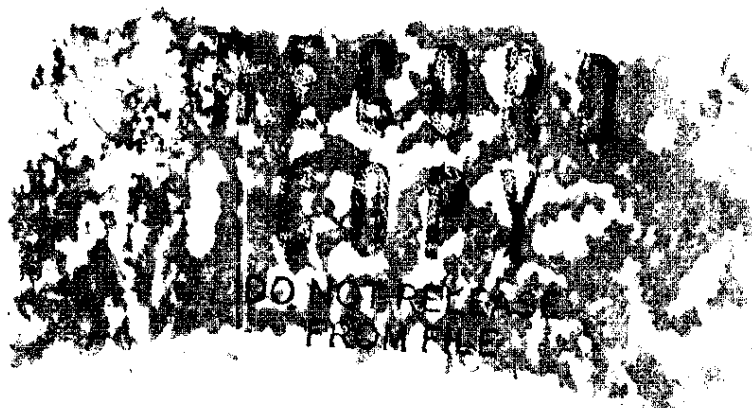


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AEC RESEARCH AND DEVELOPMENT REPORT

ENERGY SPECTRA OF NEUTRONS AND GAMMA RAYS FROM SPONTANEOUS FISSION OF ^{244}Cm

T. R. HEROLD



Savannah River Laboratory

Aiken, South Carolina

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**ENERGY SPECTRA OF NEUTRONS AND GAMMA RAYS
FROM SPONTANEOUS FISSION OF ^{244}Cm**

by

T. Richard Herold

Approved by

D. E. Waters, Manager
Laboratory Operations and Services Division

July 1965

E. I. DU PONT DE NEMOURS & COMPANY
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ABSTRACT

The energy spectrum of the neutrons from the spontaneous fission of ^{244}Cm was measured from 0.5 to 6 Mev with a solid state ^3He spectrometer. The spectrum shows a maximum at about 0.75 Mev and decreases exponentially above 2 Mev.

The energy spectrum of the gamma rays that were emitted within 180 nanoseconds of the spontaneous fission neutrons was measured from 0.1 to 2.5 Mev with a gamma ray spectrometer that included the ^3He spectrometer in a coincidence circuit. The gamma ray spectrum shows a dual-peak maximum at about 200 kev and decreases exponentially above 800 kev.

Both measured spectra are similar to the corresponding spectra from the spontaneous fission of ^{252}Cf .

The measurements were made with a purified sample of 95.5 wt % ^{244}Cm that was produced in a Savannah River reactor.

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ENERGY SPECTRA OF NEUTRONS AND GAMMA RAYS FROM SPONTANEOUS FISSION OF ^{244}Cm

INTRODUCTION

Curium-244 is an alpha emitter with a half-life of about 19 years and with a power density about five times that of ^{238}Pu . The high specific power and relatively long half-life of ^{244}Cm makes it especially attractive as a long-term nuclear heat source.

A pilot production program for ^{244}Cm is under way at the Savannah River Plant⁽¹⁾ to demonstrate that the isotope can be produced in relatively large quantities at reasonable cost. The program will also provide three kilograms of ^{244}Cm for the development and demonstration of nuclear electric power generators fueled with this isotope.

In addition to emitting alpha particles, ^{244}Cm fissions spontaneously and therefore requires protective shielding to attenuate the fast neutrons and hard gamma rays from the fission process. The present work was undertaken to measure the energy spectra of neutrons and gamma rays from the spontaneous fission of ^{244}Cm in order to provide an empirical basis for shielding calculations. The work is of general scientific interest because these spectra have not been previously reported in the literature. Some preliminary work on the neutron spectrum was done by the author at the Savannah River Laboratory in August 1964 for inclusion in a report on the radiation characteristics of ^{244}Cm .⁽²⁾

The neutron spectrum was measured with a solid state ^3He neutron spectrometer that records nuclear events in ^3He gas. This technique for determining neutron spectra is new and provides a significant improvement over the time-of-flight techniques and the proton-recoil-in-emulsion techniques previously used. The gamma spectrum was measured with a gamma ray spectrometer that included the ^3He neutron spectrometer in a (γ, n) coincidence circuit.

