MLM-2336(OP)

CONF-760615-11

GAMMA-RAY ISOTOPIC MEASUREMENTS FOR ASSAY OF PLUTONIUM FUELS

Francis X. Haas and John F. Lemming

Monsanto Research Corporation
Mound Laboratory*
Miamisburg, Ohio

Presented at the
Seventeenth Annual Meeting of the
Institute of Nuclear Materials Management
Seattle, Washington
June 22-25, 1976

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Energy Research and Development Administration under Contract No. E-33-1-GEN-53.

DISCLAIMER

GAMMA-RAY ISOTOPIC MEASUREMENTS FOR ASSAY OF PLUTONIUM FUELS

Francis X. Haas and John F. Lemming
Monsanto Research Corporation
Mound Laboratory*
Miamisburg, Ohio

ABSTRACT

The data presented in this paper summarizes our experience in obtaining plutonium and americium isotopic data using two systems: a 70 cc Ge(Li) system with 1.8 keV resolution at 1332 keV for acquisition of spectra up to 700 keV gamma-ray energy and a 1 cc planar system with 600 eV resolution at 122 keV for the acquisition of spectra up to 220 keV. Plutonium samples include ash samples with plutonium-240 concentrations of 9 to 20%; plutonium metal samples 6% - plutonium-240; ZPPR fuel pins; and various waste categories of 6% - plutonium-240 material. Nondestructive isotopic ratios obtained by gamma-ray spectroscopy are compared to chemical data obtained from these samples.

INTRODUCTION

The nondestructive assay of plutonium is important as a safeguard tool in accounting for stategic nuclear material. Several nondestructive assay techniques require a knowledge of plutonium and americium isotopic ratios to convert their raw data to total grams of plutonium. Calorimetry requires a complete knowledge of the heat producing isotopes. In addition, it must account for the decay of plutonium-241 and the ingrowth of americium-241. Spontaneous fission detectors (SFD) require a knowledge of the even isotopes of plutonium, i.e., plutonium-238, -240, -242. This paper describes the results of obtaining the isotopic ratios of plutonium-238, plutonium-240, plutonium-241 and americium-241 relative to plutonium-239 using high resolution gamma-ray spectroscopy.

The objectives of the technique are fourfold: 1) It is to be completely nondestructive. Analysis is to be performed on the material in the same container as used in the calorimeter or SFD; 2) It is applicable to bulk material including scrap and waste; 3) It is relatively fast. It should easily surpass all chemical methods of analysis and should approach the speed with which the samples can be analyzed in the calorimeter or SFD; and 4) The overall accuracy should be less than $\pm$ 3% for each isotope. In the case of calorimetry for FBR plutonium material, these accuracies would give an overall uncertainty of less than 1% in the plutonium analysis with

*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Energy Research and Development Administration under Contract E-33-1-GEN-53.

a calorimetry uncertainty of 0.25%.

INSTRUMENTATION

Two detector systems are used in our work[1]. Spectrometer A contains a 70 cc Ge(Li) detector with 1.8 keV resolution at 1332 keV with 14% efficiency. Spectrometer B contains a 1 cc Ge(Li) planar detector with 600 eV resolution at 122 keV. For the work reported, count rates of 3000 counts/sec were maintained for both systems. Bulk samples are rotated while acquiring data to reduce the effects of inhomogeneities due to the distribution of plutonium in the sample. A lead slit collimator is used to reduce the response to material located near the edge of the container. It allows the detector to view the sample only along a diameter and improve the signal-to-background ratio by absorbing photons which are Compton scattered in the sample. A 0.030 inch (0.8 mm) cadmium abosrber is used to further reduce the effects of low energy gamma rays.

