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NONDESTRUCTIVE ASSAY OF URANIUM ENRICHMENT WITH GAMMA RAYS

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ABSTRACT

An instrument has been developed and tested for nondestructive assay of ^{235}U enrichment of uranium oxide powder contained in sealed 1-gallon cans. Enrichment (ε) is measured from the count rate (D) of the 186 keV γ-ray of ^{235}U, using an HPGe γ-detector. A theoretical correlation of ε vs. D agrees well with the calibration measurements and provides guidelines for applicability. Measurements for 97 can samples with ε = 56 to 60 enrichment percent (e%) ^{235}U demonstrated accuracy of ~0.2 e% ^{235}U and precision of ~0.4 e% ^{235}U, with 10-minute count times. A microcomputer simplifies operator requirements and provides on-line enrichment results.

INTRODUCTION

Various techniques are available for assaying uranium used in nuclear fuels.[1,2] Because of potential handling hazards, nondestructive interrogation methods have been used frequently in these assays. One such method, passive gamma counting, has been successfully applied for measuring the ^{235}U enrichment of uranium oxide powders at the Savannah River Laboratory (SRL).

At SRL, much of the uranium oxide powder is stored in sealed 1-gallon (3.785 liter) steel cans. DOE requires that SRL make periodic physical inventories of the can contents. In the past, the cans were weighed for total uranium oxide content and ^{235}U enrichments were determined by mass spectroscopy. Unfortunately, the can seals had to be broken to obtain samples for mass spectroscopy. Opening the can introduces two potential problems: (1) personnel contamination and (2) uranium loss by sampling. In addition, mass spectroscopy is not an on-line or at-line method for SRL uranium oxides, and the time required for obtaining analyses is relatively long. Assay of ^{235}U enrichment by passive gamma counting does not encounter these problems.

In passive gamma counting, the ^{235}U enrichment is correlated with the count rate of the 186 keV γ-ray emitted by ^{235}U. Typical measurements with NaI(Tl) detectors have yielded enrichments with <2% relative error.[2] The present study used a high-resolution, high-purity germanium (HPGe) detector with the aim of improving the accuracy. In addition, on-line computer analyses were implemented to enhance operability.

Theoretical Basis

The detector count rate D for the 186 keV γ-ray of ^{235}U can be shown to be directly proportional to the ^{235}U enrichment ε, using the information of Fig. 1. For each point (X,Y,Z) in the detector, a γ-flux $\phi(X,Y,Z,\vec{\Omega})d\Omega$ from solid angle dΩ is experienced. (The unit vector $\vec{\Omega}$ passes through the middle of dΩ). Thus, the count rate for the detector is

$$D = \int_{V\Omega} \xi(X,Y,Z,\vec{\Omega})\phi(X,Y,Z,\vec{\Omega})dXdYdZd\Omega \tag{1}$$

where V = active volume of detector, cm^3

Ω = solid angle through which γ's are detected, steradian

$\xi(X,Y,Z,\vec{\Omega})$ = detector efficiency/volume/solid-angle-fluence, cm^{-1}-$steradian^{-1}$

The γ-flux is given by

$$\phi(X,Y,Z,\vec{\Omega})d\Omega = \int_0^\infty e^{-\int_0^R \mu(\vec{\Omega},R')dR'} \frac{s(\vec{\Omega},R)R^2d\Omega dR}{4\pi R^2} \tag{2}$$

where R = source-to-detector distance associated with incremental volumes, cm

$\mu(\vec{\Omega},R')$ = attenuation coefficient at position $(\vec{\Omega},R')$, cm^{-1}

$s(\vec{\Omega},R)$ = γ-source intensity density at $(\vec{\Omega},R)$, sec^{-1}-cm^{-3}

