ceramic-like block with a microwave melter

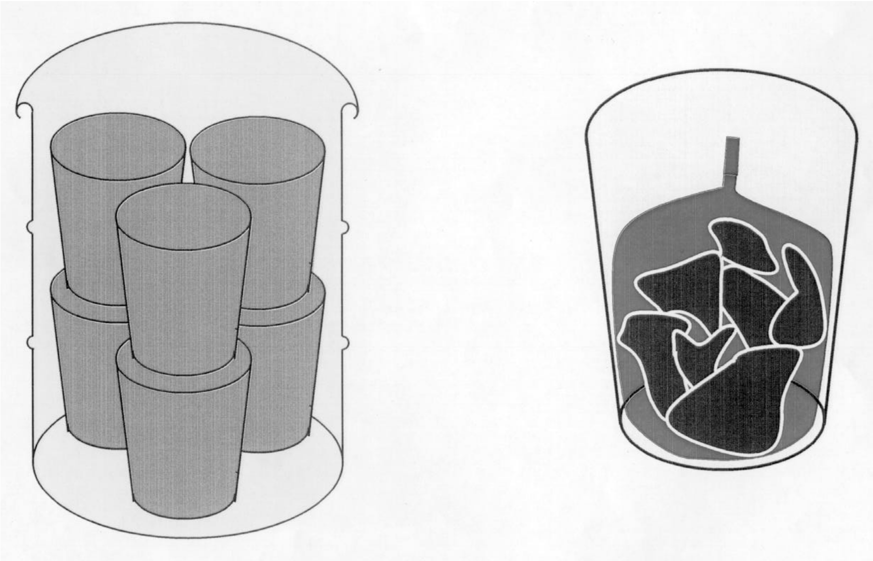

Figure 2. Container for inflammable, partial combustible and incombustible waste. The left sketch shows the 200 L drum with six cardboard boxes inside. The right sketch is an individual cardboard box.

4. System Description

The WDAS (shown in Figure 4) utilizes a 4π detector geometry consisting of ^{3}He detectors embedded in a high density polyethylene moderator to maximize efficiency and provide a uniform spatial response. A photograph of WDAS #4 in PFPF is shown in Figure 4. To distinguish the neutron emission of the plutonium isotopes from interfering (α-n) reaction neutrons, a coincidence counting technique is employed. The Add-A-Source (AAS) correction technique, which was developed by LANL, uses a small ^{252}Cf source on the outside of the drum to correct for the matrix effects on the plutonium inside of the drum, improving the assay result. In WDAS #1-3, the AAS is positioned under the drum. In WDAS #4-6, the AAS is positioned at multiple positions along the side of the drum. Detailed performance data is published for the WDAS/[2,3]/.

