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# Gamma-Ray Measurements for Simultaneous Calorimetric Assay

**David A. Rakel**

**March 2, 1982**

**Monsanto**

**MOUND FACILITY**
Miamisburg, Ohio 45342

operated by
**MONSANTO RESEARCH CORPORATION**
a subsidiary of Monsanto Company

for the
**U. S. DEPARTMENT OF ENERGY**
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# Abstract

*Gamma-ray measurements obtained in the course of developing a simultaneous calorimetric assay system are described. Gamma-ray measurements of the isotopic composition of six, well-characterized plutonium oxide samples were obtained while the samples were in the calorimeter. These samples represent a range of plutonium masses from 19 to 231 g and two isotopic compositions. The values of effective specific power determined from the gamma-ray measurements agree with the values determined from destructive assay.*

# Introduction

The determination of the total plutonium content of a sample by calorimetric assay requires the measurement of the thermal power of the sample by calorimetry and a measurement of the effective specific power of the sample. Typically, these measurements are separated in space and time and often involve the efforts of more than one investigator or technician.

Simultaneous calorimetric assay refers to a system that is capable of measuring the effective specific power and thermal power of a sample at the same time. A Simultaneous Calorimetric Assay System, which determines effective specific power from a measurement of isotopic composition by gamma-ray spectroscopy, has been developed at Mound.

This report describes the results of two sets of gamma-ray measurements obtained in the course of developing this system. First, the transmission of gamma-rays through the calorimeter was determined. Second, the effective specific power, $P_{eff}$, was calculated for six samples by using the isotopic composition determined by gamma-ray measurements of the samples in the calorimeter.

# Description of the System

The simultaneous assay system consists of two components: the calorimeter and the gamma-ray assay system. The calorimeter has been described as a transportable calorimeter. Its data acquisition and water handling systems are discussed in Ref. 1. The calorimeter used in these measurements is a right circular cylinder with an outer diameter of 28 cm and a height of 109.2 cm. Figure 1, a cut-away view of the calorimeter, shows the positions of the source and gamma-ray detector, and the composition and thickness of the materials between them. The absorbing material consists of 3.09 cm of water, 2.06 cm of plexiglass, 1.02 cm of epoxy, 0.64 cm of aluminum, and 0.08 cm of stainless steel.

The gamma-ray assay system consists of a small-volume (l-cm$^3$) intrinsic germanium detector, a multichannel analyzer, and a portable NIM bin which powers a high voltage power supply, amplifier, and ratemeter. This system has been used to assay a variety of plutonium samples [2]. Prior to the measurements described in this report, however, it had not been used to acquire data for a sample in the calorimeter.

# Transmission Measurement

The transmission through an attenuating medium is the ratio of the flux transmitted through the medium to the flux incident on the medium. To measure the transmission of gamma rays through the calorimeter, gamma-ray spectra were acquired with samples in and out of the calorimeter. For both spectra the samples were at the same distance and orientation relative to the detector. The spectra were analyzed by the computer code GRPNL2 [3]. To correct the measurements for count-rate related losses, a pulser peak was introduced into the spectrum by connecting the output of a pulser to the TEST input of the detector preamplifier and to a scaler. The transmission through the calorimeter at an energy E is given by:

