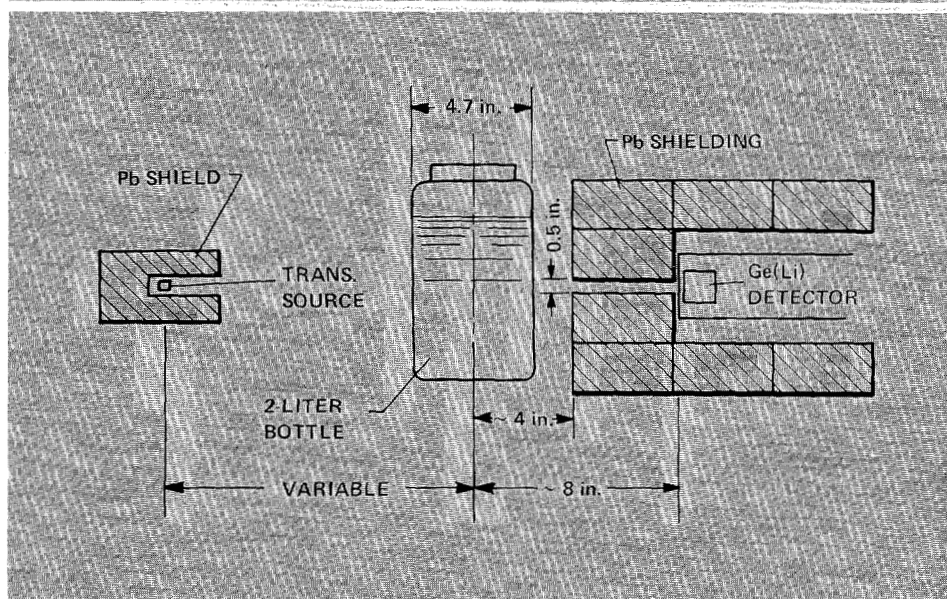


**TABLE I**  
**GAMMA-RAY MEAN FREE PATH**

Medium	186 keV ( $^{235}\text{U}$ ) (cm)	414 keV ( $^{239}\text{Pu}$ ) (cm)
H <sub>2</sub> O	7.1	9.7
Al	3.0	4.1
Fe	0.84	1.4
Pb	0.071	0.44
U	0.033	0.21



**Fig. 6.** The segmented gamma-ray scanning system used for passive gamma-ray measurement of heterogeneous uranium and plutonium scrap and waste. The sample is rotated and assayed by segments as it is translated vertically past the collimated germanium detector shown on the right. Transmissions of gamma rays from the source on the left are used to derive self-attenuation corrections for each assay segment.

decrease in the gamma-ray mean free path, especially for the lower energy gamma ray. Even small particles (~0.02 mm) of metallic uranium or plutonium are highly self-absorbing.

To determine the self-attenuation correction, most gamma-ray assays employ a separate transmission measurement of the linear absorption coefficient  $\mu_t$  of the sample. The external gamma-ray source must supply gamma rays with energies close to the energy of the signature gamma ray of interest. Once we know  $\mu_t$ , the dimensions of the sample, and its distance from the detector, we can calculate the self-absorption correction provided the mixture of the material to be assayed and the matrix

are reasonably uniform and the particles of assay material are small enough to ignore self-attenuation within individual emitting particles. Few closed forms exist for the correction factors, but often semiempirical analytical forms are sufficiently accurate. J. L. Parker and T. D. Reilly have developed analytical forms for self-attenuation corrections for most practical passive gamma-ray assay problems.

### Segmented Gamma Scanner

The simple procedures described above are inadequate to measure containers of scrap and waste because they cannot take into account the vertical

variations in SNM and matrix densities characteristic of these containers. Radial inhomogeneities are usually less pronounced, and their effects are substantially reduced by sample rotation. We developed the Segmented Gamma Scanner (SGS), which is both an instrument and a procedure, to improve the assay accuracy for a wide range of scrap and waste. It was also the first fully automated NDA instrument for Safeguards and is now used widely in fuel processing facilities.

The basic principle of the SGS is to divide the sample into thin horizontal segments and assay each segment independently using the transmission-corrected passive assay technique described above. After all the segments have been measured, the results are summed to give the total assay for the container. The method is shown in Fig. 6. A germanium detector views a segment of the container through an open slit, or collimator, in a lead shield. The transmission source is on the opposite side of the container, in line with the detector. For  $^{235}\text{U}$  assay, the transmission source is  $^{169}\text{Yb}$ . It emits 177.2-keV and 198.0-keV gamma rays, which closely bracket the 185.7-keV assay gamma ray of  $^{235}\text{U}$ . The  $^{75}\text{Se}$  400.6-keV gamma rays serve for transmission measurements in  $^{239}\text{Pu}$  assays based on detection of 414-keV gamma rays.

Computer control of SGS instruments allows automatic data acquisition, analysis, and management of all hardware. An operator places a container on the sample table, and the SGS does the rest. The SGS begins the assay sequence by positioning the sample table so that the top of the sample is just below the detector axis. Automatic controls rotate the sample continually and elevate it in discrete steps until all segments are assayed. Vertical profiles of individual segment assays and transmissions are available as data output, as well as the total SNM in the container

(Fig. 7). Typically a complete SGS assay requires 3 to 5 minutes, and accuracies lie in the 1-5% range for liter-size samples.

