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Los Alamos Los Alamos National Laboratory
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# DIRECT FISSILE ASSAY OF HIGHLY ENRICHED $UF_6$ USING RANDOM SELF-INTERROGATION AND NEUTRON COINCIDENCE RESPONSE[a]

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## ABSTRACT

A new nondestructive method for direct assay of $^{235}U$ mass contained in Model 5A uranium hexafluoride ($UF_6$) product storage cylinders has been successfully tested in the laboratory and under field conditions. The technique employs passive neutron self-interrogation and uses the ratio of coincidences-to-totals counts as a measure of bulk fissile mass. The accuracy of the method is 6.8% ($1\sigma$) based on field measurements of 44 Model 5A cylinders, 11 of which were either only partially filled or contained reactor return material. The cylinders contained $UF_6$ with enrichments from 5.96% to 97.6%. Count times were 3-6 min depending on $^{235}U$ mass. Samples ranged from below 1 kg to over 16 kg of $^{235}U$. Because the method relies primarily on fast neutron self-interrogation, complete sampling of the $UF_6$ takes place. This feature alleviates inhomogeneity problems and offers increased assurance of the presence of stated amounts of bulk fissile material as compared with current verification methods.

## INTRODUCTION

For safeguards and accountability of highly enriched uranium (HEU), determination of $^{235}U$ mass in uranium hexafluoride ($UF_6$) is required. For HEU (enrichments $\geq 20\%$ $^{235}U$), the $UF_6$ is stored in Model 5A cylinders that are nominally 128 mm in diameter and ~914 mm tall (see Fig. 1). Typical $UF_6$ fill heights are 300-400 mm. These containers, holding material of high economic and strategic value, are used for the output of enrichment plants and for the input to fuel fabrication facilities.

Fig. 1. $UF_6$ cylinder Model 5A: photographs and general data.

The conventional nondestructive method[1] for measuring $^{235}U$ enrichment of $UF_6$ in 5A cylinders employs the enrichment principle. The 186-keV gamma rays from $^{235}U$ decays are counted using NaI or germanium detectors. An ultrasonic measurement of the cylinder wall thickness (6.4-mm Monel or steel) is made to correct for variations in gamma-ray attenuation in the walls. The measured $^{235}U$ enrichment, the net $UF_6$ weight, and the purity (uranium weight fraction) must be combined to yield $^{235}U$ mass. This type of measurement has the disadvantage that only the enrichment of the surface layer (<11 mm) of the $UF_6$ is sampled. Hence, the bulk of the material in the interior of the cylinder is not verified. Also, inaccuracies can result if

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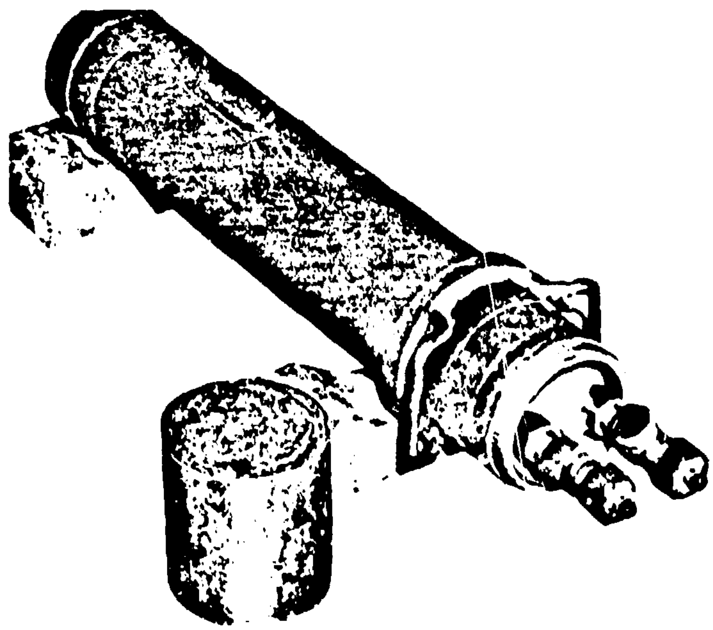


## GENERAL DATA

Other Descriptive Terminology Used · 5-in. product

| ENGINEERING DRAWING REFERENCE | GOODYEAR ATOMIC CORPORATION, DRAWING CX-761-M2011 |
|---|---|
| Nominal Diameter | 5 in. |
| Nominal Length | 36 in. |
| Wall Thickness | 1/4 in. |
| Nominal Tare Weight | 55 lb (24.95 kg) |
| Maximum Net Weight | 55 lb (24.95 kg) |
| Nominal Gross Weight | 110 lb (without cap) (49.9 kg) |
| Minimum Volume | 0.284 ft$^3$ (8.04 liters) |
| Basic Material of Construction | Monel |
| Service Pressure | 200 psig |
| Hydrostatic Test Pressure | 400 psig |
| Isotopic Content Limit | 100% U-235 max |

Valve Used · 3/4-inch Valve

the $^{235}U$ enrichment is not homogeneous. If the uranium has been irradiated in a reactor, it may contain levels of technetium that reduce the signal-to-background ratio of the gamma-ray enrichment measurement and therefore limit its precision.

Gas-phase sampling of the cylinder contents followed by mass spectrometric determination of $^{235}U$ enrichment can also be biased by inhomogeneities such as layering.

This paper describes a new passive neutron assay technique that directly samples the entire $UF_6$ volume of Model 5A product storage cylinders to determine $^{235}U$ mass. This passive technique, based on self-interrogation and coincidence counting, was identified after testing and evaluating both active and passive applications of the neutron Coincidence Collar.[2] Subsequently, the method was tested using the Active Well Coincidence Counter (AWCC)[3] in the passive mode, the Dual Range Coincidence Counter (DRCC),[4] and the High-Level Neutron Coincidence Counter (HLNCC).[5] The first field test of the method was conducted at the Goodyear Atomic Corporation (GAT) Gaseous Diffusion Plant (GDP), near Piketon, Ohio, using a high-efficiency AWCC in the passive mode.

The new method owes its simplicity to the unique neutronic properties of $UF_6$ containing HEU and the ability of the shift-register coincidence circuitry to isolate time-correlated events from those occurring randomly in time. Variations in cylinder wall thickness or the presence of reactor-return material do not significantly affect results. Variations in fill height and $UF_6$ density do affect the measurement, and a correction was developed that minimizes these effects. The correction is determined by changing the neutron reflectivity (albedo) of the sample cavity with a removable cadmium liner.

## Measurement Principles

Intrinsic neutron production in $UF_6$ containing HEU is dominated by $^{19}F(\alpha,n)^{22}Na$ reactions;[6] these are primarily due to $^{234}U$ alpha decays. Uranium-238 is the dominant source of spontaneous fission reactions in $UF_6$ containing HEU. Spontaneous fissions, however, account for only 1.1% of the total neutron production at 20% enrichment, 0.25% at 50% enrichment, and only 0.01% at 98% enrichment for typical $UF_6$ from a diffusion cascade. Figure 2 is a plot of specific neutron production in $UF_6$ vs $^{235}U$ enrichment for a diffusion cascade. For solid $UF_6$, a thick-target model of neutron production is applicable as opposed to the thin-target model[7] necessary for low-density $UF_6$ gas.

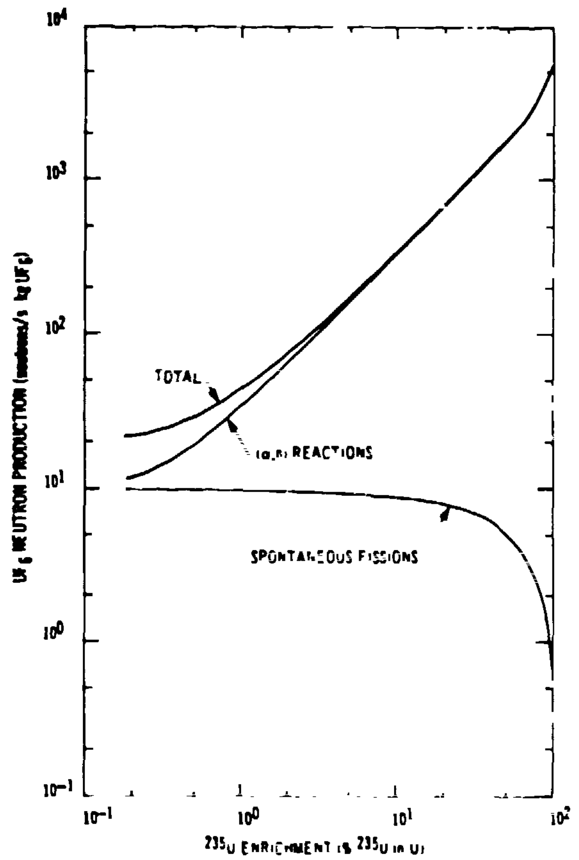



Fig. 2. Neutron production in $UF_6$ vs enrichment. Contributions from $(\alpha,n)$ reactions and spontaneous fissions are shown separately.

