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RANDOMLY PULSED NEUTRON MEASUREMENTS FOR SAFEGUARDS INTERROGATION

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UNION CARBIDE

OAK RIDGE Y-12 PLANT
OAK RIDGE, TENNESSEE

prepared for the U.S. ATOMIC ENERGY COMMISSION
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ABSTRACT

The applicability of measurements with a randomly pulsed neutron source using ^{252}Cf for safeguards interrogation has been investigated in a series of measurements with simple configurations of uranium metal, polyethylene, and stainless steel. The linear dependence of the total-time-correlated counts per californium fission on the ^{235}U enrichment of the uranium metal has shown that this method can be used for non-destructive assay determination provided measurements with standard samples of two ^{235}U enrichments are made. Accuracy of $\pm 0.5\%$ can be obtained in $\sim$30 sec with a 3.2 μg ^{252}Cf source. Since the measurements can be made with detectors sensitive only to neutrons, this technique can be used for irradiated fuel. Other measurements of the time behavior of neutrons from fission chains in plutonium, which were also initiated by neutrons from spontaneous fission of californium, indicate that this method will also be useful for determination of the ^{239}Pu content of plutonium.

INTRODUCTION

The applicability of measurements with a randomly pulsed neutron source[1] using ^{252}Cf for safeguards interrogation has been investigated in a series of measurements with simple configurations of uranium metal, polyethylene, and stainless steel.[2] The time behavior of prompt neutrons and prompt gamma rays from fission chains, which were initiated in the sample by neutrons from the spontaneous fission of californium, depends on the amount of fissile material in the sample under interrogation. This time behavior was measured by recording the counts in a detector as a function of time after the spontaneous fission of californium. Previous use of californium in safeguards has been the random source interrogation method of Foley.[3] In his method the coincidence rate of two neutrons from fission in the sample is measured. Since the method described here requires the detection of only one particle from fission in the sample, in certain cases it may require less time than the random source interrogation method of Foley although the background may be larger. This method with detectors sensitive only to neutrons can be applied to material with high fission product gamma-ray activities such as irradiated fuel, whereas gamma-ray spectroscopy techniques,[4] which have been used for the determination of ^{235}U enrichment, cannot. Other methods[5] requiring activation of the sample use sources $\geq$ 100 μg of ^{252}Cf.

This investigation was primarily concerned with the use of this technique to determine the ^{235}U enrichment of uranium metal.

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1. J. T. Mihalczo, "Use of ^{252}Cf as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements," Y-DR-41 (1970); also, J. T. Mihalczo, Nucl. Sci. Eng. 41, 296 (1970).
2. J. T. Mihalczo, Trans. Am. Nucl. Soc. 15, 156 (1972).
3. J. E. Foley and M. M. Thorpe, LA-4705-MS, p. 9, Los Alamos Scientific Laboratory (1971).
4. A. R. Flynn, "Field Determinations of Uranium-235 Enrichment by Portable Gamma-Ray Spectrometry," K-1819, Union Carbide Corporation Oak Ridge Gaseous Diffusion Plant (1972).
5. N. P. Baumann, Savannah River Laboratory, personal communication (1972).

EXPERIMENTAL

The uranium metal pieces for these measurements were rectangular parallelepipeds of varying ^{235}U enrichment. One, depleted to ~0.3 wt % ^{235}U, and another, enriched to 93.2 wt % ^{235}U, had overall dimensions of 3 x 10 x 1 in. The metal slab of uranium with 37.5 wt % ^{235}U, which was made up of three 3-1/4 x 3-1/4 x 1 in. pieces, had overall dimensions of 3-1/4 x 9-3/4 x 1 in. The uranium density in the slabs was 18.75 g/cm^3 and the uranium contained a negligible amount of impurities. The densities of polyethylene and Type 304 stainless steel were 0.916 and 7.85 g/cm^3, respectively.

Eight configurations of source, shield, detector, and sample were investigated, some with the source and detector on the same side of the sample (reflection) and others with the source and detector on opposite sides (tramsmission). In some measurements the source and detector abutted the sample, in others the source and detector were shielded from the sample by 1.0-in.-thick stainless steel or 1.0- or 2.0-in.-thick polyethylene, which abutted the large surfaces of the sample. In all but one measurement, the shield material was adjacent to both large surfaces. In the reflection configuration 3-in.-thick lead between the source and detector reduced the counts in the detector from particles directly from the source. Typical configurations of source, shield, detector, and sample are shown in Figs. 1 and 2.

The ^{252}Cf was contained in an ionization chamber[1] which was placed in contact with the sample or shield material. Three sources were used, having activities of 4,200, 17,500, and 54,000 fissions/sec. The detector was a 2-in.-diam, 2-in.-thick proton recoil scintillator, Nuclear Enterprises Type NE 213, with or without gamma discrimination. The output signal from the californium ionization chamber was amplified by a fast current amplifier[6] and then fed to a discriminator. The discriminator output was used to start a time-to-pulse height converter. The amplified and discriminated pulse from the scintillator stopped the

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6. Designed and constructed by N. W. Hill, Oak Ridge National Laboratory.

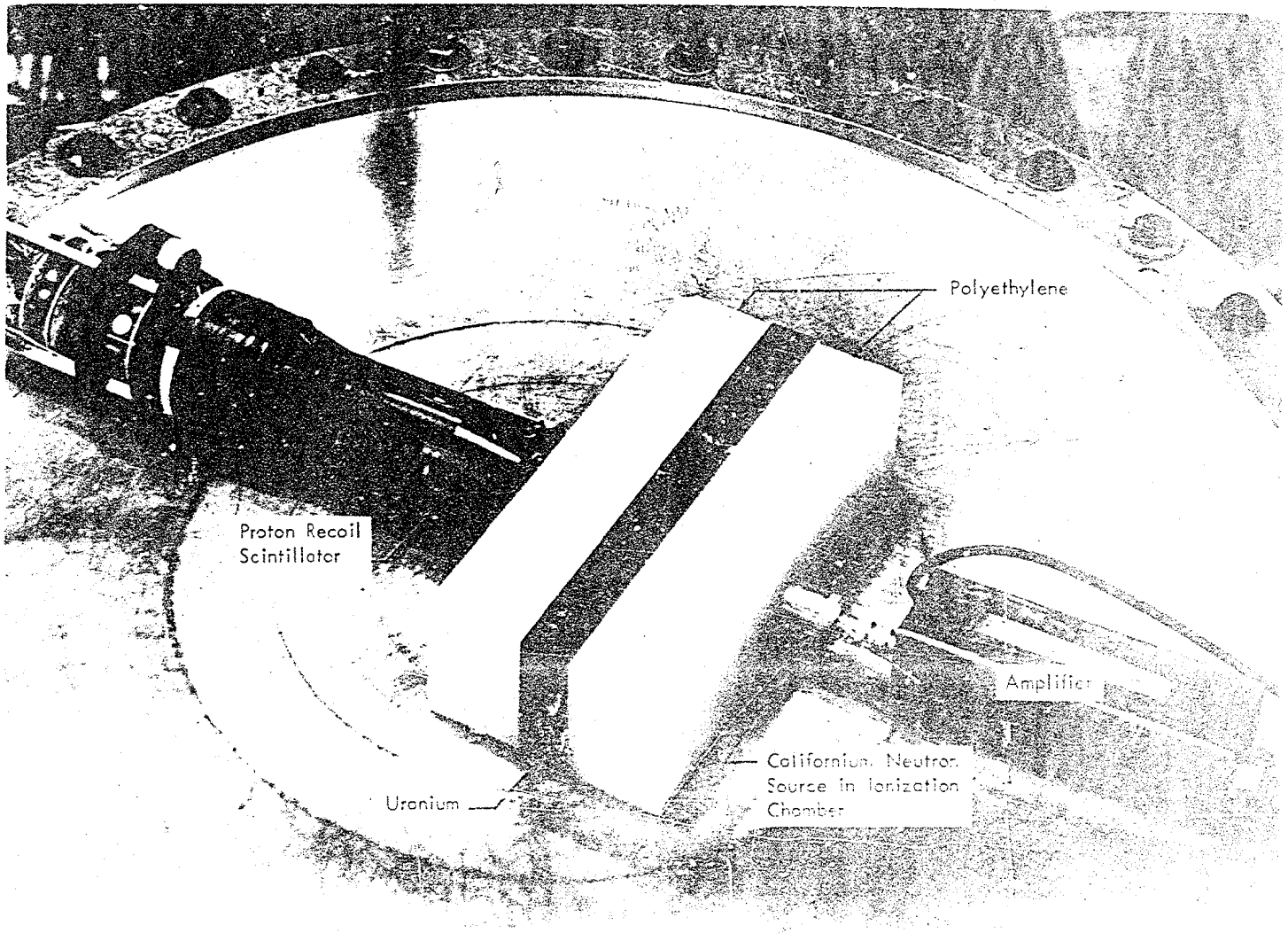

Fig. 1. Source-Detector Configuration [illegible]
Interrogation with [illegible]

