CONF-950923--8

LA-UR- 95-3297

Title: PC/FRAM: NEW CAPABILITIES FOR THE GAMMA-RAY SPECTROMETRY MEASUREMENT OF PLUTONIUM ISOTOPIC COMPOSITION

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Submitted to: American Nuclear Society
"Fifth International Conference on Facility Operations—Safeguards Interface."
Jackson Hole, Wyoming USA
September 24-29, 1995
(FULL PAPER)

Los Alamos
NATIONAL LABORATORY

PC/FRAM: NEW CAPABILITIES FOR THE GAMMA-RAY SPECTROMETRY MEASUREMENT OF PLUTONIUM ISOTOPIC COMPOSITION[a]

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ABSTRACT

We describe the new capability of and present measurement results from the PC/FRAM plutonium isotopic analysis code. This new code allows data acquisition from a single coaxial germanium detector and analysis over an energy range from 120 keV to above 1 MeV. For the first time we demonstrate a complete isotopic analysis using only gamma rays greater than 200 keV in energy. This new capability allows the measurement of the plutonium isotopic composition of items inside shielded or heavy-walled containers without having to remove the items from the container. This greatly enhances worker safety by reducing handling and the resultant radiation exposure. Another application allows international inspectors to verify the contents of items inside sealed, long-term storage containers that may not be opened for national security or treaty compliance reasons. We present measurement results for traditional planar germanium detectors as well as coaxial detectors measuring shielded and unshielded samples.

INTRODUCTION

The FRAM[b] plutonium isotopic analysis code[1,2] was initially developed during the late 1980s, programmed in FORTRAN and operated on a VAX/VMS system with Canberra Series 90 hardware. This system has been in routine operation at the Los Alamos Plutonium Facility since 1988 and has been used to assay nearly 10 000 items. The data acquisition and computer hardware are now obsolete and the requirement for a more modern and widely acceptable computer platform has led us to rewrite the code. In addition to rewriting the code in C to operate on a PC under Windows 3.1, we have significantly enhanced the functionality and flexibility of the code (now called PC/FRAM) enabling it to operate in modes never before possible with a single software package. The details and user characteristics of the PC/FRAM software are described in another paper at this meeting.[3]

The most significant new capability of the PC/FRAM code is the acquisition of a wide-range energy spectrum with a single coaxial detector. This allows not only the customary analysis range of 120-420 keV, previously used only with planar detectors, but also permits analysis of the same spectral data at energies up to and above 1 MeV. This second analysis method enables the user to obtain a complete isotopic analysis (^{238}Pu through ^{241}Pu, ^{241}Am, other isotopes, and ^{242}Pu by correlation) using only gamma rays above 200 keV. (A complete analysis with gamma rays above 300 keV has also been demonstrated.) Now users can measure the isotopic distribution of plutonium-bearing items inside thick-walled or shielded containers without having to remove the items from the container.

DATA ACQUISITION

The new code preserves all the functionality of the old code allowing data acquisition and analysis with a single planar detector in the energy range up to 420 keV. Most of our experience with both the old and new codes using planar detectors has been with spectra taken in 4096 channels at a gain of 0.1 keV/channel. However, the flexibility of PC/FRAM allows, without software changes, analysis of planar or coaxial detector spectra taken at any reasonable gain. As just one example (others will be given later), we have analyzed data taken with a single

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[a]This work is supported by the US Department of Energy, Office of Nonproliferation and National Security, Office of Safeguards and Security, and Stockpile Support Program.

[b]FRAM is a word of Scandinavian origin meaning "forward" or "onward." It is also an acronym meaning "Fixed energy, Response function Analysis with Multiple efficiency" and as such describes the working principles of the code.

planar detector in the energy range from 120-300 keV with 4096 channels at a gain of 0.075 keV/channel.

