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Calibration and Operation of the PNL Barrel Assayer

R. J. Arthur

July 1991

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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BATTELLE MEMORIAL INSTITUTE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America

Available to DOE and DOE contractors from the
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Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY

Pacific Northwest Laboratory operates a Barrel Assayer to measure the radionuclide content of 208-liter waste drums. This mobile apparatus is designed to provide quantitative information on each radioisotope in a drum and to recognize and locate "hot spots" within the drum. A lead-collimated intrinsic-germanium detector mounted on a moveable platform vertically scans a rotating drum for gamma-ray activity while 62 stationary $^{10}\text{BF}_3$ tubes measure neutrons emitted by transuranic isotopes within the waste.

Several approaches have been used to document the controls under which the Barrel Assayer has been operated from February 1989 through December 1990. Resin-filled 208-l calibration barrels containing known quantities of ^{60}Co , ^{137}Cs , and ^{134}Cs were measured by the Barrel Assayer and found to be well within 25% of the known values. A thorium nitrate standard that served as the secondary, or field, standard verified that the detector has been within calibration during the period from February 1989 through December 1990.

In previous testing, 23 waste drums were assayed at several commercial nuclear power plants. Subsequently, aliquots from the top, middle, and bottom of the barrels were analyzed. The ratio of grab-sample concentration to direct-assay concentration averaged -1.6 for one plant and +1.3 for another.

Results using the Barrel Assayer were also comparable to those obtained using Safeguards & Security's Segmented Gamma-ray Scanner (SGS) system in a cross-calibration experiment performed by a third party on several drums containing TRU wastes.

CONTENTS

SUMMARY	iii
INTRODUCTION	1
DESIGN AND OPERATION	3
CONTROLS AND PERFORMANCE TESTING	5
ANALYSIS OF TRACEABLE STANDARDS	5
CONTROL STANDARD	9
GRAB SAMPLE ANALYSIS	9
CROSS CALIBRATION EXPERIMENT	20
CONCLUSION	25
REFERENCES	27

FIGURES

1	Schematic Diagram of 208-Liter Barrel Counter Used for Quantitative Isotopic Measurement of Gamma-Ray and Neutron Activity	4
2	Geometry of 208-Liter Drum Used For Calibration With a Mixed-Isotope Source as a Function of Source Geometry and Drum Weight	7
3	208-Liter Drum Observed Efficiency Curves as a Function of Source Geometry and Drum Weight	8
4	208-Liter Drum Calibration Curves (Observed and Extrapolated) Used to Generate Appropriate Efficiency Curve for Given Barrel Weight	8
5	Computer Output From CTL Program Demonstrating that the Detector Was Within Calibration (February 23, 1989 - July 23, 1990)	17
6	Computer Output From CTL Program Demonstrating that the Detector Was Within Calibration (February 2, 1990 - December 14, 1990)	18

TABLES

1	Results from Dow-Resin Filled Gamma-Ray Standard Drum Assay	6
2	Neutron Analysis Results From ²³⁹ Pu Standard Canisters Centered in 55-Gallon Drum	10
3	Computer Results From CTL Program Used to Monitor Detector Reliability Based on Observed Peak Area and Resolution (February 23, 1989 - July 23, 1990)	11
4	Computer Results From CTL Program Used to Monitor Detector Reliability Based on Observed Peak Area and Resolution (February 2, 1990 - December 14, 1990)	14
5	Comparison of the Total Gamma-Ray Activity in Oil-Filled Drums by Grab Sampling Versus Direct Assay	19
6	Comparison of the Total Gamma-Ray Activity in Ion-Exchange-Resin Filled Drums by Grab Sample Versus Direct Assay	19
7	Comparison of Analytical Data on Critical-Mass-Laboratory Waste Drum	21
8	Material Balance Ratios	23
9	Ratio Comparison of Analytical Data on Critical-Mass-Laboratory Waste Drums	24

INTRODUCTION

The Pacific Northwest Laboratory (PNL)^(a) operates the PNL Barrel Assayer, a mobile apparatus developed for measuring the radionuclide content of 208-liter (55-gal) drums and similar smaller containers. Drums containing waste are positioned on a turntable, rotated, and directly measured using both a collimated IG gamma-ray spectrometer positioned at 11 equally-spaced intervals opposite the drum and 62 stationary $^{10}\text{BF}_3$ tubes embedded within two hemicycles of polyethylene moderator. Some of the advantages of this type of system are 1) the counting instrumentation can be transported to drum-storage locations, 2) very large sample sizes can be accommodated, 3) individual isotopic analysis and total gamma activity are determined, 4) the relative vertical distribution of activity within a drum may be determined from the scans, 5) sensitivity and counting geometry variability are improved by analyzing the sum of the 11 segmented gamma-ray scans, and 6) transuranic (TRU) levels can be determined.

This report briefly describes the operation of the Barrel Assayer and documents the controls under which it is and has been operated. The results of grab sample analyses and a cross-calibration experiment are also provided.