Passive $(\alpha,n)$ neutrons born in $UF_6$ may induce fissions before being captured or escaping the cylinder. These fissions occur predominantly in $^{235}U$ but may also occur in $^{238}U$ for neutron energies above ~1 MeV. Figure 3 shows plots of calculations of $^{234}UF_6$ neutron emission spectra, assuming two models for $^{22}Na$ level branching. These spectra are 'softer' than induced-fission spectra and are further softened by neutron scattering in the $UF_6$ sample and detector matrix material (polyethylene).

Because the shift-register coincidence electronics logic[8] measures the time-correlated events, the passive $(\alpha,n)$ neutrons do not contribute directly to the coincidence count rate since they occur randomly in time. However, they contribute indirectly to coincidence response by inducing fissions that yield prompt

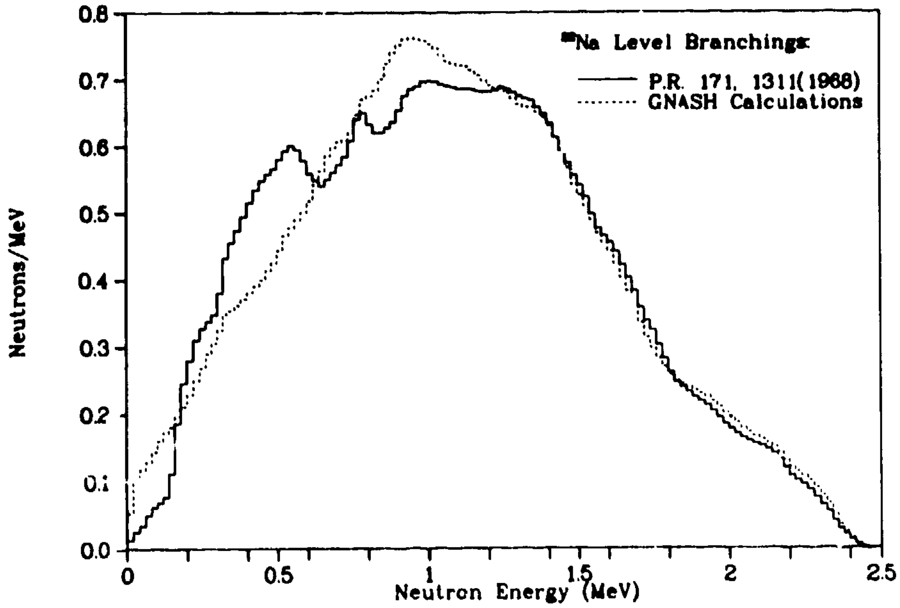



Fig. 3. Calculated neutron production spectra for the $^{234}UF_6$ ($\alpha$,n) reaction.

neutrons in bursts of 0-8. Uranium-238 spontaneous fissions make a small (and correctable) direct contribution to the measured coincidence response.

Several years ago, calculations were reported[9] that gave the neutron leakage per source neutron for bare Model 5A cylinders containing $UF_6$ at five enrichments (20, 50, 80, 93, and 97.5%) and two $UF_6$ masses, 10 and 20 kg. Results of these calculations are plotted as a function of $^{235}U$ mass in Fig. 4. The calculations assumed all passive ($\alpha$,n) neutrons had an initial energy of 1 MeV because thick-target emission spectra as shown in Fig. 3 were not available. This is not expected to significantly alter results of the calculations. For the purposes of this paper, Fig. 4 has three important features: first, induced fissions account for a significant fraction of the neutron leakage from the cylinders; second, this fraction is nearly proportional to fissile mass; and third, the half-full cylinders are more highly multiplying than full ones containing the same $UF_6$ mass. These calculations were made without any material surrounding the cylinders. They therefore give an indication of the magnitude of the induced-fission rate caused by neutrons that have not escaped the cylinder.

The passive ($\alpha$,n) neutrons and their induced-fission progeny produce a leakage neutron current from the $UF_6$ sample into the thermal neutron well counter. These instruments typically contain concentric rings of $^3He$ proportional counters in a polyethylene matrix. A fraction (albedo) of the neutron leakage is reflected back into the $UF_6$ cylinder with a shifted energy spectrum. The spectral softening is due to neutron collisions with hydrogen nuclei bound in polyethylene molecules. Because the returning neutrons have lowered energies, they are less likely to induce fissions in $^{238}U$ and more likely to induce fissions in $^{235}U$. Self-interrogation then refers to fissions induced by ($\alpha$,n) self-source neutrons before being captured or escaping the cylinder (see Fig. 4) in addition to fissions induced by returning neutrons whose average energy is lower than upon escape from the $UF_6$ cylinder.

With a Model 5A cylinder in a thermal neutron well counter and with a cadmium liner for the sample cavity, reflected neutrons with energies below 0.25 eV (thermal neutrons) do not contribute to self-interrogation, whereas with it they do. Thermal, or slow, neutrons do not penetrate deeply into $UF_6$ containing HEU. The diffusion length L is defined as the average root-mean-square distance at which a plane source of thermal neutrons is reduced by a

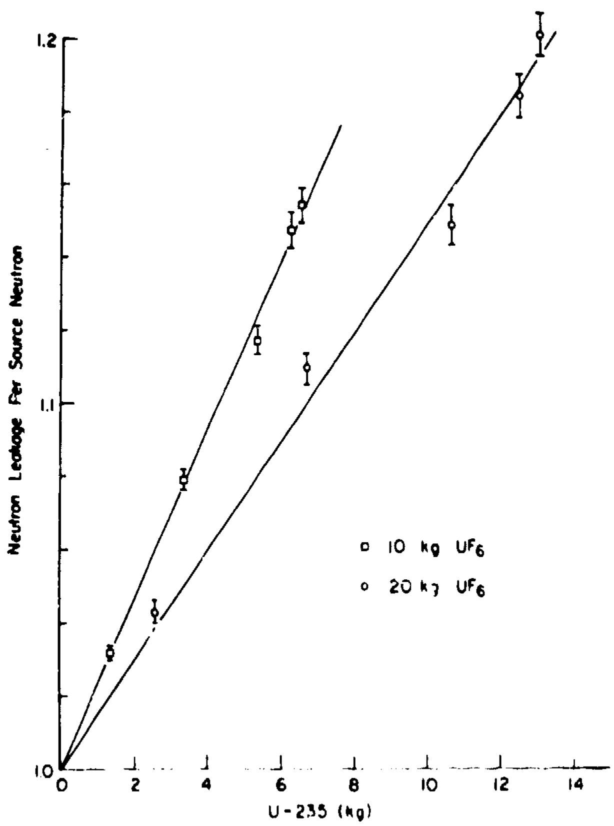



Fig. 4. Calculated neutron leakage per source neutron for $UF_6$ at five enrichments and two fill heights.

factor of e relative to initial intensity. In solid $UF_6$, L is 4.4 mm at 20% enrichment, 1.9 mm at 50% enrichment, and only 1.0 mm at 97% enrichment. Returning thermal neutrons, therefore, produce an induced-fission coincidence response indicative of only the surface layer of $UF_6$ next to the cylinder wall. However, second-generation fission spectrum neutrons produced in the surface layer are energetic enough to penetrate to the interior of the sample.

Normally, two measurements are made: one with the well liner in place and one with it removed. The second measurement is made to derive a correction for the first. The correction is used to remove effects of $UF_6$ fill-height variations.

The neutron coincidence count rate R is a measure of the induced-fission rate and thus the $^{235}U$ mass in the sample. The induced-fission rate is directly proportional to the $(\alpha,n)$ source strength S of the sample. Also, the total neutron count rate T is directly proportional to S. Because S is known to vary with enrichment and irradiation history of the sample material, a signature independent of it is desirable. We chose, therefore, coincidence count rates divided by totals count rates as a passive signature for $^{235}U$ mass. A small correction is made to coincidence rates resulting from spontaneous fissions in $^{238}U$.

