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ELECTRONIC ASSAY OF ^{252}Cf NEUTRON SOURCES

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ABSTRACT

A new electronic method was developed that requires about 10 minutes to assay ^{252}Cf sources between 1 microgram and 50 milligrams. The assay is made by placing the ^{252}Cf source in the center of a polyethylene moderator and measuring the resulting thermalized neutrons with fission counters in the moderator. All measurements are referenced to a ^{252}Cf source calibrated with a manganese sulfate bath by the National Bureau of Standards. Sources smaller than 1 microgram and larger than 50 milligrams could be similarly assayed with appropriate modifications to the moderator. The new method is routinely compared with the manganese bath method to determine the precision and sensitivity of the measurements so that the new method can be used as a secondary standard. Measurements of ^{252}Cf content agree with manganese sulfate bath determinations to within $\pm 0.6\%$. The present design is tailored for a fission neutron spectrum but could be used with proper calibration in assaying between 10^6 and 10^{11} neutrons/second from sources having different spectra.

* The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

INTRODUCTION

Californium-252 is unique among transplutonium isotopes because it emits neutrons spontaneously and copiously for several years. One gram of ^{252}Cf emits 2.31×10^{12} spontaneous fission neutrons and 1.92×10^{13} alpha particles each second with an effective half-life of 2.646 years.*

The Savannah River Laboratory (SRL) has produced ^{252}Cf neutron sources for research and development studies since 1966.^{1,2} A precise, fast method was needed for assaying these sources. The manganese sulfate bath calibration initially used required about 16 hours irradiation for each source measurement, followed by gamma-ray pulse-height analysis of bath samples.

After concluding from laboratory tests that slow neutrons could be measured more precisely than fast neutrons, various thermalization techniques for determining neutron source strength were studied. The techniques that were studied included boric acid solution,³ manganese sulfate solution,^{4,5} nuclear reactor,⁶ helium production,⁷ and beta-gamma coincidence with gold foils.⁸ These techniques do not represent the field of neutron source calibration but do represent thermalization techniques of calibration.

Extensive laboratory tests were then performed with water and polyethylene moderators and a 50 microgram ^{252}Cf calibration source to determine the optimum detection array. A fission counter and a BF_3 counter were operated in the pulse mode and in

* The effective half-life is given by $T_{\text{eff}} = \frac{T_{\alpha} T_{\text{SF}}}{(T_{\alpha} + T_{\text{SF}})}$

the integrated ionization, or current, mode in studying the thermalization of ^{252}Cf neutrons in the two moderators. These tests indicated that a minimum of three fission counters should be spaced in the moderator around the centrally located ^{252}Cf source to minimize variations in the output signal for small changes in source position. The fission counters operated more reliably in the ionization mode. One advantage of this mode is that the output signal needs no correction for resolving time of the measuring instrument or dead time of the detector.

Polyethylene was the preferred moderator because it thermalizes neutrons as efficiently as water and is easier to transport and store in shielded facilities. Because the moderator would be used occasionally in moderate external neutron fields, it was designed to shield the counters from external neutrons.

COMPONENTS

Electronic System

Figure 1 is a block diagram of the electronic system. The three fission counters in the moderator are operated in the integrated, mean-level, detection mode described by Price⁹ with output currents of the counters summed and fed to a picoammeter. The "bucking" circuit shown in Figure 2 nulls the residual currents from ^{235}U decay in the fission counters. This current totals about 3.3×10^{-8} A for the three counters. The output signal from the picoammeter is fed to a strip-chart recorder through the circuit shown in Figure 3. Polarizing potential for the counters is provided by a high voltage supply. A negative potential is applied to the counter cathodes (housings) so that the anode signals to the picoammeter input are near ground potential. Figure 4 is a photograph of the complete system.

Fission Counters

The fission counters are Westinghouse type 6376 and are 2-1/16 inches in diameter by 11-11/16 inches long with a neutron-sensitive length of 6 inches. The inner walls are coated with 2 mg/cm² of U₃O₈ enriched to >90% ²³⁵U. The counters are filled to 1 atm with an argon-nitrogen mixture. When connected for operation in the ionization mode, the thermal neutron sensitivity is about 1.4 x 10⁻¹³ A/(neutron-cm²-second) with a gamma sensitivity of about 4.2 x 10⁻¹¹ A/(R-hour). The counters are operated in electrical parallel with -600 V applied to the cathodes.

The counters are insulated with paper tape and installed with their active volumes centered about the source position in stainless steel housings in the moderator. A polyethylene plug with a center hole to accommodate the connecting electrical cable is placed over each counter to fill the volume between the counters and the moderator top. The cable from each counter terminates in a small junction box on the moderator top. This cable supplies negative potential to the counter cathode and returns the signal to the junction box. A high voltage cable and a signal cable connect the junction box to the high voltage supply and picoammeter input.

Neutron flux incident on the ²³⁵U coating produces fission products with high kinetic energy. Because the ²³⁵U cross section follows an approximate 1/v dependence, thermalized neutrons will interact more frequently in the coating than faster neutrons. Some of the fission products dissipate their energy by ionizing the filling gas of the counter. The electrons thus freed are attracted to the positively charged anode of the counter and produce additional ionization en route. The resulting current at the anode varies linearly with neutron flux over a wide range.

Moderator

Figure 5 is a photograph of the moderator top. Three fission counters are spaced 120° apart in the moderator and 4.5 inches from the axis. The central source well is surrounded by 1/2-inch-thick lead shielding to minimize the low energy gamma radiation (<200 kev from alpha decay) reaching the counters. This shielding has little effect on the prompt fission and fission product gamma activity which contribute about 0.2% of the signal obtained from a given ^{252}Cf source. Sources up to 1-1/4 inch in diameter and several inches long can be placed in the source well for testing.

The moderator dimensions were determined experimentally to obtain less than 0.1% leakage of ^{252}Cf neutrons from the moderator. The leakage of neutrons from the moderator is not significant from the standpoint of source measurement, but is important in attenuating any external neutron flux that could enter the moderator and interfere with source measurements.

Experiments were conducted to determine a reasonable distance between the source and fission counters. The best location for the counters was assumed to be where the thermalized flux was maximum, and the ratio of fast to slow flux was minimum. Placing the counters close to the source would enhance their low-level response but would limit their high-level response because of saturation effects and subsequent non-linear response at higher flux levels. The compromise then involved placing the counters in a region of adequate thermalization but retaining the desired range of linear response.

Figure 6a is a plot of counts/unit time versus moderator thickness that was obtained by placing a BF_3 counter at a fixed distance from a ^{252}Cf source in air and adding discrete thick-

nesses of water and polyethylene between the source and counter. Experimental details and dimensions are shown in Figure 6b. The peak count from thermalized neutrons was obtained for a 2.2-inch thickness of polyethylene and a 2.7-inch thickness of water. Figure 7 shows the ratio of fast neutron counts to slow neutron counts from the ^{252}Cf source; this ratio was obtained through the indicated thicknesses of water and polyethylene. The minimum ratio of fast to slow counts was obtained for a 3.1-inch thickness of polyethylene and a 3.7-inch thickness of water. The efficiencies of the Hornyak¹⁰ detector for fast neutron measurements and the BF_3 counter for slow neutron measurements were determined experimentally, and all counts were corrected for detector efficiency. Figures 6 and 7 indicate that polyethylene is about 1.2 times more efficient as a moderator than water for thermalizing ^{252}Cf neutrons. This is mainly due to the higher hydrogen concentration per unit volume in polyethylene. This "efficiency" factor was used to determine the dimensions of the polyethylene moderator from experimental data obtained in water. Figure 7 also indicates that the counters should be spaced between 2 and 5 inches from the source in polyethylene to operate in a region of adequate thermalization. The counters were spaced 4.5 inches from the source well with about 4 inches of polyethylene between counters and source.