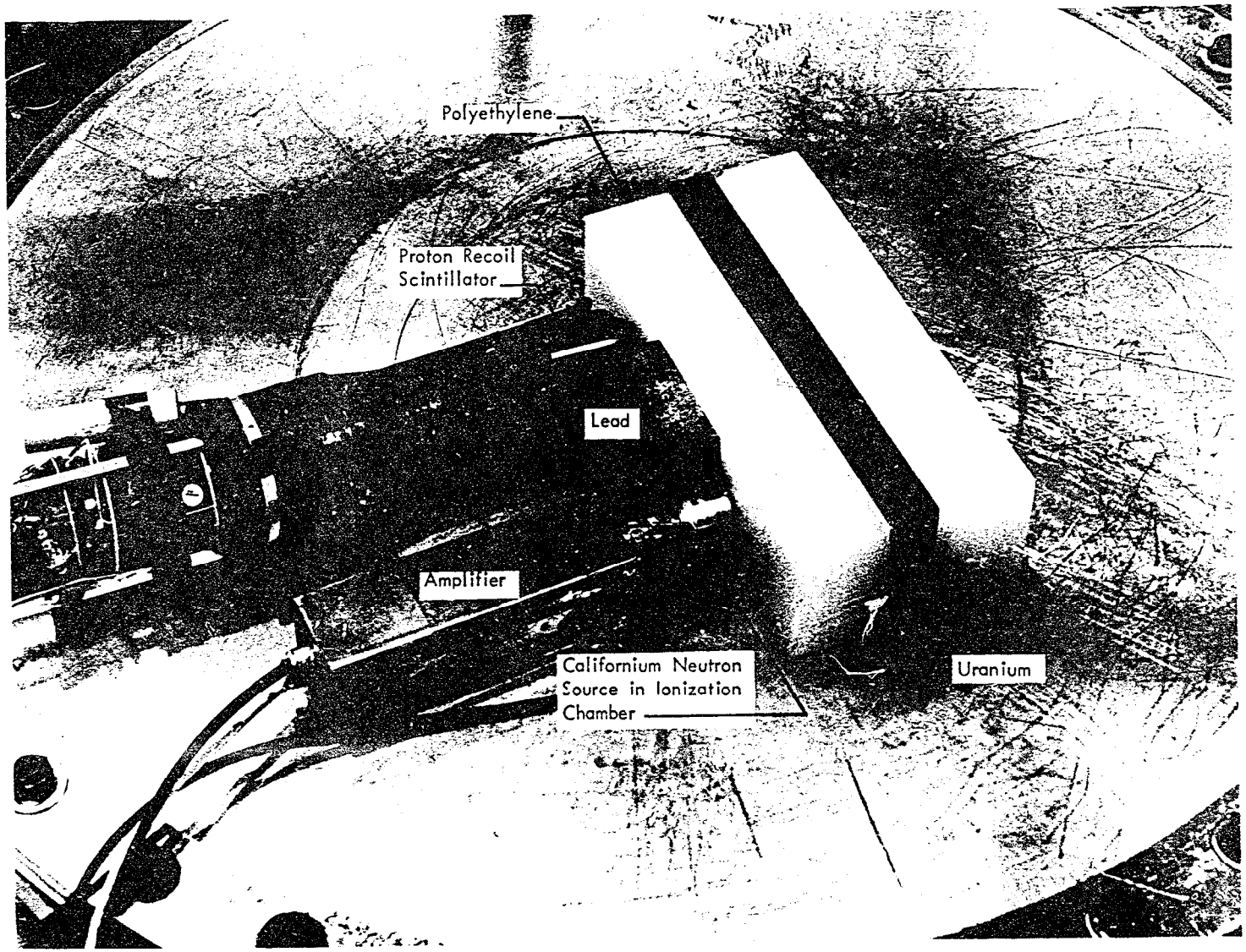

Fig. 2. Source-Detector Configuration in the Reflection Measurement for Safeguards Interrogation with 2-in.-thick Polyethylene Abutting the Uranium Metal.

converter. The converter output, which was proportional to the time between its start and stop, was stored by the pulse height analyzer. The converter-analyzer system, which processed only one count per trigger, makes up a Type III analyzer.[7] The time calibration of the channel widths was done with a pulse generator. A block diagram of the instrumentation is shown in Fig. 3. When gamma discrimination was employed, the analyzer was gated on externally by the output of a pulse shape analyzer. The shape of the input pulse determined if the scintillator detected a gamma ray or a neutron. When gamma discrimination was not used, all converter outputs were analyzed.

The results of the measurements with unshielded samples of the three enrichments and the source and the detector on opposite sides of the sample are given in Figs. 4 and 5. The counts from neutron events in the scintillator per californium fission are plotted as a function of time in Fig. 4 and those from gamma-ray interaction with the scintillator are plotted in Fig. 5. These results show a dependence of the time behavior of neutrons and gamma rays from fission in the sample on the enrichment of the sample. The total time-correlated-counts from the randomly pulsed neutron measurements are plotted as a function of ^{235}U enrichment in Fig. 6. The dependence of the neutron and of the gamma-ray count on enrichment were linear. Since both were found to be linear with enrichment, it is not essential in these measurements to employ gamma discrimination. Thus, the instrumentation may be simplified by omitting the gamma discrimination. All measurements given subsequently in this report are without gamma discrimination. Other measurements with the 37.5 wt % ^{235}U enriched sample have shown that a thin piece of aluminum placed between the sample and the detector reduces the background a factor of ~ 2 by absorption of the β particles from uranium.

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7. G. E. Hansen, H. H. Helmick, and J. D. Orndoff, "Neutron Counting Statistics in Basic Fast Critical Assemblies," Proc. of Joint Japan-United States Seminar on Reactor Noise Analysis (1968).

