LA-UR-73-880
CONF-730630--3

MEASUREMENTS OF UF_6 CYLINDERS WITH PORTABLE INSTRUMENTS*

R. B. Walton, T. D. Reilly,
J. L. Parker, and J. H. Menzel

Nuclear Analysis Research Group
University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87544

and

E. D. Marshall and L. W. Fields

Nuclear Materials Control Branch
Nuclear Materials Management Division
U. S. A. E. C. Oak Ridge Operations Office
Oak Ridge, Tennessee 37830

ABSTRACT

The applicability of portable instruments for rapid nondestructive verification of the enrichment of UF_6 in cylinders has been tested on a large number of types 30- and 5A-cylinders. Three basic techniques were used: gamma counting with NaI combined with ultrasonic measurement of cylinder wall thickness, passive neutron counting, and active neutron interrogation with thermal neutrons from a radioactive neutron source. The resulting accuracies and limitations of the techniques are presented.

* Work performed under the auspices of the U. S. Atomic Energy Commission.

I. INTRODUCTION

Means for inexpensive, rapid, nondestructive verification of UF_6 cylinder content has been a subject of considerable interest and much speculation. The international market for enriched uranium fuel and interests in enrichment processes further emphasize the need for simple verification techniques for safeguards and materials control.

A limited test of the gamma enrichment meter method[1] for determining the enrichment of UF_6 in shipping containers was conducted two years ago at the Goodyear Atomic Enrichment facility (GAT), Piketon, Ohio.[2] Measurements on five 5A and two 48A cylinders, performed with a portable NaI instrument, indicated that accuracies of $\lesssim 10\%$ might be possible. Variation in container wall thickness was noted as a source of appreciable error for the gamma method.[3] Encouraging results for cylinder assay by the gamma method were also obtained by A. R. Flynn, Oak Ridge Gaseous Diffusion Plant (K-25), Oak Ridge, Tenn.[4] Another passive technique proposed for verification of UF_6 cylinders is the detection of neutrons emitted from $^{19}F(\alpha, n)$ reactions and the spontaneous fission of ^{238}U.[3,5] This technique for UF_6 assay is used routinely at GAT for detection of holdup in the enrichment cascade.[6]

Recently, as a part of a study of perimeter safeguards for enrichment facilities, LASL Group A-2 has measured passive neutrons and gamma rays emitted from cylinders of UF_6 representative of a wide range of enrichment and container sizes.[7] Additionally for these studies, an active neutron interrogation system, devised by Caldwell,[8] was successfully tested.

The purpose of the present work was to better define the accuracies and limitations of these techniques by testing them on a large number of UF_6 cylinders, primarily of type 30 ($2\frac{1}{2}$ ton), the standard shipping container for power reactor feed material. All measurements were performed with portable instrumentation in the field and with minimal intrusion on normal plant operation. Most of the data were collected in a total of seven days at K-25 and GAT.

II. PASSIVE NEUTRON ASSAY

A. Neutron Yield of UF_6

^{234}U is the dominant α emitter in enriched uranium and hence the principal source of neutrons from $^{19}F(\alpha, n)$ neutrons in UF_6. If the ratio of $^{235}U/^{234}U$ is known, passive neutron counting can be used for ^{235}U assay. This ratio may vary by as much as a factor of two over the complete range of enrichment, but for low enriched uranium ($< 5\%$), it is more nearly constant and verification measurements of limited accuracy may be possible without specific ^{234}U isotopic data.

Table I lists α activities and neutron yields of the uranium isotopes.[3] These data were used to calculate the neutron yields of the isotopes of normal uranium (0.71% ^{235}U and $^{235}U/^{234}U$ = 125) shown in Table II.[5] An approximate expression for the neutron yield of UF_6 is

$$S \text{ (neutrons/sec)} = M_U (0.0245 + 4.68\, I_{234}) \quad (1)$$

where

M_U = mass of U in g

I_{234} = ^{234}U isotopic abundance in %.

Estimates of the neutron source strengths of 5000 lb of normal UF_6 and 3% enriched UF_6 are 8×10^4 and 2×10^5 n/sec, respectively.

B. Instrumentation and Procedure

The passive neutron measurements were performed with two shielded neutron assay probes (SNAP) designed for use with commercial portable electronics. Principal features of this portable neutron probe are shown in Fig. 1. SNAP, including its shield, weighs ~ 20 lb; has a back angle shielding factor of 4.2 and 2.3 for background neutrons from $^{238}PuLi$ ($\bar{E}_n \simeq 0.6$ MeV) and ^{252}Cf ($\bar{E} \simeq 2.2$ MeV), respectively; and has an efficiency for a detector-source distance of 1 m of ~ 8×10^{-5} and 6.5×10^{-5} for neutrons from $^{238}PuLi$ and ^{252}Cf, respectively.

One of the SNAP units was mounted on a cart with additional shielding for measurement of passive neutrons from type-30 cylinders with the detector spaced far enough away to view an entire cylinder. For these measurements, so-called "L/2", the detector was placed half the cylinder length, or 41 in., from the cylinder axis, as shown in Fig. 2. The other SNAP was used for contact measurements, in which it was placed on the ground aiming toward the center of the cylinder at 4 o'clock and 8 o'clock, relative to a 12 o'clock vertical position. Placement for the contact measurement is shown in Fig. 3. A schematic of the contact measurement, including a cross sectional view of a cylinder with a 5000 lb UF_6 filling, is shown in Fig. 4. Both neutron probes were attached to Eberline SAM I electronic units.

The background-corrected counting rates observed for a type-30 cylinder containing 5000 lb of 3% enriched UF_6 were 4950 cpm and 1370 cpm for the contact and L/2 geometries, respectively, and backgrounds for both were usually less than 350 cpm, depending on the degree of isolation from other cylinders. Counts were recorded on each side of a cylinder and the results were averaged. Asymmetries in these counts were usually much less than 10%. Typically the counting time for each side of a cylinder was two minutes.

These two measurement geometries were selected for practical reasons such as counting rates, backgrounds from neighboring cylinders,

available space, and ease of placement. It was anticipated that the L/2 measurement would provide a measure of the total mass of ^{235}U according to the expression

$$S = M_{UF_6}\left[a + \frac{c}{r} I_{235}\right] \equiv M_{UF_6}\left[a + b\, I_{235}\right] \qquad (2)$$

where

M_{UF_6} = mass of UF_6
$r = {}^{235}U/{}^{234}U$ ratio
a, c = constants
b = constant if r is fixed.

Not only was good correlation achieved for the L/2 geometry, but the contact measurements also gave very good results using the same relationship. Because of the larger uncertainty resulting from the evaluation of the background from nearby cylinders for the L/2 measurement, it was abandoned--in favor of the faster contact measurements--after about 40 cylinders were measured. Nevertheless, the L/2 measurement results indicate that the L/2 geometry is very favorable for assay if cylinders are well isolated from each other.

In order to better understand the passive neutron assay method, and, in particular, the fortuitous contact measurement results, several calculations were performed with the Monte Carlo code, MCN.[9] Since the $^{19}F(\alpha, n)$ thick target energy spectrum is not well known, two source neutron spectra were used, one with single energy neutrons at 1 MeV and the other a fission spectrum. Figure 5 shows the neutron leakage spectrum from a 5000 lb cylinder (3%) assuming a uniform spatial distribution of 1 MeV neutrons, and the corresponding results for the fission spectrum source are shown in Fig. 6. The neutron moderation by the UF_6 is quite marked for both source spectra; thus, it is apparent that a $2\frac{1}{2}$-ton cylinder is a distributed source of neutrons with energies primarily in the range 100 eV -- 100 keV.

The neutron leakage factor, defined as the fraction of the source neutrons which escape from the cylinder, was also calculated as a function of weight and enrichment of UF_6 in $2\frac{1}{2}$-ton cylinders. For a fixed weight of 5000 lb, the leakage factor changes from about 0.80 to 0.84 as the enrichment varies from 1% to 4%. The leakage factor is a stronger function of cylinder weight, varying from about 0.93 to 0.82 as the weight of $2\frac{1}{2}$% enriched UF_6 is increased from 2000 to 5000 lb. The overall effect of weight variation on the passive neutron assay is reduced because the neutron leakage spectrum is harder for lower weights and the detection efficiency decreases with increasing neutron energy.

The spatial distribution of the neutron leakage was also calculated to determine whether the average of the 4 and 8 o'clock contact measurements (cf. Fig. 4) is a direct measure of UF_6 weight. The neutron

leakage flux per source neutron at the 4 o'clock position was constant to within 7% for weights from 3000 to 5000 lb. Results for this weight range also showed that the averaging procedure cancels asymmetries caused by cylinder rotations up to ± 60° (2 h on the clock reference). Although most of the cylinders measured were in the range 4400 to 4900 lb, results for a few cylinders with weights in the range 1140 to 3000 lb indicate that even for the lighter cylinders, the contact method may be accurate to within about ± 15%.

C. Results

A total of 123 $2\frac{1}{2}$-ton cylinders were measured with the passive neutron assay technique. The enrichment distribution (number versus enrichment) of these cylinders is shown in Fig. 7.

Since there was no prior calibration procedure, the data observed for cylinders of two different known enrichments were used to determine the constants in the following expression (cf. Eq. (2)) relating the specific counting rate s, counts $[(\text{min})(\text{lb } UF_6)]^{-1}$, to the ^{235}U enrichment $I_{235}(\%)$:

$$s = a + b\, I_{235} \qquad (3)$$

For the analysis of data for enrichments greater than 1%, results for K-25 cylinders, 10 with 3.08% enrichment, and three with 2.17%, were used to determine the constants:

$$a = 0.0686 \text{ cpm/lb } UF_6$$
$$b = 0.324 \text{ cpm}/(\text{lb } UF_6 \times \%\,^{235}U)$$

A simpler procedure, which was found to be almost as accurate for enrichments greater than 1%, is to use one cylinder with enrichment between 2 and 3% to determine a single calibration constant d in the expression:

$$s = d\, I_{235}$$

The passive neutron assay results for all (91) of the licensee cylinders ($2\frac{1}{2}$ ton) measured at GAT and K-25 are presented in Fig. 8. These results are given as distributions in relative error, i.e. number of occurrences versus $(I_M - I_{Tag})/I_{Tag}$, where I_M is the measured ^{235}U enrichment and I_{Tag} is the certified value. The tag value of the net UF_6 weight, together with the measured neutron counts and the calibration constants a and b, was used to obtain I_M. The standard deviations for the K-25 and GAT data are 5.5% and 4.3%, respectively, and the results for the GAT cylinders show a bias of ~ 2.4% relative to the K-25 cylinders. This bias probably reflects a slight difference in the $^{235}U/^{234}U$ ratio for these two enrichment facilities, since the calibration was made with K-25 cylinders. It is also likely that the variation in the $^{235}U/^{234}U$ ratio also contributes

significantly to the widths of the distributions, because measurement precision was ~ 1.5%, and most of the cylinders were filled near the limit, giving uniform measurement geometries.

Figure 9 gives the results for the 91 licensee cylinders and 19 storage cylinders, all having enrichments between 1.3 and 4%. Again the same calibration was used to analyze all the data. The additional storage cylinders have a greater weight variation and probably also a greater variation in age from withdrawal (and hence $^{235}U/^{234}U$ ratio) than those of the licensee cylinders. Nevertheless, the standard deviation for this distribution is only 6.5% and there were only three measurements with errors exceeding 16%.

Results of passive neutron measurements on $2\frac{1}{2}$-ton cylinders of depleted material are presented in Table III. These cylinders were in storage and represented an age span of several years. In this case a new calibration was derived using 3.08% and 0.21% cylinders as standards and 0.106 and 0.312 as the values of a and b, respectively. Similar results were obtained for 10- and 14-ton cylinders of depleted UF_6.

Measurements of neutron yields of 25 5A cylinders with enrichments representative of the range from 1.5 to 97% reflected, as anticipated, variations in the $^{235}U/^{234}U$ ratio of ± 50%.

III. GAMMA-RAY ENRICHMENT METER MEASUREMENTS

A. Basis of the Measurement

If the concentration of uranium in a medium (which is opaque to the ^{235}U 186-keV gamma ray) is large and uniform, the 186-keV gamma activity, measured with a collimated detector, is essentially a direct measure of the enrichment of ^{235}U. The count rate of this gamma ray is:

$$C = I \frac{\Gamma \varepsilon A}{\mu_U} F, \qquad F = \left[1 + \frac{\mu_m \rho_m}{\mu_U \rho_U}\right]^{-1} \tag{4}$$

where

I = ^{235}U isotopic enrichment
Γ = specific activity of the 186-keV gamma ray (γ/sec per g ^{235}U)
ε = detection efficiency
A = area of sample viewed
μ_U, μ_m = 186-keV mass absorption coefficient of uranium, and the matrix material, respectively.
ρ_U, ρ_m = density of uranium, matrix material.

