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Rover Scrap with the Random Source  
Interrogation System

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The logo features a central circle with a '+' sign inside. This circle is surrounded by a larger circle with several dots on its circumference. A vertical line passes through the center of the circles, and a wavy line extends upwards from the top of the logo.

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ASSAY OF THE URANIUM CONTENT OF ROVER SCRAP  
WITH THE RANDOM SOURCE INTERROGATION SYSTEM

by

J. E. Foley and L. R. Cowder

ABSTRACT

A neutron interrogation instrument has been built that determines the uranium content of 10-liter containers of scrap material from the Rover nuclear rocket program. The instrument determines uranium content by producing fissions in the sample, and then detecting these fissions by coincidence counting the prompt-fission neutrons and gamma rays. A measurement procedure has been developed that gives assays that are relatively independent of the matrix material in the sample. A normalization procedure has also been developed that eliminates the effects of instrumentation drifts.

I. INTRODUCTION

The termination of the Rover nuclear rocket program in June 1973 made it necessary to do a complete assay of the entire Rover uranium fuel inventory. This inventory consisted of intact fuel elements, damaged fuel elements, and scrap from the fuel-manufacturing process that had accumulated over the many years of production. The uranium content of the intact fuel elements was well known because of the quality control on the production process, but the uranium content of the damaged elements and the scrap was not well known. The uranium in the damaged elements and in the scrap was to be recovered by the uranium recovery facility at the Los Alamos Scientific Laboratory. Uranium recovery had been going on for many years, but a large backlog of scrap containers still existed at the time of the termination of the Rover program. The damaged elements were broken into

small pieces and stored in 10-liter stainless steel "hospital cans" (208-mm-o. d. by 254-mm-high). The scrap, composed of graphite dust and small pieces of fuel elements, was also stored in these containers.

It was necessary to develop an assay instrument that could be used at the recovery facility to determine the uranium content of the incoming Rover samples, and that could be operated by the personnel at the facility.

For many years, Rover scrap samples had been assayed by the delayed-neutron technique.<sup>1</sup> This technique, which requires the use of either a Van de Graaff accelerator or a 14-MeV neutron generator, has been shown to be very accurate; but, because of the complexity of the instrumentation, is not easily used for routine in-plant assay.

The gamma-ray assay technique,<sup>2</sup> which is used for the assay of low-density samples, could

not be used because these samples, which typically contain 500 to 3000 g of uranium, were too dense to obtain proper gamma-ray transmission corrections.

The nondestructive assay instrument that could possibly do the job was the Random Source Interrogation System<sup>3</sup> (which is more commonly called the "Random Driver"). This assay system had been used successfully at the Oak Ridge Y-12 plant<sup>4</sup> for the assay of high-enriched uranium scrap in small containers (125-mm-o. d.). The Random Driver, along with other instruments of the same generic type,\* determines the  $^{235}\text{U}$  content of a sample by detecting neutron and gamma rays from fissions which are produced by neutron interrogation from a small neutron source ( $\sim 10^6$  n/s). Since the enrichment of the uranium in the Rover material was constant (93%), measurement of the  $^{235}\text{U}$  content, as done by the Random Driver, would be sufficient for the determination of total uranium content.

A Random Driver assay instrument was designed and built especially for the Rover scrap assay problem. A measurement procedure has been developed that results in assays which are relatively independent of the composition of the sample. A normalization technique has been included in this procedure which eliminates the effects of instrumentation drifts.