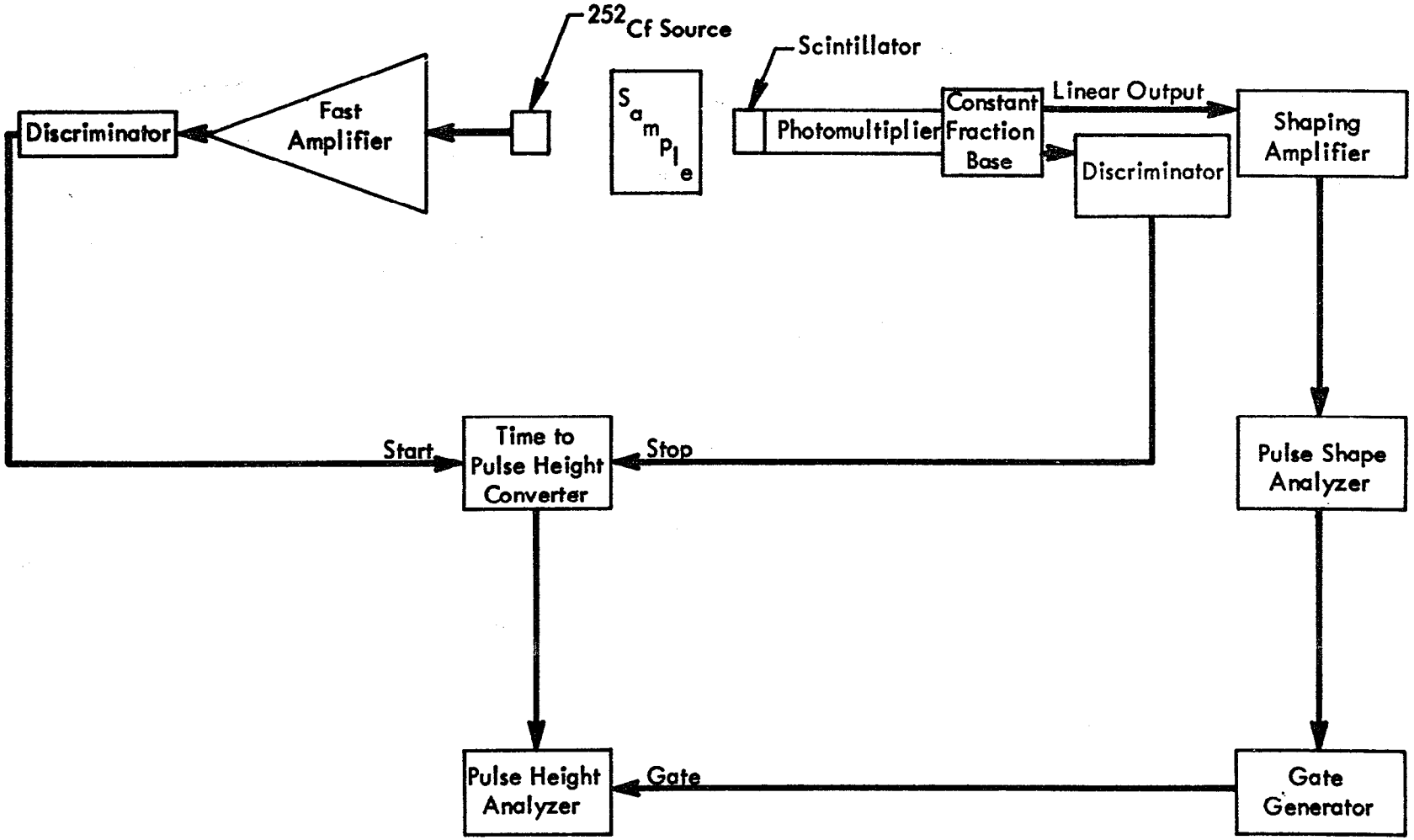

Fig. 3. Block Diagram of Instrumentation.

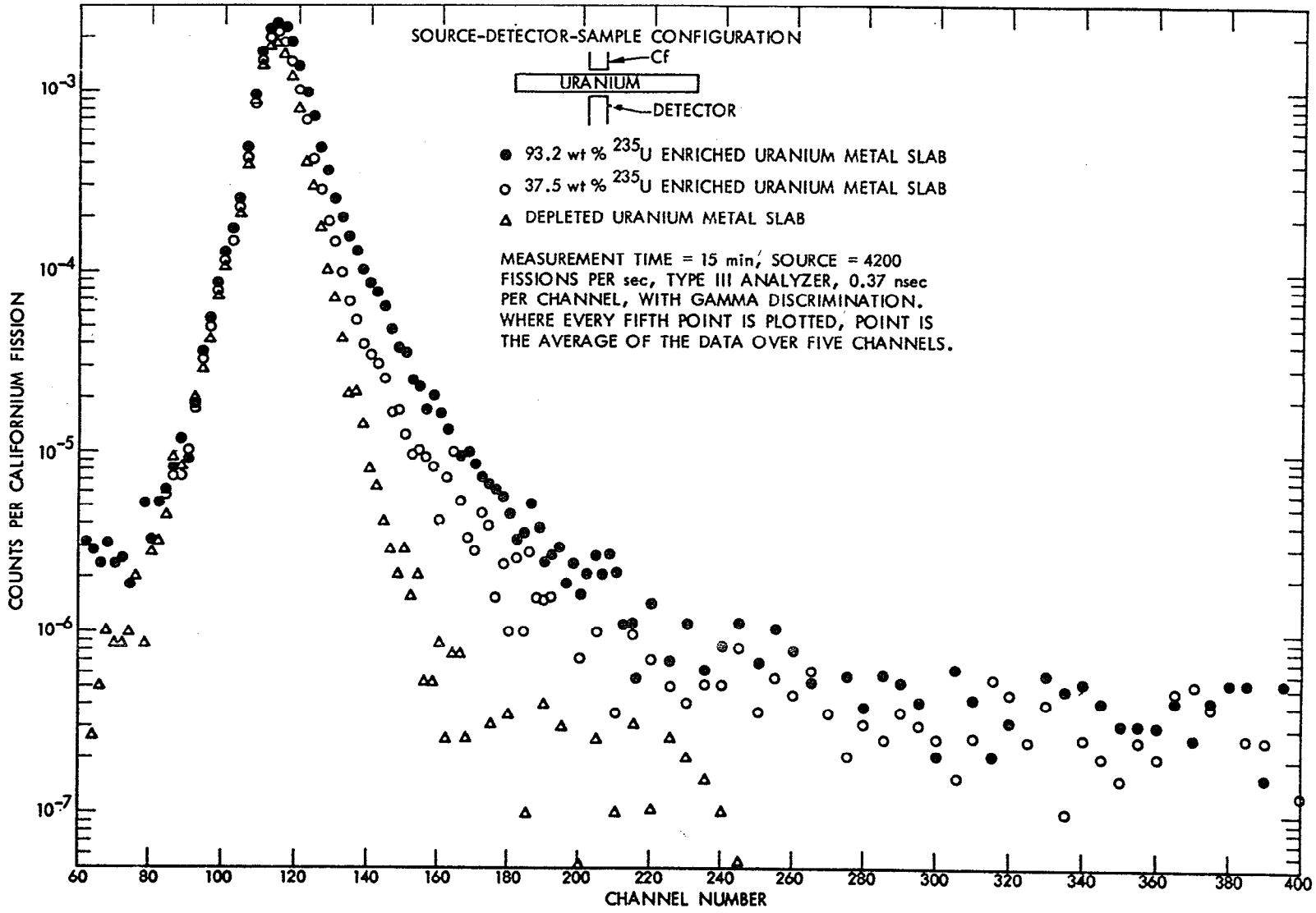

Fig. 4. Neutron Counts vs Time with the Source and Detector on Opposite Sides of the Unshielded Samples.

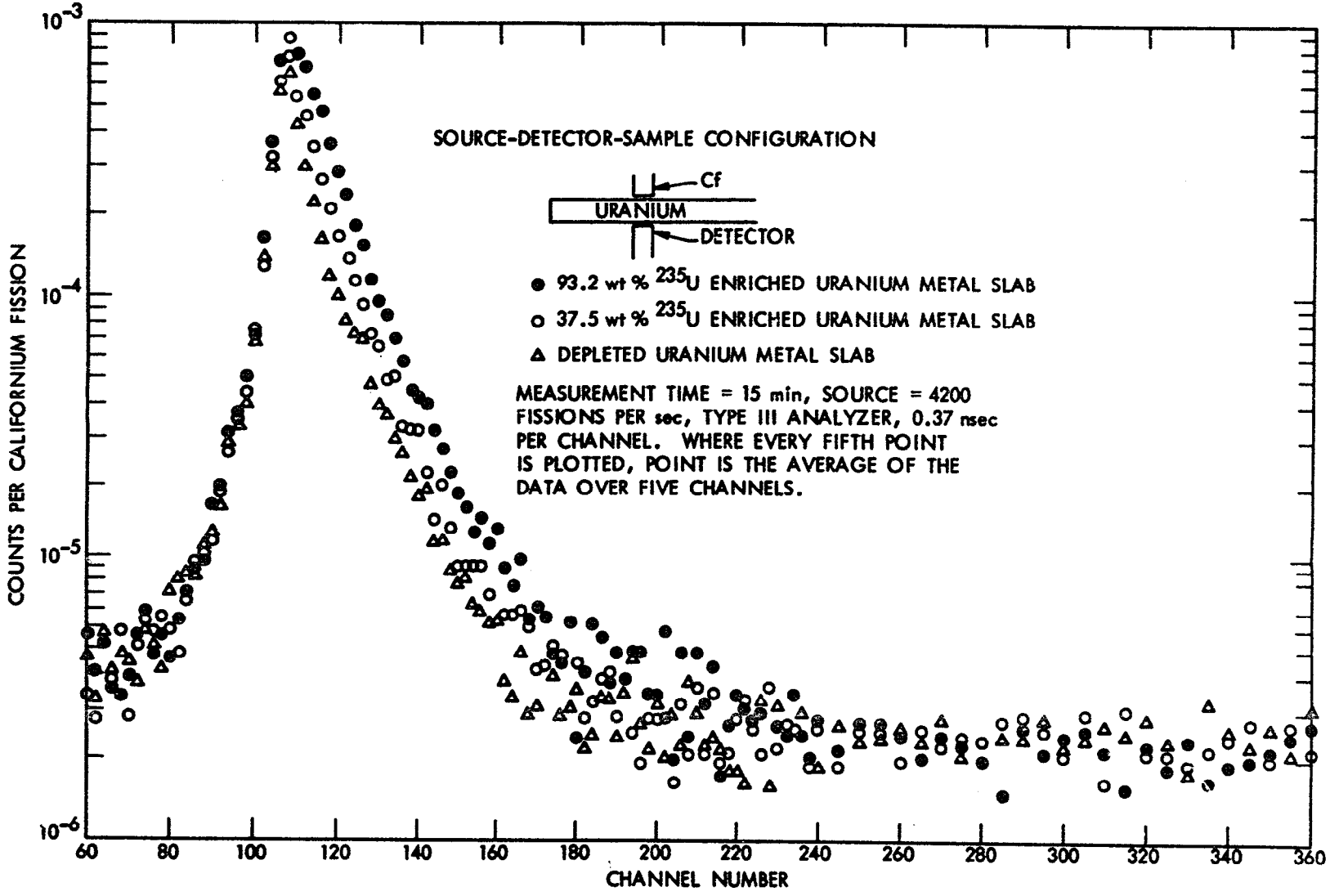

Fig. 5. Counts from Gamma Rays vs Time with the Source and Detector on Opposite Sides of the Unshielded Samples.