The current emphasis, presented here in more detail, has been to acquire one spectrum with a single coaxial detector spanning the energy range from 0 to 1024 keV in 8192 channels, a gain of 0.125 keV/channel. We use a coaxial high-purity germanium (HpGe) detector of approximately 25% relative efficiency with nominal low-rate, long-time-constant resolution of <1.75 keV at 1332 keV and <750 eV at 122 keV. We typically acquire spectra using a 2-μs (triangular shaping) time constant at an input rate of approximately 30 kHz, when possible. With this single detector and single set of data acquisition conditions, we are able to acquire and analyze data from both "normal" and "shielded" items without changing any measurement conditions. If the sample is "normal," meaning gamma rays in the 120 to 200-keV range are present with meaningful intensities, we obtain the best results by analyzing data in the energy range from 120-450 keV. If these lower-energy gamma rays are absent, usually because the item is packaged in a heavy-walled or shielded container, we analyze the spectrum remaining above 200 keV, obtaining ^{240}Pu at 642.5 keV and ^{238}Pu at 766.4 keV. We show gamma-ray spectra from a low-burnup item in a "normal" and shielded container in Fig. 1.

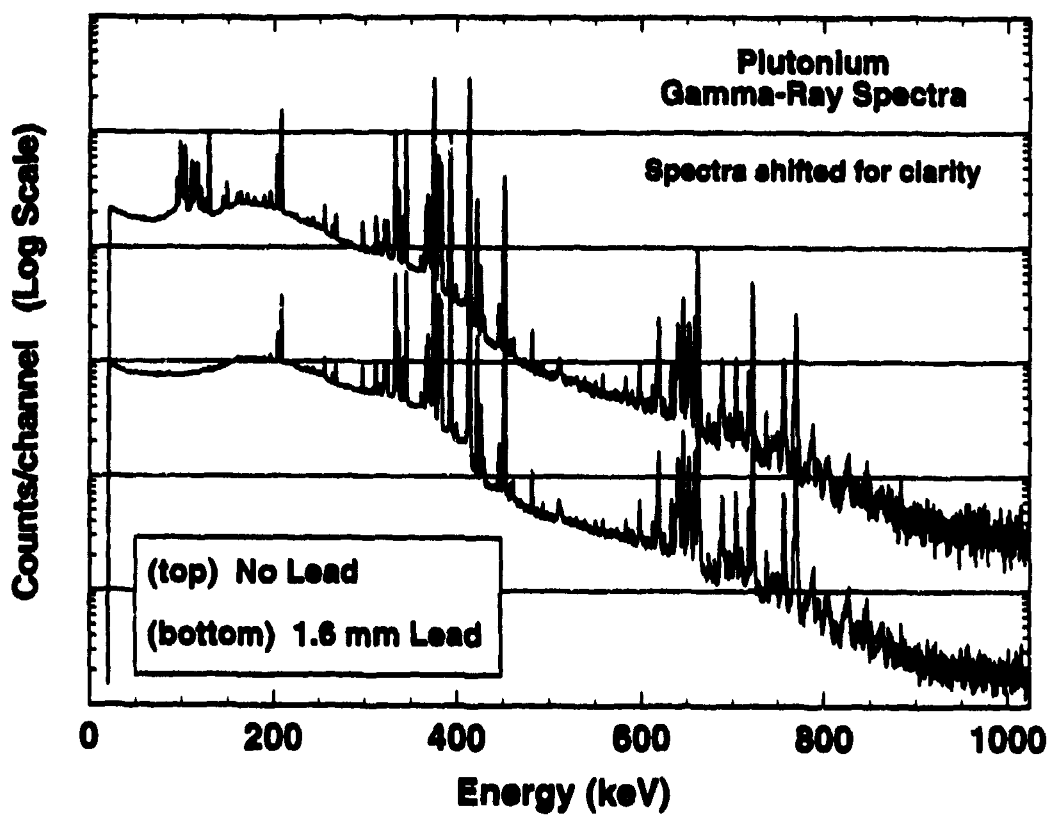

Fig. 1. Coaxial-detector gamma-ray spectra from shielded and unshielded low-burnup plutonium.

