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DP-1035

AEC RESEARCH AND DEVELOPMENT REPORT

A NEUTRON COUNTER FOR ANALYTICAL APPLICATIONS

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DU PONT
REG. U. S. PAT. OFF.

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Aiken, South Carolina

Printed in USA. Price $1.00
Available from the Clearinghouse for Federal Scientific
and Technical Information, National Bureau of Standards,
U. S. Department of Commerce, Springfield, Va. 22151

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Instruments
(TID-4500)

A NEUTRON COUNTER FOR ANALYTICAL APPLICATIONS

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August 1966

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CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION

ABSTRACT

A neutron counter was developed to assay rapidly and nondestructively such radionuclides as ^{244}Cm and ^{252}Cf by counting the spontaneous fission neutrons that these isotopes emit. Neutrons are thermalized in a polyethylene moderator assembly and counted with BF_3 detectors spaced in the moderator. The counter background is 4 counts/minute and the counting efficiency is 4%. The reliable detection limit is 4×10^{-7} g for ^{244}Cm and 1.2×10^{-12} g for ^{252}Cf.

CONTENTS

LIST OF FIGURES

A NEUTRON COUNTER FOR ANALYTICAL APPLICATIONS

INTRODUCTION

The increased production of transplutonium isotopes at the Savannah River Plant created a need for an analytical technique to assay rapidly and nondestructively such radionuclides as ^{244}Cm and ^{252}Cf. Since these two alpha-emitting nuclides also decay by spontaneous fission, neutron counting with an assembly of BF_3 detectors offered a promising method of assay. In addition, because of the interest in neutron emission of other alpha-emitting isotopes, ^{238}Pu and ^{210}Po, a neutron counting system was needed for measurement of neutrons from α,n reactions.

A study was undertaken to design a sensitive neutron counting system capable of accepting a variety of sample types, volumes, and shapes.

SUMMARY

A neutron counter was designed specifically to count fission neutrons with high efficiency and low background. Spontaneous fission neutrons from a ^{244}Cm source were used to define optimum moderator dimensions and source-to-detector spacing. Background levels were minimized by selecting BF_3 detectors with high sensitivity and low noise, and by incorporating an annulus of cadmium in the reflection zone of the polyethylene moderator. The compact assembly, 16 inches in diameter by 16.8 inches in height, has a background of only 4 counts/minute and a counting efficiency of 4% from the ten BF_3 detectors. Reliable detection limits are 4×10^{-7} g for ^{244}Cm and 1.2×10^{-12} g for ^{252}Cf. A removable lead annulus between the sample well and the detectors reduces interference from gamma radiation.

The counter is used to determine ^{252}Cf solutions in which the $^{252}Cf/^{244}Cm$ weight ratios vary from 10^{-4} to 10^{-7}. This nondestructive method for assay of ^{252}Cf is sensitive, precise, and rapid. The counter has been used for process control, in tracer-scale studies of the analytical chemistry of californium, and for assaying neutrons produced from α,n reactions in light elements.

DISCUSSION

DESIGN OF NEUTRON COUNTER

Geometry of Moderator and Detectors

The test arrangement shown in Figure 1 was used to determine optimum source-to-detector and moderator dimensions with a ^{244}Cm standard as a source of spontaneous fission neutrons. The energy spectrum of spontaneous fission neutrons from ^{244}Cm was recently measured at the Savannah River Laboratory[1] and found to be similar to the reported ^{252}Cf spectrum.[2] Water was used as moderator in these tests instead of polyethylene so that the source-to-detector distance could be easily varied. The data in Figure 2 show that the maximum count rate is obtained with 3/4 inch of water between the source and detector.

The relative efficiencies of polyethylene and water in moderating fast neutrons were obtained with the test arrangement shown in Figure 3 by varying the moderator thickness between a fixed, unmoderated neutron source and fixed neutron detectors. The data in Figure 4 show that 17/32 inch of polyethylene produced the same amount of thermalization of source neutrons as did 3/4 inch of water. Polyethylene was selected for the moderator because it moderates fast neutrons with high efficiency and is easily machined and handled. The shorter source-to-detector distance in polyethylene also increases the fraction of source neutrons intercepted by the detectors.

Some of the source neutrons are not thermalized when they reach the BF_3 detector position and will travel past the detector. Figure 4 indicates that the ratio of slow to fast neutrons from the source reaches a plateau at about 3-1/4 inches of polyethylene moderator. An additional 3-1/4 inches of polyethylene moderator is needed beyond the detector position to ensure maximum thermalization of these neutrons and to "reflect" a fraction of them back to the BF_3 detector position to enhance the overall counting efficiency.