## II. THEORY OF OPERATION

A top view of the Random Driver is shown in Fig. 1. A small fraction of the neutrons from the AmLi source ( $\sim 10^6$  n/s) interacts with the uranium in the sample, inducing fissions in the  $^{235}\text{U}$  atoms. Relatively few fissions occur in the  $^{238}\text{U}$  because the neutron-energy spectrum of the AmLi source is subthreshold ( $< 1$  MeV) for  $^{238}\text{U}$ . About two and one-half neutrons and about six gamma rays are

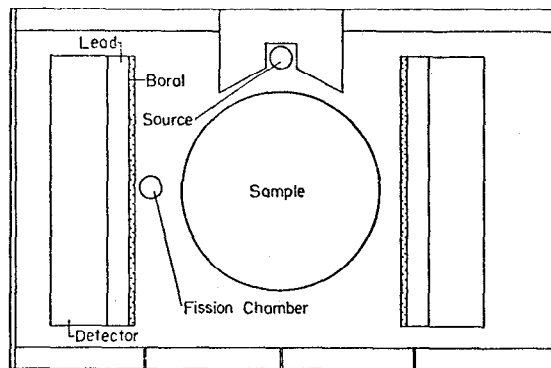


Fig. 1. Top view of the Random Driver.

produced when a  $^{235}\text{U}$  atom fissions. These neutrons and gamma rays are detected, in the presence of the relatively intense neutron flux from the AmLi source, by coincidence counting with two fast plastic scintillation detectors located on opposite sides of the sample. The fission neutrons and gamma rays are time-correlated (produced two or more at a time), whereas the AmLi source neutrons are produced randomly (uncorrelated, produced one at a time). The coincidence-counting technique, which requires the detection of two neutrons or gamma rays within a very short period of time, is used to distinguish the time-correlated, induced-fission neutrons and gamma rays from the uncorrelated, or random, source neutrons. If a neutron or gamma-ray detection in one of the fast plastic scintillation detectors is followed closely ( $< 40$  ns) by a detection in the second detector, then a coincidence event has been observed. The probability that such an event occurs is high for the time-correlated neutrons and gamma rays from fission, and it is low for the uncorrelated neutrons from the random source. The coincidence rate, being proportional to the quantity of the  $^{235}\text{U}$  in the sample, therefore provides a "signature" for determining the uranium content of the sample.

The energy spectrum of the interrogating neutrons from the source is kept "hard" (high energy) in order to obtain good penetration into the sample. A thermalized-neutron source will

\*The Isotopic Source Assay Systems manufactured by Intelcom Rad Tech, San Diego, CA; the Random Driver manufactured by National Nuclear Corp., Redwood City, CA; and FEFI manufactured by Texas Nuclear Corp., Austin, TX.

increase the induced-fission rate by a factor of at least 10, but assays made with this type of source are very sensitive to the density of the uranium and the matrix material in the sample<sup>5</sup> because of the poor neutron penetration into the sample. Hard-spectrum interrogation is justified, in spite of the lower induced-fission rate, because the assays, though not as precise, are consistently more accurate than those made by thermal-neutron interrogation.

Three corrections must be made to the measured coincidence rate before an accurate assay can be given. Corrections must be made for (1) a background change due to the presence of the sample in the counter, (2) neutron thermalization within samples containing hydrogenous materials, and (3) attenuation of the fission neutrons and gamma rays within the sample.

#### A. Background Correction

A background coincidence rate of 12.0 counts/s is observed in the Random Driver designed for the assay of the Rover scrap samples. Most of this background is from cosmic rays that, in passing through the system, produce simultaneous counts in both detectors, resulting in a coincidence count. A lower background rate is observed when the sample is placed into the counter, because some of the cosmic rays that ordinarily would travel through both detectors, and thus produce coincidence counts, are stopped within the sample; i. e., the sample shields the second detector from a fraction of the cosmic rays. The background coincidence rate with the sample in the counter cannot be measured in the presence of the AmLi source because this background coincidence rate cannot be distinguished from the induced-fission rate. Since it would be very time-consuming to remove the source and to measure the background for each sample, a different approach is necessary. It was determined experimentally that the change in the background due to the sample is a function of only the mass of the sample and not a function of the

composition of the sample for the low- and intermediate-Z materials investigated. Figure 2 illustrates this for the Rover scrap container filled with various densities of materials that are expected to be found in the samples: metal, graphite, and graphite flour. The background rate can be calculated from the weight of the sample. A separate background measurement thus does not need to be made for each sample.