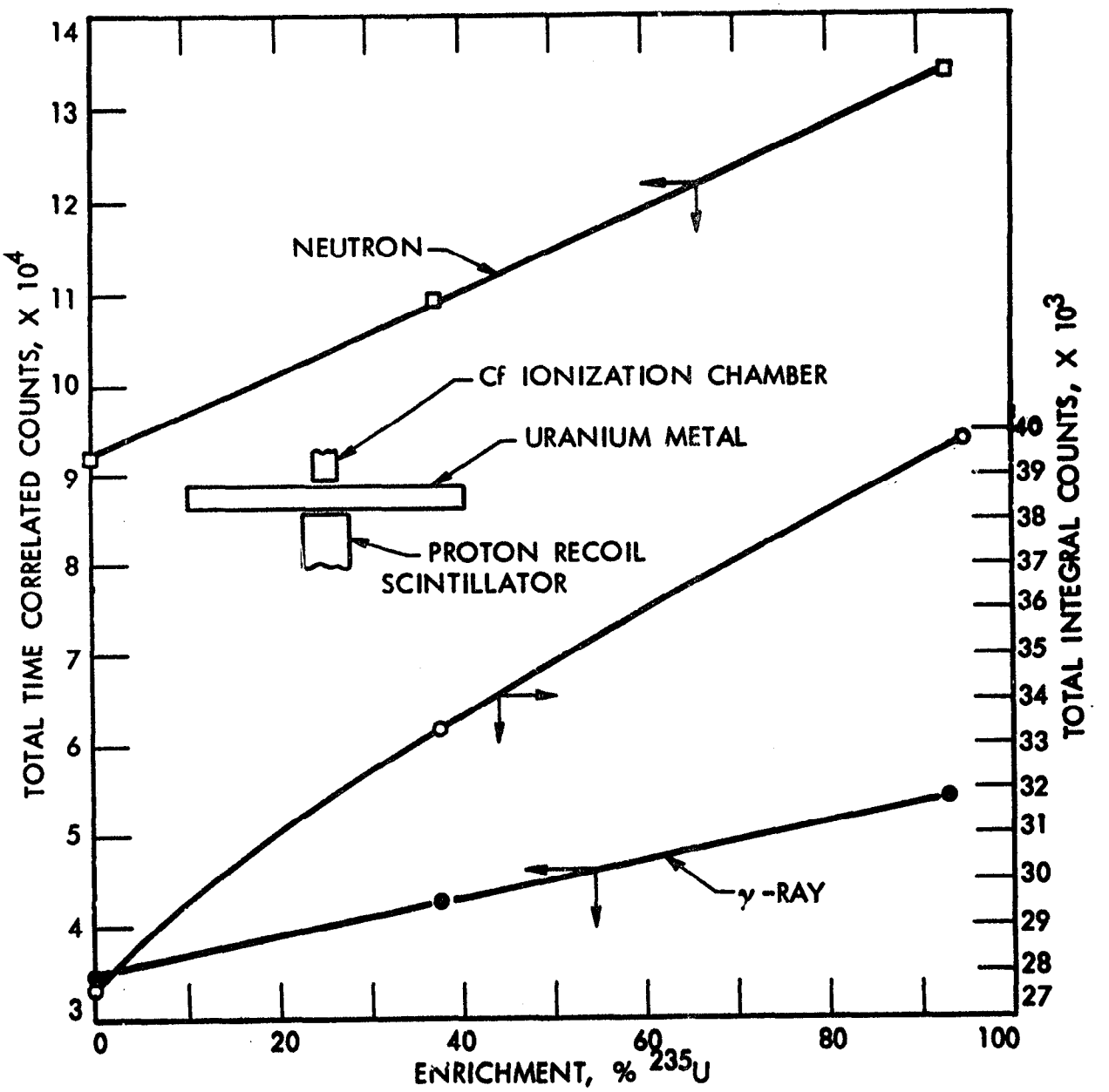

Fig. 6. Total Time Correlated Counts from a Randomly Pulsed Neutron Measurement with Californium and Total Integral Counts from a Source Neutron Multiplication Measurement as a Function of ^{235}U Enrichment of Unshielded Samples with the Source and Detector on Opposite Sides.

The total integral counts from a source neutron multiplication measurement for the various enrichment slabs of uranium are given in Fig. 6 also. In this case and in all cases subsequently observed, the source neutron multiplication was not linear with enrichment. Thus, the randomly pulsed neutron measurement has some advantage over the source neutron multiplication since an intermediate enrichment could be determined with only two standard samples.

In applying this technique to an in-plant determination, it may be desirable not to move the samples from their standard storage locations. In this case it may not always be possible to place the source and the detector on opposite sides of the sample. It may be preferable to apply this technique with both the source and the detector on the same side of the sample. The results of measurements without gamma discrimination and with the source and the detector on the same side of the unshielded depleted and 93.2 wt % ^{235}U enriched samples are shown in Fig. 7. The peak in these distributions, at about channel 106, is from gamma rays that travel without collision through the lead separating the source and the detector. The counts in channels greater than 106 are primarily from neutrons and gamma rays produced in fission induced in the sample by neutrons from californium fission. Some of the data recorded in channels immediately after channel 106 results from californium fission gamma rays and neutrons diffusing through the lead to the detector.

In many in-plant situations there may be other material surrounding the uranium so that the surface of the uranium is not accessible. In order to establish the applicability of the technique for cases where the surface of the uranium is not exposed, measurements were made with polyethylene or stainless steel abutting the sample. The results of the measurements with 1- and 2-in.-thick polyethylene for both configurations of source and detector are plotted in Figs. 8 to 11. In the reflection configuration with a very thick shield, most of the particles arriving at the detector will come from the source and not from fission in the sample, while in the transmission configuration the reverse will be true. Thus for very thick polyethylene shields the transmission configuration is preferred to the reflection configuration.