Figure 5 shows top and side sectional views of the moderator assembly that was designed on the basis of the above data. The ten BF_3 detectors in the moderator were the maximum number that could be placed at the optimum radius from the sample. The lead annulus surrounding the sample tube was designed to absorb the low energy gamma radiation associated with ^{244}Cm and ^{252}Cf

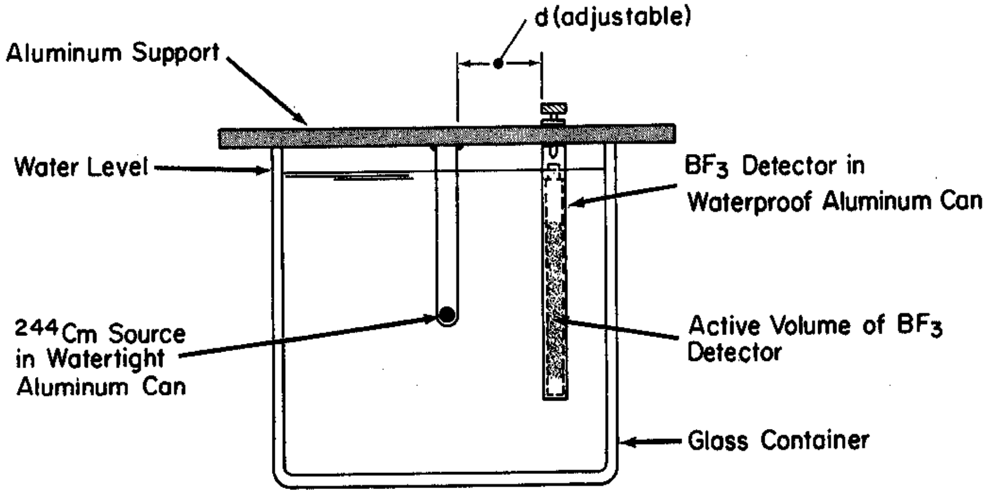


FIG. 1 TEST ARRANGEMENT TO DETERMINE SOURCE-TO-DETECTOR DISTANCE

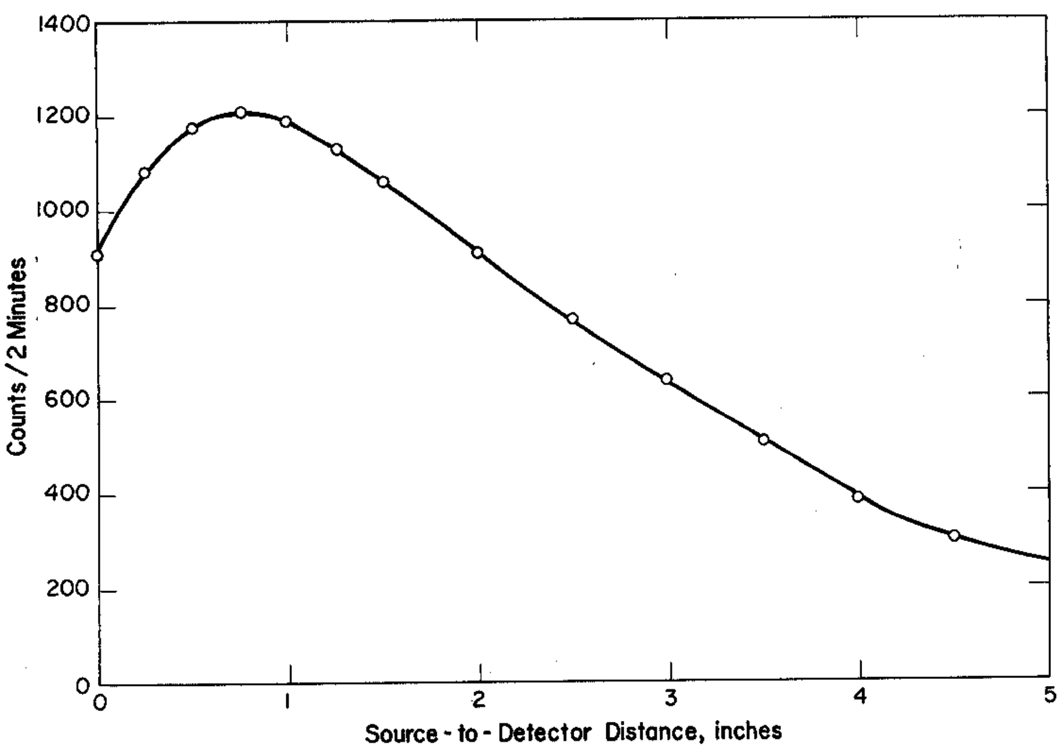
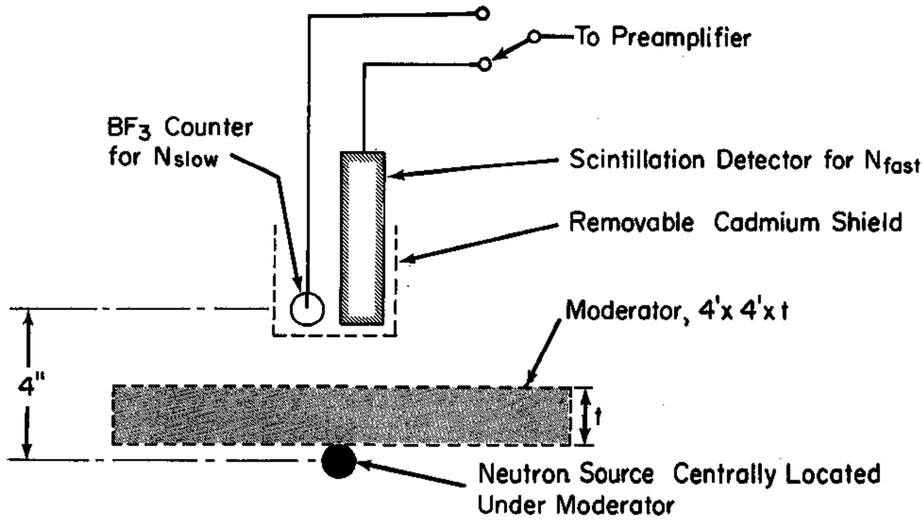