$\Gamma \varepsilon A/\mu_U$ is a constant and F equals 0.962 and 0.985 for UF_6 and U_3O_8, respectively. This method has been very successfully applied with NaI and Ge(Li) detectors to U metal, fuel pins and plates, cans of uranium oxide feed material, etc., for the range of enrichments from 0.7 to 97%.

B. Instrumentation

An Eberline SAM II portable gamma-ray assay unit was used for the measurements of enrichment of UF_6 in cylinders. The gamma-ray probe consists of a $\frac{1}{2}$-in. thick by 2-in. diam NaI and photomultiplier tube housed in a tungsten and lead collimator. This size crystal is optimum for enhancing the photopeak of 185-keV ^{235}U gamma rays relative to the background activity which arises principally from Compton scattering of the 765- and 1001-keV gamma rays from ^{234m}Pa, a daughter of ^{238}U. The unique feature of this unit is the use of α-particle pulses from an ^{241}Am-doped NaI seed implanted in the crystal for gain stabilization. Another feature of the electronic unit which is convenient for enrichment measurements is the availability of two separate single channel analyzers (SCA) with an up-down scaling option for automatic background subtraction.

A schematic of the NaI probe and the collimator for this study is shown in Fig. 10. A 0.030-in. thick Cd foil was placed over the front surface of the NaI detector to attenuate x rays from the uranium as well as secondaries produced in the collimator. For all measurements, the collimation depth was 3 in., as measured from the front surface of the collimator to the surface of the NaI unit. The collimated NaI probe, an Eberline SAM II, and a Branson Model 104 portable ultrasonic thickness gage were mounted on a push cart for the field measurements. Figure 11 shows this field system as it was used to measure the enrichment of $2\frac{1}{2}$-ton UF_6 cylinders.

C. Calibration

To measure enrichment with a portable NaI system, the net ^{235}U counting rate is obtained from a two-window analysis with one window bracketing the 186-keV peak and the other set just above the peak to evaluate the background. The expression for ^{235}U enrichment I is then:

$$I = aC_a - bC_b \tag{5}$$

where

C_a = counts in peak window
C_b = counts in background window
a, b = constants obtained from measurement of two standards of different enrichments.

The absorption of gamma rays by container walls must be taken into account by calibration with standard containers and/or multiplying the above expression by $e^{\alpha t}$, where t is the wall thickness and α is the effective linear absorption coefficient for the nominal thickness of interest. Since the peak window accepts 186-keV gammas Compton-scattered by the container walls, α depends on the wall thickness, as well as the measurement geometry. A variation in wall (Fe) thickness of .010 in. reflects a relative change of $\sim$ 2.5% in the measured enrichment.

Calibrations for measurements of the $2\frac{1}{2}$-ton cylinders were performed in the laboratory (LASL) using two 4-in. diam cans of U_3O_8 (10% and 0.71% ^{235}U) and iron plates to simulate the container walls and to determine α for iron. α for Monel was also determined in preparation for measurements of UF_6 in 5A cylinders ($\frac{1}{4}$-in. thick Monel wall).

The two-window calibration used for this study is illustrated in Fig. 12. The plots are pulse height spectra of 1.93% U_3O_8 with no absorber and with 0.5 in. of iron simulating the nominal thickness of a 30B cylinder. A well aged (relative to chemical separation) U_3O_8 sample was used so that the high energy background gammas from ^{234m}Pa were in equilibrium. It is apparent that the iron almost completely suppresses the x rays and also distorts the 186-keV photopeak.

Figure 12 also shows the positions and widths of the photopeak and background windows. For the field measurements, the width of the background window was adjusted so that the constants a, b in Eq. (5) were equal. This was done using the two standards (10% and 0.71% ^{235}U) for iron absorbers from $\frac{1}{4}$ to $\frac{3}{4}$ in. thick, and Monel plates 0.2 and 0.3 in. thick. For all cases, including no absorber, the following set of window widths, W_A, W_B, and SCA thresholds E_A, E_B, gave the equality a = b:

$$W_A = 49 \text{ keV}$$

$$W_B = 1.424\, W_A = 69.8 \text{ keV}$$

$$E_A = 162 \text{ keV}$$

$$E_B = 235 \text{ keV}$$

Thus the up-down scaling feature of the SAM II could be used for automatic background subtraction with the following calibration formula:

$$I = a_o R e^{+\alpha t} \qquad (6)$$

where

R = net counting rate, $C_A - C_B$, observed through wall thickness t
a_o = calibration constant.

The measured value of α for iron in the thickness range from 0.4 to 0.6 in. is 2.42 $in.^{-1}$, while that of Monel in the range 0.2 to 0.3 in. is 2.97 $in.^{-1}$.

The accuracy of this calibration was approximately 3% over the relevant range of absorber thicknesses, as established by measurements with U_3O_8 standards with enrichments of 0.71, 1.93, 3.02, and 10.0%. For a counting time of 5 min, the statistical uncertainties in the net counts with a $\frac{1}{2}$-in. thick iron absorber were 4, 1.5, 1, and 0.5% for ^{235}U enrichments of 0.71, 1.93, 3.02, 10.0%, respectively. The net counting rate per $\%^{235}U$ with the $\frac{1}{2}$-in. thick iron absorber was 1730 cpm, as contrasted with

a rate of 6936 cpm in the background channel. A correction to the calibration was made for the small difference between UF_6 and U_3O_8, according to Eq. (4). Corrections for the thickness of the actual cylinder walls, measured with the ultrasonic thickness gage, were calculated during the course of the measurements. The uncertainty in the thickness measurement was about ± .002 in.

D. Results for 2½-ton Cylinders

Enrichment results were obtained for 107 30A and 30B cylinders of UF_6 with enrichments between 1.3 and 3.1%. With the automatic background subtraction and a 5-min count, a measurement rate of six cylinders per hour was achieved. The wall thickness measurement took only a few seconds but required sanding the paint off a spot within the area viewed by the gamma probe.

The laboratory calibration, based on measurements with U_3O_8 standards, gave results for the first few cylinders measured which were about 3% too high, so the calibration constant was adjusted by this amount for the remainder of the measurements.

Figure 13 shows the results for 98 of the 107 cylinders. Truncating the distribution at ± 16% relative error because of obvious outliers, one obtains a standard deviation for the distribution of 5.3%. The 17 outliers (with relative assay error > 16%) were easy to recognize because measurements at different points on an outlier cylinder gave varied results, in contrast to a "well behaved" cylinder. Quick confirmation of an outlier was also obtained from the passive neutron measurement.

Table IV is a list of the outliers observed in the gamma assay of 107 cylinders. Except for one result, -17%, all are larger than the tag value.

Among the possible reasons for the observed failures of the assay method are:

1) Age of UF_6 from withdrawal--The ^{234m}Pa activity grows into equilibrium essentially with the time constant of its precursor ^{234}Th ($T_{\frac{1}{2}}$ = 24.1 d), the ^{238}U decay product. If the shape of the background spectrum is not time-dependent, then the two-window analysis method should adequately account for the background, even for large changes in the background intensity. Good results were obtained for cylinders aged from less than 48 h to a few years after filling.

2) Presence of ^{233}Pa and fission products from UF_6 product from reprocessing of spent reactor fuel--Although gamma rays from ^{137}Cs and ^{233}Pa (a daughter of ^{237}U) have been observed in the Ge(Li) spectra of

gamma rays from cylinders,[7] only five cylinders in the present study were known to contain reactor returns and the assay results for these were very good.

3) Uranium in the "heel" remaining from the previous filling--A uranium deposit, either in volatile form (UF_6) or as UO_2F_2, UF_4, or UF_5, if present on the walls and sufficiently thick could, because of the short mean free path of the 186-keV gamma (~ .075 in.), mask the signal from the actual UF_6 filling. The magnitude of the effect of a heel with enrichment differing from that of the fill has been estimated, and the results are shown in Fig. 14. To simplify the calculations, it was assumed that the U heel was uniformly deposited over the surface covered by the filling. If, for example, a heel with 3% enrichment weighs 25 lb, the maximum allowable limit, and the enrichment of the UF_6 filling is 2%, then the gamma assay will be ~ 16% too high because of the heel. Data on the heel weight and enrichment of all the licensee cylinders measured at K-25 showed no correlation with the relative measurement errors, including the outliers.

4) Plating of ^{234}Th from the previous UF_6 filling--It is well known that this nonvolatile component of the heel is deposited when a cylinder is emptied (by volatilization of the UF_6) and is largely responsible for significant radiation levels frequently observed for empty cylinders. Because all of the ^{234}Th which was in equilibrium with the previous UF_6 filling can be concentrated on the walls, the background radiation from this source (its daughter, ^{234m}Pa) may be much greater than that from the ^{234m}Pa which grows into the new filling uniformly and is therefore strongly self-shielded. The amplitude of this background will depend on the spatial distribution of the heel, which generally has highest concentration on the bottom of the cylinder, and the age, or "turn-around time", of the cylinder. Not only can the background intensity vary because of plated activity, but the shape of the gamma ray spectrum may be different because the ^{234m}Pa is located at the interface of the cylinder wall and the UF_6, in contrast to the uniformly distributed ^{234m}Pa associated with the fill. Two processes which would be sensitive to the location of the material are Compton scattering of the high energy gammas from ^{234m}Pa and Bremsstrahlung production from the 2.3-MeV beta particles emitted copiously in the decay of ^{234m}Pa. A change in the shape of the background spectrum would, of course, cause a significant assay error.

Of the above possible sources for the observed outliers in the gamma assay, the last, namely plated ^{234}Th-^{234m}Pa, is the most likely. Gamma spectra from three outlier cylinders and a well behaved one, measured with the gamma assay unit using a narrow SCA window, showed no extraneous peaks in the range up to 800 keV and were consistent with the distorted background explanation. Additionally, for five sets of cylinders, each set filled from a single large storage cylinder, there were in each set both outliers and well behaved cylinders, indicating that the problem is associated with the cylinder heel and not the UF_6 fill.

Use of a Ge(Li) gamma-ray detector, which has a resolution ~ 20X greater than NaI, should obviate problems of background caused by plated ^{234}Th-^{234m}Pa (and other possible sources of radiation), and thereby obtain complete reliability for the gamma enrichment meter.

E. Results for 5A Cylinders

The 5A cylinder has a diameter of only 5 in. and is used for storage of highly enriched uranium. Because of the greater 186-keV activity relative to the background and the thinner wall (0.25 in.) of this cylinder, the reliability and accuracy of the NaI enrichment meter for these cylinders are much better than for the type-30 cylinder.

Results for 24 5A cylinders covering a range of enrichments from 3.1 to 97.7% are presented in Table V. The counting time for each assay was one minute and the cylinder assay rate, including the wall thickness measurement and cylinder placement, was 20 per hour. One Monel cylinder with 58.3% enrichment was designated as the standard. The attenuation correction of the iron cylinders was calculated relative to the Monel standard using an intercalibration of the thickness gage for the two materials. The precision of the results for cylinders with enrichments less than 7% was limited by the statistical uncertainties incurred in the 1-min count. Excluding the four cylinders with enrichments less than 7%, the standard deviation of the results for the remaining 19 cylinders is 2.1%. Excluding the data for the two iron cylinders in this group, a standard deviation of 1.8% is obtained.

IV. ACTIVE NEUTRON ENRICHMENT METER

The present measurements were undertaken to extend the development of Caldwell's assay system,[8] adapting it for portable instrumentation and concentrating on application of the technique to low enriched UF_6 in $2\frac{1}{2}$-ton cylinders. The Caldwell unit for enrichment assay utilizes low energy neutrons, from a radioactive source and moderating assembly, to produce ^{235}U fissions and energy-biased ^{4}He gas proportional counters for detection of the prompt fission neutrons.[9] Principal modifications to Caldwell's system were the addition of more moderator around the source to enhance the thermal fraction of the interrogating neutrons and the implementation of a cadmium subtraction technique to obtain a net response from thermal neutron fission only.

The use of strictly thermal neutrons for the assay is advantaneous because the region of the cylinder interrogated is confined to the thermal neutron mean free path (~ few inches), thus essentially eliminating variations in response from different fill heights. The Cd subtraction technique also provides an operationally simple method for subtracting fast neutron backgrounds from the radioactive source, from the cylinder being measured, and from neighboring cylinders.