The solid curve of Figure 8 shows the theoretical distribution of thermal neutron flux in an infinite H_2O moderator from a point source of fission neutrons. This curve was plotted from the two-group diffusion theory formula and constants shown in the figure.¹¹ Experimental points obtained with a miniature BF_3 counter in a large water tank in which a source of ^{252}Cf was centrally located agree very well with the theoretical curve. This curve shows that about 3.6 inches of water moderator attenuates the thermalized flux from the ^{252}Cf source to a tenth of its

original value. The corresponding thickness of polyethylene moderator is about 3 inches, as calculated with the "efficiency" factor for polyethylene given above. Approximately 10.5 inches of polyethylene was provided external to the fission counters to obtain the <0.1% leakage value previously mentioned.

Figure 9 shows the experimental response from a fission counter operated in the integrated, mean-level, ionization mode as the counter was moved outward from a ^{252}Cf source centrally located in a large volume of water moderator. The response curve deviates from linearity on the semilog plot as the counter is brought close to the source, due possibly to the relatively large volume of moderator displaced by the counter. The curve indicates that at least 4 inches of water (or 3.3 inches of polyethylene) moderator should be placed between the counter and source to operate the counter in a region of fairly uniform thermalization, and substantiates the use of 4 inches of polyethylene between the source well and counters.

Figure 10 is a scale drawing showing the top and side-sectional views of the moderator that was designed and built from the laboratory tests. The moderator was constructed from 1/2-inch-thick sheets of stress-relieved polyethylene. The sheets were tightly bolted together. Thicker sheets of polyethylene or solid polyethylene could be used at an increased cost, but would not significantly improve the overall performance. Cadmium sheeting 0.030-inch-thick was installed around the counters to further shield them from external neutrons. Laboratory tests indicated that the location of the cadmium shield in the moderator outside the counter housings was not critical, but that at least 5 inches of polyethylene was required around the cadmium shield for effective absorption of external neutrons in the cadmium. Approximately 9 inches of polyethylene was provided around the

cadmium shield.

SOURCE DESCRIPTION

The source material is californium oxide. The isotopic composition changes with time because ^{252}Cf decays more rapidly than ^{249}Cf , ^{250}Cf , and ^{251}Cf . Small amounts of ^{253}Cf and ^{254}Cf are also present. Only ^{252}Cf and ^{254}Cf emit neutrons, and the small fraction of the total neutrons emitted by ^{254}Cf is considered during calibration measurements.

A typical source is doubly encapsulated in platinum-rhodium and stainless steel as described by Boulogne and Faraci¹² and has an active volume of source material of less than 25 mm³. Sources of this type are shown in Figure 11 and are screw-attached to the end of a brass rod and placed in a polyethylene insert for calibration in the moderator.

The ^{252}Cf content of other configurations up to 1-1/4 inch in diameter and several inches long can also be determined by removing the polyethylene insert. Ion exchange columns and shipping assemblies are measured routinely.

CALIBRATION

Measurements are referenced to a nominal 50 microgram source of ^{252}Cf that was previously calibrated with the manganese sulfate bath technique by the National Bureau of Standards.¹³ The NBS determined that the total neutron emission from the source was 1.13×10^8 neutrons/second on 12/8/69 with an estimated "total error" of 1.4% for the measurement.

Numerous sources of the type shown in Figure 11 have been measured with the new electronic technique, and some of these

sources were also measured in one or more of the four manganese sulfate baths at the SRL with overall agreement between the two techniques of $\pm 0.6\%$. The NBS calibration source was also measured in the SRL baths at irregular intervals. Sources measured to date have contained between 4 microgram and 11 milligrams of ^{252}Cf . Sources containing between 1 microgram and 50 milligrams of ^{252}Cf may be measured with the present system.

The initial measurement involves placing the calibrated ^{252}Cf source at the center of the polyethylene moderator and recording the output current. The calibration source is then removed, and the unknown source is placed in the moderator and the output current is recorded. The ratio of the unknown source current to the calibrated source current is multiplied by the ^{252}Cf content of the calibrated source corrected to the time of measurement to obtain the equivalent ^{252}Cf content of the unknown source. The equivalent ^{252}Cf content is then corrected for ^{254}Cf content and gamma contribution to yield the actual ^{252}Cf content.

The manganese sulfate bath technique used as a reference for the electronic method was described by Marion and Fowler.¹⁴ The SRL baths are 4 feet in diameter and 4 feet high, and the manganese atoms are activated to ^{56}Mn in about 900 liters of manganese sulfate solution. A 0.45 M solution of MnSO_4 is used for up to 100 micrograms of ^{252}Cf , and a 0.075 M solution is used for larger amounts of ^{252}Cf to avoid excessive solution activity. After a timed irradiation of about 16 hours by the source being measured, the MnSO_4 solution is thoroughly stirred, and two aliquots are taken for assay. The assay is made by counting the gamma rays emitted from ^{56}Mn (produced by $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$) with a $\text{NaI}(\text{Tl})$ crystal, and averaging the results from the two aliquots. Corrections are made for neutron leakage and for neutron capture by hydrogen,

oxygen, and sulfur atoms in the solution. The accuracy of this technique with the SRL baths is estimated to be $\pm 3\%$ with a precision of $\pm 0.5\%$.

TEST RESULTS

Nine sources were measured with the electronic technique and also with an SRL manganese sulfate bath. These measurements are summarized in Table I and indicate an average variation of $\pm 0.6\%$ from the bath determinations. Five of these sources were remeasured between eight and thirty-five days later with the electronic method, and these results are shown in Table II. The "decayed value" in Table II is the original measured value (electronic) corrected for decay for the appropriate time using a half-life of 2.646 years for ^{252}Cf . The percent difference shown in Table II between the decayed value and the corresponding measured value is less than $\pm 0.3\%$.

The effectiveness of the moderator in shielding the fission counters from external neutrons was determined experimentally. While measuring a 93 microgram source of ^{252}Cf in the moderator, an unshielded 1 milligram source of ^{252}Cf was brought in contact with the outside of the moderator at midheight. The current from the fission counters increased from 0.403×10^{-7} to 0.408×10^{-7} A, for an error in the 93 microgram source measurement of $+1.2\%$.

The sensitivity of the fission counters to ^{252}Cf gamma radiation was determined experimentally in air with the same source-to-counter spacing as is used in the moderator. A 1/2-inch-thick lead shield was placed between the source and the counter and 0.030-inch-thick cadmium sheeting was placed around the counter for these tests. Several sources of varying strength were then measured to determine that about 0.2% of the signal

from a given source was due to high energy gamma radiation from the source. These tests neglected gamma ray absorption in the polyethylene moderator and a small amount of gamma radiation from thermal neutron capture in the cadmium surrounding the fission counter.

When sources in the milligram range are measured, the aluminum outer housings and electrodes of the fission counters become sufficiently activated to interfere with the measurement of subsequent sources. The activation decays with the characteristic half-life of ^{28}Al and is negligible after about 10 minutes. This effect is important only when measuring weak sources after measuring strong sources and the usual procedure is to measure sources in order of increasing strength. The time between successive measurements is normally more than 10 minutes because of the source-handling techniques and procedures used.