#### B. Fission-Chamber Correction

Samples containing hydrogenous matrix materials show higher induced responses per gram of  $^{235}\text{U}$  than do samples without such materials. A  $^{235}\text{U}$  fission chamber, positioned inside the Random Driver close to the sample, is used to monitor the thermal flux within the sample. An increase in the response of the fission chamber indicates that there is a similar increase in the response from the sample itself. Normalization of the induced coincidence rate of the sample to the count rate of the fission chamber corrects the assay by eliminating the thermalization effects of the hydrogen.<sup>6</sup>

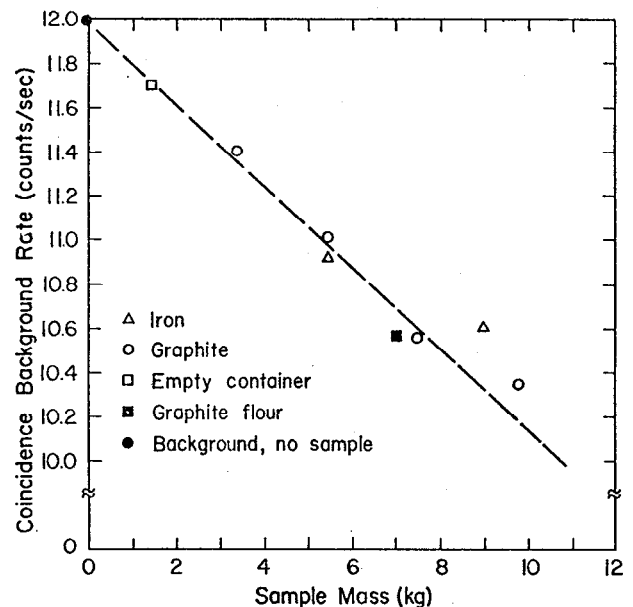


Fig. 2. Background coincidence rate of the Random Driver for various sample compositions and masses.

### C. Attenuation Correction

A fraction of the fission neutrons and gamma rays that are produced within the sample from the neutron interrogation are captured by the material inside the sample. In addition, elastic scattering of a fission neutron by the material inside the sample can reduce the energy of the neutron below the discrimination threshold level of the plastic scintillation detector, resulting in a loss of a coincidence count. Both processes, capture and elastic scattering, work to reduce the coincidence rate to a value that is lower than it should be; the resulting assay will then be low.

The  $^{252}\text{Cf}$  source-addition technique<sup>7</sup> is used to correct the assay for these losses. The attenuation correction is made in the following manner: the coincidence rate of a small spontaneous fission  $^{252}\text{Cf}$  source ( $\sim 10^4$  n/s) is determined by placing the source into the Random Driver; the coincidence rate of the source is then determined with the sample in the counter; the ratio of the coincidence rate obtained with the sample in the counter to that obtained without the sample gives an estimate of the decrease in the response due to the material in the sample. The attenuation is a function of both the density and the type of material in the sample. A heavy sample containing a matrix of 8 kg of iron, for example, exhibits a coincidence rate that is about 20% lower per gram of  $^{235}\text{U}$  than does a lightweight sample.<sup>8</sup> The  $^{252}\text{Cf}$  source is placed on the outside of the sample about 100 mm above the rotating turntable on which the sample sits (the source rotates with the sample). Measurements show that the attenuation correction will be approximately the same no matter whether the  $^{252}\text{Cf}$  source is located on the outer surface of the sample, or whether it is located anywhere inside the sample; i. e., the measured response is nearly independent of the position of the fission location within the sample.