Figure 15 is an isometric drawing of the active neutron system used for this study. A polyethylene moderating block, indicated by the sliding arrow in the figure, contains a 5×10^5 n/sec Am-Li neutron source which is enclosed in a 5-in. long, 2-in. diameter tungsten shield. The source is surrounded in all directions with $1\frac{1}{2}$ to 2 in. of polyethylene, the thickness in the direction of the UF_6 cylinder being 2 in. The source-moderating assembly is located between an array of four 2-in. diameter, 24-in. long (active), 18-atmosphere ^4He counters which are connected in parallel. An Eberline SAM I electronic unit supplies the high voltage ($\sim$ 1900 V) and processes the pulses.

To perform an assay, the unit is placed in contact with the cylinder and as far under the cylinder as possible, and measurements are then performed with and without a cadmium shield between the assay unit and the cylinder. The difference between these two counts is the response from thermal neutron interrogation.

The compositions of the signal and background are shown in Fig. 16, which presents plots of integral counting rates as a function of pulse discriminator (integral bias) setting. The background as obtained with cadmium, denoted as "active Cd" in Fig. 16, results principally from neutrons from epithermal (but subthreshold for ^{238}U) fissions in the UF_6 being assayed and the high energy neutrons from the Am-Li source. The passive neutrons from the UF_6 (see Fig. 16, "passive no Cd"), as measured with the source removed from the assay assembly, are a weaker background. Although the net signal (active no Cd minus active Cd) is only about 40% of the background, the signal and the background from epithermal fissions are essentially proportional to the enrichment, and consequently meaningful results can be obtained even for highly depleted U. The constant background from the source, although a measurement impediment, provides a convenient internal standardization to correct for instrument drifts. A discriminator setting of 2.0 was used for all field measurements.

B. Results and Interpretation

Data were accumulated for 38 type-30 cylinders at a rate of four per hour. Two 2-min counts, one with and one without Cd, were taken on each side of a cylinder.

The results of these measurements expressed as net counts versus UF_6 enrichment, are shown in Figure 17. It is apparent that a straight line fit (least squares) adequately represents the response up to 3.0% enrichment. The dispersion of individual points probably resulted from the sensitivity to the physical positioning of the assay unit relative to the cylinder--a problem which could be eliminated by designing the assay unit with a contour matching the cylinder. In spite of the scatter of the data, the single point obtained for an enrichment greater than 3.1%, namely 3.95%,

The response calculated with this simple model is indicated by the dashed curve in Fig. 17. Qualitatively, the calculations predict the effective saturation of the response at 4% enrichment. The rapid increase in the fast neutron path length in the iron cylinder wall, i.e., $t/\cos\theta$, contributes to this saturation effect.

The Los Alamos Monte Carlo Code MCN[10] was also used to predict the response of this assay system for four discrete enrichments; the results, shown in Fig. 17, are consistent with the measurements and corroborate results of the simple analytical model.

This analysis indicates that the response of the active thermal neutron method is a nonlinear function of enrichment and depends sensitively on the physical layout of the source, detector, and sample. On the other hand, it may be possible to use the simple analytical model, in lieu of expensive Monte Carlo calculations, to optimize the configuration of the active neutron method for specific applications.

V. CONCLUSIONS

Of the three methods investigated, passive neutron counting is the fastest, simplest and least expensive for verification of low enriched UF_6. The standard deviation of results obtained for 110 type-30 cylinders is 6%, indicating that for this group of GAT and K-25 cylinders, the $^{235}U/^{234}U$ ratio is almost constant. Nevertheless, since the $^{235}U/^{234}U$ ratio for product in this enrichment range can vary significantly more than this study might imply, the passive neutron counting method, used by itself, would be correspondingly less accurate. If either the specific neutron yield of the UF_6 or the ^{234}U isotopic abundance were included in the cylinder specifications, passive neutron counting would provide a very accurate verification procedure. The specific neutron yield could be obtained at the originating product station by counting the neutrons from either a sample of UF_6 or the actual cylinder. Neutron counting also provides a nondestructive method for ^{234}U isotopic analysis, providing ^{238}U and ^{234}U are the dominant neutron sources.[5]

The NaI gamma enrichment meter, together with the ultrasonic thickness gage, can be used to measure the enrichment of $2\frac{1}{2}$-ton cylinders to an accuracy of $\sim 5\%$ (1σ); however, based on results obtained for 107 cylinders, this method fails (relative error $> 16\%$) about 15% of the time. An outlier is easy to recognize because measurements at different points on the cylinder give significantly different results. Information on cylinder heels, cylinder fill dates, and gamma spectra, together with some calculations, indicate that outlier results are probably caused by the background radiation from ^{234}Th-^{234m}Pa deposited on the cylinder walls when the cylinder was emptied of its previous UF_6 fill. The shape of this background spectrum should differ from that of ^{234m}Pa uniformly distributed in the UF_6 and would therefore cause an error in the simple two-window

analysis. Use of a high resolution detection system should obviate this problem and give complete reliability to the method. For higher enrichments of UF_6, stored in 5A cylinders, no problems were encountered with the NaI system and the standard deviation of results obtained for 19 cylinders was 2%. This reliability and accuracy was expected for the higher enrichments because of the inherently larger signal relative to background.

Active neutron interrogation, using thermal neutrons to generate fissions in UF_6 and ^4He counters to detect the prompt fission neutrons, is a promising method for measuring low enrichment UF_6 but still needs more development to reach a fully operational status. The response of the system used in the present study varies almost linearly as a function of enrichment up to 2.5% and then effectively saturates at 4%. Analysis of the data showed that the response is sensitive to relative locations of the neutron source, the UF_6 cylinder, and the neutron detectors; consequently these parameters may be used to optimize the technique for specific applications.

These three basic techniques provide a capability for the verification of UF_6 product which is based completely on portable instrumentation and minimal calibration. Measurement accuracies can be improved with further development.

VI. ACKNOWLEDGMENTS

Any attempt to make a complete acknowledgment would necessarily fall short because of the large number of people who contributed to this study; only some will be mentioned here. The study would not have been possible without the hospitality of the GAT and K-25 facilities. In particular, Messrs. V. DeVito, R. Saltsman and Associates at GAT and Messrs. N. Weisner and G. Conner and Ms. J. Brown at K-25 assisted in the logistics of the field operation and supplied the necessary cylinder data. We are grateful to Dr. J. C. Barton for an illuminating discussion on the chemistry of cylinder heels. Thanks are due to the following LASL Group A-1 personnel: Dr. T. L. Atwell for developing a practical procedure for field assay with the active neutron system, Drs. R. A. Forster and D. B. Smith for Monte Carlo calculations, Mrs. C. E. Burns for assisting with data reduction, and Messrs. L. G. Speir, T. R. Capelli and J. W. Woolsey for design of equipment. We are especially grateful to Dr. J. T. Caldwell, LASL Group A-2, for timely communication of his active interrogation method and many subsequent discussions. Finally, Dr. T. E. Sampson contributed significantly to the interpretation of gamma-enrichment results through discussions of the Ge(Li) spectra obtained by LASL Group A-2.

REFERENCES

1. T. D. Reilly, R. B. Walton, and J. L. Parker, Los Alamos Scientific Laboratory report LA-4605-MS, pp. 19-21 (1970).

2. T. D. Reilly and R. B. Walton, Los Alamos Scientific Laboratory report LA-4705-MS, p. 16 (1971).

3. T. D. Reilly, J. L. Parker, A. E. Evans, and R. B. Walton, Los Alamos Scientific Laboratory report LA-4794-MS, pp. 16-19 (1971).

4. A. R. Flynn, Union Carbide report K-1819 (1972).

5. R. B. Walton, Los Alamos Scientific Laboratory report LA-5091-PR, pp. 20-22 (1972).

6. J. L. Feuerbacher and R. L. Newvahner, Goodyear Atomic Corp., private communication.

7. J. M. Bieri, J. T. Caldwell, W. H. Chambers, J. L. Evans, P. E. Fehlau, W. Kunz, N. Nicholson, T. E. Sampson, and G. M. Worth, LASL Group A-2, private communication.

8. J. T. Caldwell, INMM Newsletter, Vol. II, No. 2, to be published (1973).

9. H. O. Menlove and R. A. Forster, Los Alamos Scientific Laboratory reports LA-4794-MS, pp. 10-13 (1971) and LA-4994-PR, p. 6 (1972).

10. E. D. Cashwell, et al., Los Alamos Scientific Laboratory report LA-4751 (1972).

TABLE I

α-DECAY DATA AND UF_6 NEUTRON YIELDS FOR U ISOTOPES

Isotope	$T_{\frac{1}{2}}$ (yr)	α Activity (dps/g)	f_α [a]	Neutrons/g-sec
^{234}U	2.48×10^5	2.27×10^8	1.00	4.6×10^2
^{235}U	7.13×10^8	7.90×10^4	0.56	8.8×10^{-2}
^{236}U	2.39×10^7	2.35×10^6	0.75	3.6×10^0
^{238}U	4.51×10^9	1.23×10^4	0.38	9.5×10^{-3}

[a] f_α is the F(α, n) yield correction factor (relative to ^{234}U) which accounts for effect of different alpha particle energies.

TABLE II

UF_6 NEUTRON YIELDS OF NORMAL URANIUM

Isotope	Neutrons per sec-g U	Relative Yield (%)
^{234}U	2.6×10^{-2}	51.0
^{235}U	6.3×10^{-4}	1.2
^{238}U	9.4×10^{-3}	18.5
^{238}U s.f. [a]	1.5×10^{-2}	29.3
Total:	5.1×10^{-2}	

[a] spontaneous fission.

TABLE III

PASSIVE NEUTRON ASSAY OF DEPLETED UF_6 IN $2\frac{1}{2}$-TON CYLINDERS

Cylinder Number	Tag Isotopic (% ^{235}U)	Net Weight (lb)	Measured Isotopic (% ^{235}U)	$\frac{I_M - I_{Tag}}{I_{Tag}} \times 100\%$
D76825	0.653	4704	0.54	-17.
D31597	.423	4439	.40	- 6.7
D86071	.388	3201	.34	-12.
D78575	.385	2885	.48	+25.
D31721	.208	2051	.18	-11.
D26333	.201	4703	.20	+ 0.5
D26882	.200	3190	.23	+14.
D27098	.200	4696	.20	0.0
D24054	.200	4587	.18	-10.
D44910	.199	4888	.20	0.0

TABLE IV

OUTLIERS IN THE GAMMA ASSAY OF

107 2½-TON CYLINDERS

Cylinder No.	I_{Tag} (% ^{235}U)	$\frac{I_M - I_{Tag}}{I_{Tag}} \times 100\%$
GEW 063	1.332	- 17
GEW 250	2.940	16
GEW 076	2.164	17
GEW 060	2.926	18
RB 2393	2.515	18
GEW 116	2.885	22
GEW 1024	2.940	26
GEW 144	2.885	32
D86172	1.353	36
GEW 282	2.940	37
GEW 259	2.940	63
A 1480	1.353	81
GEW 122	2.93 (t)[a]	87
RB 2370	1.937	93
GEW 096	2.926	109
RB 2394	1.35	113
GEW 252	1.330	130

[a] t denotes target enrichment.