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DESIGN AND OPERATION

The PNL Barrel Assayer is schematically illustrated in Figure 1. A lead-collimated, shielded intrinsic-germanium (IG) detector mounted on a movable platform vertically scans a drum from top to bottom. A magnetic position sensor accurate to 0.0025 cm is employed by the software controlling program, "SGS," to locate the detector platform within 0.25 cm of the specified height. A barrel is mounted on a turntable which rotates the drum at approximately 30 revolutions per minute during the vertical scanning. The vertical gamma-ray scanning is normally performed in eleven 7.6 cm segments for 208-l drums, and the segment counts are summed to provide both total gamma-ray activity and specific activity, as well as individual isotopic activity. This information is useful in the event that some portion of the waste in the drum contains a highly unusual radionuclide composition.

The counting system contains a relatively large IG detector, typically with an efficiency for gamma-ray detection of between 19% and 30% (relative to a 7.6 cm by 7.6 cm right-circular sodium-iodide cylindrical detector). The detector is shielded on the side with 2.54 cm of lead. A 5-cm lead collimator is used on the face of the diode with a slit of height 0.635 cm or 2.54 cm, depending on the activity of the waste, exposing the full width of the diode.

The neutron detectors that surround the drum measure neutrons emitted by TRU isotopes within the waste. The detectors are embedded in polyethylene moderator to enhance the efficiency of the measurement. Additionally, TRU concentrations for Pu, Am, and Cm can be directly measured with the IG detector if their concentrations exceed about 1 nCi/g and if their gamma-ray emissions are not dominated by fission or activation product radiations.

Gamma-ray spectral data from the IG detector are collected and stored using a commercial (EG&G ORTEC) ADCAM software program. The resulting 11 spectra from the analysis of a typical barrel are then summed together. The summed data are then reduced using the RAYGUN gamma-ray analysis program and experimentally determined efficiency curves.

The contents of a barrel are assumed to be packed homogeneously in the first analysis. For barrels with detectable activity, the analyst checks this

FIGURE 1. Schematic Diagram of 208-Liter Barrel Counter Used for Quantitative Isotopic Measurement of Gamma-Ray and Neutron Activity

assumption by observing the count rate for each segment and by looking at the resulting disintegration-per-second (dps) factors on radioisotopes with gamma-rays of several energies, such as ^{239}Pu , ^{60}Co , ^{125}Sb , ^{134}Cs , ^{152}Eu , and ^{154}Eu . If a discrepancy is noted in the dps factor, then several methods are available to correct the discrepancy. For example, the segments can be analyzed individually using an appropriate technique to approximate a more accurate attenuation correction.

Resulting gamma-ray spectra are deconvoluted using the RAYGUN gamma-ray analysis code (Hensley et al. 1988), which is based on the GAMANAL (Gunnink and Niday 1972) and GRPANL (Gunnink and Ruhter 1980) codes. Radioisotopes that are difficult to measure are determined on the basis of previously measured analog ratios (Cline and Noyce 1985).

CONTROLS AND PERFORMANCE TESTING

Several approaches have been used to document the controls under which the Assayer has been operated from February 1989 through December 1990. These include the analysis of traceable standards, use of a secondary or control standard tied to the traceable-standards analyses, analysis of waste barrels for a nuclear power plant in which quantitative radiochemical analyses were subsequently performed on grab samples from the barrels (Robertson et al. 1989), and a cross-calibration experiment performed by a third party using the Barrel Assayer and PNL Safeguards and Security's SGS scanning system.

ANALYSIS OF TRACEABLE STANDARDS

Two sets of calibration barrels were manufactured to 1) document the performance of the gamma-ray spectrometer, 2) establish efficiency curves, and 3) document the response when a "hot spot" or discrete radioactive particle is contained within the drum.

The first set of barrels was made by homogeneously mixing and solidifying Dow Resin with known quantities of ^{60}Co , ^{137}Cs , and ^{134}Cs . The cobalt was obtained from Amersham, and the two cesium radioisotopes were obtained from New England Nuclear Corp. as certified solutions in sealed glass ampules. The concentrated solutions were reconstituted to 25 ml and the ampule was rinsed a minimum of 30 times with distilled water until all traces of radioisotope were removed from the glass ampule walls.

An aliquot of the reconstituted ^{137}Cs solution was counted on a gamma-ray spectrometer that was calibrated against USGS and/or NIST standards. The computed activity agreed with the stated activity.

PNL technicians were specially trained by Dow in the procedures for solidifying Dow Resin, and the approved Dow procedure was followed for solidifying the Dow Resin.

Table 1 presents the gamma-ray activity levels expected for the resin-filled calibration barrel and the levels determined most recently by a re-analysis using the Barrel Assayer. All values are well within the 25% stated accuracy of the Barrel Assayer and are conservative concentrations.

