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In-Plant Experience with Automated Gamma-Ray Spectroscopy Systems for Plutonium Isotopic Composition Measurements

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by

T. E. Sampson, E. L. Sandford, D. F. Bowersox, S. S. Johnson,
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ABSTRACT

Two gamma-ray spectroscopy systems for the nondestructive measurement of the isotopic composition of plutonium are described. One system is at the Savannah River Plant and the other is at the TA-55 Plutonium Facility at Los Alamos National Laboratory. Results from a year's operation of the Los Alamos system are presented.

I. INTRODUCTION

At Los Alamos National Laboratory, techniques for the measurement of plutonium isotopic composition using gamma-ray spectroscopy techniques were proposed by Parker and Reilly in 1974.[1] Only in recent years were these techniques put into routine use at Los Alamos,[2,3] although they are widely used in other laboratories. References 2 and 3 list references to other isotopic measurement methods. The analysis methods used at Los Alamos are described in Refs. 2 and 3 and are repeated in Appendix A for completeness. The software is described in Appendix B.

This report describes how the methods of Refs. 2 and 3 were implemented in systems suitable for routine in-plant use. The two systems described are (1) a single-detector system installed at the Savannah River Plant (SRP) and (2) a multiple-detector system installed at the Los Alamos TA-55 Plutonium Facility. Results from a year's use at TA-55 are presented.

II. CHRONOLOGY

The work described in Refs. 2 and 3 was started in 1979 with an experimental system installed at the Los Alamos TA-55 Plutonium Facility late that year. Its operation was so successful that process measurements took over the system for the next 2 years.

Early in 1981, we started working on a system to be delivered to the JB-line at SRP for a test and evaluation program. Some improvements were made in the physics analysis originally implemented in the experimental Los Alamos system. The SRP system was completed at the end of May 1981 but was not shipped to SRP until August 1981. Installation took place in October 1981. Figure 1 shows the system before shipment to SRP. To date, SRP has not started up the system because of numerous problems with the building air conditioning and electrical utilities in the installation area. The system was removed from the air-conditioned cabinet shown in Fig. 1 and installed in a conventional electronics rack because of the planned use of room air conditioning. We can present calibration results for this system but cannot show any results from process samples.

After the initial success of the experimental system at TA-55, a new system was built for permanent installation in the Los Alamos Plutonium Facility. This system has two detectors (expandable

Fig. 1. SRP plutonium isotopics system in air-conditioned cabinet.

to four detectors) that can measure two samples simultaneously. This project culminated in the installation of the multiple-detector plutonium isotopic system (MUDPI) at TA-55 in April 1982 (Fig. 2). We present calibration results from this system and isotopic results from several batches of material measured during the past year.

The TA-55 system has the same physics analysis as the SRP system. The software was modified to allow acquisition and analysis from up to four detectors. Some additional features allow easier use of an auxiliary plutonium isotopic decay correction program for application to calorimetry measurements.

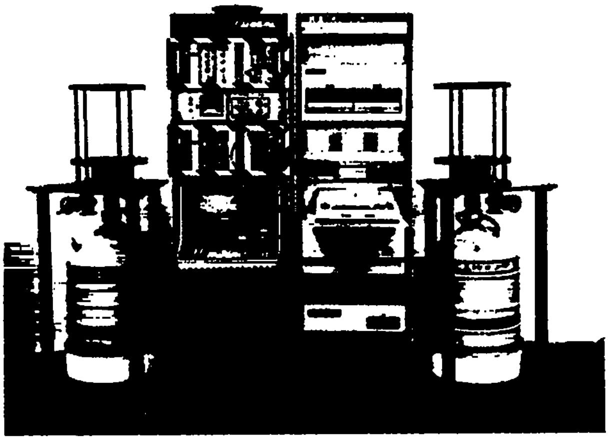

Fig. 2. Multiple-detector plutonium isotopics system for the Los Alamos TA-55 Plutonium Facility.

III. SYSTEM CONFIGURATIONS

The SRP and TA-55 systems are configured similarly with one major difference: the TA-55 system can operate up to four independent detectors simultaneously, whereas the SRP system is a single-detector unit. Major system components are listed in Table I, hardware interconnections are shown in Figs. 3 and 4, and the detector shield, filters, and sample platform for the TA-55 system are shown in Figs. 5 and 6.

Even though the detectors are low-energy photon spectrometers (LEPS), we specified that they be delivered without beryllium windows in order to be more rugged. This does not affect performance because all measured gamma rays are above 100 keV. The detector end cap can be seen in the center of the shield in Fig. 5.

Filters, shown lying on the table in Fig. 5, are placed in a holder that slides into the shield just above the end cap of the detector. We use 75 mils (1.9 mm) of cadmium and 10 mils (0.25 mm) of copper for all samples. These filters can be easily changed, if desired.

Samples are placed on a Lucite platform above the detector (Fig. 6). A hand-crank mechanism, visible in Figs. 5 and 6, moves the platform up and down to adjust the gross count rate to an acceptable value. Operators are instructed to position large samples so that the gross count rate, monitored with a recycling counter/timer in the electronics rack, is 15 kHz. Small samples can be set directly on the filters above the end cap to maximize count rate.

The system for SRP is similar to that shown in Figs. 3-6. The sample platform is moved manually, being held in position by thumbscrews.

IV. CALIBRATION

A. General Procedure

Methods using relative efficiency curves generated from the spectrum under study do not, in principle, require any calibration.[1-3] This

TABLE I

MAJOR SYSTEM COMPONENTS

Component	SRP	TA-55
Detector	Ortec Planar 16 mm diam by 10 mm deep	2 - Ortec Planar 16 mm diam by 10 mm deep
Resolution	<495 eV at 122 keV	<498 eV at 122 keV
Count rate	>4000 MeV/s	Exceeds 40 kHz for filtered ^{57}Co
NIM bin	Ortec 401A/402D	2 - Ortec 401A/402D
Bias supply	Ortec 459	2 - Ortec 459
Amplifier	Ortec 572	2 - Ortec 572
Count-rate monitor	Ortec 776	2 - Canberra 1772
Oscilloscope	Tektronix T922R	Tektronix T922R
MCA	Canberra Series 80 8-k channel	Canberra Series 80 16-k channel
Digital stabilizer	2 - Canberra 8200	4 - Canberra 8200
ADC	Canberra 8621	2 - Canberra 8621
Expansion chassis		Canberra 8683A
Computer	DEC PDP-11/03	DEC PDP-11/23
Mass storage	DEC RX02	DEC RX02 DEC RL01
Terminal	DEC LA120	Texas Instruments, TI-743
Operating system	RT-11 Version 3B	RT-11 Version 3B
Rack	DEC H960	2 - DEC H960
Power controller	DEC 861C	2 - DEC 861C
Line filter and regulator	Topaz Line 2, Model 70306	2 - Topaz Line 2, Model 70306

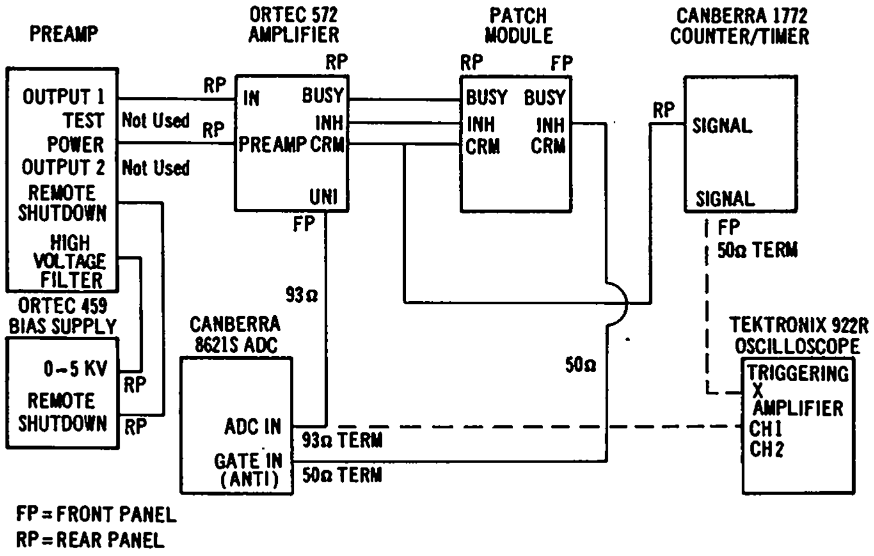

Fig. 3. Block diagram of counting electronics for one detector of the TA-55 system.

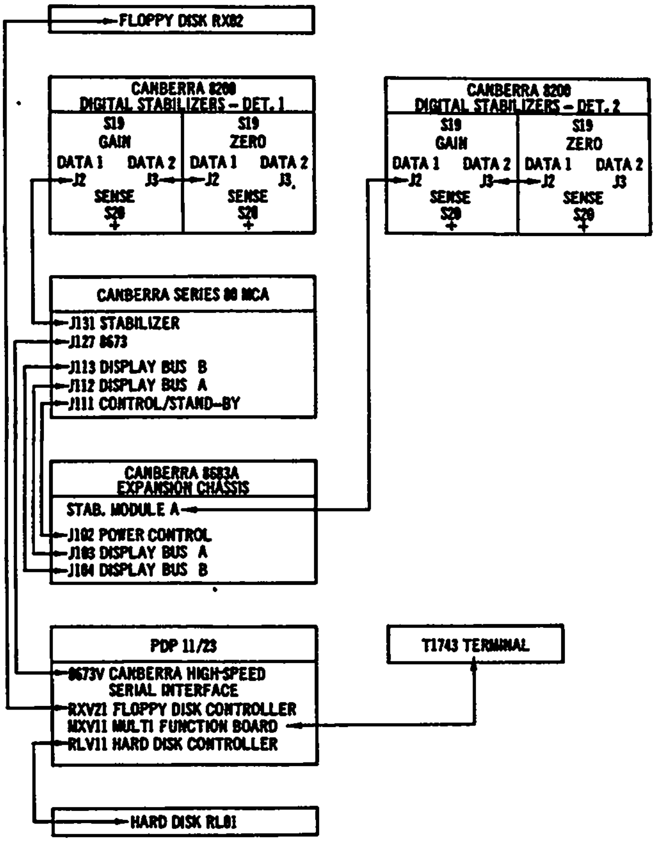

Fig. 4. Computer-MCA interconnections for the TA-55 multiple detector system.

Fig. 5. Top of detector stand for the TA-55 system; filters and filter holder have been removed from detector shield.

assumes, however, that branching ratios and half-lives are bias free and that all peak areas can be determined in an error-free manner. In practice, these assumptions do not always hold. Although half-life[4] uncertainties do not contribute significantly to the errors, there are some recognized errors[5] in the best branching-ratio data available.[6] The simple, region-of-interest (ROI) peak area determination used in the Los Alamos instruments can give biased peak areas for poorly resolved peaks. Additional biases can result from the exact methods used to interpolate and extrapolate for relative efficiency. These effects can cause biases of, typically, 1 to 5% in the measured isotopic ratios.

We greatly reduce these biases by calibrating the system using measurements on samples with known isotopic composition. To date, we have used a wide range of sample masses and isotopic content to produce a wide-range, nearly universal, system suitable for measurements on any sample. The general success of this approach is demonstrated by the results that follow in this report. Areas for improvement also are apparent.

In Tables II and III, we display the FORTRAN statements used to compute isotopic ratios for both equilibrium (or aged) samples and nonequilibrium (freshly separated) samples. Variables are

A(IXXX)	Net peak area of peak with energy XXX keV. May include interferences.
CA(IXXX)	Net peak area of ^{237}U component of a peak with both ^{237}U and ^{241}Am contributions.
REFF(IXXX)	Relative efficiency at energy XXX keV.
FF XXYY	Calibration constant for equilibrium isotopic ratio of isotope XX to YY. May be dimensioned.
NE XXYY	Calibration constant for nonequilibrium isotopic ratio.