SAMPLES

The ^{244}Cm sample was prepared by dissolving a 7.1-mg sample of purified curium⁽³⁾ in 2.2 ml of HNO_3 (1M) and placing the resulting solution in a small glass pipet. Mass spectrometric analyses of the sample showed that it contained 95.5 wt % ^{244}Cm , 2.7 wt % ^{246}Cm , 1.6 wt % ^{245}Cm , 0.12 wt % ^{242}Cm , 0.04 wt % ^{247}Cm , 0.04 wt % ^{248}Cm , 0.03 wt % ^{243}Cm , and trace amounts of ^{243}Am and ^{252}Cf . Most of the neutrons emitted from the sample derive from the spontaneous fission of

^{244}Cm , with minor contributions from the spontaneous fission of ^{242}Cm , ^{246}Cm , and ^{252}Cf . A second minor source of neutrons is from the reaction of alpha particles with light elements, for example, the (α, n) reaction with oxygen in CmO_2 .

About 1.2 nanograms of ^{252}Cf was obtained from the curium purification. The ^{252}Cf sample was radiochemically free of other alpha- and gamma-emitting nuclides. The fission gamma spectrum of this sample was measured for comparison with the similar ^{244}Cm spectrum measured and with the ^{252}Cf spectrum recently reported by another laboratory. (12)

NEUTRON SPECTRUM

The spectrum of spontaneous fission neutrons from ^{244}Cm was measured with the ^3He neutron spectrometer and associated electronic equipment described in the Appendix, which also contains details of the instrument checkout and calibration.

The ^{244}Cm sample was taped tightly to the ^3He spectrometer, which was covered with 0.030 inch of cadmium to shield the detectors from thermal neutrons. The spectrometer and sample were then suspended from the laboratory ceiling so that the spectrometer-sample assembly was at least 5 ft away from any large scattering object.

The neutron spectrum of ^{244}Cm was obtained in energy increments of approximately 0.5 to 1.1 Mev, 1.0 to 2.2 Mev, 2.0 to 3.5 Mev, and 3.0 to 6.0 Mev, with the biased amplifier technique described in the Appendix. The data were normalized to produce the spectrum shown in Figure 1. The spectrum shows a maximum at about 0.75 Mev and decreases exponentially above 2 Mev. This spectrum has been corrected for background. Background counts in each energy increment were obtained by separating the spectrometer and sample by 6 inches to reduce the direct count rate from the sample by about 99%. Signal-minus-background counts were corrected for the variation in the $^3\text{He}(n, p)$ cross section with neutron energy, with the curve shown in Figure 2, which represents the average of data reported by several investigators. (4-7)

Figure 3 shows the ^{244}Cm spectrum along with the reported experimental spectra of ^{233}U , (8) ^{235}U , (9) ^{239}Pu , (10) and ^{252}Cf (11) for comparison. The normalization of the curves shown in Figure 3 was chosen for clear representation and is otherwise arbitrary.