SGS instruments are now commercially available and are used in most major fuel cycle facilities to measure scrap and waste. Figure 8 shows an SGS instrument for 220-L and other large containers of low-level waste. We also have designed smaller units to assay containers less than 20 L.

### The Enrichment Meter Principle

The end products of uranium fuel production are high-concentration, homogeneous forms of enriched uranium, such as metal, uranium oxide in powder and ceramic fuel forms, and rich uranium scraps. We have developed a very simple method to measure the  $^{235}\text{U}$  enrichment in these materials, which is widely used in nuclear facilities not only to safeguard nuclear fuels but also to assure their quality. Called the *enrichment meter* technique, it uses to advantage the severe attenuation of the  $^{235}\text{U}$  186-keV gamma ray by high-Z materials. For homogeneous samples whose thickness is essentially infinite relative to the mean free path for this gamma ray, the intensity of the 186-keV gamma ray emitted will be independent of the sample thickness because only the gamma rays emitted near the surface will reach the detector. With reference to the sample-detector geometry in Fig. 9, performing the integration over the sample thickness indicated in the figure shows that for a metallic uranium sample, the counting rate (CR) of 186-keV gamma rays is given by the following expression.

$$\text{CR} = (\epsilon I A \underline{^{235}\text{U}}/\mu_1) \propto (\underline{^{235}\text{U}}/\mu_1),$$

where

$\epsilon$  = detection efficiency,

$I$  = intensity of 186-keV gamma ray ( $\gamma/\text{s per g } ^{235}\text{U}$ ),

$\underline{^{235}\text{U}}$  =  $^{235}\text{U}$  mass fraction (enrichment),

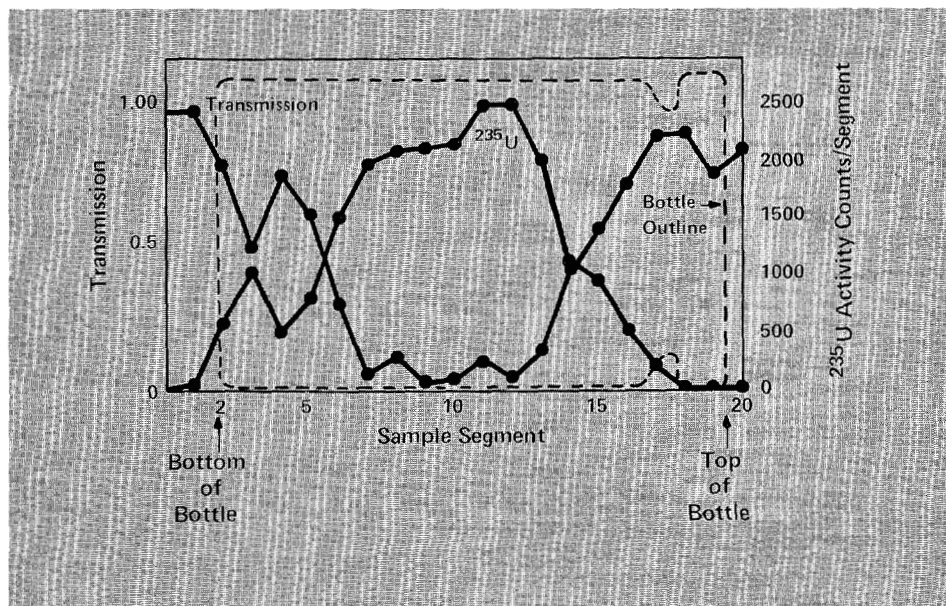


Fig. 7. Sample of data produced by segmented gamma scanner. The transmission of external rays (left-hand ordinate) and  $^{235}\text{U}$  186-keV gamma-ray activity (right-hand ordinate) are shown as a function of vertical segment of a 2-L bottle of high-enriched uranium. Note the profile of the bottle (lying on its side). The mirror effect in the transmission and activity data indicates that the sample attenuation is caused principally by its uranium content.