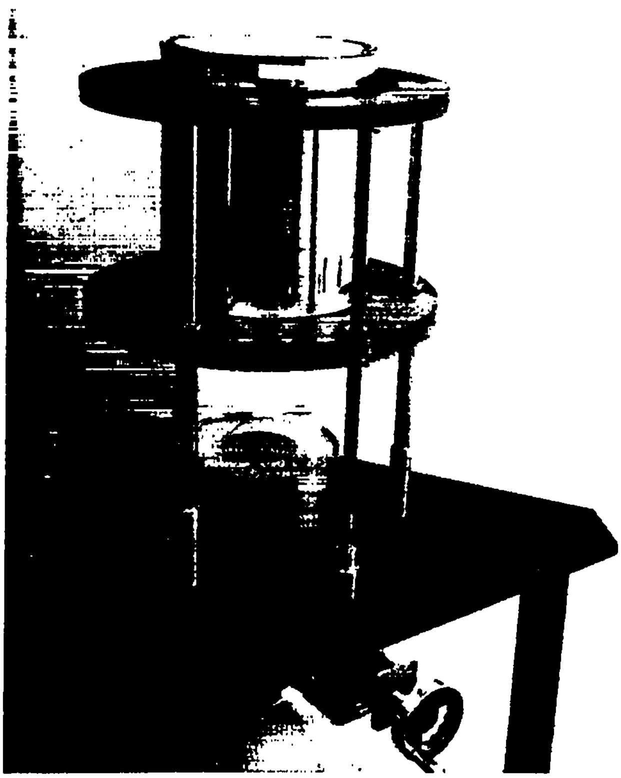

Fig. 6. Sample in position on detector stand for the TA-55 system.

FRACT(N)	Isotopic fraction of isotope N. ^{238}Pu, N = 1; ^{239}Pu, N = 2; ... .
AMPURA	Ratio of mass ^{241}Am to mass total plutonium.

The numerical constants in the formulas arise from combining the branching ratios and half-lives as outlined in Appendix A. Slight differences between constants in like expressions in Tables II and III arise from the use of different half-lives when the calculations were done. They are not significant.

We see in Tables II and III that six of the ratio expressions have only a single term. Two expressions, ^{241}Am/^{239}Pu at 125.3/129.3 keV, have two terms arising from the subtraction of a ^{239}Pu interference. The expressions for ^{240}Pu/^{241}Pu have three terms arising from subtraction of two interferences, ^{239}Pu and ^{241}Pu.

Calibration proceeds by measuring a wide variety of samples with either multiple runs or long counting times to minimize uncertainties arising from the counting statistics. Spectra from the various samples are carefully examined in the vicinity of each peak of interest in order to choose the proper windows for the background and peak ROIs. High ^{241}Am content samples are necessary at this step to ensure that weak ^{241}Am peaks do not appear in any of the background ROIs. A consensus set of ROIs is then fixed.

After the ROIs have been set, the calibration runs are analyzed with all calibration constants set to unity. For the isotopic ratios with one term we form, for each sample, the ratio of the isotopic ratio measured by gamma rays to the accepted ratio (usually from mass spectrometry). If the gamma-ray results are bias free, then the average ratio of these two ratios will be unity. If the average ratio differs from unity, the multiplicative calibration constant is adjusted to eliminate the average bias.

For the ^{241}Am/^{239}Pu ratios that have two terms, the procedure is a little more complex. For each sample, the accepted value of the ratio and the measured values for the peak areas and relative efficiencies are substituted into the formulas in Tables II and III, yielding an equation with two unknowns, the two calibration constants. This series of equations is solved by weighted least squares with the weighting factors determined from the uncertainty in the accepted values.

A similar procedure is used for the ^{240}Pu/^{241}Pu ratio that has three terms. The accepted value of the ratio and the measured peak areas and relative efficiencies are substituted into the formulas in Tables II and III. This gives a series of equations of the form

$$Z = A_0 + A_1X + A_2Y \ , \qquad (1)$$

where the constants A_0, A_1, and A_2 are desired. These equations are solved by multiple linear regression techniques using the applied statistics module with a Texas Instruments TI-59 calculator.

At this point, with the calibration completed, it is important to note that all calibration adjustments are carried out on the measured isotopic ratios, ^{238}Pu/^{241}Pu, ^{239}Pu/^{241}Pu, ^{240}Pu/^{241}Pu, and ^{241}Am/^{239}Pu, not on the final isotopic fractions.

TABLE II

ISOTOPIC RATIOS FOR EQUILIBRIUM ANALYSIS

Ratio	Energies (keV)	Expression	Comment
$^{238}\mathrm{Pu}/^{241}\mathrm{Pu}$	152.7/148.6	$\mathrm{RATIO(1) = FF3841 * 1.182 * \frac{A(I152) * REFF(I148)}{A(I148) * REFF(I152)}}$	
$^{239}\mathrm{Pu}/^{241}\mathrm{Pu}$	345.0/332.3	$\mathrm{RA3941(1) = FF3941(1) * 88.59 * \frac{A(I345) * REFF(I332)}{CA(I332) * REFF(I345)}}$	Plutonium-239 interferences at 332 stripped using 345. 332 from ^{237}U corrected for ^{241}Am.
$^{239}\mathrm{Pu}/^{241}\mathrm{Pu}$	203.5/208.0	$\mathrm{RA3941(2) = FF3941(2) * 1586.37 * \frac{A(I203) * REFF(I208)}{CA(I208) * REFF(I203)}}$	208 from ^{237}U corrected for ^{241}Am.
$^{240}\mathrm{Pu}/^{241}\mathrm{Pu}$	160.3/164.6	$\mathrm{RA4041(1) = FF4041(1) * 51.08 * \frac{A(I160) * REFF(I164)}{CA(I164) * REFF(I160)}}$	Plutonium-239 interference at 160.2 stripped using 161.5. Plutonium-241 interference at 160.0 stripped using 164.6. Note three terms.
		$\mathrm{-FF4041(2) * 7.60581}$	
		$\mathrm{-FF4041(3) * 2.64084 * \frac{A(I161) * REFF(I164)}{CA(I164) * REFF(I161)}}$	
$^{241}\mathrm{Am}/^{239}\mathrm{Pu}$	125.3/129.3	$\mathrm{RAAM39(1) = FFAM39(1) * 0.02771 * \frac{A(I125) * REFF(I129)}{A(I129) * REFF(I125)}}$	Plutonium-239 interferences at 125.2 and 124.5 stripped using 129. Two terms.
		$\mathrm{-FFAM39(2) * 5.861\ E\text{-}4}$	
$^{241}\mathrm{Am}/^{239}\mathrm{Pu}$	169.5/171.3	$\mathrm{RAAM39(2) = FFAM39(3) * 0.011703 * \frac{A(I169) * REFF(I171)}{A(I171) * REFF(I169)}}$	
$^{242}\mathrm{Pu}$	Correlation	$\mathrm{PER242 = FF242(1) * \frac{FRACT(3) * [FRACTN(4) + AMPURA(2) * FRACTN(2)]}{FRACTN(2)**2}}$	Correlation for wt% ^{242}Pu.

TABLE III

ISOTOPIC RATIOS FOR NONEQUILIBRIUM ANALYSIS

Ratio	Energies (keV)	Expression	Comment
$^{238}\mathrm{Pu}/^{240}\mathrm{Pu}$	152.7/148.6	RATIO(1) = NE3841 * 1.1807 * $\frac{\mathrm{A(I152) * REFF(I148)}}{\mathrm{A(I148) * REFF(I152)}}$	
$^{239}\mathrm{Pu}/^{241}\mathrm{Pu}$	129.3/148.6	RATIO(2) = NE3941 * 49.77 * $\frac{\mathrm{A(I129) * REFF(I148)}}{\mathrm{A(I148) * REFF(I129)}}$	
$^{240}\mathrm{Pu}/^{241}\mathrm{Pu}$	160.3/148.6	RATIO(3) = NE4041(1) * 210.949 * $\frac{\mathrm{A(I160) * REFF(I148)}}{\mathrm{A(I148) * REFF(I160)}}$ -NE4041(2) * 7.6032 -NE4041(3) * 10.899 * $\frac{\mathrm{A(I161) * REFF(I148)}}{\mathrm{A(I148) * REFF(I161)}}$	Plutonium 239 interference at 160.2 stripped using 161.5. Plutonium-241 interference stripped using 148.6. Three terms.
$^{241}\mathrm{Am}/^{239}\mathrm{Pu}$	125.3/129.3	RAAM39(1) = NEAM39(1) * 0.02771 * $\frac{\mathrm{A(I125) * REFF(I129)}}{\mathrm{A(I129) * REFF(I125)}}$ -NEAM39(2) * 5.861 E-4	Would not be expected to be useful for fresh samples after americium separation.
$^{242}\mathrm{Pu}$	Correlation	PER242 = FF242(1) * $\frac{\mathrm{FRACT(3) * [FRACTN(4) + AMPURA(2) * FRACTN(2)]}}{\mathrm{FRACTN(2)**2}}$	Correlation for wt% $^{242}\mathrm{Pu}$.

If biases are observed in an isotopic fraction, one must return to the above ratios in order to correct them.

B. SRP System Calibration

The SRP system calibration was carried out during April 1981. A total of 12 samples, described in Table IV, were used. Measurements were typically 24 h on each sample, usually divided into six 4-h runs. Averaged isotopic ratios, peak areas, and relative efficiencies were obtained, as necessary, for each sample. The calibration procedure described above was followed. After the calibration constants were determined, the data were reanalyzed with the proper constants to see how well the procedure worked. The final comparison was made on the isotopic fractions (weight per cent, actually), not on the ratios. This comparison, after calibration, is shown in Table V for equilibrium analysis and in Table VI for nonequilibrium analysis. A plot of the equilibrium data of Table V is given in Fig. 7. We point out that the nonequilibrium analysis was done on equilibrium samples. True nonequilibrium samples were not available to us. That the nonequilibrium analysis should work on equilibrium samples would seem to be a necessary, but not necessarily a sufficient condition for its general operation.

For the plutonium isotopic fractions, the equilibrium analysis produces somewhat more precise results than those from the nonequilibrium analysis. This is probably related to the fact that fewer peaks are used for the nonequilibrium analysis, and relative efficiency extrapolations and interpolations are made over a greater range.

We must also look at the results for the three 0.25-g samples (946, 947, 948) with great care. Even though these samples are National Bureau of Standards (NBS) reference material, they are probably the poorest samples in the group for calibration. First, their small size yields low count rates and poor counting statistics. Their small size also gives a sharply peaked relative efficiency curve that peaks around 200 keV, making interpolation for relative efficiency at 203 and 208 keV uncertain. Third, their small size makes them totally unrepresentative of the bulk sample sizes customarily found in processing plants. Finally, the ^{238}Pu content of 948 is known to be in error, and the ^{241}Am content of all the NBS samples is not well known because it is not a certified value. It

TABLE IV

CHARACTERISTICS OF SAMPLES USED FOR CALIBRATION OF SRP ISOTOPICS SYSTEM[a]

Sample	Pu (g)	Chemical Form	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	^{241}Am (μg/g Pu)
CALEX	400	oxide	0.0100	93.76	5.862	0.3375	0.027	380
J001325	500	metal	0.0119[b]	93.77	5.900	0.2557	0.028	479
8GMETA	8	metal	0.0151	93.67	5.948	0.3370	0.033	1 474
117	2	oxide	0.0157	93.49	6.152	0.3017	0.039	332
948	0.25	sulfate	0.0102	91.69	7.952	0.3115	0.033	4 473
118	2	oxide	0.0275	90.20	8.986	0.6828	0.104	756
119	2	oxide	0.0394	86.99	11.75	1.054	0.167	1 170
639	1	oxide	0.0511	86.69	11.87	1.179	0.218	9 689
946	0.25	sulfate	0.2316	84.32	12.28	2.583	0.580	17 425
116	2	oxide	0.3797	78.71	15.25	4.624	1.035	4 914
120	2	oxide	0.3798	78.70	15.25	4.631	1.035	12 087
947	0.25	sulfate	0.2780	76.92	18.65	2.942	1.214	19 024

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[a] Values for April 15, 1981.
[b] Gamma-ray measurement.