TABLE V

GAMMA ASSAY OF UF_6 IN 5A MONEL CYLINDERS

Cylinder Number	Tag Isotopic (% 235U)	Measured Isotopic (% 235U)	$\frac{I_M - I_{Tag}}{I_{Tag}} \times 100\%$
1	3.132	3.30	+ 5.4
2	3.329	3.49	+ 4.8
3[a]	3.421	3.79	+ 11.
4	3.715	3.70	- 0.4
5	7.609	7.62	+ 0.1
6	18.63	19.2	+ 3.1
7	19.20	18.8	- 2.1
8	23.12	23.4	+ 1.1
9	27.06	27.6	+ 2.2
10[a]	33.50	33.5	+ 4.5
11[a]	39.56	40.4	+ 2.0
12	58.30	Standard	
13	59.06	60.2	+ 1.9
14	59.09	57.9	- 2.1
15	79.61	77.6	- 2.5
16	93.21	93.6	+ 0.4
17	93.28	93.6	+ 0.3
18	95.21	92.6	- 2.7
19	96.70	95.0	- 1.8
20	97.00	97.5	+ 0.5
21	97.63	96.1	- 1.6
22	97.65	95.7	- 2.0
23	97.65	96.0	- 1.7
24	97.65	97.8	+ 0.2
		Mean =	0.9%
		σ =	3.2%

[a] Steel cylinders

TABLE VI

VALUES OF λ AND f AS A FUNCTION OF ^{235}U ENRICHMENT

I_{235} (%)	λ (cm)	f (%)
0	43.4	0
0.2	28.8	34
0.5	19.1	56
1.0	12.2	72
2.0	7.1	84
3.0	5.0	88
4.0	3.9	91

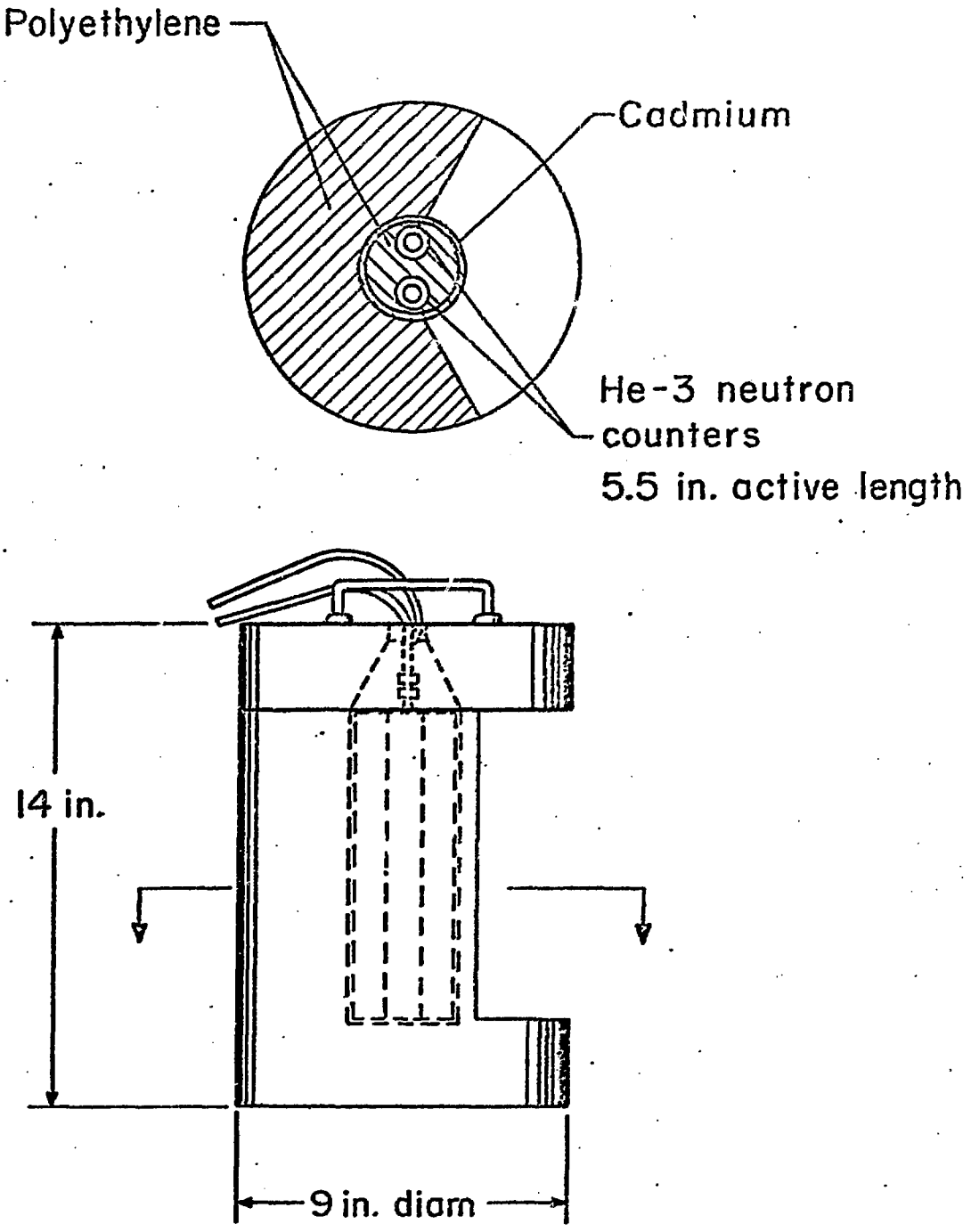

Fig. 1. Schematic of SNAP, the portable shielded neutron assay probe used for measurements of neutrons from UF_6.

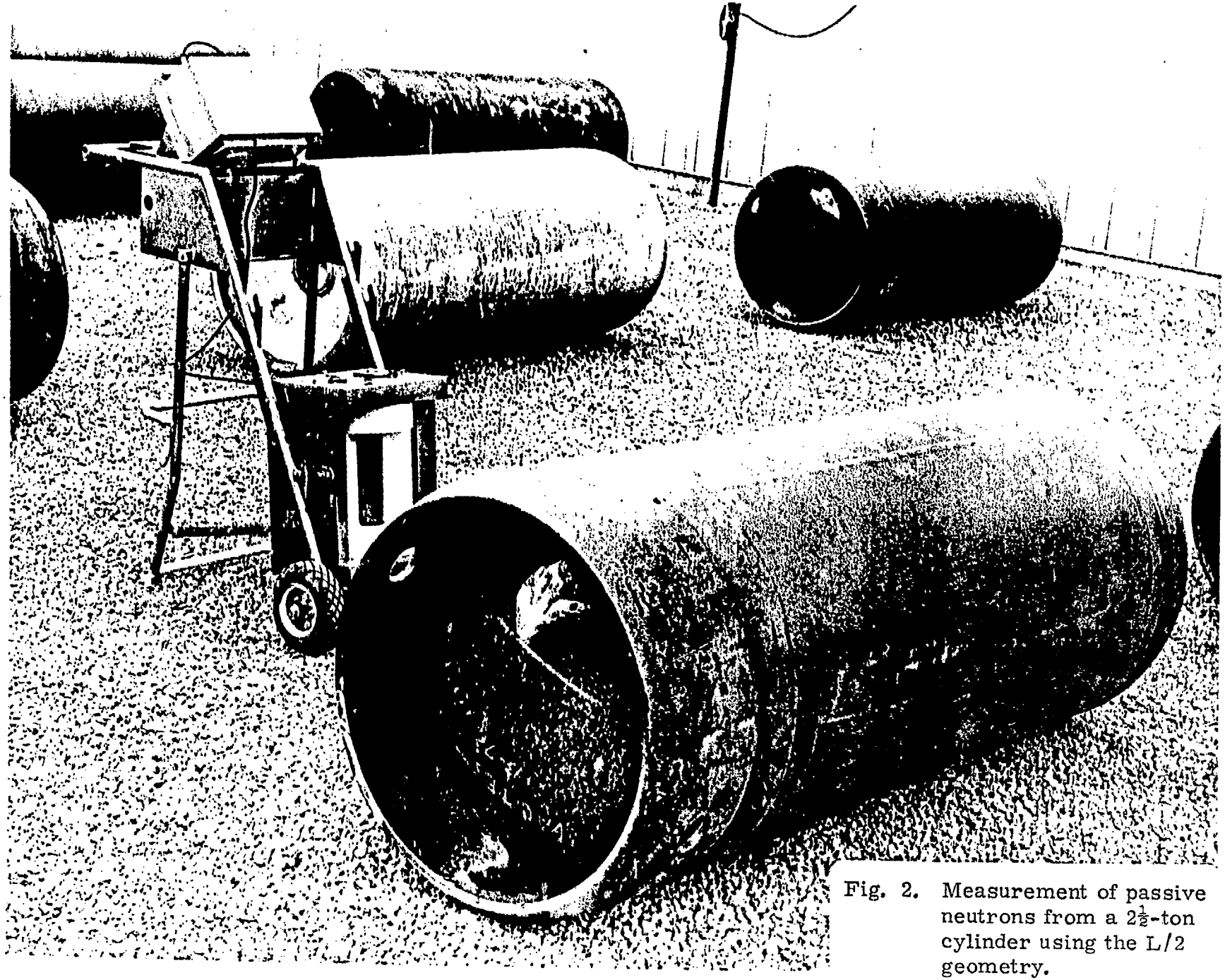

Fig. 2. Measurement of passive neutrons from a $2\frac{1}{2}$-ton cylinder using the L/2 geometry.

Fig. 3. Contact measurement of passive neutrons from a $2\frac{1}{2}$-ton cylinder.

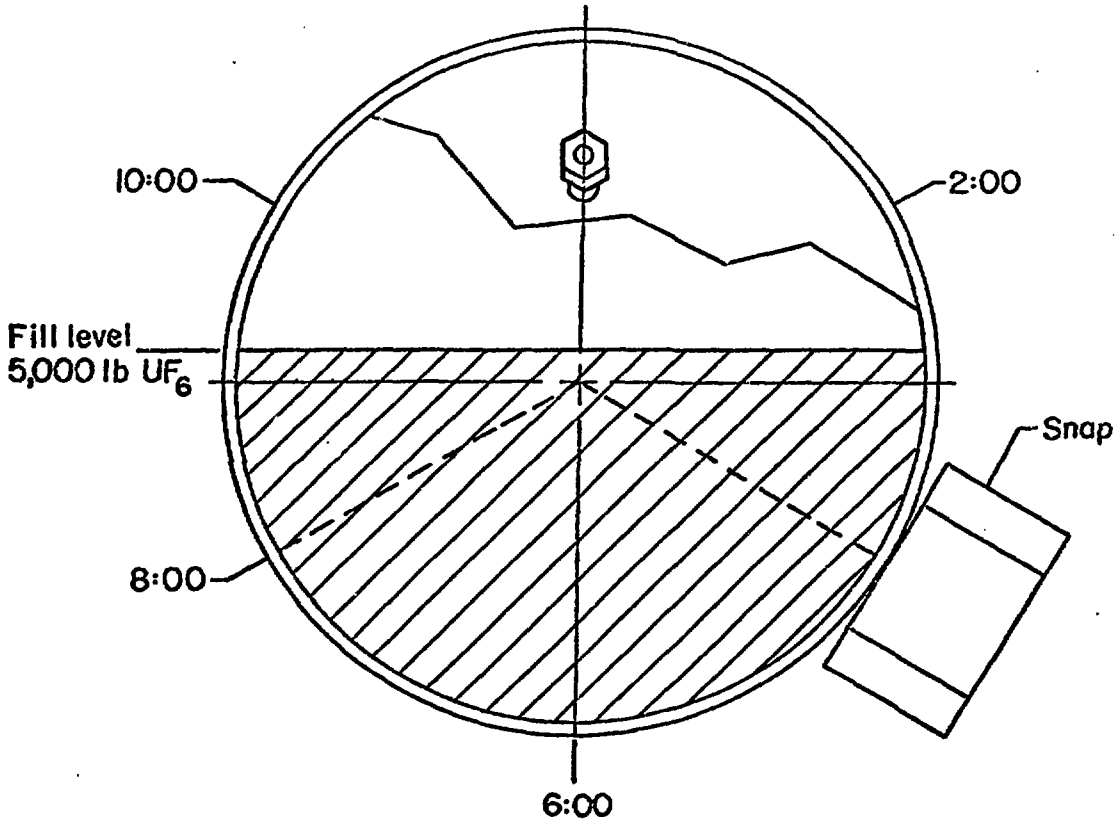

Fig. 4. Schematic of contact measurement, showing 5000-lb UF_6 fill level. The assay is derived from the average of measurements at the 4 and 8 o'clock positions.

