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AUTHOR(S): T. L. Atwell, J. E. Foley, and L. V. East

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los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

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NDA OF HTGR FUEL USING THE RANDOM DRIVER*

T. L. Atwell, J. E. Foley, and L. V. East

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

ABSTRACT

This paper discusses the use of an active neutron source interrogation system known as Random Driver for the nondestructive assay of uranium in HTGR (in-process) fuel particles. The system employs a random source of fast neutrons consisting of four AmLi (α, n) sources to induce fissions in the ^{235}U present. Fission events are detected by coincidence counting of prompt-fission neutrons using a pair of fast plastic scintillation detectors. A PDP-11/05 minicomputer is used for data acquisition and reduction. Details of the assay system and preliminary results on the assay of HTGR fuel particles are presented.

INTRODUCTION

As part of a joint program between General Atomic Co. and the USAEC for development and application of nondestructive assay techniques for HTGR materials safeguards analysis, LASL Group A-1 has developed an automated instrument referred to as Random Driver Mod-III for assaying containers of HTGR in-process fuel particles for ^{235}U content. The Mod-III is an upgraded version of the original Random Driver,[1-5] which was developed for assaying high concentrations of highly enriched uranium. Illustrations of the new instrument are given in Figs. 1 and 2. Shown in Fig. 3 is the data acquisition system which includes a minicomputer and teletype for data analysis and assay printout. The device is scheduled for installation at GAC's Sorrento Valley plant in September 1974 for field evaluation.

What is a Random Driver? It is an active neutron interrogation system which employs an ^{241}AmLi (α, n)-reaction neutron source to induce fissions in the fissile material within a sample; hence, it "drives" the sample. Since the neutrons emanating from the AmLi source ($\bar{E}_n \approx 0.65$ MeV) have energies below the fission threshold of ^{232}Th, the technique is

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

insensitive to the ^{232}Th and ^{238}U content in a sample. Yet the source energy is high enough to achieve the penetrability required for assaying samples of high fissile mass, i. e., the problem of sample self-shielding is minimized. The "randomness" of the driver pertains to the nature of the source; i. e., alpha-decay in ^{241}Am is purely random in time; and since one and only one neutron is emitted per (α, n) reaction with Li, the neutrons produced are also random and therefore not correlated with one another. This characteristic enables a pair of fast neutron scintillation detectors to distinguish noncorrelated source neutrons from fission-produced neutrons (which have high probabilities of being emitted in pairs and triplets) by demanding that two events be detected within a short coincidence interval, typically 30 nanoseconds.

Although radioactive sources other than AmLi exist which produce a single neutron per decay, and these include (γ, n) as well as (α, n) sources, AmLi is the only high yield source for which the energies of the gamma rays accompanying the decay and neutron-producing reaction are small ($\lesssim$ 160 keV). This is important since fast neutron detectors, even when shielded with several centimeters of lead, are nevertheless still slightly sensitive to very energetic gamma rays and, therefore, have a finite probability of recognizing source neutrons and coincident source gamma rays as coincident events. The latter leads to an undesirable, real-coincidence background which, in turn, decreases the measurement precision and decreases the sensitivity of the instrument.

BRIEF DESCRIPTION OF HTGR-COATED FUEL PARTICLES

Production containers for in-process fuel range from 2 to 4 liters in volume and contain from 100 to 400 g of uranium (93% ^{235}U). The fuel is in the form of carbon- and silicon carbide-coated particles of UC_2 and ThC_2. The Th/U blending ratios range from 3.6 to 4.3 by weight, and the coated particle diameters range from 240-560 microns in size. Figure 4 is a sketch of the three types of containers commonly used for the various stages of the coated-particle product. The type CU-4 particle is a ThC_2/UC_2 fissile bead (100-300 μ) coated with a 50 μ buffer coating and a 20 μ pyrolytic carbon coating. The CU-5 particle contains an additional 20 μ SiC coat, onto which another 40 μ of carbon is added to form the TRISO-coated CU-6 type particle. After cleaning, the latter are designated CU-7 particles.

From information given above it is clear that if a high degree of NDA measurement accuracy is to be achieved the response of any such instrument must be either insensitive to or able to correct for variations in: a) heavy-element concentrations (Th and U), b) light element matrix (C and Si), c) sample height and diameter, and d) container material (Al or CH_2).

THE UPGRADED MOD-III RANDOM DRIVER

The Mod-III Random Driver differs from the original in several aspects. First, four stationary sources ($\sim 5.5 \times 10^5$ n/s each), mounted in

cylindrical tungsten gamma shields, are arranged in a nickel-reflected irradiation cavity to achieve a spatially "flat" fast neutron flux distribution. The steel-backed nickel reflector provides very good source-to-sample coupling and increases the effective source strength by a factor of two over a 100%-steel-reflected cavity. A vertical, center-to-center source spacing of 235 mm was found tobe optimum for the typical range of sample fill heights (75 to 200 mm). A plot of the vertical response versus sample fill height is given in Fig. 5, along with a sketch of the relative locations of the sources and sample rotator. This plot was generated by integrating the instrument's point-wise response over the sample fill height. As can be seen, the change in overall response is less than 0.6% from 75 to 200 mm.

The second area of concentration in the adaptation of the Random Driver to the NDA of HTGR fuel particles was to find a method by which the sample's induced response could be corrected for perturbations on neutron flux due to the presence of light-element matrix and container materials. To monitor the neutron flux, four 12.7-mm-diam by 178-mm-active length ^{3}He proportional counters (10-atm fill pressure) were installed in the corners of the sample chamber.

After a series of experiments it was determined that for a given sample the change in response (net coincidence count rate) due to a change in either the container material or exterior bagging was directly proportional to a correction factor K given by the relation

$$K = \frac{1+F}{2} \quad , \tag{1}$$

where F is the flux monitor ratio defined as

$$F = \frac{^{3}\text{He Count Rate With Sample}}{^{3}\text{He Count Rate With Empty Chamber}} \tag{2}$$

Presented in Table I are the results of a series of measurements on variations in response due to the absence, presence, or addition of polyethylene around an HTGR sample. The mass of an empty polyethylene bottle is ~ 58 g and has the overall effect of increasing the induced signal by 10-15%. The relatively good agreement between the "flux-corrected" responses within the sets of data for each sample confirms the validity of Eq. 1. To be discussed later, an additional correction factor for ^{235}U mass is needed to completely linearize the response.