$R^2d\Omega dR$ = incremental source volume, cm^3

To evaluate the integral in Eq. (2), we first note that

$$e^{-\int_o^R \mu(\vec{\Omega},R')dR'} = e^{-\mu_d t_d(\vec{\Omega})} e^{-\mu_c t_c(\vec{\Omega})} e^{-\int_o^r \mu(\vec{\Omega},r')dr'} = A(\vec{\Omega})\, e^{-\int_o^r \mu(\vec{\Omega},r')dr'} \qquad (3)$$

where μ_d = attenuation coefficient of detector, cm^{-1}

$t_d(\vec{\Omega})$ = portion of R within detector, cm

μ_c = attenuation coefficient of can bottom wall, cm^{-1}

$t_c(\vec{\Omega})$ = portion of R within can bottom wall, cm

$\mu(\vec{\Omega},r')$ = attenuation coefficient at position $(\vec{\Omega},r')$ of source, cm^{-1}

r = portion of R within ^{235}U source region, cm

$A(\vec{\Omega})$ = attenuation along R not associated with ^{235}U source regions, unitless

Furthermore, we note that

$$\mu(\vec{\Omega},r) = \rho(UO_x)\Sigma(UO_x), \qquad (4a)$$

$$s(\vec{\Omega},r) = k\rho(^{235}UO_x) = k\varepsilon\rho(UO_x) = \frac{k\varepsilon\mu(\vec{\Omega},r)}{\Sigma(UO_x)} \qquad (4b)$$

where $\Sigma(UO_x)$ = molecular γ-cross section for uranium oxide (UO_x), cm^2

$\rho(UO_x)$ = UO_x molecular density at $(\vec{\Omega},R)$, cm^{-3}

k = specific activity per $^{235}UO_x$ molecule, sec^{-1}

ε = ^{235}U enrichment

Substituting Eq. (3) and Eq. (4) into Eq. (2), we obtain

$$\phi(X,Y,Z,\vec{\Omega}) = \frac{A(\vec{\Omega})k\varepsilon}{4\pi\Sigma(UO_x)} \int_o^\infty e^{-\int_o^r \mu(\vec{\Omega},r')dr'} \mu(\vec{\Omega},r)dr \qquad (5)$$

where dr has replaced dR with no loss of generality. Now, because we may write

$$I(\vec{\Omega},r) \equiv \int_0^r \mu(\vec{\Omega},r')dr', \quad \text{and} \tag{6a}$$

$$dI(\vec{\Omega},r)/dr = \mu(\vec{\Omega},r), \tag{6b}$$

the major integral in Eq. (5) reduces to

$$\begin{aligned} \int_0^\infty e^{-I(\vec{\Omega},r)}(dI(\vec{\Omega},r)/dr)dr &= \int_0^{I_{max}} e^{-I}dI = 1-e^{-I_{max}} \\ &= 1 \text{ for typical } I_{max} \end{aligned} \tag{7}$$

Normally, the cans will be filled to a height > several cm of UO_x. Thus, because $\mu(UO_x)$ is ~ 10 cm^{-1} for 186 keV γ-rays, the value of $e^{-I_{max}}$ is usually negligible.

Substituting Eq. (5) and Eq. (7) into Eq. (1),

$$D = \frac{k\varepsilon}{\Sigma(UO_x)} \int_{V\Omega} \frac{\xi(X,Y,Z,\vec{\Omega})A(\vec{\Omega})dXdYdZd\Omega}{4\pi}, \quad \text{and} \tag{8}$$

$$D = \frac{k\varepsilon}{\Sigma(UO_x)} \bar{\xi}, \tag{9}$$

where $\bar{\xi}$ is proportional to the detector efficiency for this geometry. For calibration purposes, we arrange Eq. (9) as follows:

$$\varepsilon = \frac{\Sigma(UO_x)D}{k\bar{\xi}} \tag{10}$$

Equation (10) illustrates that the ^{235}U enrichment ε is directly proportional to the 186 keV count rate D, regardless of the density fluctuations (due to packing) within the UO_x powder.