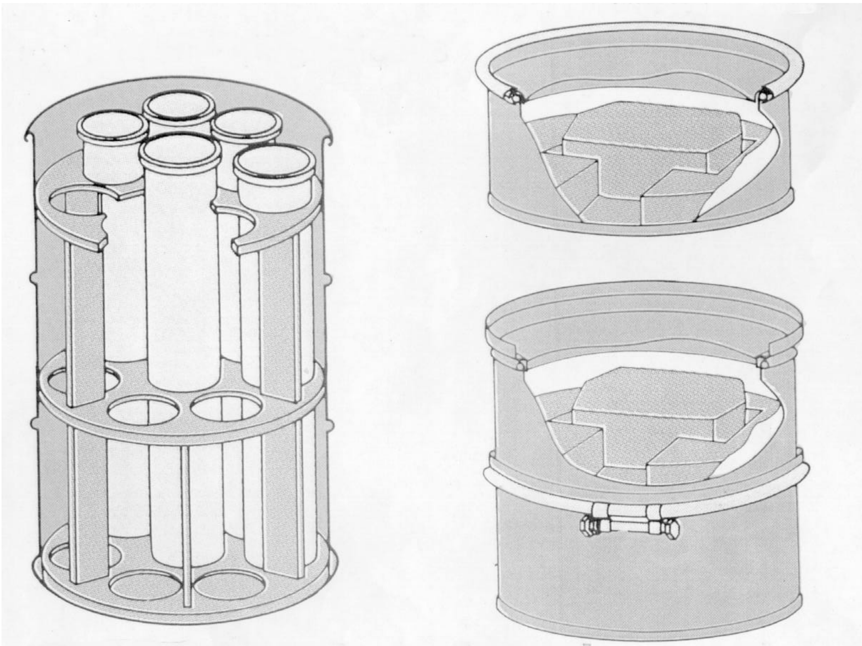

Figure 3. Containers for metal ingots (left sketch) and ceramic-like blocks (right sketch).

Each WDAS is integrated with a gamma spectroscopy system that measure the isotopics of the plutonium allowing calculation of the total plutonium mass in the waste drums. The gamma spectroscopy systems with WDAS #1-3 use the FRAM/[4]/ code developed at LANL and WDAS #4-6 use the enhanced Multi-Group Analysis Code (MGA)/[5,6]/ that was developed by Dr. Ray Gunnink under contract to Canberra, to measure the plutonium isotopics.

Figure 4. WDAS #4 installed in PFPF

5. Calibration Procedure

The six WDAS were installed and calibrated by LANL, Canberra, and PNC. Two calibration procedures were performed: AAS calibration for the matrix correction and mass calibration. These calibrations are described in detail in reference 7. A summary of the results follows. The AAS calibration was performed on known MOX mass loadings that were positioned inside six cardboard boxes in the matrix drums. The MOX standards were positioned ~250 mm above the bottom and halfway between the center and outside of the cardboard box.

The AAS perturbation and the plutonium assay perturbation for WDAS #1 were plotted for various low-density matrices. The calculated coefficients of this curve, $f(x) = a_0 + a_1 x + a_2 x^2$, are:

a_0 = -0.0201
a_1 = +0.60106
a_2 = +0.82810

For samples of unknown plutonium mass, The WDAS software uses the calculated coefficients along with the AAS measurement of the sample, and automatically calculates the AAS correction factor, 1+f(x), to correct the measured reals rate for matrix effects. Moderating and absorbing matrices such as the 27.4 kg H_20 in paper have the largest AAS correction factor (see Table 2). Figure 5 shows the relative Reals rate before and after AAS correction. These data indicate that significant improvement in the assay accuracy can be achieved, even for drums containing large amounts of moderators and absorbers for uniform source distributions.

Table 2. AAS Correction Data for Various Matrices

Matrix Type	Net Weight (kg)	Average Reals/g (counts/s-g ^{240}Pu)	f(x)	AAS Correction Factor (CF)	Corrected Reals/g (counts/s-g ^{240}Pu)	Matrix versus Empty Drum % Difference
Empty	0	17.10	0	1.00	17.10	0
Dry paper	12.9	17.17	0.022	1.022	17.54	+2.6
9.5 kg H_20 in paper	12.9 + 9.5	15.09	0.165	1.165	17.43	+2.5
15.7 kg H_20 in paper	12.9 + 15.7	12.75	0.301	1.301	16.59	-3.1
27.4 kg H_20 in paper	12.9 + 27.4	9.23	0.865	1.865	17.21	+0.6
26 kg rubber gloves	26	14.61	0.133	1.133	16.55	-3.3
15 kg PVC bags	15	16.31	0.054	1.054	17.19	+0.5
15 kg PVC + 6 kg rubber	15 + 6	15.72	0.111	1.111	17.46	+2.1
HEPA filter	not applicable	16.09	0.093	1.093	17.58	+2.8

Once the AAS calibration was completed, the mass calibration was performed for WDAS #1 using MOX standards from 0.243 to 62.7 g ^{240}Pu-effective in matrices similar to the AAS calibration. The calculated calibration coefficient from these measurements was 17.4 +/- 0.5 Reals/g ^{240}Pu-effective. The WDAS #1 was put into routine use at PFPF in November 1991. In October 1994, the calibration was repeated by the IAEA to authenticate the system for routine use. The IAEA calibration used large metal boxes in addition to the drum containing the six cardboard boxes. The MOX standards used by the IAEA included 10 each 1-g plutonium standards and 6 each 30-g plutonium standards. After correcting the larger standards for multiplication, the calibration coefficients were calculated to be 17.3 +/- 0.4 for the drums containing cardboard boxes and 19.18 for the large metal boxes.