IMPROVED ANALYSIS CAPABILITY

These important new analysis capabilities are made possible by improvements in the background-continuum subtraction routines. We define the background underneath each peak region by specifying up to four background regions of interest (ROI) of arbitrary size and location in the spectrum. For each peak region, the user can choose from seven functional forms for the shape of the background continuum underneath that region. The chosen functional form is then fit by least squares techniques to the data in the specified background ROIs. The seven functional forms are (1) zero-slope straight line, (2) sloping straight line, (3) zero-slope smoothed step function, (4) smoothed step function on a sloping straight line, (5) quadratic, (6) exponential, and (7) bilinear smoothed step function (different slopes at the entrance and exit to the region). The previous FRAM code forced all background ROIs to be the same size for a given peak region and limited the maximum number of channels defined for the background. Both of these significant restrictions have been removed in PC/FRAM.

While one may legitimately question the accuracy of the background continuum calculated over a wide energy region, nevertheless, we calculate this continuum over a 55-keV-wide region encompassing nearly 450 channels of data to fit the complex of peaks between 640 and 662 keV. This region is vital for measurements on shielded samples because it contains the only ^{240}Pu peak above 200 keV. Such a background calculation and peak fit was not possible in the previous FRAM code. The ability of the PC/FRAM code to fit the data in this region is displayed in Fig. 2.

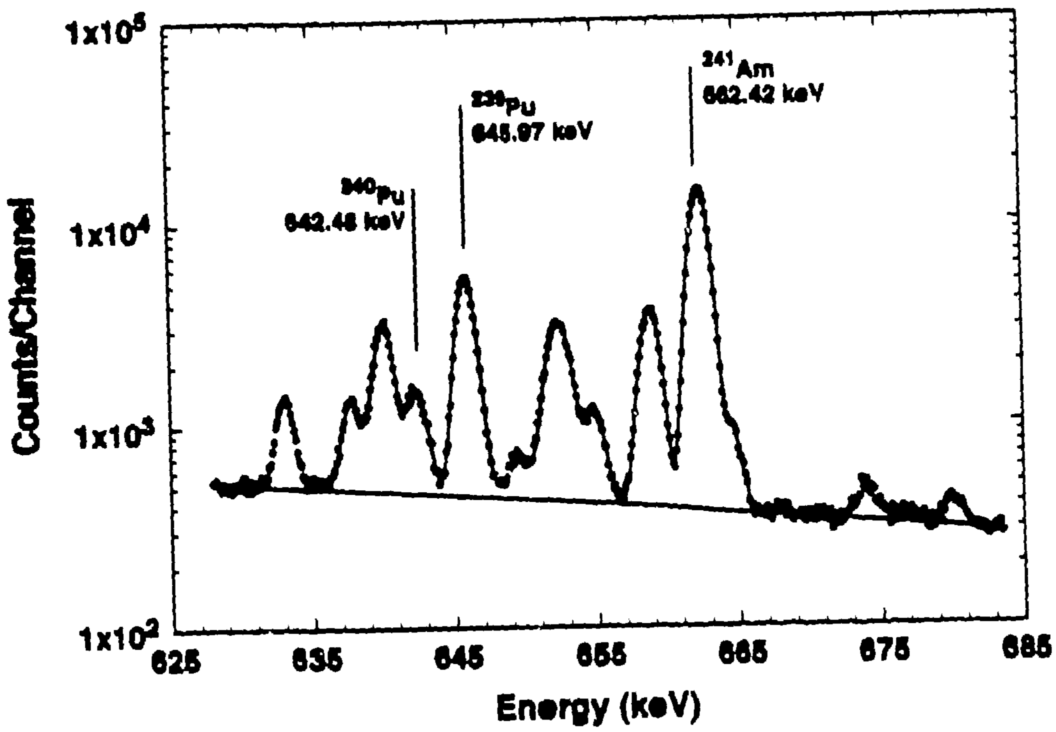

Fig. 2. PC/FRAM fit to data in the 640-keV region. The background continuum under the peak region is calculated from ROI data at 628, 650, 671 and 683 keV. Thirteen peaks are fitted between 637 and 664 keV.