The  $^{252}\text{Cf}$  source-addition technique also automatically corrects the assay for long-term

changes in the response caused by instrumentation drifts. A response increase of 10%, for example, perhaps caused by a severe temperature change or by a shift in photomultiplier tube voltage, will produce the same increase in response for both the sample and the  $^{252}\text{Cf}$  spontaneous-fission source. The normalization process thus removes the effects of this response change. It is necessary, of course, to periodically correct the normalization factors because of the decay of the  $^{252}\text{Cf}$  source (960-d half-life).

### III. ASSAY INSTRUMENT

The completed assay instrument is shown in Fig. 3. The turntable on which the sample is placed is located in the center of the counter between the two fast-plastic scintillation detectors. The fission chamber (Reuter-Stokes model RS-P6-1608-110) is located to the left of the turntable. In practice, two neutron sources, seen at the rear of the counter, are used instead of a single source. A uniform vertical response from the sample ( $\pm 3\%$ ) is obtained with the two sources (each 50, 8-mm-o. d. by 140-mm-long) spaced 250 mm between centers (see Appendix A). A narrower spacing between these sources results in a peaking of the

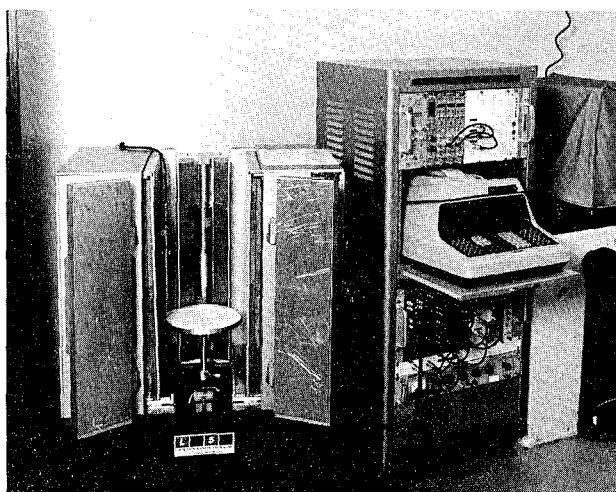


Fig. 3. Completed Random Driver assay system for assay of 10-liter containers.

response at the center of the sample, whereas a wider spacing results in a peaking of the response at the ends of the sample. The proper spacing for the flat response was determined experimentally.

The fast-plastic scintillation detectors are 50.8-mm-thick, 254-mm-wide, and 610-mm-long. The inner face of each detector is covered with a 25.4-mm-thick layer of lead and a 6.35-mm-thick layer of BORAL. The purpose of the lead is to reduce assay nonlinearities in dense samples by preventing the low-energy fission gamma rays, which suffer severe attenuation in these samples, from reaching the detectors. The BORAL keeps the neutron spectrum inside the assay system at high energy by absorbing neutrons that are thermalized in the plastic scintillation detectors and preventing them from reaching the sample.

The two doors, which are shown open in Fig. 3, make it easy for the operator to put the sample into the counter. These doors, which are closed during the assay, are covered with BORAL to eliminate thermalized room-return neutrons.

The coincidence logic circuit is shown in Fig. 4. Signals from the two photomultiplier tubes (RCA 8575) on each plastic scintillation detector are added together with an EG&G model AN102/N fast mixer. These signals are then converted to standard fast-logic pulses ( $\sim 40$ -ns-wide) by an EG&G model T121/N quad discriminator. The coincidence logic consists of (1) a prompt channel that records both the coincidences from the induced fissions and the accidental (chance) coincidences produced by the random source and the sample itself, and (2) a delayed-coincidence channel that records only the accidental coincidences. The difference between the count rates in these two channels is the induced-coincidence rate, which is proportional to the  $^{235}\text{U}$  content of the sample. The delay for the accidental-coincidence channel is produced by a delay line made of 30.5 m of 50- $\Omega$  coaxial cable (RG223). The coincidences are determined by an EG&G C102B/N dual-overlap coincidence unit. The coincidence-