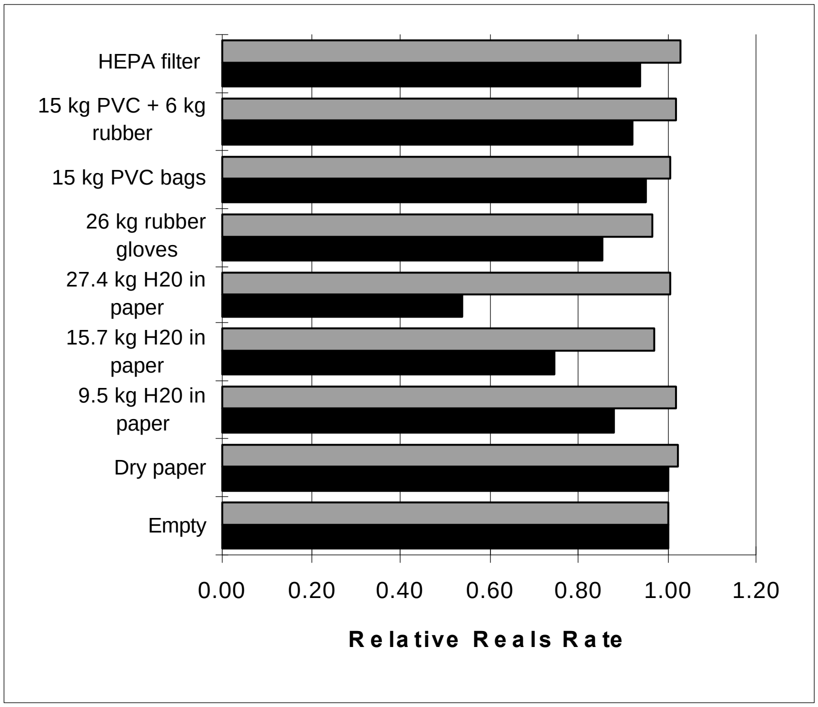

Figure 5. Relative reals rate before and after AAS correction. The dark line is the uncorrected relative reals rate and the light-colored line is the corrected relative reals rate.

A ^{252}Cf neutron source was used to verify that the spatial responses and AAS calibrations are similar on WDAS #1-3. The low-density matrices perform similarly in WDAS #1-3. As described above, WDAS #3 which is installed at PWCA, also must be able to measure high-density metal ingots and ceramic-like blocks. These matrices exhibit no moderation, but high neutron scattering properties. The system software automatically sets the AAS correction factor to 1 for these drums. The same ^{252}Cf neutron source was used to cross-calibrate WDAS #2 and #3 to WDAS #1. The position of the AAS in WDAS #4-6 required a new AAS and mass calibration. The same calibration procedures with the same matrices used for WDAS #1-3, described above, were repeated for WDAS #4-6. The operating parameters and calibration coefficients for WDAS #1-6 are listed in Table 3.