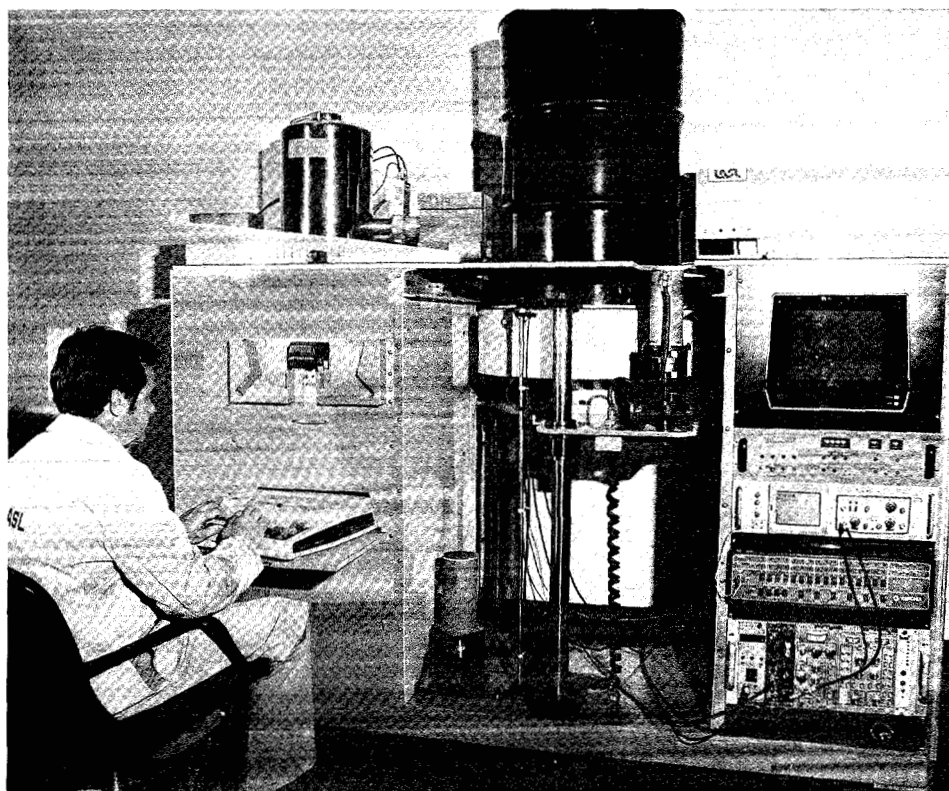


Fig. 8. An automated segmented gamma scanner for assay of uranium and plutonium in large containers. The germanium detector is located on the left of the 55-gallon drum sample, and the transmission source is on the right.

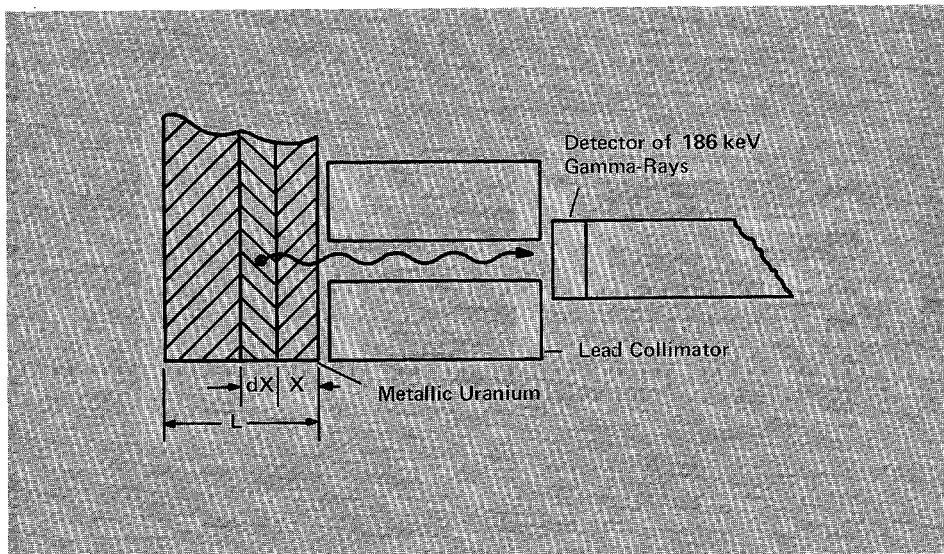


Fig. 9. The measurement geometry used for the enrichment meter principle. A measurement of 186-keV  $^{235}\text{U}$  gamma rays from a thick high-concentration uranium sample can yield the  $^{235}\text{U}$  enrichment directly.

$A$  = area of sample ( $\text{cm}^2$ ) defined by collimator, and  
 $\mu_1$  = mass attenuation coefficient of uranium for 186-keV gammas ( $\text{cm}^2/\text{g}$ ).

The counting rate is directly proportional to the  $^{235}\text{U}$  mass fraction, or enrichment, and does not depend on sample density or other factors as long as a fixed detector and collimation geometry is maintained for calibration and measurements.

For uranium compounds such as  $\text{UO}_2$  or  $\text{UF}_6$ , the expression for CR is