Thirdly, the Mod-III version of Random Driver was made less sensitive to variations in heavy-element matrix by reducing its sensitivity to prompt fission gamma rays which are emitted in coincidence with a $\bar{\nu}$ of about 6. This compares to a fission neutron $\bar{\nu}$ of about 2.5 for ^{235}U. Although coincidence detection of prompt gamma rays would increase the response and therefore improve the counting precision, it would tend to worsen the accuracy of a measurement in that one then becomes sensitive to gamma ray attenuation within the sample. For example, if samples A and B have

the same ^{235}U mass but the Th/U ratios are 3.5 and 4.3, respectively, sample A will assay higher than B because the gamma attenuation due to the thorium will be lower in A than B. Discrimination against coincident gamma rays in the Mod-III Random Driver is accomplished by time discrimination, the circuit for which is given in Fig. 6. Only events which are separated by a minimum interval of 5 ns up to a maximum interval of 30 ns are sensed. Since coincident gamma rays have very short flight times (< 2 ns) from sample to scintillator, the technique works quite well. Shown in Fig. 7 are time-dependent response curves for a ^{60}Co source, a random background source, and an HTGR sample with and without time discrimination. The amount of time discrimination is adjusted by varying the length of the "short delay".

A PDP-11/05 minicomputer with 8K of memory is used for automated scaler readout and data reduction. In addition, a library of subroutines are available for data fitting routines and statistical analysis. In normal operation the operator keys in the sample identification, the tag mass, the desired assay time, and other pertinent information, and then starts the assay. At the conclusion of the assay, the computer calculates the assay mass, the associated uncertainty, and other parameters asked for, and returns control to the operator. Hard copy and papertape output is generated by the teletype.

CORRECTION FACTORS

The following formulation provides a basis for the application of first order correction on the Random Driver response. Let us define

Y = perturbation in neutron flux due to sample's presence
F = ^{3}He flux monitor ratio
R = random driver response; M = mass of ^{235}U in sample
A, B, C, D = constants

Now assume that the response is directly proportional to the perturbed neutron flux such that

$$R = AM(1 + Y) \quad , \tag{3}$$

and that the ^{3}He flux monitor responds to the change in neutron flux and to the induced fission rate as

$$F = 1 + CY + DR \quad . \tag{4}$$

The flux perturbation can now be expressed in terms of F and R as

$$Y = \frac{F - 1 - DR}{C} \quad . \tag{5}$$

Upon substitution of Eq. 5 into Eq. 3 and rearranging terms,

$$R = \frac{AM[(C + F - 1)/C]}{1 + (AD/C)M} \tag{6}$$

The term in brackets in the numerator is the flux monitor correction factor K defined by Eq. 1. This factor was determined experimentally, as discussed earlier. The constant C has a nominal value of 2. Rewriting Eq. 6 in terms of K and substituting the constant B for AD/C,

$$R = \frac{AKM}{1 + BM} \quad . \tag{7}$$

The response per gram is then given by

$$R/M = \frac{AK}{1 + BM} \tag{8}$$

The constant A is simply the "linearized response per gram".

ASSAY RESULTS ON HTGR SAMPLES†

Five containers of HTGR-coated particles and the LASL A-1 Universal Standard (HEU metal) were assayed in the Random Driver, with a compilation of the results given in Table II. The tag values for ^{235}U listed in Table II are based upon Van de Graaff assay results on six small samples from each container. For each of the samples the response obtained with the Random Driver was first corrected by the flux monitor factor defined by Eq. 1 and then plotted in Fig. 8. The data in the flux-corrected response curve were then fit to the response relation given by Eq. 7 with K = 1. The result of the fit yielded values for A and B of 0.2623 and 0.0003885, respectively.

The value of the corrected response per gram for each of the samples was then compared with A, the linearized response parameter and a percent difference were calculated, with the results tabulated in the last column of Table II. Differences ranged from +1.42% to -1.99% with uncertainties (one estimated standard deviation) ranging from ± 0.46% to ± 0.91%.

When looking at the relative response differences and the standard deviations of the measurements it is difficult to make a statement at this time as to where the error lies, especially in view of the fact that the Van de Graaff small sample assay results revealed sample-to-sample variations in grams ^{235}U per gram sample up to 4% for material taken from the same batch.

PRECISION, ACCURACY, AND STABILITY

In our present high altitude cosmic-ray background, assay precisions of less than 1% can be obtained for 300 g ^{235}U samples in counting times of 1000 s. Although the data thus far indicate that the Random Driver, after appropriate corrections, is relatively insensitive to thorium content, light-element matrix material, and container type and material, more experience is needed to make a definitive statement on its accuracy at this time.

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†Results obtained with prototype of MOD-III in which the scintillator thickness was only 51 mm.

A plot of the stability of the Mod-III random driver over a 34-hour period is presented in Fig. 9. The fact that the observed standard deviation was slightly higher than the estimated standard deviation associated with the counting statistics indicates slight drifts in stability. It is very likely that the drift is associated with a temperature effect since the room temperature varied over a span of ~ 5°C, as a function of the time of day, over a 24-hour period. Later studies will be made to determine whether or not the diurnal cosmic ray effect might also be a contributor.

For a 1000-s counting time the ^{235}U sensitivity is 12 g for a net signal equivalent to three standard deviations of the background.

REFERENCES

1. J. E. Foley, U. S. Patent #3,736,429, issued May 29, 1973.
2. J. E. Foley, "Random Source Interrogation System," Los Alamos Scientific Laboratory report LA-4883-PR (1971), p. 9.
3. J. E. Foley, "Random Driver Measurements at the LASL Uranium Recovery Facility (CMB-8)," Los Alamos Scientific Laboratory report LA-5431-PR (1973), p. 5.
4. J. E. Foley, "Application of the Random Source Interrogation System (Random Driver) at the Oak Ridge Y-12 Plant, Preliminary Results," Los Alamos Scientific Laboratory report LA-5078-MS (1972).
5. J. E. Foley, "Assay of Uranium Content of Rover Scrap With the Random Source Interrogation System," Nuclear Technology (to be published).