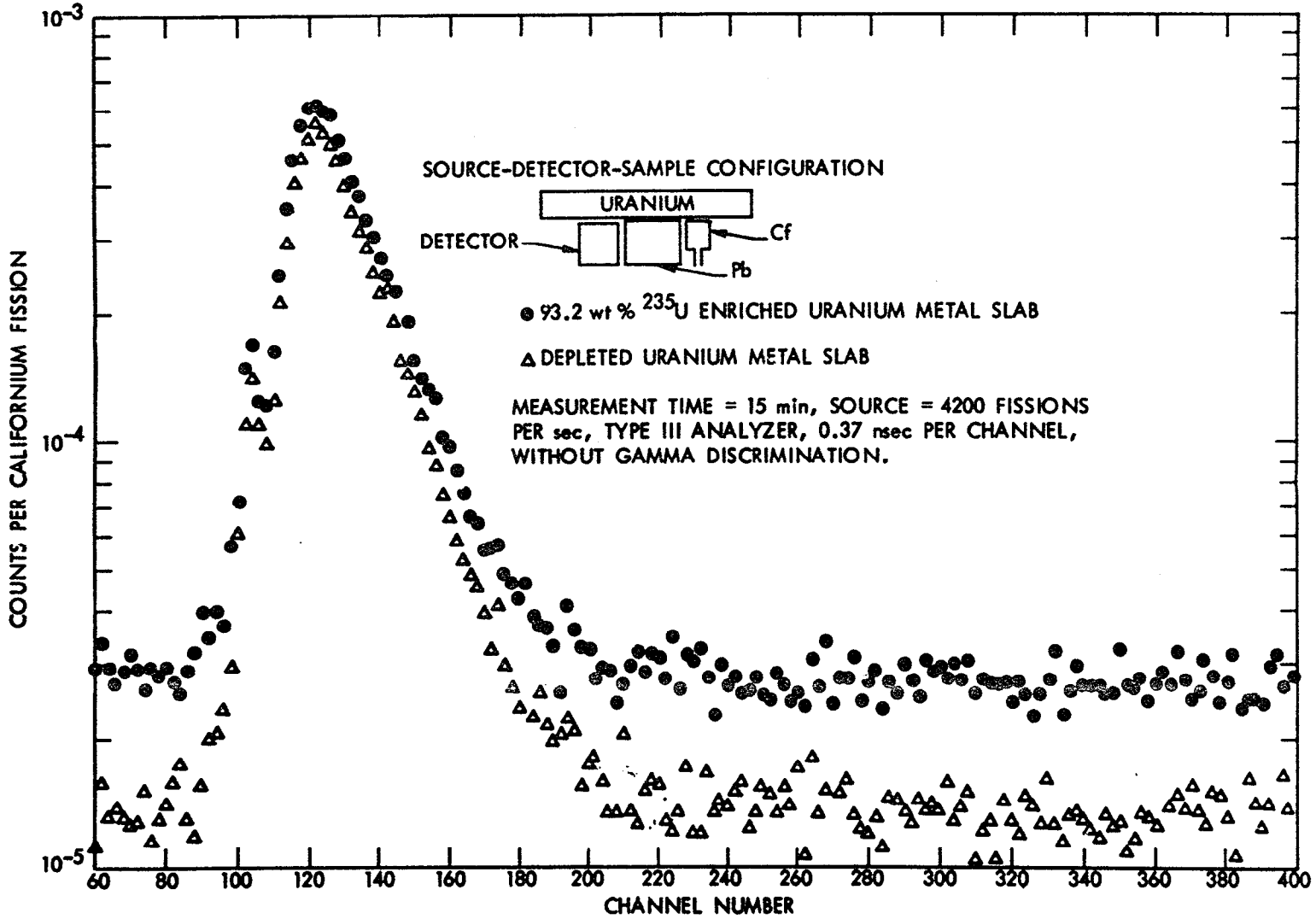

Fig. 7. Counts vs Time with the Source and Detector on the Same Side of Unshielded Samples.

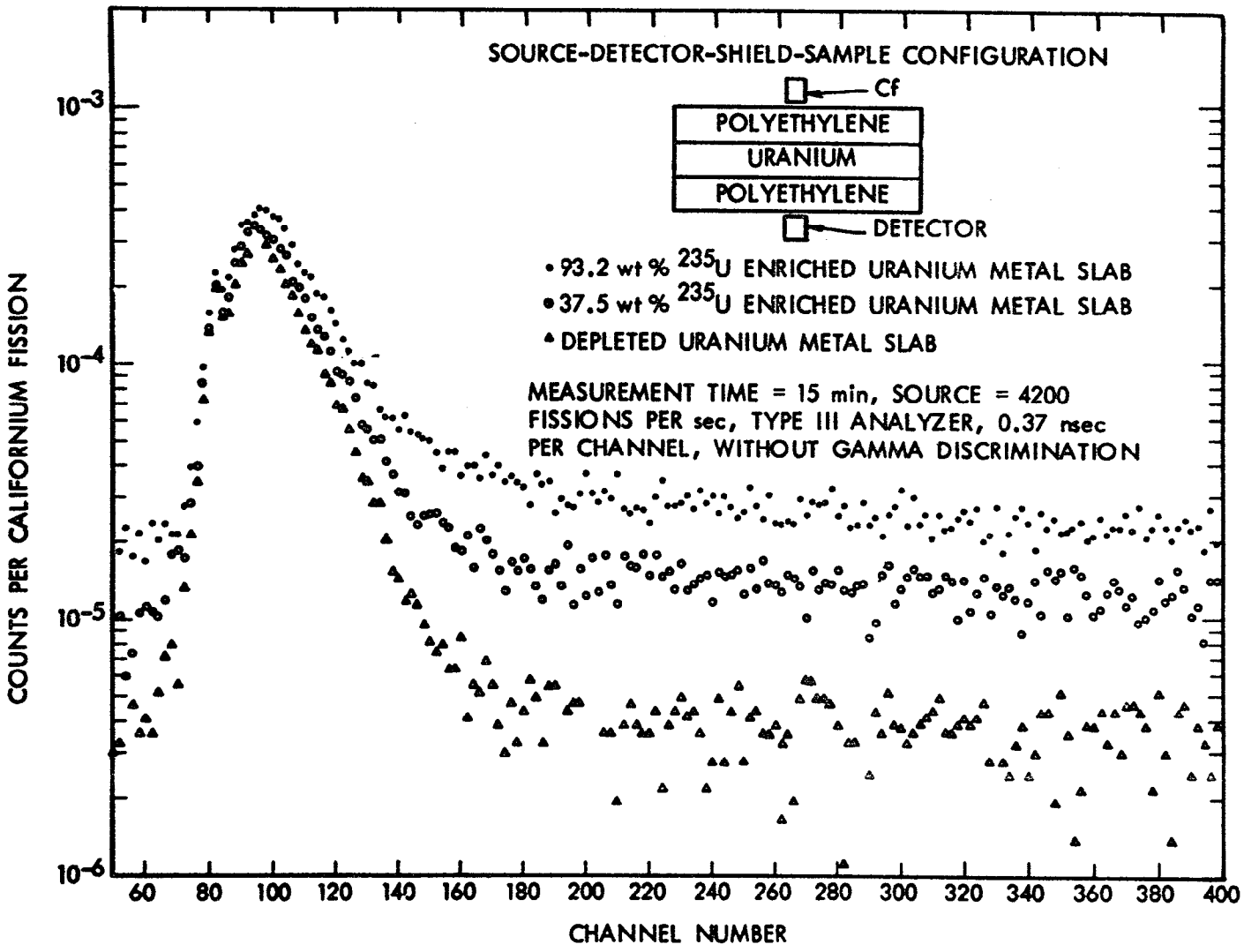

Fig. 8. Counts vs Time with the Source and Detector on Opposite Sides of the Sample and with a 1-in.-thick Polyethylene Shield.