$$\text{CR} \propto ({}^{235}\text{U}/\mu_1) F,$$

where

$$F = [1 + (\mu_2\rho_2/\mu_1\rho_1)]^{-1},$$

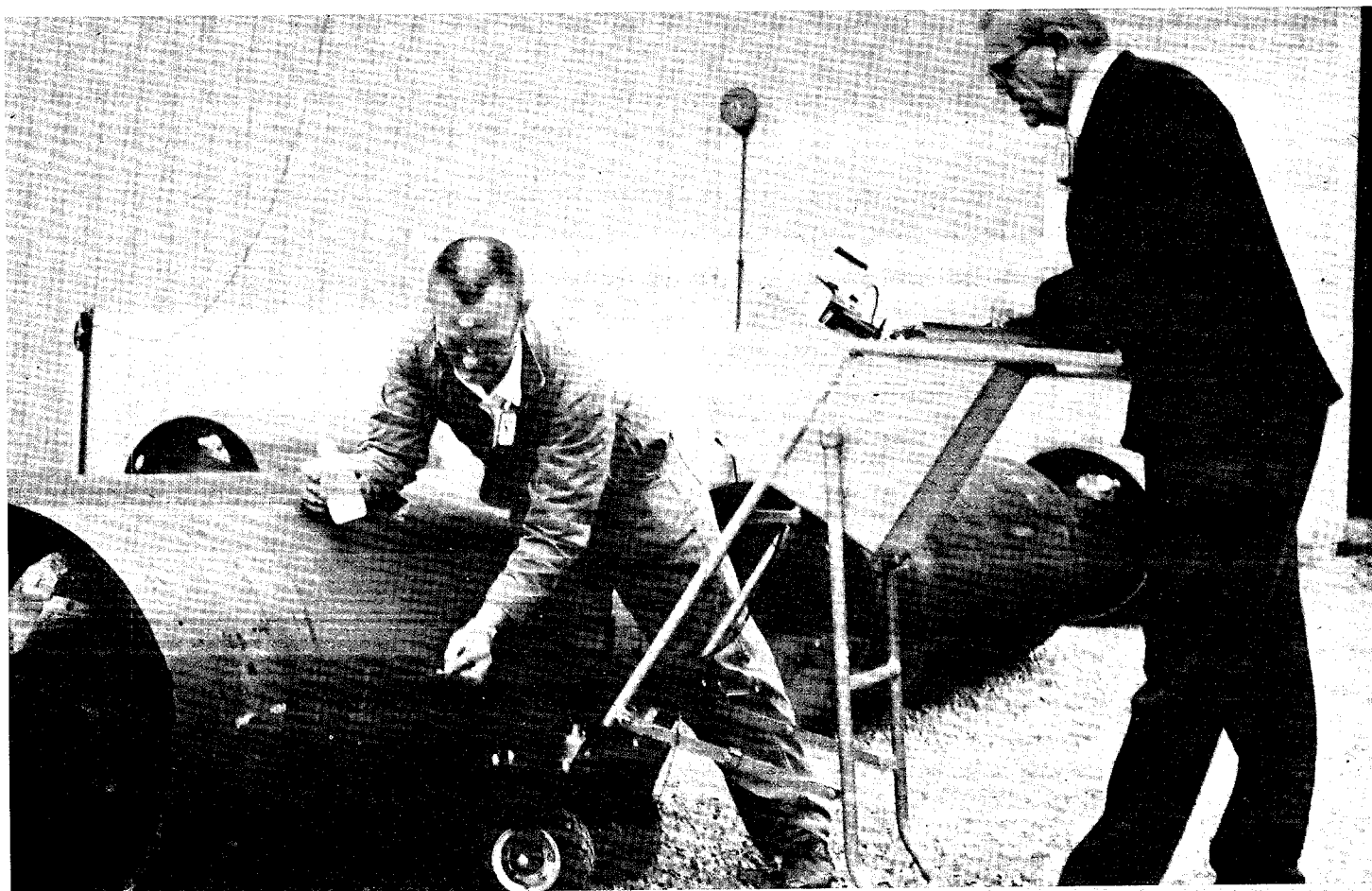


Fig. 10. Field measurement of  $^{235}\text{U}$  enrichment of  $\text{UF}_6$  in a 2-1/2-ton shipping container. A portable NaI detector is used to measure 186-keV  $^{235}\text{U}$  gamma rays. A portable ultrasonic gauge is used to determine the cylinder wall thickness for the attenuation correction.

$\mu_2$  = mass attenuation coefficient of the low-Z element, and  
 $\rho_1, \rho_2$  = the density (g/cm<sup>3</sup>) of uranium and the low-Z element, respectively.

For matrix materials with  $Z < 30$ ,  $\mu_2/\mu_1 \leq 0.11$  and, in many practical cases,  $F$  is nearly equal to unity, being 0.988 for  $\text{UO}_2$  and 0.930 for residues containing only 50 wt% uranium. Note also that  $F$  depends on concentration ratios instead of absolute quantities.

The enrichment meter principle may be used with a NaI or germanium detector; the latter gives more accurate results. Figure 10 shows a field measurement of the enrichment of  $\text{UF}_6$  in a standard 2 $\frac{1}{2}$ -ton cylinder used to ship this material to light water reactor (LWR) fuel fabricators.

### Plutonium Isotopic Analysis

The isotopic abundances of plutonium are needed for safeguards and accounting, particularly to distinguish weapons-grade from reactor-grade material and to assure the quality of product fuel. Furthermore, all NDA methods developed so far for the quantitative assay of plutonium in bulk materials depend on a prior knowledge of the isotopic abundances of the isotopes whose signatures are to be used for the measurement technique, that is,  $^{239}\text{Pu}$  for gamma-ray assay based on the 414-keV gamma ray, the fissile isotopes  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  for active neutron methods, and all plutonium isotopes and  $^{241}\text{Am}$  for calorimetry. In many instances, particularly for scrap and waste materials and recycle streams, the isotopic abundances of plutonium are not specified reliably.

To meet the need for a rapid non-destructive isotopic analysis, we are developing and adapting gamma-ray spectrometry. The methods involve analysis of pulse-height spectra of

plutonium gamma rays measured with high-resolution germanium detectors. Peak areas are obtained by using either a simple channel summation procedure with a straight-line background subtraction or sophisticated peak-fitting algorithms. As evident from Fig. 3, the plutonium isotopes  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ , as well as the  $^{241}\text{Pu}$  daughters  $^{241}\text{Am}$  and  $^{237}\text{U}$ , emit gamma rays that are useful for isotopic analysis. Unfortunately, the intensities of the gamma rays from  $^{240}\text{Pu}$  are weak and often obscured by gamma rays from other isotopes. Plutonium-242 does not have a useful gamma ray, but its isotopic abundance is small for low-burnup plutonium. For high-burnup fuels such as LWR spent fuels, it can be estimated from correlations of other plutonium isotopes or inferred from results of an independent measurement of elemental plutonium concentration combined with gamma-ray spectrometry data for the other isotopes.