All of the parameters governing the definition of the background continuum are stored in an easily accessible, easily edited database accessed by the PC/FRAM software. This user-editable parameter database is one of the principal features of PC/FRAM that makes the code versatile and adaptable without expensive, time-consuming software modifications. This versatility allows the user to switch among different detectors or data acquisition conditions or both without reprogramming. An example of the many data acquisition conditions that have been used with PC/FRAM is given in Table I.

TABLE I. Example of Data Acquisition Conditions for PC/FRAM (most widely used conditions in bold)

Detector	Analysis (keV)	Gain (keV/ch)
Planar	**120-420**	**0.1**
	120-307	0.075
Coaxial	**120-460**	**0.125**
	120-460	0.25
(Shielded items)	**200-800**	**0.125**
(Shielded items)	300-800	0.125

APPLICATION OF PC/FRAM

The most significant new application permitted by the new PC/FRAM code is the complete isotopic analysis of spectra acquired with a single coaxial detector from items in shielded or heavy-walled containers (Fig. 3). Now, for the first time, facility operators will not have to unpackage items in shielded containers before performing an isotopic measurement. This avoids the additional radiation exposure that this extra handling produces in facilities that employ other isotopic analysis methods. This new application also permits safeguards inspectorates to verify or measure items in heavy-walled storage containers without opening the container or handling the items: actions that might compromise sensitive or classified information. Below we show measurement results for the two most important parameters extracted from plutonium isotopic measurements, the effective specific power P_{eff} used with calorimetry measurements and the effective ^{240}Pu fraction $^{240}Pu_{eff}$ used with neutron coincidence counting measurements.

We display in Fig. 4 the bias of P_{eff} and $^{240}Pu_{eff}$ for data analyzed from 200–800 keV. Some of the measurements were taken with lead shielding surrounding the sample (see bottom spectrum in Fig. 1) while others had no shielding but were analyzed as if they were shielded. We saw no differences in the two sets of analyses. These data typically consisted of 10-15 repeated measurements of 1 hour each. The accepted value of the ^{242}Pu fraction was entered by the operator for each measurement sequence. The P_{eff} data show essentially no bias with ^{240}Pu content while the $^{240}Pu_{eff}$ data show a small but definite ^{240}Pu-dependent bias arising from a bias in the measurement of the ^{240}Pu fraction. We do not completely understand the reason for this bias and are studying it. The % relative standard deviation (RSD) shown in Figs. 4-6 is the RSD of the distribution of the bias data points.

The same coaxial detector data (unshielded samples only) shown in Fig. 4 were also analyzed in the more traditional lower-energy range: 120-450 keV for this data. The results for P_{eff} and $^{240}Pu_{eff}$ are shown in Fig. 5. Here we see excellent results for P_{eff} and somewhat poorer results for $^{240}Pu_{eff}$ on low-burnup samples. The poorer results for $^{240}Pu_{eff}$ probably arise from the difficulty in calculating the correct background for the weak peak complex at 160 keV in the presence of a changing slope for the background continuum under this region. We are working on this problem.

The benchmark to which the coaxial detector measurements should be compared is "conventional" planar detector data from the 120-420 keV region. The new PC/FRAM system with a planar detector is every bit as good as the previous Vax-based software system. We display bias results for PC/FRAM with a planar detector in Fig. 6.

These results for bias (Fig. 4-6) show that overall a planar detector is still the PC/FRAM detector of choice for samples that are not shielded, although the coaxial detector is at least as good, if not somewhat better, for P_{eff} with unshielded items. With shielded items there is no comparison to be made because PC/FRAM is the first code to demonstrate a complete plutonium isotopic analysis using only gamma rays above 200 keV. The RSDs displayed in Figs. 4-6 can be viewed as a typical "accuracy" for the measurement of the specified parameter. These values are tabulated in Table II.

Similar plots (Fig. 7) can be displayed for the measurement precision or repeatability. The values for

Fig. 3. Storage containers that no longer have to be opened to perform isotopic analysis measurements. Container on right is lead-lined while container on left can store dismantled weapons components.