$$T(E) = \frac{A_i(E)}{A_o(E)} \frac{PP_o}{PP_i} \frac{SC_i}{SC_o}$$

where T(E) = transmitted fraction of gamma-ray flux at energy E,

$A_i(E)$ = area of peak at energy E with source in calorimeter,

$A_o(E)$ = area of peak at energy E with source out of calorimeter,

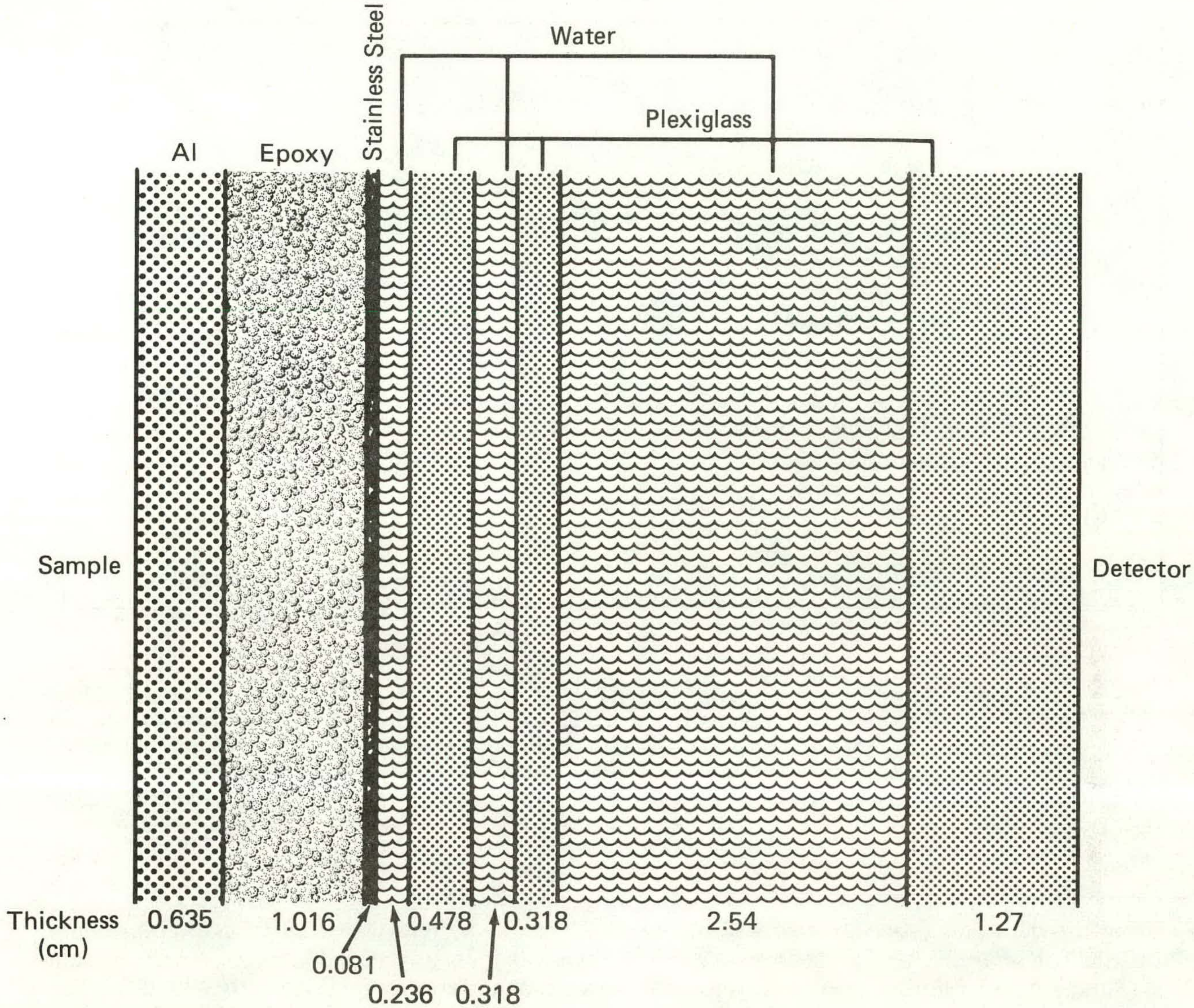

FIGURE 1 - Cut-away view of calorimeter showing materials between the plutonium sample and the gamma-ray detector.

$PP_i$ = pulser peak area with the source in the calorimeter,

$PP_o$ = pulser peak area with source out of calorimeter,

$SC_i$ = scaler counts for in-calorimeter spectrum, and

$SC_o$ = scaler counts for out-of-calorimeter spectrum.

Transmission measurements spanning the energy range from 53 to 451 keV were obtained by using point sources of $^{57}$Co and $^{133}$Ba and cans of plutonium oxide. For each source, three measurements of the transmission were obtained. In Figure 2, the weighted averages of the transmission values are plotted as a function of gamma-ray energy. The transmission values obtained with the cans of plutonium oxide are systematically less than the values obtained with the point sources. This is attributed to the effective increase in the thickness of the calorimeter walls for the finite-sized samples.

The gamma-ray transmission through the calorimeter was calculated using the mass attenuation coefficients of Ref. 4. The attenuation coefficients for plexiglass and epoxy were estimated