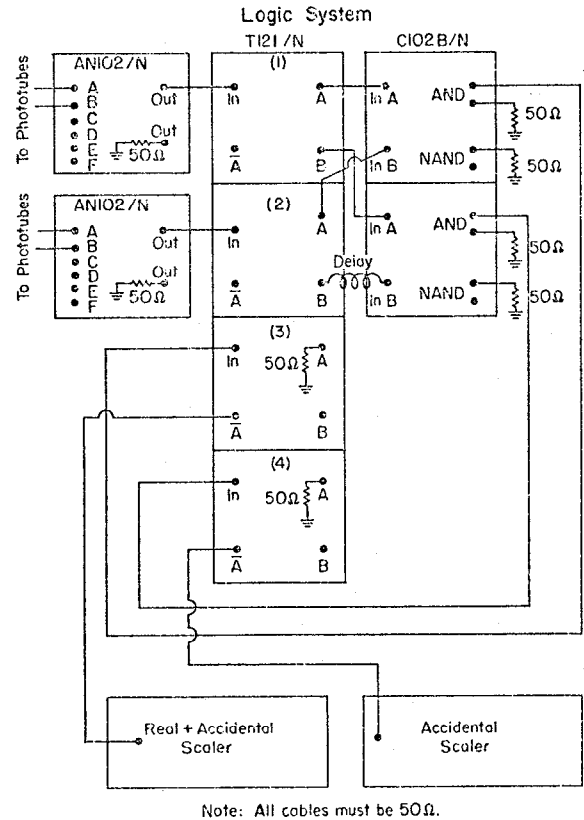


Fig. 4. Coincidence logic system for the Random Driver.

resolving time is determined by the output pulse widths of the discriminators. The outputs of the coincidence units are shaped by the remaining two discriminators in the T121/N module for proper acceptance by the Tencel model TC 546P scalers.

The calculations for the assay are done automatically by a Hewlett-Packard 9100B programmable calculator that is interfaced to the scalers. The sequence for the assay of a sample is

1. the sample is weighed and its weight is entered into the calculator,
2. the induced response is determined in a 200-s count,
3. the  $^{252}\text{Cf}$  source is then placed into the counter at the side of the sample,
4. the response of the  $^{252}\text{Cf}$  source is determined in a second 200-s count, and

5. the assay is automatically calculated; the value is presented on the display, and it is printed on a paper tape for a permanent record. The equations used for the calculations are presented in Appendix B.

#### IV. CALIBRATION

Four standards, which simulate the Rover samples, were used to calibrate the instrument. These standards, which contain 500, 1000, 2000, and 4000 g of enriched uranium in "hospital cans," span the mass range of the samples to be assayed. They are composed of a mixture of 93% enriched  $U_3O_8$  powder and graphite flour, filled to within  $\sim 25$  mm of the top of the containers.

The calibration curve that was generated using these standards, and using two neutron sources ( $\sim 5 \times 10^6$  n/s each) spaced 250 mm apart, is shown in Fig. 5. Background, fission chamber, and  $^{252}Cf$  source-addition corrections are included in this calibration curve (see Appendix B).

The nonlinearity in the calibration curve indicates that the assay, unfortunately, is a function of not only the amount of uranium in the sample, but it is also a function of the density of the uranium

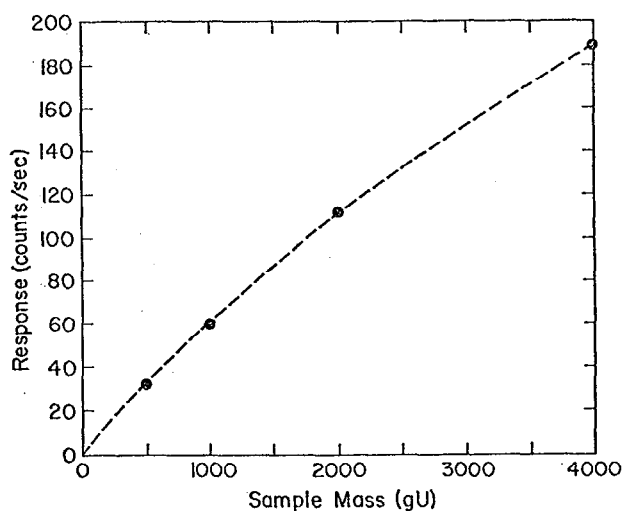


Fig. 5. Calibration curve for 10-liter containers filled with a mixture of 93% enriched  $U_3O_8$  and graphite flour.