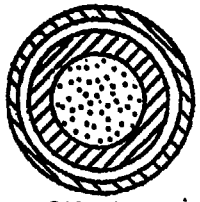
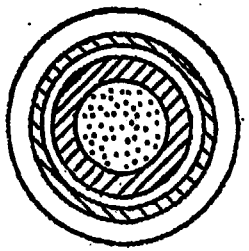
TABLE I

FLUX MONITOR CORRECTION FOR RESPONSE VARIATION DUE TO THE INFLUENCE OF CONTAINER MATERIALS AND POLYETHYLENE BAGGING ON THE INTERROGATING NEUTRON FLUX

Sample	Container	Uncorrected Response (counts/s-g)	Flux Monitor Ratio	Flux-Corr. Response (counts/s-g)
CU4B-10191	Al can	0.2338 ± 0.0015	1.0266 ± 0.0018	0.2307 ± 0.0019
	Al + 58-g bag	0.2574 ± 0.0016	1.2374 ± 0.0018	0.2301 ± 0.0019
CU5B-399	Al can	0.2566 ± 0.0045	1.1188 ± 0.0018	0.2422 ± 0.0042
	Al + 116-g bag	0.3369 ± 0.0045	1.5434 ± 0.0016	0.2424 ± 0.0032
CU7B-10244	Poly bottle	0.2929 ± 0.0036	1.3220 ± 0.0019	0.2523 ± 0.0031
	+ 58-g bag	0.3201 ± 0.0036	1.5844 ± 0.0016	0.2477 ± 0.0028
CU7B-10244	Al can	0.2584 ± 0.0036	1.0784 ± 0.0017	0.2487 ± 0.0035
	Al + 58-g bag	0.2978 ± 0.0036	1.3363 ± 0.0020	0.2549 ± 0.0031
CU7B-721	Poly bottle	0.3050 ± 0.0050	1.3587 ± 0.0020	0.2586 ± 0.0042
	Poly + 58-g bag	0.3201 ± 0.0036	1.5844 ± 0.0016	0.2477 ± 0.0028
CU7B-721	Al can	0.2697 ± 0.0050	1.1155 ± 0.0017	0.2550 ± 0.0047
	Al + 58-g bag	0.3018 ± 0.0050	1.3794 ± 0.0020	0.2537 ± 0.0042

TABLE II

RANDOM DRIVER ASSAY RESULTS ON HTGR SAMPLES

Coated Particle	Th/U Ratio	Matrix Ratio	U-235 (grams)	Corr. Response (counts/s-g)	Rel. Response Diff. (%)*
 CU-4	3.57	2.91	399.9	0.2660 ± 0.0017	+1.42 ± 0.64
	Matrix ratio: grams carbon and silicon per gram ^{235}U				
 CU-5	3.57	10.66	292.2	0.2575 ± 0.0019	-1.85 ± 0.74
	4.18	17.09	156.2	0.2571 ± 0.0022	-1.99 ± 0.86
 CU-6,7	3.57	9.47	154.3	0.2635 ± 0.0024	+0.46 ± 0.91
	4.29	16.82	110.4	0.2659 ± 0.0020	+1.35 ± 0.90
	Linearized response value: (from data fit)			0.2623 ± 0.0058	*Reference
Standard (LASL)	0	0	470.2	0.2620 ± 0.0012	-0.12 ± 0.46

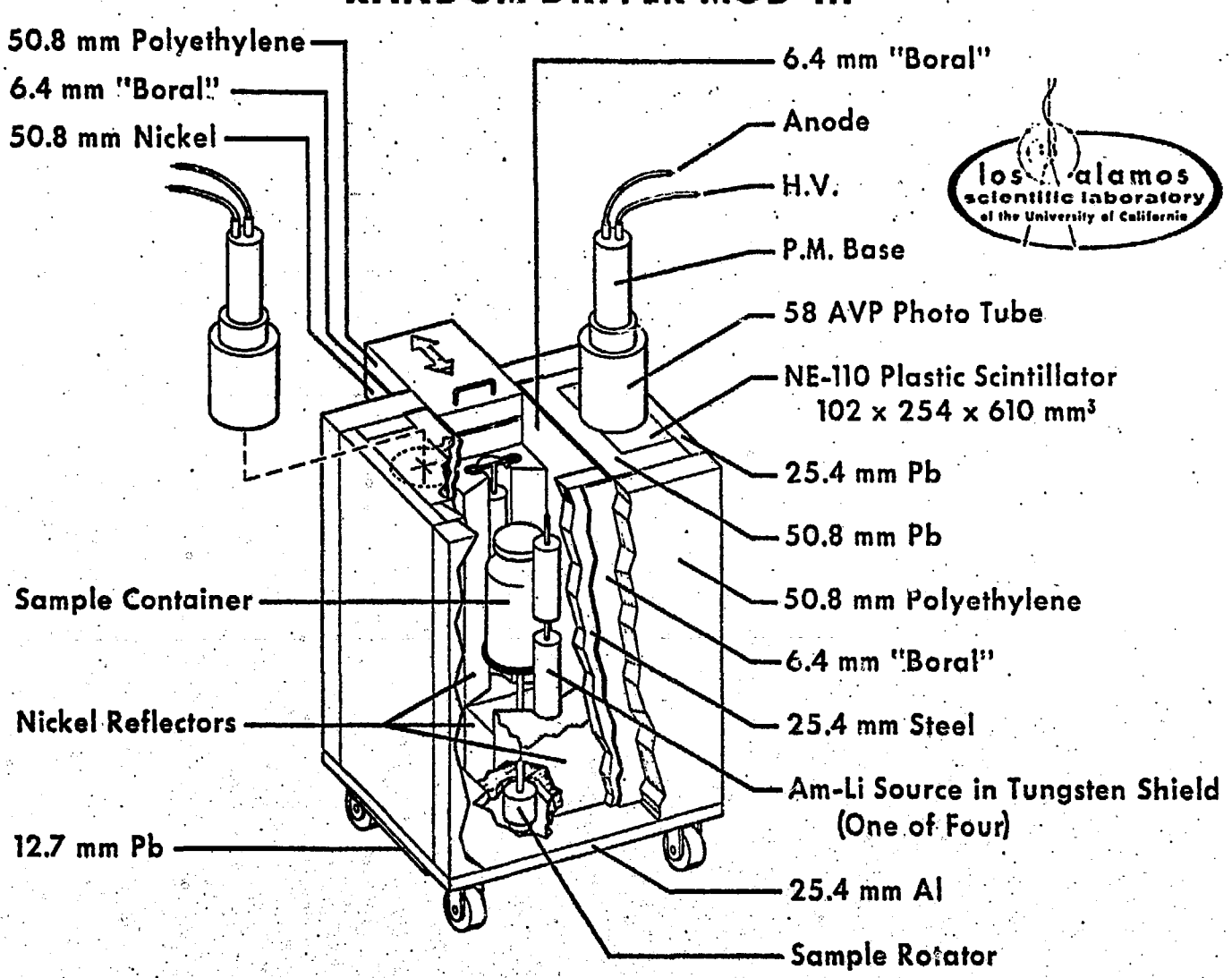


Fig. 1. Cutaway view of Mod-III Random Driver. Not shown are the four 12.7-mm-diam by 178-mm-active length ^{3}He proportional counters which are located along the four corners of the sample cavity.