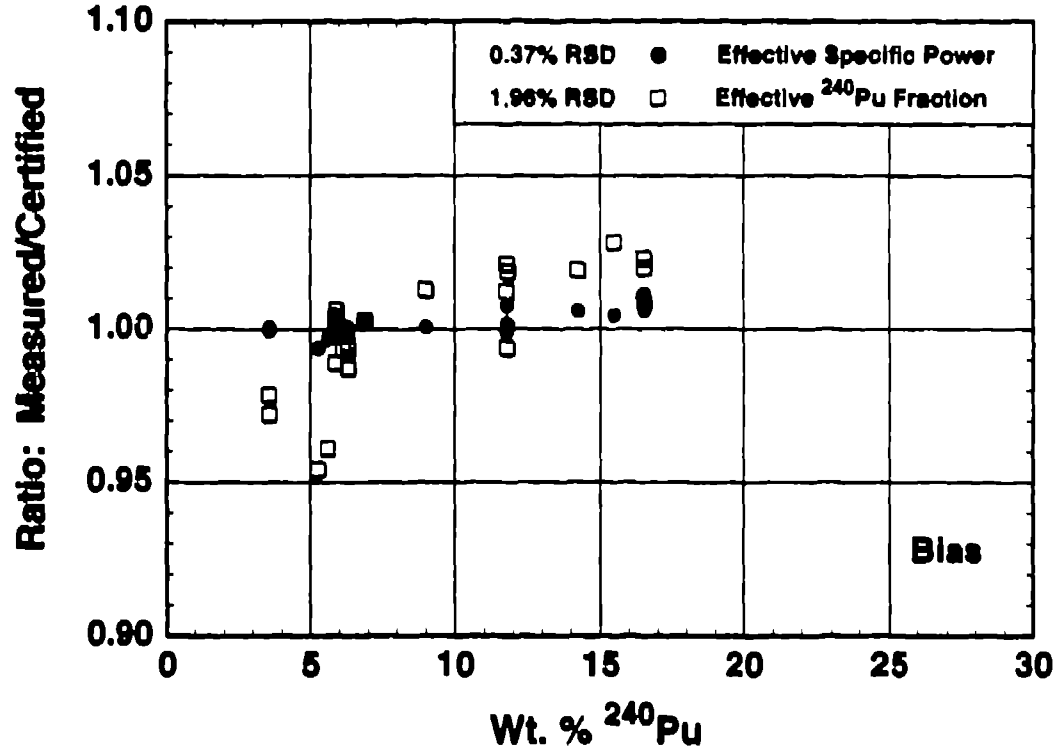

Fig. 4. Bias of coaxial detector measurements of effective specific power and effective ^{240}Pu fraction with analysis of gamma rays from 200-800 keV.