Table 3. Operating parameters for WDAS #1-6

Operating Parameter	WDAS #1	WDAS #2	WDAS #3	WDAS #4	WDAS #5	WDAS #6
Predelay (μsec)	4.5	4.5	4.5	4.5	4.5	4.5
Gate length (μsec)	128	128	128	128	128	128
Die-away time (μsec)	80.0	73.0	73.0	79.9	78.0	82.0
Calibration coefficient, a	-0.02010	-0.02010	-0.02010	+0.0000	+0.0000	+0.0000
Calibration coefficient, b	+0.60106	+0.60106	+0.60106	+1.5500	+1.5500	+1.5500
Calibration coefficient, c	+0.82810	+0.82810	+0.82810	-0.2490	-0.2490	-0.2490
Mass calibration coefficient	17.30	16.80	16.82	18.09	18.08	17.95

6. Measurement Results

Waste samples were measured by PNC in WDAS #1 and WDAS #4 to allow comparison of performance data (Table 4). Sample identification numbers (i.d.) that begin with S-A and S-N, refer to inflammable and partial combustibles waste types, respectively. The calibrations were verified against the IAEA standard drum and agreement was within 2.9%.

Table 4 . Comparison of Plutonium Mass for WDAS #1 and WDAS #4.

Sample ID	Plutonium mass (g)		Deviation (%)
	WDAS #1	WDAS #4	((#1-#4)/#1)x100
S-A-0498	0.777 (0.036)	0.788 (0.039)	-1.42
S-A-0452	0.313 (0.016)	0.353 (0.013)	-12.8
S-N-0607	0.493 (0.021)	0.500 (0.021)	-1.42
S-N-0610	1.241 (0.064)	1.255 (0.063)	-1.13
S-A-0457	0.732 (0.031)	0.764 (0.033)	-4.37
S-A-0500	0.061 (0.005)	0.075 (0.007)	-23.0
S-N-0599	0.761 (0.033)	0.828 (0.038)	-8.8
S-N-0597	0.257 (0.017)	0.235 (0.017)	8.8
S-N-0605	0.753 (0.031)	0.775 (0.034)	-2.92

Minimum Detectable Activity

The minimum detectable activity (MDA) is measured to determine the activity range and environmental conditions (e.g., background) where the system can be applied. The MDAs for the coincidence mode are reported in Table 5 with the measurement conditions. The totals MDAs can be significantly lower than that obtained from coincidence counting. But because the Totals background can vary based on movement of waste near the counter, the Totals MDA is normally used only as a conservative screening technique in reporting the total alpha activity in the drum.

Table 5. MDA for WDAS

Measurement Parameters	WDAS #1 PFPF Floor 1	WDAS #2 PFFF	WDAS #3 PWCA	WDAS #4 PFPF Floor 1	WDAS #5 TRP	WDAS #6 PFPF Basement
B_T = Totals background rate	4.74	10.61	2.07	5.12	47.64	25.68
B_R = measured net Reals background rate	0.017	0.035	0.011	0.043	0.04	0.016
Effective overhead shielding (cm)	60	30	90	60	0	90
MDA for Reals (mg ^{240}Pu-effective)	1.07	1.54	0.92	1.40	3.26	1.92
MDA for Totals (mg ^{240}Pu-effective)	0.64	0.96	0.43	0.68	2.03	1.50

The Totals background rates for WDAS #1-6 vary depending on the amount of plutonium stored in the area near the counters. WDAS #6 is installed in the basement of PFPF but several hundred of plutonium-bearing waste drums are stored nearby. The same is true for WDAS #5 in TRP where waste drums and crates are stored in the room near the counter. The Reals background rates are more dependent on the effective overhead shielding (e.g., thickness of concrete shielding over the counter). Even though the Totals background rate for WDAS #6 is large, the Reals background rate is among the lowest because of the 90 cm of concrete above the counter. The large amount of concrete reduces the background from cosmic ray spallations.

7. Conclusions

Operating experience at PNC with WDAS #1-6 indicate that passive coincidence counting with AAS correction can provide accurate assay results for input into the facility's MC&A program. This conclusion is reinforced by the IAEA's authentication and routine use of the WDAS systems. Installation of six similar systems throughout Tokai Works provides data for comparison of the performance including the MDA. MDAs approximately equal to 1 mg ^{240}Pu-effective are achievable even in areas with high totals backgrounds such as storage areas.

8. References

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