Laboratory Measurements

To test a variety of techniques for fissile assay of $UF_6$ in Model 5A cylinders, initial measurements were made at Los Alamos National Laboratory using the neutron Coincidence Collar[2] in both active and passive modes. The contents of the Los Alamos cylinders are described in Table I. Each cylinder contains approximately the same uranium mass, but the enrichments vary widely. Data on Model 5A and other standard United States $UF_6$ cylinders can be found in Ref. 10.

TABLE I

DESCRIPTION OF LOS ALAMOS MODEL 5A $UF_6$ CYLINDERS

| Item | $UF_6$ (kg) | $^{235}U$ (wt%) | U (kg) | $^{235}U$ (kg) |
|---|---|---|---|---|
| 1 | 22.262 | 19.94 | 15.031 | 2.997 |
| 2 | 20.449 | 46.80 | 13.793 | 6.455 |
| 3 | 22.621 | 97.67 | 15.224 | 14.869 |

In the active mode, the collar employs an external AmLi source that produces random neutrons (not time correlated) with an average energy of ~300 keV. These external source neutrons are slowed in the polyethylene matrix before interrogating the cylinder. The source used for these studies was MRC-70 with a yield of $4.67 \times 10^5$ n/s. This yield is approximately four times the maximum passive $(\alpha,n)$ yield of the cylinders. In the active method, we simultaneously measure the coincidence rate R and totals rate T using the AmLi source to drive the fission reactions. The AmLi source is then removed from the system to measure the passive R and T values from the $UF_6$. The net coincidence rate is used for fissile assay and the totals rates are used to correct the coincidence rates for deadtime effects. For active mode measurements, the Coincidence Collar was configured around the Model 5A cylinders, as shown in Fig. 5.

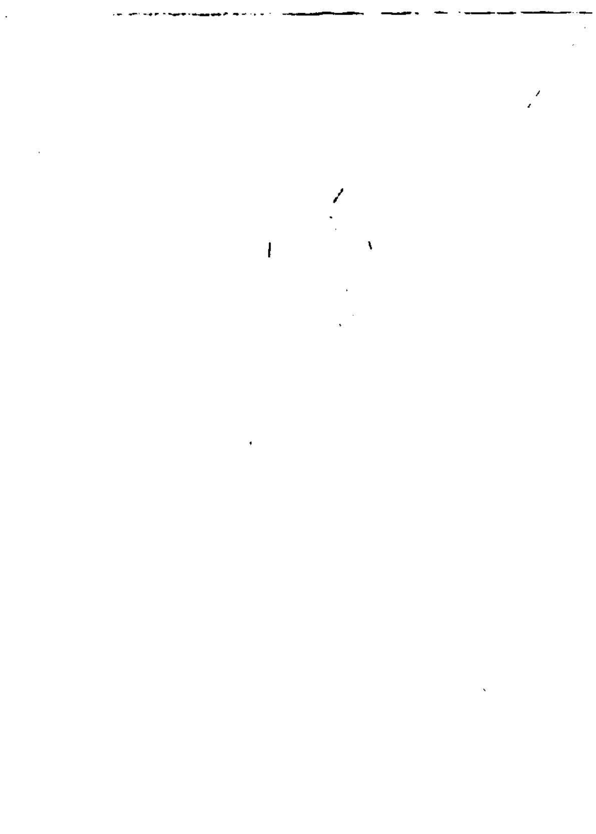


Fig. 5. Neutron Coincidence Collar (active mode) surrounding a Model 5A $UF_6$ cylinder. The external AmLi interrogation source rests on its polyethylene holder.

In the passive configuration of the collar, the polyethylene block housing the AmLi source is replaced by a fourth bank of six polyethylene-moderated $^3He$ counters. For the passive/active mode or self-interrogation measurements, the cylinders were centered in the sample cavity as shown in Fig. 6. For both active and passive/active applications of the collar, measurements were made with and without a cadmium liner for the cavity.

The following techniques for fissile assay were tested using the collar.