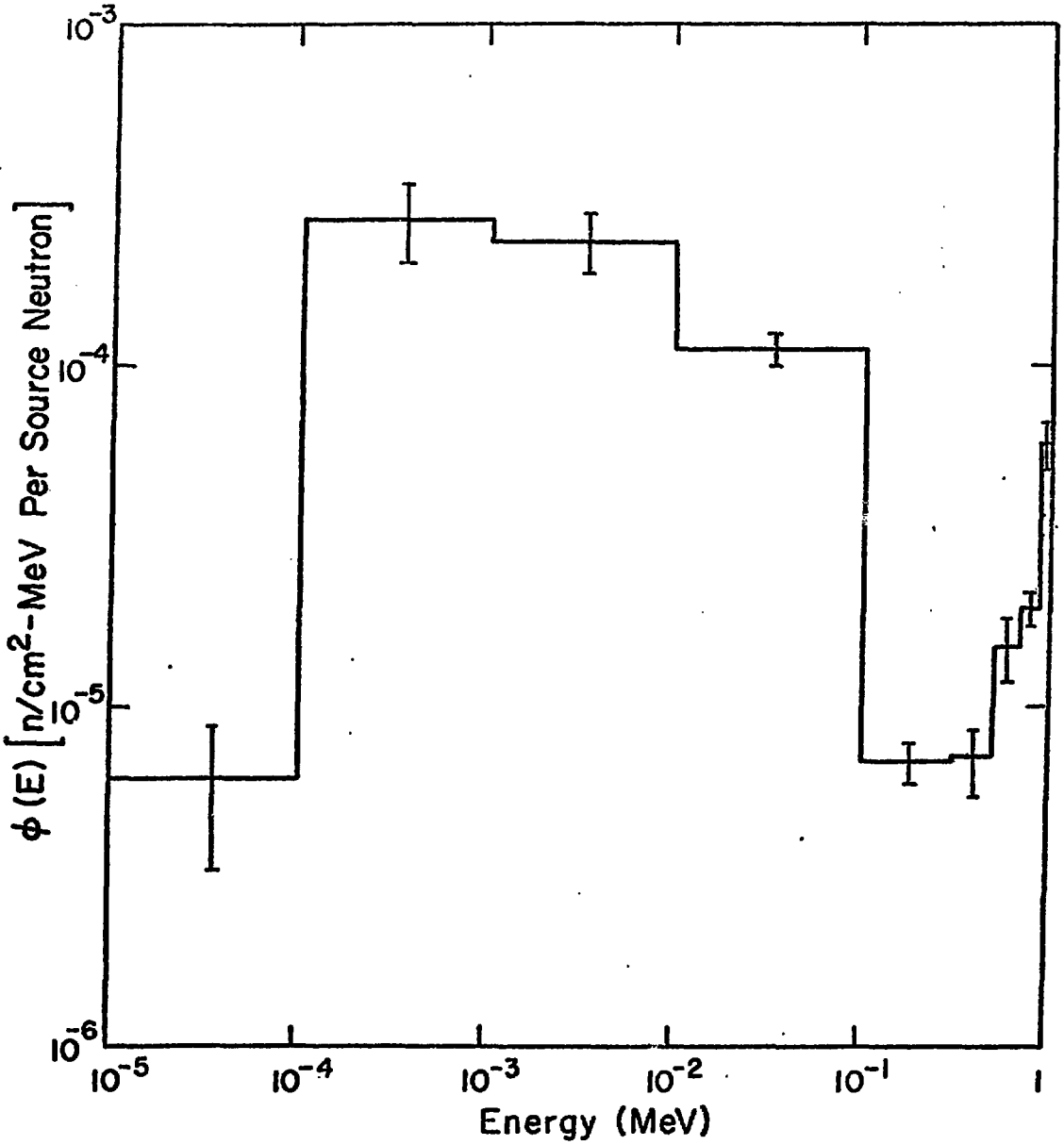

Fig. 5. Neutron leakage spectrum from a 5000 UF_6 ($2\frac{1}{2}\%$ ^{235}U) cylinder calculated assuming a neutron spectrum with single energy neutrons at 1 MeV.

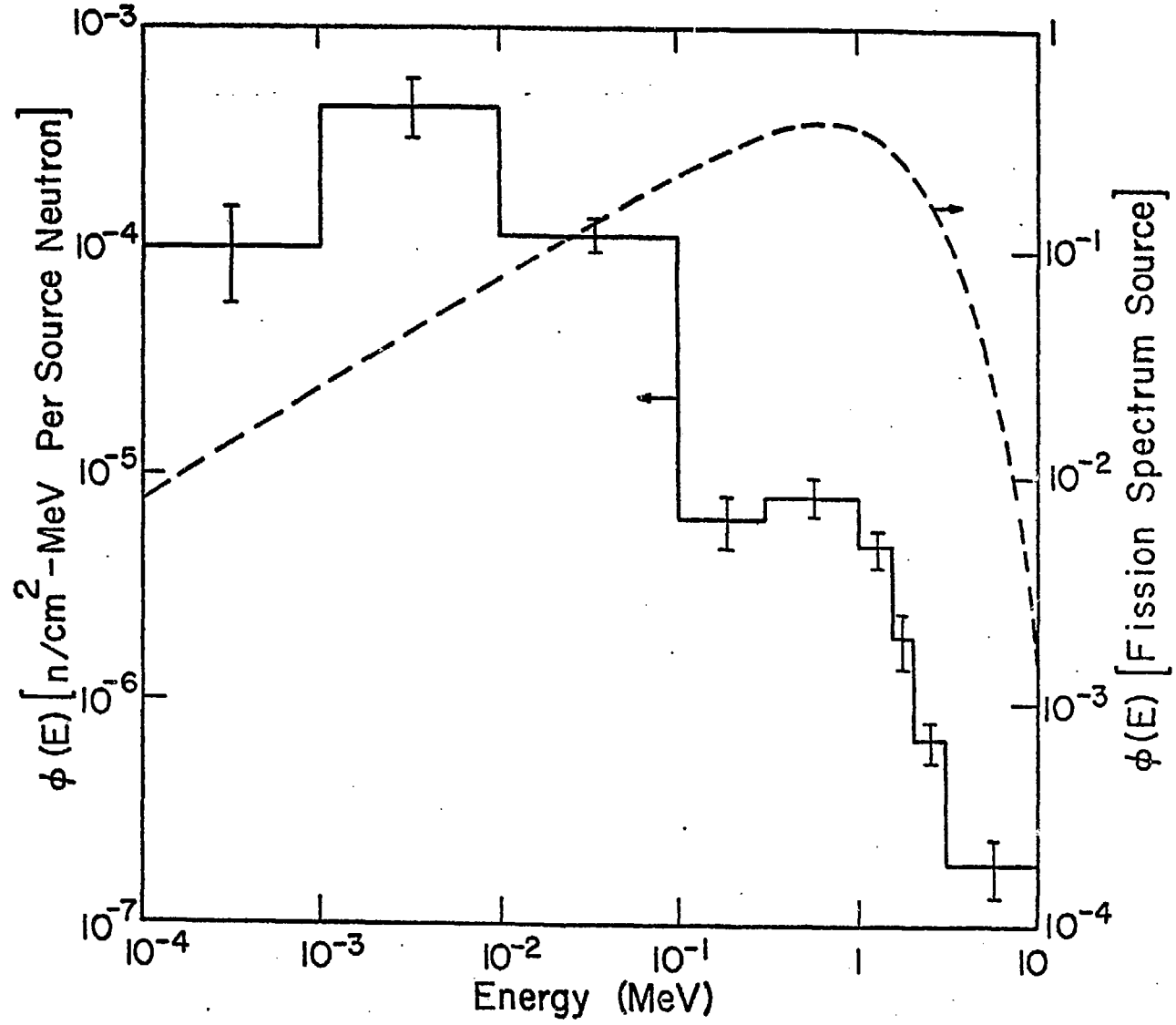

Fig. 6. Neutron leakage from a 5000 lb UF_6 ($2\frac{1}{2}\%$ ^{235}U) cylinder calculated assuming a fission source spectrum. Both spectra are plotted.

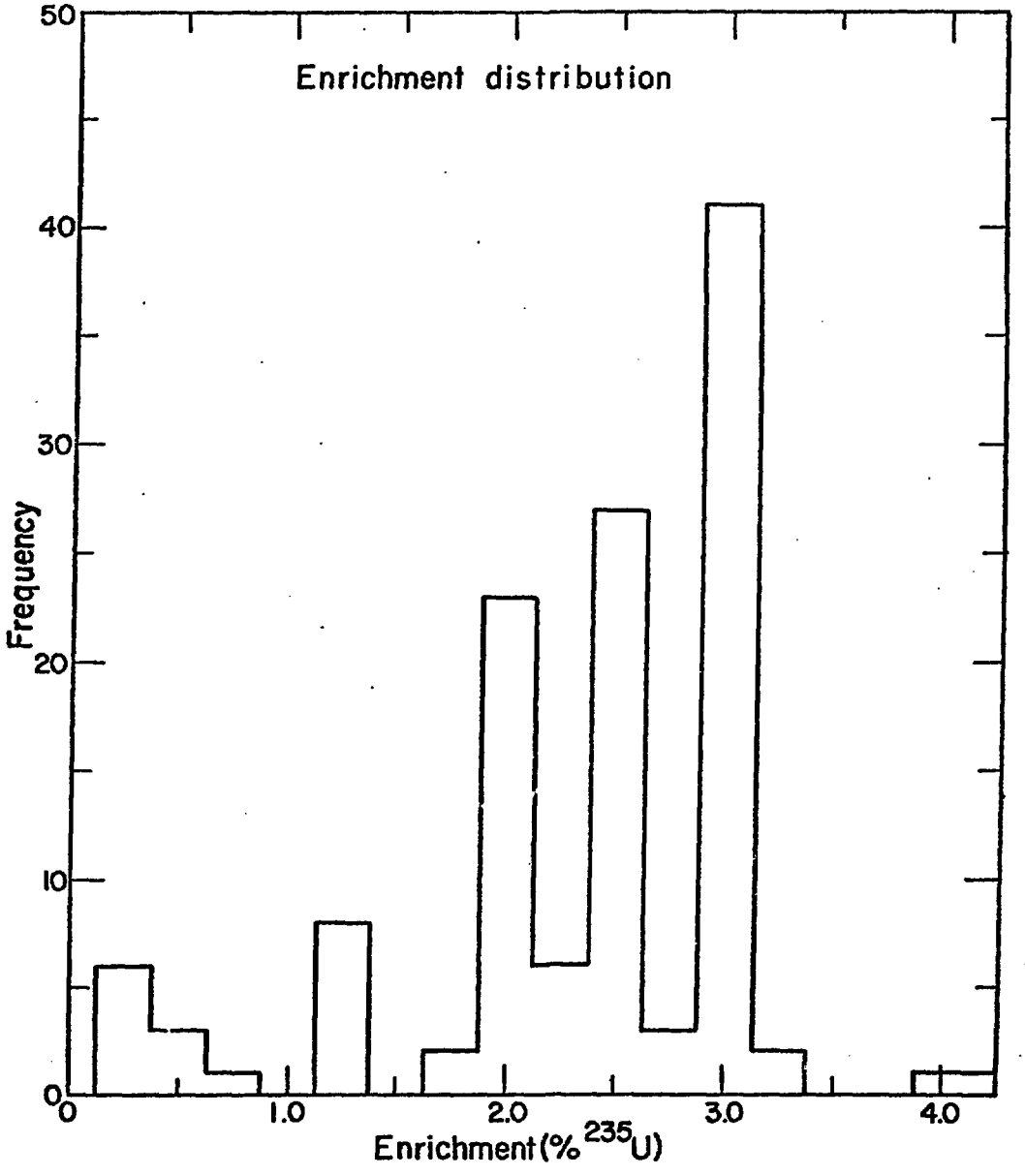

Fig. 7. Enrichment distribution of 123 type-30 cylinders which were measured by the passive neutron assay technique. Number of cylinders versus enrichment are shown.