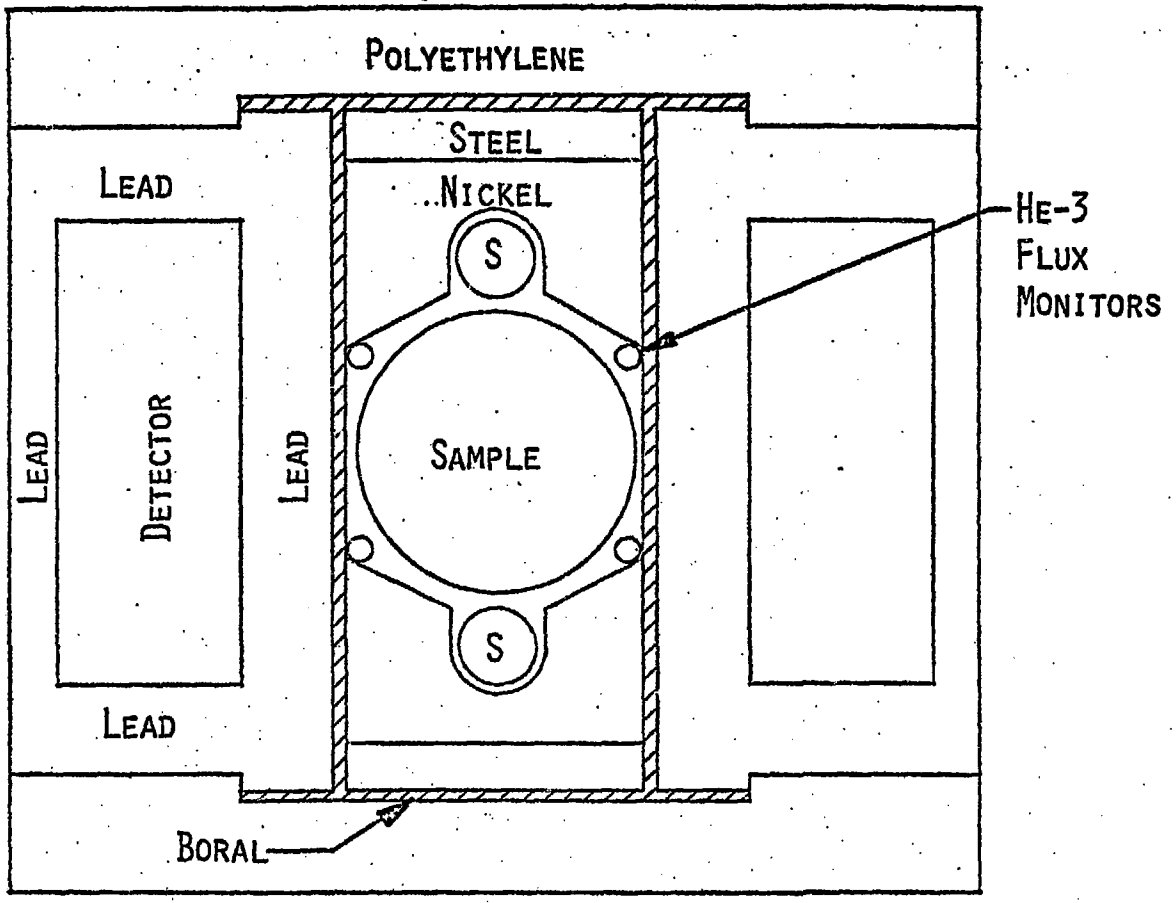


Fig. 2. Top view of Mod-III Random Driver. The instrument will accommodate sampler up to 140 mm diam by 300 mm high.