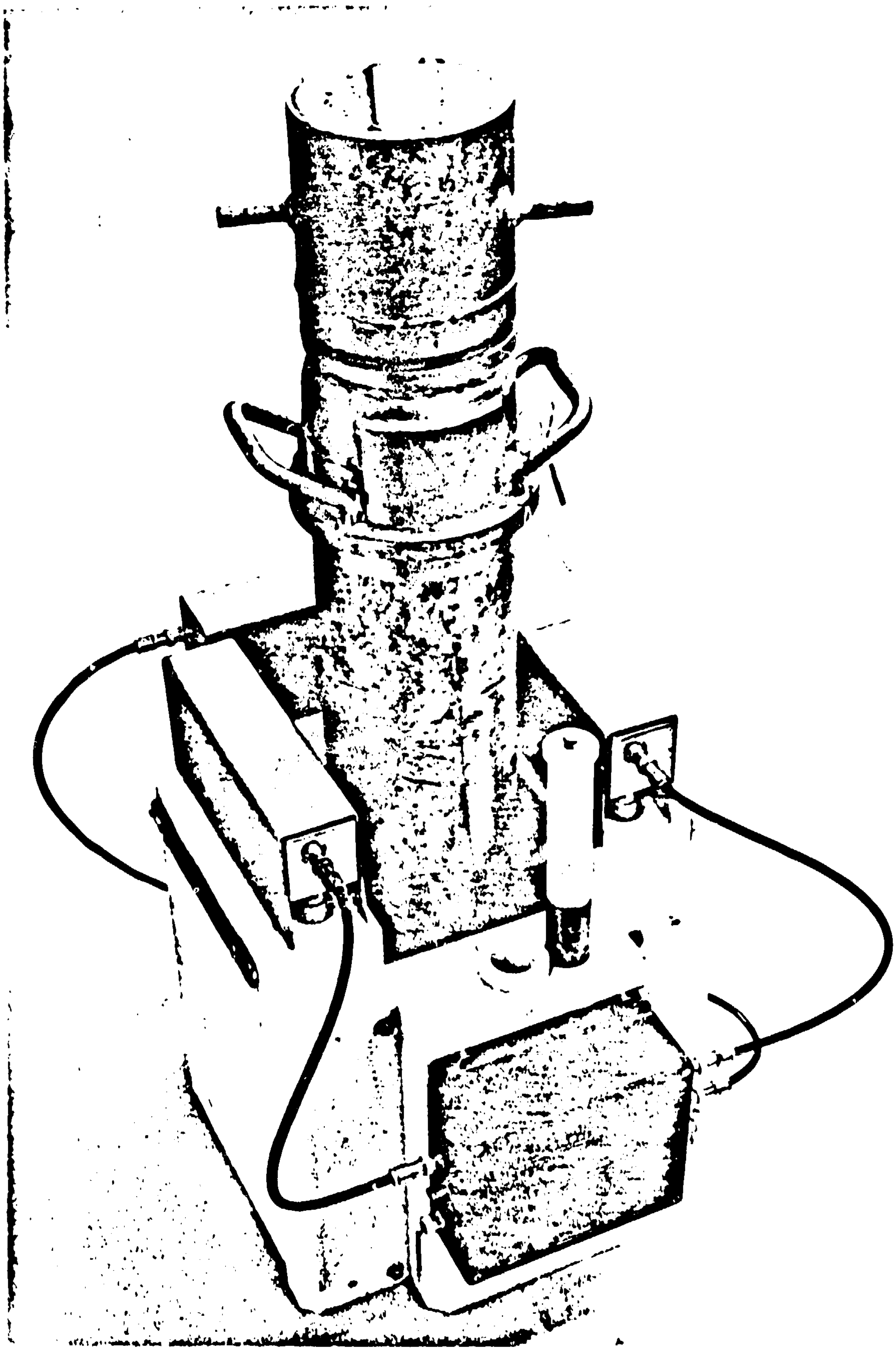
<u>Active (External AmLi)</u>

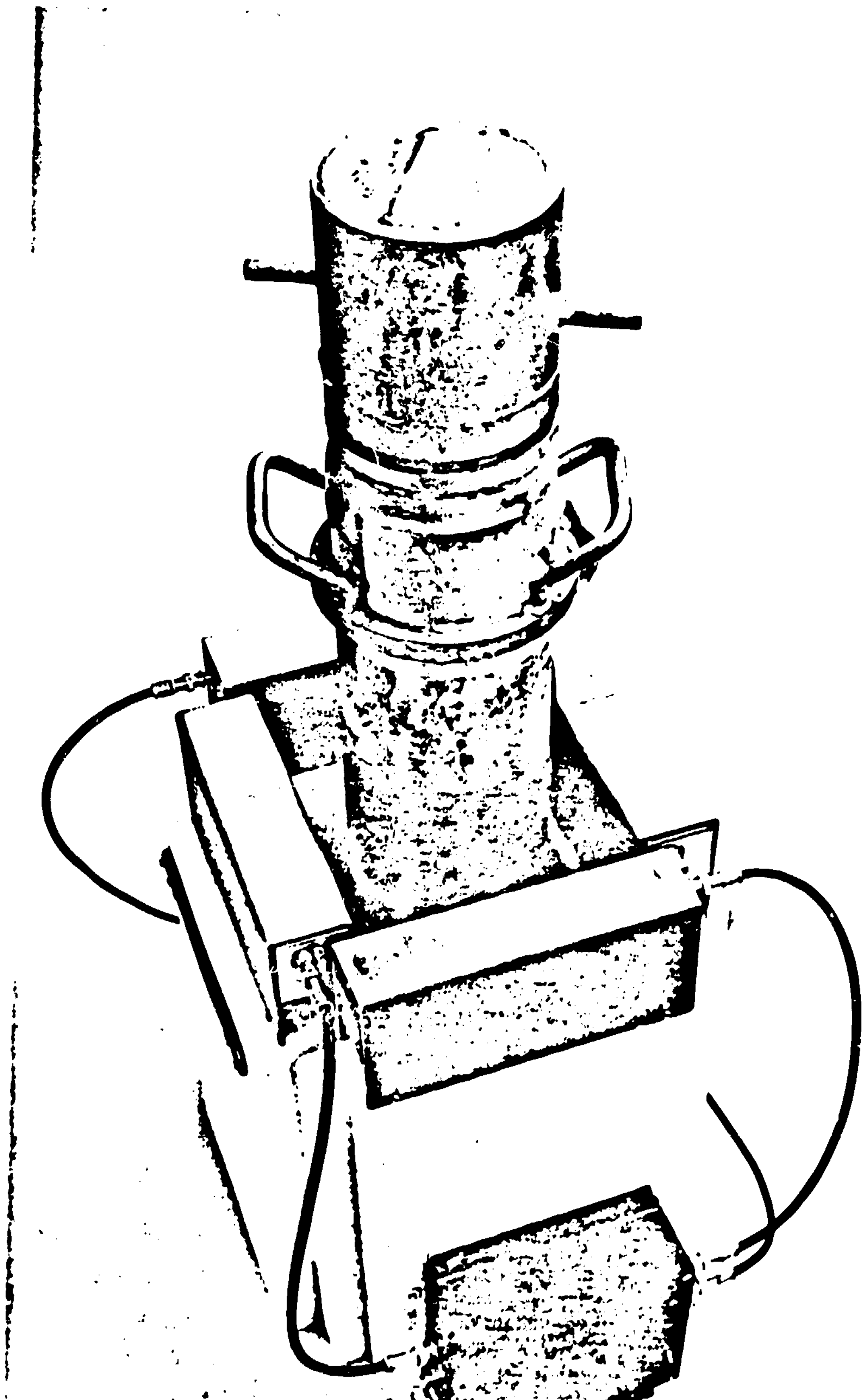
- Net coincidence; epicadmium ($\Delta R_{Cd}$)
- Net coincidence; no cadmium ($\Delta R_{no\ Cd}$)

<u>Passive/Active (Self-interrogation)</u>

- Normalized coincidence; epicadmium [$(R/T)_{Cd}$]
- Normalized coincidence; no cadmium [$(R/T)_{no\ Cd}$]
- Normalized net coincidence; thermal neutron return ($\Delta R/T_{no\ Cd}$)

Fig. 6. Neutron Coincidence Collar (passive mode) surrounding a Model 5A $UF_6$ cylinder. A fourth bank of polyethylene-moderated $^3He$ counters replaces the AmLi source assembly in the passive mode.





The results of initial laboratory measurements indicated viability of all the above techniques for measuring $^{235}U$ content of Model 5A cylinders. Each of the methods resulted in monotonically increasing response as a function of $^{235}U$ mass. Features of the various methods are compared in Table II. The precisions quoted in Table II are due to counting statistics only.

Table II allows an intercomparison of techniques with regard to counting precision, and the simplicity and length of the measurement procedure. In order of preference, the techniques are

1. $(R/T)_{Cd}$ - passive
2. $(R/T)_{no\ Cd}$ - passive
3. $(\Delta R)_{Cd}$ - active
4. $(\Delta R)_{no\ Cd}$ - active
5. $(\Delta R/T_{Cd}$ - passive

The first two passive techniques proved reliable signatures for bulk $^{235}U$ mass and are preferred over active methods primarily because of simplicity. The last passive technique lacks penetrability for bulk assay, but a related signature proved useful for fill-height corrections to field measurements.

Using only the $(R/T)_{Cd}$ method, further cylinder measurements were made at Los Alamos using the HLNCC, the DRCC, and two versions of the AWCC in the passive mode. A schematic of the two-ring AWCC (active mode) is shown in Fig. 7. The three-ring AWCC uses 60 $^3He$ tubes in three concentric rings in a polyethylene matrix. Compared with the two-ring AWCC, it has a higher efficiency and a smaller diameter cavity. Figure 8 shows measured values of $(R/T)_{Cd}$ vs $^{235}U$ mass obtained using the four well counters. These plots have been corrected for the small $^{238}U$ contribution to R. This correction is ~7% of the induced-fission signal for the 20%-enriched sample, ~1% for the 50%-enriched sample, and ~0.01% for the 98%-enriched sample.

The differences in the slopes of the plots for the four instruments are due to differences in counting efficiency and coupling between sample and detector. $(R/T)_{Cd}$ is directly proportional to the counting efficiency and increases with the fraction of returning neutrons intercepting the cylinder. Based on the results of the laboratory intercomparison of the four well counters, the three-ring AWCC in the passive mode was determined to have the best combination of efficiency and cavity size for field measurements of Model 5A cylinders.

### Piketon GDP Exercise

Following laboratory measurements that identified optimum choices of counting technique and instrument, arrangements were made for the first field exercise at the GAT GDP enrichment facilities near Piketon, Ohio. Before shipment of the three-ring passive AWCC (henceforth known simply as the PWCC), the fixed cadmium liner was

TABLE II

COMPARISON IN FEATURES OF COINCIDENCE COLLAR MEASUREMENT METHODS FOR MODEL 5A $UF_6$ CYLINDERS

| | Measurement Technique | | | | |
|---|---|---|---|---|---|
| | Passive | | | Active | |
| Feature | $(R/T)_{Cd}$ | $(R/T)_{no\ Cd}$ | $\Delta R/T_{Cd}$ | $(\Delta R)_{Cd}$ | $(\Delta R)_{no\ Cd}$ |
| AmLi source | No | No | No | Yes | Yes |
| Cadmium liner change | No | No | Yes | No | No |
| Uranium-238 correction | Yes | Yes | No | No | No |
| No. measurements[a] | 1 | 1 | 2 | 2 | 2 |
| Precision of $^{235}U$ measurement (%)[b] | 0.6 | 0.5 | 2.0 | 1.1 | 0.9 |

[a]Measurements of 1000 s.
[b]Item 3 (97.67% enrichment).

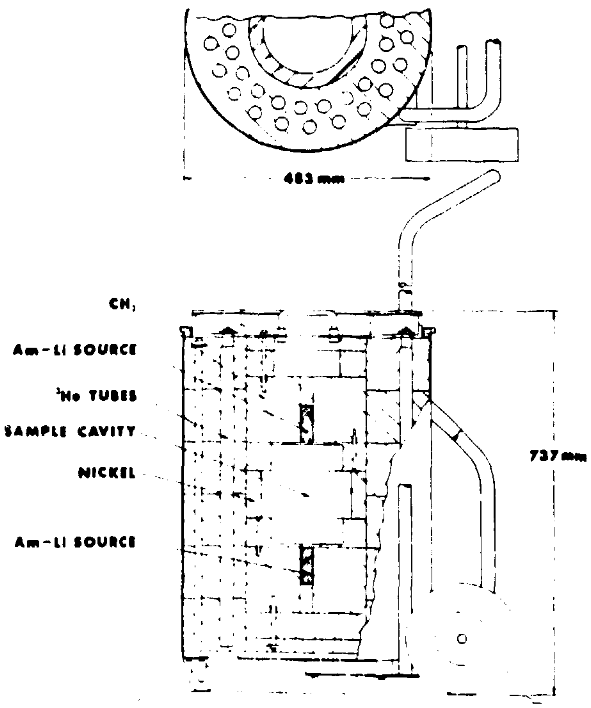



Fig. 7. Schematic of the AWCC Model II. This unit has two rings of $^3$He detectors. The unit used for field measurements has three rings of detectors.

