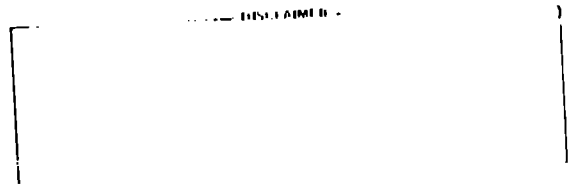# MASTER THE DETERMINATION OF PLUTONIUM ISOTOPIC COMPOSITION BY GAMMA-RAY SPECTROSCOPY

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THE DETERMINATION OF PLUTONIUM ISOTOPIC COMPOSITION BY
GAMMA-RAY SPECTROSCOPY

by

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ABSTRACT

We discuss the general approach, computerized data analysis methods, and
results of measurements used to determine the isotopic composition of
plutonium by gamma-ray spectroscopy. The simple techniques are designed to be
applicable to samples of arbitrary size, geometry, age, chemical, and isotopic
composition. The combination of the gamma spectroscopic measurement of
isotopic composition coupled with calorimetric measurement of total sample
power is shown to give a totally nondestructive determination of sample
plutonium mass with a precision of 0.6" for 1000-g samples of PuO_2 with 12"
^{240}Pu content. The precision of isotopic measurements depends upon many
factors, including sample size, sample geometry, and isotopic content.
Typical ranges are found to be ^{238}Pu, 1 to 10"; ^{239}Pu, 0.1 to 0.5";
^{240}Pu, 2 to 5"; ^{241}Pu, 0.3 to 0.7"; ^{242}Pu (determined by isotopic cor-
relation); and ^{241}Am, 0.2 to 10".

I. INTRODUCTION

Work at the Los Alamos National Laboratory in the area of plutonium iso-
topic analysis by gamma-ray spectroscopy started in 1974 when J. L. Parker and
T. D. Reilly outlined a method using ratios of neighboring peaks with relative

efficiency corrections determined from the spectrum under study.[1] These techniques, using gamma rays in the 120 to 420-keV range, have received wide acceptance and have been put into routine use at several laboratories.[2,3] Results from an interlaboratory comparison have been published.[4] Different laboratories generally use the same techniques with the main difference being the methods used for determining photopeak areas.

A somewhat different approach has been used by Gunnink[5] who fits the complex 100-keV region with response functions of each isotope. This method has been used very successfully for dilute solutions, where attenuation corrections are small.

Gunnink and coworkers,[6,7] and Cowder et al.,[8] have used gamma rays in the 60-keV region for isotopic analysis on freshly separated solutions in the 100 to 300-g/ℓ range. Use of gamma rays in the 60-keV region has also been discussed by Umezawa et al.[9]

Techniques similar to those of Ref. 1 have been described by Dragnev and co-workers,[10-12] and Reilly et al.[13]

The work reported here will enable routine application of this method for verification of plutonium isotopic composition and for determination of plutonium isotopic composition, which is necessary for proper interpretation of total plutonium measurements obtained by calorimetry or neutron coincidence counting.

The procedures to be described require no peak fitting thus minimizing computer core and speed requirements. Little training is needed for the method to be routinely used by technicians. The method represents a simple, and perhaps the most versatile, single detector approach to plutonium isotopics.

II. 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

$$\frac{N_1}{N_2} = \frac{C_1}{C_2} \times \frac{T_{1/2}(1)}{T_{1/2}(2)} \times \frac{B_2}{B_1} \times \frac{RE_2}{RE_1}$$

where

N = number of atoms of the indicated isotope

C = photopeak counts from 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

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.[14,15] 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. ^{239}Pu, ^{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.[16] 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 is used in the data acquisition electronics. This is important to ensure that the peaks don't wander out of their assigned ROIs. The 129.3-keV 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 in order to be able to 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 only limited by count-rate and counting time considerations and can range from ~0.1 g to as large as critic- ality considerations allow. We attempt to keep count rates at about 10 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 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.

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

III. ANALYSIS METHOD

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 com- plexes 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, the two peak areas and two isotopic unknowns are used 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

also 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 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 x 10 mm planar detector are shown in Fig. 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 relative efficiency values.