(This is true, provided that $e^{-I_{max}} \ll 1$, which is usually the case.) $\bar{\xi}$ is constant for reproducible counting geometries and k, the specific activity, is also constant. The value of $\Sigma(UO_x)$ can differ, depending on the oxide form; however, $\Sigma(UO_x)$ calculated for different cases shown in Table 1 agree within 2%. In the present work, UO_3 and U_3O_8 were examined and their $\Sigma(UO_x)$ differ by less than 0.2%. Table 1 indicates that moisture effects should not be severe.

Instrumentation and Procedures

The instrumentation for these γ-measurements is shown in Fig. 2. It consists of (1) a γ-detector and shielding, (2) detector electronics, and (3) a programmable multichannel analyzer.

The detector is a high-purity germanium (HPGe) detector supplied by ORTEC. It has a closed-end coaxial volume of 76.7 cm^3, resolution of 0.8 keV FWHM for the 186 keV gamma of ^{235}U, and an efficiency of 16.9%.* The detector views an area on the bottom of the 1-gallon uranium can, via the two lead collimator-shields shown in the figure. The lower collimator-shield reduces the room background, and the upper one defines the can area viewed by the detector. Tests showed that different areas of the same can produced indistinguishable 186 keV count rates. The procedure of visually centering the can over the collimator was adopted for the measurements.

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* Efficiency relative to 1333 keV γ-detection by 3" x 3" NaI(Tl) detector, with source-to-detector distance of 25 cm.

The detector electronics was supplied by ORTEC and included a low noise cryogenic charge-sensitive preamplifier, a Model 572 Spectroscopy Amplifier, a Model 459 High Voltage Supply, and a Model 729A Liquid Nitrogen Level Monitor. The amplified detector signals were processed by a Model 2048 Nucleus Multichannel Analyzer and Computer (MCA-computer).

The MCA-computer stores 2048-channel gamma spectra, which are analyzed by a BASIC program. The BASIC program was written to automatically count the sample for 10 minutes, calculate the intensity and statistical error of the 186 keV γ-peak, and then calculate and print out the enrichment. A typical spectral analysis of the 186 keV peak is outlined in Fig. 3. Calibration information is requested by the program before measurements commence. Once the program is initialized, the operator needs to answer only three questions for each sample. These questions concern the sample identification (name), the count mode (auto/manual), and the peak plot option (yes/no). Usually the samples are counted in automatic mode and the peak is not plotted. However, the 186 keV peak is plotted occasionally to check that it has not drifted significantly. The peak drift was insignificant, as it was less than 0.5 channels from the average during the measurement period of one week.

Measurements

The measurements involved (1) calibration with UO_x standards for the ε vs. D correlation and (2) assay of 97 cans for ^{235}U

enrichment. Each individual measurement was obtained with 10-minute counting time, with corrections for deadtime effects.

Calibrations involved measuring D for six UO_x samples of known ε ranging from 0.2 to 69.3 enrichment percent (e%) ^{235}U. Of these, four were U_3O_8 and two were UO_3 oxide standards. The resulting ε vs. D data are given in Fig. 4 and correspond to the least squares fit of

$$\varepsilon = (0.0222 \pm 0.0001)D - (0.003 \pm 0.247) \tag{11}$$

where the respective units of ε and D are e% ^{235}U and counts/minute. The calibration of Eq. (11) agrees well with the functional form predicted by Eq. (10).

The 97 cans assayed had enrichments ranging from 56 to 60 e% ^{235}U. The calibration of Fig. 4 was used to obtain the values corresponding to the D measurements. Table 2 compares the measured results with enrichment values ε_o which were obtained with mass spectroscopy at the time of original assay. Several different cans with the same ε_o were measured for their individual ε_i. For the n measurements of ε_i corresponding to a given ε_o, Table 2 includes the following entries:

$$\bar{\varepsilon} = \sum_{i=1}^{n} \frac{\varepsilon_i}{n} , \tag{12a}$$

$$\Delta\varepsilon = \bar{\varepsilon} - \varepsilon_o , \quad \text{and} \tag{12b}$$

$$\sigma = \sqrt{\frac{\sum_{i=1}^{n} \left(\varepsilon_i - \bar{\varepsilon}\right)^2}{n - 1}} \tag{12c}$$

where $\bar{\varepsilon}$ is the average γ-measurement corresponding to ε_o, $\Delta\varepsilon$ provides a measurement of accuracy, and σ corresponds to a measure of precision for an individual measurement. Including the data of all 97 cans yields $\Delta\varepsilon = -0.16$ e% ^{235}U and $\sigma = 0.44$ e% ^{235}U. For comparison, the σ due to counting statistics alone was 0.41 e% ^{235}U.