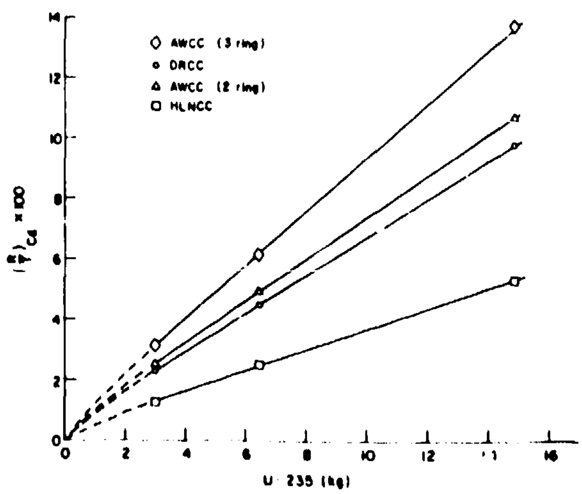


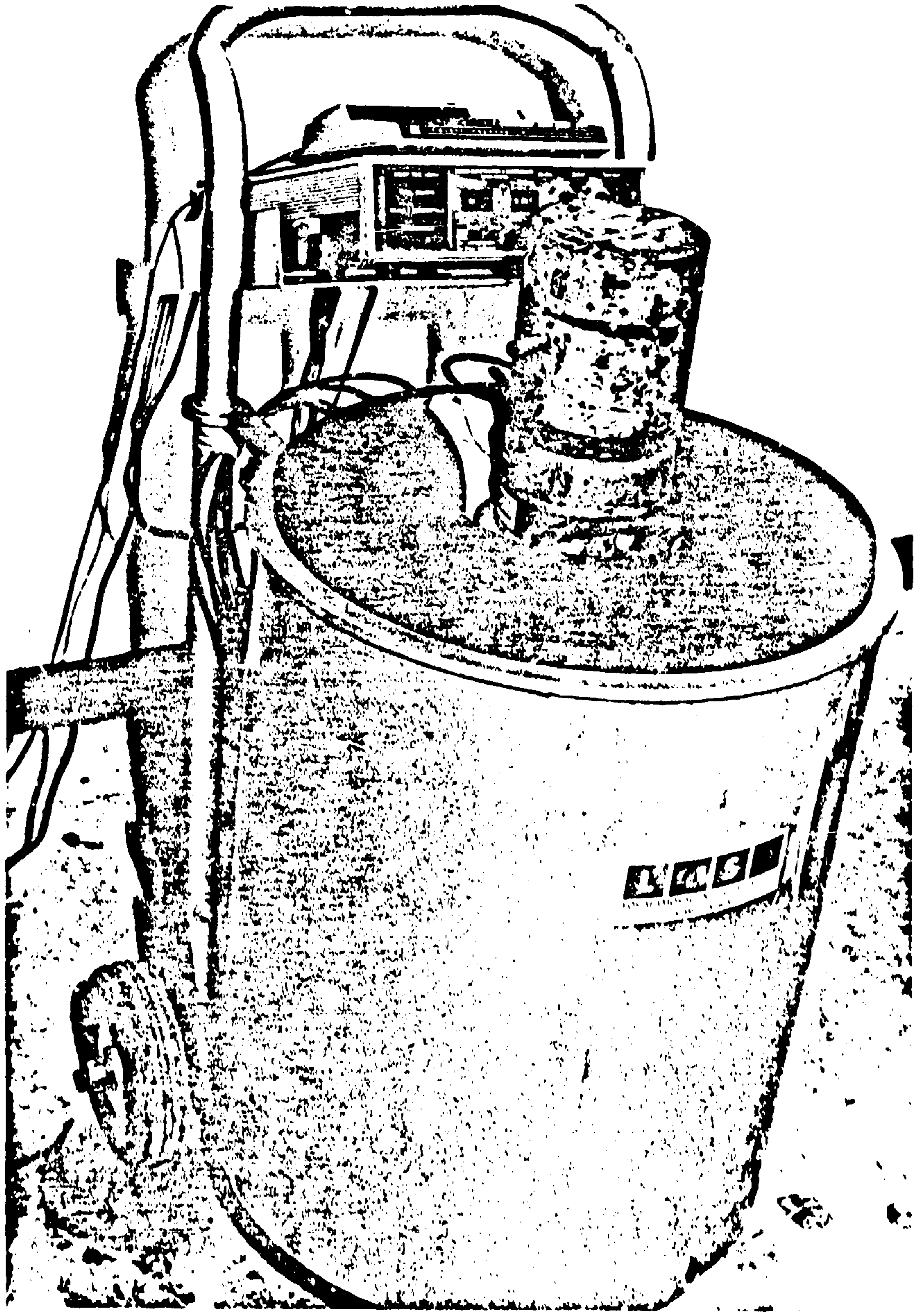

Fig. 8. The passive $(R/T)_{Cd}$ signature vs $^{235}U$ mass for laboratory measurements of three Model 5A cylinders in four separate thermal neutron coincidence counters.

replaced by one that could be easily removed and inserted. The capability to measure both R and T with and without the liner would be desirable to interpret results obtained with partially filled cylinders.

For a $^{252}Cf$ neutron source placed in the center of the cavity, the efficiency of the PWCC is ~40%. This is compared with the two-ring AWCC at ~30%, the DRCC at 21%, and the HLNCC at 12%.

Figure 9 is a photograph of the PWCC with a Model 5A cylinder in the cavity during the field exercise at GAT. The electronics unit used with the PWCC is identical to that used with the HLNCC.[5] The unit contains high- and low-voltage power supplies, six amplifier-discriminator lines, a microprocessor, and

Fig. 9. Field photograph of the three-ring AWCC during a passive measurement of a Model 5A $UF_6$ cylinder.



shift-register[8] coincidence logic. The unit is interfaced directly to a Hewlett-Packard HP-97 programmable calculator. The microprocessor is used to read out the run time, total counts, real-plus-accidental coincidence counts, and accidental coincidence counts into the HP-97. A special data reduction program for the HP-97 was prepared for the exercise.

A total of 53 cylinders was measured at the average rate of 4 to 5 per hour during 2.5 days at GAT. These included four small Model 1S sample cylinders. These measurements were made to calibrate $UF_6$ and $UO_2F_2$ neutron yields for cascade holdup surveys. One Model 5A cylinder containing 0.711% (normal) enrichment $UF_6$ was measured to verify the $^{238}U$ correction determined at Los Alamos. Four cylinders were measured only with the cadmium well liner in place. The remaining 44 cylinders were measured both with and without the liner. These are listed in order of measurement in Table III along with tag assay values, masses of $UF_6$, uranium, and $^{235}U$. Special features of the cylinders are also noted.