TABLE 1. Gamma-Ray Analysis Results from Dow-Resin Filled
Gamma-Ray Standard Drum (226.2 kg) Assay

<u>ID Number</u>	<u>Isotope</u>	<u>Expected Activity (uCi)</u>	<u>Observed Activity (uCi)</u>	<u>% Diff.</u>
50037-29-1	^{137}Cs	2411	2704	+12%
50037-29-2	^{134}Cs	236.2	276.1	+17%
50037-29-4	^{60}Co	863.9	925.4	+7.1%

Figure 2 illustrates the second type of calibration barrel, one which was used to gauge the effects of various densities of waste material (i.e., matrix effects) on counting efficiencies. A certified mixed-radionuclide gamma-ray reference solution was obtained from Amersham and sealed in a 2 cm diameter x 77 cm long thin-wall PVC tube. This standard was inserted into a series of PVC guide tubes placed inside an empty drum, as illustrated in Figure 2. The calibration tube was counted in each guide tube with the collimated IG detector as the drum rotated. Counts were performed for equal time intervals on equally-spaced segments along the length of the drum.

Efficiency curves were generated by filling the drum with materials of different density, as illustrated in Figure 3. Counts from each guide-tube position were then averaged to obtain the counting-efficiency curves for a "homogeneously" distributed source. Figure 4 shows the resulting counting-efficiency curves for a homogeneous sample. A series of interpolated curves are shown to provide the necessary counting efficiency for any weight of drum up to 318 kg.

A mixed-radionuclide calibration standard was used to simulate the presence of a "hot spot" or discrete radioactive particle in the drum. The source, serving essentially as a point, was counted at a number of positions inside the 208-l drum and the response compared with the 77-cm long standard for which the drum had been extensively calibrated. For relatively low sample densities, the point-source, mixed-radionuclide calibration standard produced counting-efficiency curves that were within $\pm 10\%$ of those provided by the 77-cm long standard. The approximate vertical locations of "hot spots" were

FIGURE 2. Geometry of 208-Liter Drum Used For Calibration With a Mixed-Isotope Source as a Function of Source Geometry and Drum Weight

FIGURE 3. 208-Liter Drum Observed Efficiency Curves as a Function of Source Geometry and Drum Weight

FIGURE 4. 208-Liter Drum Calibration Curves (Observed and Extrapolated) Used to Generate Appropriate Efficiency Curve for Given Barrel Weight

identified within the drum by examining the spectra for individual segments along the drum.

To assess the reliability of the neutron detection system, several sets of standards were analyzed. For example, one set of standards is the NDA Safeguards Standards, consisting of two cans containing 7 liters of Absorb-It, thoroughly mixed with known quantities of 88.77% ^{239}Pu nitrate stock solution, purified of ^{241}Am (analyzed by HEDL as sample #Q7543). These standards are placed alone or together at various positions within an empty drum. Results of such analyses are presented in Table 2.

CONTROL STANDARD

A thorium nitrate standard (52733-38-1) was prepared and counted. This standard served as the secondary or control standard, with the resin-filled barrels serving as primary standards. Tables 3 and 4 and Figures 5 and 6 present values for the peak areas of selected thorium-standard gamma-rays obtained from the commercial ORTEC MCA software when applied to the control standard. The full-width at half-maximum (FWHM) values are merely for monitoring the energy-resolving characteristics of the IG detector and not for control purposes.

The control is typically run a second time if one of the values exceeds the 8% control limit. The system is out of control if the second run does not show the value to be in control. In the figures, a 4% limit is illustrated as a dotted line. The 8% limit is represented by the solid lines above and below the 4% limit lines.

GRAB SAMPLE ANALYSIS

Tables 5 and 6 contain values obtained by assaying 23 waste barrels at several commercial nuclear power plants. The barrels contained ion-exchange resins or oil. After being counted by the Barrel Assayer, aliquots were removed from the top, middle, and bottom of the barrels for radiochemical analysis.

Analysis of the segmented scans obtained from the Barrel Assayer used on oil-filled drums indicates that the activity is homogeneously distributed throughout the drum. The activity in the drum is likely to be present as

TABLE 2. Neutron Analysis Results from ^{239}Pu Standard
Canisters Centered in 55-Gallon Drum

<u>ID Number & Position Within Drum</u>	<u>Net Barrel Weight (g)</u>	<u>Expected ^{239}Pu Weight (g)</u>	<u>Observed ^{239}Pu Weight (g) (Neutron)</u>	<u>Maximum ^{239}Pu Weight (g) (Neutron)</u>
#3 std (Centered)	14515	30.3	30.5 (+0.7%) +/- 4.2	<37.1
#4 std (Centered)	16103	10.2	10.3 (+1.0%) +/- 1.5	<12.6
#3 & #4 (Centered)	21773	40.5	37.6 (-7.2%) +/- 5.3	<46.0
#3 std (Outer Middle)	4082.4	30.3	28.54 (- 5.8%) +/- 4.07	<35
#3 std (Outer Bottom)	4082.4	30.3	27.26 (-10.0%) +/- 3.88	<35
#3 & #4 (Outer Middle)	8618.4	40.5	38.01 (- 6.1%) +/- 5.42	<46.5

discrete particulates suspended in a uniform oil matrix, however, and a good deal of variability is expected in small grab samples. Even greater variability is expected in ion-exchange resin. It should also be noted that sampling only three regions in any one drum introduces significant error in the analysis.