DISCUSSION

The passive γ-measurement technique for ^{235}U enrichment assay is an effective nondestructive method. Both operational efficiency and measurement accuracy are obtained with the method. However, limits for applicability should be recognized.

A single operator can use the instrument effectively after only 10 minutes of training. With 10-minute counting time and allowing for data readout and sample changing, uranium oxide cans may be assayed in 15-minute intervals. The 97 cans of the present study were assayed in one week, which is several times faster than that normally accomplished with mass spectroscopy.

A relative accuracy of 0.28% and a 1-σ precision of 0.76% is deduced for the results in Table 2. These estimates assume that the original enrichments ε_o are the true values. This is a reasonable assumption because the good agreement between ε_o and ε_i argues that the can contents have not been altered from their original amounts.

The accuracy probably could be reduced below 0.1% with suitable calibrations; however, the calibration standards and unknown samples should all have the same type of UO_x. For example, the systematic error of -0.28% for the 97 UO_3 measurements is due partly to calibrating with both UO_3 and U_3O_8 cans. Using Table 1, the average $\Sigma(UO_x)$ for the calibrations differ from $\Sigma(UO_3)$ by -0.13%. Thus, the calibration based on Eq. (10) yields UO_3 measurements of ε that would be low by this amount, if all other systematic errors were eliminated.

The precision can be improved by obtaining more 186 keV γ-counts. Normally, the can could be placed closer to the detector; however, this was not done in the present work because of significant deadtime effects caused by ^{228}Th in the uranium oxides. However, longer count times could be used to improve the counting statistics. For example, the precision could be improved by a factor of 2 by counting for 40 minutes. However, this is accomplished at the cost of a longer sample processing interval. The demonstrated precision of 0.76% is for ε in the 56 to 60 e% ^{235}U range. For other ε, the precision would be approximated as $(0.76\%)\sqrt{58/\varepsilon}$ for 10-minute counting. The counting error of 0.71% is the major influence on the precision.

It is worthwhile to appraise the above measurement results collectively. First, the source of the non-counting error contribution to ε is examined. To do this, we calculate the error of each term of $\overline{\Delta\varepsilon} = \bar{\varepsilon} - \bar{\varepsilon}_0$ for the average over all 97 measurements.

These errors are related as

$$\sigma(\overline{\Delta\varepsilon})^2 = \sigma(\bar{\varepsilon})^2 + \sigma(\bar{\varepsilon}_o)^2$$

or

$$\frac{\sigma_i(\Delta\varepsilon)^2}{97} = \frac{\sigma_i(\varepsilon)^2}{97} + \frac{\sigma_i(\varepsilon_o)^2}{20}$$

where the standard deviations of the means are calculated from the typical σ_i of individual measurements. (Note that only 20 measurements are recorded for the original ε_o values from mass spectroscopy.) With $\sigma_i(\Delta\varepsilon) = 0.76\%$ and assuming that $\sigma_i(\varepsilon) = 0.71\%$ due to counting statistics alone, the resulting value of $\sigma_i(\varepsilon_o) = 0.12\%$, which agrees well with the ~0.1% precision known for the ε_o measurements. Thus, the non-counting error is essentially caused by the uncertainty in the assumed true values of ε_o. It is also interesting to note that because $\overline{\Delta\varepsilon} \pm \sigma(\overline{\Delta\varepsilon}) = -0.28 \pm 0.08\%$, the residual $\overline{\Delta\varepsilon}$ after correcting for the known -0.13% for the $\Sigma(UO_x)-\Sigma(UO_3)$ effect is $-0.15 \pm 0.08\%$, which is within 2-σ of 0.0%. Thus, the observed discrepancies, which are relatively small to begin with, are well understood and can be addressed in further refinements of the method.