TABLE III

LIST OF CYLINDERS ASSAYED AT PIKETON GASEOUS DIFFUSION PLANT

| Cylinder | $UF_6$ (kg) | U (kg) | Tag Assay (wt% $^{235}U$) | $^{235}U$ (kg) | Special Features |
|---|---|---|---|---|---|
| 1S | 22.892 | 15.457 | 26.19 | 4.048 | R($T_c$) |
| 2M | 23.721 | 16.012 | 31.75 | 5.084 | |
| 3M | 24.530 | 16.545 | 57.61 | 9.532 | |
| 4M | 24.859 | 16.730 | 97.26 | 16.272 | |
| 5M | 23.700 | 15.960 | 76.78 | 12.254 | |
| 6M | 23.235 | 15.649 | 82.39 | 12.925 | |
| 7M | 24.966 | 16.802 | 97.26 | 16.342 | |
| 8M | 24.700 | 16.645 | 60.04 | 9.994 | |
| 9M | 23.659 | 15.948 | 66.67 | 10.633 | |
| 10M | 22.846 | 15.421 | 22.22 | 3.427 | |
| 11M | 22.738 | 15.312 | 93.15 | 14.263 | |
| 12M | 21.080 | 14.218 | 48.83 | 6.943 | |
| 13M | 24.737 | 16.675 | 72.81 | 12.141 | |
| 14M | 23.925 | 16.130 | 56.15 | 9.057 | |
| 15M | 16.119 | 10.884 | 26.37 | 2.870 | P(69%) |
| 16M | 24.702 | 16.647 | 60.04 | 9.995 | |
| 17M | 9.780 | 6.591 | 72.85 | 4.802 | P(42%) |
| 18M | 22.318 | 15.027 | 93.22 | 14.008 | |
| 19M | 24.044 | 16.237 | 26.44 | 4.293 | |
| 20M | 23.864 | 16.113 | 33.50 | 5.398 | |
| 21M | 24.016 | 16.177 | 79.04 | 12.786 | |
| 22M | 23.975 | 16.124 | 73.10 | 11.787 | |
| 23M | 25.008 | 16.830 | 97.20 | 16.359 | A |
| 24M | 24.262 | 16.355 | 72.95 | 11.931 | |
| 25M | 23.463 | 15.826 | 54.62 | 8.644 | |
| 26M | 23.597 | 15.919 | 51.25 | 8.158 | |
| 27M | 23.910 | 16.108 | 79.97 | 12.882 | A |
| 28M | 20.562 | 13.883 | 20.39 | 2.831 | |
| 29S | 21.119 | 14.268 | 5.96 | 0.851 | |
| 30S | 24.350 | 16.397 | 89.23 | 14.631 | |
| 31S | 20.[illegible]8 | 14.044 | 95.65 | 13.433 | |
| 32S | 22.799 | 15.392 | 26.53 | 4.083 | R($T_c$) |
| 33S | 24.178 | 16.315 | 37.52 | 6.121 | R($T_c$) |
| 34S | 17.950 | 12.105 | 58.30 | 7.057 | R($T_c$),R(234),P(76%) |
| 35M | 21.395 | 14.418 | 73.1. | 10.542 | A |
| 36M | 22.528 | 15.164 | 97.62 | 14.803 | |
| 37M | 23.830 | 16.038 | 97.26 | 15.599 | |
| 38M | 24.559 | 16.528 | 97.27 | 16.077 | |
| 39M | 24.572 | 16.537 | 97.28 | 16.087 | |
| 40M | 6.723 | 4.531 | 79.11 | 3.584 | P(29%) |
| 41S | 11.767 | 9.956 | 72.04 | 7.172 | P(63%) |
| 42S | 13.653 | 9.213 | 24.04 | 2.215 | P(58%) |
| 43S | 23.664 | 15.978 | 23.55 | 3.763 | |
| 44M | 24.494 | 16.521 | 57.86 | 9.559 | |

Special feature codes:

- R($T_c$) - reject code for $T_c$ > 1.5 ppm uranium.
- R(234) - reject code for abnormal $^{234}U$ concentration.
- P(XX) - partially filled cylinder (XX full).
- A - analysis requested, outlier.
- M - Monel cylinder.
- S - steel cylinder.

The list includes cylinders only partially filled, reject cylinders with high concentrations of the fission product technetium ($T_c$) and cylinders that gave discrepant responses for no apparent reason. The three cylinders in the last category we labeled outliers and analysis has been requested. Of the 44 cylinders listed in Table III, 3 have high $T_c$ and 6 were partially filled. One cylinder (34S) shared both features as well as having abnormal $^{234}U$ content. The remaining 35 we called full, although their $UF_6$ masses varied from 20.6 to 25 kg.

The PWCC was located in the rear of the vault building where Model 5A cylinders are stored. Cylinders were lowered into the counter using a fork-lift truck modified for cylinder-handling operations. Typically, a measurement was made first without the cadmium liner; the cylinder was raised while the liner was inserted, the cylinder was lowered, and another measurement was started. Figure 10 is a photograph of a measurement in progress.

Count times for the two measurements were ~3 min for high fissile mass samples and ~6 min for low $^{235}U$ mass samples. Cylinder-handling operations typically required approximately the same time as for counting.

Figure 11 displays real coincidence counts (reals) divided by total counts (totals) with the cadmium well liner in place plotted vs

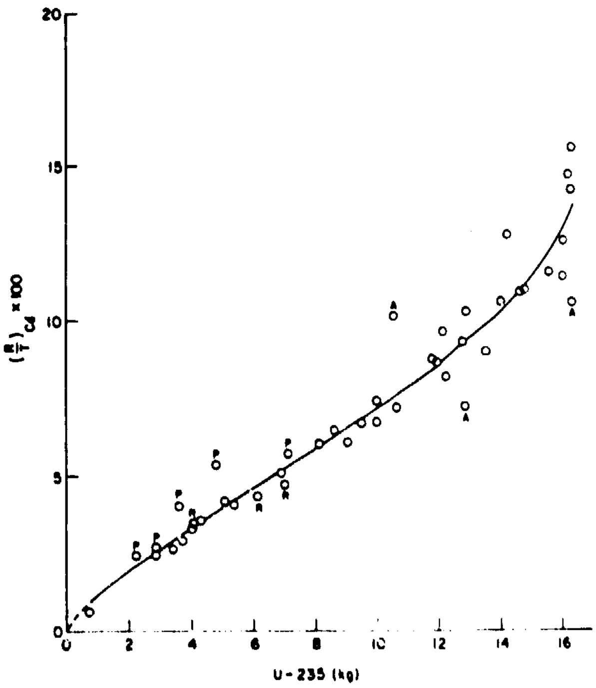
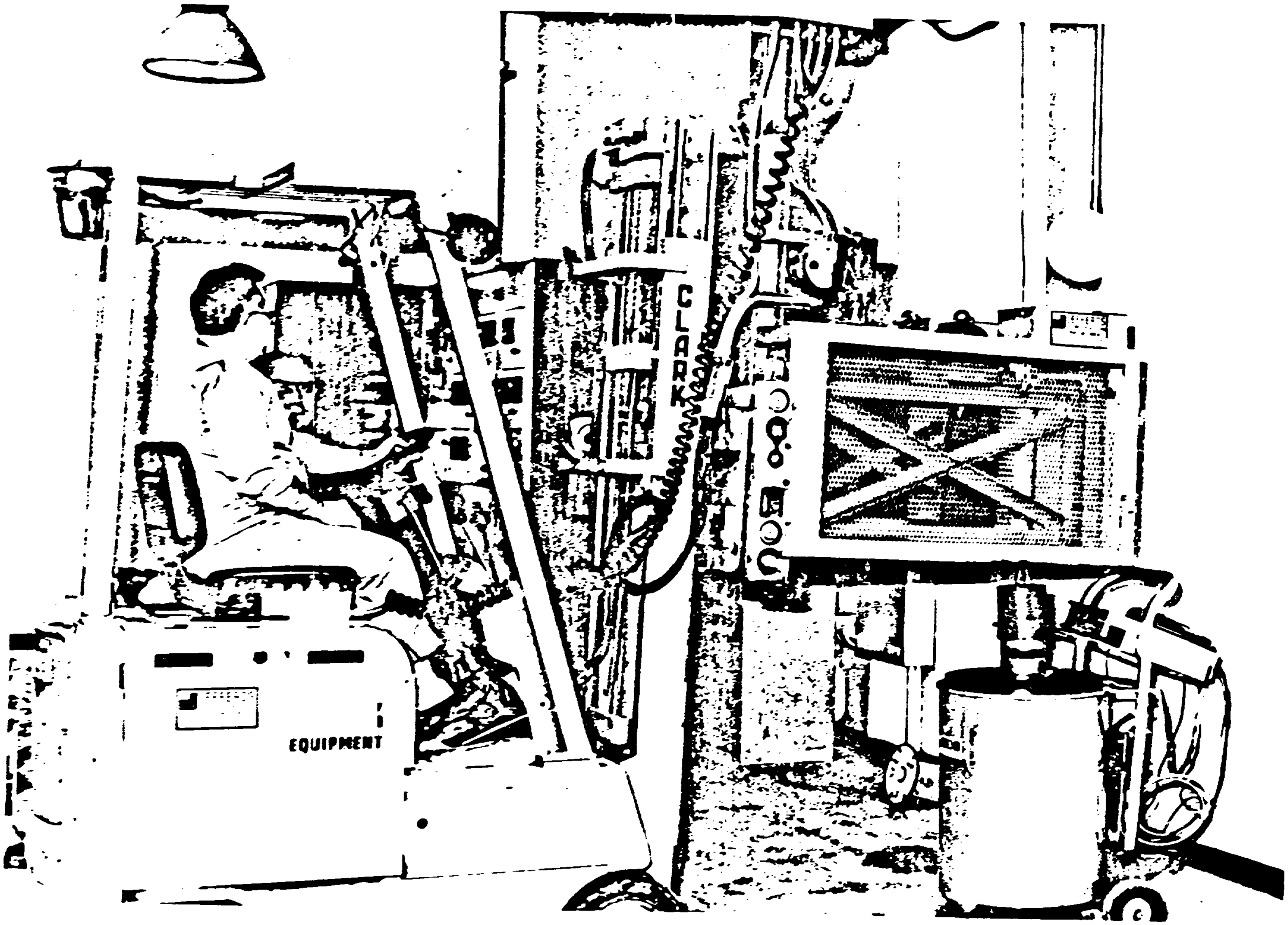



Fig. 11. The passive $(R/T)_{Cd}$ signature (uncorrected) vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid curve is an unweighted least squares cubic polynomial fit to the data.