The ratio of grab-sample concentration to direct-assay concentration presented in Table 5 shows acceptable agreement between the two measurement methods.

TABLE 3. Computer Results From CTL Program Used to Monitor Detector Reliability Based on Observed Peak Area and Resolution (February 23, 1989 - July 23, 1990)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
23-FEB-89	6.3124E+04	2.560	1.8721E+04	2.840	1.0534E+04	3.900
23-FEB-89	6.2568E+04	2.580	1.8512E+04	2.860	1.0561E+04	3.870
23-FEB-89	6.2552E+04	2.590	1.8632E+04	2.820	1.0532E+04	3.710
23-FEB-89	6.2297E+04	2.550	1.8710E+04	2.810	1.0399E+04	3.710
23-FEB-89	6.3335E+04	2.560	1.8738E+04	2.830	1.0583E+04	3.690
24-FEB-89	6.3280E+04	2.540	1.8258E+04	2.810	1.0586E+04	3.860
24-FEB-89	6.2274E+04	2.530	1.8409E+04	2.750	1.0574E+04	3.720
24-FEB-89	6.2522E+04	2.530	1.8547E+04	2.720	1.0555E+04	3.870
24-FEB-89	6.2891E+04	2.370	1.8485E+04	2.850	1.0554E+04	3.720
24-FEB-89	6.2285E+04	2.550	1.8768E+04	2.820	1.0553E+04	3.740
15-MAR-89	6.2310E+04	2.260	1.8447E+04	2.510	1.0534E+04	3.520
21-MAR-89	6.2714E+04	2.500	1.8964E+04	2.780	1.0491E+04	3.820
28-MAR-89	6.2070E+04	2.670	1.9221E+04	2.960	1.0592E+04	3.830
28-MAR-89	6.2146E+04	2.630	1.8461E+04	2.840	1.0556E+04	3.890
11-JUL-89	6.2855E+04	3.260	1.8893E+04	3.280	1.0280E+04	4.170
12-JUL-89	6.3198E+04	2.740	1.8838E+04	2.880	1.0441E+04	3.880
13-JUL-89	6.3863E+04	2.550	2.0326E+04	2.870	1.0648E+04	3.840
17-JUL-89	6.3386E+04	2.540	1.8473E+04	2.790	1.0568E+04	3.910
18-JUL-89	6.3317E+04	2.490	1.8653E+04	1.810	1.0654E+04	3.680
19-JUL-89	6.2214E+04	2.570	1.8505E+04	2.890	1.0514E+04	3.900
31-JUL-89	6.2668E+04	2.160	1.8588E+04	2.470	1.0789E+04	3.680
17-AUG-89	6.3826E+04	2.430	1.9861E+04	2.380	1.0668E+04	3.960

TABLE 3. (Continued)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
22-AUG-89	6.3034E+04	2.360	1.8753E+04	2.810	1.0431E+04	4.060
22-SEP-89	6.2914E+04	2.590	1.8385E+04	2.930	1.0638E+04	3.970
25-SEP-89	6.0161E+04	3.240	1.8740E+04	3.310	1.0381E+04	4.180
27-SEP-89	6.2598E+04	2.770	1.7816E+04	3.010	1.0588E+04	4.160
2-OCT-89	6.1674E+04	2.430	1.7864E+04	2.730	1.0405E+04	3.750
3-OCT-89	6.3549E+04	2.370	1.8767E+04	2.720	1.0606E+04	3.710
19-OCT-89	6.2767E+04	2.210	1.9123E+04	2.450	1.0788E+04	3.630
24-OCT-89	6.2999E+04	2.150	N/A	N/A	1.0444E+04	3.480
31-OCT-89	6.2917E+04	2.250	1.7859E+04	2.510	1.0563E+04	3.610
2-NOV-89	6.2217E+04	2.190	1.8735E+04	2.560	1.0632E+04	3.630
8-NOV-89	6.4419E+04	2.430	1.8588E+04	2.680	1.0659E+04	3.650
9-NOV-89	6.3120E+04	2.560	1.8986E+04	2.890	1.0618E+04	3.940
27-DEC-89	6.4890E+04	2.190	N/A	N/A	1.0670E+04	3.810
28-DEC-89	6.3566E+04	2.180	1.9014E+04	2.360	1.0738E+04	3.400
5-JAN-89	6.4113E+04	2.320	1.9141E+04	2.620	1.0672E+04	3.610
10-JAN-90	6.3513E+04	2.280	1.8684E+04	2.600	1.0573E+04	3.630
1-FEB-90	6.5527E+04	2.270	1.9380E+04	2.620	1.0745E+04	3.640
11-APR-90	6.2430E+04	2.470	N/A	N/A	1.0543E+04	3.830
20-APR-90	6.2790E+04	2.660	1.8543E+04	2.890	1.0158E+04	3.970
1-MAY-90	6.3503E+04	2.550	1.8583E+04	2.840	1.0660E+04	3.840
8-MAY-90	6.3100E+04	2.610	1.8304E+04	2.920	1.0575E+04	3.890
11-MAY-90	6.3E356+04	2.380	1.8070E+04	2.760	1.0282E+04	3.800
17-MAY-90	6.3107E+04	2.750	1.8162E+04	2.920	1.0332E+04	4.120