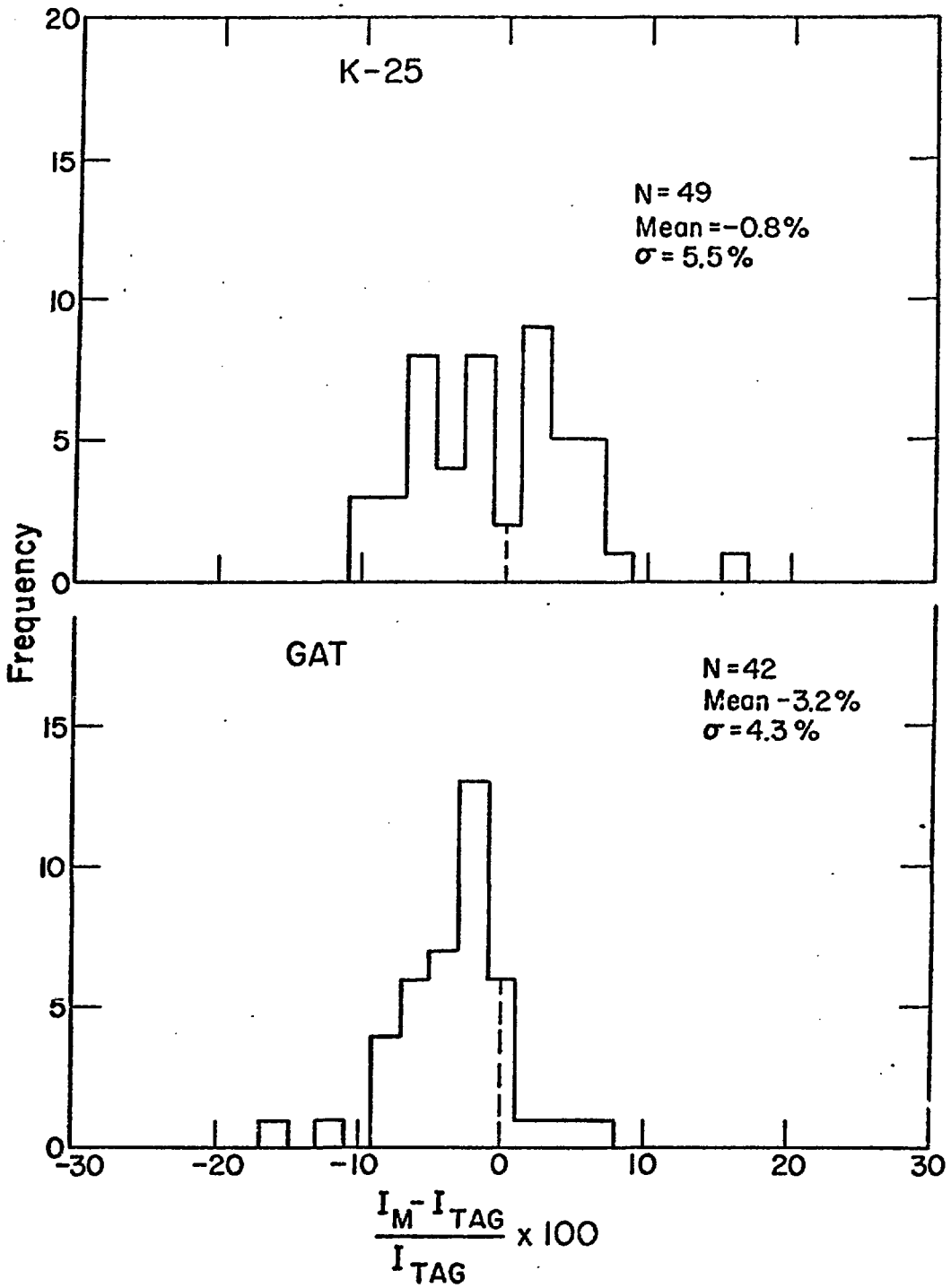

Fig. 8. Passive neutron assay results for 91 licensee, $2\frac{1}{2}$-ton cylinders (1.3 - 3.1% enrichment) at GAT and K-25.

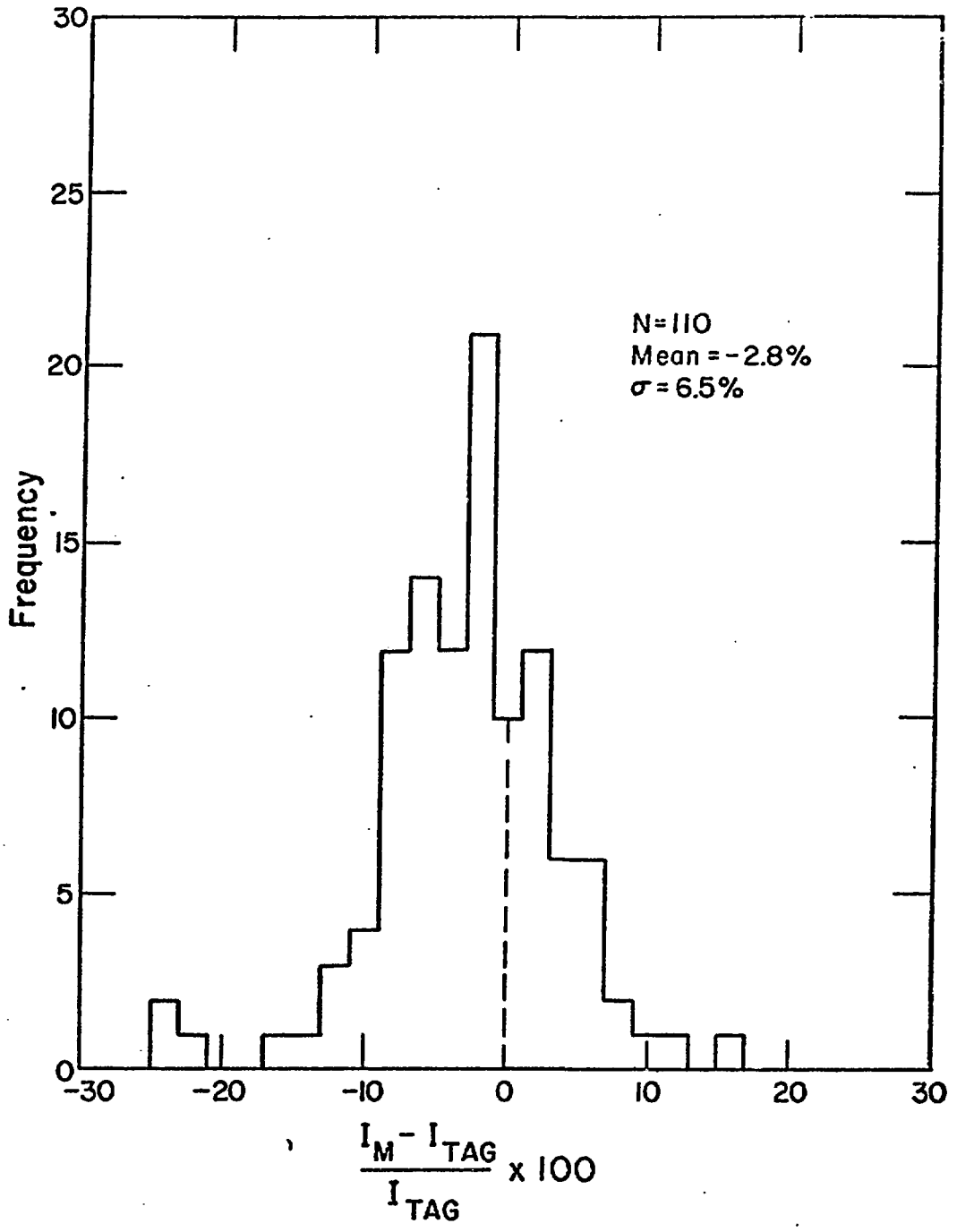

Fig. 9. Passive neutron results for all type-30 cylinders with enrichment greater than 1%. This group includes 91 licensee cylinders (cf. Fig. 8) and 19 storage cylinders.

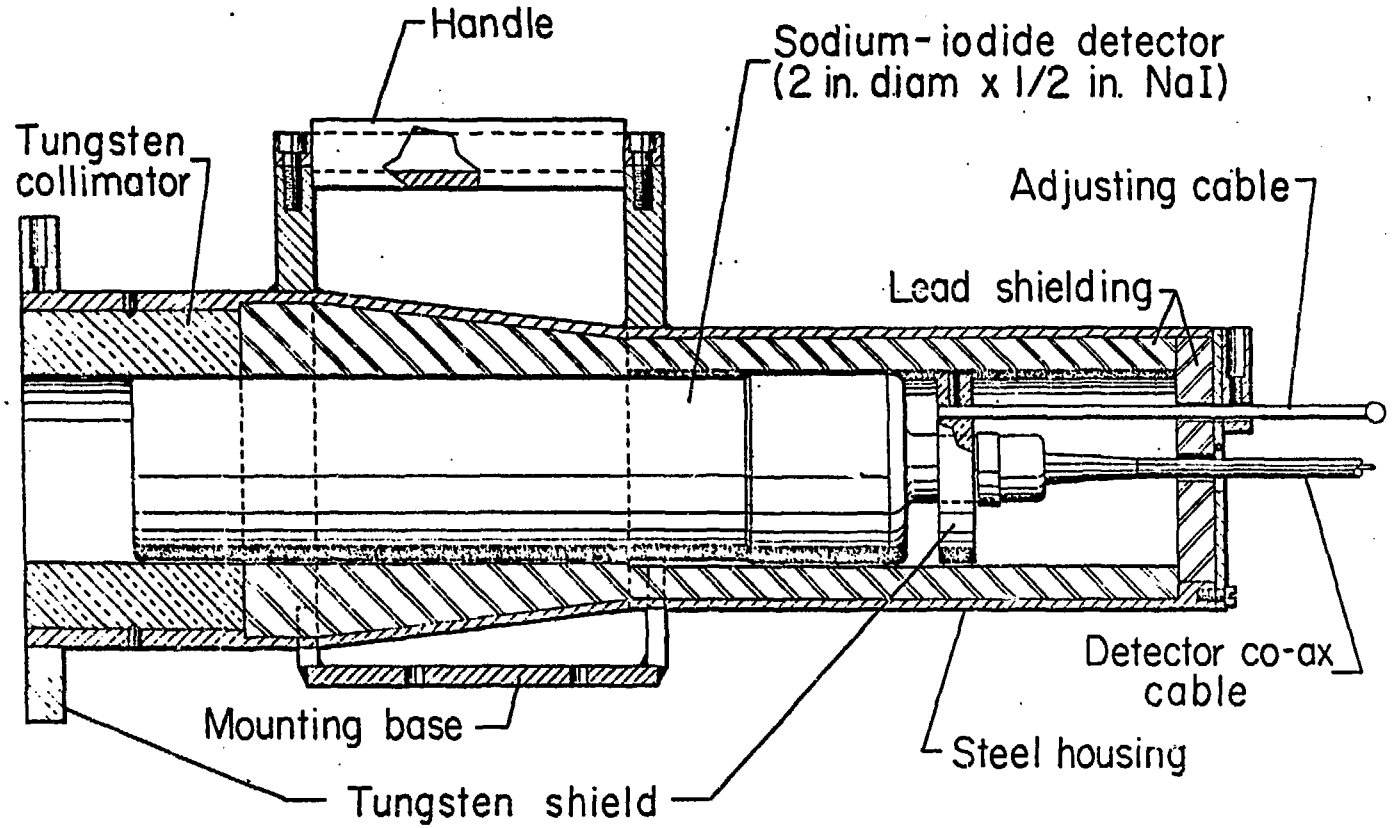

Fig. 10. Details of NaI probe and collimator assembly used for gamma enrichment measurements.

Fig. 11. Gamma-ray enrichment measurement with Eberline SAM II gamma unit and Branson ultrasonic thickness gage.

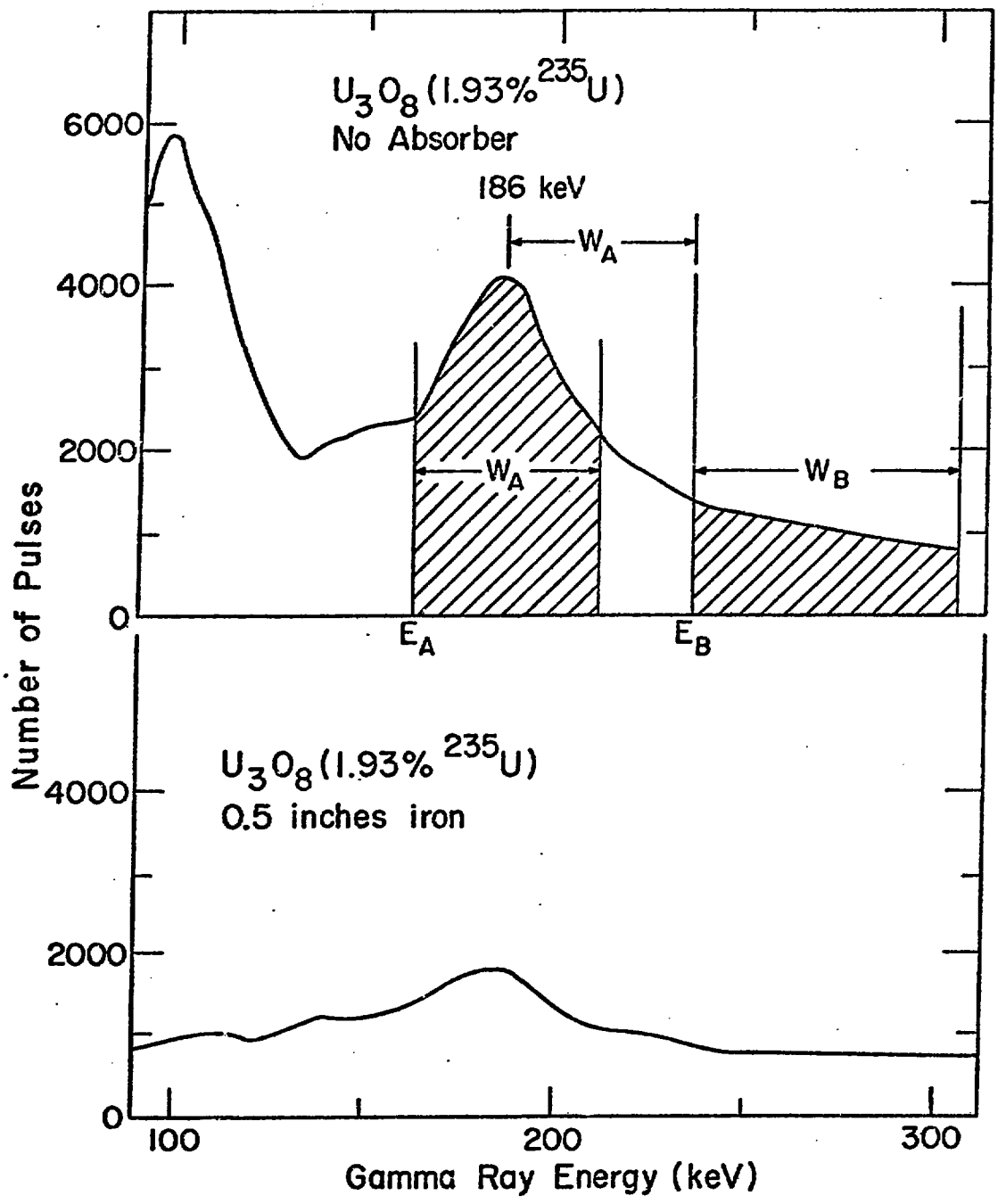

Fig. 12. Pulse height distributions of gamma rays from 1.93% U_3O_8 with no absorber (upper spectrum) and with 0.5 in. of iron (lower spectrum). Positions and widths of the photopeak and background windows for enrichment analysis are also shown.