FIG. 2 THERMAL NEUTRON DISTRIBUTION IN THE H_2O MODERATOR



[Counter and Source Positions Fixed During Tests]

FIG. 3 TEST ARRANGEMENT TO DETERMINE MODERATOR EFFICIENCY

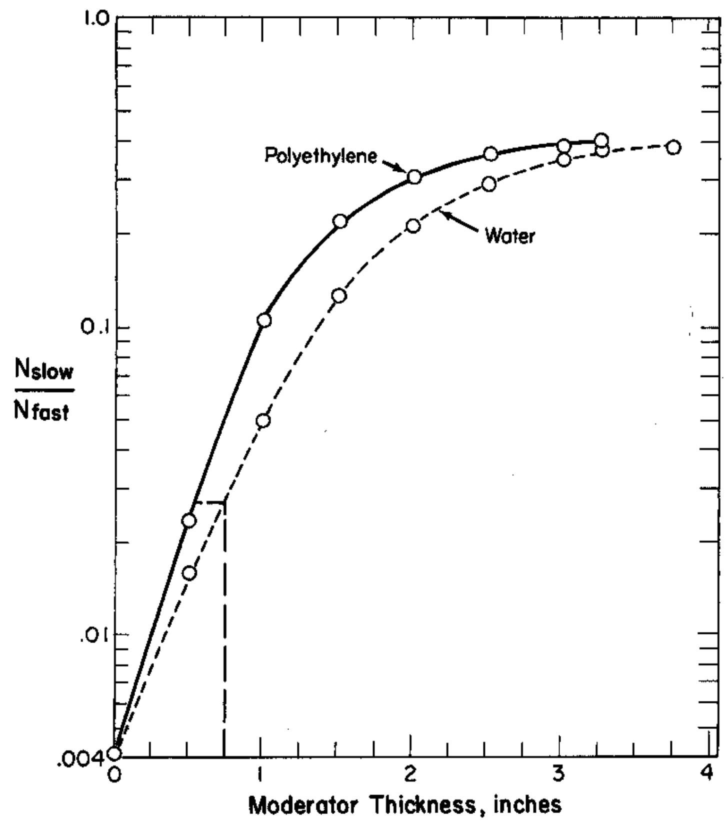


FIG. 4 RELATIVE EFFICIENCIES OF POLYETHYLENE AND WATER AS NEUTRON MODERATORS

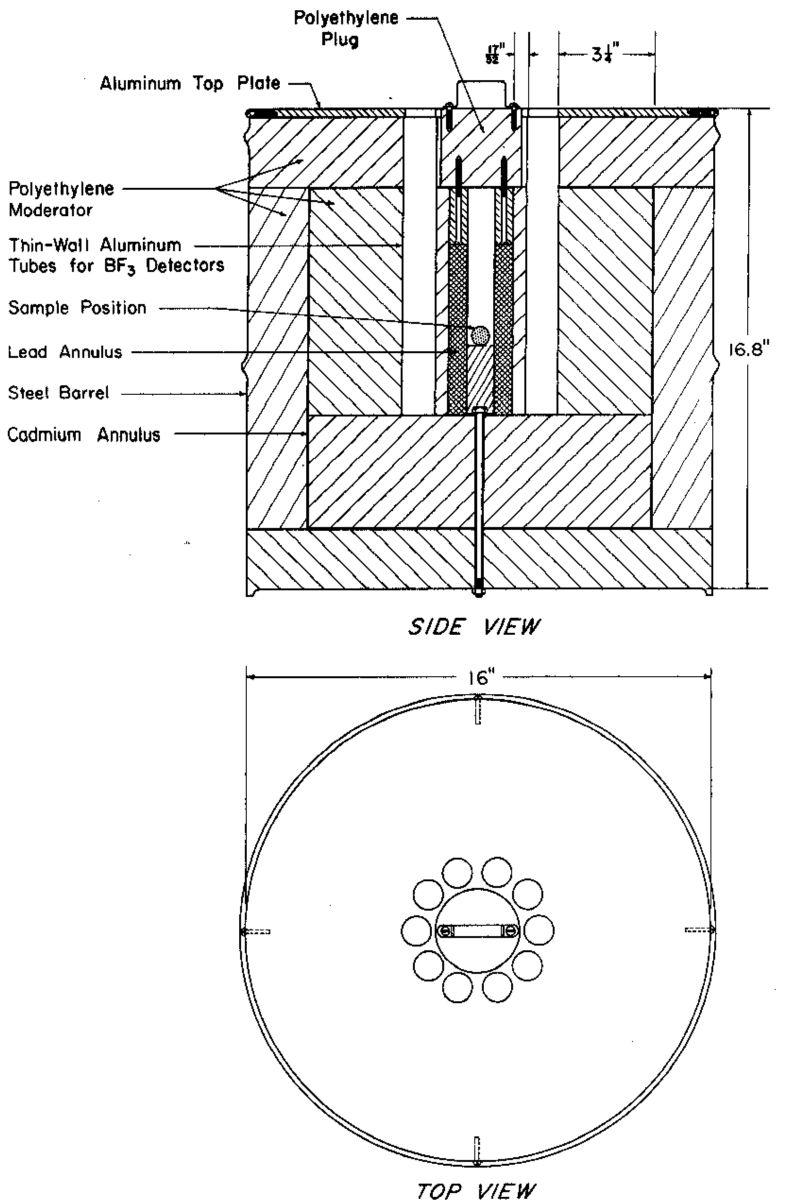


FIG. 5 TOP AND SIDE SECTIONAL VIEWS OF MODERATOR ASSEMBLY

samples. Figure 6 shows how the absorption efficiency of the lead annulus varies with gamma ray energy. Attenuation of the source neutrons by the lead is negligible. The cadmium annulus shown in Figure 5 reduces the background count from external neutrons at the BF_3 detector positions. External neutrons are thermalized in the outer volume of polyethylene and absorbed by the cadmium.

BF_3 Detectors

Commercially available BF_3 detectors were selected on the basis of size, uniformity of response, and sensitivity. Tubes with an active volume 1 inch in diameter and 4-1/2 inches long and filled with enriched BF_3 (96% ^{10}B) to 30 cm Hg pressure were of the proper size to permit an efficient annular arrangement compatible with optimum moderator dimensions. The detectors were arranged in the moderator assembly so that samples could be positioned near the midpoint of the active volumes. The BF_3 detectors were selected from stock to have the same nominal operating high voltage and were then individually tested with a neutron source. The final selection was made on the basis of individual pulse height versus count rate curves, neutron sensitivity, and noise level. Ten acceptable tubes were found among the first fourteen tested.

Electronic Components

Figure 7 is a block diagram of the commercially available electronic components. The ten BF_3 detectors in the moderator tank are connected in electrical parallel. Individual shielded cables connect each detector to a common terminal box located on top of the moderator assembly. A single shielded cable connects the terminal box to the input of the charge-sensitive preamplifier and serves to provide high voltage to the detectors and also return the neutron signals to the preamplifier. The voltage pulses at the preamplifier output are further amplified and fed through the discriminator to the scaler. The time base of the scaler is controlled by the electronic timer.

The electronic system was adjusted so that neutrons were counted in the plateau region of each BF_3 detector. A slope of 1.2% change in count rate per 100-volt change in high voltage was obtained in the plateau region for all ten detectors connected in electrical parallel. Figure 8 shows the discriminator curve, and Figure 9 shows the plateau for the ten detectors operated together.