TABLE 3. (Continued)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
25-MAY-90	6.2773E+04	2.450	1.8435E+04	2.740	1.0287E+04	3.920
31-MAY-90	6.2542E+04	2.430	1.8836E+04	2.020	1.0339E+04	3.680
6-JUN-90	6.4431E+04	2.540	1.8525E+04	2.810	1.0335E+04	3.920
13-JUN-90	6.1172E+04	2.400	1.8775E+04	2.700	1.0602E+04	3.770
19-JUN-90	6.3375E+04	2.890	1.9133E+04	2.900	1.0529E+04	4.190
34-JUN-90	6.2710E+04	2.990	1.8610E+04	3.090	1.0202E+04	4.080
28-JUN-90	6.2607E+04	2.590	1.8790E+04	3.890	1.0204E+04	4.400
5-JUL-90	6.1831E+04	2.420	1.8832E+04	2.710	1.0052E+04	3.720
9-JUL-90	6.2408E+04	2.780	1.8615E+04	2.840	1.0334E+04	3.750
12-JUL-90	6.4354E+04	3.040	1.8781E+04	3.170	1.0581E+04	4.090
13-JUL-90	6.3732E+04	2.720	1.6847E+04	2.000	1.0676E+04	3.980
16-JUL-90	6.2230E+04	2.710	1.8770E+04	2.860	1.0542E+04	3.070
19-JUL-90	6.3270E+04	3.020	1.9057E+04	3.150	1.0765E+04	4.020
23-JUL-90	6.2252E+04	2.620	1.9079E+04	2.930	1.0670E+04	3.830

Average	6.2713E+04	2.536	1.8578E+04	2.811	1.0570E+04	3.779

TABLE 4. Computer Results From CTL Program Used to Monitor Detector Reliability Based on Observed Peak Area and Resolution
(February 2, 1990 - December 14, 1990)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
2-FEB-90	6.4560E+04	2.250	1.8907E+04	2.420	1.0766E+04	3.520
5-FEB-90	6.4456E+04	2.330	1.9426E+04	2.550	1.0563E+04	3.660
29-MAR-90	6.2415E+04	2.170	N/A	N/A	1.0534E+04	3.430
30-MAR-90	6.3097E+04	2.230	1.8636E+04	2.610	1.0641E+04	3.690
5-APR-90	6.3464E+04	2.400	1.8641E+04	2.630	1.0611E+04	3.840
9-APR-90	6.2923E+04	2.640	1.8495E+04	2.730	1.0505E+04	3.760
11-APR-90	6.2438E+04	2.470	N/A	N/A	1.0543E+04	3.830
20-APR-90	6.2790E+04	2.660	1.8543E+04	2.890	1.0158E+04	3.970
1-MAY-98	6.3503E+04	2.550	1.8503E+04	2.840	1.0660E+04	3.840
8-MAY-98	6.3180E+04	2.618	1.8304E+04	2.928	1.0575E+04	3.850
11-MAY-90	6.3835E+04	2.380	1.8070E+04	2.760	1.0282E+04	3.800
17-MAY-98	6.2107E+04	2.750	1.8162E+04	2.920	1.0332E+04	4.120
25-MAY-90	6.2779E+04	2.450	1.8435E+04	2.770	1.0397E+04	3.930
31-MAY-90	6.2542E+04	2.430	1.8836E+04	2.800	1.0589E+04	3.680
6-JUN-90	6.4451E+04	2.540	1.8525E+04	2.810	1.0555E+04	3.920
13-JUN-90	6.1172E+04	2.408	1.8775E+04	2.780	1.0602E+04	3.770
19-JUN-98	6.3375E+04	2.890	1.9133E+04	2.900	1.0529E+04	4.190
24-JUN-90	6.2710E+04	2.990	1.8618E+04	3.090	1.0502E+04	4.080
28-JUN-90	6.2607E+04	2.590	1.8750E+04	3.090	1.0284E+04	4.400
5-JUL-90	6.1031E+04	2.420	1.8832E+04	2.710	1.0607E+04	3.720
9-JUL-90	6.2408E+04	2.700	1.8615E+04	2.840	1.0334E+04	3.750
12-JUL-90	6.4354E+04	3.040	1.8701E+04	3.170	1.0581E+04	4.090