TECHNIQUE

Figures 1 and 2 show a gamma-ray spectrum of a high fissile ash sample acquired using spectrometers A and B, respectively, showing the gamma-ray peaks of interest. Figure 3 shows the energy region at 600 keV obtained with spectrometer A. The basis for the nondestructive measurement of the plutonium isotopic ratios using gamma-ray spectroscopy involves the analysis of a spectral group whose members belong to different isotopes. Ratios of the areas of the neighboring gamma-ray peaks are related to the isotopic abundance ratios by the expression

$$\frac{I_i}{I_j} = \frac{N_i \lambda_i B_i \varepsilon_i S_i}{N_j \lambda_j B_j \varepsilon_j S_j}$$

where I, N, λ, B, ε, and S are the measured peak area, the number of nuclei, the nuclear decay constant, the absolute branching intensity, the detector efficiency and the self-absorption for gamma rays with energies E_i and E_j from isotopes i and j, respectively. By selecting gamma rays whose energies differ by less than 10 keV, the ratios of the relative efficiencies ($\varepsilon_i S_i / \varepsilon_j S_j$) is assumed to be 1. The ratio for the isotopic values then reduces to the form

$$\frac{N_i}{N_j} = K \frac{I_i}{I_j},$$

where K is calculated using nuclear decay constants. The gamma-ray peaks and the values for the conversion factors are listed in Table I.

RESULTS

A. Routine Analysis

The accuracy of nondestructive techniques can be established by comparing the results on aliquots with their subsequent chemical analysis. Spectrometer A is used for "routine" analysis of samples received as part of a Plutonium Metal Exchange Program and the Plutonium Verification Program[2]. Isotopic values are obtained from data in a single spectrum. The 640 keV and 660 keV regions are used for plutonium-240 and americium-241 analysis, respectively.

Over the past three years, we have compared the nondestructive (NDA) and chemical analysis (DA) results for forty-five 9-g plutonium metal samples (93% plutonium-239). The results of this comparison are shown in Table II. Column 2 shows the average percent differences of the two techniques.

The bulk samples investigated come from the Plutonium Verification Program[2]. Samples are selected at random by the Albuquerque Operations Office of the U. S. Energy Research and Development Administration and sent to Mound Laboratory for calorimetric assay. Sample weights have varied from 20-g to several kilograms of plutonium in various matrix materials. Analytical aliquots are chosen from selected samples prior to shipment to Mound. Because of the overall time commitments of the program, samples are counted for only four hours. In the most recent sample exchange, 20 aliquots and 79 samples were processed. The results of the analysis are shown in Table III. Column 2 compares the results of the analysis on the aliquots and Column 3 compares the gamma-ray analysis of the bulk sample to the aliquots.

The agreement between the NDA and DA for the data in these two tables is good for plutonium-241 and americium-241. The large uncertainties on the plutonium-238/plutonium-239 ratios is due to the weak intensity of the 153 keV peak at the 100 ppm level in the 93% plutonium-239 material. The large uncertainty on the plutonium-240 arises from poor counting statistics due to the weak intensity of the 600 keV peaks, which are two to three orders of magnitude less in intensity than the peaks in the 100 to 200 keV region, and the four-hour data acquisition time. If we were to use uncertainties of 15% for plutonium-238/plutonium-239 and plutonium-240/plutonium-239, 3% for plutonium-241/plutonium-239 and 7% for americium-241/plutonium-239, the resulting uncertainty in the calorimetric assay would be 3%. However, for the Verification Program, isotopic ratios and their uncertainties from chemical analysis are used for the plutonium-238 and plutonium-240 values to give assay uncertainties of less than 1%. Gamma-ray isotopic data is used for plutonium-241 and americium-241 and to verify that the aliquot is representative of the bulk sample.

We have extended these techniques to analyze five reactor fuel pins containing 86% plutonium-239. Table IV shows the comparison of the gamma-ray analysis of the pins with chemical analysis of the fuel at the time of fabrication. Counting times of eight hours were used for spectrum acquisition. The use of gamma-ray isotopic data in a calorimetric assay would yield total plutonium values which are 0.43% higher than corresponding values using chemical isotopics.

B. Low Energy Analysis

Spectrometer B has been used to develop areas of analysis for the isotopic values in the energy region from 120 to 220 keV. This is the high count rate region of the spectrum and could yield all the isotopic data in a shorter period of time.