Applicability of these enrichment measurements is limited to homogeneous materials. However, as pointed out in Eq. (10), the material density does not have to be uniform. The bottom of the can containing the uranium must be similar to that of the calibration standards, so that the associated attenuation $A(\vec{\Omega})$ of Eq. (8) is constant as assumed in the analysis. It is best to both calibrate and assay using similar cans and contents, because of the strong attenuation for the 186 keV γ-ray. Also, the uranium attenuation limits the assay to detecting 186 keV γ-rays within a few mm of the bottom surface of the sample. Thus, if the assayed surface of the sample is not representative of the entire sample, the measured ε will not be reliable. Fortunately, the uranium oxide powders in 1-gallon cans do satisfy the above criteria. For less homogeneous samples, neutron interrogation with an Active Well Coincidence Counter (AWCC) yields much better results, because it probes the entire body of the sample.[4-5] However, when applicable, the γ-technique is attractive because its instrumentation is <50% as expensive as the AWCC.

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TABLE 1 $\Sigma(UO_x)$ Examples

U-Oxide	UO_x	$\Sigma(UO_x)$*	Relative Difference
UO_2	UO_2	$591.7\mathrm{x}10^{-24}$ cm^2	-0.56%
$UO_4 \cdot 2H_2O$	$UO_6(H_4)$	$605.9\mathrm{x}10^{-24}$ cm^2	1.84%
UO_3 (Samples)	UO_3	$595.0\mathrm{x}10^{-24}$ cm^2	0.00%
U_3O_8	$UO_{8/3}$	$593.9\mathrm{x}10^{-24}$ cm^2	-0.19%
$1/3(UO_3)+2/3(U_3O_8)$ (Calibration)	$UO_{25/9}$	$594.6\mathrm{x}10^{-24}$ cm^2	-0.13%

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* $\Sigma(UO_x) = \Sigma(U) + X\Sigma(O) + Y\Sigma(H)$, for UO_xH_y.

Individual $\Sigma(Z)$ obtained from Ref. 3 are:

$\Sigma(U) = 5.85\mathrm{x}10^{-22}$ cm^2

$\Sigma(O) = 3.35\mathrm{x}10^{-24}$ cm^2

$\Sigma(H) = 4.15\mathrm{x}10^{-25}$ cm^2

TABLE 2 Enrichment Measurements

	Enrichment Values (e% ^{235}U)*			
Can IDs	Original (ε_o)	Measurement** ($\bar{\varepsilon}$)	Difference ($\Delta\varepsilon$)	Sample Error (σ)
1-5	56.03	55.94	-0.09	0.47
6-10	56.34	55.85	-0.49	0.47
11-13	56.43	55.95	-0.48	0.12
14-18	56.90	56.73	-0.17	0.28
19-23	57.01	57.11	0.10	0.35
24-28	57.07	57.13	0.06	0.44
29-33	57.17	56.97	-0.20	0.24
34-38	57.35	57.32	-0.03	0.35
39-43	57.50	57.51	0.01	0.49
44-48	57.63	57.43	-0.20	0.39
49-53	57.90	57.87	-0.03	0.43
54-58	58.20	58.01	-0.19	0.35
59-63	58.28	58.25	-0.03	0.85
64-68	58.61	58.28	-0.33	0.60
69-73	58.85	58.84	-0.01	0.41
74-78	59.59	59.81	0.22	0.39
79-83	59.81	59.28	-0.53	0.16
84-88	59.85	59.31	-0.54	0.46
89-93	59.88	59.75	-0.13	0.31
94-97	60.03	59.74	-0.29	0.21
1-97	-	-	-0.16	0.44
			(-0.28%) relative	(0.76%) relative