TABLE V

SRP ISOTOPICS CALIBRATION RATIO OF MEASURED/ACCEPTED VALUES
(Equilibrium Analysis)

Sample	^{240}Pu (wt%)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power
CALEX	5.86	1.0647	1.0002	0.9977	0.9929	1.0171	1.0016
J001325	5.90	----[a]	1.0007	0.9954	0.9901	0.9996	----
8GMETA	5.95	0.7905[a]	1.0008	0.9891	0.9798	1.0222	0.9929
117	6.15	0.9238	1.0010	0.9884	0.9977	0.9906	0.9956
948	7.95	1.0927[a]	0.9987	1.0100	1.0112	----	----
118	8.99	0.9778	0.9997	1.0068	0.9991	0.9829	0.9995
119	11.75	0.9939	1.0008	0.9947	0.9963	1.0068	0.9988
639	11.87	1.0178	0.9990	1.0008	0.9934	0.9629	0.9906
946	12.28	1.0235	0.9989	0.9961	1.0127	1.0255	1.0132
116	15.25	0.9924	1.0015	0.9871	1.0000	0.9981	0.9947
120	15.25	0.9909	0.9970	0.9993	0.9984	0.9848	0.9927
947	18.65	1.015	0.9942	1.0121	1.0069	0.9963	1.0037
	$\bar{x}$	1.000	0.9994	0.9981	0.9982	0.9988	0.9983
	S_i	0.038	0.0021	0.0082	0.0091	0.0186	0.0067

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[a] Omit from average, accepted value known to be in error.

TABLE VI

SRP ISOTOPICS CALIBRATION RATIO OF MEASURED/ACCEPTED VALUES
(Nonequilibrium Analysis)

Sample	^{240}Pu (wt%)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power
CALEX	5.86	1.0836	0.9994	1.0015	1.0027	1.0055	1.0022
J001325	5.90	----[a]	1.0001	1.0047	0.9964	0.9893	----
8GMETA	5.95	0.8077[a]	0.9999	1.0031	0.9911	1.0186	0.9950
117	6.15	0.9218	1.0017	0.9781	0.9909	1.0021	0.9943
948	7.95	1.1044[a]	0.9983	1.0140	1.0278	----	----
118	8.99	0.9804	0.9983	1.0199	0.9998	0.9868	1.0021
119	11.75	0.9985	0.9972	1.0207	0.9985	1.0092	1.0046
639	11.87	1.0272	0.9962	1.0194	1.0053	0.9654	0.9949
946	12.28	1.0521	1.0007	0.9764	1.0478	1.0170	1.0148
116	15.25	0.9987	1.0014	0.9859	1.0044	1.0081	0.9981
120	15.25	0.9970	0.9966	0.9992	1.0025	0.9936	0.9968
947	18.65	1.0350	0.9892	1.0258	1.0324	0.9839	1.0063
	$\bar{x}$	1.0105	0.9982	1.0041	1.0083	0.9981	1.0009
	S_i	0.046	0.0033	0.0169	0.0178	0.0160	0.0065

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[a] Omit from average, accepted value known to be in error.

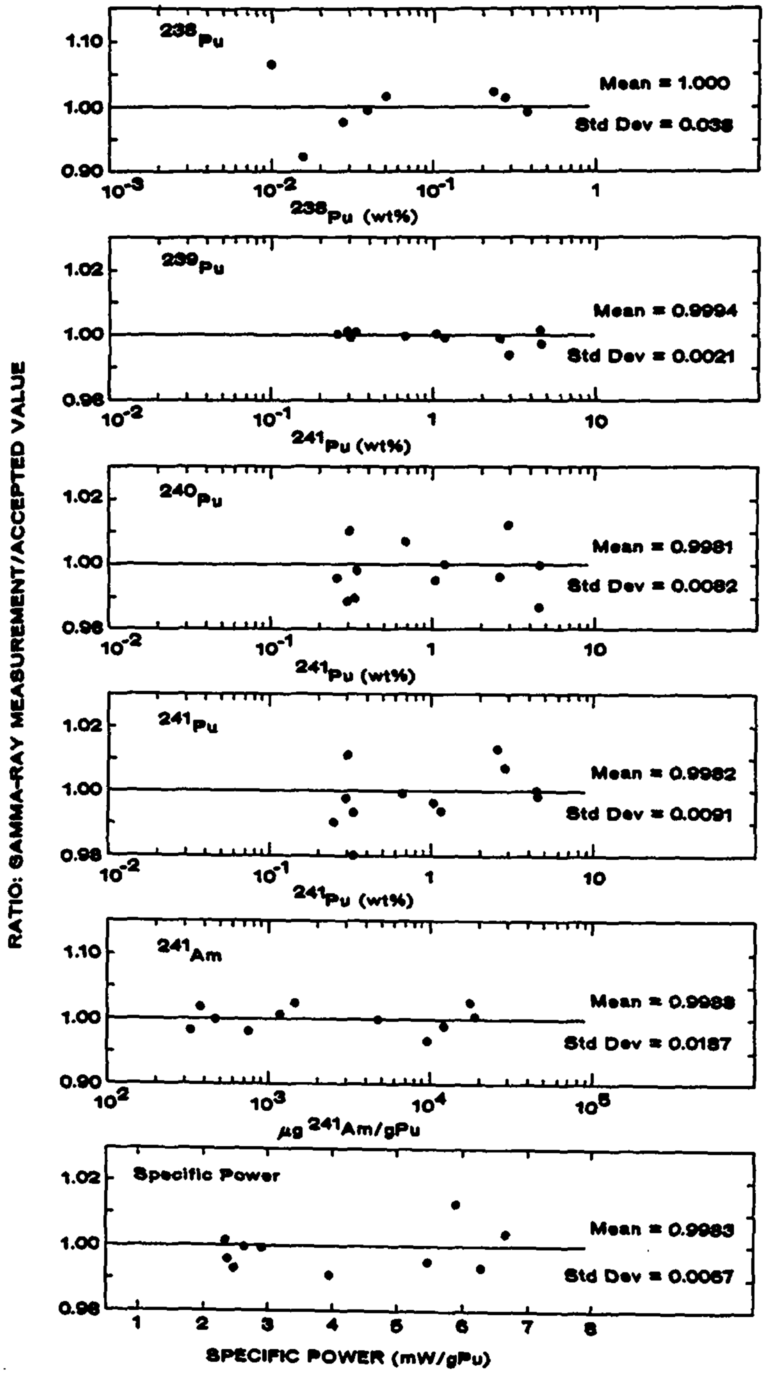

Fig. 7. SRP isotopics calibration, equilibrium data analysis.

is best to use these samples for their intended purpose, that is, to calibrate mass spectrometers. Then the mass spectrometer can be used to characterize larger, more useful, samples to check the gamma-ray isotopics.

While on this subject, it does not hurt to reemphasize some well-known facts about mass spectrometry as it compares with gamma-ray isotopics. First, because measurement of ^{238}Pu by mass spectrometry for low concentrations of ^{238}Pu (<0.1%) can be biased high by contamination with environmental ^{238}U, alpha counting should be relied on for ^{238}Pu results in these concentrations. Second, before a sample is measured on a mass spectrometer, a chemical separation is carried out to remove any americium so that ^{241}Am will not interfere with the measurement of ^{241}Pu. If this separation is not complete, the ^{241}Pu result will be biased high. Finally, we are not aware of any significant problems with the measurement of ^{239}Pu and ^{240}Pu. Also, just because problems can arise with ^{238}Pu and ^{241}Pu does not mean that these problems are always present. Most mass spectrometer measurements are more accurate and precise than the gamma-ray results described here.

C. TA-55 System Calibration Verification

This procedure, carried out during May 1982, followed the same steps as outlined for the SRP system. Twice as many data were generated because two detectors were checked. As before, 24 h of data were collected for each sample. The characteristics of the samples are given in Table VII. Nine of the fifteen samples are the same as those used for the SRP calibration. The remainder are different, including some with only 2% ^{240}Pu.

After the data were available, they were analyzed with the same windows and calibration constants that were determined for the SRP system over a year previously. The results for equilibrium analysis on detectors 1 and 2 are displayed in Tables VIII and IX and plotted in Fig. 8. Overall, the average values look quite good for both detectors, although the ^{241}Am results for concentrations <10^3 μg/g Pu may be biased. Considering that we have different detectors, electronics, and multichannel analyzer, the fact that the linearity is

TABLE VII

CHARACTERISTICS OF SAMPLES USED FOR CALIBRATION VERIFICATION OF TA-55 MUDPI SYSTEM

Sample	Pu (g)	Chemical Form	^{238}Pu (wt%)	^{239}Pu (wt%)	^{240}Pu (wt%)	^{241}Pu (wt%)	^{242}Pu (wt%)	Am (μg/g Pu)	Date
151	2	oxide	0.0024	97.96	2.010	0.0199	0.003	11	4/29/82
HRA19	2200	metal	0.0027[a]	98.01	1.960	0.0173	0.001	457	4/18/82
HRA33	2200	metal	0.0023[a]	97.83	2.134	0.0204	0.002	506	4/18/82
CALEX	400	oxide	0.0108	93.78	5.863	0.3219	0.270	544	4/16/82
J001325	500	metal	0.0118[a]	93.81	5.902	0.2431	0.028	605	5/4/82
117	2	oxide	0.0156	93.51	6.152	0.2866	0.039	482	4/19/82
STD8	240	oxide	0.0104[a]	93.41	6.326	0.2165	0.025	801	4/20/82
STD3	60	oxide	0.0237[a]	91.82	7.609	0.4745	0.077	1 933	5/4/82
118	2	oxide	0.0273	90.23	8.989	0.6492	0.104	1 093	4/27/82
STD40	875	oxide	0.0677	86.88	11.74	1.106	0.200	1 549	4/20/82
119	2	oxide	0.0391	87.03	11.76	1.002	0.167	1 689	4/27/82
946	0.25	sulfate	0.232	84.43	12.30	2.461	0.582	19 080	4/24/82
116	2	oxide	0.3776	78.89	15.28	4.416	1.037	7 099	4/22/82
120	2	oxide	0.3776	78.89	15.28	4.416	1.037	14 350	4/22/82
947	0.25	sulfate	0.278	77.03	18.67	2.803	1.215	20 750	4/24/82

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[a] Gamma-ray measurement.

TABLE VIII

TA-55 MULTIPLE-DETECTOR PLUTONIUM ISOTOPICS
(Detector 1)

Ratio: Measured/Accepted Isotopic Values
Equilibrium Analysis

Sample	^{240}Pu (wt%)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power	Comment
151	2.01	1.160	1.0005	0.9747	0.9645	----	0.9998	Americium too low to measure.
HRA33	2.13	---	1.0019	0.9185	0.637	1.474	0.9864	Accepted values high for 238 and 241.
CALEX	5.86	0.9935	0.9996	1.0073	0.9944	0.9265	0.9989	
117	6.15	0.9033[a]	0.9998	1.0051	1.0074	0.9453	0.9960	
STD8	6.33	---	1.0012	0.9834	0.9866	0.9783	0.9964	Gamma value used for 238
J001325	5.90	---	1.0010	0.9867	0.9895	0.9836	0.9980	Gamma value used for 238.
STD3	7.61	---	1.0006	0.9955	1.0036	0.9955	0.9993	Gamma value used for 238.
118	8.99	0.9839	1.0002	1.0011	1.0015	0.9632	0.9978	
119	11.76	1.0150	0.9996	1.0031	0.9977	0.9755	1.0003	
STD40	11.74	1.0014	1.0014	0.9922	0.9943	1.0034	0.9988	
946	12.30	1.0107	0.9975	1.0060	1.0084	1.0035	1.0033	
116	15.28	0.9958	1.0003	0.9923	1.0017	1.0007	0.9973	
120	15.28	0.9915	0.9969	0.9995	0.9994	0.9879	0.9931	
947	18.67	1.0082	0.9969	0.9967	1.0056	0.9873	0.9974	
	$\bar{x}$	0.9999	0.9996	0.9974	0.9992	0.9792	0.9980	Two per cent ^{240}Pu samples not used in averages.
	S_i	0.0107	0.0016	0.0077	0.0069	0.0238	0.0025	

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[a] Not used in average.