TABLE 4. (Continued)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
13-JUL-90	6.3732E+04	2.720	1.8847E+04	2.880	1.0676E+04	3.900
16-JUL-90	6.2238E+04	2.710	1.8770E+04	2.850	1.0542E+04	3.640
19-JUL-90	6.3270E+04	3.020	1.9027E+04	3.150	1.0765E+04	4.090
23-JUL-90	6.2252E+04	2.620	1.9079E+04	2.930	1.0649E+04	3.830
27-JUL-90	6.3129E+04	2.420	1.8773E+04	2.770	1.0801E+04	3.560
2-AUG-90	6.4262E+04	2.490	1.8777E+04	2.740	1.0545E+04	3.720
27-AUG-90	6.2479E+04	3.070	1.8576E+04	3.280	1.0582E+04	4.450
30-AUG-90	5.9445E+04	2.700	1.8619E+04	2.930	1.0573E+04	3.960
31-AUG-90	6.3334E+04	2.850	1.8791E+04	2.920	1.0690E+04	3.860
4-SEP-90	6.2736E+04	3.040	1.9153E+04	3.190	1.0558E+04	3.980
5-SEP-90	6.1939E+04	3.020	1.8759E+04	3.110	1.0574E+04	4.060
12-SEP-90	6.3417E+04	2.760	1.8844E+04	3.080	1.0518E+04	3.940
19-SEP-90	6.3757E+04	2.770	1.8565E+04	2.890	1.0515E+04	3.860
21-SEP-90	6.2303E+04	2.640	1.8979E+04	2.970	1.0652E+04	3.910
25-SEP-90	6.3170E+04	3.030	1.7862E+04	3.170	1.0613E+04	4.100
26-SEP-90	6.2179E+04	2.960	1.9021E+04	3.860	1.0802E+04	3.940
27-SEP-90	6.4044E+04	2.940	1.9293E+04	3.120	1.0702E+04	3.930
3-OCT-90	6.3554E+04	2.820	1.9161E+04	2.880	1.0726E+04	3.840
4-OCT-90	6.3386E+04	2.860	1.9030E+04	2.940	1.0669E+04	3.890
17-OCT-90	6.2903E+04	2.200	1.8402E+04	2.550	1.0754E+04	3.500
29-OCT-90	6.1816E+04	2.860	1.8540E+04	3.010	1.0728E+04	3.980

TABLE 4. (Continued)

DATE	Decay Corrected C/M					
	TH (238)	FWHM 238	TH (911)	FWHM 911	TH(2615)	FWHM2615
14-DEC-90	6.0324E+04	2.820	1.8796E+04	2.980	1.0419E+04	3.810
14-DEC-90	6.2074E+04	2.400	1.8496E+04	2.720	1.0515E+04	3.620
Average	6.2713E+04	2.536	1.8578E+04	2.811	1.054E+04	3.779

FIGURE 5. Computer Output from CTL Program Demonstrating that the Detector Was Within Calibration (February 23, 1989 - July 23, 1990)

FIGURE 6. Computer Output from CTL Program Demonstrating that the Detector Was Within Calibration (February 2, 1990 - December 14, 1990)

TABLE 5. Comparison Of The Total Gamma-Ray Activity in Oil-Filled Drums By Grab Sampling Versus Direct Assay

Plant ID	Drum No.	Concentration (pCi/g)		
		Range or Ave. of Grab Sampling	Direct Assay	Ratio of Grab/Direct
L	10	(0.9-1.4)	0.60	+1.8
L	15	(0.5-4.2)	0.28	+6.8
L	29	(1.2-6.3)	0.47	+7.4
L	30	(6.1-7.0)	17.0	-2.7
L	31	(8.3-10.9)	1.6	+6
L	32	(16-143)	20	+2.6
L	203	0.57	4.2	-7.4
L	235	40	73	-1.8
L	236	5.4	13	-2.4
L	237	5	74	-15
Average for Plant L		12.4	20.4	-1.6

TABLE 6. Comparison of the Total Gamma-Ray Activity in Ion-Exchange-Resin Filled Drums By Grab Sampling Versus Direct Assay

Plant ID	Drum No.	Concentration (pCi/g)		
		Grab Sampling	Direct Assay	Ratio of Grab/Direct
B	R-58	5.2	9.2	-1.8
B	R-59	36	11	+3.3
B	R-60	19	5.1	+3.7
B	R-61	2.1	15.	-7.1
B	R-62	4.5	7.6	-1.7
B	R-63	4.9	8.5	-1.7
B	R-64	4.6	7.6	-1.7
B	R-65	0.81	7.2	-8.9
B	R-66	1.4	5.1	-3.6
B	R-67	1.5	3.9	-2.6
B	R-68	24	1.7	+14.1
B	R-69	0.68	3.6	-5.3
B	R-70	28.	8.2	+3.4
Average for Plant B		10.2	7.76	+1.3

CROSS-CALIBRATION EXPERIMENT

In Table 7, results using Safeguards & Security's SGS scanning system are compared with those using the PNL Barrel Assayer^(a). Safeguards & Security's SGS system uses segment-by-segment transmission corrected data to account for different matrix densities. Essentially all drums contained TRU wastes. Drums which were assayed without replication are listed first. The contents of some drums were rearranged and analyzed twice (such as Drum 2 and 2A, Drum 83 and 83A, Drum 85 and 88, and Drum 86 and 89) which provides information on variability due to geometry. The contents of several of the drums were assayed as pieces in addition to being assayed as a whole; the various combinations are noted in Table 7. These data provided information for a material balance comparison.