For samples with ^{241}Pu–^{237}U equilibrium, we calculate needed relative efficiencies as follows. Efficiencies at 152.7 (^{238}Pu), 160.3 (^{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. The efficiency for ^{241}Am at 125.3 keV is determined by linear extrapolation from 148.6 and 129.3 keV.

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 Pu while the 345/332 ratio is better for reactor grade Pu.

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 below ^{241}Am concentrations of about 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 a few hundred ppm americium concentrations. 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. ^{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.

IV. CALIBRATION

The basic expression for isotopic ratios from gamma spectroscopy (Eq. 1) produces results using published fundamental constants that, in principle, can be used without recourse to standards. We have incorporated adjustable calibration constants into these ratios to adjust for branching ratio errors and biases in peak-area and relative efficiency algorithms. By calibration we mean the adjustment of these constants to give the best agreement with measurements on "known standards."

Obtaining known standards is more difficult than it may seem. There are problems with even the National Bureau of Standards reference material.[4] The determination of ^{241}Am concentrations in many of the samples available to us is not as precise as desired. We find that the best ^{241}Am values, for our purposes, are obtained by allowing the ^{241}Am to grow into a high ^{241}Pu content sample and calculating the ^{241}Am knowing the half-lives and chemical separation date.

The constants are adjusted by comparing the measured isotopic ratios determined from repeated high-precision, long runs with those ratios determined from the best appropriate chemical techniques. Such calibration results are shown in Fig. 2. Here a wide range of sample sizes and types were counted ranging from several hundred grams to 0.25 g. The ^{240}Pu content ranged from 6 to 18%. The best values for the calibration constants were used to recompute the isotopics. It is the recomputed isotopics, after adjustment of the calibration constants, that are displayed in Fig. 2. The fact that the mean values are not 1.0 illustrates the difficulty in making these adjustments for such a wide range of isotopics, especially for ^{240}Pu which has three terms in its ^{240}Pu/^{241}Pu-ratio expression.

Over a wide range of sample sizes, configurations, and isotopic composition, the isotopics measured by gamma-ray spectroscopy show essentially no bias. Poor alpha spectrometer determinations of ^{238}Pu may contribute to the relatively poor precision observed in Fig. 2 for that isotope. The extent of this contribution is not known.

V. PLUTONIUM MASS MEASUREMENTS WITH CALORIMETRY AND GAMMA-RAY SPECTROSCOPY

The isotopics calibration that produced the results in Fig. 2 was used with gamma-ray spectroscopy measurements of thirteen 1000-g samples of Fast Flux

Test Facility (FFTF) PuO_2 and one 500-g sample of low burn up metal. All samples were also measured in a calorimeter. The plutonium isotopic composition determined from the gamma-ray measurement was used to determine the sample's specific power in W/g plutonium, which was combined with the calorimeter measurement of sample power in watts to give the sample mass in grams of plutonium.[14] The results for the plutonium mass are compared with the accepted values determined by destructive chemical analysis for g plutonium/g sample, ignition for moisture content, weighing, and mass spectrometry for isotopic composition.

This combination technique is not new, having been in use at Mound Laboratory[18] and Rockwell-Hanford[3] for some time. This is, however, the first application of this powerful combination of NDA techniques at Los Alamos.

The PuO_2 had nominal isotopic composition of 0.06% ^{238}Pu, 86.6% ^{239}Pu, 11.8% ^{240}Pu, 1.3% ^{241}Pu, 0.2% ^{242}Pu, and 600 ppm ^{241}Am. The low burnup metal's isotopic as determined by mass spectrometry was 0.04% ^{238}Pu, 93.8% ^{239}Pu, 5.9% ^{240}Pu, 0.27% ^{241}Pu, 0.03% ^{242}Pu, and 360 ppm americum (radiochemically measured).