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* Refer to Eq. (12) for details

** Average of cans identified in first column

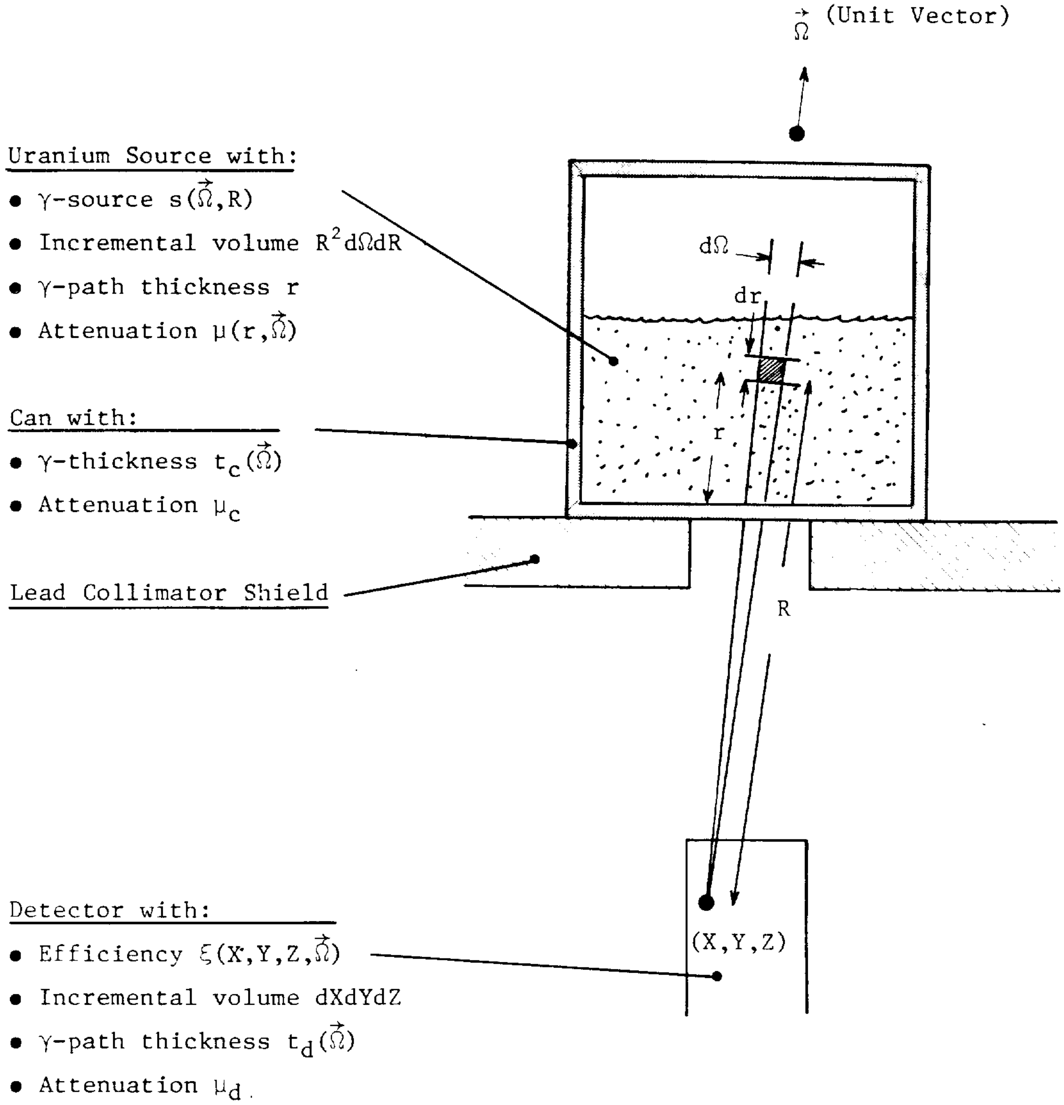

Figure 1 —— Geometrical features for γ-detection.