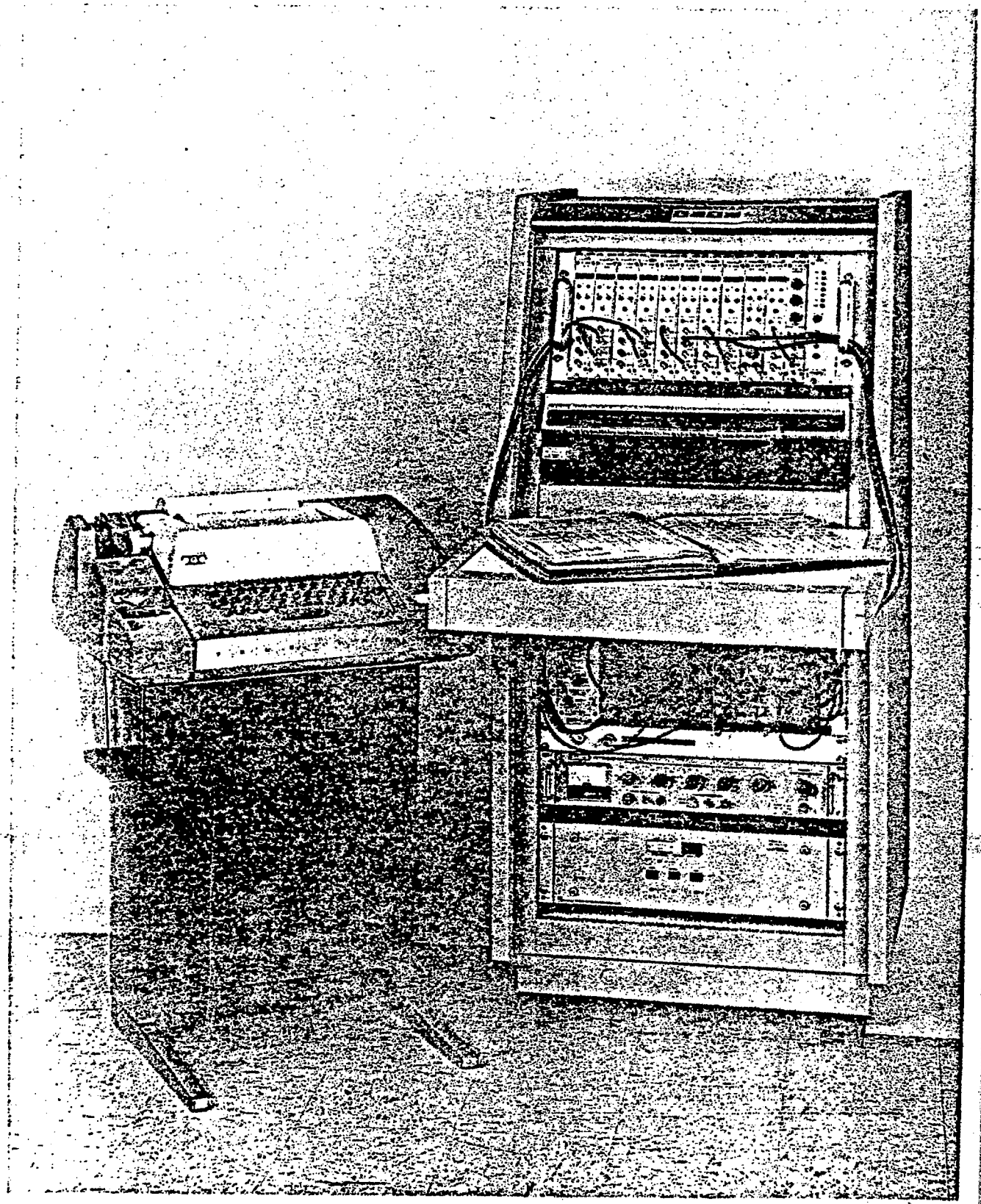


Fig. 3. Electronics console for Mod-III Random Driver, including PDP-11/05 minicomputer and teletype unit.

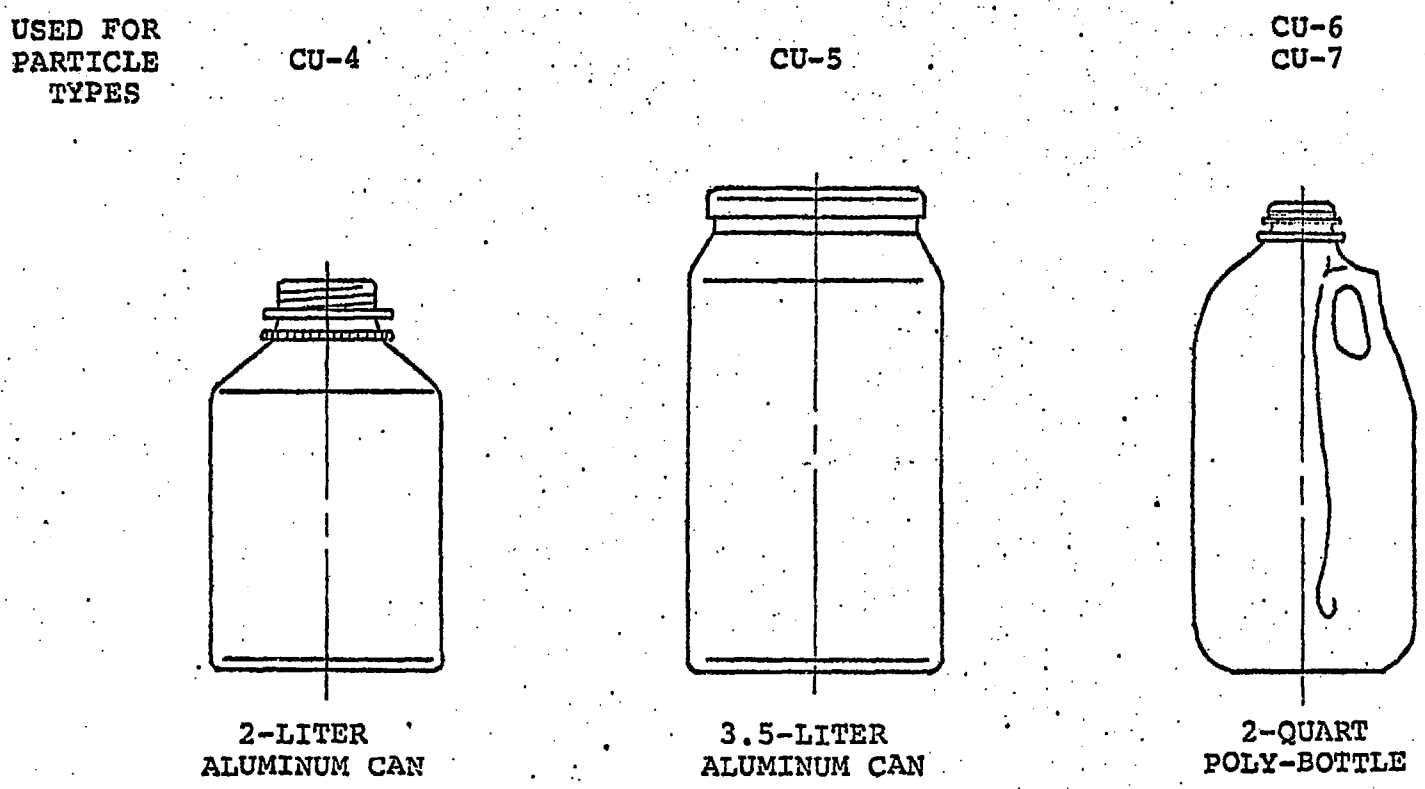


Fig. 4. Containers used for HTGR-coated fuel particles.