In Table I the 4-h gamma-ray isotopic measurements are presented as ratios to the mass spectrometry values. Agreement is seen to be excellent for the major plutonium isotopes with the exception of ^{238}Pu. Here, ^{238}U contamination is always a problem for mass spectrometry and can produce large errors at these relatively low concentrations (0.05 to 0.07%). The gamma spectroscopy results are more reliable than mass spectrometry measurements of ^{238}Pu in general. The bias in the ^{242}Pu results arises from the isotopic correlation technique used. Over a narrow burnup range the bias may be substantially reduced by choosing a different correlation. In any case, this bias is not significant for total plutonium determination because ^{242}Pu contributes negligibly to the plutonium specific power.

The relatively large bias and standard deviation for ^{241}Am is caused by the poor statistical precision of the gamma spectroscopy measurement at these lower americium levels (300 to 700 ppm) and the imprecision of the radio-analytical techniques used to determine the "standard" values. For these low americium content samples, the americium contributes only a minor portion (1 to 2%) of the sample power. The gamma-ray measurement precision improves for higher americium concentrations, which can contribute significantly to the sample power.

The gamma-spectrometer isotopics were used to interpret a calorimetry measurement on each sample. The calorimetry-gamma spectroscopy comparison with chemical analysis is shown graphically in Fig. 3. The observed precision of 0.6% for the plutonium mass determination comparison in Fig. 3 also contains a contribution from the precision of the weighing and destructive chemical analysis techniques used to determine the "accepted" chemical analysis values. Experience indicates that this contribution may be 0.1 to 0.2%.[18]

In Table II we summarize some of the precisions that have been observed on measurements of three different sample types. The first entries for FFTF oxide summarize the measurements already discussed. The other two entries (reactor-grade oxide and low-burnup oxide) summarize the results of repeated measurements of the specific power from gamma-ray isotopics. These two samples did not have enough sample mass (a few grams) or a precise enough plutonium mass value to enable them to be measured in our calorimeter. Larger samples than these would give poorer precision because of the increased effect of the Compton continuum in the 100 to 200-keV region arising from the 300 to 400-keV plutonium gamma rays.

The Table II entries illustrate how the precision of the specific power can vary, especially as a function of the isotopic composition. For low americium samples the uncertainty in the ^{240}Pu isotopic dominates the predicted precision. As the americium content increases, the ^{240}Pu power contribution becomes less important and results in improved overall precision.

VI. APPLICATION TO URANIUM-PLUTONIUM MIXED OXIDES AND MATERIALS WITH FISSION PRODUCT CONTENT

We have made measurements on uranium-plutonium mixed oxides to determine if the ^{235}U gamma rays at 163.4, 202.1, and 205.3 keV interfere with the ^{240}Pu/^{241}Pu measurement at 160.2/164.6 keV and the ^{239}Pu/^{241}Pu ratio measured at 203.5/ 208.0 keV. With proper selection of regions of interest one can perform unbiased measurements of the ^{240}Pu/^{241}Pu and ^{239}Pu/^{241}Pu ratios on mixed oxides up to ^{235}U/plutonium ratios of 0.3 for the ^{239}Pu/^{241}Pu ratio and 3.0 for the ^{240}Pu/^{241}Pu ratio. For comparison we note that mixed oxides with natural uranium:plutonium ratios of

3 or 4 to 1 (typical of US fast breeder reactor fuel) will give a
^{235}U/plutonium ratio in the range of 0.02 to 0.03, well within the range of
this technique.

Additional measurements have been made on plutonium samples plus added
^{137}Cs to mockup fission product activity. The measurement of plutonium
isotopic ratios in the 100 to 200-keV region suffers from the increased
Compton continuum arising from 600 to 800-keV fission-product gamma rays. The
degredation is enough to make ^{238}Pu/^{241}Pu and ^{240}Pu/^{241}Pu measurements
on low burnup plutonium (~6% ^{240}Pu) not meaningful above fission product
concentrations of about 10 μCi/g Pu. For higher burnup plutonium these
measurements can tolerate fission product concentrations of around 20 μCi/g Pu.

REFERENCES

1. J. L. Parker and T. D. Reilly, "Plutonium Isotopic Determination by Gamma-Ray Spectroscopy," in "Nuclear Analysis Research and Development Program Status Report," Jan. – April 1974, Los Alamos National Laboratory report LA-5675-PR (August 1974).