STATISTICAL CONSIDERATIONS

The accuracy of the electronic method is difficult to define because absolute calibrations are not now possible. Measurements are referenced to a source of ^{252}Cf free of ^{254}Cf that was calibrated by the NBS with a manganese sulfate bath.

The thermalization technique of neutron source calibration including the manganese bath is discussed in an NBS report, and the results of a large number of neutron source calibrations of this type are tabulated.¹⁵ The errors quoted for various separate determinations range between ± 2 to ± 3 percent. The report concludes that careful measurements of absolute neutron emission are capable of about ± 1.5 percent accuracy.

The four manganese sulfate baths at SRL were cross-compared with the nominal 50 microgram calibration source. Single measurements were made in each bath on different days, and the earlier measurements were mathematically decayed to the last measurement day. The spread among the results was ± 1.6 percent from the average of the four determinations.

Independent measurements of a nominal 5 milligram source were made with the electronic method by three laboratory personnel on the same day to avoid decay corrections. Each person obtained five readings from the unknown source and five readings from the calibration source and calculated the ^{252}Cf content of the unknown source according to procedure. All the results were within ± 0.2 percent of the average of the three determinations.

The statistical variation in the fission counter detection system was studied with the method outlined by Price.¹⁶ The expected standard deviation from the three counters for a current of 4×10^{-7} A from a 1 milligram source of ^{252}Cf is 0.1 percent, based on 2.7×10^6 ion pairs per fission product and an input electrometer time constant of 0.5 second. Similarly, the expected standard deviation is about 0.3% from a 100 microgram source and about 1% from a 10 microgram source.

More intense calibration sources containing up to 5 milligrams of ^{252}Cf will be prepared and sent to the NBS for calibration. The sources will be used with our present calibration source in a continuing program to compare the electronic method with the manganese sulfate bath method until sufficient data are accumulated for a reliable least squares fit. From the least squares fit, we can deduce the sensitivity of the electronic measurement system in micrograms of ^{252}Cf /microampere. The measurement of an unknown ^{252}Cf source will then be reduced to multiplying its

output current by the sensitivity and correcting the result for ^{254}Cf content and gamma contribution. The electronic system will then be a secondary standard, and a calibration source will no longer be necessary except as an occasional cross-check on the electronic system. The ultimate precision of measurement with the electronic method is expected to be better than ± 0.5 percent.

The sensitivity that is deduced by this technique will apply only to the one electronic system. When additional electronic systems are built they will require the same cross-comparison with the NBS calibration sources to determine their measurement sensitivity.

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TABLE I

Electronic Measurements versus Manganese Sulfate Bath

<u>Source Number</u>	<u>²⁵²Cf Content, μg</u>		<u>% Diff</u>
	<u>Electronic*</u>	<u>Bath**</u>	
A	5235 $\pm 0.3\%$	5215	+0.4
B	1030 $\pm 0.6\%$	1032	-0.2
C	224 $\pm 0.4\%$	226	-0.9
D	360 $\pm 0.5\%$	358	+0.5
E	29.9 $\pm 0.7\%$	30.1	-0.7
F	130 $\pm 0.4\%$	131	-0.8
G	4960 $\pm 0.4\%$	4930	+0.6
H	595 $\pm 0.3\%$	590.5	+0.7
I	4.80 $\pm 1.0\%$	4.77	+0.6

* Average of five measurements with spread

** Single measurements

TABLE II
Later Electronic Measurements

<u>Source Number</u>	<u>Days Between Measurements</u>	<u>²⁵²Cf Content, μg</u>		<u>% Diff</u>
		<u>Decayed Value</u>	<u>Measured Value</u>	
A	35	5093	5103	+0.2
B	22	1012	1015	+0.2
D	8	359	358	-0.3
E	8	29.7	29.6	-0.3
F	8	130.1	130.3	+0.2