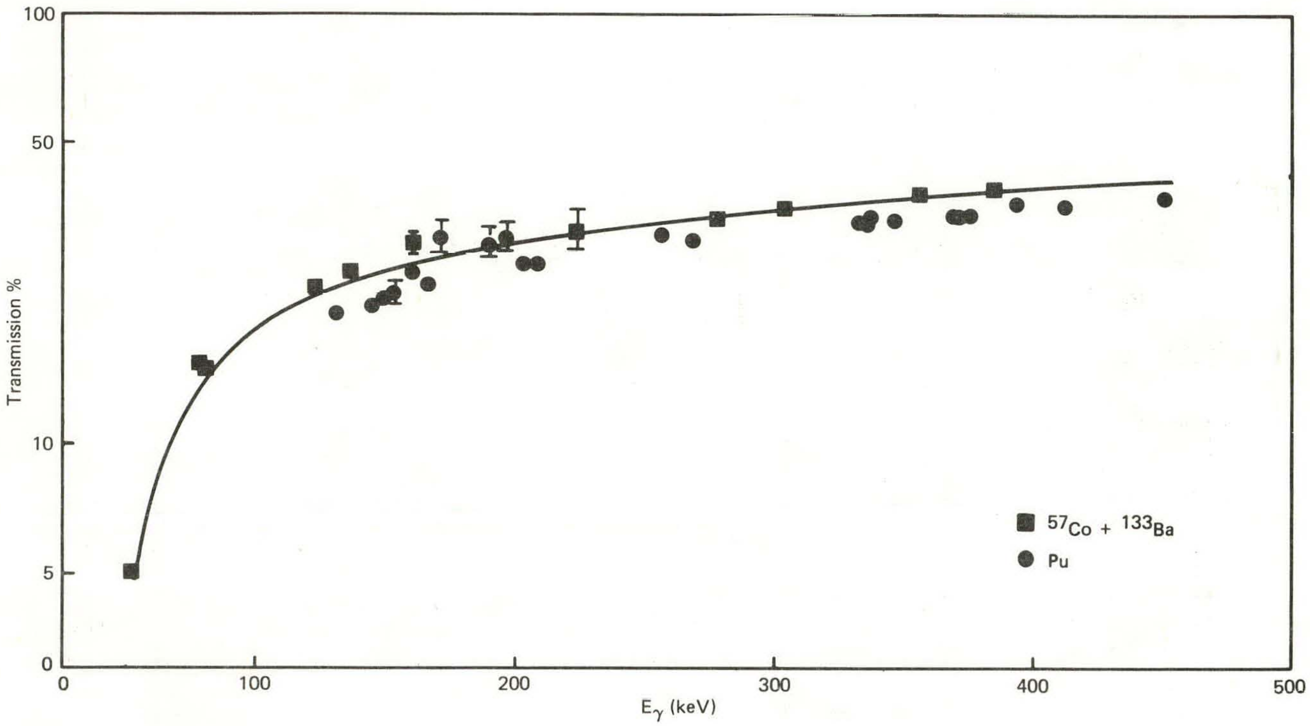

FIGURE 2 - Calculated values for transmission through calorimeter plotted as a function of gamma-ray energy (smooth curve). Measured values are also plotted.

by using the data and procedures of Ref. 4. A plot of the calculated value for the transmission as a function of photon energy forms the smooth curve in Figure 2.

Above 200 keV the calculated values agree with the measurements with point sources. Below that energy, the calculated transmission values underestimate the transmission when compared to the values measured with point sources. This could be attributed to overestimating the attenuation coefficients for plexiglass and epoxy. However, the calculated values are an acceptable representation of the transmission for a qualitative discussion.

The transmission through the various components of the calorimeter was calculated. In Figure 3 these values are plotted as a function of gamma ray energy along with the calculated values of the transmission through the calorimeter. Above 100 keV, the calorimeter component which produces the greatest attenuation is water. When the system was used to assay plutonium samples out of the calorimeter, spectral data were acquired from 20 keV to 470 keV. But peaks in the 125-208 and 332-375 keV regions only were used in the peak-pair ratios to determine isotopic composition. The relative efficiency curve was constructed using peaks from 125 to 451 keV. Peaks below 120 keV were not analyzed to determine isotopic composition. These peaks, particularly the 59.5 keV gamma ray from $^{241}$Am, can produce interference in a spectrum which prohibits a meaningful interpretation of the data. A judiciously chosen absorber can alleviate this problem. For the detector-sample combination used in this study, an absorber composed of 9-mil Pb and 32-mil Cd has been appropriate for out-of-calorimeter measurements of isotopic composition. Henceforth, this combination of Pb and Cd will be referred to as the absorber pack. The transmission of the transportable calorimeter and that of the absorber pack are plotted as a function of photon energy in Figure 4. The discontinuity in the