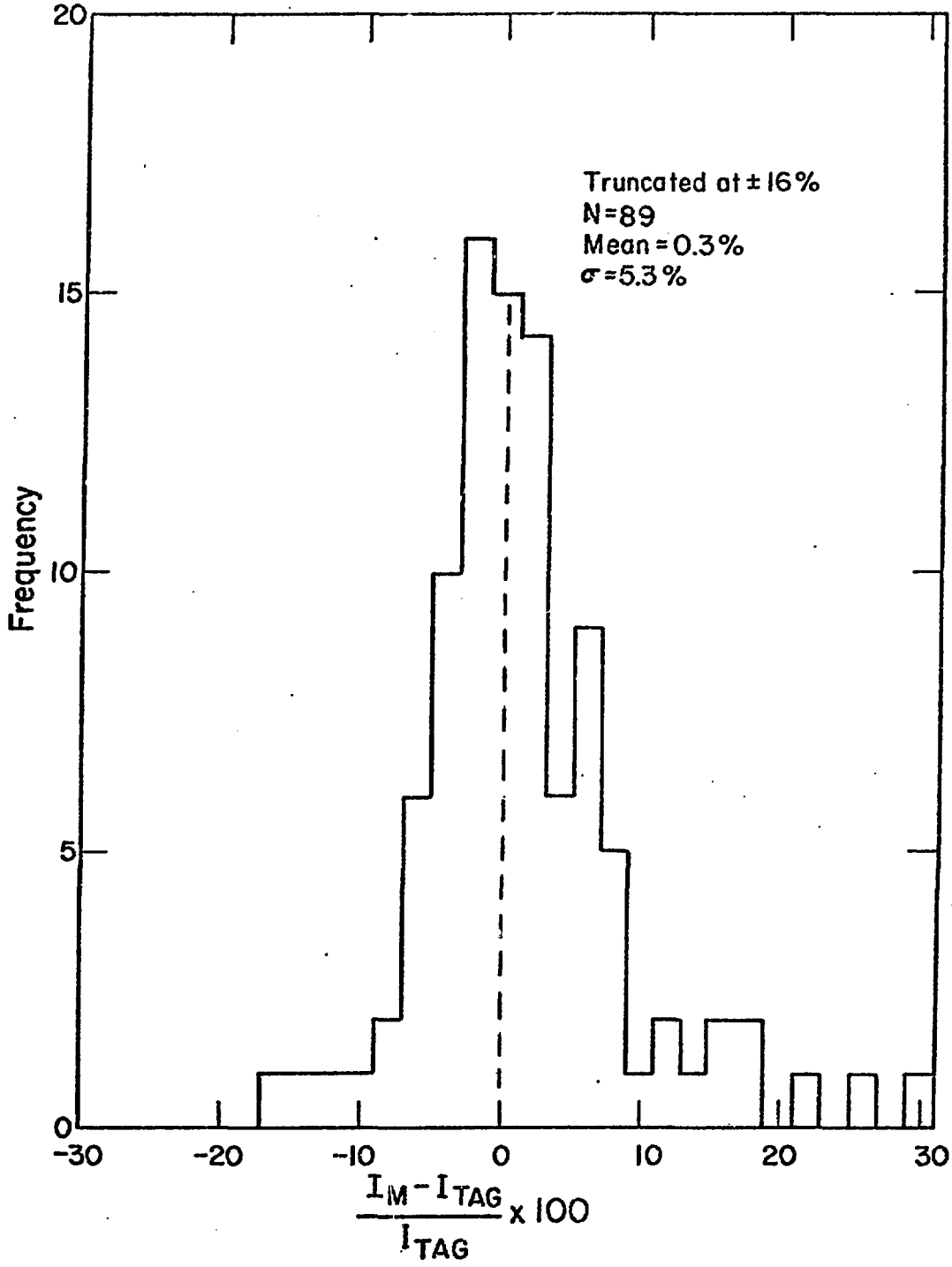

Fig. 13. Gamma enrichment assay results for 2½-ton cylinders (1.3 - 3.1% enrichment). The distribution of relative errors was truncated at 16%, omitting obvious outliers. Of a total of 107 cylinders measured, there were 17 outliers (relative error greater than 16%), and these are listed in Table IV.

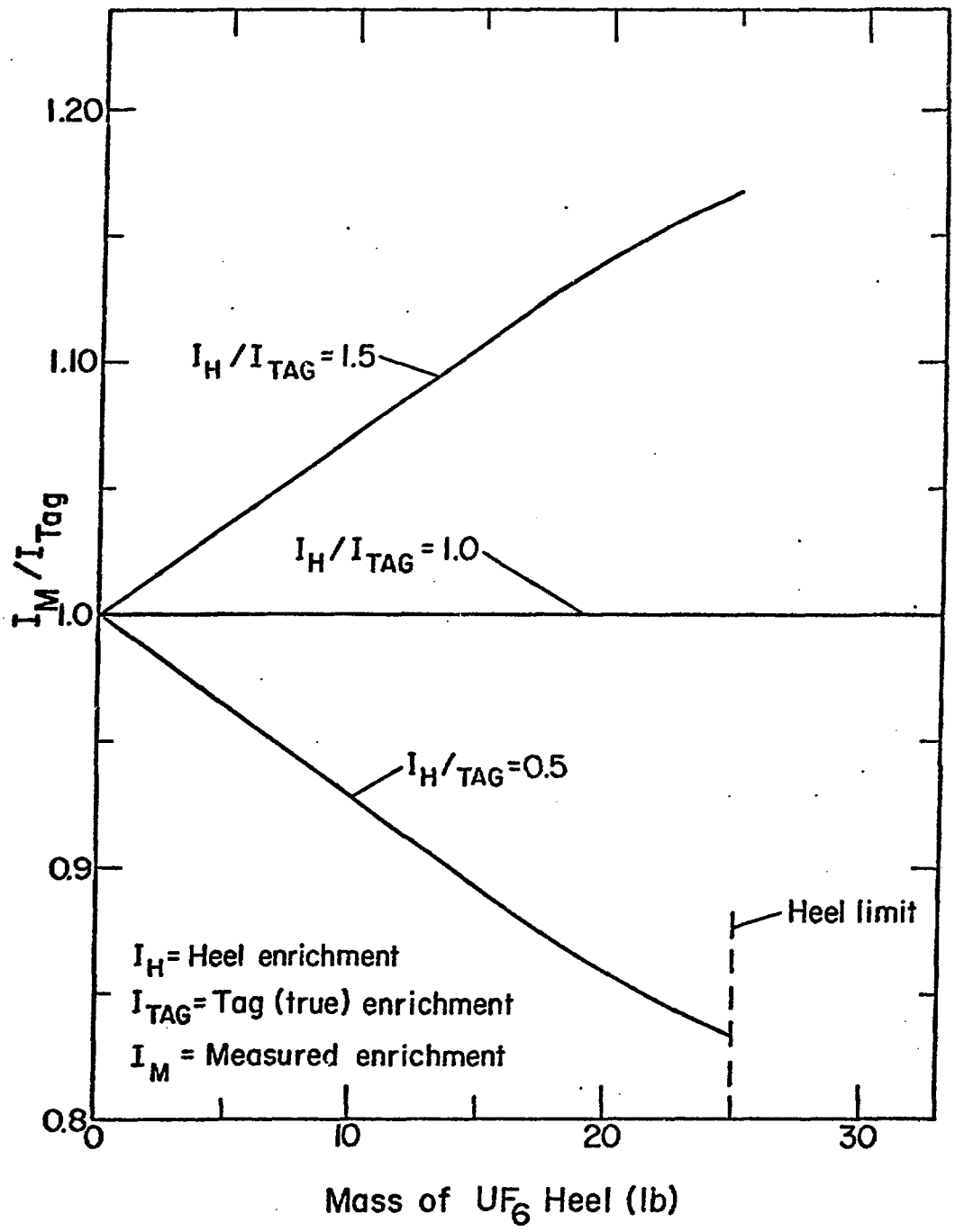

Fig. 14. Calculated effect of UF_6 heel on gamma enrichment measurement versus heel weight.

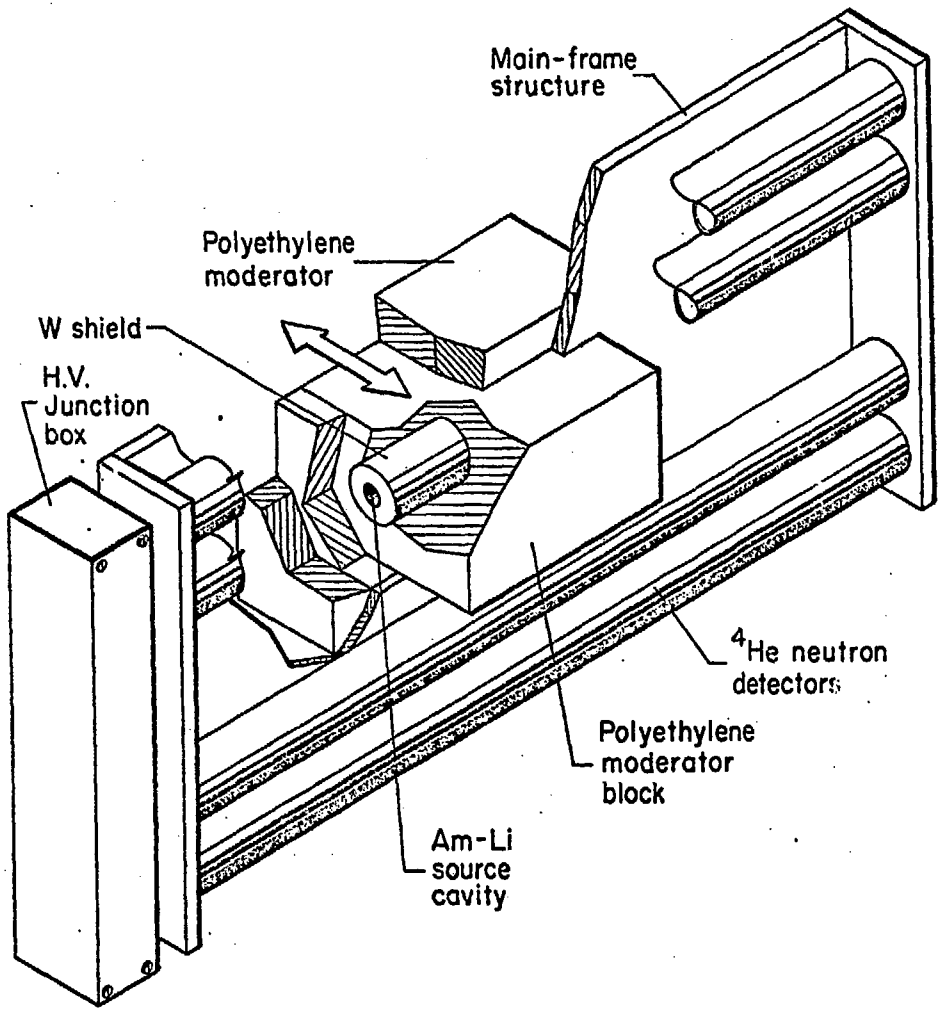

Fig. 15. Modified Caldwell assay unit. Thermal neutrons from the source-moderator assembly produce fissions in the UF_6 and the prompt fission neutrons are detected by the ^{4}He counters.