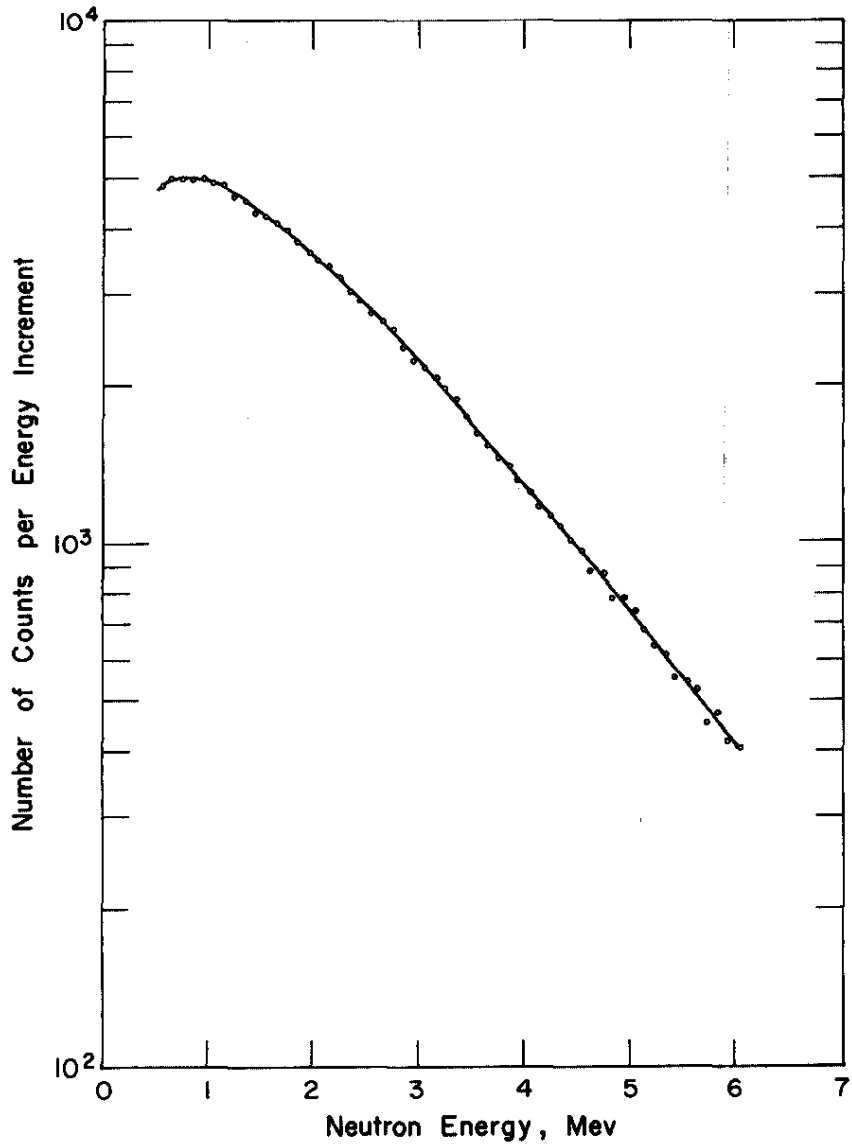


FIG. 1 SPONTANEOUS FISSION NEUTRON SPECTRUM OF ^{244}Cm

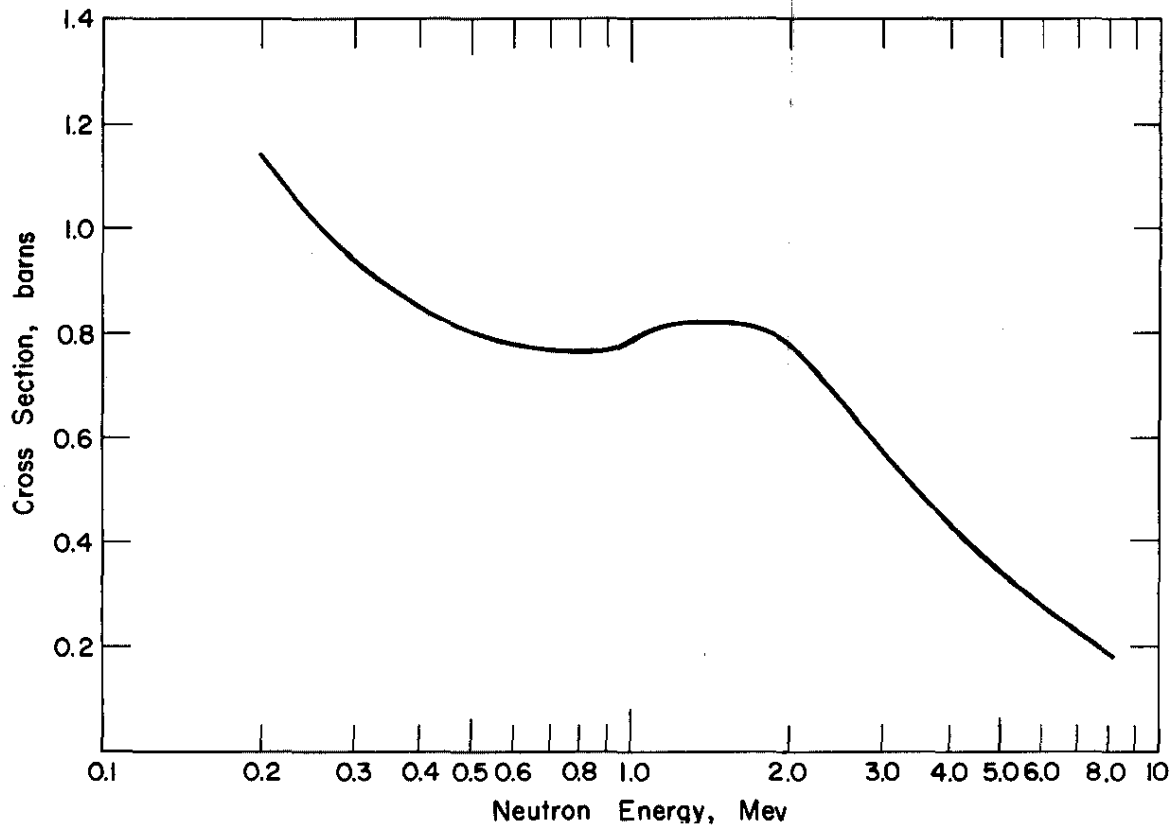


FIG. 2 NEUTRON CROSS SECTION FOR THE ${}^3\text{He}(n,p)$ REACTION

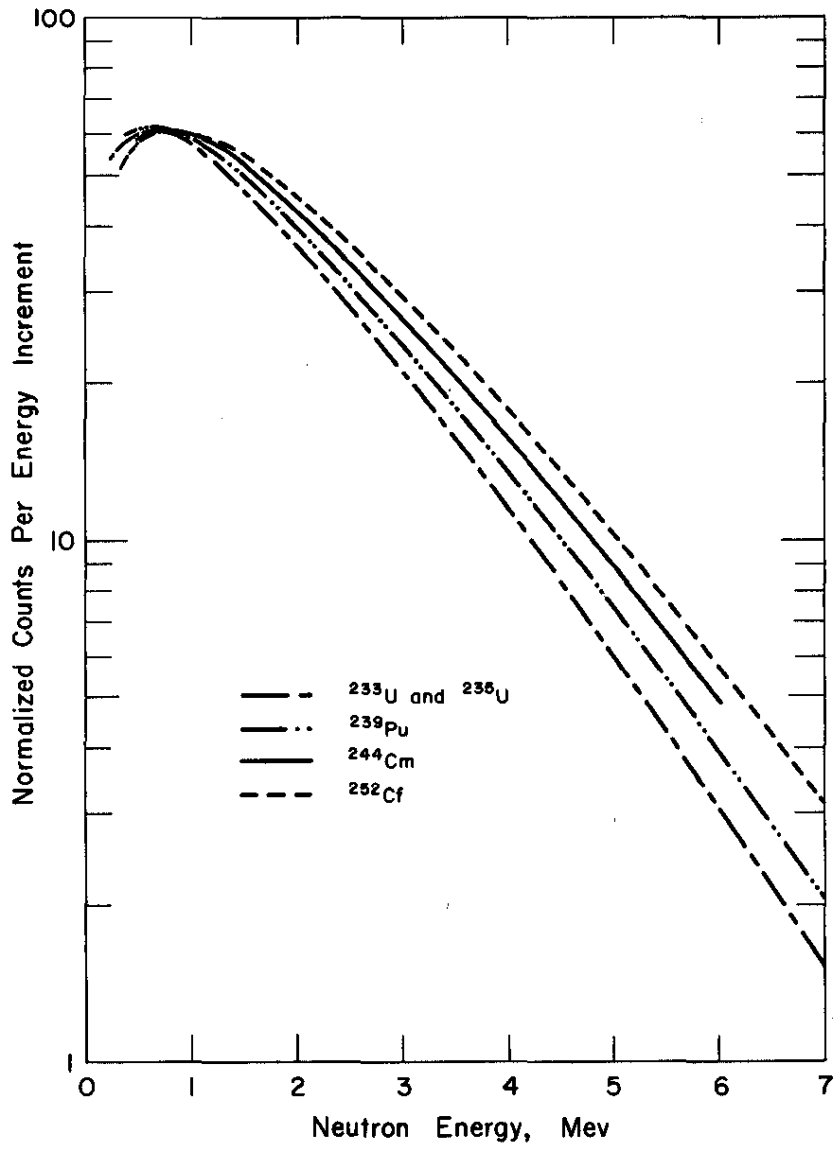


FIG. 3 FISSION NEUTRON SPECTRA

GAMMA SPECTRUM

The ^3He neutron spectrometer and a conventional NaI(Tl) gamma spectrometer were arranged in a (γ, n) coincidence circuit to record the energy spectrum of gamma rays that were emitted within 180 nanoseconds of spontaneous fission neutrons. This equipment is described in the Appendix.