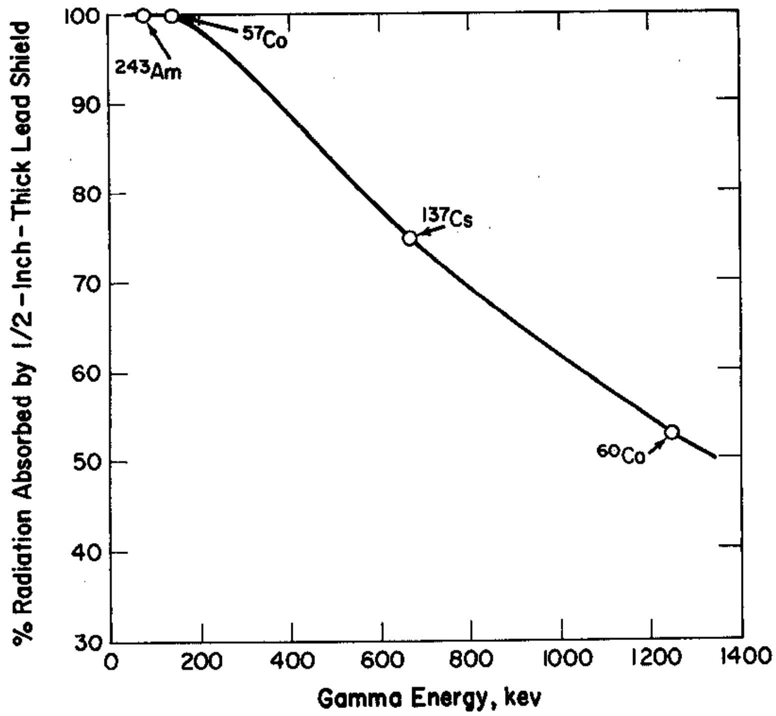


FIG. 6 GAMMA ABSORPTION EFFICIENCY OF LEAD ANNULUS

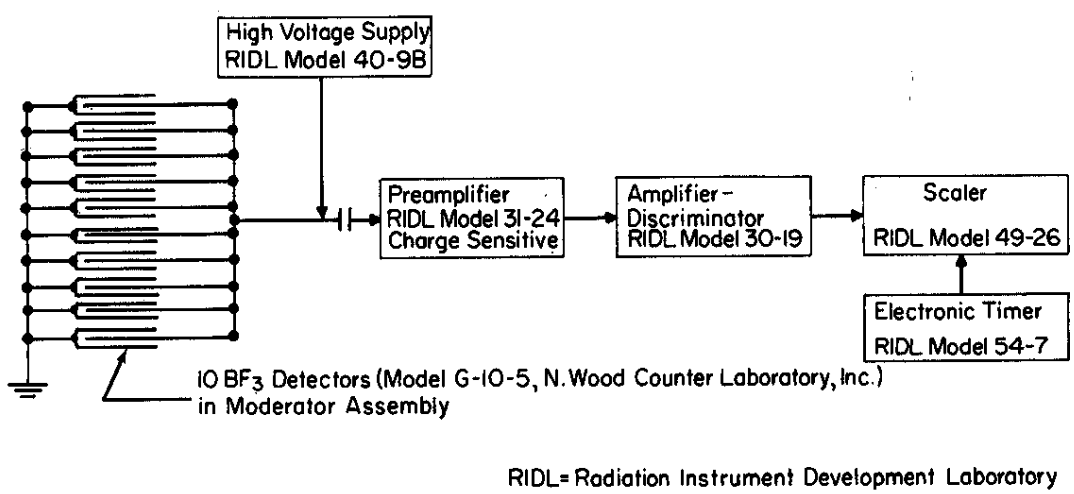


FIG. 7 BLOCK DIAGRAM OF ELECTRONIC COMPONENTS

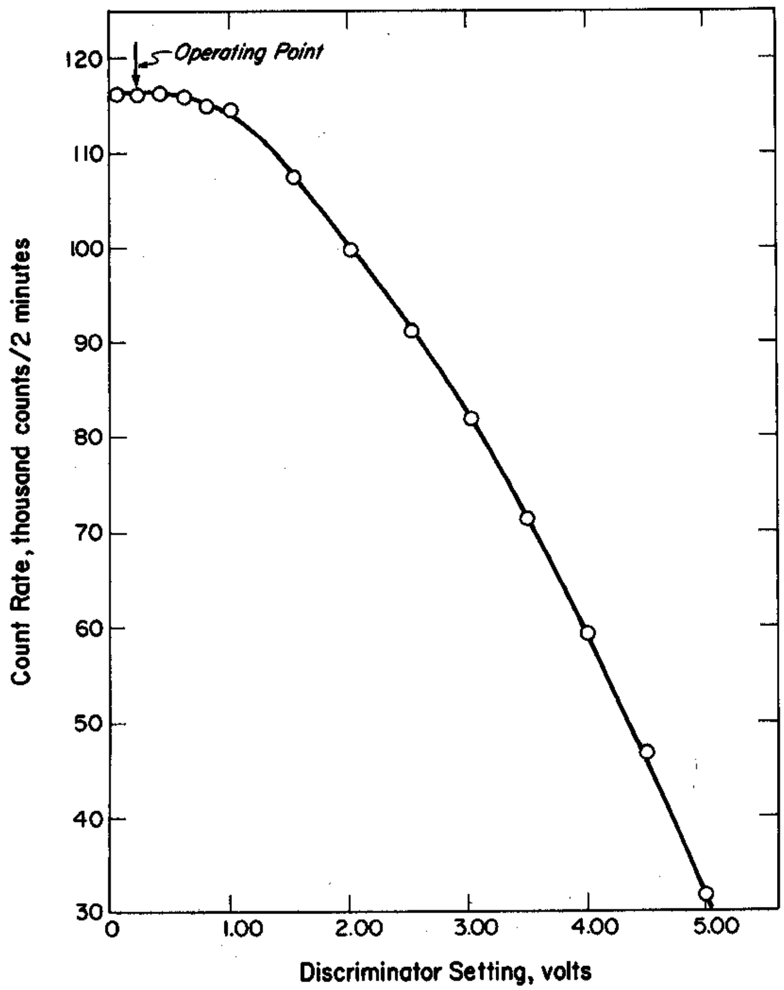


FIG. 8 DISCRIMINATOR CURVE FOR TEN BF_3 DETECTORS IN PARALLEL

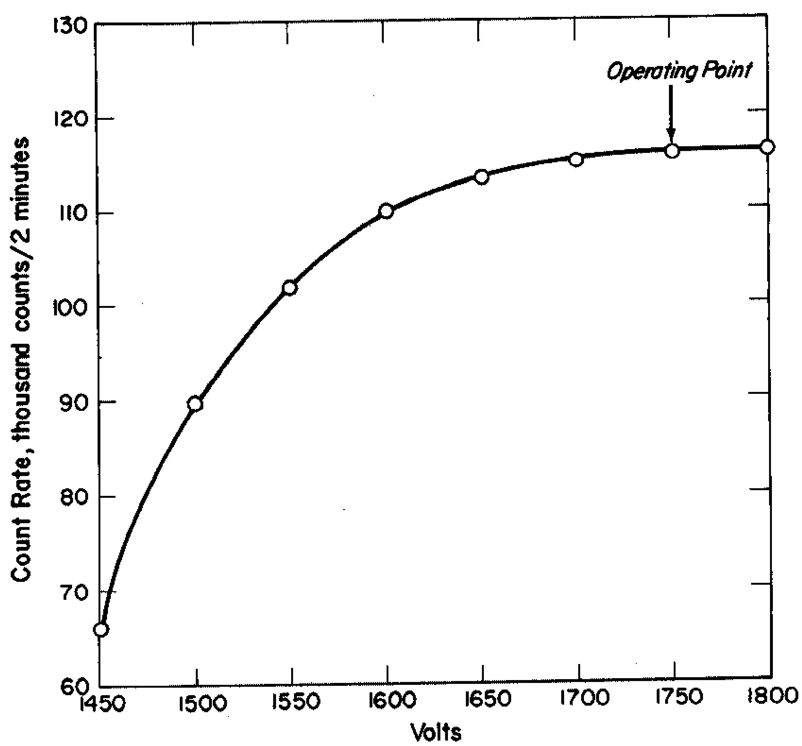


FIG. 9 VOLTAGE PLATEAU FOR TEN BF_3 DETECTORS IN PARALLEL

Description of Assembled Neutron Counter

The neutron counter, pictured in Figure 10, consists of the polyethylene moderator assembly, 16 inches in diameter by 16.8 inches in height, and the associated electronic system. The source or sample to be counted is placed in the sample tube at the center of the moderator assembly. A lead annulus surrounds the sample tube. The ten BF_3 detectors are spaced concentrically in the polyethylene around the lead annulus. Pulses produced by neutron interactions with the BF_3 detectors are fed to the electronic system composed of a preamplifier, amplifier-discriminator, and scaler.