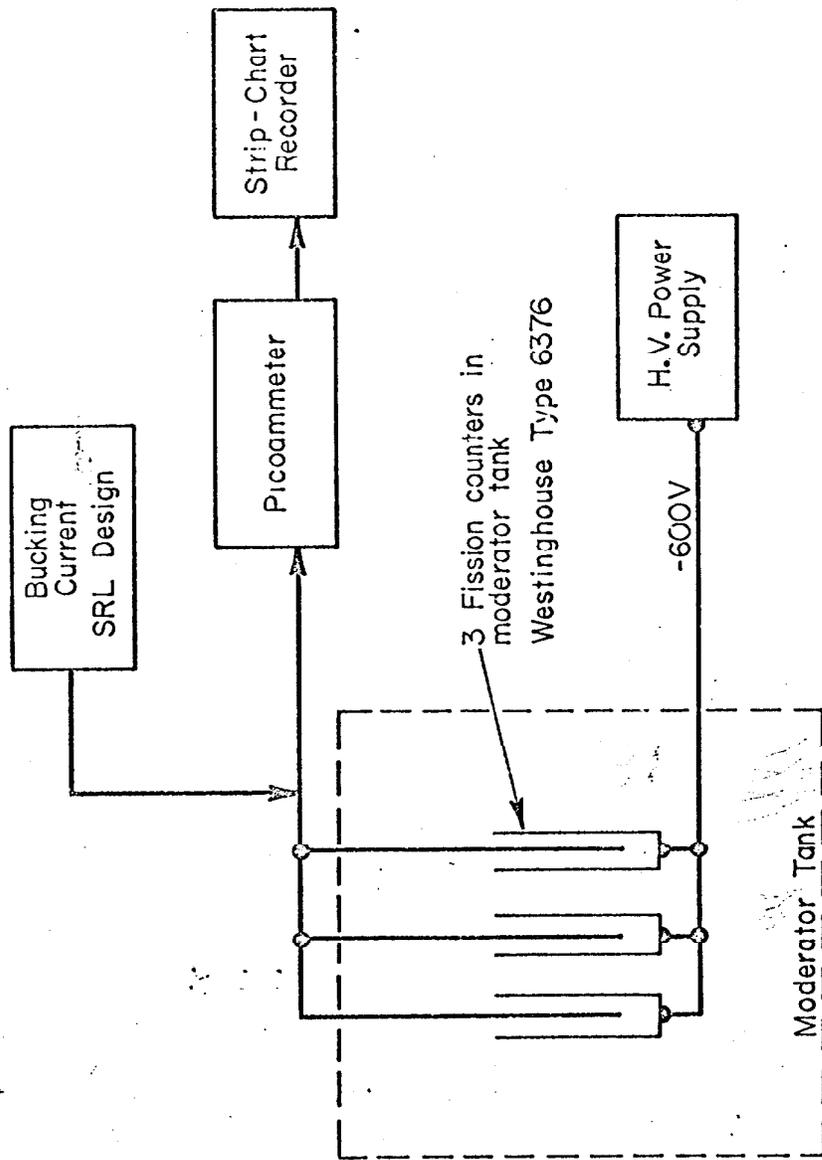


FIG. 1 ELECTRONIC SYSTEM

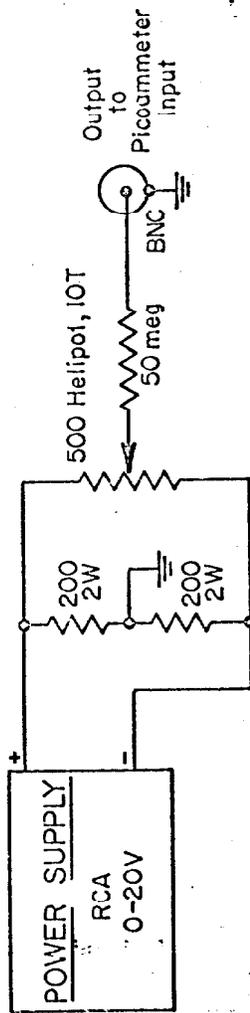


FIG. 2 BUCKING CIRCUIT

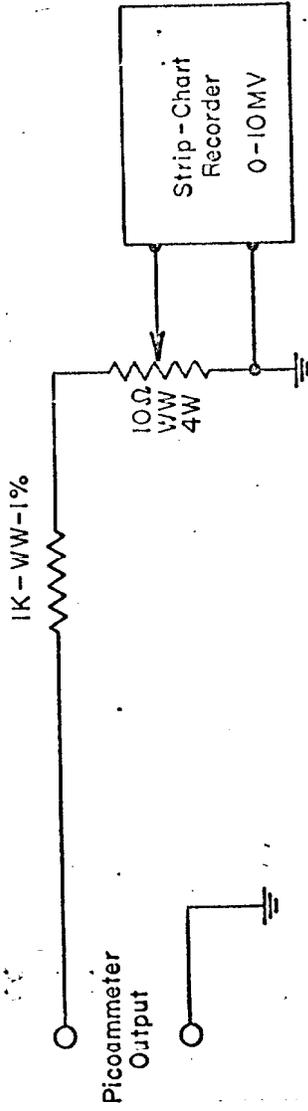


FIG. 3 RECORDER HOOKUP

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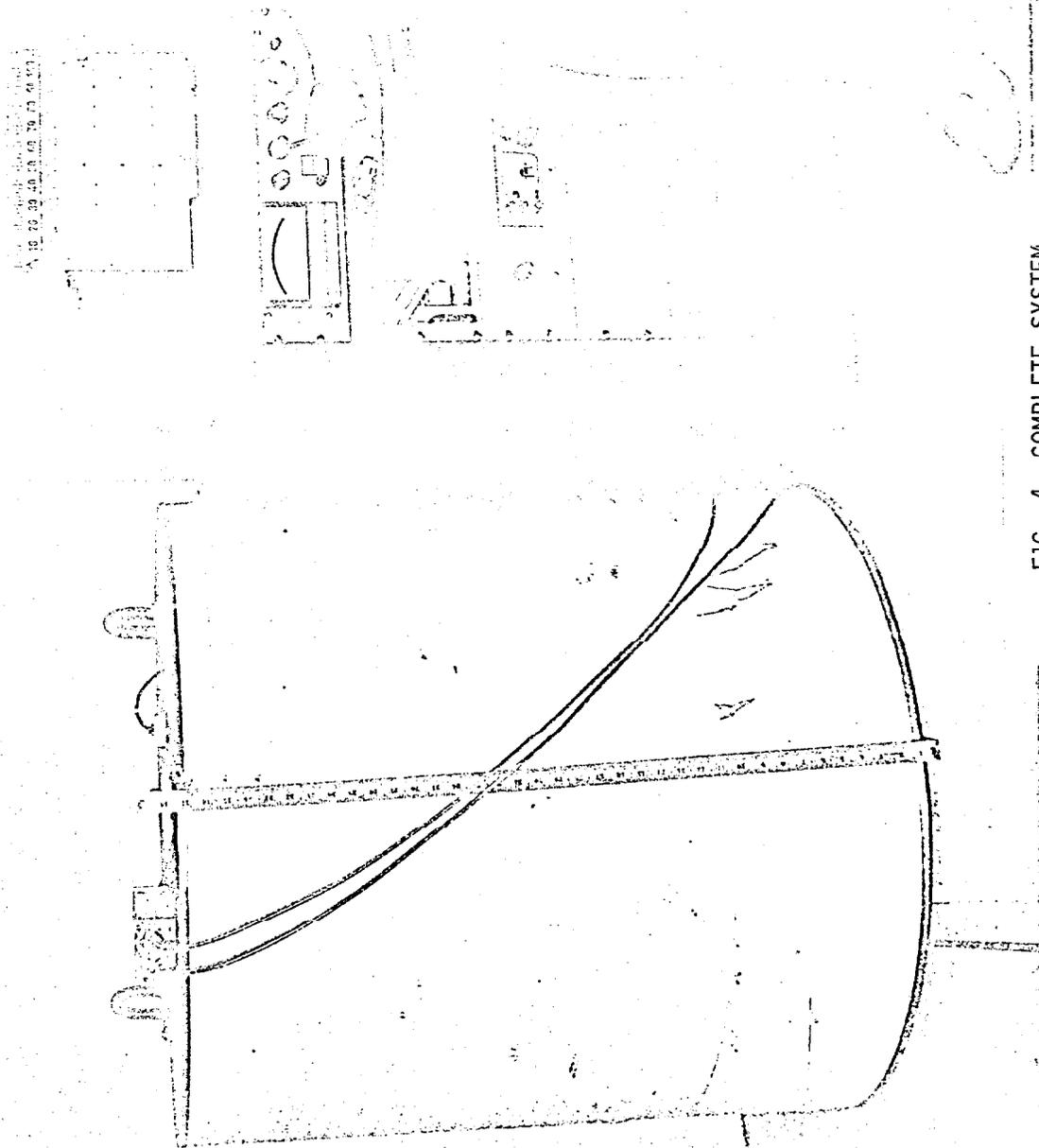