Samples of controlled and constant form and known chemical composition are the easiest for isotopic analysis. For example, if we prepare "thin" samples of low-concentration solutions in precision vials, we can obtain quantitative abundances of individual plutonium isotopes by comparing gamma-ray peak areas from standard solutions and applying self-attenuation corrections.

We have also developed a relatively simple method for the isotopic analysis of plutonium in samples of arbitrary shape and composition. The procedure requires no detailed peak fitting; thus it is fast, and computer cost and speed requirements are minimal. This method can be programmed into portable multiple-channel analyzers for use in field inspections. We measure areas under the peaks of closely spaced gamma-ray lines in the region from 120 to 414 keV to determine the following isotopic ratios.

$^{238}\text{Pu}/^{241}\text{Pu}$	152.7 keV/148.6 keV
$^{239}\text{Pu}/^{241}\text{Pu}$	203.5 keV/208.0 keV or 345 keV/332.3 keV
$^{240}\text{Pu}/^{241}\text{Pu}$	160.3 keV/164.6 keV
$^{241}\text{Pu}/^{241}\text{Pu}$	332.3 keV/335.4 keV
$^{242}\text{Am}/^{239}\text{Pu}$	125.0 keV/129.3 keV

In some energy regions, the measured lines are not clean peaks; that is, they must be corrected for contributions from neighboring lines of other isotopes. For example, the measured  $^{240}\text{Pu}$  peak at 160.3 keV includes contributions from the  $^{241}\text{Pu}$  line at 160.0 keV and from the  $^{239}\text{Pu}$  line at 160.2 keV. By measuring the clean peak at 164.6 keV from the  $^{241}\text{Pu}$  daughter  $^{237}\text{U}$ , using the known intensity branching ratios for the 160.0- and 164.6-keV lines, and applying small corrections for changes in relative efficiencies, we can determine the 160.0-keV contribution and subtract it from the measured peak; similarly the  $^{239}\text{Pu}$  peak at 161.5 keV is used to obtain the  $^{239}\text{Pu}$  contribution to the 160-keV complex. The corrected peak is then divided by the  $^{241}\text{Pu}$  ( $^{237}\text{U}$  daughter) peak at 164.6 keV to give the  $^{240}\text{Pu}/^{241}\text{Pu}$  isotopic ratio.

For each sample, the relative detection efficiency is derived empirically from the areas of isolated, clean peaks of  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  in different regions of the spectrum, adjusted for their respective gamma-branching intensities. The isotopic ratios are then combined to yield the actual plutonium mass fractions using the constraint that their sum is unity. Results can be obtained by this method in 1 hour and, in some cases, with accuracies comparable to those obtained from mass spectrometry.

### Active Gamma-Ray Assay

We are adapting two active gamma-ray assay methods for measuring fissionable materials in plant environments: x-ray edge densitometry and x-ray fluorescence. These methods com-