The ^{244}Cm sample was taped snugly to the cadmium-covered ^3He neutron spectrometer as it was for the neutron spectrum measurements. The sample also rested on top of the 3-inch NaI(Tl) crystal of the scintillation detector.

Figure 4 shows the measured gamma energy spectrum of ^{244}Cm and also shows the ^{252}Cf spectrum measured by the same method. Both spectra

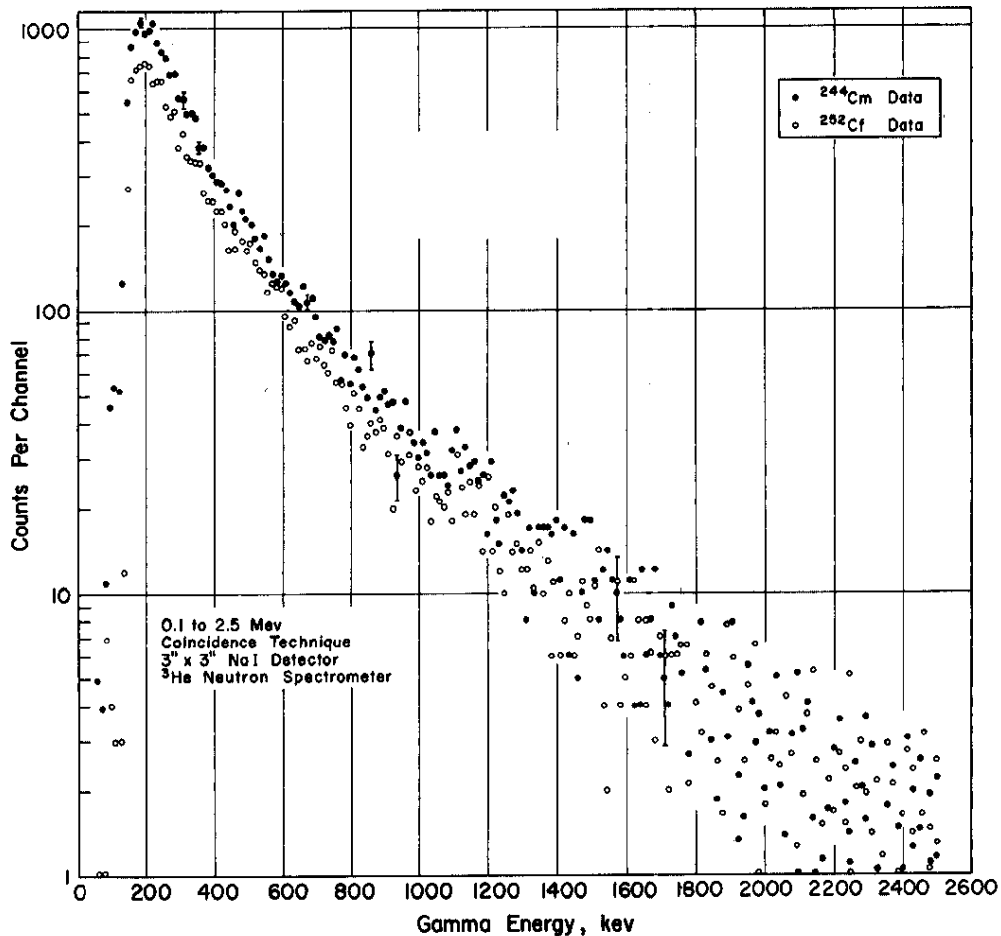


FIG. 4 GAMMA SPECTRA OF ^{244}Cm AND ^{252}Cf

exhibit a dual-peak maximum at about 200 kev and decrease exponentially above 800 kev. In these complex spectra, there is some evidence of peaks which would possibly become statistically significant if more counts per energy interval were accumulated.

Figure 5 shows the ^{244}Cm and ^{252}Cf spectra that resulted from correcting the raw data of Figure 4 for detector efficiency and fitting them to a smooth curve. Figure 6, a plot of NaI(Tl) detector efficiency versus gamma energy, was used to make the corrections.