Table V shows the improvement in analyzing the 160 keV region for plutonium-240/plutonium-239 using spectrometer B. Table VI compares the values obtained for americium-241 isotopic data using the 125 keV/129 keV and 169 keV/ 171 keV peaks. With the improvement in plutonium-240 isotopic data at 160 keV and the equivalence of using the americium-241 data at 125 keV, a series of spectra were acquired using 100-g ash samples of high fissile, FBR and LWR material. Ten spectra were acquired for acquisition times of 1000, 2000, 3000, 4000, 10000, 20000 and 40000 seconds for each sample. Figure 4 shows graphically the ratio of the gamma-ray data to the chemical data supplied with the samples. The plutonium-238 values are 25% below the chemical values for this sample and this large difference is assumed, based on our previous experience with high fissile material, to arise from incorrect chemical values for the plutonium-238. This difference is being investigated further. For the other samples, the difference is -5% for FBR and +1% for LWR material. The differences for plutonium-240 values are -5% for high fissile material and -10% for FBR and LWR material. Plutonium-241 values are within 2% for all categories.

Recognizing that these differences exist, one might correct for them by comparing data to a well characterized standard of the same material category or possibly by reexamination of the nuclear decay constants. This data does show, however, the precision which can be obtained at the counting times chosen. Figure 5 shows the plot of the precision at one sigma, based on counting statistics alone, which can be obtained under the experimental arrangements described. The dashed line at 3% indicates the goal line we have set for our objective. This objective cannot be reached for plutonium-238 for high fissile material even after 40,000 seconds. It can be reached for plutonium-238 after 10,000 seconds for FBR material and after 4,000 seconds for LWR. The 3% objective for plutonium-240 can be reached after about 20,000 seconds and plutonium-241 after 3,000 seconds for these material types.

CONCLUSION

Gamma-ray isotopic measurements are reliable techniques for nondestructively characterizing plutonium material. The large volume detector can be used to obtain the necessary isotopic data using gamma rays from 140 keV to 660 keV. This technique is in routine use in Mound's Verification Program. With Spectrometer B, the energy region from 125-208 keV is viable because it decreases the acquisition time in obtaining the necessary isotopic data.

Another experimental arrangement would be to use a two detector system looking at both energy regions. Spectrometer B would yield the plutonium-238 and plutonium-241 data; spectrometer A, with the introduction of lead absorbers to eliminate the low energy portion of the spectrum, would yield the plutonium-240 and americium-241 in a shorter time than required for obtaining all of the isotopic data from the same spectrum.

REFERENCES

1. Mound Laboratory Nuclear Safeguards Progress Reports MLM-2302 (April 2, 1976), MLM-2286 (December 9, 1975), MLM-2186 (December 27, 1974).

2. Raymond S. George and Roy B. Crouch, "Inspector Measurement Verification Activities", Jour. INMM IV, No. III, (1975), p. 327.

LIST OF FIGURES

TABLE I

GAMMA RAYS AND CONVERSION FACTORS USED FOR ISOTOPIC DETERMINATIONS

Isotopic Ratio	Gamma Rays	Conversion Factors to Express Ratio in ppm
$\frac{^{238}Pu}{^{239}Pu}$	$\frac{153\ keV}{144\ keV}$	$1010\ \frac{I_{153}}{I_{144}}$
$\frac{^{238}Pu}{^{241}Pu}$	$\frac{153\ keV}{148\ keV}$	$1.185\ \frac{I_{153}}{I_{148}}$
$\frac{^{240}Pu}{^{239}Pu}$	$\frac{642\ keV}{640\ keV}$	$148300\ \frac{I_{642}}{I_{640}}$
$\frac{^{240}Pu}{^{241}Pu}$	$\frac{160.35\ keV}{164.6\ keV}$	$50.7\ \frac{I_{160.35}}{I_{164.6}}$
$\frac{^{241}Pu}{^{239}Pu}$	$\frac{208\ keV}{203\ keV}$	$623.7\ \frac{I_{208}-.0373I_{662}}{I_{203}\qquad I_{659}}$
$\frac{^{241}Pu}{^{239}Pu}$	$\frac{148\ keV}{144\ keV}$	$852.7\ \frac{I_{148}}{I_{144}}$
$\frac{^{241}Am}{^{239}Pu}$	$\frac{662\ keV}{659\ keV}$	$487.5\ \frac{I_{662}}{I_{659}}$
	$\frac{125\ keV}{129\ keV}$	$27880\ \frac{I_{125}-540}{I_{129}}$