FIG. 4 COMPLETE SYSTEM

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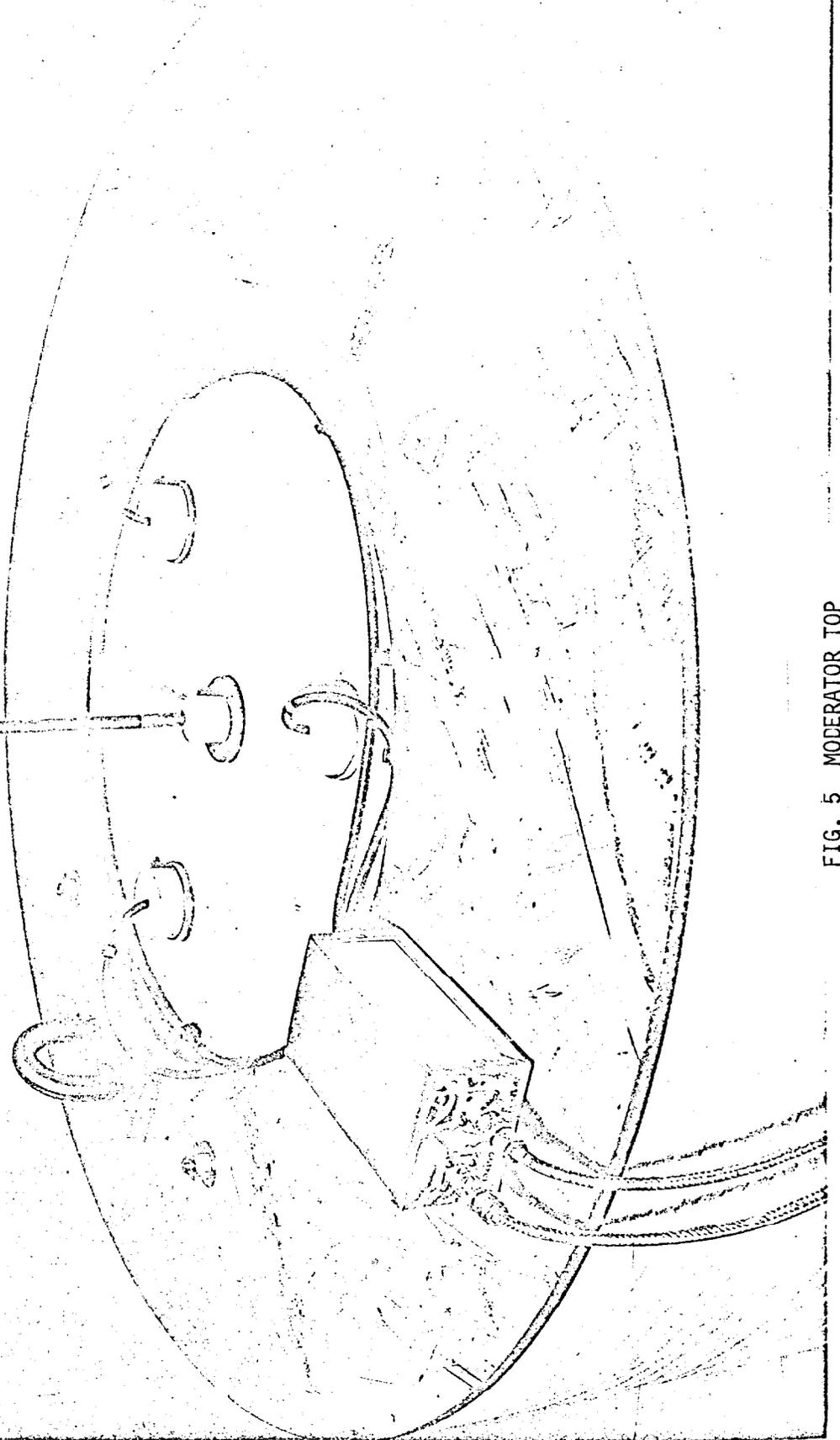