Fig. 10. Field photograph showing the three-ring AWCC during a passive measurement of a Model 5A cylinder suspended by the cylinder-handling truck.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stated $^{235}U$ mass. The reals have been corrected for $^{238}U$ spontaneous fission contributions and the totals have been corrected for vault background. The $^{238}U$ correction was a constant 19.8 counts/s for all measurements. This value was obtained from laboratory measurements of depleted uranium metal cubes and agrees well with the value obtained using the normal enrichment cylinder at GAT. This correction is actually dependent on $^{238}U$ mass in the sample, but the correction is relatively so small at high enrichments that the constant value taken for a 20%-enrichment cylinder (full) is adequate for other cases. The vault totals background varied from 80-150 counts/s during the exercise and was frequently updated during remote cylinder-handling operations.

Random errors ($\pm 1\sigma$) in measured quantities are indicated in Figs. 11-16 by the size of the circles. Excellent reproducibility was obtained for repeated measurements. Also, with a cylinder centered in the sample cavity and with the same cylinder touching the cavity wall, essentially the same results were obtained.

The labeled points of Fig. 11 correspond to the special feature codes of Table III, that is, P stands for partial fills, R stands for reject cylinders (for example, with high $T_c$), and A stands for outliers on which analysis will be done. Measured values of $(R/T)_{Cd}$ for 44 cylinders were fitted with an unweighted, least squares cubic polynomial that is shown also in Fig. 11. The partial-fill data points all lie above the fitted curve of Fig. 11. This is because a given $^{235}U$ mass contained in a partially filled cylinder will result in a higher leakage multiplication than the same $^{235}U$ mass in a full cylinder. Figure 4 displays this effect well. Uranium-235 mass density and sample geometry are the two key parameters determining sample multiplication. Because $(R/T)_{Cd}$ is a measure of sample multiplication, the partially filled cylinders have higher response than full cylinders for the same $^{235}U$ mass. The reject cylinders that are problems for NaI enrichment measurements are not obvious problems with the $(R/T)_{Cd}$ technique. Figure 11 also displays significant scatter in the data points for the high-mass samples that is not due to counting statistics. The fitted curve is nearly linear from 2-14 kg $^{235}U$, but for the higher mass samples, multiplication effects become more pronounced.

Figure 12 displays measured values of $(R/T)_{no\ Cd}$ plotted vs $^{235}U$ tag masses as well as the cubic polynomial fit to these data. Compared with the $(R/T)_{Cd}$ data, the partial fills would be more accurately assayed using the

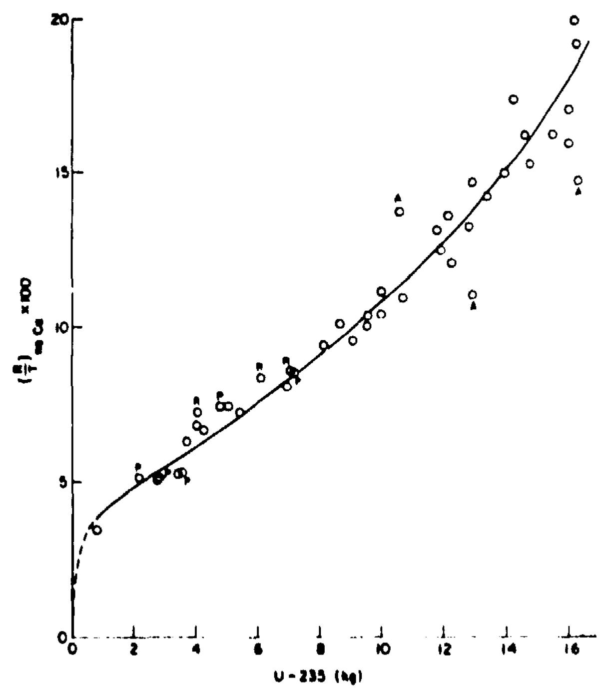


Fig. 12. The passive $(R/T)_{no\ Cd}$ signature vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid curve is an unweighted least squares cubic polynomial fit to the data.

$(R/T)_{no\ Cd}$ fit, but the scatter in the high-mass sample data is nearly the same for the two signatures.

Figure 13 is a plot of the difference in reals rates (with or without cadmium liner) divided by the totals rate with cadmium. The plot is included for illustration only because it displays response to thermal neutron albedo and is roughly proportional to the surface area of the sample. The significant feature of Fig. 13 is that the partial-fill data points all lie below the fitted curve except for one point. This is consistent because the partial fills all have smaller sample surface areas than the full cylinders. Figures 12 and 13 exhibit the effects of sample self-shielding in the lower $^{235}U$ mass region and the effects of sample multiplication in the high-mass region. These same features are known from active applications of the AWCC.

Another signature of interest, $R_{Cd}/\Delta T$, is plotted in Fig. 14. The measured quantity $\Delta T$ is the difference in totals rates because of the cadmium liner. $R_{Cd}/\Delta T$ is inversely proportional

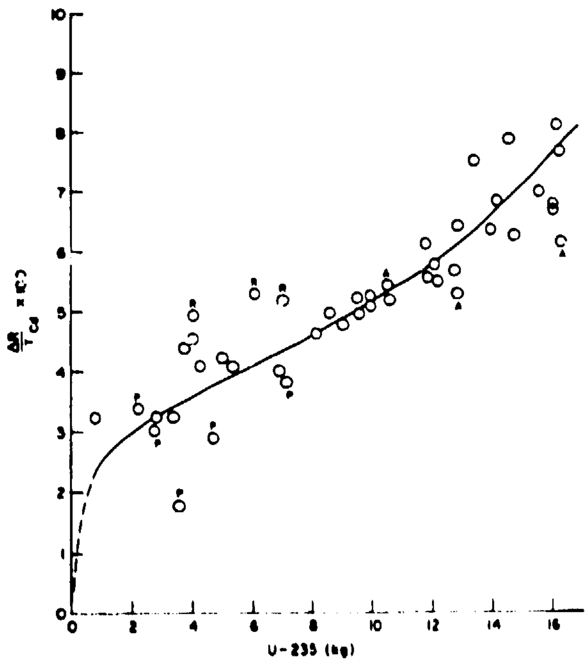


Fig. 13. The passive $\Delta R/T_{Cd}$ signature vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid curve is an unweighted least squares cubic polynomial fit to the data.