Figure 5 also shows for comparison the total prompt gamma energy spectrum from ^{252}Cf that was recently measured by another technique. (12)

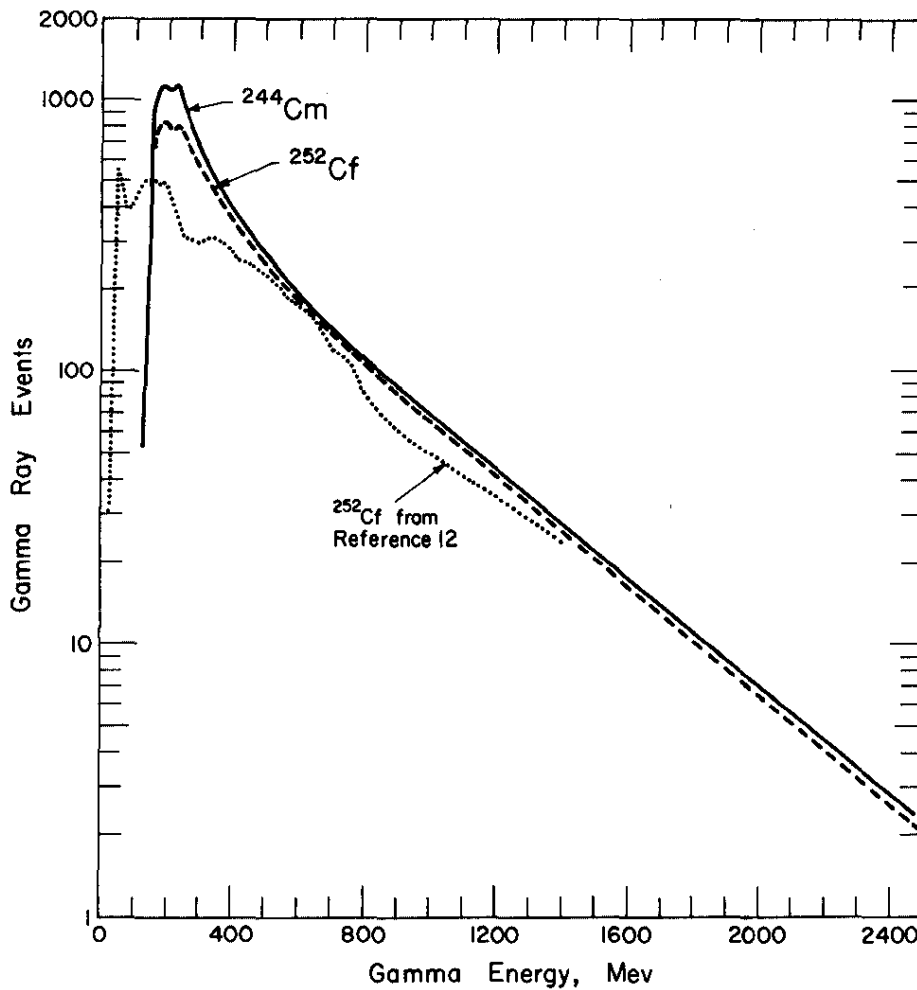


FIG. 5 CORRECTED GAMMA SPECTRA OF ^{244}Cm AND ^{252}Cf

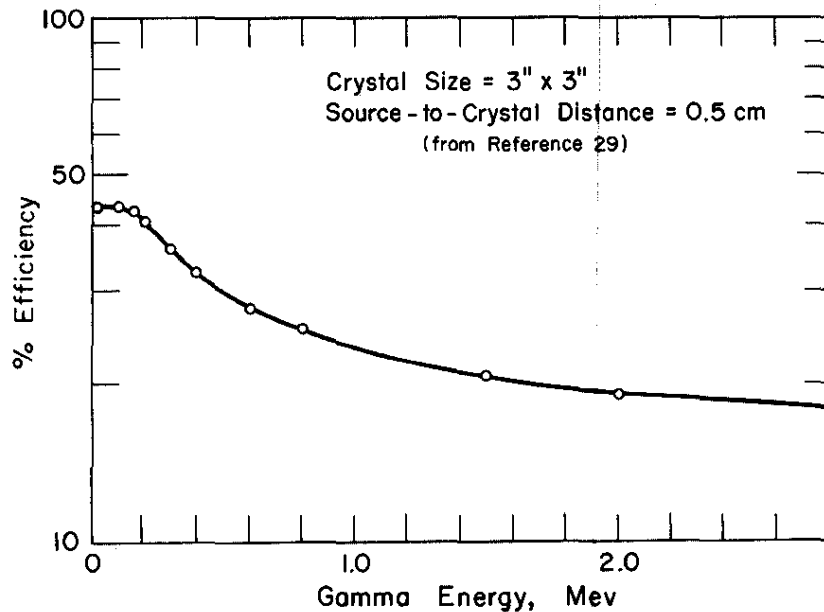


FIG. 6 NaI(Tl) DETECTOR EFFICIENCY VERSUS GAMMA ENERGY

SIGNIFICANCE OF THE SPECTRA

All experimental energy distributions for fission neutrons are similar whether the neutrons derive from induced fission as in the case of ^{235}U , ^{238}U , or ^{239}Pu , or from spontaneous fission as in the case of ^{244}Cm or ^{252}Cf . In all cases a Maxwellian distribution is obtained as predicted by Weisskopf's evaporation theory.⁽¹³⁾ This theory is a thermodynamic analogy that compares the energy stored in a compound nucleus with the heat energy of a solid or a liquid. Frenkel⁽¹⁴⁾ first emphasized that the subsequent explosion of particles from the compound nucleus was analogous to an evaporation process from a solid or a liquid. All predictions of fission neutron spectra assume that the neutrons are emitted from moving fission fragments.⁽¹⁵⁾ This assumption is in agreement with all reported experimental data, both as to the general shape of the spectra and as to the directional correlation of neutrons and fragments.⁽¹⁶⁾