2. J. G. Fleissner, J. F. Lemming, and J. Y. Jarvis, "Study of a Two-Detector Method for Measuring Plutonium Isotopics," Proc. Am. Nucl. Soc. Conf. on Measurement Technology for Safeguards and Materials Control, Kiawah Island, S.C., November 26-29, 1979.

3. R. A. Hamilton, Atlantic Richfield Hanford Co., Letter and enclosures to W. W. Rodenburg, Mound Laboratory (February 18, 1977).

4. H. Ottmar and E. Eberle, "Determination of Plutonium Isotopic Composition by Gamma Spectrometry: Results from Interlaboratory Comparison Measurements Organized by ESARDA," 1st Annual ESARDA Symposium on Safeguards and Nuclear Materials Management, April 10, 1979. See also INMM Journal, VIII (1979).

5. R. Gunnink, J. B. Niday, and P. D. Siemens, "A System for Plutonium Analysis by Gamma-Ray Spectrometry, Part I: Techniques for Analysis of Solutions," Lawrence Livermore Laboratory report UCRL-51577 (April 1974).

6. R. Gunnink and J. E. Evans, "In-Line Measurement of Total and Isotopic Plutonium Concentrations by Gamma-Ray Spectrometry," Lawrence Livermore Laboratory report UCRL-52220 (1977).

7. R. Gunnink, A. L. Prindle, J. B. Niday, A. L. VanLehn, and Y. Asakura, "TASTEX Gamma Spectrometer System for Measuring Isotopic and Total Plutonium Concentrations in Solutions," Journal INMM, VIII (1979), p. 429. See also Lawrence Livermore Laboratory report UCRL-82335 (1979).

8. L. R. Cowder, S.-T. Hsue, S. S. Johnson, J. L. Parker, P. A. Russo, J. K. Sprinkle, Y. Asakura, T. Fukuda, and I. Kondo, "Gamma-Ray NDA Assay System for Total Plutonium and Isotopics in Plutonium Product Solutions," Proc. ANS Topical Conf. on Measurement Technology for Safeguards and Materials Control, Kiawah Island, S.C., November 26-29, 1979.

9. H. Umezawa, T. Suzuki, and S. Ichikawa, "Gamma-Ray Spectrometic Determination of Isotopic Ratios of Plutonium," J. Nucl. Sci. Technology, 13, 327 (1976).

10. T. N. Dragnev, "Intrinsic Self-Calibration of Non-Destructive Gamma Spectrometric Measurements. Determination of U, Pu, and ^{241}Am Isotopic Ratios," J. Radioanal. Chem., 36, 491 (1977).

11. T. Dragnev and K. Scharf, "Non-Destructive Gamma Spectrometry Measurement of ^{238}Pu/^{240}Pu and Pu/^{240}Pu Ratios," Int. J. Appl. Radiat. Isot., 26, 125 (1975).

12. T. N. Dragnev and B. P. Damjanov, "Methods for Precise Absolute Gamma-Spectrometic Measurements of Uranium and Plutonium Isotopic Ratios," Nuclear Safeguards Technology 1978, Vol. 1, IAEA, Vienna, Austria, (1979), p. 739.

13. T. D. Reilly, D. D'Adamo, I. Nessler, and M. Cuypers, "Gamma-Ray Spectrometric Determination of Plutonium Isotopic Composition," Nuclear Safeguards Technology 1979, 1, IAEA, Vienna, Austria (1979), p. 727.

14. "American National Standard Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," American National Standards Institute N15.22-1975 (June 1975).

15. R. Gunnink, J. E. Evans, and A. L. Prindle, "A Reevaluation of the Gamma-Ray Energies and Absolute Branching Intensities of ^{237}U, $^{238,239, 240,241}Pu$, and ^{241}Am," Lawrence Livermore Laboratory report UCRL- 52139 (October 1976).

16. R. H. Augustson and T. D. Reilly, "Fundamentals of Passive Nondestructive Assay of Fissionable Materials," Los Alamos National Laboratory report LA-5651-M (September 1974).

17. M. S. Krick, "^{240}Pu-Effective Mass Formula for Coincidence Counting of Plutonium with SR Electronics, in "Nuclear Safeguards Research and Development Program Status Report," May-August 1977, Los Alamos National Laboratory report LA-7030-PR (March 1978).