Table II

COMPARISON OF ISOTOPIC DATA
METAL EXCHANGE SAMPLES
(93%, ^{239}Pu)

ISOTOPE	NDA-DA / DA
238-Pu	1.0±11.7%
240-Pu	−5.2±6.4
241-Pu	4.2±5.7
241-Am	−2.9±4.9

Table III
COMPARISON OF RESULTS
PLUTONIUM VERIFICATION PROGRAM

ISOTOPE	NDA-DA / DA ALIQUOTS	BULK SAMPLE / ALIQUOTS
238-Pu	1±7%	1±14%
240-Pu	−7±11	1±18
241-Pu	−2±3	1±2
241-Am	1±7	1±7

Table IV
COMPARISON OF ISOTOPIC DATA
ZPPR FUEL PINS

ISOTOPE	CHEMICAL VALUE (ppm)	GAMMA-RAY VALUE (ppm)	DIFFERENCE (%)
238-Pu	523	550±40	5.2
240-Pu	131990	131920±7700	−0.8
241-Pu	16170	16210±230	0.2
241-Am	4470	4540±100	−5.1
Specific Power (mW/g)	3.245	3.231	−.43

TABLE VI

COMPARISON OF $^{241}AM/^{239}PU$ ISOTOPIC DATA FOR LISTED PEAK AREAS RELATIVE TO 662-KEV/659-KEV VALUE

SAMPLE	SPECTRA	125-KEV/129-KEV	169-KEV/171-KEV
SRM-946	10	0.98 ± 0.01	0.99 ± 0.08
SRM-947	10	1.02 ± 0.01	1.00 ± 0.08
SRM-948	10	0.99 ± 0.02	1.00 ± 0.13
VERIFICATION ALIQUOTS	20	0.97 ± 0.04	

TABLE V

AVERAGE RATIOS OF GAMMA-RAY TO MASS SPECTROSCOPY VALUES FOR ^{240}PU CONCENTRATIONS USING 160-KEV PEAKS

SAMPLE	NUMBER OF SPECTRA	DETECTOR	AVERAGE RATIO NDA/DA
METAL	25	A	0.80 ± 0.15
EXCHANGE	8	B	0.94 ± 0.04
VERIFICATION	20	A	0.70 ± 0.13
ALIQUOTS	15	B	0.95 ± 0.07

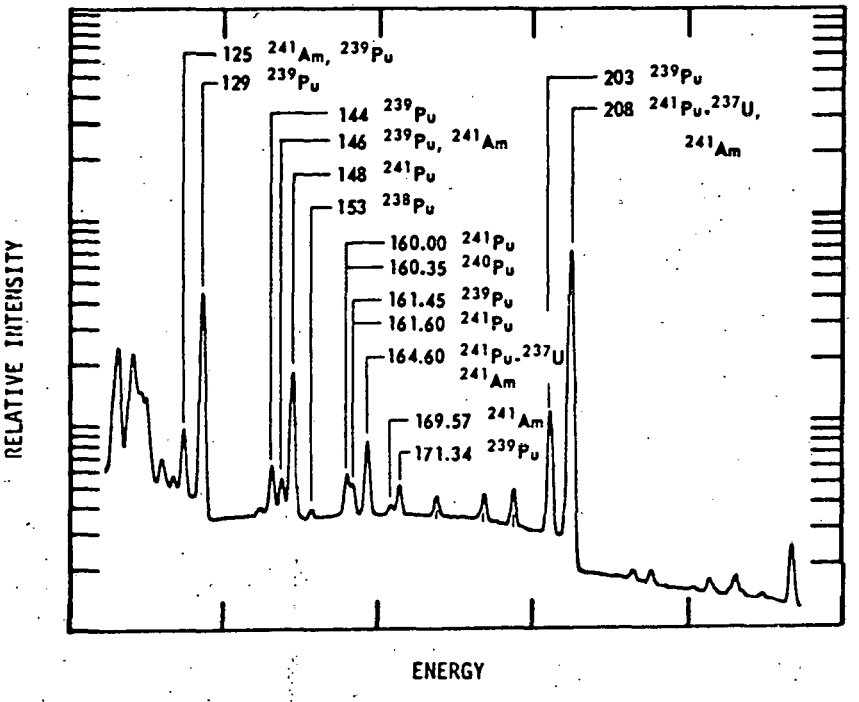
125 ^{241}Am, ^{239}Pu
129 ^{239}Pu
144 ^{239}Pu
146 ^{239}Pu, ^{241}Am
148 ^{241}Pu
153 ^{238}Pu
160.00 ^{241}Pu
160.35 ^{240}Pu
161.45 ^{239}Pu
161.60 ^{241}Pu
164.60 ^{241}Pu-^{237}U
^{241}Am
169.57 ^{241}Am
171.34 ^{239}Pu
203 ^{239}Pu
208 ^{241}Pu-^{237}U,
^{241}Am
RELATIVE INTENSITY
ENERGY