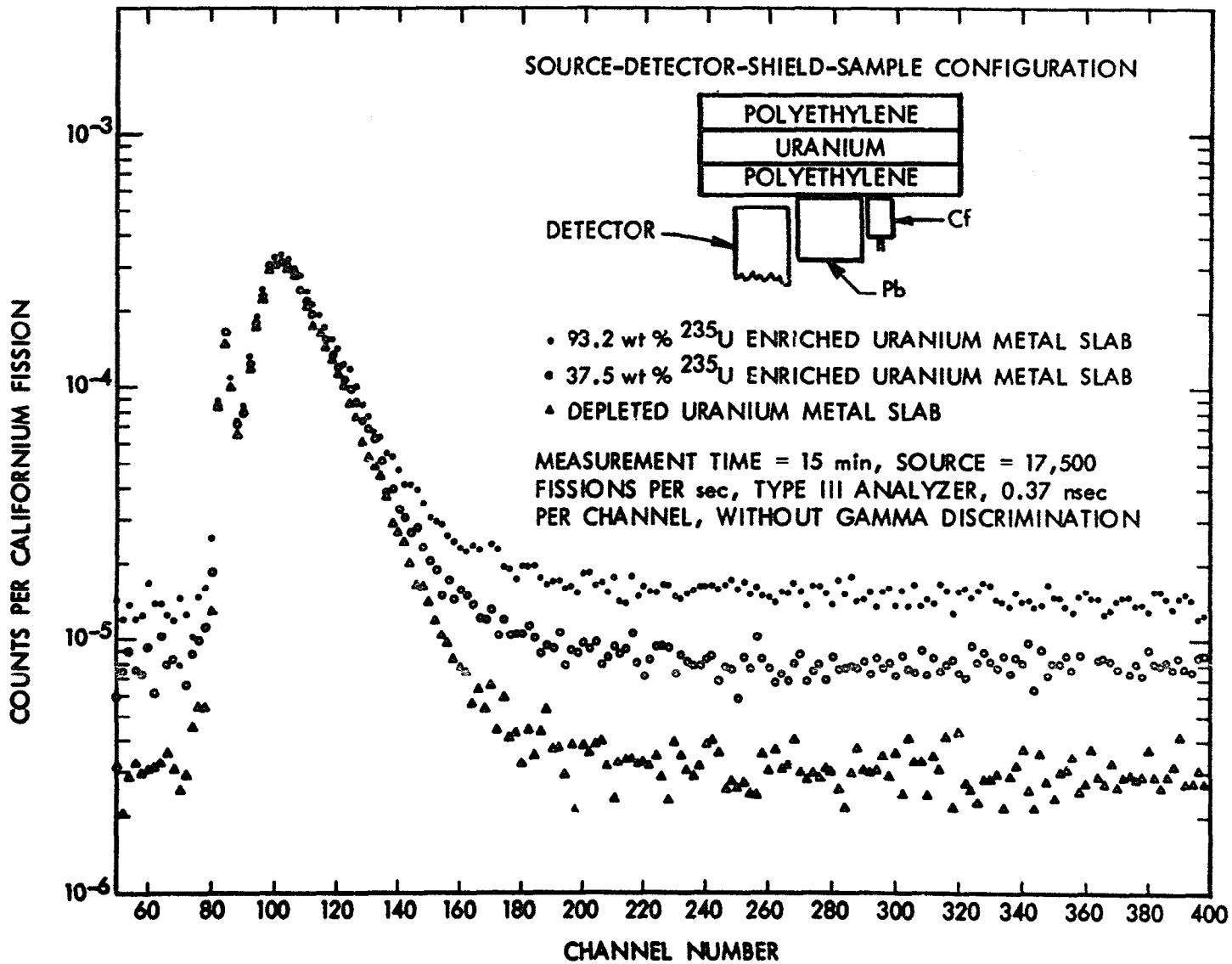

Fig. 9. Counts vs Time with the Source and Detector on Opposite Sides of the Sample and with a 2-in.-thick Polyethylene Shield.

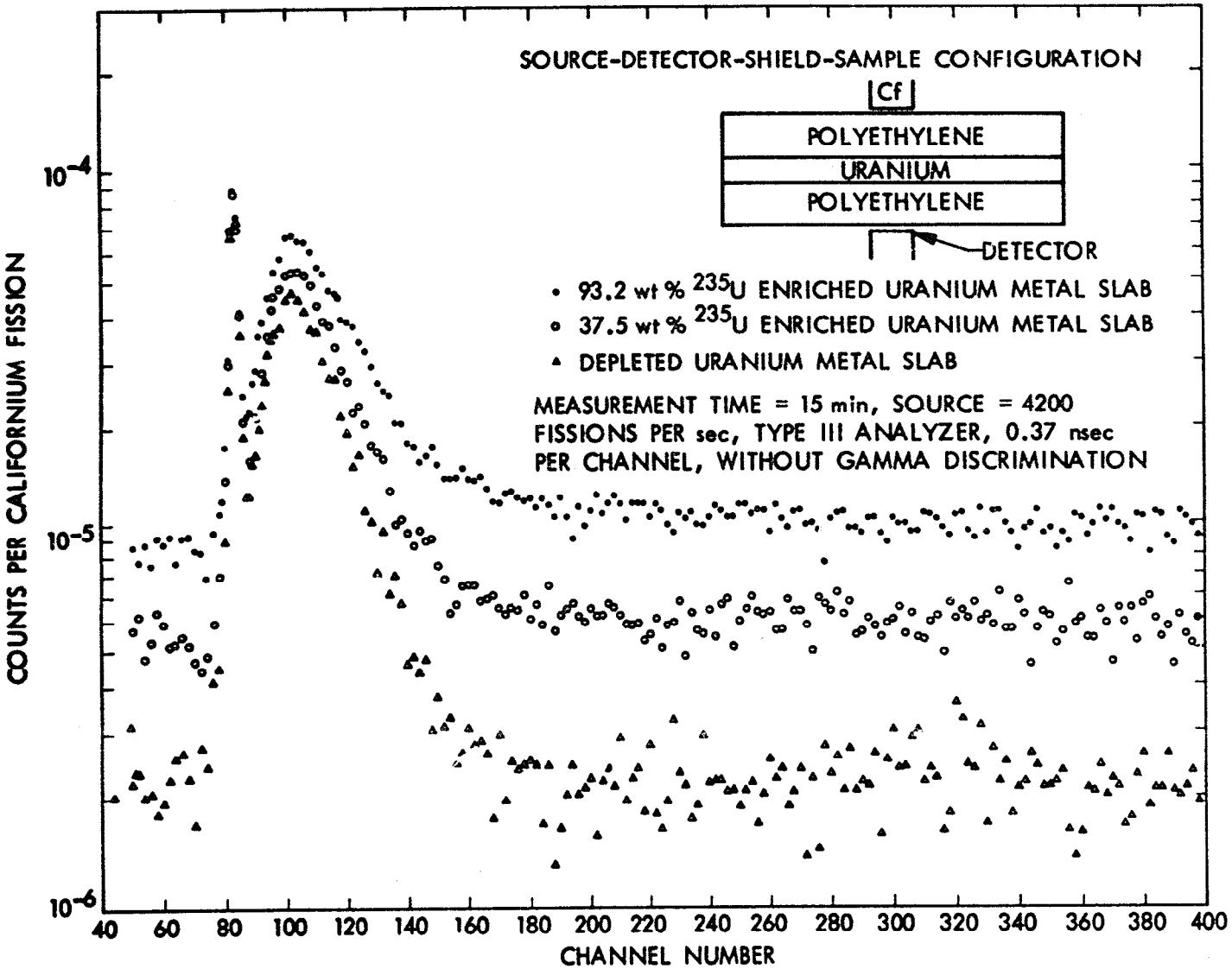

Fig. 10. Counts vs Time with the Source and Detector on the Same Side of the Sample and with a 1-in.-thick Polyethylene Shield.