18. W. W. Rodenburg, "An Evaluation of the Use of Calorimetry for Shipper-Receiver Measurements of Plutonium," Monsanto Research Corporation, Mound Laboratory report MLM-2518 (1978).

FIGURE CAPTIONS

Fig. 1   Typical relative efficiency curves for two sample sizes using a
              200-mm^2 x 10-mm deep planar detector. Solid circles are
              points from ^{239}Pu. Triangles are points from ^{241}Pu and
              $^{241}Pu\text{-}^{237}U$.
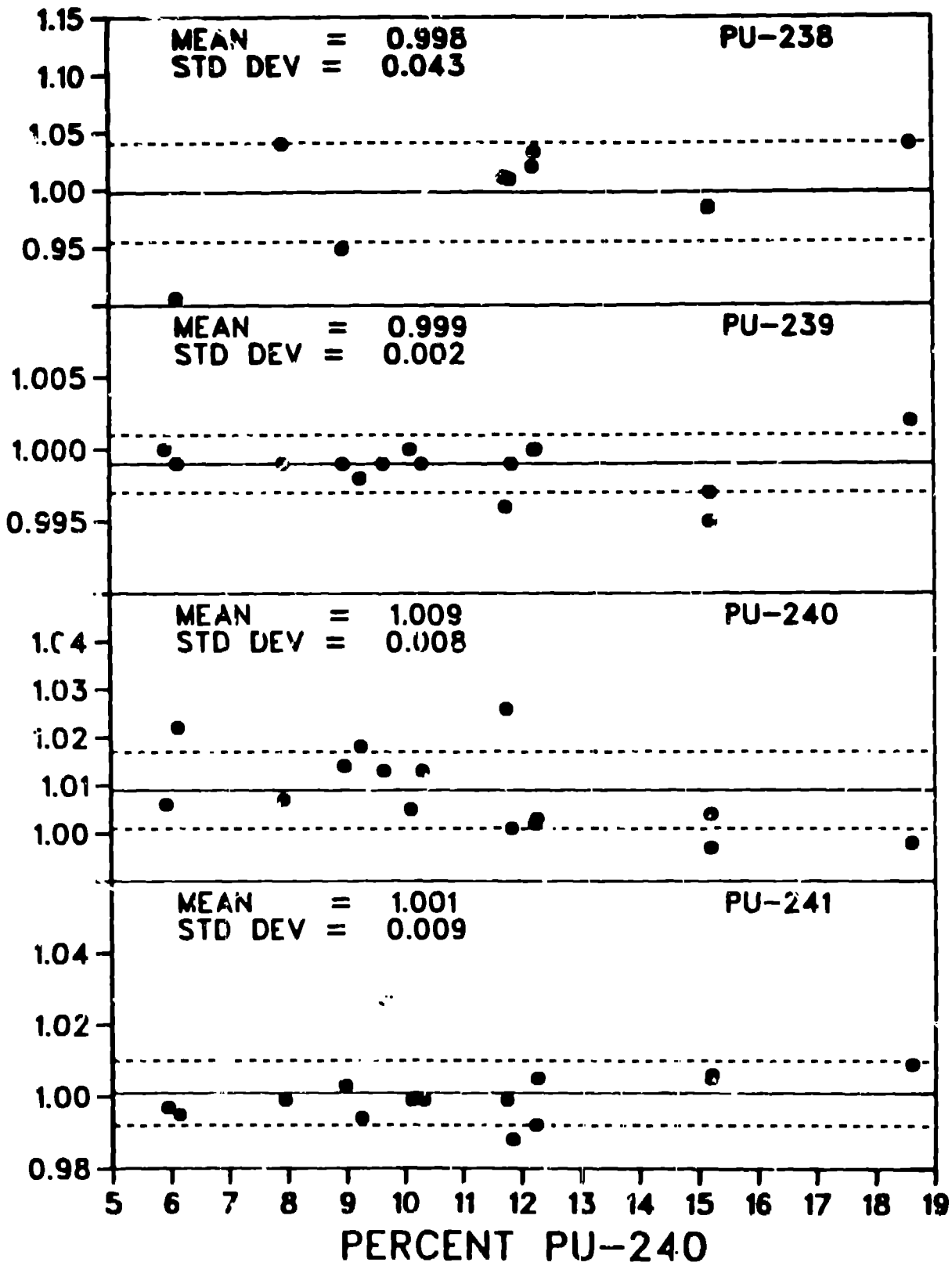
Fig. 2   Comparison of measured plutonium isotopic with mass spectrometry.