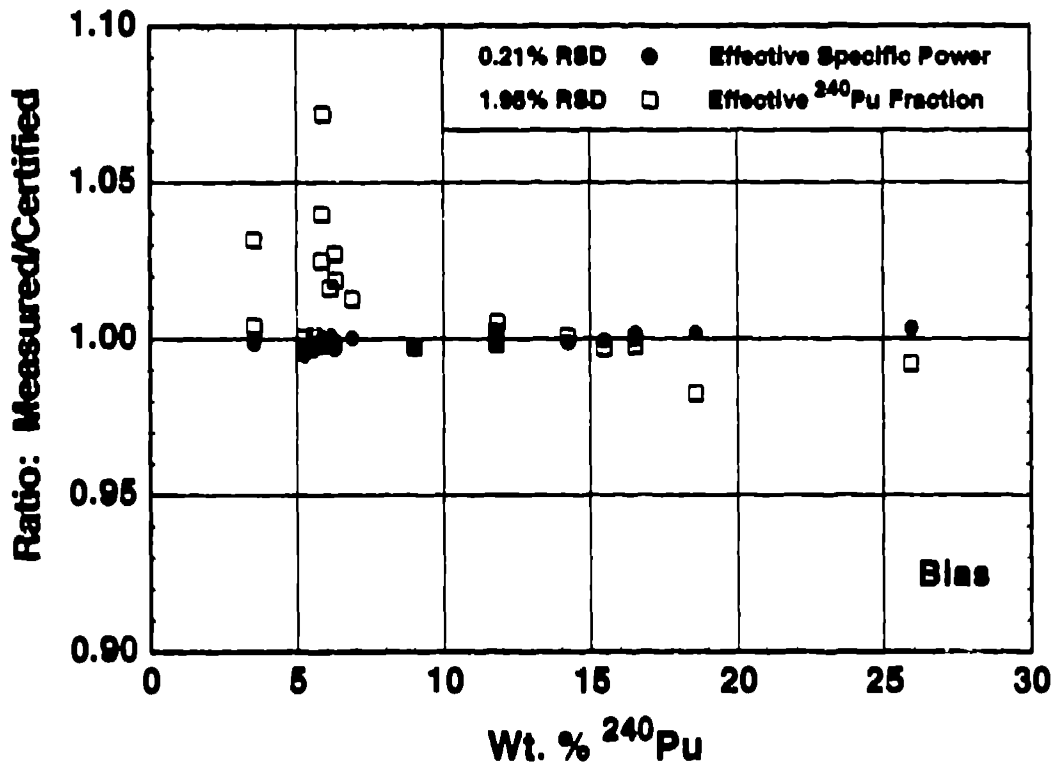

Fig 5. Bias of coaxial detector measurements of effective specific power and effective ^{240}Pu fraction with analysis of gamma rays from 120-450 keV.

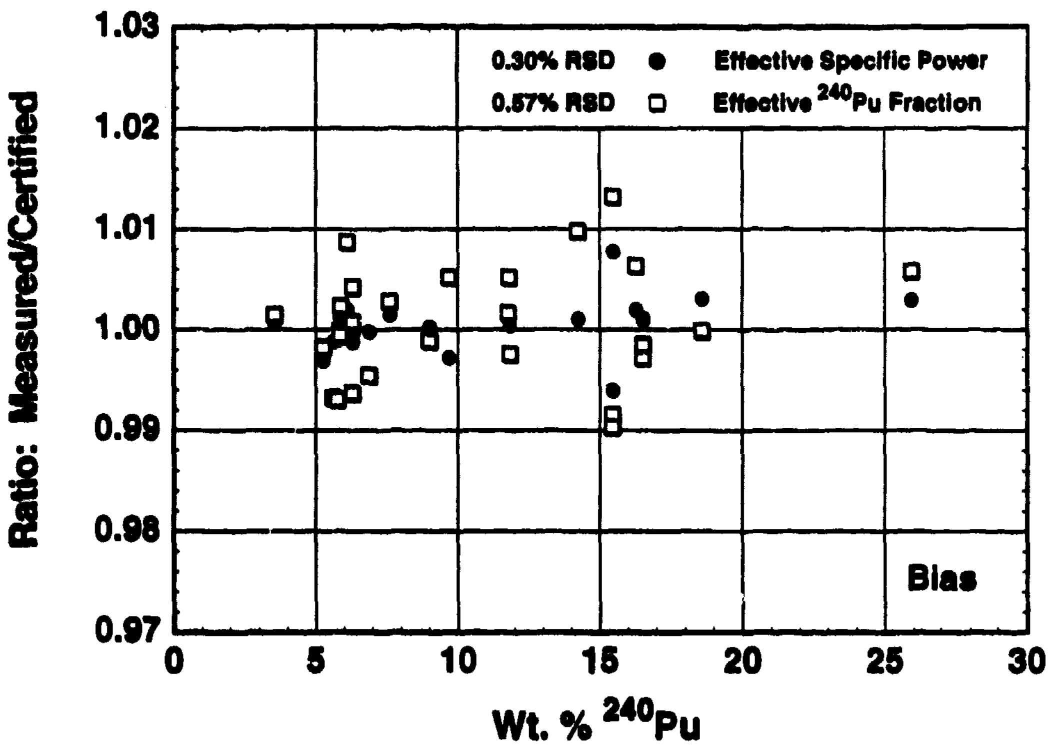

Fig 6. Bias of planar detector measurements of effective specific power and effective ^{240}Pu fraction with analysis of gamma rays from 120-420 keV.