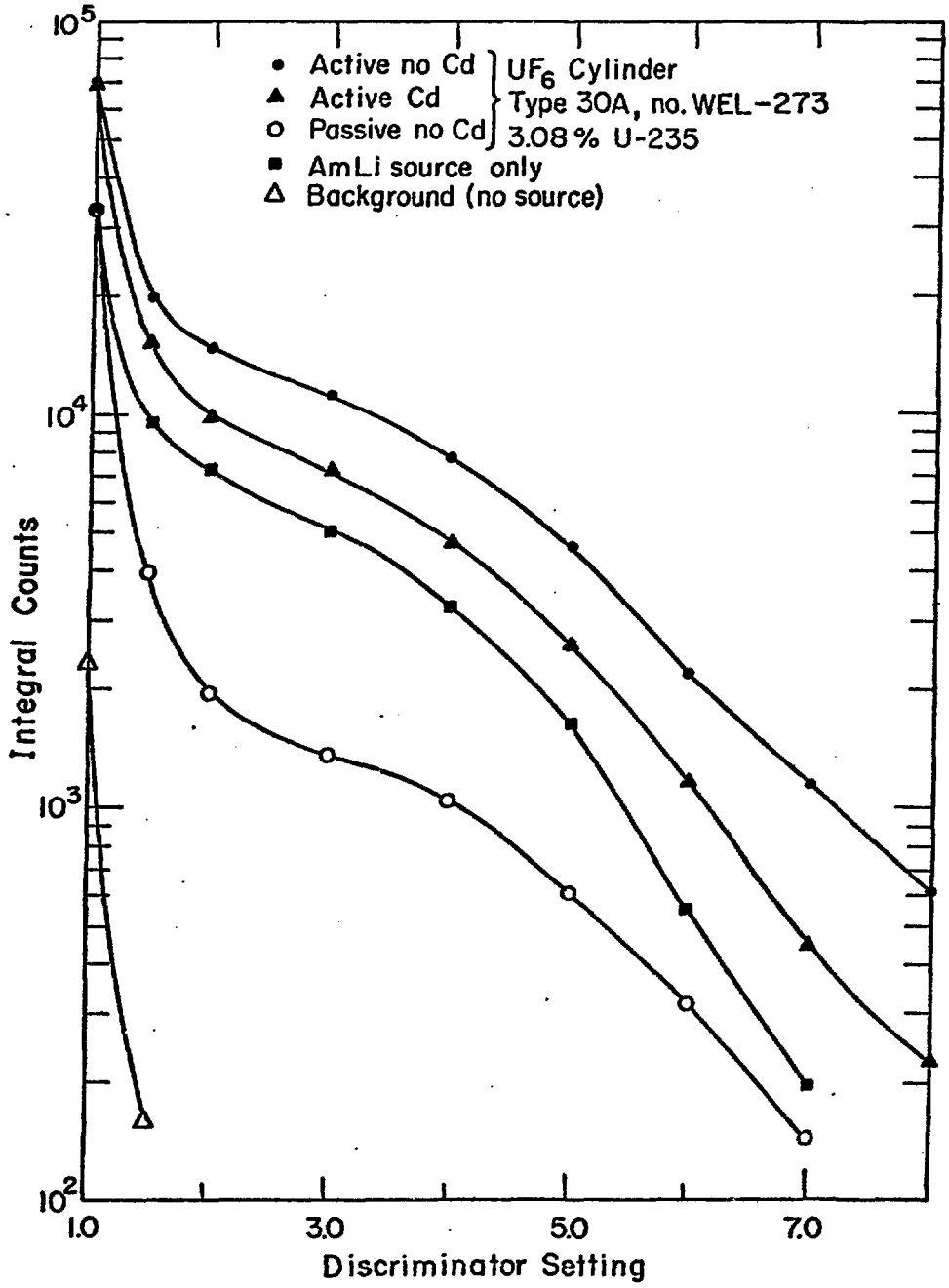

Fig. 16. Signal and background rates of the active neutron interrogation unit versus pulse discriminator setting. The net signal used for assay is the "active no Cd" minus "active Cd".

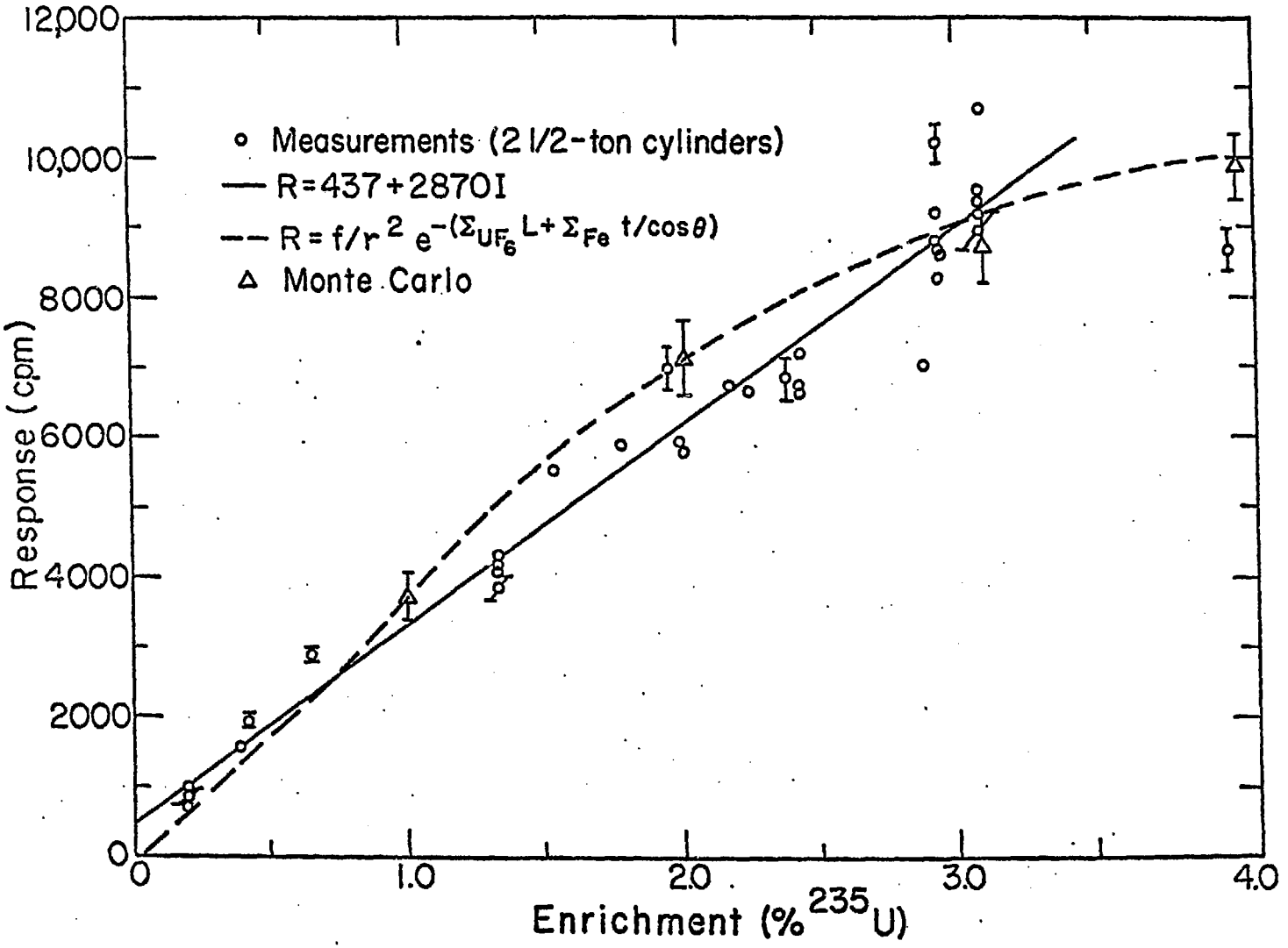

Fig. 17. Response of the active neutron assay system versus enrichment of $2\frac{1}{2}$-ton cylinders of UF_6. Results of Monte Carlo calculations and of a simple analytical model (cf. Fig. 18), together with a straight line fit, are also shown.

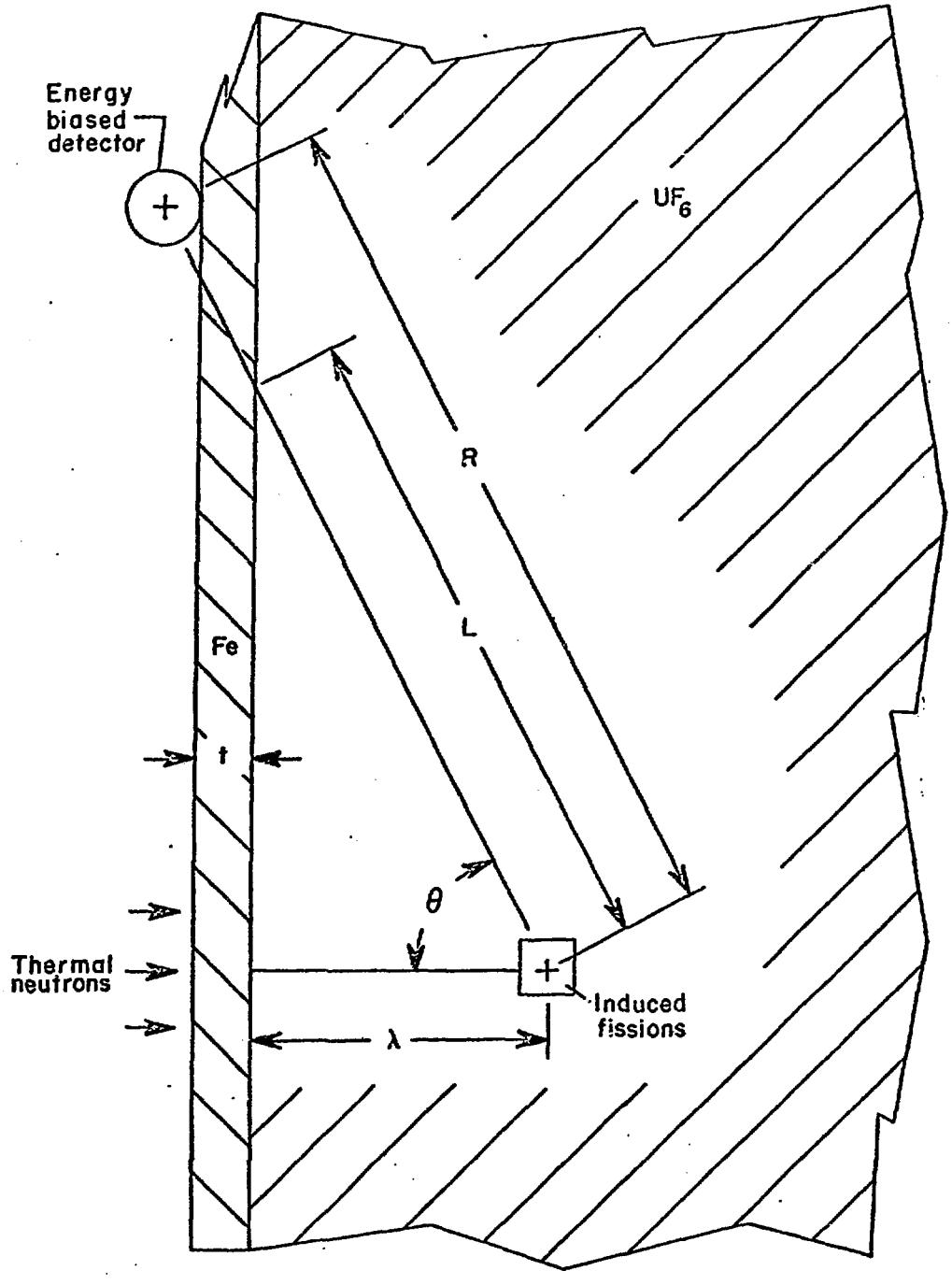

Fig. 18. Schematic representation of the active neutron assay method.