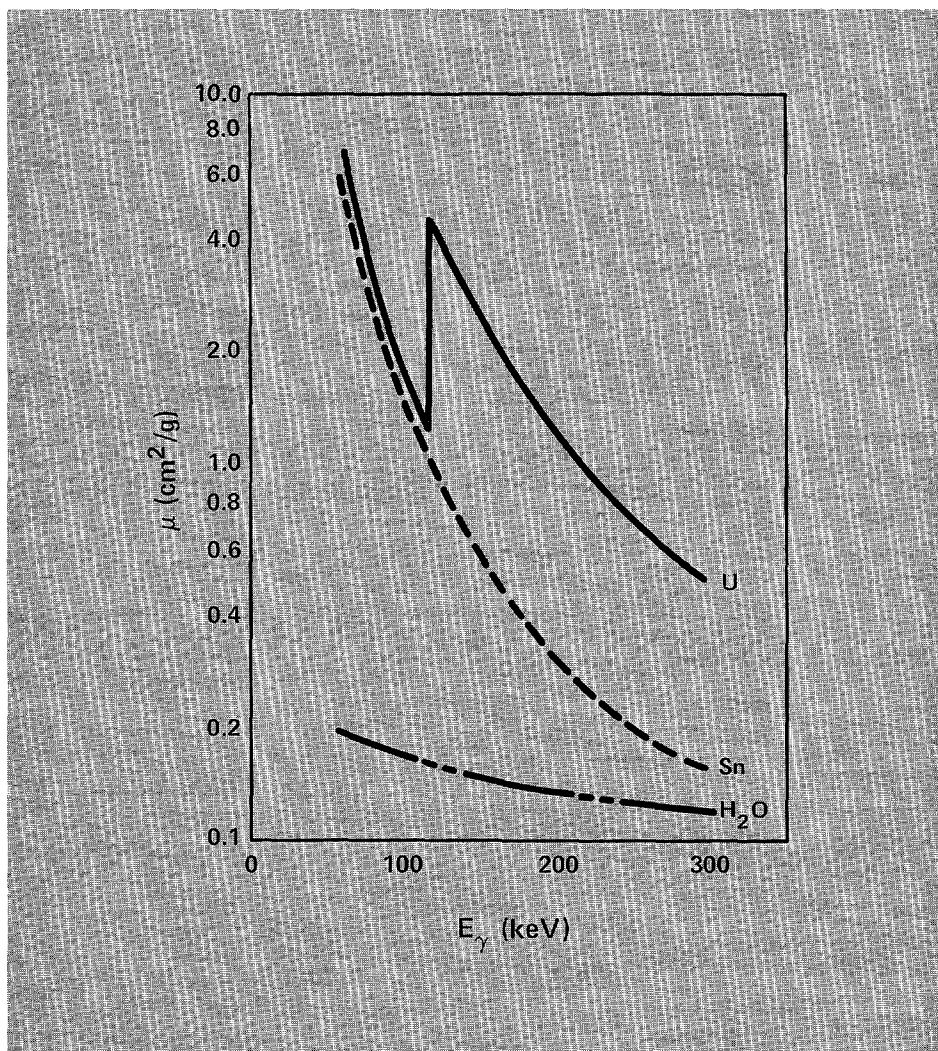


Fig. 11. Gamma-ray mass attenuation coefficient versus energy. The sharp K-edge discontinuity in the coefficient of uranium is contrasted with the monotonically varying coefficients of lower-Z materials.

TABLE II  
ABSORPTION EDGES AND  $\Delta\mu$  VALUES FOR URANIUM AND PLUTONIUM

Absorption Edge	Element	Edge Energy (keV)	$\Delta\mu$ ( $\text{cm}^2/\text{g}$ )
$L_{III}$	U	17.17	56.6
	Pu	18.05	51.9
K	U	115.6	3.65
	Pu	121.76	3.39

plement passive gamma-ray assays in that they measure total elemental content rather than specific isotopes. The sample is bombarded with an external beam of photons or particles at energies tailored to induce specific atomic interactions characteristic of the element, such as removal of an electron from an inner atomic shell. Traditionally, analytical chemistry laboratories have implemented x-ray edge densitometry and x-ray fluorescence with x-ray generators and high-resolution wavelength dispersive techniques. Our adaptation uses high-resolution gamma-ray detectors to measure uranium and plutonium concentrations in solutions. The measurements can be made either in the process line itself (in-line) or by removing a sample and making the measurement alongside the process line (at-line). These techniques can be very useful in reprocessing plants to assay low-concentration solutions containing both uranium and plutonium.

We will limit this discussion to x-ray edge densitometry. The technique utilizes the fact that gamma rays with energies just above the threshold for removing an electron from the K or  $L_{III}$  atomic shells of uranium and plutonium are absorbed with much greater probability than lower energy gamma rays. These thresholds create jumps in the mass attenuation coefficient, known as K and  $L_{III}$  edges. By bombarding a sample with gamma rays just above and below the K or  $L_{III}$  edges, and measuring the difference in transmissions, we can measure the amount of plutonium or uranium in the sample.

The K edge for uranium is shown in Fig. 11, together with the smoothly varying mass attenuation coefficients of lower-Z elements in the same energy region. Table II presents the positions of the energies of the K and  $L_{III}$  edges of uranium and plutonium and the magnitudes of the corresponding mass attenuation discontinuities,  $\Delta\mu$ .

The basic experimental arrangement for densitometry is shown in Fig. 12. Narrow-beam collimators eliminate the detection of multiple-scattered gamma rays and thereby minimize matrix effects. The transmission source is either an x-ray generator or appropriate radioactive isotopes. X-ray generators have the advantages of variable intensity and a continuous bremsstrahlung spectrum that can be tailored to cover the absorption edge, but radioactive sources are simpler and less expensive. High-resolution germanium detectors normally are used for K-edge measurements, whereas Se(Li) and germanium detectors must be used for L-edge measurements.

The transmissions  $T_1$  and  $T_2$  just below and above the absorption edge are related to the total concentration  $\rho$  of the element of interest by the relation

$$T_1 = T_{m_1} e^{-\mu_i \rho X},$$

where

$\mu_i = \mu_1$  or  $\mu_2$ , the mass attenuation coefficient of the element below and above its edges, respectively;

$T_{m_1}$  = corresponding transmissions of the matrix (all other materials); and

$X$  = sample thickness.

If the energies of the transmission gamma rays very closely bracket the edge, then  $T_{m_1} \cong T_{m_2}$ , and the concentration of the subject element is given by

$$\rho = \frac{-\ln T_2 / T_1}{\Delta \mu X}; \quad \Delta \mu = \mu_2 - \mu_1.$$

Corrections for effects of matrix materials may be applied when the transmission gamma rays are not so closely spaced.

X-ray edge densitometry measurements for multiple simultaneous elemental determinations are shown in Fig. 13.