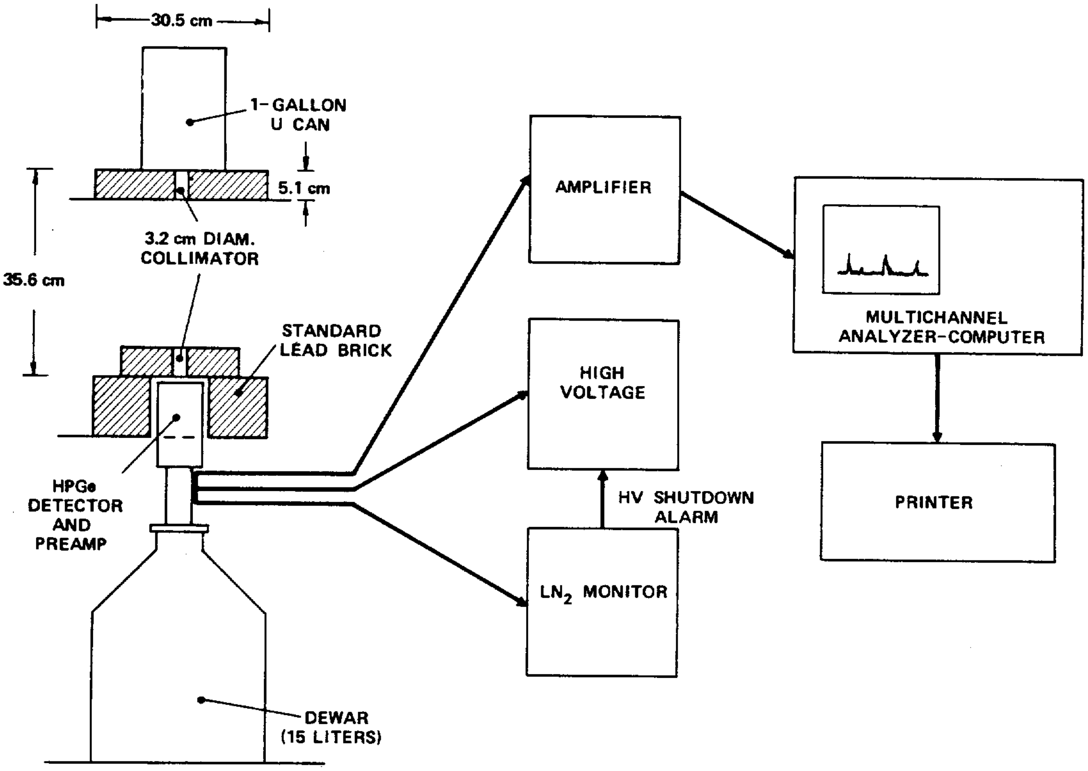

Figure 2 — ^{235}U γ-monitor instrumentation.