Predictions of the fission-associated gamma ray spectra have not been reported on the basis of theories of fission. Therefore, experiments provide the only access to these spectra. The general assumption is that the observed continuous spectrum is produced by the decay of a statistical ensemble of many levels.⁽¹⁷⁾

APPENDIX

EQUIPMENT

The development of semiconductor charged-particle detectors^(18,19) has enabled the development of fast neutron spectrometers that provide 100- to 150-keV energy resolution over a wide range of neutron energies. Several spectrometers have been reported that detect neutrons by such exothermic secondary reactions as $^3\text{He}(n,p)$, $^6\text{Li}(n,\alpha)$, $^{10}\text{B}(n,\alpha)$, and $^1\text{H}(n,p)$.⁽²⁰⁻²⁴⁾ Such spectrometers can be used to measure neutron spectra rapidly and rather simply.^(25,26)

^3He NEUTRON SPECTROMETER

A ^3He neutron spectrometer was used to measure the spectrum of the spontaneous fission neutrons from ^{244}Cm . Figure 7 is a sectional view of the spectrometer, which consists basically of two closely spaced silicon surface-barrier detectors with high-pressure ^3He gas between and surrounding the detectors. The assembly is sealed in an aluminum can about 1-1/2 inches in diameter and 1-1/2 inches long. The can will withstand pressures up to 20 atmospheres.