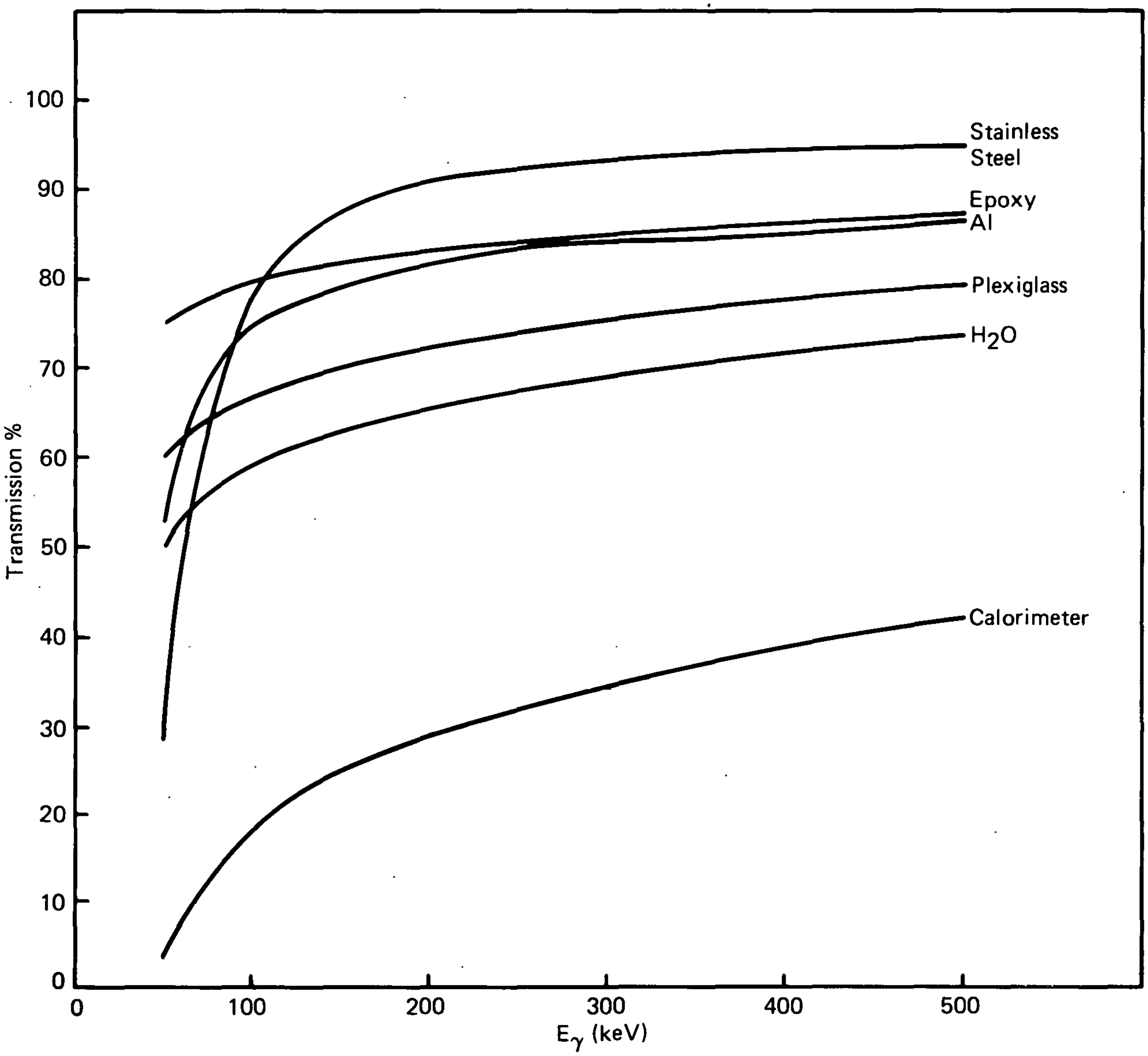

FIGURE 3 - Calculated values for transmission through calorimeter components plotted as a function of gamma-ray energy. The thicknesses of the various elements are: 3.09 cm of water, 2.06 cm of plexiglass, 1.02 cm of epoxy, 0.64 cm of aluminum, and 0.08 cm of stainless steel.

transmission curve of the absorber pack is due to the K-edge of Pb at 88 keV. Around 120 keV the transmission through the calorimeter is greater than that of the absorber pack. Above 120 keV the transmission by the absorber pack is greater than that by the calorimeter. Consequently, additional absorbers should be added to the simultaneous assay system to prevent spectral interferences from the 59.5 keV gamma ray.

Figure 5 depicts the transmission of the calorimeter and the transmission calculated for the calorimeter plus three combinations of Pb and Cd absorbers which reproduce the transmission of the absorber pack at 59.5 keV. Of these three, the additional 21-mil Cd absorber yields the maximum transmission in the 100-208 keV region, the region used for the Pu assay.

The ratio of the transmission through the absorber pack to the transmission through the calorimeter and a 21-mil Cd absorber is plotted in Figure 6. From 120 to 200 keV the ratio varies from 1 to 2.5. From 200 to 500 keV the ratio is approximately constant.