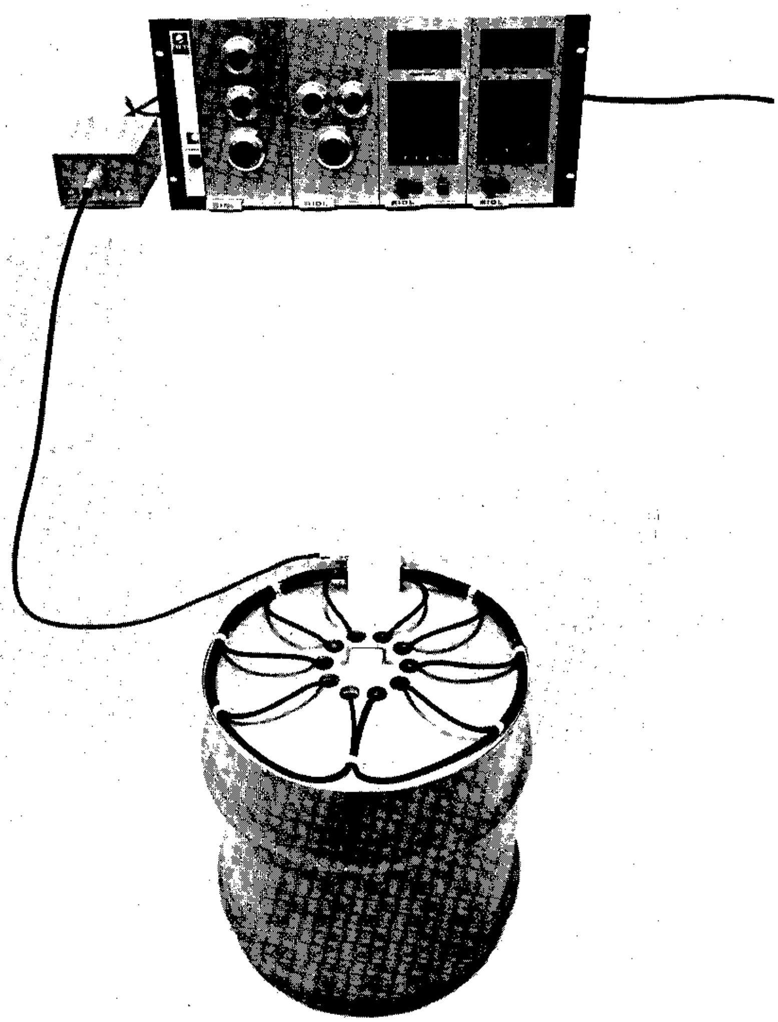


FIG. 10 ASSEMBLED NEUTRON COUNTER

Precision and Calibration

The precision of the counter was determined by a series of twenty counts of two-minutes duration each with a 10-nanogram ^{252}Cf source (1.5×10^6 n/m) in the sample holder. The relative standard deviation of the counts thus obtained was 0.26%.

The stability of the counter was determined by obtaining 30-minute counts of a 0.3-nanogram ^{252}Cf source (5×10^4 n/m) several times throughout each day for eight days, with the electronic system operating continuously and with the source removed from the moderator after each count. The relative standard deviation of the 30 counts was 0.43%.

The efficiency of the neutron counter was 4.06%, based on counting a purified ^{244}Cm standard whose neutron emission rate was determined from alpha counting, alpha pulse height analysis, and the reported value for the ^{244}Cm specific neutron activity.[3]

ANALYTICAL APPLICATIONS

Curium Purification Process

The neutron counter has been used primarily to follow the performance of the curium purification process,[4] which was designed to separate actinides from lanthanides by solvent extraction of the former from a high concentration of LiCl into an organic phase containing a high-molecular-weight tertiary amine. This process has been undergoing development and testing at the Savannah River Laboratory as part of the program to produce 4.5 kg of ^{244}Cm. In recent tests of this process with miniature mixer-settlers, a californium rejection step was incorporated to lower the neutron emission rate of the final curium product. From these tests, a total of 127 process samples were analyzed nondestructively for ^{252}Cf by counting the spontaneous fission neutrons. Also, 16 stage samples were analyzed for ^{252}Cf to determine its behavior in the curium stripping bank. In all cases where ^{244}Cm and ^{252}Cf were present in the same sample, a correction derived from alpha counting was made for neutrons from the spontaneous fission of ^{244}Cm. In most of the samples the neutrons from ^{252}Cf were at least 40% of the total neutron count. Each sample required a total of approximately two man-hours for the nondestructive ^{252}Cf analysis, whereas determination by the usual ion exchange chromatographic separation[5] followed by alpha counting and pulse height analysis would require at least 20 man-hours. Analysis of ^{252}Cf by the measurement of prompt gamma rays from spontaneous fission[6] was not possible on many of the samples since the relatively high

(>50 mrad/hr) beta-gamma activity present from fission products would interfere with the gamma counting.