Figure 1

Fig. 2

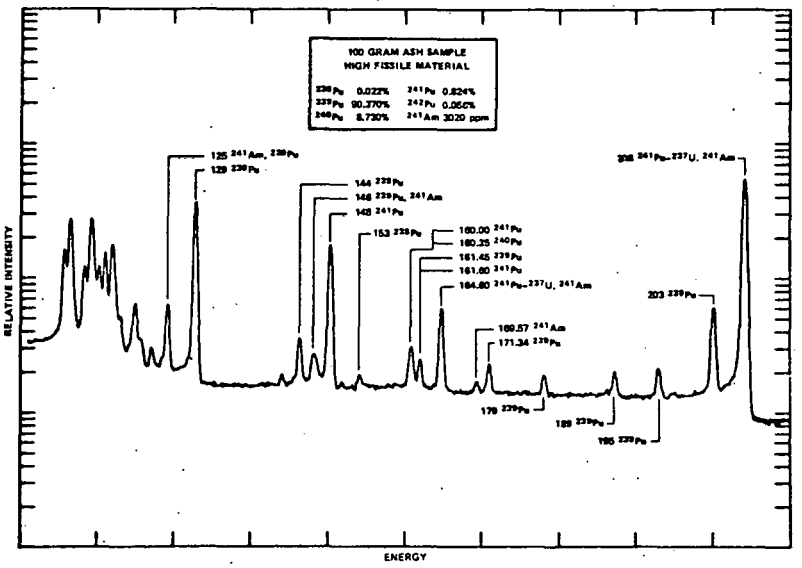

Figure 2

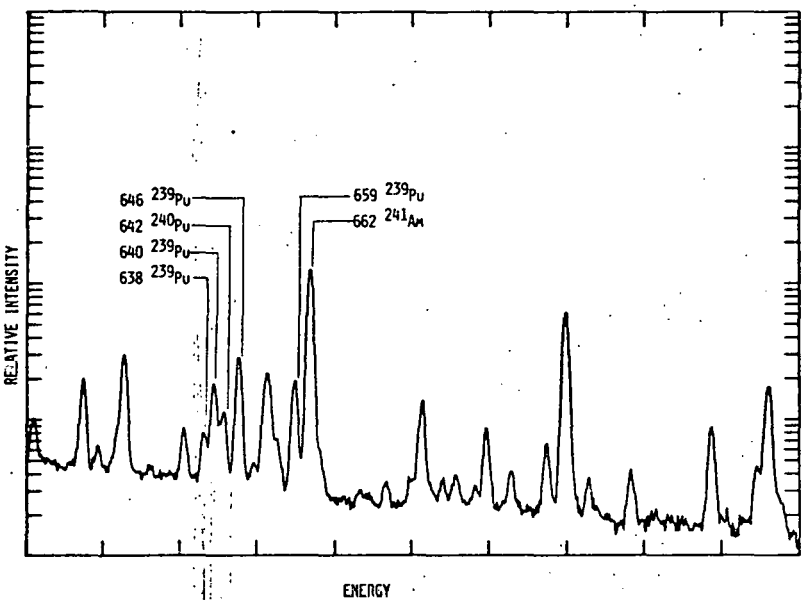

Figure 3

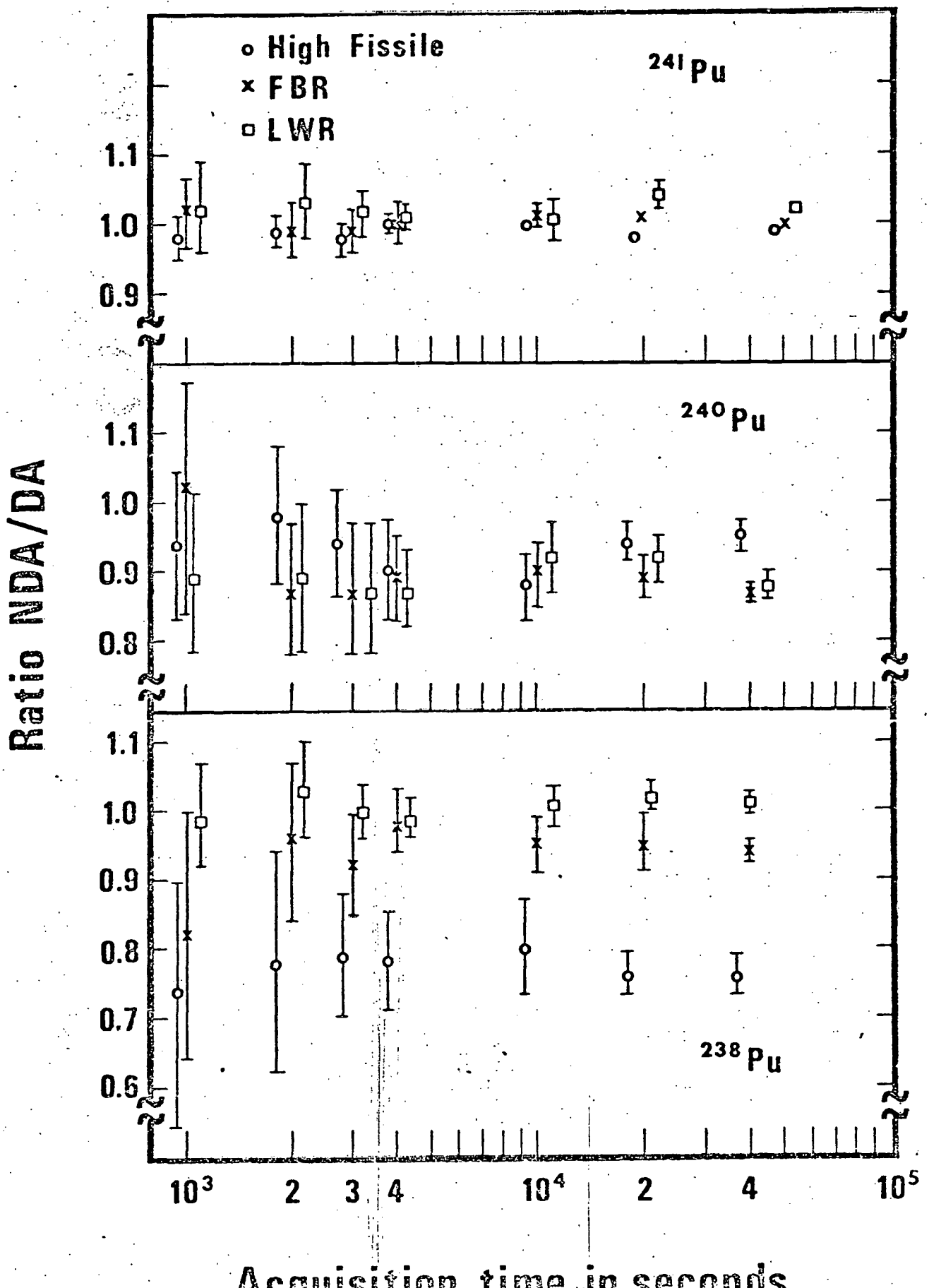
o High Fissile
× FBR
□ LWR
^{241}Pu
^{240}Pu
^{238}Pu
Ratio NDA/DA
1.1
1.0
0.9
0.8
0.7
0.6
10^3
2
3
4
10^4
2
4
10^5
Acquisition time in seconds