TABLE IX

TA-55 MULTIPLE-DETECTOR PLUTONIUM ISOTOPICS
(Detector 2)

Ratio Measured/Accepted Isotopic Values
Equilibrium Analysis

Sample	^{240}Pu (wt%)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power	Comment
151	2.01	0.9832	1.0002	0.9913	0.9602	---	1.0002	Americium too low to measure.
HRA19	1.96	---	1.0019	0.9097	0.7457	1.347	0.98409	Accepted values for 238 and 241 high.
CALEX	5.86	0.9851	1.0001	0.9985	0.9964	0.9616	0.9985	
J001325	5.90	---	1.0005	0.9941	0.9920	1.0351	0.9998	Gamma value used for 238.
117	6.15	0.8910[a]	1.0001	1.0014	1.0019	0.9859	0.9960	
STD8	6.33	---	1.0006	0.9940	0.9871	0.9733	0.9982	Gamma value used for 238.
STD3	7.61	---	1.0011	0.9898	1.0034	1.0026	0.9990	Gamma value used for 238.
118	8.99	0.9828	1.0002	1.0012	1.0002	0.9608	0.9976	
STD40	11.74	0.9922	1.0022	0.9867	0.9937	1.0275	0.9981	
119	11.76	1.0074	1.0002	0.9993	0.9950	0.9871	0.9996	
946	12.30	1.0134	0.9980	1.0028	1.0089	0.9985	1.0024	
116	15.28	0.9918	1.0007	0.9908	1.0011	1.0041	0.9960	
120	15.28	0.9995	0.9991	0.9872	1.0060	0.9872	0.9945	
947	18.67	1.0095	0.9950	1.0091	1.0084	0.9843	0.9975	
	$\bar{x}$	0.9977	0.9998	0.9962	0.9995	0.9923	0.9981	Two per cent ^{240}Pu samples not used used in averages.
	S_i	0.0115	0.0018	0.0069	0.0068	0.023	0.0021	

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[a] Not used in average.

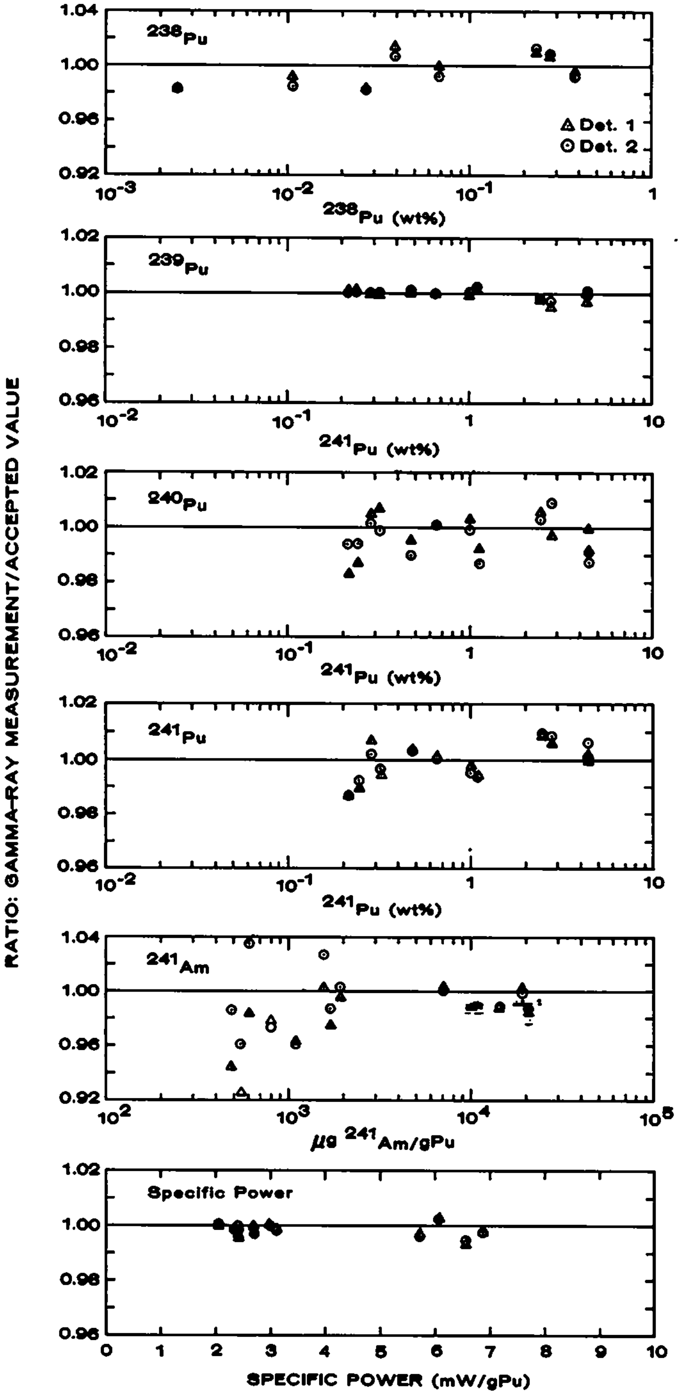

Fig. 8. Calibration verification for TA-55 multiple-detector plutonium isotopics system. Equilibrium analysis.

good enough to make the same windows usable is quite satisfying. The calibration done for the SRP system probably can be used as a generic calibration for similar systems.

The TA-55 system was put into routine operation with the same calibration and window parameters as those used in the SRP system. A check of the calibration for nonequilibrium analysis was not done.

D. Measurement Precision

As part of calibration procedures, many samples were measured repeatedly to verify measurement precision estimates in the computer codes.

Measurement precision is affected by many variables. The effects of counting rate and counting time are obvious. For samples containing more than a few grams of plutonium, we usually can obtain maximum desirable system count rates. Sample size has another effect. Large samples generally give poorer precision than do small samples of the same material, because scattering of higher energy gamma rays within the sample increases the continuum under the 100- to 200-keV region and thus yields higher backgrounds and poorer statistical precision. Examples of precision of the isotopic fractions and specific power from two reasonably large samples with different isotopic fractions are given in Figs. 9-11. These are representative of large samples of this type and show reasonable agreement between the code predictions and experimental observations. Specific power can be measured with 1% or better precision with a 30-min count time. Routine counting is done for 2 h at 15 kHz.

Three examples of precision for small samples of widely varying isotopic composition are given in Table X. All of these samples produce count rates lower than 15 kHz.

V. RESULTS FROM PROCESS CAMPAIGNS AT TA-55

A. Scope of Results

Hundreds of samples have been measured on the TA-55 system in the year since its installation. For most of these samples, detailed comparisons with conventional destructive techniques cannot be made because conventional analyses are not available.

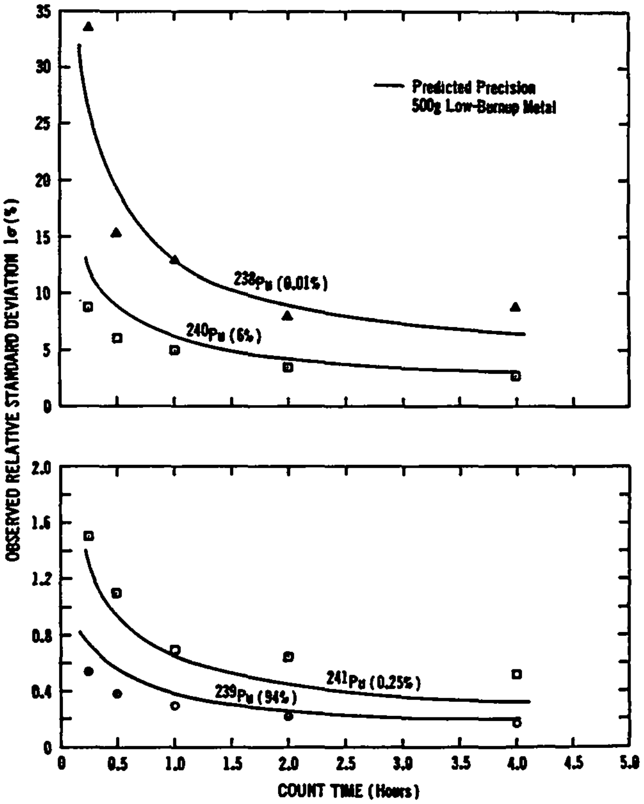

Fig. 9. Precision of isotopic fractions determined from 30 measurements on 500 g plutonium metal with isotopic fractions listed in parentheses. Solid line is prediction from analysis code.

Two groups of samples were extensively analyzed, both destructively and nondestructively, and the results are discussed below. All isotopic measurements were made by process-level technicians who had been trained in the use of the system.

B. LZB120 Series, PuO_2, 6% ^{240}Pu

Forty-nine samples were measured on the isotopics system. The measurements were compared with results from mass spectrometry, alpha counting for ^{238}Pu, calorimetry, and destructive analysis for plutonium concentration. These samples contained ~1500 g PuO_2 with the approximate isotopic composition being ^{238}Pu, 0.01%; ^{239}Pu, 93.6%; ^{240}Pu, 5.9%; ^{241}Pu, 0.45%; ^{242}Pu, 0.035%; ^{241}Am, 1500 μg/g Pu.

The combination of plutonium isotopic measurements (by way of mass spectrometry or gamma-ray spectroscopy) with calorimetry to produce total plutonium content is difficult to analyze. Offsetting errors in the two methods can result in apparently correct answers for total plutonium, which

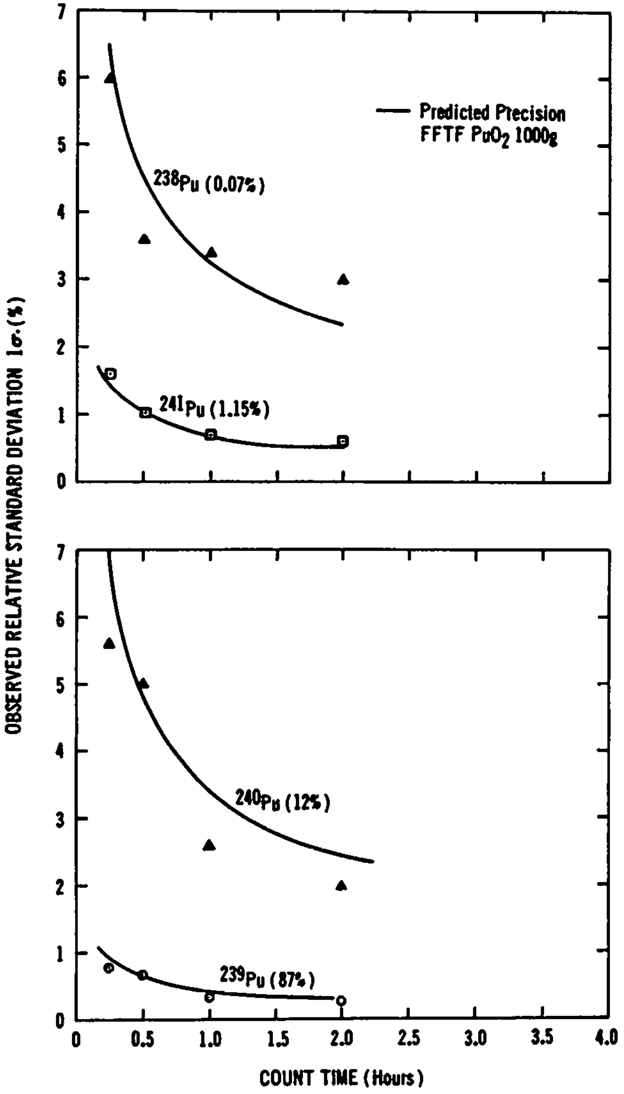

Fig. 10. Precision of isotopic fractions determined from 30 measurements on 1000 g PuO_2 with isotopic fractions indicated in parentheses. Solid line is prediction from analysis code.

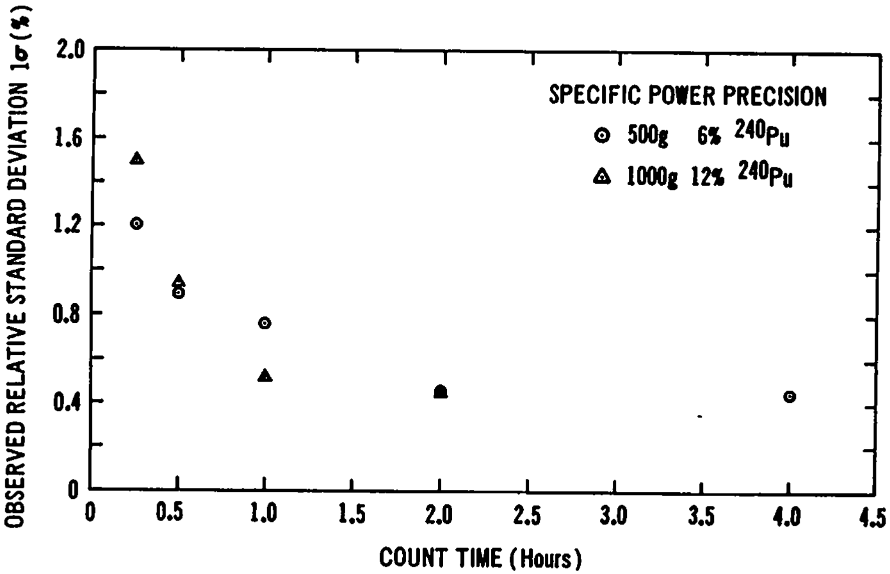

Fig. 11. Precision of specific power determined from 30 measurements on 500-g metal and 1000-g PuO_2 samples.