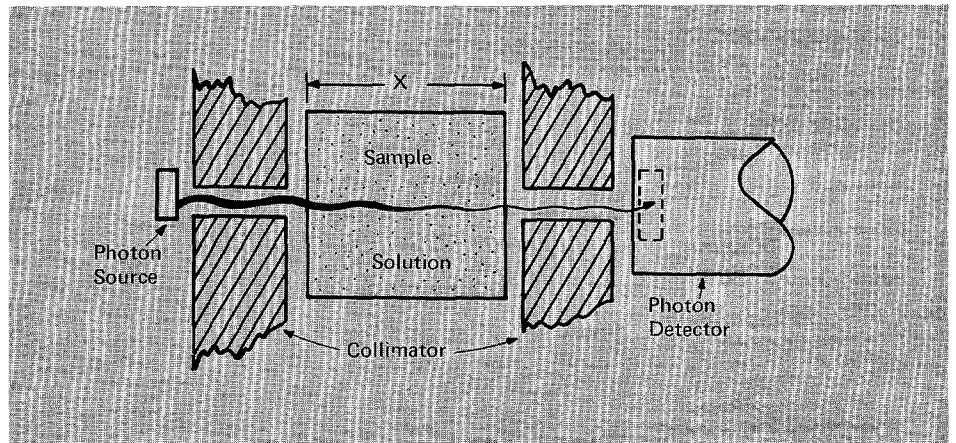


Fig. 12. Typical experimental setup for x-ray absorption-edge measurement of uranium and plutonium solution concentrations. The gamma-ray source is collimated, and the detector measures the intensity of gamma rays transmitted through the sample (thickness  $X$ ) at energies above and below the characteristic x-ray absorption edges of the elements of interest.

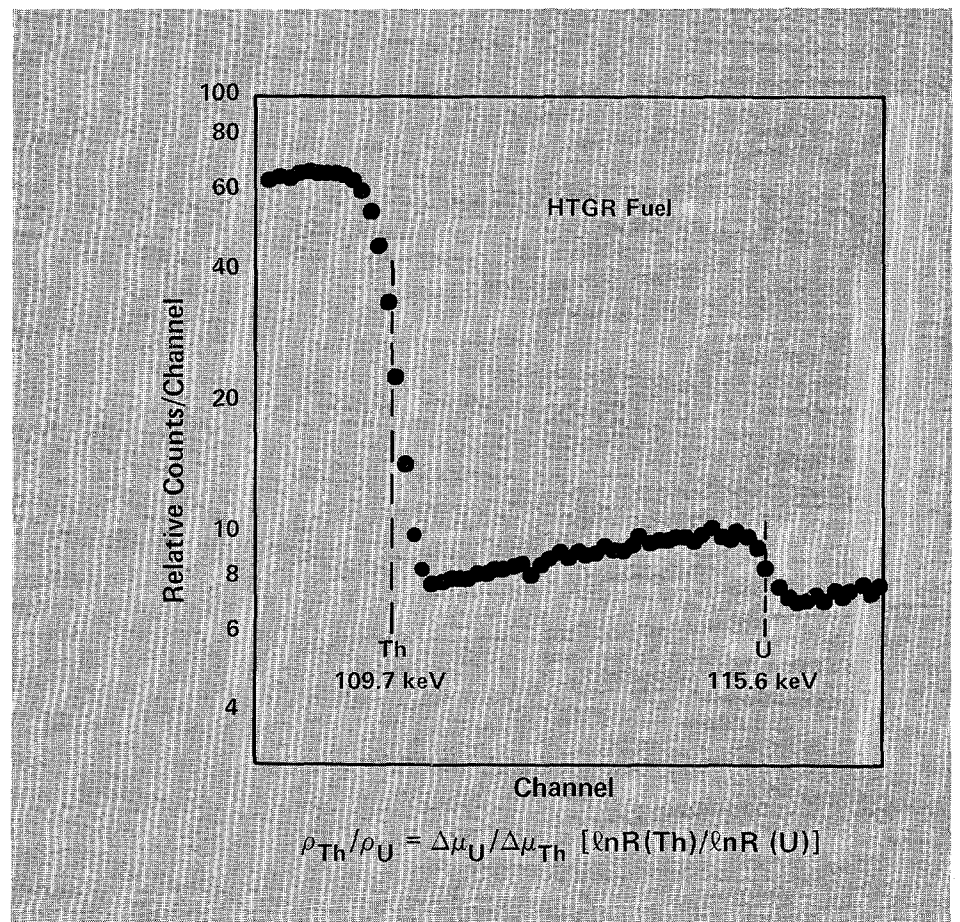
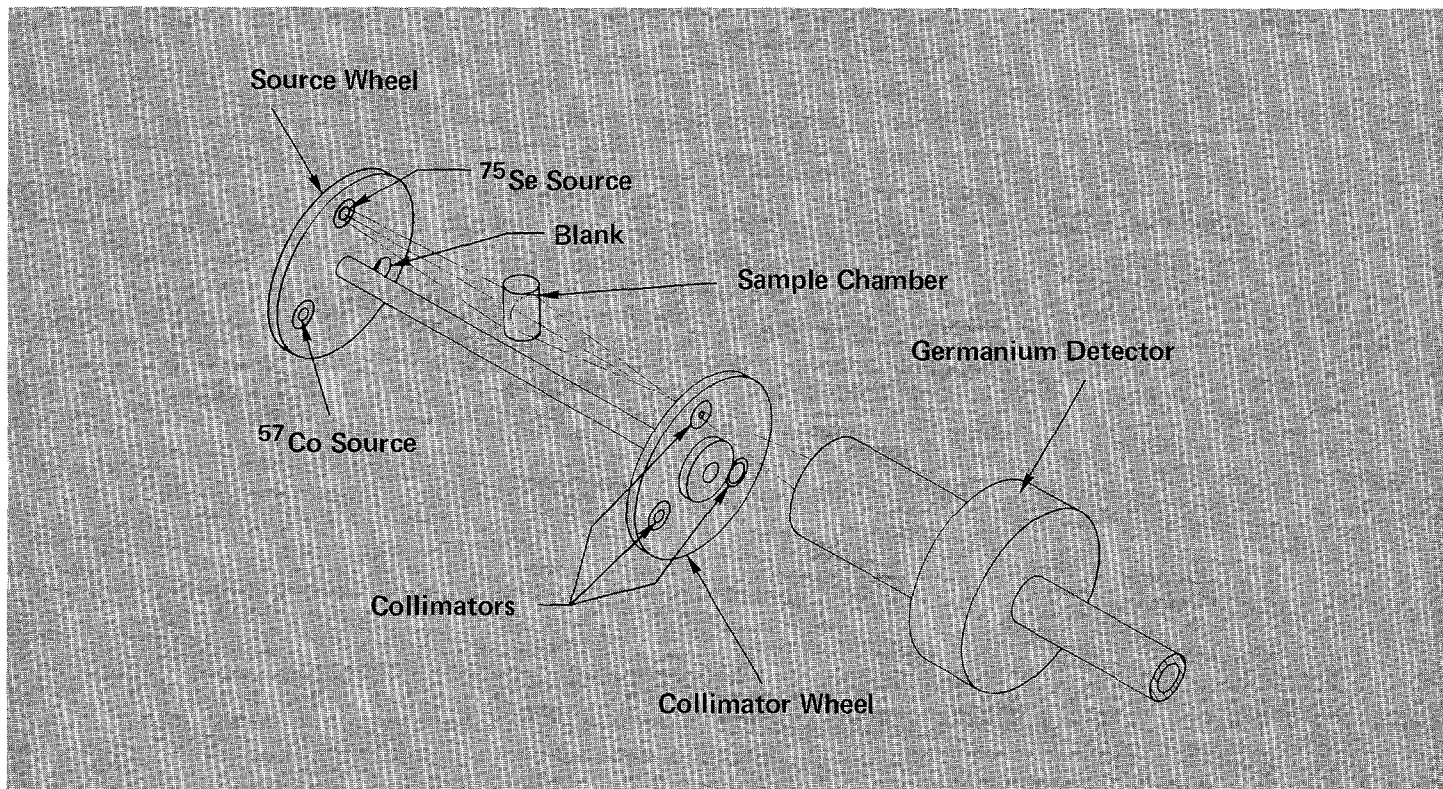