in the sample. The response per gram of uranium in the 4000-g standard, for example, is  $\sim 80\%$  of the response per gram of the 1000-g standard. This problem can be overcome by filling the samples to be assayed to the same fill height as that of the standards; i. e., the samples will all have the same volume. If a sample is reasonably homogeneous, then its mass and its density are proportional, with the proportionality constant being the fixed volume. The calibration curve is then unique and the nonlinearity causes no problem. Accurate assays thus require constant fill heights.

The calibration curve of Fig. 5 is fitted, by the least-squares technique, to an equation of the form<sup>9</sup>

$$R = A(1 - e^{BU}), \quad (1)$$

where R is the corrected coincidence rate (see Appendix B) of the sample containing U grams of 93% enriched uranium. A and B are calibration constants determined by the least-squares fit. Inversion of this equation to

$$U = \frac{1}{B} \ln \left( 1 - \frac{R}{A} \right) \quad (2)$$

gives the assay in grams of uranium.

#### V. ASSAY PRECISION

The precision of the assay can be obtained by propagating the uncertainties associated with the count rates through all of the equations. It is easier, however, to obtain estimates of the precision by making repeated measurements on the standards. Table I gives the results of such measurements; these values can be used as estimates of the precision of measurements of actual samples.

Since the precision of the assay of samples with a uranium content of less than 500 g is governed almost totally by the background rates, an estimated standard deviation of  $\sim 20$  g must be



TABLE I  
ESTIMATED STANDARD DEVIATION  
OF ROVER SAMPLES

Sample	Standard Deviation
500 g	20 g
1000 g	60 g
2000 g	110 g
4000 g	290 g

applied to all such samples. The practical minimum sensitivity of the instrument for assay of samples in 10-liter "hospital cans," using a 200-s assay time, is therefore  $\sim 50$  g of uranium. A lower sensitivity can be obtained, if necessary, by increasing the assay time. An 800-s assay, for example, will give a sensitivity of  $\sim 25$  g of uranium.

Because the actual content of a sample is generally not known accurately, it is difficult to make quantitative evaluations of the accuracy of the Random Driver. However, comparisons have been made between assays made with this instrument and assays made with the delayed-neutron technique. The agreement between the two techniques, as shown in Fig. 6, is quite good.

Further evaluations of the performance of the instrument will be made as more information is obtained about sample content from the uranium recovery operations. The major problem encountered in comparing the assay values with the recovery values is that recovery samples are generally

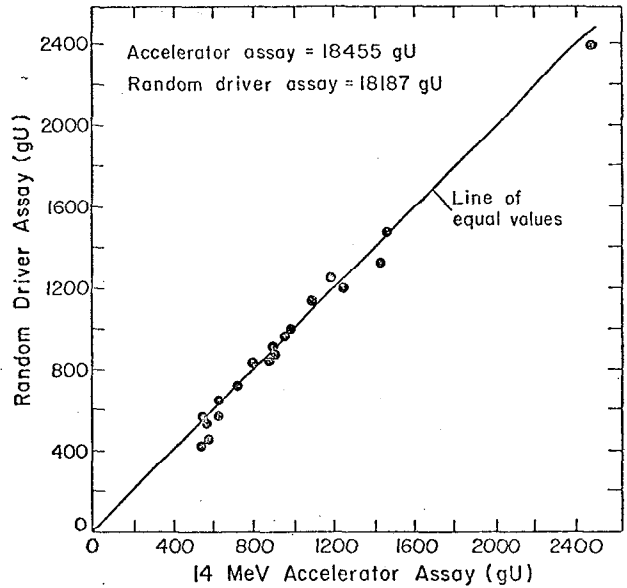


Fig. 6. Comparison of assays of the Random Driver with assays made by the delayed-neutron technique.