Figure 4

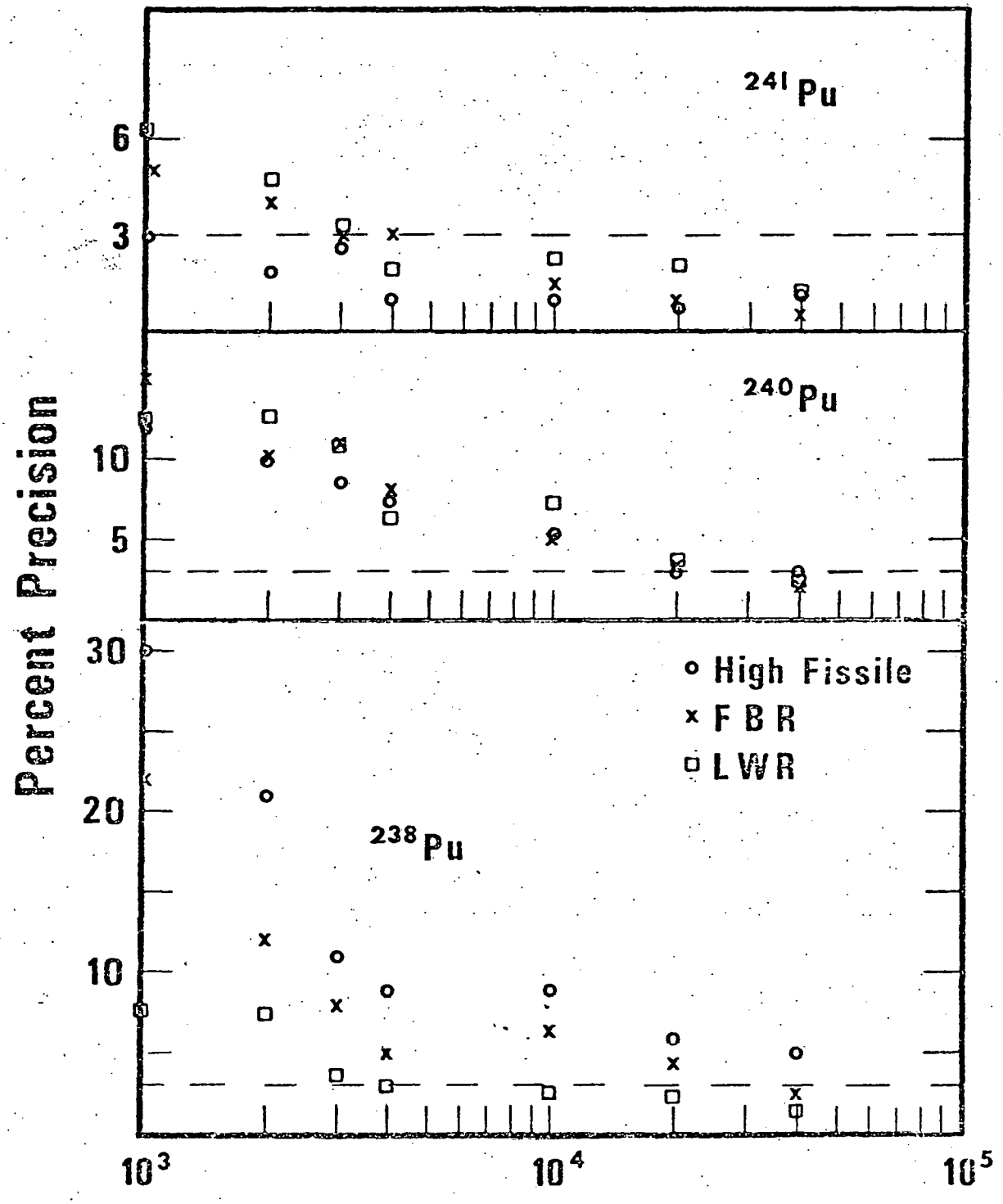
^{241}Pu
^{240}Pu
^{238}Pu
6
3
10
5
30
20
10
Percent Precision
o High Fissile
x F B R
□ LWR
10^3
10^4
10^5
Acquisition time in seconds

Figure 5