Fig. 13. Spectrum of x rays transmitted through a sample of high-temperature gas-cooled reactor fuel. The K edge of thorium at 109.7 keV and uranium at 115.6 keV are shown. Elemental concentrations are derived from the observed discontinuities at those edges.



**Fig. 14. Conceptual view of measurement configuration of the Tokai-Mura densitometer. The plutonium solution sample is shown between the source ( $^{75}\text{Se}$ , in this case) and the collimator in the detector line of sight. Sources and collimators are mounted on separate wheels driven synchronously.**

The x-ray spectrum is transmitted through a sample of high temperature gas-cooled reactor (HTGR) fuel, which contains thorium and uranium in a graphite matrix. The discontinuities produced by the K-absorption edges of both uranium and thorium are clearly evident, allowing accurate determination of the respective densities of thorium and uranium, in this case in proportion 4:1.

To resolve low-concentration solutions of uranium and plutonium, we must use the  $L_{III}$  edge with its larger  $\Delta\mu$  values rather than the K edge. The  $L_{III}$  edge is useful for solution concentrations ranging from 2 to 100 g/L. However, an x-ray generator is required for the transmission source because no adequate radioactive sources are available. We have developed an x-ray generator-based absorption-edge densitometer for the simultaneous measurement of uranium and plutonium in solutions con-

taining both elements. Some future reprocessing and nitrate-to-oxide conversion plants will coprocess uranium and plutonium in such solutions. We have conducted a preliminary test of this instrument, operating in-line, at the Savannah River Laboratory coprocessing demonstration facility, as well as at LASL. The instrument performed well for mixed solutions with plutonium concentrations between 2 and 10 g/L and uranium concentrations 4 to 10 times higher and is thus a candidate for use in advanced, near-real-time accounting systems for fuel reprocessing and conversion facilities.

For K-edge measurements of higher concentration plutonium solutions, we can use two discrete gamma-ray transmission sources, the 121.1- and 122.2-keV gamma rays from  $^{75}\text{Se}$  and  $^{57}\text{Co}$ , respectively. These gamma-ray energies very closely bracket the 121.8-keV electron-binding energy of the K

shell of plutonium.

We have recently developed such a densitometer to measure 150-350 g/L plutonium product solutions of the spent-fuel reprocessing in Tokai-mura, Japan. The instrument was installed and calibrated in this facility in the latter part of 1979 and is being evaluated for cooperative use by the facility operator and the IAEA for rapid off-line plutonium analysis. The Tokai densitometer rotates transmission sources in and out of position (Fig. 14) to measure the x-ray edge for plutonium concentration. This instrument also measures isotopic abundances by passive gamma-ray assay. Results obtained with the Tokai densitometer indicate that it will measure in times less than 1 hour plutonium concentrations to an accuracy of  $\pm 0.3\%$  (1 standard deviation) or better, and isotopic abundances in the accuracy range 0.3-1.5%, depending on the isotope and its abundance.