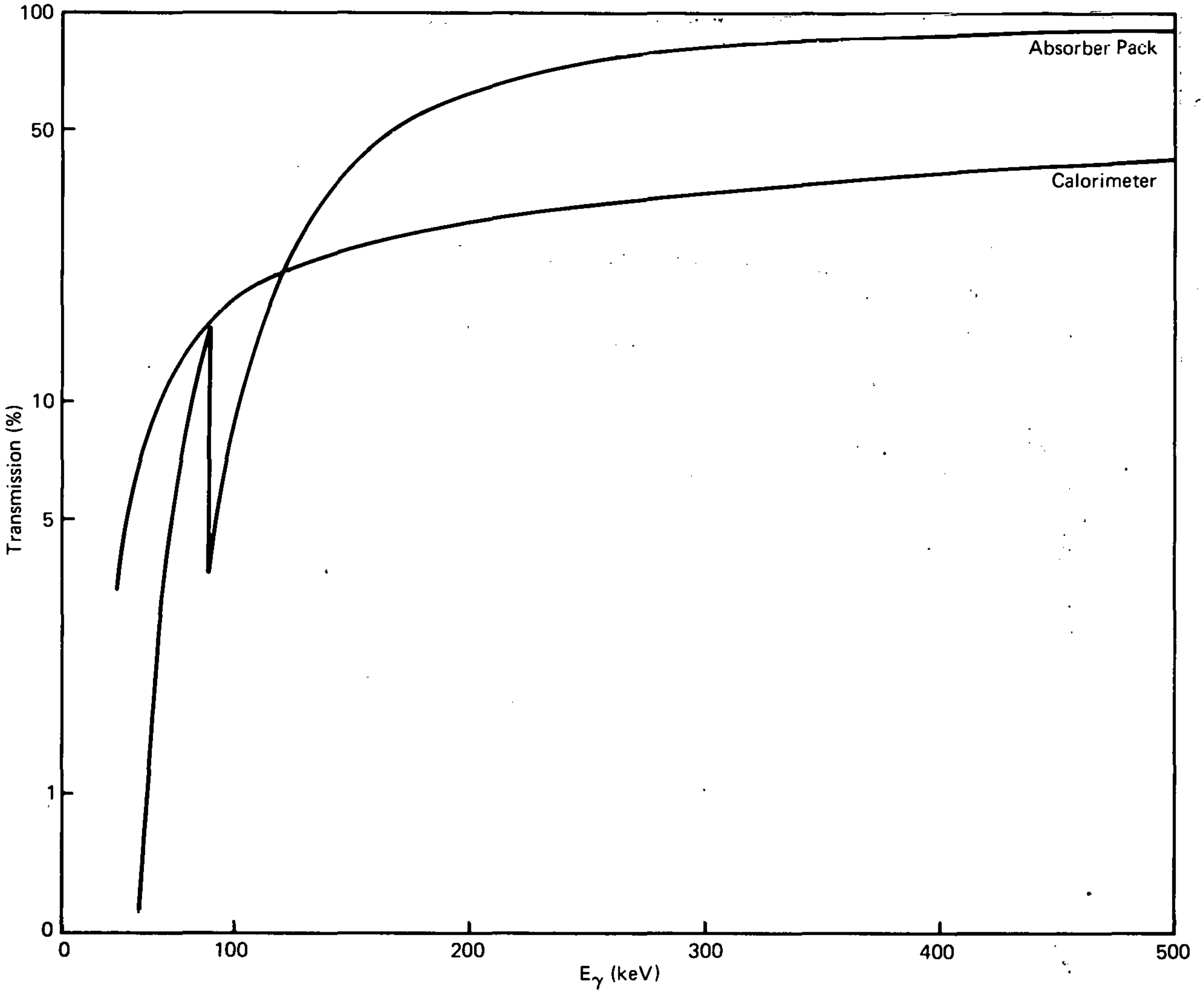

FIGURE 4 - Transmission through absorber pack composed of 9-mil lead and 32-mil cadmium, and transmission through calorimeter plotted as a function of gamma-ray energy.

The preceding discussion of transmission through the absorber pack and through the calorimeter plus additional absorbers has been quite specific about the elemental composition and thickness of the absorbers. The effectiveness of these absorbers in preserving spectral quality is dependent on the detector and sample combination. The values quoted are valid for the detector and the samples used in this study. The important result of these transmission measurements, which is specific to the calorimeter, is that the gamma-ray intensity in the energy region important for plutonium assay is reduced for in-calorimeter measurements by a small factor. The fact that the reduction is small indicates that gamma-ray measurements of isotopic composition can be performed with the plutonium sample in the calorimeter.