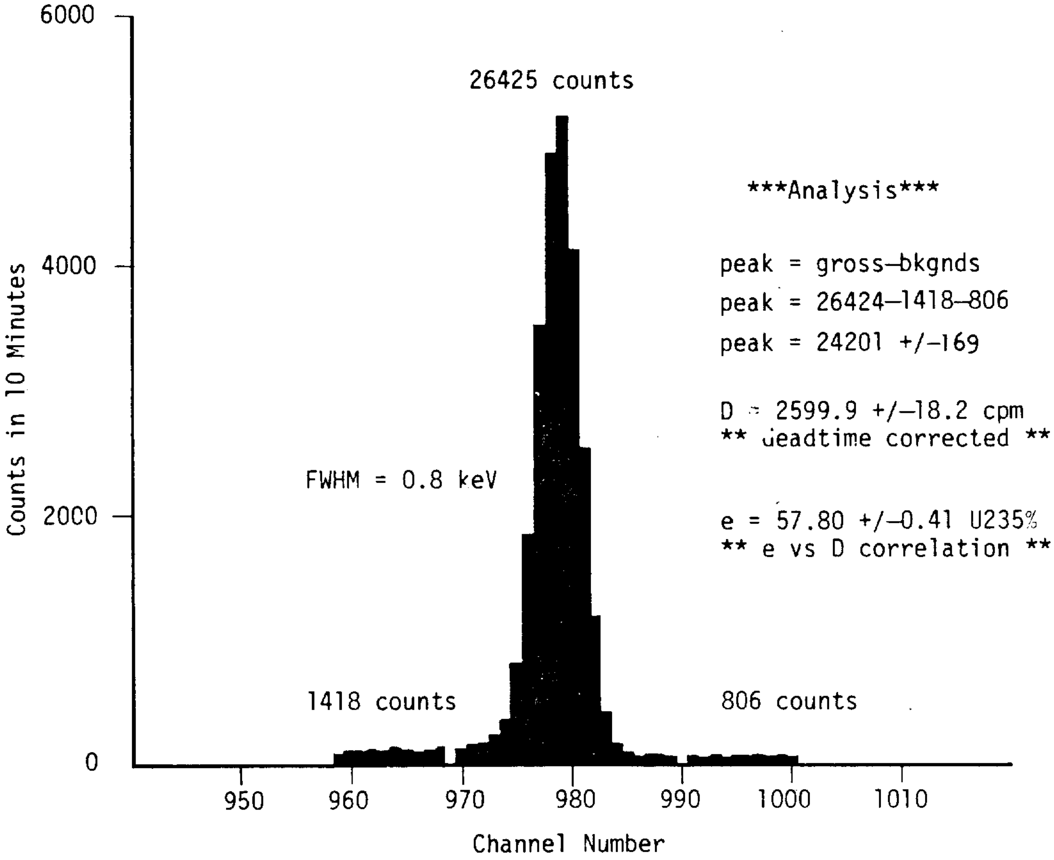

FIGURE 3. Analysis of typical 186 keV γ-peak.

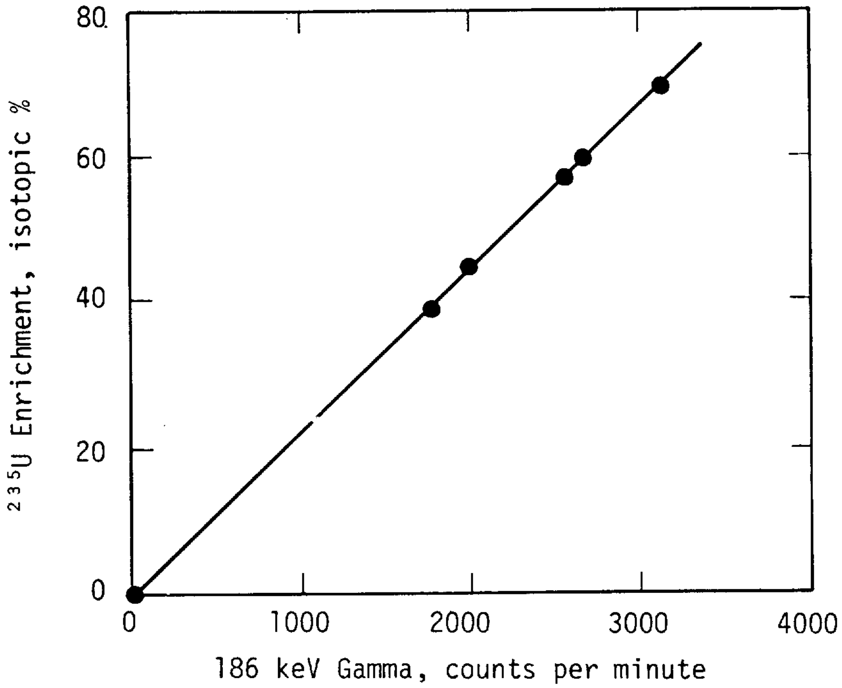

Figure 4 — ε vs. D correlation.

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