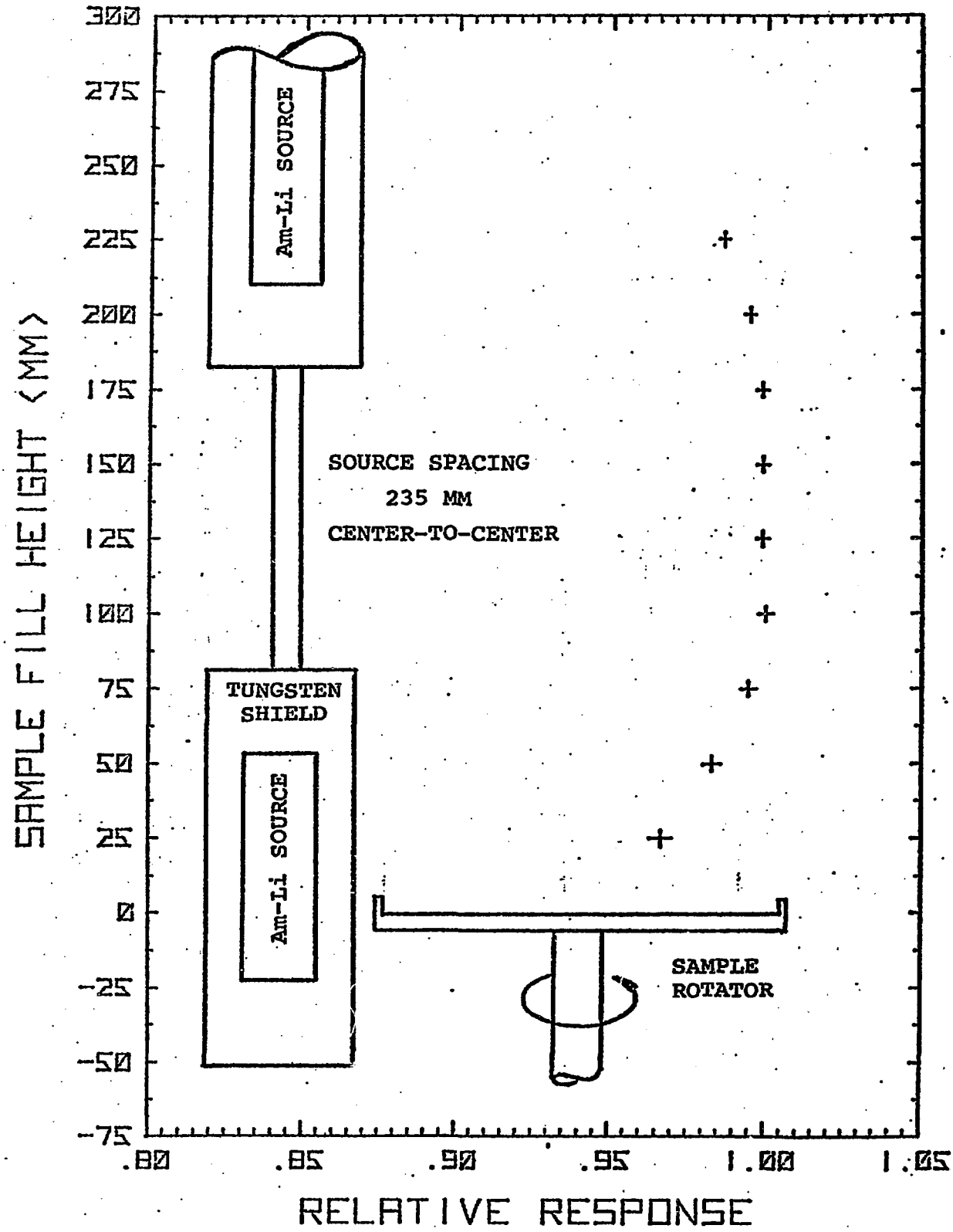


Fig. 5. Plot of relative vertical response versus sample fill height. The "flatness" in response for fill heights between 75 and 200 mm was achieved by vertically separating the sources as shown.

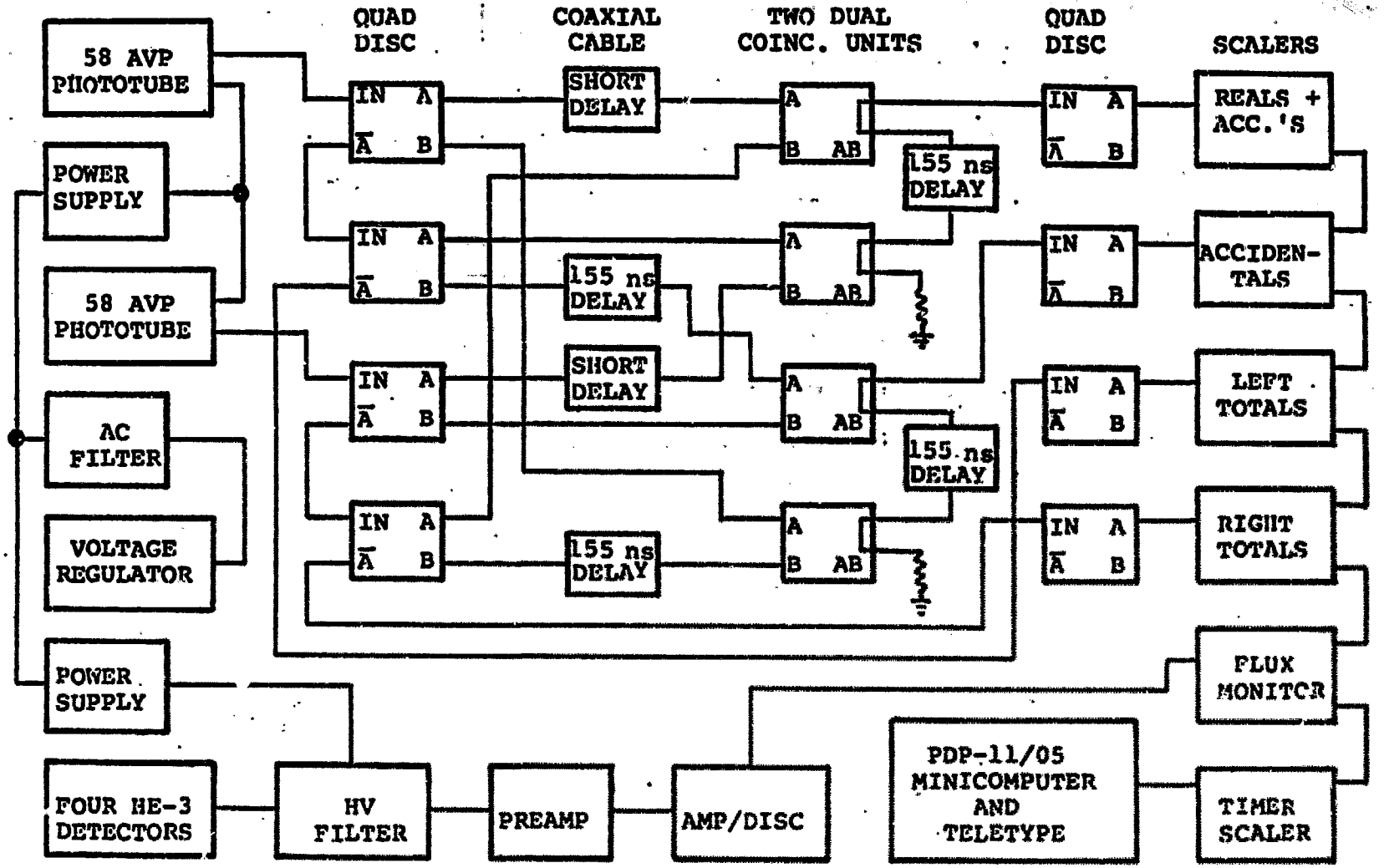


Fig. 6. Block diagram of the electronics system for the Mod-III Random Driver.