## In-Calorimeter Isotopic Measurements

In this section the effective specific power is determined for a variety of samples from the results of in-calorimeter measurements of isotopic composition. The effective specific power, $P_{eff}$, is defined [5] as:

$$P_{eff} = \sum_i R_i P_i$$

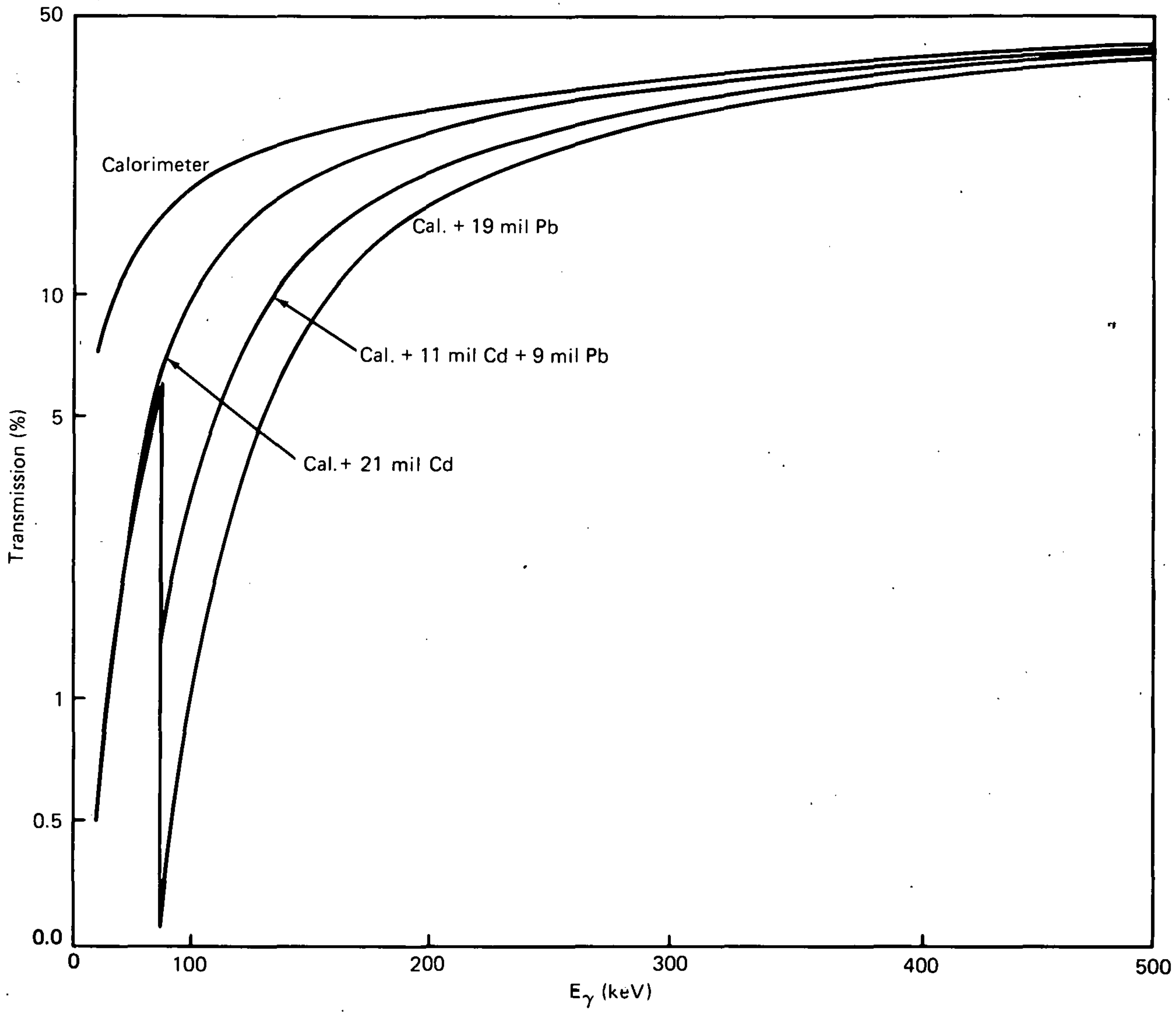

FIGURE 5 - Transmission through calorimeter and various absorbers plotted as a function of gamma-ray energy. The absorber thicknesses were chosen to reproduce at 59.5 keV the transmission of the absorber pack of 9-mil lead and 32-mil cadmium.

where

$R_i$ = abundance of $i^{th}$ nuclide (in weight percent),

$P_i$ = specific power of $i^{th}$ nuclide (in watts per gram), and i extends from $^{238}$Pu to $^{242}$Pu and includes $^{241}$Am.