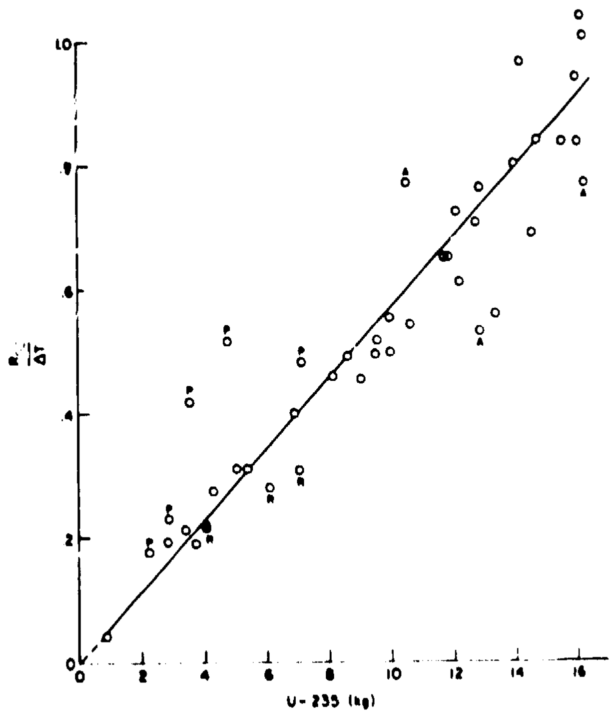


Fig. 14. The passive $R_{Cd}/\Delta T$ indicator vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid line is a linear fit to the data.

to the sample surface area and directly proportional to the sample coincidence multiplication. The $R_{Cd}/\Delta T$ indicator has the interesting property of magnifying the deviations of the partial fills and outliers from the straight-line fit. This feature suggested $R_{Cd}/\Delta T$ for use in a correction algorithm for the $(R/T)_{Cd}$ data. Several forms of correction algorithms were considered, but the one found most effective has the form

$$CF = 1 - \frac{k}{T_{Cd}^{\,n}} \frac{R_{Cd}}{\Delta T} ,$$

where CF represents the correction for application to $(R/T)_{Cd}$ signature data and k and n are empirical constants. $T_{Cd}$ is plotted vs $^{235}U$ mass in Fig. 15. Values of k and n were determined by minimizing the standard deviation of differences in tag $^{235}U$ masses and assay values determined from fitted curves through the corrected $(R/T)_{Cd}$ values. The "best" values of k and n were found to be 8.0 and 0.25, respectively, using an iterative procedure.

With the chosen form of the correction factor, both R and T are measured with the cadmium liner in place, and only the totals rate is required with the liner removed. For the cylinders of Table III, totals rates (no cadmium liner) varied between 1800 and 85 000 counts/s. Thus, the form of the correction factor chosen minimizes counting time because the no-cadmium measurement need require only a few seconds as a result of the high totals rates. Generally, the precision of the $R_{Cd}$ measurement will dominate the precision of CF $\ast$ $(R/T)_{Cd}$.

Corrected values of $(R/T)_{Cd}$ signature data are plotted in Fig. 16. When these data are compared visually with those of Fig. 11 [uncorrected $(R/T)_{Cd}$ data], much less scatter is evident. Table IV is a quantitative comparison of $(R/T)_{no\ Cd}$ and corrected $(R/T)_{Cd}$ signatures for $^{235}U$ assay. The correction algorithm improved the assay accuracy (1σ) from 10.6% to 6.8%, or a factor of 1.6. Also, the algorithm markedly improved accuracy for partial fills and high-mass samples. Table IV applies to all 44 cylinders measured at GAT, both with and without the

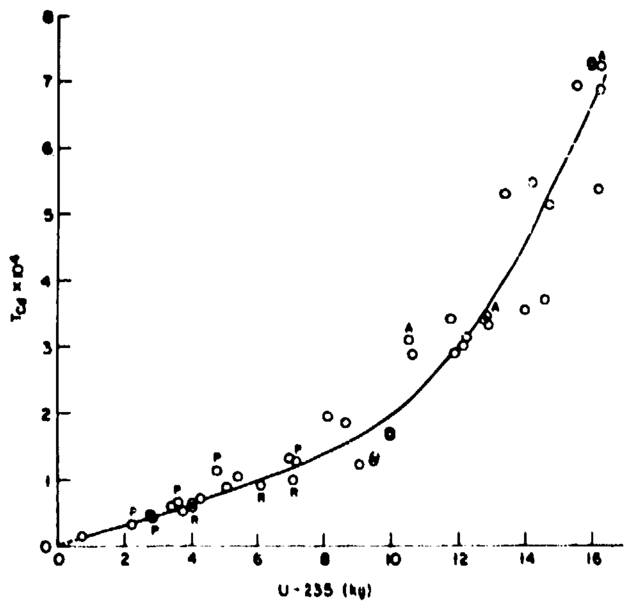


Fig. 15. Total count rate (with cadmium liner) vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid curve is an unweighted least squares cubic polynomial fit to the data.

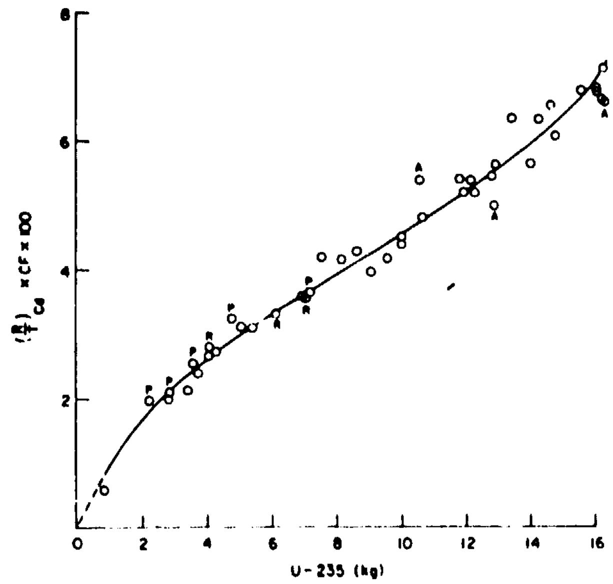


Fig. 16. The corrected, passive $(R/T)_{Cd}$ signature vs $^{235}U$ mass for field measurements of 44 Model 5A cylinders. The solid curve is an unweighted least squares cubic polynomial fit to the data.

TABLE IV

COMPARISON OF ASSAY ACCURACY FOR THREE SELF-INTERROGATION SIGNATURES OF $^{235}U$ MASS IN MODEL 5A CYLINDERS

| | Fitted Cubic Polynomial Coefficients[a] | | | | |
|---|---|---|---|---|---|
| Signature | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $\sigma_d(\%)$ |
| $(R/T)_{Cd}$ | -3.38411 E-01 | 1.06083 E+00 | 9.93987 E-02 | -6.53091 E-03 | 10.6 |
| $(R/T)_{no\ Cd}$ | -6.73982 E+00 | 2.02818 E+00 | -4.76277 E-02 | 2.89142 E-04 | 10.0 |
| $CF * (R/T)_{Cd}$ | 8.44352 E-01 | -6.49284 E-01 | 9.13669 E-01 | -7.28569 E-02 | 6.8 |

[a] $M = A_0 + A_1x + A_2x^2 + A_3x^3$,
$M$ = $^{235}U$ assayed mass,
$x$ = signature.

cavity liner, that is, $\sigma_{\bar{d}}$ represents one standard deviation (relative) of the differences in assay and tag values for all cylinders. Table V shows relative differences in assay and tag values for problem cylinders, that is, partial fills or those with reject codes. Assay values were determined using the CF * $(R/T)_{Cd}$ calibration curve. The standard deviation of relative differences between assay and tag values for problem cylinders is 9.8%. The partially filled cylinder assays were not as accurate as those for the reject cylinders that have been difficult to assay using the NaI/enrichment meter technique. Generally, however, the assay accuracies for problem cylinders were nearly as good as for the 44-cylinder population as a whole.

## Conclusions

The feasibility of a new technique for direct fissile mass verification of $UF_6$ containing HEU using random source self-interrogation and neutron coincidence response has been successfully demonstrated in the laboratory and during the first field test. The corrected $(R/T)_{Cd}$ signature is the most accurate of those evaluated. This signature yields verification of bulk $^{235}U$ mass in the $UF_6$ sample within only a few minutes of counting without the use of an external interrogation source and with an assay accuracy of 6.8% (1σ). The passive $(R/T)_{Cd}$ signature method is the first nondestructive assay technique able to verify the entire bulk fissile mass in a Model 5A cylinder. As such, it should be used in conjunction with gamma-ray enrichment measurements for routine verification of product storage cylinders. The neutron and gamma-ray methods together provide a much more powerful verification tool than either method alone.

TABLE V

ASSAY ACCURACIES OF "PROBLEM" CYLINDERS

| Cylinder | Special Features[a] | $d = \frac{\text{Assay} - \text{Tag}}{\text{Tag}}$ (%) |
|---|---|---|
| 1B | $R(T_c)$ | +3.7 |
| 15M | P(69%) | -1.2 |
| 17M | P(42%) | +21.9 |
| 32B | $R(T_c)$ | +[illegible] |
| 33B | $R(T_c)$ | +0.2 |
| 34B | $R(T_c)$,R(234),P(76%) | -3.7 |
| 40M | P(29%) | +8.2 |
| 41B | P(63%) | -0.7 |
| 42B | P(58%) | +21.9 |
| | | $\bar{d}$ = 7.0% |
| | | $\sigma_{\bar{d}}$ = 9.8% |

[a]Defined in Table III.


## ACKNOWLEDGMENTS

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