TABLE II. Typical Measurement "Accuracy" From the Distribution of Observed Measurement Bias on Individual Items (%RSD)

Detector/Measurement	P_{eff}	$^{240}Pu_{eff}$
Planar, 120-420 keV	0.30	0.57
Coaxial, 120-450 keV	0.21	1.95
Coaxial, 200-800 keV	0.37	1.96

the measurement precision are calculated for each item as the RSD of the 10-15 (typical) repeated measurements of the tabulated parameter. The measurements were made without replacing the sample. All measurement precision data are for 1-hour measurements.

For lower mass items (<30 g) the trend is fairly clear with the shielded coax analysis in the 200–800-keV range giving poorer precision than the other two methods. For the <30-g mass range, the 120–450-keV analyses with the coaxial and planar detectors are similar. The poorer precision of the 200–800-keV analysis is expected because the important gamma rays at 642 and 766 keV are much less intense than their counterparts below 200 keV. The higher mass items gain back some of this deficit in the analysis above 200 keV because the greater penetrability of the gamma rays with energies greater than 600 keV allows the detector to effectively sample a larger fraction of the item than that sampled at lower energies. The planar detector may still have a slight edge in precision for higher-mass items while the two coaxial detector analysis modes appear to be similar.

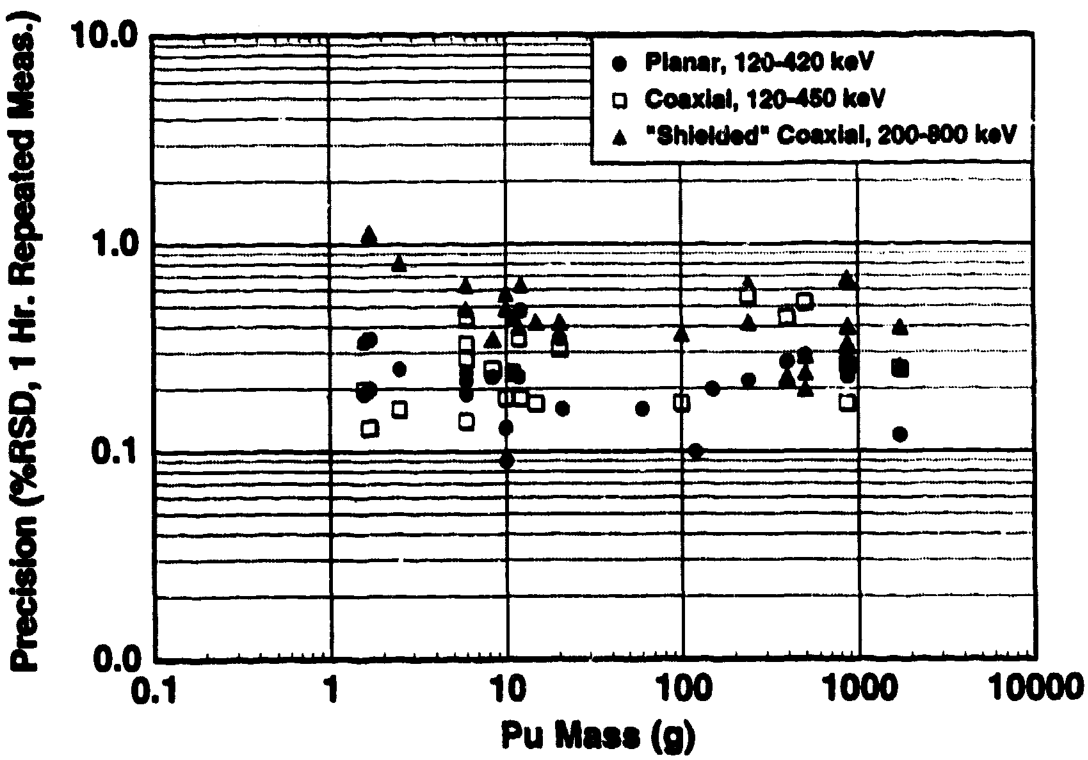

Fig 7. Precision of repeated measurements of effective specific power for measurements taken with three detector/measurement combinations.