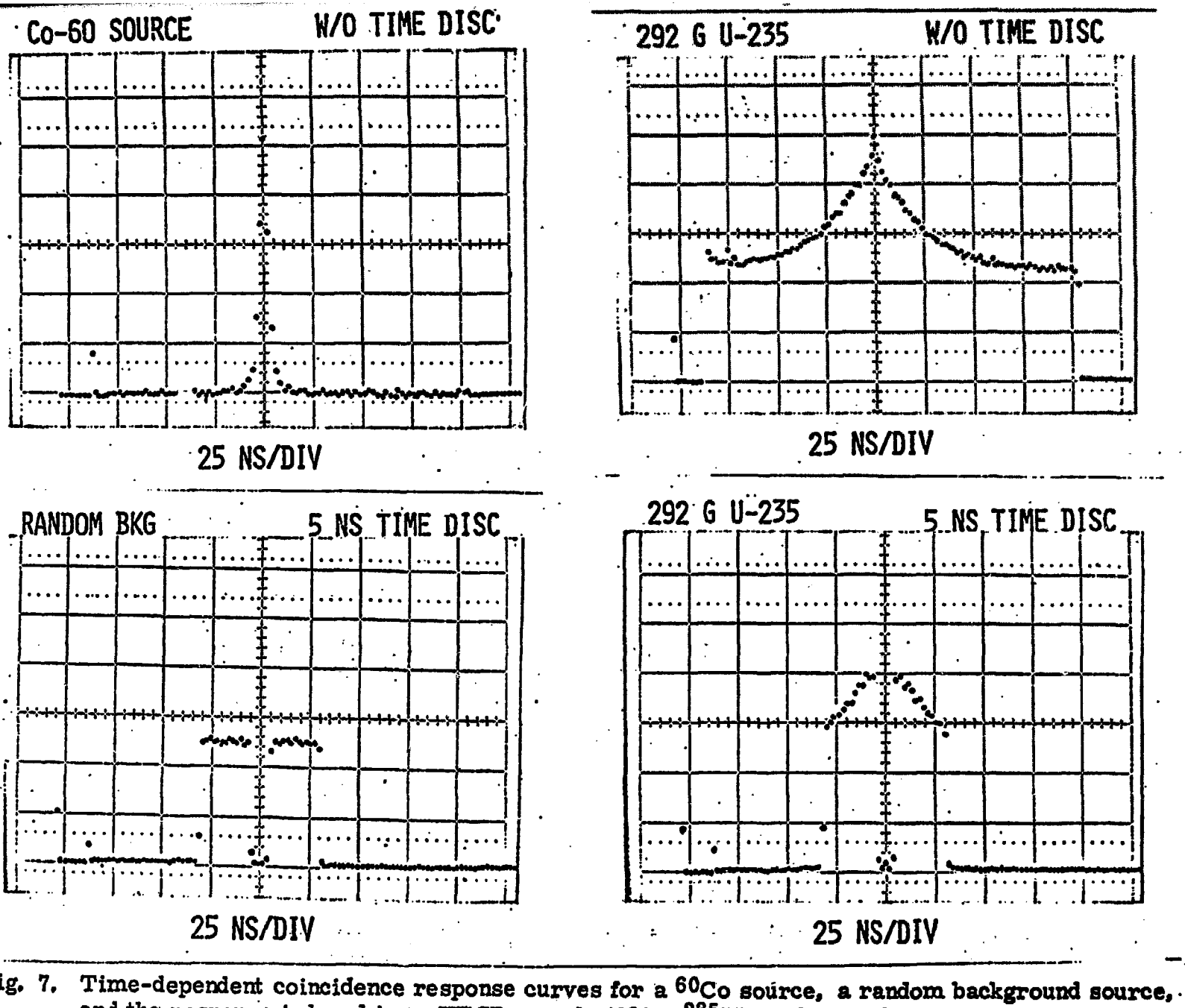


Fig. 7. Time-dependent coincidence response curves for a ^{60}Co source, a random background source, and the response induced in an HTGR sample (292 g ^{235}U) by the Random Driver with and without time discrimination.