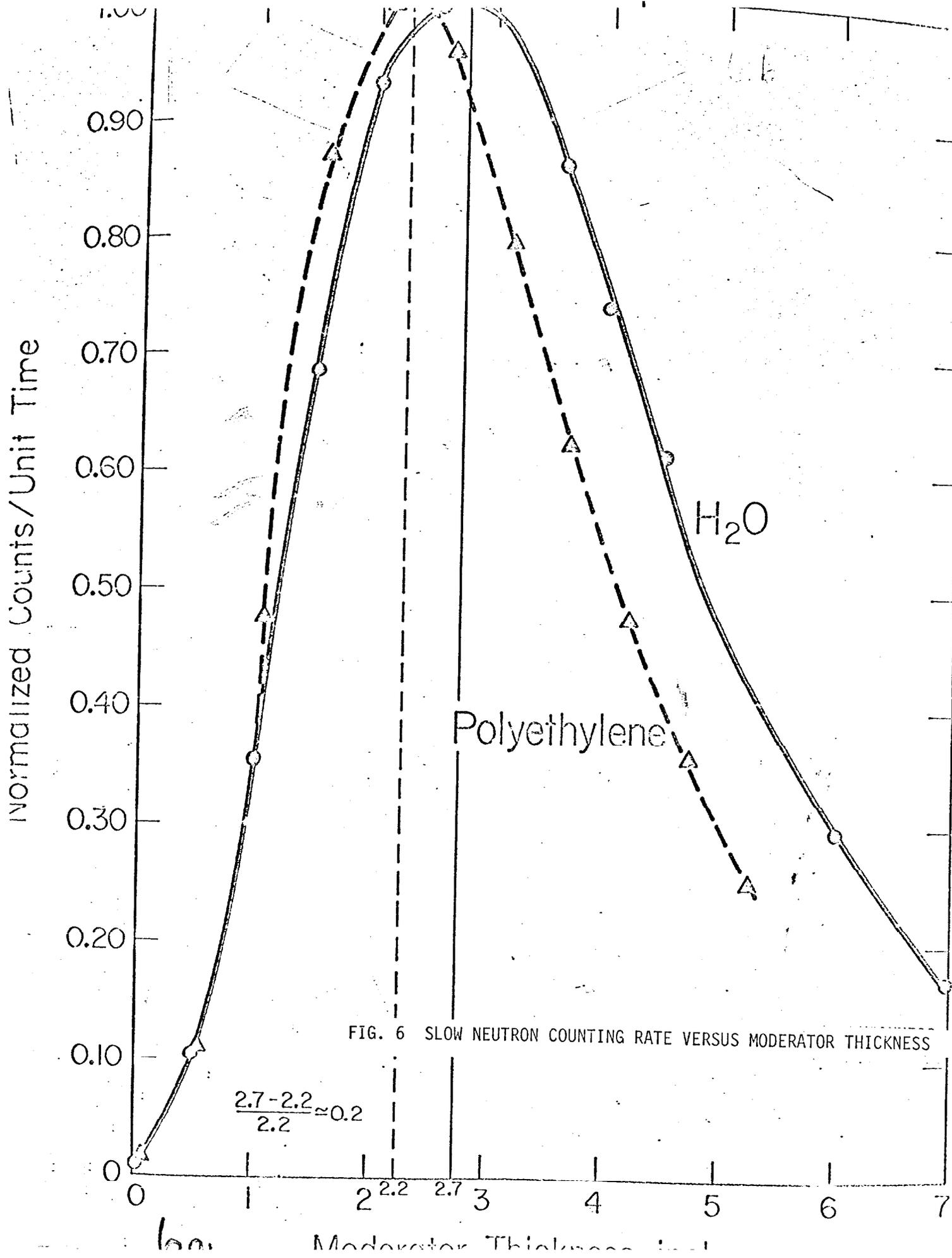
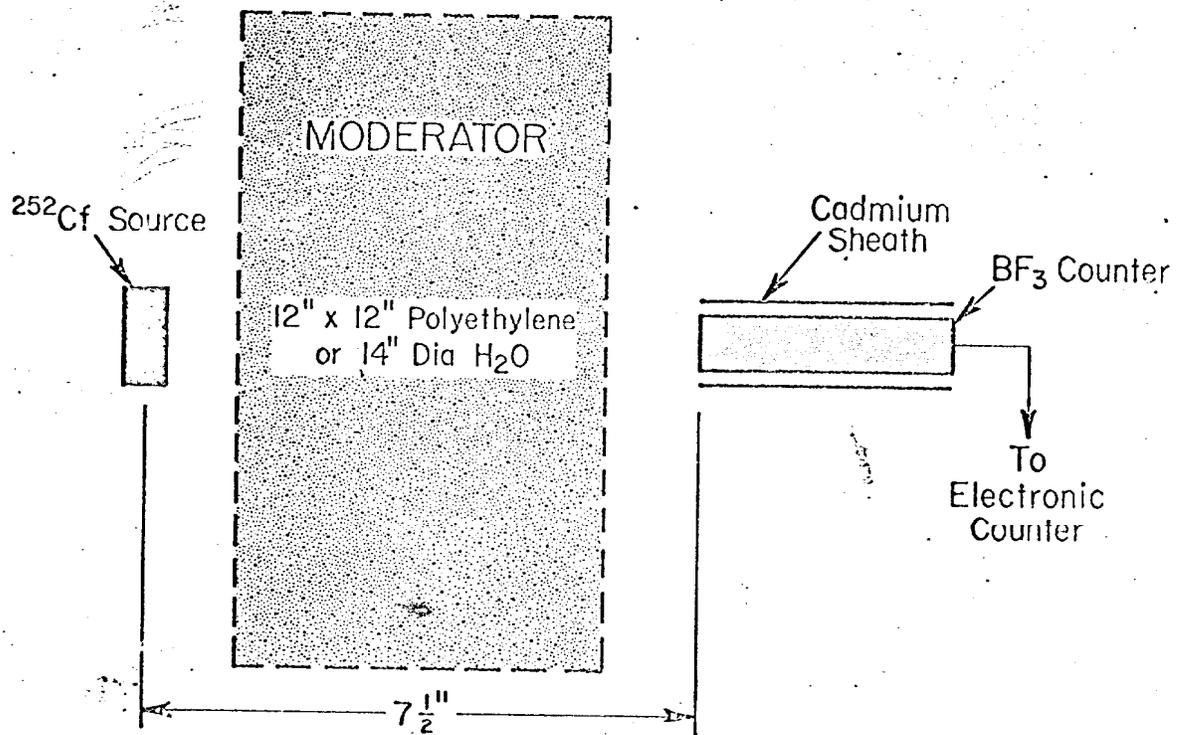
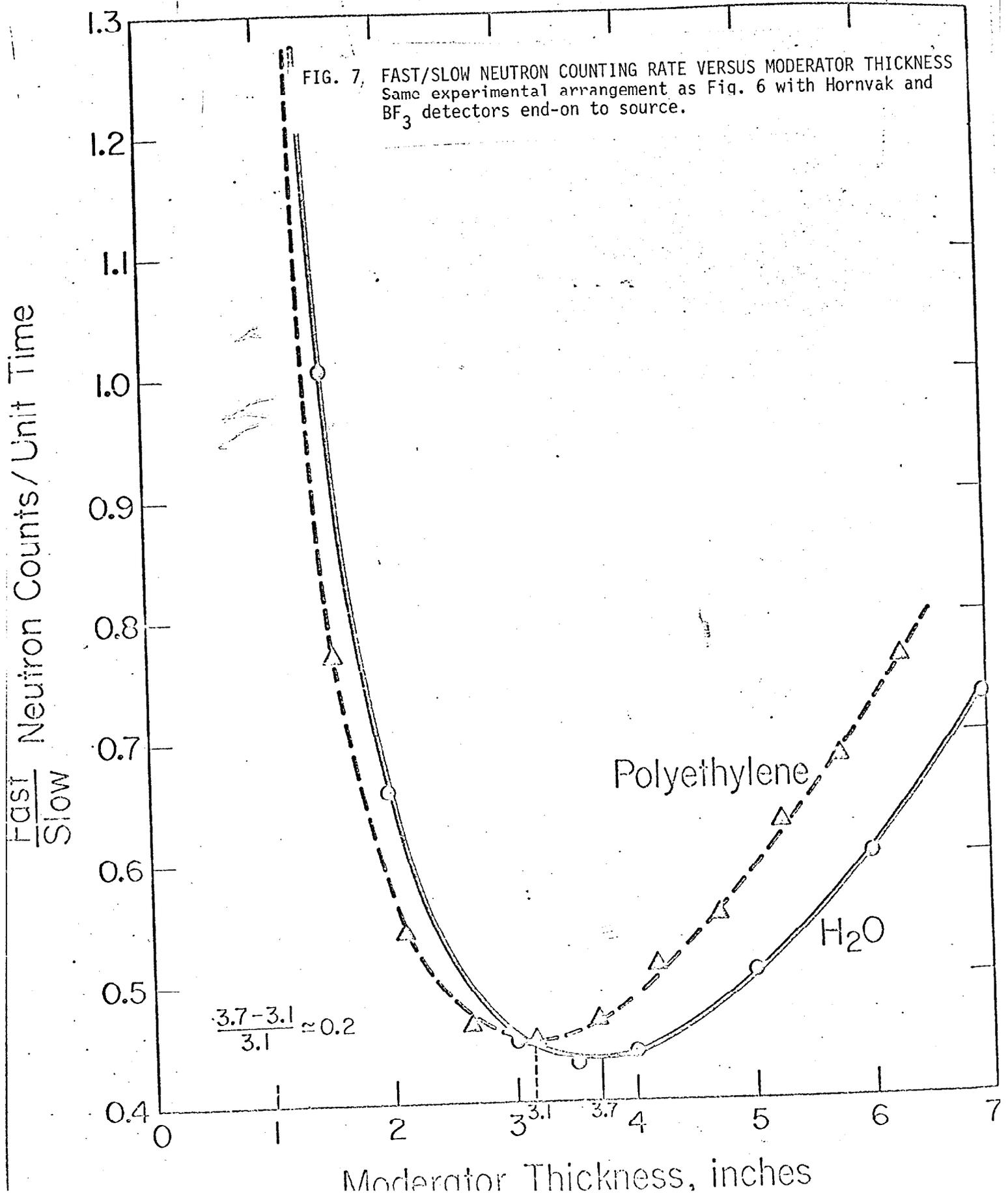