"batched" together into large groups for processing; thus, the individual sample identity is lost and individual comparisons cannot be made.

## VI. CRITICALITY PROCEDURES

The criticality procedures followed in assaying the Rover scrap material are given in Appendix C. These procedures apply only to this Random Driver and this particular application. New procedures must be developed for different assay situations.

## APPENDIX A

### VERTICAL RESPONSE OF SYSTEM

Early Random Drivers<sup>10</sup> used a source that moved up and down to help flatten the vertical response of the sample. The response, however, was still higher at the center of the sample than it was at the ends. The response at the center of the sample can be reduced, and at the same time the

response at the ends increased, if two sources are used. These sources are spaced such that the response at the center and at the ends of the sample are the same. This technique works well for source spacings of up to 250 mm. When spacings are larger than this, the response in the center of the

sample is lower than at the ends. Tall samples that require large source spacings may require an additional source ("shim source") at the center of the counter to boost the response of the center of the sample. Figure A-1 shows the vertical response of the Random Driver for three source configurations: (1) a source fixed in the center of the counter, (2) a

source moving up and down at a uniform speed with amplitude of  $\pm 125$  mm, and (3) two sources with centers spaced 250 mm apart. These measurements were made by placing the center of a 70-mm-thick layer of material (250-mm-o. d.) at various distances from the center of the counter. Each data point was measured to a precision ( $1\sigma$ ) of  $\sim 1\%$ .

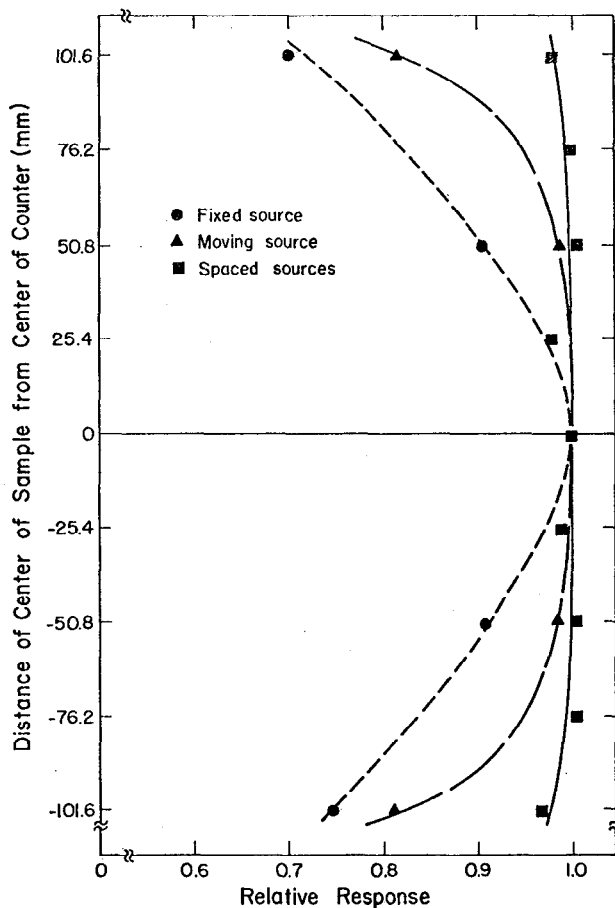


Fig. A-1. Vertical response profiles of the Random Driver for different source conditions.

## APPENDIX B

### CALCULATIONS

#### A. Background Correction

The background coincidence rate  $\dot{B}$  is determined from the weight of the sample using Fig. 2.