Figure 8 displays, in the same fashion, the measurement precision for $^{240}Pu_{eff}$.

The trends and comparisons among the three measurement sets are similar to those in Fig. 7 for P_{eff}, except the absolute magnitude of the precision is about 4-5 times larger.

CONCLUSIONS

We can accurately and precisely measure the plutonium isotopic composition of arbitrary samples contained in heavy-walled or shielded containers, packaging configurations heretofore unmeasurable. We carry out these measurements with a single coaxial HpGe detector which, without changing instrument settings, can acquire and analyze data from 120 keV to above 800 keV. The flexibility of the new PC/FRAM code allows these measurements without software reprogramming.

This enhanced capability now allows operators to measure shielded containers without handling and unpacking them before measurement. This significantly reduces the radiation dose to plant personnel and increases safety margins through less handling of plutonium-bearing containers.

The measurement accuracy and precision for P_{eff} and $^{240}Pu_{eff}$ for coaxial detectors are entirely adequate for accountability measurements in DOE facilities and in some cases meet or exceed the quality of measurements from traditional planar detectors. This capability is now available in the new PC/FRAM code with over a dozen installations in place or planned worldwide.

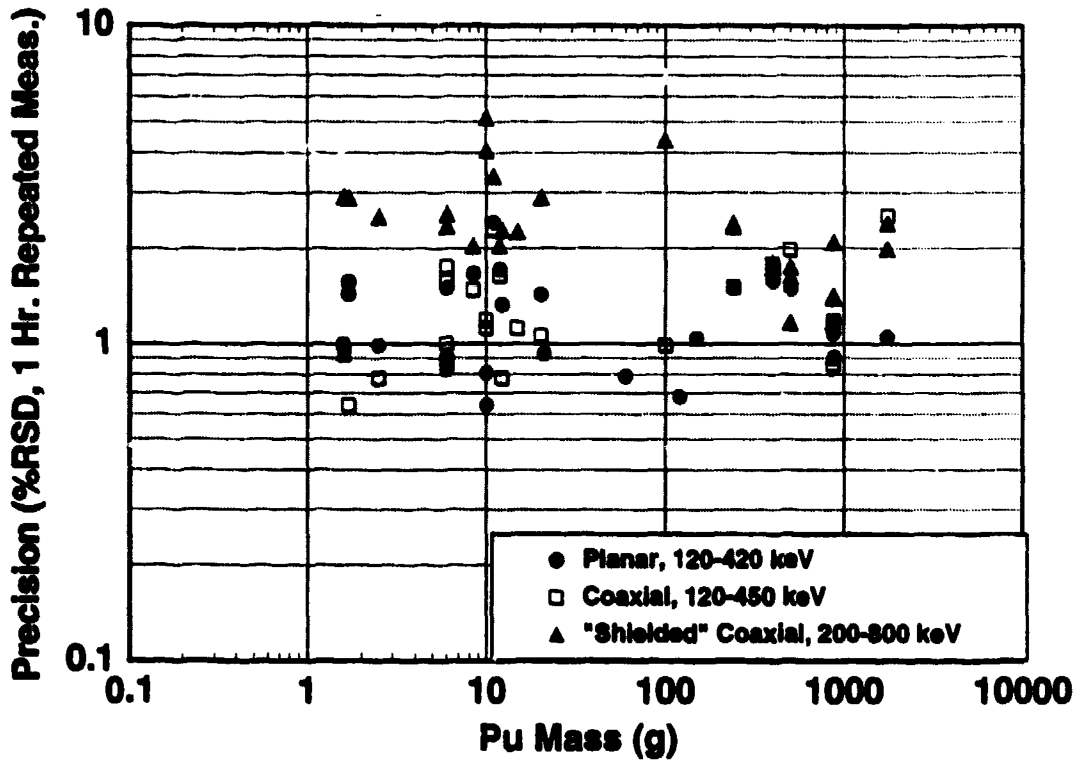

Fig 8. Precision of repeated measurements of the effective ²⁴⁰Pu fraction for measurements taken with three detector/measurement combinations.

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