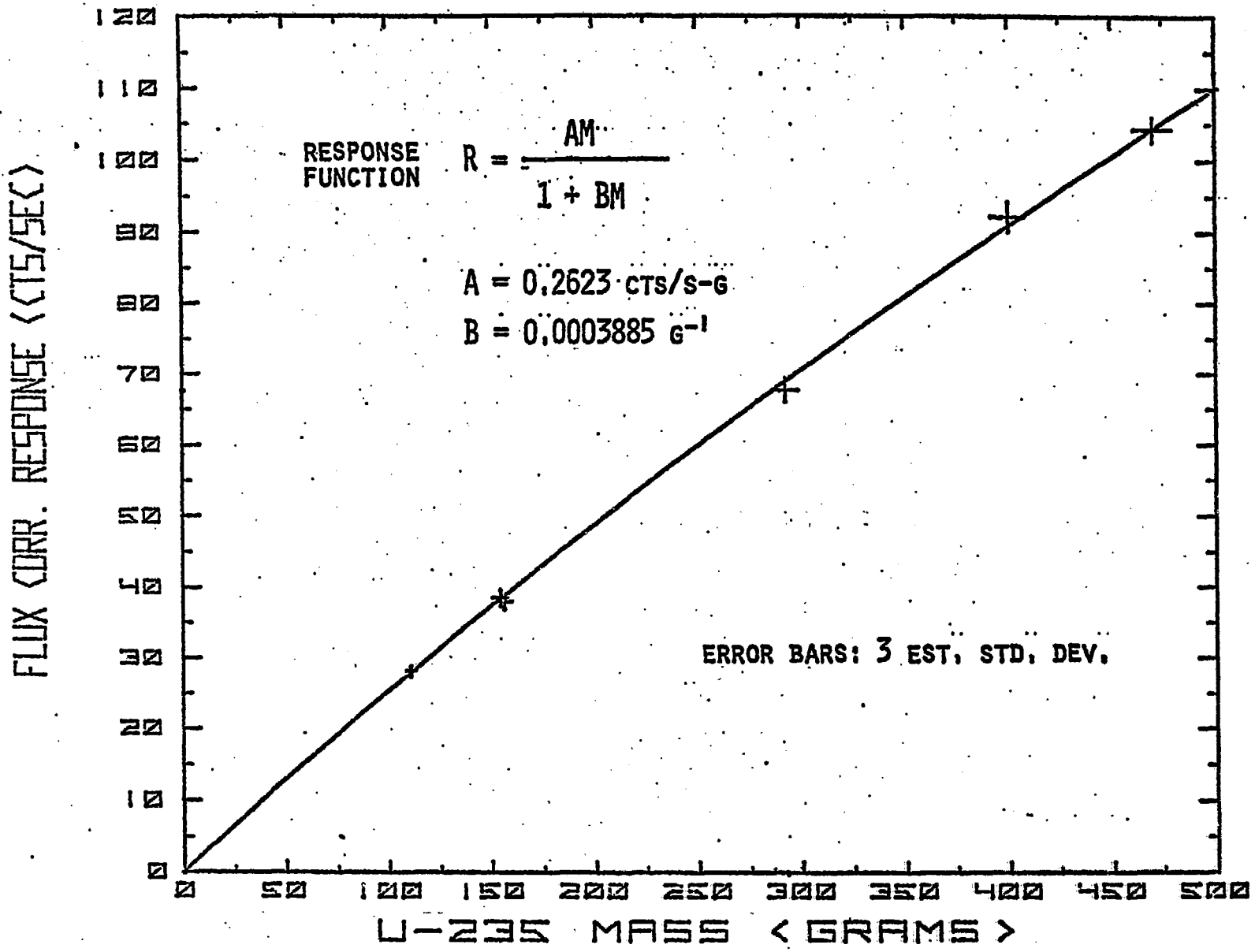


Fig. 8. Mass-dependent response curve for HTGR fuel particles after flux monitor correction.

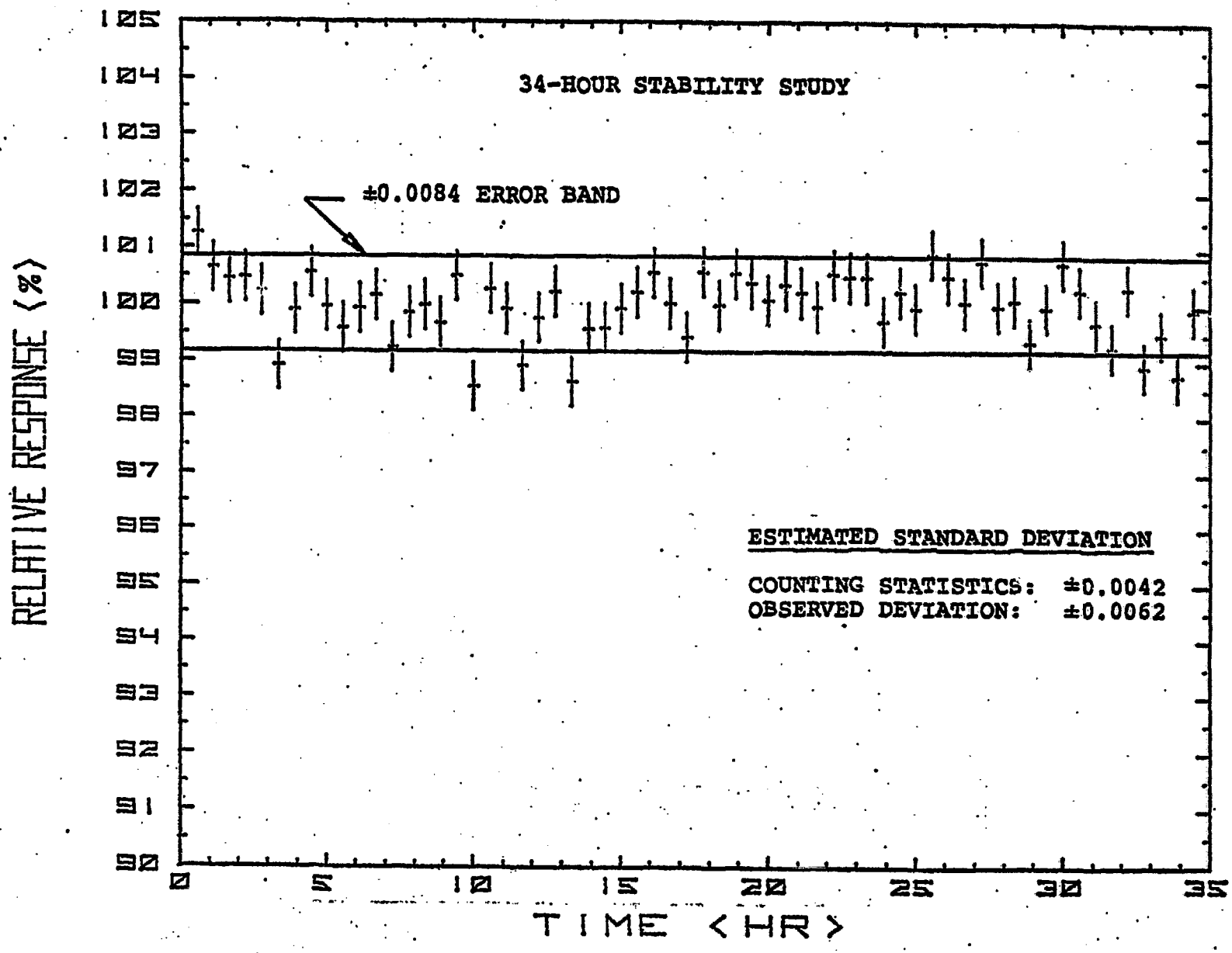


Fig. 9. Random Driver response variations over 34-hour period for counting intervals of 33⅓ minutes.