TABLE X

EXAMPLES OF MEASUREMENT PRECISION FOR SMALL SAMPLES

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power (mW/g Pu)	Comment
Isotopic composition (wt%)	0.068	87.01	11.59	1.15	0.11	3.048	Count-rate factor of 3 lower than usual maximum.
Observed precision, 19 meas., %RSD, 4-h count, 4200 counts/s, 15 g PuO_2	2.5%	0.29%	2.1%	0.56%	4.5%	0.55%	
Isotopic composition (wt%)	0.12	82.40	15.09	1.77	1.29	4.893	Count rate lower than ideal.
Observed precision, 30 meas., %RSD, 2-h count, 6100 counts/s 1 g PuO_2	0.96%	0.20%	1.0%	0.60%	0.61%	0.15%	
Isotopic composition (wt%)	0.0026	98.04	1.93	0.020	--	2.050	Americium too low to measure.
Observed precision, 14 meas., %RSD, 4-h count, 2700 counts/s, 3 g PuO_2	27.5%	0.18%	8.9%	0.74%		0.75%	Very low count rate.

lead to a false sense of security for the individual techniques. In Table XI, gamma-ray isotopics are compared to those determined by mass spectrometry and radiochemistry (^{238}Pu, ^{241}Am). Results for the two detectors are evenly represented and no differences were observed between them.

There are four sets of numbers in the first set of data in Table XI. The "ratio" shows the average of the ratio of the individual gamma-ray results to mass spectrometry data. This gives a measure of the bias when the standard deviation of the average ratio ($S_{\bar{x}}$, third line) is taken into account. The second line, S_i, is the standard deviation of an individual ratio, calculated from the observed ratios. This is to be compared with the last line, the predicted precision from the MUDPI code. S_i includes the precision of the mass spectrometry result folded with the gamma-ray precision. For most isotopes the mass spectrometry precision is enough smaller than the gamma-ray precision that it does not contribute to S_i, and a direct comparison between S_i and the MUDPI prediction can be made. Typical mass spectrometer precision (alpha counting for ^{238}Pu and ^{241}Am) in percentage relative standard deviation (RSD) is on the order of ^{238}Pu, 0.5%; ^{239}Pu, 0.03%; ^{240}Pu, 0.3%; ^{241}Pu, 0.7%; and ^{241}Am, 2%. The mass spectrometry/alpha counting precisions are typically an order of magnitude better than these gamma-ray measurements for ^{238}Pu, ^{239}Pu, and ^{240}Pu. They are comparable to the gamma-ray measurements for ^{241}Pu and ^{241}Am. These relations generally hold for this approach to gamma-ray isotopics with the exception that higher burnup samples give better precision for gamma-ray results on ^{238}Pu and ^{241}Am than do the routine conventional techniques.

The detailed comparison of observed and predicted precision cannot easily be made for the other three series in Table XI because the last three series combine different numbers of measurements of the same length for different samples.

An isotope-by-isotope discussion follows:

Plutonium-238

An average difference of ~6% is seen for these data where alpha counting is used for ^{238}Pu. This is quite good compared to the differences of 50 to 100% that usually appear when mass spectrometry is used for ^{238}Pu. It is difficult to ascertain whether MUDPI is in error, alpha counting is biased, or some combination of both exists.

TABLE XI

COMPARISON OF GAMMA-RAY ISOTOPICS WITH MASS SPECTROMETRY AND RADIOCHEMISTRY

$$\text{Ratio: } \frac{\text{Gamma-ray measurement}}{\text{Mass spectrometry + radiochemistry}}$$

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Specific Power
2- to 2.5-h Measurements **26 Samples, 1 measurement each sample**							
Average ratio	1.0419	1.0003	0.9955	0.9887	1.100	1.0394	1.0030
S_i	0.107	0.0024	0.037	0.0219	0.139	0.0392	0.0054
$S_{\bar{x}}$	0.021	0.0005	0.007	0.004	0.027	0.008	0.0011
MUDPI (Predicted precision, 1 RSD)	$\sim$0.1	$\sim$0.0023	0.037	$\sim$0.004	---	$\sim$0.025	$\sim$0.004
4-h Measurements **15 Samples, 43 Total Measurements**							
Average ratio	1.0764	1.0007	0.9893	0.9968	0.985	1.0322	1.0026
6- to 8-h Measurements **3 Samples, 16 Total Measurements**							
Average ratio	1.055	1.0007	0.9874	1.0167	0.939	1.024	1.0016
16- to 24-h Measurements **4 Samples, 12 Total Measurements**							
Average ratio	1.0795	1.0006	0.9927	0.9809	0.933	1.0227	1.0021

Plutonium-239

The essentially negligible positive bias for ^{239}Pu is probably a result of the small negative bias for ^{240}Pu shown by MUDPI.

Plutonium-240

The negative bias shown by MUDPI is likely to be real. Mass spectrometry has always been reliable for this isotope.

Plutonium-241

Mass spectrometry can produce results biased high if the ^{241}Am separation is incomplete. This can cause the MUDPI/accepted ratio to be low, consistent with most of our observations. The observed precision of the ratio ($\sim$2% RSD) is about five times that predicted from the statistics of the MUDPI measurement. Because we expect mass spectrometry precision to be $\sim$0.5 to 1.0% RSD, there seems to be an unresolved component in the precision of the ratio. This could be evidence for fluctuations in the mass spectrometry results caused by varying degrees of incomplete ^{241}Am separations. If so, this could explain the small negative bias seen for the MUDPI/accepted ratio. In any event, this bias is not significant in determining the specific power.

Plutonium-242

The ^{242}Pu ratio to accepted values is very satisfactory considering that a correlation is used for the MUDPI values and that mass spectrometry precision for a single measurement is likely to be $\sim$5 to 10% RSD.

Americium

The ~3% difference seen in Table XI is consistent with the measurement control value for STD40, whose absolute americium concentration is nearest to those of the LZB120 series. We also do not believe that the MUDPI system can be checked to much better than ±2% with the standards we now have because the radiochemical values are not as accurate as we could use.

Power

The 0.2 to 0.3% agreement of the power ratio seen in Table XI seems to indicate that neither MUDPI nor mass spectrometry is in serious trouble. This agreement is much better than that seen previously when mass spectrometry was used to determine ^{238}Pu. The difference is probably acceptable if it is translated to total plutonium mass. However, because of some uncertainties as to the absolute accuracy of the mass spectrometry values, we examined calorimetry data in order to get a further idea of the accuracy of MUDPI.

During the series of gamma-ray measurements on the LZB120 samples, all items were also measured by calorimetry. Two standards were also measured several times during the period to check on calorimeter performance.

We then computed the plutonium mass in each sample by combining the calorimetry with the gamma-ray spectroscopy isotopic results after correcting the calorimetry for biases determined from the standards measurements. We compared these numbers with the plutonium mass determined from conventional destructive chemistry plus weighing. This comparison yielded the bias in the specific power arising from the gamma-ray measurement. This bias was found to be 0.9978 ± 0.0020, essentially the same as that found during system calibration (Tables VIII and IX).

C. Two Per Cent ^{240}Pu Metal

These samples have proved to be one of the most difficult measurement problems for this system because of the very weak ^{241}Pu and ^{237}U peaks in the spectrum (~0.03 wt% ^{241}Pu, typically). These weak peaks are shown in a spectrum (Fig. 12) in the 120- to 215-keV range from this type of material.

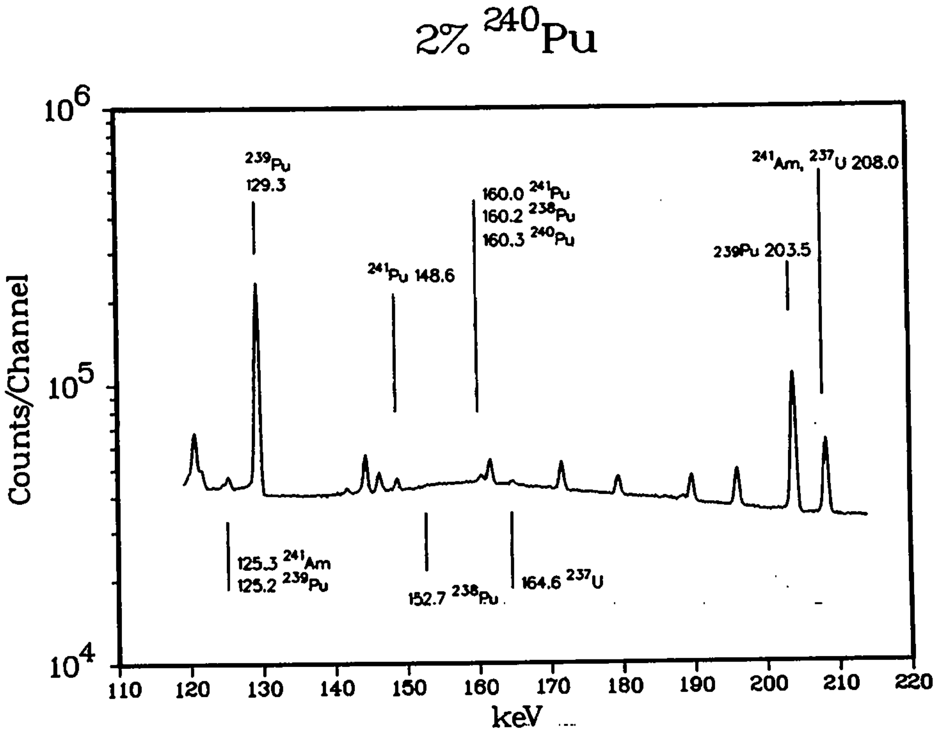

Fig. 12. The 120- to 215-keV region of a spectrum from a metal button containing ~2% ^{240}Pu.

All isotopes except ^{239}Pu are difficult to measure because of poor statistical precision arising from low abundance, even with 4-h count times. Typical isotopic abundances in these nominal 2-kg metal buttons were ^{238}Pu, 0.004%; ^{239}Pu, 97.50%; ^{240}Pu, 2.46%; ^{241}Pu, 0.033%; ^{242}Pu, 0.003%; and ^{241}Am, 770 µg/g Pu.

Table XII compares data from 34 samples with mass spectrometry data (alpha counting for ^{238}Pu and ^{241}Am).

These kinds of samples also push the limits of mass spectrometry on the low abundance of ^{241}Pu. Discussion of the biases for each isotope follows:

Plutonium-238

The almost nonexistent ^{238}Pu gamma-ray peak makes a quantitative gamma-ray measurement nearly impossible. Typical MUDPI measurement precision was 21%. It is no surprise that the gamma-ray data could be biased.

Plutonium-239

The essentially zero bias result is influenced by the fact that ^{239}Pu composes almost the entire sample. Errors in its measurement wash out when isotopic fractions are normalized to unity.

Plutonium-240

The bias of 1% is probably as small as this gamma-ray technique can provide for this small concentration.

Plutonium-241

The 10% bias here is somewhat of a puzzle. The precision of the ratio, 4.5%, is much greater than that of the gamma-ray measurement, 0.6%. This may mean that the mass spectrometry precision at these low abundances is significant or that variably incomplete ^{241}Am separations are influencing the mass spectrometry data. The latter would cause a bias in the observed direction. The very weak ^{241}Pu peaks may result in a gamma-ray bias. At the abundance level of 0.03%, the best answer may be, "Who cares?"

Americium-241

As for ^{241}Pu, we observe a biased ratio (18%) with a large standard deviation, 38% relative, but with a gamma-ray measurement precision of ~4%. The ^{241}Am concentrations are within the measurement range of the MUDPI system, and the radiochemistry precision is expected to be in the 2 to 3% RSD range. The MUDPI/radiochemistry ratios vary from 0.62 to 2.17. These considerations indicate that sample inhomogeneity may be the problem. This has been observed in these kinds of buttons in the past. Note that if this is the case, neither the MUDPI measurement nor the radiochemistry result is "wrong," but neither one may be representative of the average properties of the sample.