Fig. 3   Total plutonium determination by combination of calorimetry and
              gamma spectroscopy compared to conventional chemistry tech-
              niques. Error bars are predicted uncertainties arising from
              gamma-ray isotopic determination.

TABLE 1

PLUTONIUM ISOTOPIC BY GAMMA-RAY SPECTROSCOPY

Sample	238Pu	239Pu	Ratio: 240Pu	$\frac{\text{Gamma Spectroscopy (4 h)}}{\text{Mass Spectroscopy *}}$ 241Pu	242Pu	241Am
Plutonium Oxide:						
LA0134C1	0.8738	0.9936	1.0090	0.9934	1.171	0.994
LA0134C2	0.8486	1.0022	0.9825	0.9986	1.138	0.932
LA0134C3	0.8850	0.9998	1.0000	0.9954	1.159	1.036
LA0134C4	0.8810	0.9992	1.0042	0.9965	1.167	1.003
LA0134C5	0.8292	0.9975	1.0169	0.9946	1.184	0.989
LA0134C6	0.8746	0.9972	1.0191	0.9943	1.187	1.037
LA0134C8	0.9052	0.9987	1.0074	0.9996	1.173	1.117
LA0135CB	0.9729	0.9945	1.0398	0.9998	1.129	1.047
LA0146C3	0.8990	0.9965	1.0243	0.9945	1.169	0.843
LA0148C3	0.7331	1.0077	0.9402	1.0122	1.200	1.239
LA0153C3	0.8149	0.9955	1.0324	1.0071	1.091	1.239
LA0154C3	0.9152	1.0012	0.9951	0.9991	1.229	1.221
LA0150C3	0.9587	0.9964	1.0231	0.9942	1.224	1.031
Low Burnup Metal:						
J001325	0.3437**	1.0014	0.9844	0.9955	0.689**	1.114
mean (bias)	0.8762	0.9990	1.0056	0.9982	1.171	1.060
observed std dev	0.062	0.0034	0.025	0.0054	0.037	0.116

*americum determined by radioanalytical techniques

**omitted from average

TABLE II

PRECISION OF DETERMINAT'ON OF SPECIFIC POWER
USING GAMMA-RAY SPECTROSCOPY

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Am
FFTF Oxide (Low Am)						
Typical Isotopic (wt")	0.061	86.58	11.79	1.35	0.20	595 ppm
Typical Isotopic Precision						
4-h Measurement (")	3.5	0.38	2.8	0.57	--	6.3
Per cent Total Power	11./	56.3	28.2	1.5	0.01	2.3

Specific Power ː 0.00298 W/g Pu

Observed Precision of Specific Power: Thirteen 4-h Measurements = 0.6"

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Am
Reactor-Grade Oxide (High Am)						
Typical Isotopic (wt")	0.24	84.10	12.36	2.77	0.53	16 000ppm
Typical Isotopic Precision						
7.4-h Measurement (:)	0.70	0.24	1.70	0.54	--	0.15
Per cent Total Power	23.6	28.3	14.6	1.6	0.01	31.9

Specific Power = 0.00577 W/g Pu

Observed Precision of Specific Power: Nine 7.4-h Measurements = 0.15"

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Am
Low-Burnup Oxide (Low Am)						
Typical Isotopic (wt")	0.014	93.34	6.30	0.32	0.026	189 ppm
Typical Isotopic Precision						
5.9-h Measurement (:)	5.0	0.14	2.2	0.31	...	5.5
Per cent Total Power	3.3	76.5	18.7	0.46	0.001	1.07