Several results were available from the PNL Barrel Assayer besides the "best" value, such as low, median, and high values as determined by the gamma-ray and the neutron analysis. A ratio was determined based on replicate analyses to gauge how well results from the analysis of a whole compared to the analyses of the parts. The results of the analysis are presented in Table 8. An additional more detailed breakdown is given in Table 9. Except for the singly measured drums 17A and 91, the agreement is generally excellent.

The results of the "low value determined by neutron analysis" under heading 4 of Table 8 agree the best with the results of the Safeguards and Security SGS scanning system and show the smallest standard deviation. Such an outcome is consistent with the highly penetrating nature of neutrons and the strong dependence on matrix of gamma-ray detection. These data also emphasize one of the strong features of the PNL Barrel Assayer design: for TRU waste the neutron analysis provides an excellent absolute measure of TRU content while the gamma-ray analysis provides an excellent measure of the relative ratios of the associated isotopes.

(a) H.H. Van Tuy1, "Update on CML Waste Drums." Memo to W.D. Richmond dated December 2, 1989, (Project Number HHV33/6).

TABLE 7. Comparison of Analytical Data on Critical-Mass-Laboratory Waste Drums

Drum	Safeguards & Security (g Pu)	Barrel Assayer (g Pu)		
	Gamma	Gamma	Neutron	"Best"
13	44.7	30.9 - 79.3	34.2 - 36.4	36.4
17A	204	>71.3	52.2 - 256	72
20		1.39	0.842	1.39
21			<0.0034	<0.0034
52	61.0	63.1 - 187	57.5	63.1
69	<0.002		0.00531	0.00531
79	50.1	42.1 - 59.4	47.1 - 47.3	47.2
80	78.3	58.4 - 75.8	63.4 - 63.5	63.5
81	49.4	30.2 - 30.3	25.2	30.2
82	74.0	57.4 - 117	33.6 - 35.8	57.4
90	2.6	>1.26	<0.0086	1.3
91	3.6	>1.94	0.527	1.9
93	14.9	>14.3	14.3 - 40.8	15
Drum 2 = Drum 2A				
2	34.2	35.9 - 111	31.1 - 81.7	36.8
2A	28.0	48.6	31.7 - 76.8	49
Drum 67 = Drums 86 + 83 + 2 + 85				
67	127.3	251 - 1790	252 - 254	254
86	34.2	42.9 - 90.3	37.9 - 107.7	42.9
83	65.2	48.8 - 125.8	44.7 - 127.0	68.8
2	34.2	35.9 - 111	31.1 - 81.7	36.8
85	273	229 - 993	267 - 709	267
Drum 70 = Drums 70A + 62A + 83B				
70	185.8	27.9 - 943.4	93.0 - 123	106
70A	35.3	49.9	34.3 - 96.7	50
62A	34.8	>46.4	24.6 - 72.3	47
83B	62.1	121	62.8 - 202	120
Drum 72 = Drums 84 + 72A				
72	215.3	147 - 829	200 - 202	202
84	94.8	140 - 756	99.8 - 295	140
72A	145.6	189 - 898	137 - 447	189

TABLE 7. (Continued)

Drum	Safeguards & Security (g Pu)	Barrel Assayer (g Pu)		
	Gamma	Gamma	Neutron	"Best"
Drum 83 = Drum 83A				
83	65.2	48.8 - 125.8	44.7 - 127.0	68.8
83A	52.7	110	54.3 - 138	110
Drum 85 = Drum 88				
85	273	229 - 993	267 - 709	267
88	272	162 - 370	253 - 649	253
Drum 86 = Drum 89				
86	34.2	42.9 - 90.3	37.9 - 107.7	42.9
89	39.6	29.4 - 111	36.2 - 96.5	59
Drum 86B = Drums 86A + 89 + 83A				
86B	95.9	187	93.5 - 251	190
86A	3.3	5.37	5.20	5.3
89	39.6	29.4 - 111	36.2 - 96.5	59
83A	52.7	110	54.3 - 138	110
Drum 87A = Drums 87 + 62 + 2A				
87A	109.1	>159	103 - 336	160
87	<0.09	1.08- 1.59	1.04- 3.72	1
62	53.1	112 - 240	59.6 - 59.9	112
2A	28.0	48.6	31.7 - 76.8	49
Drum 88 = Drums 55A + 89A				
88	272	162 - 370	253 - 649	253
55A	115	104 - 534	135 - 381	220
89A	111	>160	105 - 293	160
Drum 90A = Drums 55 + 92				
90A	104.3	206	97.8 - 310	210
55	72.9	51.3 - 144	81.6 - 82.1	81.9
92	9.0	8.57	5.73- 16.4	8.6