Calorimetry data from 20 of the samples were also analyzed after the calorimetry results were corrected for bias by measurements of a 2.75-W

TABLE XII

COMPARISON WITH MASS SPECTROMETRY, TWO PER CENT ^{240}Pu METAL BUTTONS

$$\text{Ratio: } \frac{\text{MUDPI (gamma-ray spectroscopy)}}{\text{Mass spectrometry (radiochemistry for } ^{238}\text{Pu and } ^{241}\text{Am)}}$$

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	Specific Power
Average ratio	1.201	1.0003	0.9900	0.9015	0.840	1.184	1.0067
S_i	0.308	0.0014	0.055	0.045	0.297	0.38	0.0139
MUDPI (Predicted precision, 1 RSD)	0.21	0.0032	0.13	0.006	--	0.04	0.0065

standard (a typical button produced ~4.2 W). The corrected calorimetry was combined with the gamma-ray isotopics to give total plutonium mass. The total plutonium mass was compared with the mass obtained from conventional destructive analysis. The comparison is given in Table XIII.

The average bias of 18% on the ^{241}Am content can account for about 0.7 of the 1.3% plutonium mass bias if one assumes that the radiochemistry results for ^{241}Am are representative of the average content of the samples.

If sample inhomogeneity (Am/Pu not constant throughout the sample) is indeed the problem with the ^{241}Am measurement, little can now be done to improve the nondestructive analysis results. Measurements on the material after conversion to oxide and blending would presumably eliminate the problem.

Note that the bias in total plutonium mass determined by comparison with destructive analysis (1.3%) is not the same as the bias in the specific power determined by comparison to mass spectrometry (0.67%, Table XII). This could indicate some bias in the mass spectrometry results, effects from ^{241}Am inhomogeneity, or incorrect bias corrections on the calorimetry. The destructive chemistry is accepted to be the most accurate component compared.

These comparisons illustrate the difficulty of reconciling three techniques (gamma-ray spectroscopy, calorimetry, and mass spectrometry/radiochemistry) at the sub-1% accuracy level.

D. Mixed Oxide

Measurements have shown that the program can handle mixed uranium-plutonium oxide samples as long as the ^{235}U/Pu ratio is below 0.3 (Ref. 7). This ratio covers typical United States breeder oxide with 2 to 4 parts depleted or natural uranium to 1 part plutonium (^{235}U/Pu ~0.004-0.03) but does not hold if the uranium is more highly enriched in ^{235}U (>10-20% ^{235}U). A few samples of enriched uranium/plutonium mixed oxides have been measured at TA-55. Because of too few samples and the unavailability of reliable analytic data, we have been unable to produce a special calibration file for mixtures of highly enriched uranium and plutonium.

TABLE XIII

TOTAL PLUTONIUM COMPARISON

	MUDPI + Calorimetry	Destructive Chemical Analysis + Weight
Plutonium mass (g) 20 samples	39 022	39 538

Difference = 1.3%

E. Salts, Slag, and Crucibles

Another difficult problem involves the measurement of residues from electrochemical processes. Two conditions complicate these measurements. The first is that ^{241}Am concentrations may be very high (>10%), much higher than the 2% maximum values for which the calibration has been verified. These high levels require some adjustment of ROI windows, which, in turn, implies an adjustment of calibration constants. Because these types of residues are not normally analyzed destructively, accepted values to use to verify a calibration are not available. The second condition is much more serious, potentially fatal, for this type of measurement. This condition is the observation that the americium is not homogeneously mixed with the plutonium in some of these residues. If this is the case, the ^{241}Am/Pu ratio will not be correct and the specific power will be biased. It is difficult to determine the extent of these biases.

VI. MEASUREMENT CONTROL RESULTS

During the year that the TA-55 system has been in operation, two well-characterized samples have been measured on a regular basis. The measurements have not been used in a formal measurement control program because the instrument was not being used for formal accountability measurements. However, all the data have been analyzed and give a good indication of the performance stability over an extended period of time. There were no adjustments made to the system during this 1-year period. The only maintenance required (see Sec. VII) did not affect the system performance. All measurements, except those of April 1982, were done by process technicians.

The two samples are CALEX1, a 400-g plutonium as PuO_2 calorimeter standard with 6% ^{240}Pu, and

STD40, an 875-g plutonium as PuO_2 standard with 12% ^{240}Pu. Their isotopic characteristics are given in Table VII, where CALEX1 is called "CALEX."

All measurements are plotted in Figs. 13 and 14 with averages for three time intervals, denoted by vertical lines, tabulated in Tables XIV and XV. All measurements and standard values were decay corrected to a common date before ratios were calculated. Measurements from detector 1 are denoted by triangles and those from detector 2 by boxed X's. The measurements for the first period, April 1982, are the calibration verification data already discussed and tabulated in Tables VIII and IX. These were 4-h runs and show less scatter than do the 2-h runs used for all the rest of the results. Differences between the average ratio for ^{238}Pu in CALEX1 shown in Table XIII for the April 1982 data and those shown in Tables VIII and IX reflect a re-evaluation of the accepted value for that parameter between the times the two sets of tables were prepared.

Evidence of small biases is apparent and reflects the difficulty of calibrating for a wide range of sample sizes and isotopic composition with one set of parameters.

No significant long-term trends are apparent, so we conclude that system operation has been stable for the 1 year of operation during which no adjustments were made to the detectors, electronics, or calibration parameters. This level of accuracy was also obtained using calibration parameters determined on an entirely different system (SRP system) a year before the initial operation at TA-55.

Implementation of a formal measurement control program has been discussed. One suggestion is to monitor the fundamentally measured isotopic ratios (not the fractions) and the specific power. Implementation of such a program awaits further definition.

VII. MAINTENANCE EXPERIENCE AND TROUBLE SPOTS

In the 1-year operation at TA-55, maintenance experience has been excellent. Only two items have required maintenance during this period. The first was one of the DEC RX02 floppy disk drives (one drive still remained available), and the second was one of the fans circulating air through the instrument racks.

Even though much effort has been expended to make the system easy to operate, there always remains room for improvement. The flexibility of being able to choose or reject the recording of data onto disks has presented the most problems for operators. Confusion between names of disk units, file names, and local names for the individual floppy diskettes has caused some problems. Additional training has cleared up most of these troubles. A system with less flexibility or with a more structured flexibility would probably cause fewer operator errors. However, disk recording of data is an adjunct to the hard-copy output that is always available, so no analysis results were compromised by any of these problems.

Several instances of very poor precision for the isotopic fractions were traced to measurements made on containers that had been lead lined to reduce external dose rates. This internal lead shielding reduced the 100- to 200-keV gamma-ray intensities to such a low level that meaningful measurements were impossible.

Suggestions have been made to incorporate warning messages to flag poor precision on the isotopic fractions and to flag the presence of ^{235}U in the sample. Operating problems with inhomogeneous ^{241}Am concentrations and with highly enriched uranium have been discussed in Sec. V.

After 1 year's operation, a check of detector resolution indicates that the operational resolution measured at 129 keV with 15-kHz count rate has deteriorated from ~530 eV to ~600 eV. The probable cause for this is neutron damage to the detector from its continuous exposure to kilogram quantities of plutonium. We observed the same effect on the experimental system used previously at TA-55. This would seem to indicate a useful detector lifetime of 1-1/2 to 2 years, with a recalibration and change of ROIs necessary after ~1 year.

Samples containing unexpected radioisotopes also present measurement problems for the system. Peaks from these isotopes can cause interferences with the peak-and-background ROIs normally used for analysis. We have observed such interferences from samples containing thorium and also ^{243}Am-^{239}Np. Other interferences may also cause problems but have not been specifically identified.

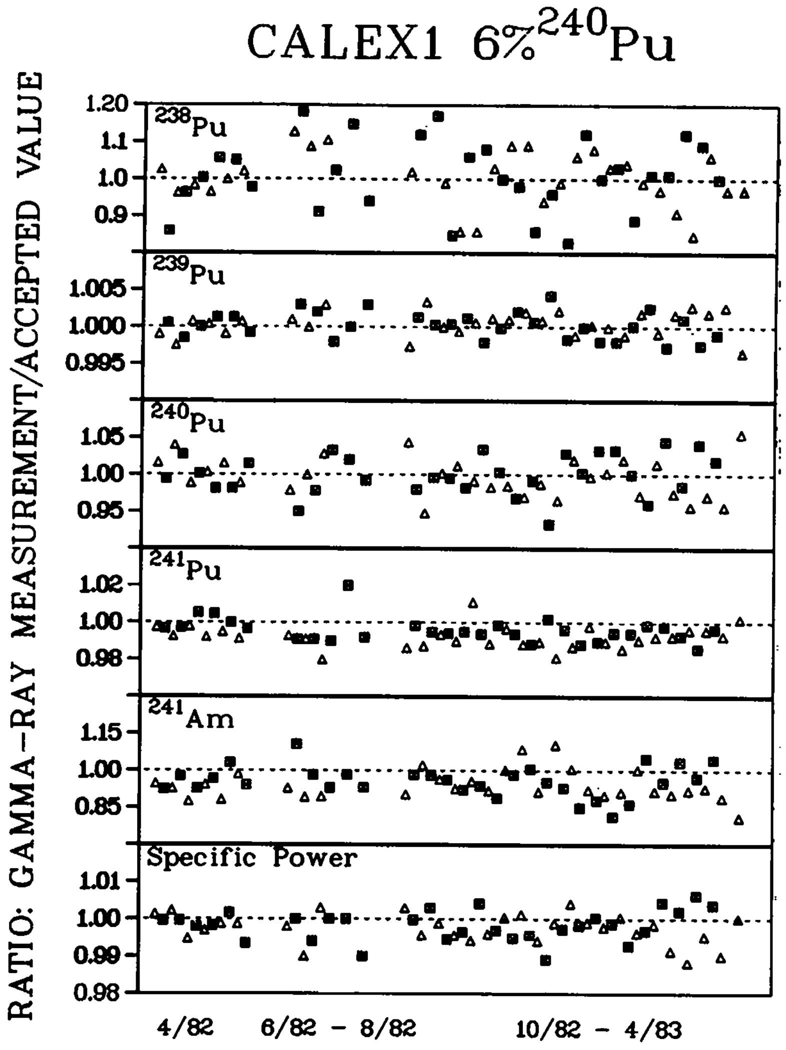

Fig. 13. Measurement on the standard CALEX1 6% ^{240}Pu for a 1-year period. Triangles: detector 1; squares: detector 2.

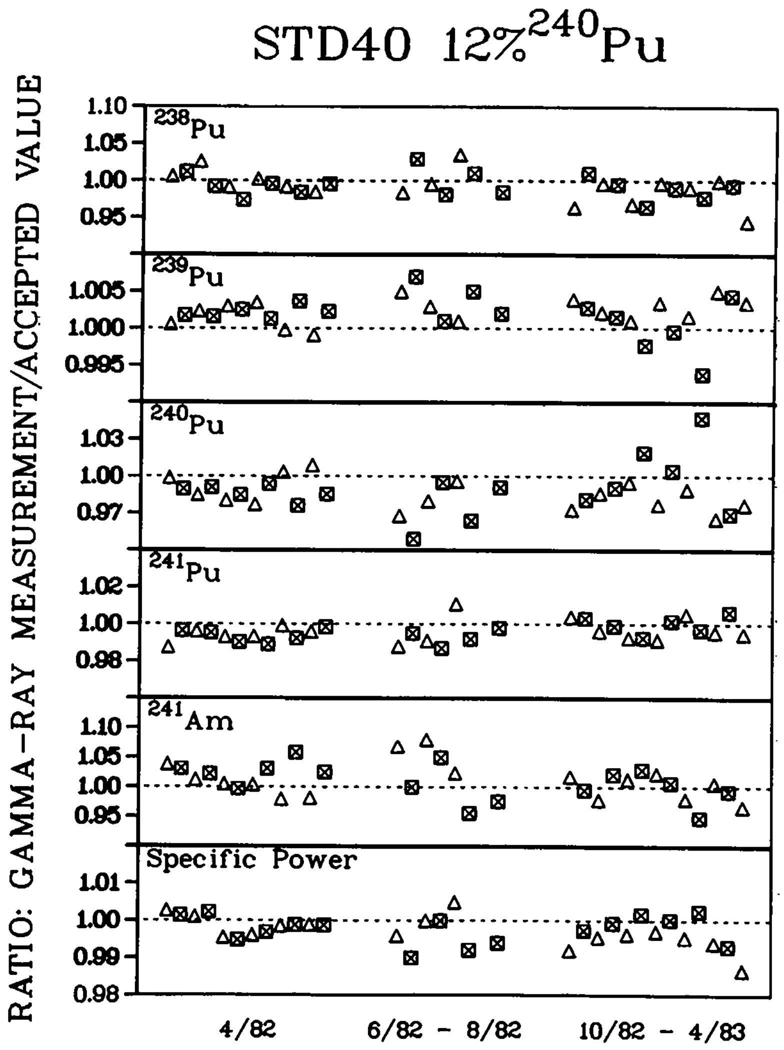

Fig. 14. Measurements on the standard STD40 12% ^{240}Pu for a 1-year period. Triangles: detector 1; squares: detector 2.