Specific Power = 0.00235 W/g Pu

Observed Precision of Specific Power: Fourteen 5.9-h Measurements - 0.23"
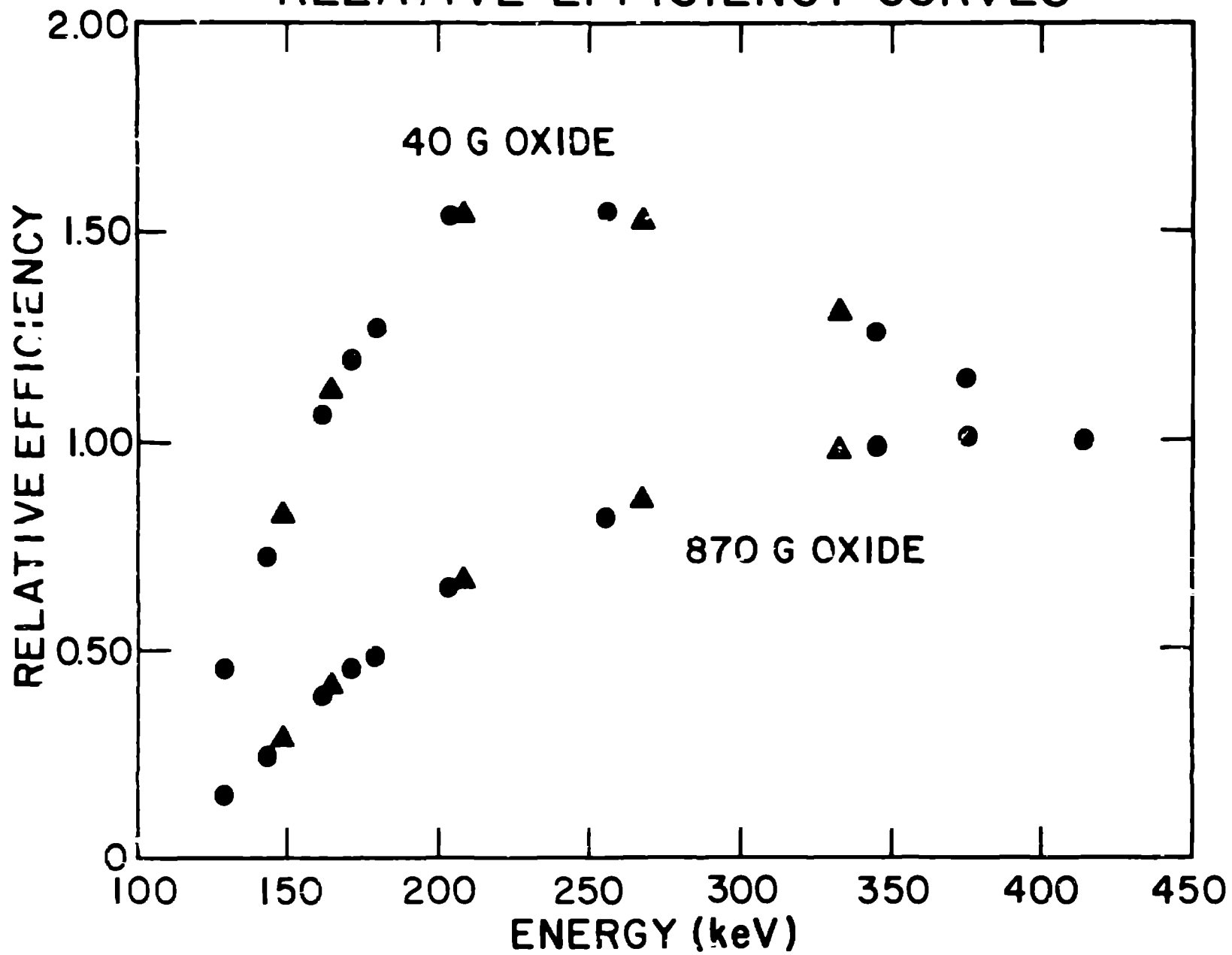
RELATIVE EFFICIENCY CURVES

40 G OXIDE

870 G OXIDE

Fig. 2   Comparison of measured plutonium isotopic with mass spectrometry.