TABLE 8. Material Balance Ratios*

Drum	Safeguards & Security	Barrel Assayer						
		1	2	3	4	5	6	7
70	1.41	7.70	2.23	4.34	1.31	2.28	3.02	2.05
72	1.12	2.24	2.03	2.00	1.18	2.43	3.67	1.63
67		1.42	1.22	1.36	1.51	1.78	4.04	1.64
86	1.16	1.46	1.05	1.23	1.05	1.10	1.12	1.38
83	1.24	2.25	1.26	1.14	1.21	1.12	1.09	1.60
2	1.22	1.35	1.51	2.28	1.02	1.04	1.06	1.33
85	1.00	1.41	2.30	2.68	1.06	1.08	1.09	1.06
88	1.20	1.63	1.80	1.88	1.05	1.01	1.04	1.50
868	1.00	1.29	1.01	1.21	1.02	1.03	1.05	1.09
87A	1.35	1.02	1.42	1.83	1.12	1.89	2.39	1.01
90A	1.23	3.37	1.92	2.34	1.12	2.19	3.15	2.29
Avg.	1.19	2.29	1.61	1.94	1.15	1.63	2.07	1.51
Std. Dev.	0.13	1.94	0.47	0.94	0.15	0.69	1.21	0.40

* Ratio required to bring replicate analyses of parts vs. whole into agreement

1. low value determined by gamma-ray analysis
2. median value determined by gamma-ray analysis
3. high value determined by gamma-ray analysis
4. low value determined by neutron analysis
5. median value determined by neutron analysis
6. high value determined by neutron analysis
7. "best" value.

**TABLE 9. Ratio Comparison of Analytical Data on
Critical-Mass-Laboratory Waste Drums**

<u>Drum</u>	<u>Safeguards & Security Gamma(g Pu)</u>	<u>Barrell Assayer "Best" (g Pu)</u>	<u>Ratio Best/S&S</u>	<u>Barrell Assayer Neutron (g Pu)</u>	<u>Ratio Neutron/S&S</u>
2	34.2	36.8	+1.08	31.1	-1.10
2A	28.0	49.0	+1.75	31.7	+1.13
13	44.7	36.4	-1.23	34.2	-1.31
17A	204.0	72.0	-2.83	52.2	-3.91
52	61.0	63.1	+1.03	57.5	-1.06
55	72.9	81.9	+1.12	81.6	+1.12
55A	115.0	220.0	+1.91	135.0	+1.17
62	53.1	112.0	+2.11	59.6	+1.12
62A	34.8	47.0	+1.35	24.6	-1.41
67	127.3	254.0	+2.00	252.0	+1.98
70	185.8	106.0	-1.75	93.0	-2.00
70A	35.3	50.0	+1.42	34.3	-1.03
72	215.3	202.0	-1.07	200.0	-1.08
72A	145.6	189.0	+1.30	137.0	-1.06
79	50.1	47.2	-1.06	47.1	-1.06
81	49.4	30.2	-1.64	25.2	-1.96
82	74.0	57.4	-1.29	33.6	-2.20
83	65.2	68.8	+1.06	44.7	-1.46
83A	52.7	110.0	+2.09	54.3	+1.03
83B	62.1	120.0	+1.93	62.8	+1.01
84	94.8	140.0	+1.48	99.8	+1.05
85	273.0	267.0	-1.02	267.0	-1.02
86	34.2	42.9	+1.25	37.9	+1.11
86A	3.3	5.3	+1.61	5.2	+1.58
86B	95.9	190.0	+1.98	93.5	-1.03
87A	109.1	160.0	+1.47	103.0	-1.06
88	272.0	253.0	-1.08	253.0	-1.08
89	39.6	59.0	+1.49	36.2	-1.09
89A	111.0	160.0	+1.44	105.0	-1.06
90	2.6	1.3	-2.00	<.009	
90A	104.3	210.0	+2.01	97.8	-1.07
91	3.6	1.9	-1.89	0.53	-6.83
92	9.0	8.6	-1.05	5.7	-1.57
93	14.9	15.0	+1.01	14.3	-1.04
	<hr/> 2877.8	<hr/> 3466.8	<hr/> +1.20	<hr/> 2610.5	<hr/> -1.10

CONCLUSION

Several approaches have been presented to validate the operation of the PNL Barrel Assayer on both TRU and non-TRU waste drums.

One test describes the re-analysis of resin-filled calibration barrels containing known quantities of ^{60}Co , ^{137}Cs , and ^{134}Cs . The observed activity levels matched the expected levels, while an analysis of the data generated from a thorium nitrate secondary standard verified that the detector was within calibration for the period from February 1989 through December 1990.

Data were also presented describing the assay of 23 non-TRU waste drums at two separate commercial nuclear power plants. Comparison of grab samples from oil and ion-exchange-resin filled drums validate the operation of the Barrel Assayer on non-TRU waste drums.

The Barrel Assayer was also employed to analyze several barrels containing TRU wastes. The results are compared to results obtained with an alternative technology and validate the operation of the PNL Barrel Assayer on TRU waste drums.

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