TABLE XIV

AVERAGE RATIO OF MEASURED/ACCEPTED VALUES FOR CALEX1 FOR THREE TIME PERIODS DURING THE FIRST YEAR OF OPERATION

Ratio: Measured/Accepted

Time	No. Meas.	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power
Detector 1							
4/82	6	1.071 ± 0.011	0.9996 ± 0.0005	1.0073 ± 0.008	0.9944 ± 0.0011	0.9265 ± 0.018	1.0007 ± 0.0011
6/82-8/82	3	1.107 ± 0.011	0.9997 ± 0.0009	1.0023 ± 0.014	0.9910 ± 0.0011	0.906 ± 0.011	1.0000 ± 0.0015
10/82-4/83	21	0.993 ± 0.017	1.0006 ± 0.004	0.9910 ± 0.0063	0.9953 ± 0.0014	0.946 ± 0.016	0.9970 ± 0.0009
Detector 2							
4/82	6	1.063 ± 0.03	1.0001 ± 0.0004	0.9985 ± 0.007	0.9964 ± 0.0016	0.962 ± 0.017	1.0003 ± 0.0011
6/82-8/82	5	1.042 ± 0.05	1.0012 ± 0.0010	0.9836 ± 0.015	0.9968 ± 0.0058	0.987 ± 0.032	0.9968 ± 0.0020
10/82-4/83	19	1.009 ± 0.023	1.0000 ± 0.0004	1.0007 ± 0.007	0.9977 ± 0.0009	0.946 ± 0.016	0.9985 ± 0.0010
Accepted Values (wt%)							
4/1/83		0.00990	93.795	5.858	0.3084	0.06875	2.36928 mW/g Pu

TABLE XV

AVERAGE RATIO OF MEASURED/ACCEPTED VALUES FOR STD40 FOR THREE TIME PERIODS DURING THE FIRST YEAR OF OPERATION

Ratio: Measured/Accepted

Time	No. Meas.	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Specific Power
Detector 1							
4/82	6	1.0004 ± 0.006	1.0014 ± 0.0007	0.9922 ± 0.0053	0.9943 ± 0.0016	1.0034 ± 0.0086	0.9988 ± 0.0011
6/82-8/82	3	1.0047 ± 0.015	1.0030 ± 0.0011	0.9813 ± 0.0081	0.9967 ± 0.0072	1.058 ± 0.017	1.0003 ± 0.0026
10/82-4/83	7	0.9803 ± 0.008	1.0030 ± 0.0005	0.9806 ± 0.0039	0.9972 ± 0.0020	0.9981 ± 0.0084	0.9939 ± 0.0014
Detector 2							
4/82	6	0.9922 ± 0.0053	1.0022 ± 0.0004	0.9867 ± 0.0026	0.9737 ± 0.0015	1.0275 ± 0.0078	0.9981 ± 0.0011
6/82-8/82	4	1.001 ± 0.011	1.0037 ± 0.0014	0.9747 ± 0.011	0.993 ± 0.002	0.9955 ± 0.020	0.994 ± 0.002
20/82-4/83	6	0.9884 ± 0.006	1.0000 ± 0.0016	1.0020 ± 0.012	1.0000 ± 0.012	0.9989 ± 0.012	0.9991 ± 0.0014
Accepted values (wt%) 4/1/83		0.0675	86.93	11.74	1.056	0.2052	3.1625 mW/g Pu

APPENDIX A

ANALYSIS

I. GENERAL APPROACH

The philosophy that has governed this approach is one of simplicity. The approach uses the simplest data acquisition and analysis techniques and yet has the widest applicability to arbitrary sample configurations.

The atom ratio of isotopes 1 and 2 is determined from a gamma-ray spectrum by means of

$$\frac{N_1}{N_2} = \frac{C_1}{C_2} \cdot \frac{T_{1/2}(1)}{T_{1/2}(2)} \cdot \frac{B_2}{B_1} \cdot \frac{RE_2}{RE_1} \quad , \tag{A.1}$$

where

N = number of atoms of the indicated isotope,

C = photopeak counts from a selected gamma ray from the indicated isotope,

$T_{1/2}$ = half-life of the indicted isotope,

B = branching ratio of the selected gamma ray from the indicated isotope, and

RE = relative efficiency at the selected gamma-ray energy, including geometry, sample self-absorption, attenuation, and detector efficiency.

The isotopic ratios are measured using gamma rays in the 120- to 420-keV range. Half-lives and branching ratios are taken from the literature.[4,6] The relative efficiency is determined from the spectrum under study by determining the quotient of the photopeak counts and the branching ratio for a series of gamma rays from one of the isotopes in the sample. Plutonium-239, ^{241}Pu, and ^{241}Pu-^{237}U gamma rays are used in this method with the ^{241}Pu and ^{241}Pu-^{237}U relative efficiency points being normalized to those from ^{239}Pu.

Photopeak areas are determined by region of interest (ROI) summation. Background regions are selected above and below each photopeak. A linear background is interpolated under the photopeak from the centroids of the background regions. Background regions are carefully selected to avoid neighboring peak interferences, particularly from ^{241}Am, which can vary greatly from sample to sample.

Digital gain and zero stabilization are used in the data acquisition electronics. This is important to ensure that the peaks do not wander out of their assigned ROIs. The 129.3- and 413.7-keV peaks of ^{239}Pu are used for zero and gain stabilization.

The ROI summation method puts great emphasis on good detector resolution so that we can resolve the peaks of interest from close-lying neighbors. A high-resolution (~500 eV at 122 keV) planar detector is used for these measurements. The techniques discussed here are applicable to a very wide range of sample types. Sample size is limited only by count-rate and counting-time considerations and can range from ~0.1 g to as large as criticality considerations allow. We attempt to keep count rates at about 10 to 15 kHz as a compromise between optimal data collection rates and best resolution. Counting times are influenced by the statistical precision desired and the ultimate application of the isotopic results. Simple verifications of, say, the ^{239}Pu/^{241}Pu ratio may take only a few minutes. Applications that require ^{238}Pu and/or ^{240}Pu isotopics generally require at least several hours.

Filters are used to reduce the count rate from ^{241}Am at 59.5 keV and the 100-keV x-ray and gamma-ray complex in order to remove any pile-up peaks from the 150- to 165-keV region.

A requirement for this method is that the isotopic distribution of all plutonium in the sample must be homogeneous. The sample itself may contain a nonhomogeneous plutonium distribution, but all plutonium should have the same isotopic composition. Americium must also be uniformly distributed with the plutonium.

The methods utilized apply to both freshly separated plutonium (no ^{241}Pu-^{237}U equilibrium) and aged plutonium (>45 days from uranium separation).

II. METHODS

Relative efficiency values are calculated from the peak area/branching ratio for ^{239}Pu lines at 129.3, 143.4 + 144.2, 171.3, 179.2, 189.3, 195.7, 203.5, 255.4, 297.5, 345.0, 375.0, and 413.7 keV. All relative efficiencies are normalized to a value of 1.0 at 413.7 keV.

Next, the ^{239}Pu relative efficiency values at 345.0 and 375.0 keV are linearly extrapolated to give values at 332.4 and 335.4 keV. The peak complexes at 332 and 335 keV contain contributions from ^{241}Pu-^{237}U, ^{241}Am, and ^{239}Pu. The ^{239}Pu component is subtracted from both complexes using the 345.0-keV ^{239}Pu line. The remaining peak areas at 332 and 335 keV contain contributions from ^{241}Pu-^{237}U and ^{241}Am. Assuming ^{241}Pu-^{237}U equilibrium, we use the two peak areas and two isotopic unknowns to solve for the ^{241}Am/^{241}Pu ratio.[1] This ^{241}Am/^{241}Pu ratio is used to correct other ^{241}Pu-^{237}U peaks at 164.6, 208.0, 267.5, 332.4, 335.4, 368.6, and 370.9 keV for their ^{241}Am content.[1] The same procedure applies to nonequilibrium samples, although in that case the ratio solved for is proportional to ^{241}Am/^{237}U. The ^{241}Am correction formalism remains unchanged. The ^{239}Pu relative efficiency points at 129.3, 143.4 + 144.2, 171.3, and 203.5 keV are fit to a quadratic to determine the relative efficiency at 208 keV. Relative efficiency points at 148.6 keV from ^{241}Pu and at 164.6, 208.0, 267.5, and 332.4 from ^{241}Pu-^{237}U are normalized to the values determined from ^{239}Pu lines by using a weighted average of normalization factors at 332.4 and 208.0 keV. The resulting relative efficiency curves for two sample sizes and a 200-mm^2 by 10-mm-deep planar detector are shown in Fig. A-1.

In keeping with our goal of simplicity, we do not attempt to fit the entire relative efficiency curve. Interpolation and extrapolation over limited ranges are used to calculate the needed values.

For samples with ^{241}Pu-^{237}U equilibrium, we calculate needed relative efficiencies as follows. Efficiencies at 152.7 keV (^{238}Pu), 160.3 keV (^{240}Pu), and 161.5 keV (^{239}Pu) are determined by linear interpolation between 148.6 and 164.6 keV. The efficiency at 169.6 keV (^{241}Am) is set equal to that at 171.3 keV. The efficiency for ^{241}Am at 125.3 keV is determined by linear extrapolation from 148.6 and 129.3 keV.

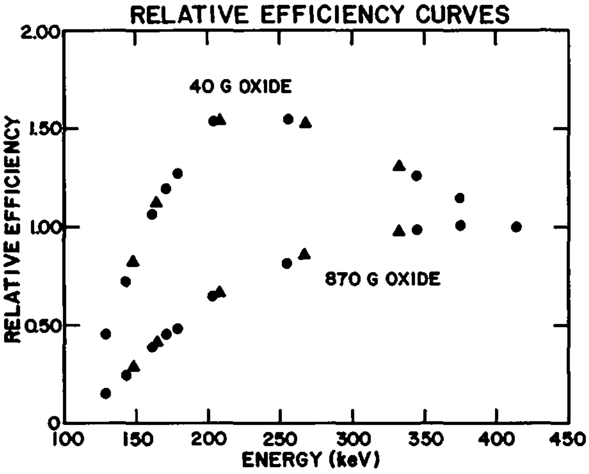

Fig. A-1. Typical relative efficiency curves for two sample sizes using a 200-mm^2 by 10-mm-deep planar detector. Solid circles are points from ^{239}Pu. Triangles are points from ^{241}Pu and ^{241}Pu-^{237}U.

The isotopic ratios for equilibrium (aged) samples are calculated as follows. The ^{238}Pu/^{241}Pu ratio is calculated from clean single lines at 152.7 keV (^{238}Pu) and 148.6 keV (^{241}Pu).

Two values are calculated for the ^{239}Pu/^{241}Pu ratio: first, from the 345.0-keV (^{239}Pu) and 332.4-keV (^{241}Pu-^{237}U) lines and second, from the 203.5-keV (^{239}Pu) and 208.0-keV (^{241}Pu-^{237}U) lines. The final value for the ^{239}Pu/^{241}Pu ratio is determined from a weighted average of the two ratios. The 203/208 ratio gives more precise results for low-burnup plutonium, whereas the 345/332 ratio is better for reactor-grade plutonium.

The ^{240}Pu/^{241}Pu ratio is calculated from the 160.3-keV (^{240}Pu) and 164.6-keV (^{241}Pu-^{237}U) lines. The 160.3-keV ^{240}Pu line is isolated from the 160-keV complex by stripping out the ^{241}Pu component at 160.0 keV with the 164.6-keV ^{241}Pu-^{237}U line and stripping out the small ^{239}Pu component at 160.2 keV with the 161.5-keV ^{239}Pu line.