FIG. 5 MODERATOR TOP





66.



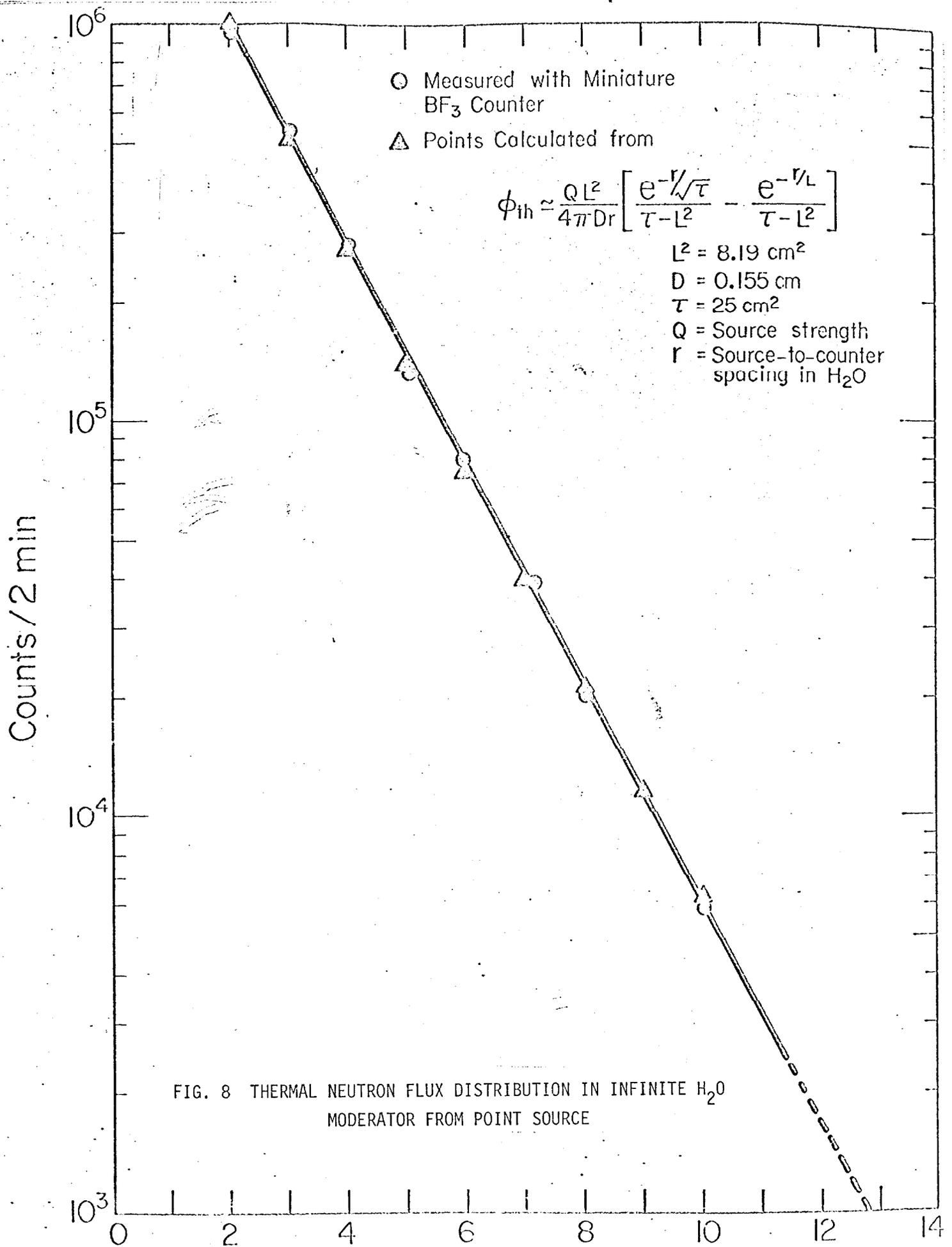


FIG. 8 THERMAL NEUTRON FLUX DISTRIBUTION IN INFINITE H₂O MODERATOR FROM POINT SOURCE

Fission Counter Current, amperes

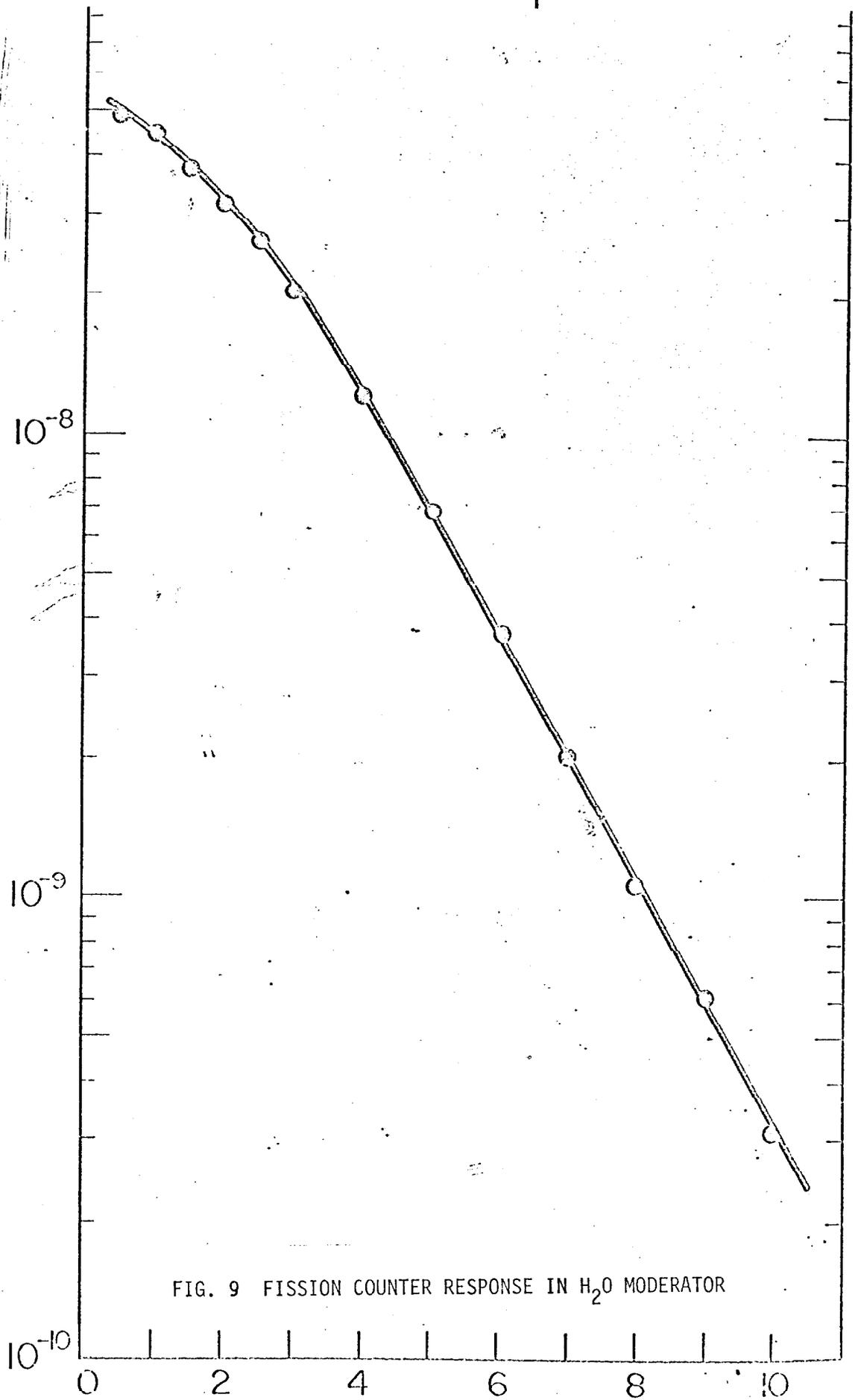
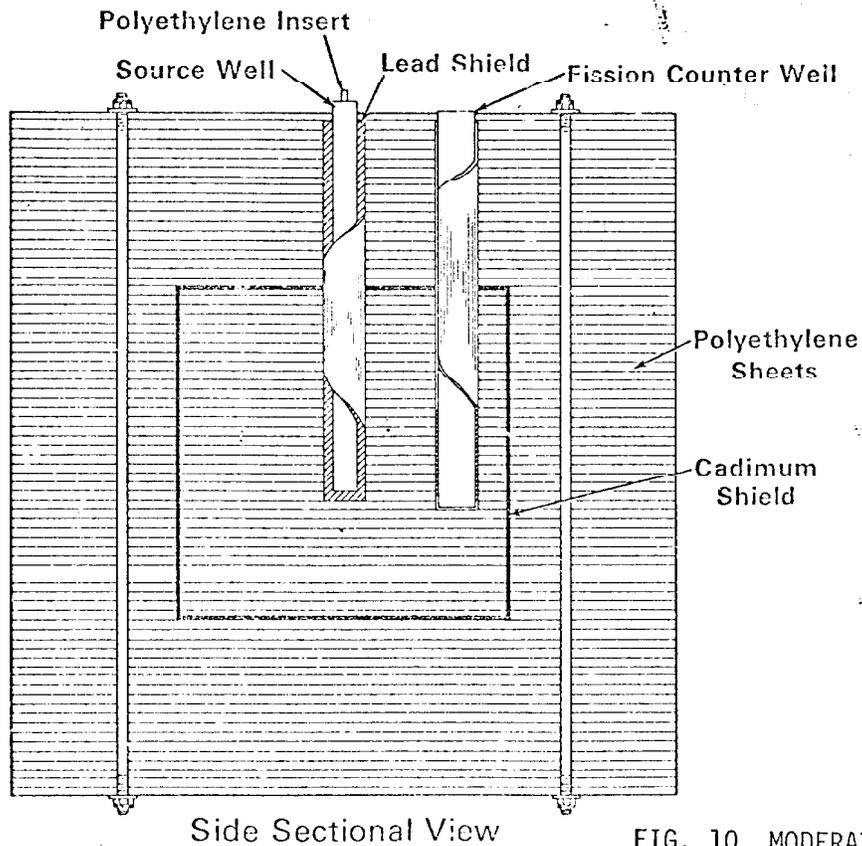
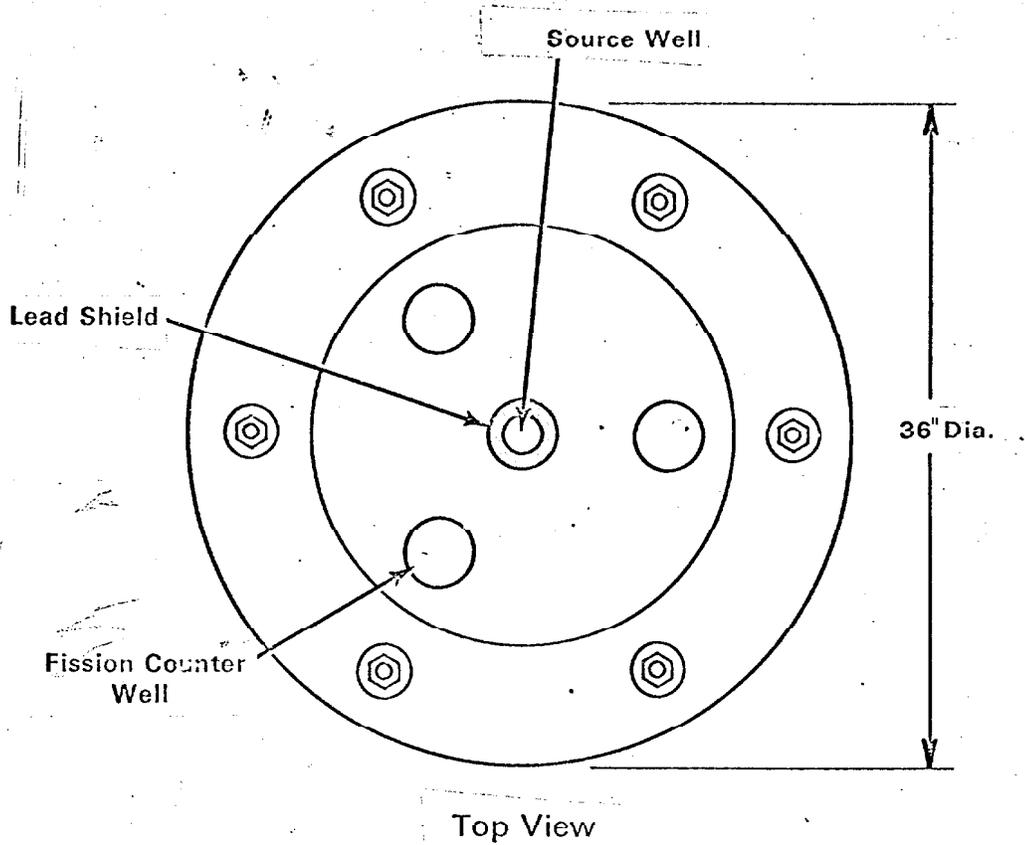