#### B. Induced-Coincidence Rate

The induced-coincidence rate when the sample is in the counter is given by

$$\dot{I} = (\dot{R}_1 - \dot{R}'_1 - \dot{B}),$$

where  $\dot{R}_1$  is the count rate of the prompt-coincidence scaler, and  $\dot{R}'_1$  is the count rate of the delayed-coincidence scaler.

#### C. Fission Chamber Correction

The fission chamber response when there is no sample in the counter is given by  $\dot{A}_O$  and the response with the sample is  $\dot{A}_S$ . The fission chamber correction factor  $G$  is then

$$G = \frac{\dot{A}_O}{\dot{A}_S}.$$

#### D. Attenuation Correction

The  $^{252}\text{Cf}$  source-addition correction is made in the following manner.

Let  $\dot{R}_2$  and  $\dot{R}'_2$  be the coincidence rates of the prompt and the delayed scalers, respectively, when the  $^{252}\text{Cf}$  source is in the counter without the sample.

The coincidence rate of the  $^{252}\text{Cf}$  source  $\dot{C}_O$  is then

$$\dot{C}_O = (\dot{R}_2 - \dot{R}'_2 - \dot{B}).$$

Let  $\dot{R}_3$  and  $\dot{R}'_3$  be the coincidence rate of the prompt and delayed scalers, respectively, when the  $^{252}\text{Cf}$  source is in the counter with the sample.

The coincidence rate of the  $^{252}\text{Cf}$  source in the presence of the sample  $\dot{C}_S$  is then

$$\dot{C}_S = (\dot{R}_3 - \dot{R}'_3 - \dot{B} - \dot{I}).$$

The  $^{252}\text{Cf}$  source-addition correction factor  $F$  is then given by

$$F = \frac{\dot{C}_O}{\dot{C}_S}.$$

#### E. Corrected Sample Response

The sample coincidence rate with background correction, fission chamber correction, and attenuation correction is then

$$R = G \cdot F \cdot \dot{I}.$$

The final assay is then determined from Eq. (2).

## APPENDIX C

### CRITICALITY PROCEDURES\*

The following procedure shall apply for the assay of uranium ( $\leq 93.5\%$   $^{235}\text{U}$ ) in the Random Driver.

Any container having gross weight less than 30 lb ( $\sim 13.6$  kg) and volume less than 20 liters may be placed in the Random Driver for assay, providing the air gap between the sides of the container and the Random Driver is at least 2 cm.

These limits are based on a series of DTF71 calculations (using Hansen-Roach 16-group cross sections) in which the Random Driver was modeled as a series of spherical shells (Boral, Fe,  $\text{H}_2\text{O}$ ) surrounding a spherical volume containing U ( $93.5\%$   $^{235}\text{U}$ ) +  $\text{H}_2$ ). A 2-cm gap was maintained between the U +  $\text{H}_2\text{O}$  mixture and the innermost shell of the reflector. The calculations indicate the minimum total mass of a critical mixture of U ( $93.5\%$   $^{235}\text{U}$ ) and water in this system to be  $\sim 14.6$  kg.

The 30-lb ( $\sim 13.6$ -kg) limit is considered conservative for the following reasons.

1. Calculations by Stratton<sup>11</sup> for unreflected and  $\text{H}_2\text{O}$  fully reflected U ( $93.5\%$ ) +  $\text{H}_2\text{O}$  + C mixtures indicate that U +  $\text{H}_2\text{O}$  (no C) results in the lowest critical mass of  $^{235}\text{U}$  and the lowest gross weight for any given volume in the range that will fit into the Random Driver.

2. The Random Driver has hydrogenous reflectors only on two sides; the front and rear are reflected only by 25.4 mm of steel, and the top is completely unreflected. Thus the spherical geometry used in the calculations should be quite conservative.

3. The gross weight limit set above is  $\sim 7\%$  below the minimum total mass indicated by the calculations and includes the weight of the empty container.

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\*This appendix written by D. B. Smith, A-1 (LASL).