Several curium process samples were analyzed by the ion exchange chromatographic separation, as well as by neutron counting. The results are compared in the following table.

Comparison of ^{252}Cf Analyses

Sample	^{252}Cf Concentration, dis/min-ml: Neutron Counting	^{252}Cf Concentration, dis/min-ml: Ion Exchange and Alpha Counting	% Difference
1	3.8×10^5	3.7×10^5	2.6
2	4.6×10^5	5.0×10^5	-8.7
3	4.8×10^5	4.2×10^5	12
4	5.7×10^5	5.3×10^5	7.0
5	6.1×10^5	5.4×10^5	11
6	5.8×10^5	5.0×10^5	14
7	6.3×10^5	5.1×10^5	19
8	7.5×10^5	8.2×10^5	-9.3
9	3.4×10^5	3.1×10^5	8.8
10	9.8×10^4	9.8×10^4	0
11	4.0×10^4	5.1×10^4	-28
12	2.6×10^4	2.1×10^4	19
13	5.3×10^5	6.0×10^5	-13
14	6.3×10^3	3.7×10^3	41
		Avg (excluding Sample 14 and disregarding signs):	12

The general agreement between the two independent methods is quite satisfactory considering the various sources of error. Excluding sample 14 in which ^{252}Cf contributed only 0.6% of the total neutrons, the average per cent difference (sign disregarded) between the two methods was 12%. In three (11-13) of the five samples where the per cent difference was greater than 12%, the neutrons from ^{252}Cf spontaneous fission were only a small percentage (from 5 to 25%) of the total neutrons from the sample. When the test of paired replicates was applied to the data, no evidence of bias between the two methods was apparent at the 95% confidence level.

Chemistry of Californium

The neutron counter has been used to determine tracer amounts of ^{252}Cf in studies of its chemical behavior in highly salted systems containing both a solid and a liquid phase. To determine californium by alpha counting in such systems, dilution of the liquid phase by a factor of 10^3 to 10^4 would be necessary. Such dilutions would necessitate use of a major portion of the current supply of californium tracer for a single experiment. The solid phase would require dissolution and mounting to determine the californium content. However, since either the entire liquid or solid phase could be used for neutron counting, only 1/20 of the californium tracer stock solution was needed per experiment, and therefore, time-consuming purification of the tracer between experiments was eliminated.

Determination of Neutrons from α,n Reactions

Neutrons from α,n reactions in the following systems have been determined with the neutron counter:

- A ^{228}Th source prepared for use in the determination of deuterium by γ,n reaction
- ^{238}Pu solutions and compounds containing ^{17}O and ^{18}O
- ^{210}Po solutions

Other Applications

The effect of α,n reactions from ^{244}Cm on the determination of ^{252}Cf in mixtures of these two nuclides is also being determined. In this study, alpha emitters with low spontaneous fission rates, as well as ^{244}Cm, will be used to study the effects of the source matrix (e.g., salt concentration, anion influence) on the α,n reaction. The neutron counter should be especially useful in these studies because of its low background and constant counting efficiency.

The counter will be used in continuing support of process tests and of studies of californium chemistry. Also, the instrument will be used extensively as part of the analytical control instrumentation for the forthcoming separation of 4.5 kg of ^{244}Cm.

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EXTERNAL RELEASE OF TECHNICAL INFORMATION

Description of Material   No. DP-1035   Date: 8/10/66

Title: A Neutron Counter for Analytical Applications

Author: T. R. Herold and H. P. Holcomb

Type of Material

Classified DP Report [ ]   Classified Paper [ ]

Unclassified DP Report [x]   Unclassified Paper [ ]

Letter [ ]

Technical Content

Approved by /s/ E. L. Albenesius   Date: 8/3/66

Classification

Approved by S. W. O'Rear /HSH   Date: 8/10/66
S. W. O'Rear

Authority:

Topic 1.1 CG-UF-2

Topics 151.5.1, 151.5.8, and 151.5.10 SROO Classification Guide

SROO Classification Notice 31

Category if DP Report

Approved by S. W. O'Rear /HSH   Date: 8/10/66
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Final Du Pont Release

Approved by [signature]   Date: 8/22/66
Coordinating Organization Director

Released by R. G. Erdley: 8/15/66