The effective specific power and the thermal power (W) combine to give the plutonium content (M) of the sample:

$$M = \frac{W}{P_{eff}}$$

Six cans containing various quantities of plutonium oxide were assayed. Two isotopic compositions (Batch A and Batch B) were represented: there were three samples of each isotopic composition. The amount of Pu ranged from 19 to 231 g. The isotopic compositions for Batch A and Batch B and the plutonium content of the individual samples are listed in Table 1. The samples were doubly encapsulated in stainless steel cans; the outer can was 6.6 cm in diameter and 16.4 cm high. Further details of the determination by destructive assay of the isotopic composition of these samples and their encapsulation can be found in Ref. 6.

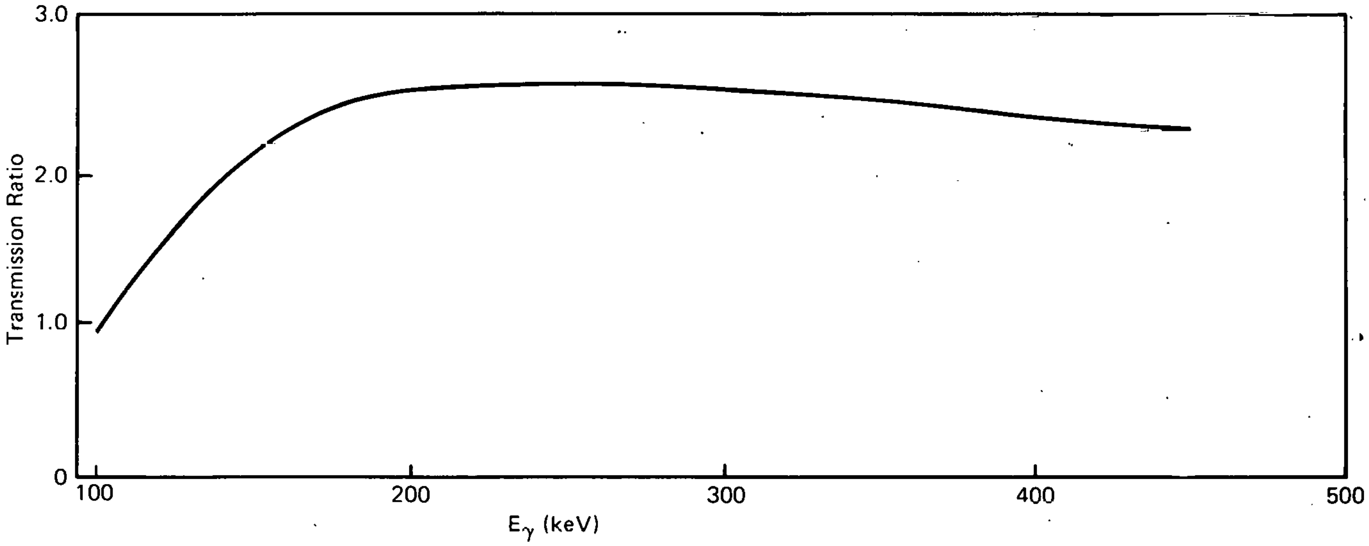

FIGURE 6 - Ratio of the transmission through the absorber pack (9-mil lead and 32-mil cadmium) to the transmission through the calorimeter and an added 21-mil of cadmium plotted as a function of gamma-ray energy.

Table 1 - PROPERTIES OF MIXED OXIDE SAMPLES[a]

| Batch | Can | Pu Content, (g)[b] |
|---|---|---|
| A | 5 | 19.21921 + /-0.00600 |
|  | 11 | 44.21980 + /-0.01682 |
|  | 17 | 192.27346 + /-0.06077 |
| B | 4 | 231.41571 + /-0.08202 |
|  | 20 | 28.28636 + /-0.00990 |
|  | 21 | 59.15817 + /-0.02437 |

Plutonium Isotopic Composition (wt %)

|  | Batch A | Batch B |
|---|---|---|
| $^{238}$Pu | 0.0539 | 0.2430 |
| $^{239}$Pu | 86.7189 | 79.1160 |
| $^{240}$Pu | 11.7152 | 16.8230 |
| $^{241}$Pu | 1.3038 | 2.9263 |
| $^{242}$Pu | 0.2081 | 0.8916 |
| $^{241}$Am | 0.1824 | 0.6496 |

[a] Ref. 6.

[b] Common decay date March 1, 1978.

Two spectra were acquired for each sample; one for 50 ksec and another for 14.4 ksec. The 14.4 ksec period approximately matched the assay time of the calorimeter. The 50 ksec period is representative of an overnight assay. All of the spectra were acquired at the same source to detector distance and with a 21-mil cadmium absorber between the source and detector. The detector can was flush against the outer wall of the calorimeter. This geometry provides the highest count rate attainable for each sample with this set of hardware.