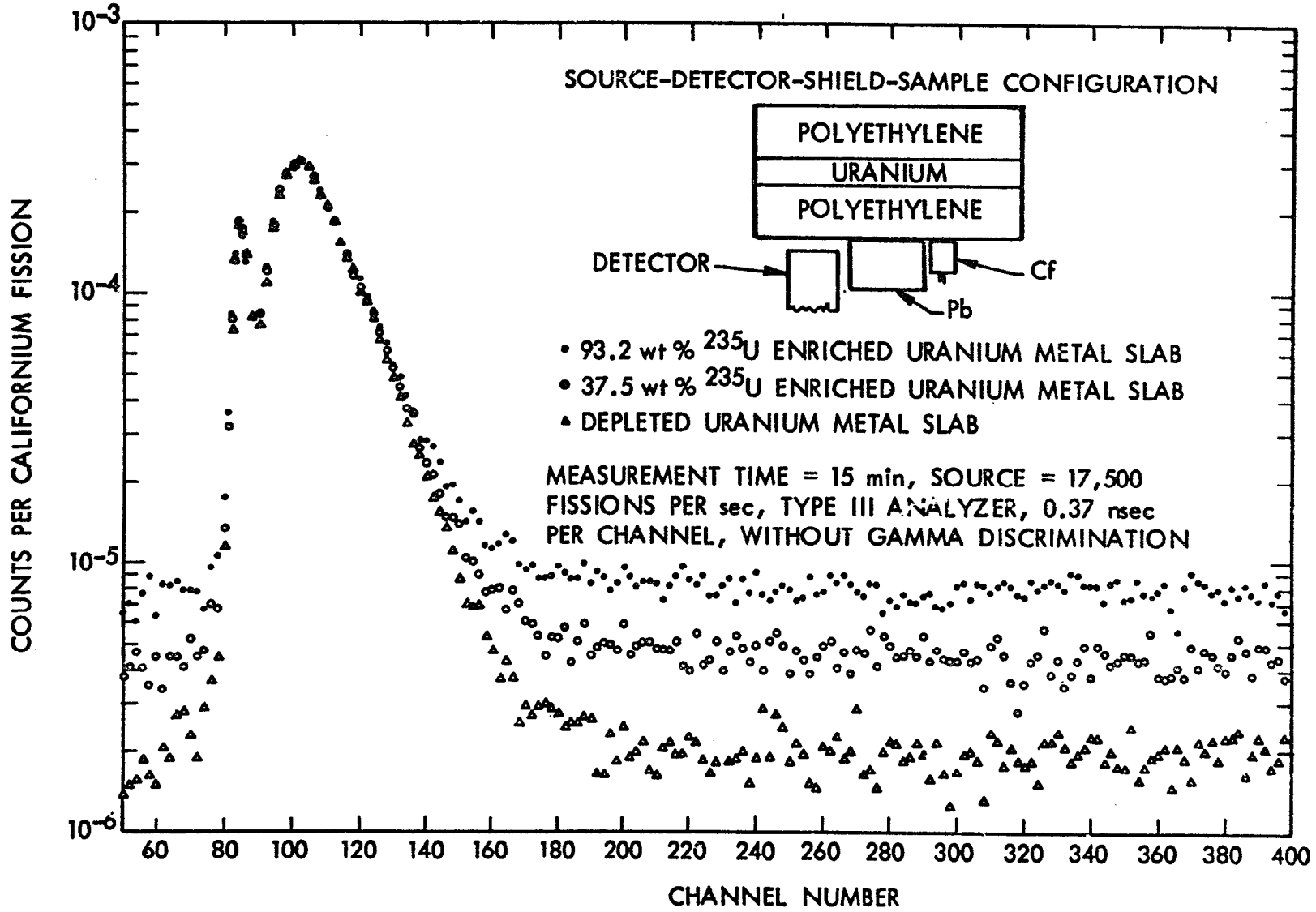

Fig. 11. Counts vs Time with the Source and Detector on the Same Side of the Sample and with a 2-in.-thick Polyethylene Shield.

The data for the measurements with a stainless steel shield are given in Figs. 12 and 13 for a transmission and a reflection configuration. The reflection configuration stainless steel abutted only one large surface of the sample, that on the same side as the source and detector.

The dependence of the total time-correlated-counts on the ^{235}U enrichment is shown in Fig. 14 for some of the measurements. For all of the configurations examined with this technique, the dependence of the total time-correlated-counts on enrichment was linear. These measurements have shown that the enrichment of uranium metal can be determined easily using this method by comparing the results of measurements with the unknown sample (in this case 37.5 wt % ^{235}U) with the results with standard samples of the same geometry and of known enrichment (in this case depleted in ^{235}U and enriched to 93.2 wt % ^{235}U).

All measurements presented so far in this report were performed in 15 min. In order to determine the precision of a measurement made in a short time, a series of 10 measurements requiring 30 sec each was performed for samples with a 1-in.-thick polyethylene shield. The source and detector were on opposite sides of the sample (see configuration on Fig. 8). The results of this series of measurements with a californium source of 4200 fissions/sec showed that the enrichment of the 37.5 wt % ^{235}U enriched sample could be determined to about ± 15% in 30 sec. Similar measurements with a 54,000 fission/sec source showed that the enrichment of the 37.5 wt % ^{235}U enriched sample could be determined to about ±3% in 30 sec. The errors given are one standard deviation and can be reduced by the use of larger sources. A limit on the amount of californium that can be used in this type of measurement exists since the individual spontaneous fissions of californium must be resolved. Present use of californium in ionization chambers with existing electronics would result in being able to resolve ~98 % of the spontaneous fissions in an ionization chamber with 2×10^6 californium fissions/sec (3.2 μg of ^{252}Cf). Thus, using a 3.2 μg ^{252}Cf source, a measurement could be performed in 30 sec to an accuracy of about ± 0.5%.

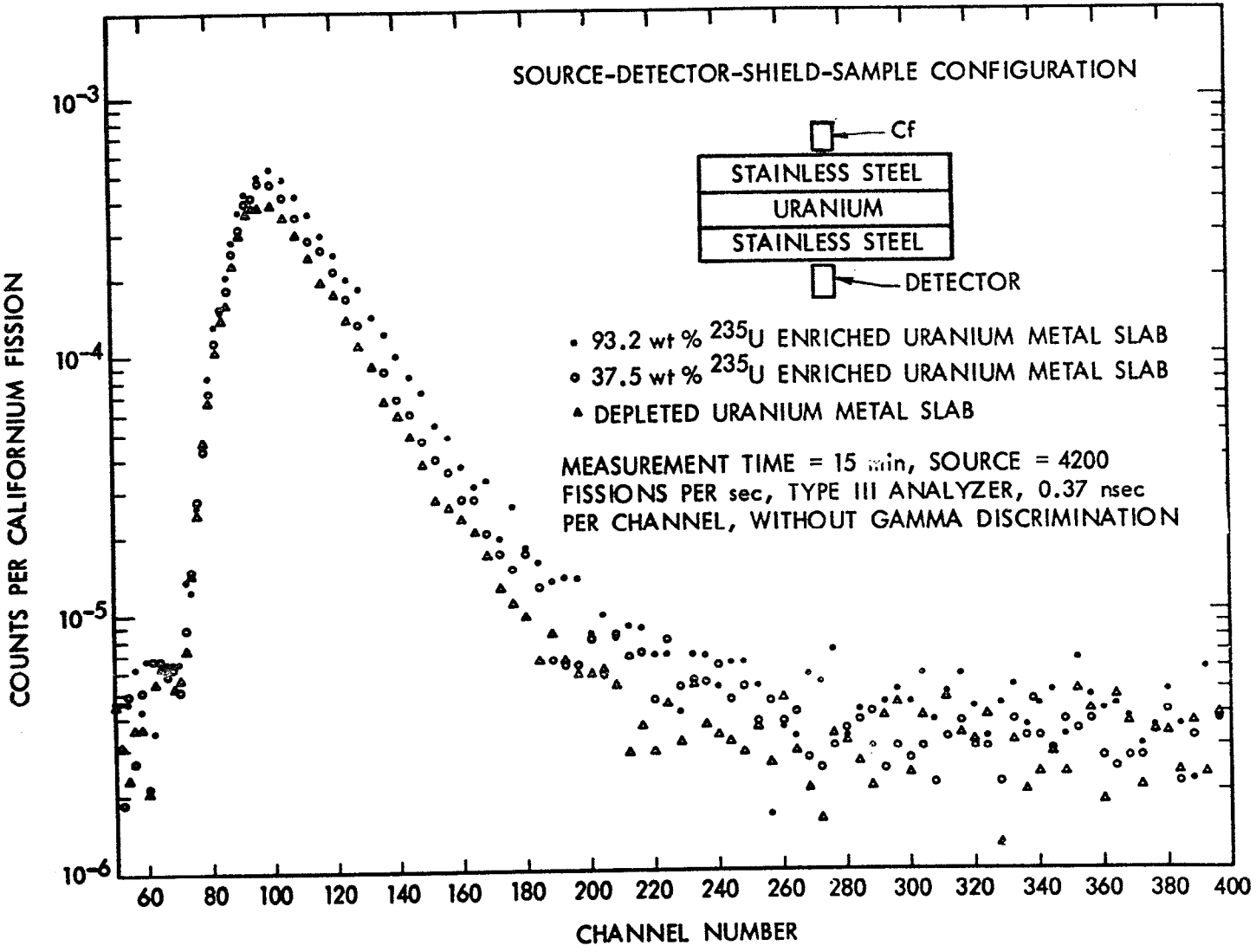

Fig. 12. Counts vs Time with the Source and Detector on Opposite Sides of the Sample and with a 1-in.-thick Stainless Steel Shield.