The ^{241}Am/^{239}Pu ratio is calculated two ways. The first uses the 125.3-keV ^{241}Am line and the 129.3-keV ^{239}Pu line with ^{239}Pu interferences at 124.5 and 125.2 keV being stripped using the 129.3-keV ^{239}Pu line. The second ratio uses the clean single lines at 169.5 keV (^{241}Am) and 171.3 keV (^{239}Pu). The latter ratio is not useful for ^{241}Am concentrations below ~1000 ppm because the 169.5-keV ^{241}Am line is not visible. The 125-keV/129-keV ratio can give ^{241}Am/^{239}Pu ratios down to americium concentrations of a few hundred parts per million. The final ^{241}Am/^{239}Pu ratio is calculated from a weighted average of the two ratios.

For freshly separated samples (no ^{241}Pu-^{237}U equilibrium), all isotopic ratios are calculated with respect to the 148.6-keV ^{241}Pu line. The relative efficiency at 148.6 keV is found from a quadratic fit to the efficiency points at 129.3, 143.4 + 144.2, 164.6, 171.3, 203.5, and 208.0 keV. The ^{238}Pu line at 152.7, the ^{239}Pu line at 129.3, and the ^{240}Pu line at 160.3 keV are used for the three isotopic ratios. The correction for the 160.0-keV ^{241}Pu line is made from the 148.6-keV line. The ^{241}Am is determined as before. Comparing the relative efficiency at 148 keV determined in this fashion with that found by assuming that the plutonium is aged enables one to determine if the sample is in ^{241}Pu-^{237}U equilibrium.

All ratios contain one or more adjustable constants (depending on the algebraic form), which are adjusted using measurements with known standards. These adjustable constants correct for several effects. One effect is possible errors in the branching ratios.[4] Other effects can arise from systematic errors introduced by the specific peak-area analysis and the relative efficiency interpolations and extrapolations used. Typically, adjustments are a few per cent.

The ratios of ^{238}Pu, ^{239}Pu, and ^{240}Pu to ^{241}Pu are converted to weight fractions of those isotopes. The ^{242}Pu fraction is determined by isotopic correlation techniques. The plutonium isotopic weight fractions are then recomputed incorporating the ^{242}Pu value. The ^{241}Am content is computed from the measured ^{241}Am/^{239}Pu ratio and the final ^{239}Pu weight fraction. Statistical precision estimates are propagated from the estimates for the individual isotopic ratios.

APPENDIX B

PLUTONIUM ISOTOPICS SYSTEM SOFTWARE

I. THE SYSTEM

The SRP plutonium isotopics program (LAPIS) and the TA-55 program (MUDPI) run on Digital Equipment Corporation (DEC) PDP-11 series computers as background programs under the RT-11 Version 3B FB monitor. For the SRP system, the operating system and LAPIS program reside on the system floppy disk (DEC-RX02) along with a parameter file used by the program. The TA-55 system uses a hard disk (DEC-RL01) as its system unit. Both systems write spectral data to a second floppy disk. The PDP-11 computer communicates with the Canberra Series 80 multichannel analyzer (MCA) through a Canberra Model 8673 high-speed interface.

The programs are written entirely in FORTRAN (66) and use the FORTRAN-callable routines in the Canberra FT8673 library (Version 1.07) to communicate with the MCA firmware (Version 6.04).

II. USE OF MULTICHANNEL ANALYZER

The Canberra Series 80 MCA has its own firmware responsible for acquiring and storing data in the MCA and controlling the MCA display. The subroutines in the FT8673 library allow the FORTRAN program to talk to the firmware and to invoke some of the functions normally performed at the MCA keyboard: clearing memory, presetting time, setting ROIs, and starting and stopping acquisition. In addition, the subroutines permit copying MCA data to computer memory and obtaining MCA status information.

After the MCA is powered up, the operator must put it into the REMOTE mode using ADC setup dialogue at the MCA keyboard. The REMOTE mode allows the MCA to communicate with the computer. When the program initializes the MCA, the program alerts the operator if the MCA is not in REMOTE and waits. Once the program is able to send commands to the MCA, it clears out any outstanding tasks in the MCA's task list.

During power up, the first 4096 channels of memory are automatically assigned to ADC1. For the TA-55 system, the operator must additionally assign ADC2 to the second 4-k channels in the 16-k MCA memory. These assignments remain in effect until the MCA is powered off or until they are manually changed.

The final operator action is to use the ADC baseline controls to offset the spectrum by 100 channels. These are one-time adjustments that rarely have to be changed.

During routine operation, the operator does not have to interact with the MCA in any fashion. The operator may manipulate the MCA display, if desired, to view spectral data as appropriate. To interrupt the data collection, the operator must get the attention of the program at the console terminal. The program then sends stop commands to the MCA. During data acquisition, the program is constantly polling both the MCA for termination of acquisition and the console terminal for operator input.

The MCA is set to stop acquisition if the contents of any channel exceed 2^{20}. When this is sensed by the program, the overflow is tallied in computer memory and acquisition is restarted. The program can accept overflows in up to 50 different channels.

III. OPERATOR INTERFACE

Communication with the operator is designed to be as simple and error free as possible, consistent with the required input. To command the program, the operator types one-letter commands from a menu of options. The commands are designed to be suggestive of the operation as much as possible. The operator is prompted if decimal points are required in the input.

Operator responses are checked for validity. There are no default answers. A YES/NO question must be answered by 'Y' or 'N'; any other response causes the question to be repeated. One reason for requiring specific responses is that the RT-11 type-ahead feature causes havoc if an operator repeatedly hits the RETURN key. The program then stacks up input lines and uses default values if allowed.

The operator dialogue incorporates escapes for the situation where the operator changes plans or has made a mistake. A negative number entered for the counting time will also return the operator to the options menu.

The program is designed so that the operator specifies all input before the assay and then walks away, returning to view the printed results at the end of the counting time. Data analysis takes about 15 s after acquisition has been completed.

IV. PROGRAM OPTIONS

Program options have been designed so that all routine operations can be performed without exiting from the program. The program options are described briefly below.

A -- Assay

Performs a complete assay sequence, including operator dialogue, data acquisition, data analysis, and output of results. Operator dialogue is somewhat longer than desired in order to incorporate maximum flexibility into the system. An example of the maximum dialogue required is shown below. Sample power and disk recording answers can reduce the total dialogue. Operator responses are underlined.

```
21-APR-83  10:15:03   ENTER OPTION -> A

ASSAY          ---    DETECTOR # ?  -> 1


          ASSAY - DETECTOR # 1

THE CONSTANTS FILE READ WAS: SY:SRP001.CON   WIDE RANGE FOR SRP, 5/6/81
DO YOU WANT TO SELECT ANOTHER CONSTANTS FILE? (Y/N) -> N

EQ = EQUILIBRIUM ASSAY - AGE > 45 DAYS
NE = NON-EQUILIBRIUM ASSAY
UN = UNKNOWN

ENTER ASSAY TYPE (CARRIAGE RETURN FOR ESCAPE) -> EQ
TYPE SAMPLE ID -> SAMPLEID
TYPE REMARK (< 20 CHARACTERS) -> DIALOG DEMO

IS SAMPLE POWER KNOWN? (Y/N) -> Y
ENTER SAMPLE POWER IN WATTS
USE DEC. PT. (CARRIAGE RETURN FOR ESCAPE) -> 4.2357
ENTER DATE OF POWER MEASUREMENT (MM/DD/YY) -> 04/16/83
PRESET TIME (SECS) PER CYCLE   (USE DEC. PT.)  -> 7200.

DETECTOR # 1 - DO YOU WANT TO WRITE SPECTRA TO DISK? (Y/N) -> Y
DEVICE FOR FILES?  -> DY1
FILE NAME? (UP TO 6 CHARS) -> SAMPID
PAUSE -- HIT RETURN WHEN SAMPLE IS READY FOR ASSAY ->

21-APR-83  10:16:22   STARTING ADC # 1   CYCLE    1     7200. SECS

WAITING FOR ADC(S) TO FINISH,  ENTER 'O' FOR NEW OPTION
```

C -- Status

Prints out status of ADCs in multiple detector system, including on/off, memory region assignment, conversion gain, ADC range, digital offset, cycles completed of total requested, elapsed time, preset time, data file requested.

H -- Help

Produces a terminal listing of operator's option menu.

I -- ISOPOW

Runs the ISOPOW code, which calculates decay corrections for plutonium and americium isotopic results.

K -- Disk Directory

Enables operator to obtain directory or free-space listing for any disk in the system without exiting program.

L -- Last Cycle

Enables operator to terminate an autocycle run (see option U) before the requested number of cycles has been completed.

M -- Select Long/Short Listing

Enables operator to select a long or short version of the analysis output. Short form is the default on program startup. An example of the short output is shown below for analysis from disk (option D). The long output includes, in addition, peak areas, errors, americium corrections, relative efficiency, peak positions, widths, and measured isotopic ratios.

N -- Analyze Current Series 80 Data

Analyzes a spectrum already in MCA memory either from a previous acquisition or having been read in from a disk (option R).

O -- Operator Change

Takes operator through new operator identification dialogue to ensure that proper operator identification is output in header block.

S -- Stop Acquisition

Allows operator to stop acquisition during a measurement. Three suboptions allow operator to "abort" and return to Enter Option mode with no analysis, to "terminate" and proceed to normal data analysis before returning to Enter Option mode, or to "suspend"; that is, to stop acquisition and wait for manual resumption.

Q -- Resume Acquisition

Restarts data acquisition after it has been "suspended."

R -- Read Spectrum From Disk

Reads a spectrum from a disk unit into the MCA memory.

I -- Time Check/Change

Allows operator to check or change time and date on system clock.

U -- Autocycle

Enables operator to repeat a measurement a selected number of times without operator intervention between measurements.

```
                       **** EQUILIBRIUM ANALYSIS ****

 DETECTOR # 1  SY:SRP001.CON            WIDE RANGE FOR SRP, 5/6/81

 INPUT FILE:   STD40D.001              DATE:  02-MAY-83
 OPERATOR ID:  TES                     TIME:  14:39:58
 LIVE TIME:     5605.                  CLOCK TIME:      7201.


                PU-238    PU-239    PU-240    PU-241    PU-242    AM-241

  WT. PCT.      0.06711  87.03324  11.58297   1.14327   0.17341   1167.3 UG/GPU
   1 SIGMA      0.00153   0.27597   0.27889   0.00568   0.04335     53.7 UG/GPU
     (%)        2.2772    0.3171    2.4078    0.4967   25.0000    4.6015

  % TOTAL PWR  12.46284  54.97715  26.91865   1.26895   0.00658   4.36582


 SPECIFIC PWR (W/G PU) =  3.05424E-03 +/- 1.38525E-05 ( 0.454%)
  02-MAY-83

 SAMPLE POWER (WATTS)  =  2.66900
  28-APR-83

 PLUTONIUM MASS (G PU) =  874.055 +/-  3.964 ( 0.454%)
  28-APR-83

 PU-240 EFF. FRACTION  =   0.12022 +/- 0.00287 (  2.388%)
```

W -- Write Spectrum to Disk

Writes spectral data from MCA memory to a selected disk unit.

X -- Exit From Program

Returns to RT-11 monitor.

D -- Analyze Disk Data

Analyzes data stored on disk. Data are read from disk directly to computer, by-passing MCA. Can be used for off-line analysis; MCA is not needed in system.

E -- Autocycle Disk Analyses

Analyzes from disk multiple runs stored during an autocycle (option U) acquisition. No operator intervention is required between analyses.

P -- Parameter List/Change

Allows changing or updating of calibration parameters including windows for ROIs and calibration constants. This option involves a submenu that addresses the particular parameters to be changed.

V -- Change Output Device

Allows operator to direct output to line printer if available in system.

V. PROGRAM FILES

The program files residing on the system disk that are required for the TA-55 multiple detector system are

MUDPI.SAV: Analysis program.

CONSTA.DC1: Text file giving list of available constants files to be presented to operator for ADC1.

CONSTA.DC2: Same for ADC2.

Also required is the actual constants file that is selected. This file may be on any available device. The only files used to date are SY:SRP001.CON for analysis and SY:CALIB.CON for calibration analysis (all constants = 1.0).

VI. PROGRAM STRUCTURE

The PDP-11 32-k-word address space is insufficient to contain the entire program and operating system. The 51 subroutines occupy 43.5-k words of memory, resulting in a program that is highly modularized and structured for overlays. The program runs with two overlay levels and takes approximately 21-k words of memory.

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