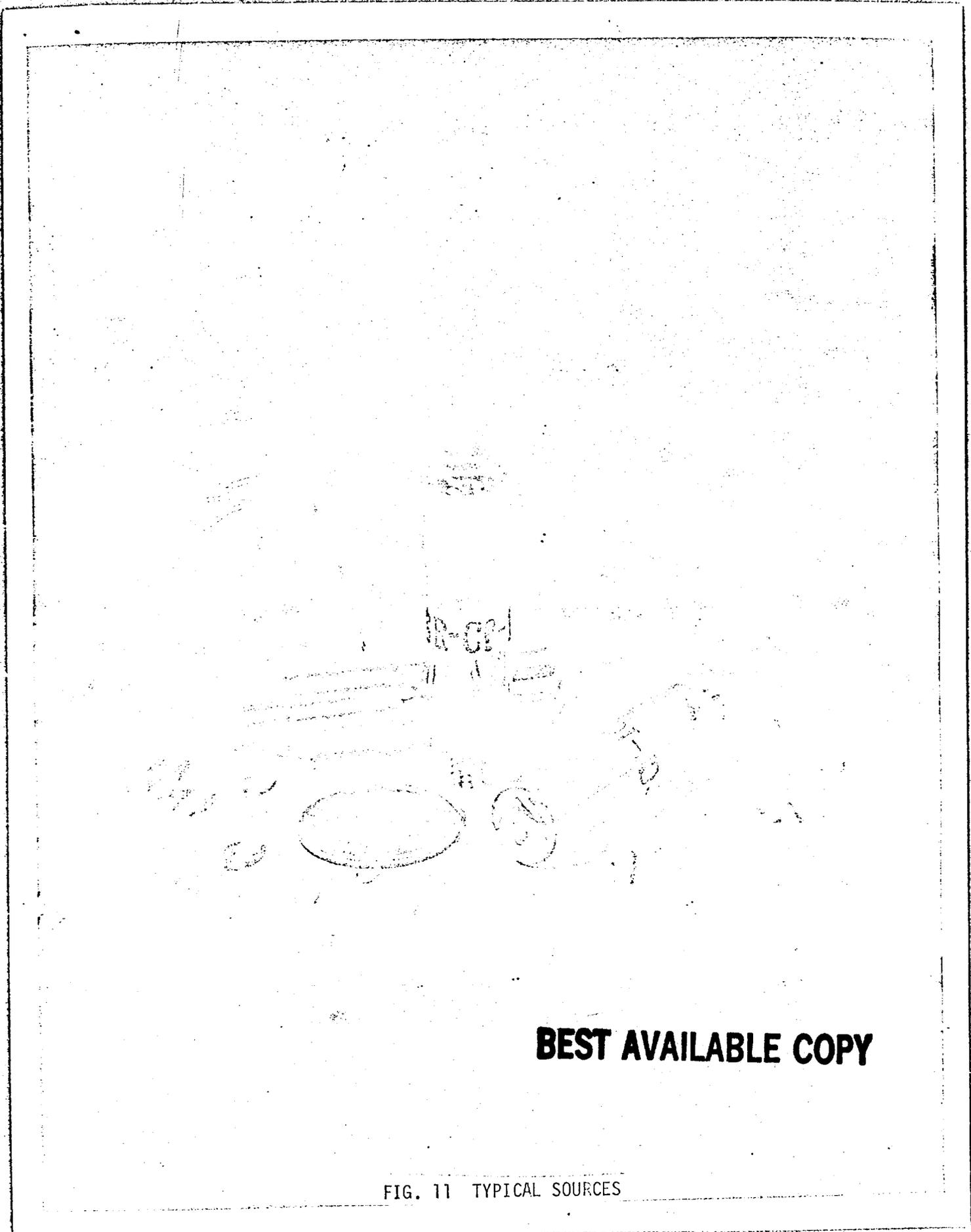
FIG. 9 FISSION COUNTER RESPONSE IN H₂O MODERATOR

Distance between Source and Counter, inches



Side Sectional View

FIG. 10 MODERATOR DESIGN



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FIG. 11 TYPICAL SOURCES