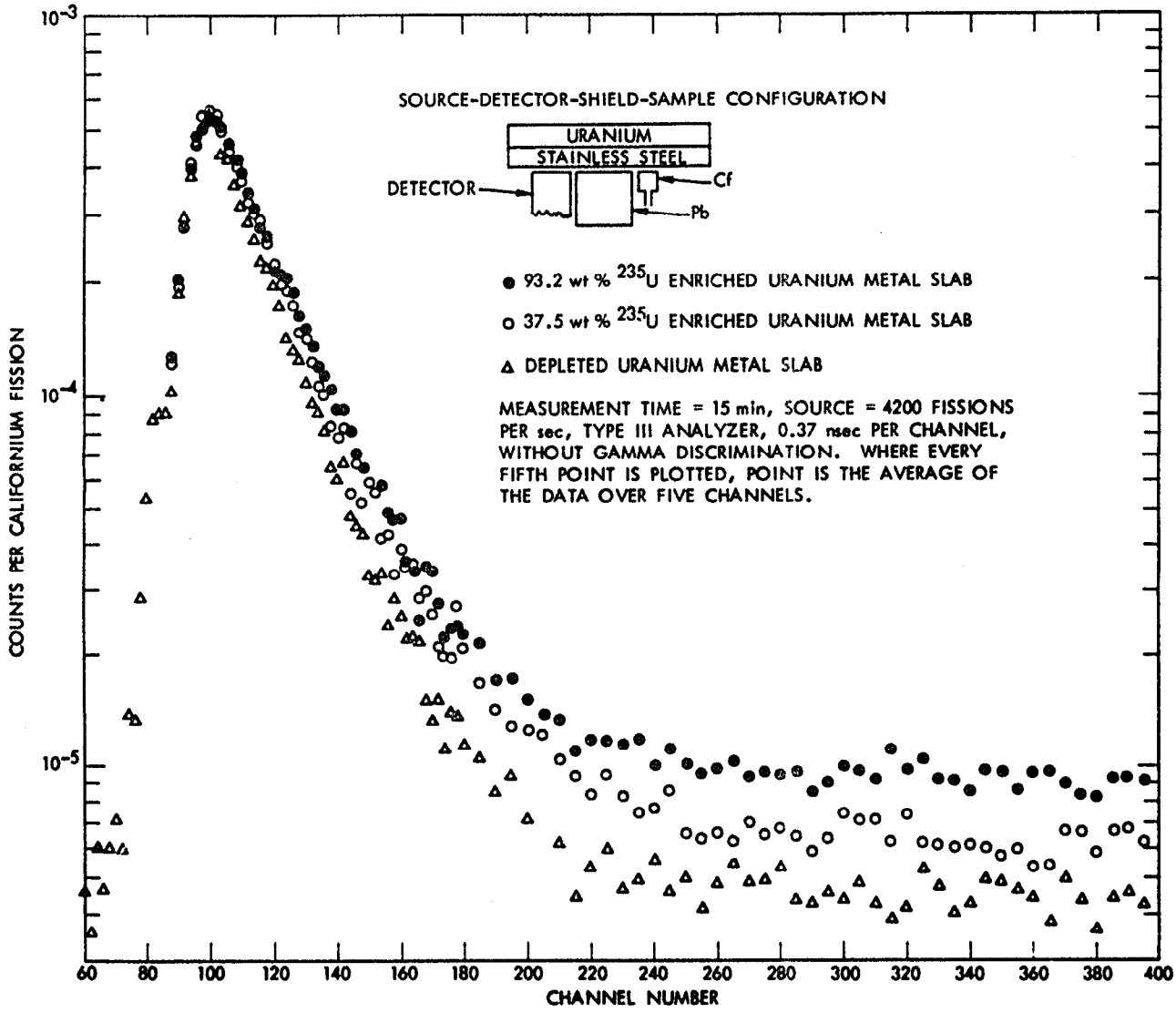

Fig. 13. Counts vs Time with the Source and Detector on the Same Side of the Sample and with a 1-in.-thick Stainless Steel Shield.

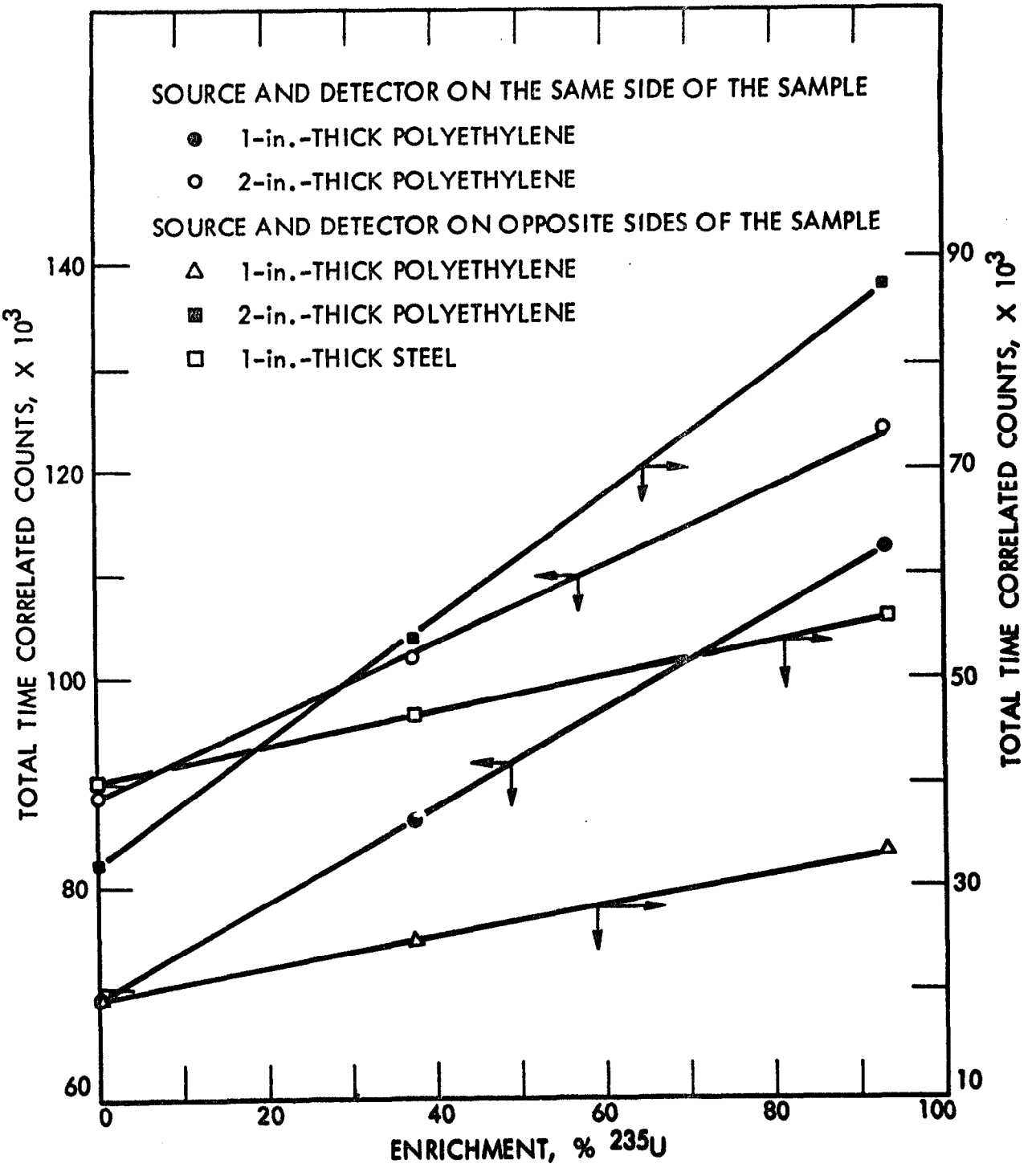

Fig. 14. Total Time Correlated Counts as a Function of ^{235}U Enrichment for Various Source-Detector-Shield Configurations.

CONCLUSIONS

The linear dependence of the total time correlated counts in a randomly pulsed neutron measurement on the enrichment of ^{235}U for uranium metal samples has shown that this method may be used for nondestructive assay determination provided measurements with two standard samples of the same geometry and of known enrichments preferably bracketing the unknown enrichment are made. Where the uranium is enclosed by large amounts of other materials, the source and detector must be placed on opposite sides of the sample (transmission measurement) since in the measurements with the source and the detector on the same side of the sample most of the neutrons and gamma rays reaching the detector are scattered by the other materials rather than from fission chains in the uranium. For shield material thicknesses up to 2 in. around the sample, the source and the detector may be placed on the same side of the sample. For thicknesses of shield material up to 2 in., the assay of a uranium metal sample can be measured to an accuracy of $\pm 0.5\%$ in 30 sec. Since these measurements can be made with detectors sensitive only to neutrons, the technique may also be useful for irradiated fuel.

For large amounts of shield material around the sample, larger californium sources would be required. A limit on the amount of californium that can be used in this type of measurement exists since the individual spontaneous fissions of californium must be detected. Present use of californium in ionization chambers with existing electronics results in being able to resolve $\sim 98\%$ of the spontaneous fissions in an ionization chamber with 2×10^6 californium fissions/sec (3.2 μg ^{252}Cf).

Although these measurements were performed with uranium, this technique will also be useful for determination of the ^{239}Pu content of plutonium since other measurements have shown that the prompt neutron decay can easily be measured in plutonium metal with up to 20 wt % ^{240}Pu.[8]

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8. J. T. Mihalczo, Trans. Am. Nucl. Soc. 15, 471 (1972).