The spectra were analyzed with the automated program GRPAUT [7]. The program provides multiple determinations of the isotopic ratios: $^{238}$Pu/$^{239}$Pu, $^{240}$Pu/$^{239}$Pu, $^{241}$Pu/$^{239}$Pu, and $^{241}$Am/$^{239}$Pu. These multiple determinations of an isotopic ratio are used for consistency checks and were averaged to specify the isotopic composition of the sample.

Using these results, the computer code CALGAM calculates the effective specific power for the sample by the computational method described in ANSI N15.22-1975 [5]. For each sample, three values of the effective specific power for the date of the gamma-ray measurement were computed by using the values of the isotopic composition determined by destructive assay and the values determined by the gamma-ray measurements of 50 and 14.4 ksec. The values of effective specific power are listed in Table 2 for Batch A samples and in Table 3 for Batch B samples. Also listed in Tables 2 and 3 are the uncertainties, at the one sigma level, in the values of the effective specific power determined from the gamma-ray measurements. The uncertainty in the effective specific power determined by destructive assay is 0.2% for Batch A and Batch B samples. Tables 2 and 3 also list the percent difference between the effective specific power values determined by gamma-ray assay and those determined by destructive assay. The effective specific power values determined by gamma-ray assay and destructive assay agree within their uncertainties.

The uncertainties in the effective specific power and the thermal power combine to give the uncertainty in the plutonium content of the sample. The uncertainty in the effective specific power dominates the uncertainty in the overall assay. The uncertainty in the power measurement is typically 0.3% for the calorimeter used in this study. Using this power measurement uncertainty, the uncertainty in the plutonium content of the samples is estimated to range from 1.3% (192-g sample, 50-ksec gamma-assay period) to 4.4% (28-g sample, 14.4-ksec gamma-assay period).

Table 2 - MEASURED AND CALCULATED VALUES OF THE EFFECTIVE SPECIFIC POWER FOR BATCH A SAMPLES

| Can No. | Pu (g) | Count Time (ksec) | Measured $P_{eff}$ (mW/g) | % Error | Calculated $P_{eff}$ (mW/s) | M-C % Difference |
|---|---|---|---|---|---|---|
| 17 | 192 | 50 | 3.219 | 1.22 | 3.232 | −0.4 |
| | | 14.4 | 3.290 | 2.40 | 3.232 | +1.79 |
| 11 | 44 | 50 | 3.249 | 1.85 | 3.232 | +0.52 |
| | | 14.4 | 3.201 | 3.10 | 3.232 | −0.96 |
| 5 | 19 | 50 | 3.268 | 2.25 | 3.232 | +1.11 |
| | | 14.4 | 3.363 | 3.93 | 3.232 | +4.05 |

Table 3 - MEASURED AND CALCULATED VALUES OF THE EFFECTIVE SPECIFIC POWER FOR BATCH B SAMPLES

| Can No. | Pu (g) | Count Time (ksec) | Measured $P_{eff}$ (mW/g) | % Error | Calculated $P_{eff}$ (mW/s) | M-C % Difference |
|---|---|---|---|---|---|---|
| 4 | 231 | 50 | 5.324 | 1.92 | 5.347 | −0.43 |
| | | 14.4 | 5.326 | 2.29 | 5.347 | −0.39 |
| 21 | 59 | 50 | 5.258 | 1.58 | 5.314 | −1.05 |
| | | 50 | 5.314 | 1.54 | 5.314 | 0. |
| | | 14.4 | 5.306 | 3.18 | 5.314 | −0.15 |
| 20 | 28 | 50 | 5.462 | 2.24 | 5.316 | +2.75 |
| | | 14.4 | 5.274 | 4.38 | 5.316 | −0.79 |

The precision of the effective specific power is dependent on the isotopic composition of the sample, the mass of the sample, and the gamma-ray assay period. For a particular sample, the uncertainty in the effective specific power scales with the inverse square root of the gamma-ray assay time, if all other measurement parameters remain unchanged. The average ratio of the uncertainties in the effective specific power for the 14.4 and 50 ksec measurements is 1.8, which is comparable to the square root of the ratios of the measurement periods, 1.9.

# Summary

Two sets of gamma-ray measurements have been performed as part of the development of a simultaneous calorimetric assay system. The transmission of the gamma-rays in the energy region of interest for Pu assay through the transportable calorimeter has been determined. A dependence on sample size was noted. To maintain spectral quality, even further attenuation of the low energy spectrum was required.

The effective specific power was determined from in-calorimeter measurements of the isotopic composition of six samples. These samples included two different isotopic compositions and a range of Pu masses from 19 to 231 g. For these samples, and for this set of measurement hardware, overall calorimetric assay precision of 1.3% to 4.4% could be anticipated from these results.

# Acknowledgement

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