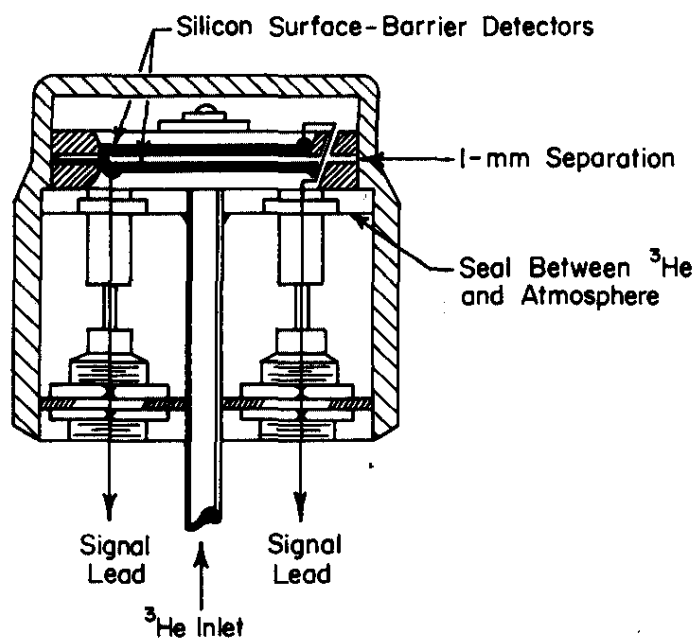


FIG. 7 SECTIONAL VIEW OF THE ^3He NEUTRON SPECTROMETER

Fast neutrons incident on the assembly cause $^3\text{He}(n,p)\text{T}$ events in the ^3He gas, releasing a total energy to the proton and triton equal to the neutron energy plus the reaction Q-value (0.76 Mev). Some of the protons and tritons enter the silicon detectors and create free electron-hole pairs at the rate of 3.4 ev/electron-hole pair. The detector response is linear with energy over a wide range, provided the sensitive depth of the detector exceeds the range of the particle, and the electric field in the sensitive region is large enough to separate the charged pairs before they recombine. The total energy of the proton and triton from a given reaction is obtained by summing the output pulses from the two detectors. Thus, the amplitude of the summed pulse is proportional to the energy of the incident neutron. The Q-value of the reaction provides a built-in bias against low-energy background events.

Detectors

The two silicon surface-barrier detectors in the ^3He neutron spectrometer were made of nominal 5000-ohm-cm silicon and were carefully matched by the vendor. Each detector had an active area of 280 mm², a FWHM noise width of 21 kev, a reverse current of 0.2 microampere, and a depletion depth of 250 microns at 50-volts operating bias. The depletion depth was sufficient to absorb totally either 20-Mev tritons or 5.5-Mev protons.

Associated Electronic Components

Figure 8 shows the instruments used to test the spectrometer and to measure the neutron spectrum.

Charge-sensitive preamplifiers must be used with semiconductor detectors because these detectors do not have the internal multiplication of signal that Geiger counters or proportional counters have. Only the electric charge created along the path of the charged particle in the depleted silicon is available as an input signal to the electronic system. This circumstance imposes a severe low-noise requirement on the input stage of the preamplifier to obtain a satisfactory signal-to-noise ratio and to minimize the resolution spread due to electronic system noise. The preamplifiers shown in Figure 8 were satisfactory in this regard and converted input charge bursts from nuclear events into voltage pulses, which were fed to the respective amplifiers.

The output pulses from the amplifiers were added algebraically in the passive sum network and fed to the biased amplifier. The biased amplifier was necessary to obtain a sufficient number of analyzer channels over a spectrum peak or other region of interest. By this

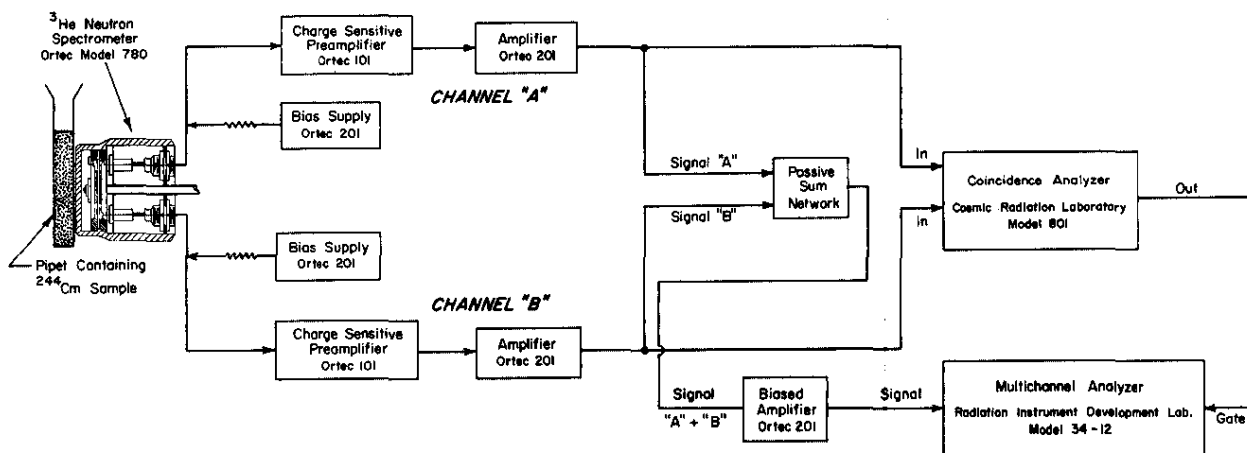


FIG. 8 INSTRUMENTS USED TO MEASURE NEUTRON SPECTRA

means all of the spectrum that was below a chosen energy was suppressed, and the remaining region was expanded as desired with biased amplifier gain before pulse height analysis.

The output pulses from the biased amplifier were fed to the input of the multichannel analyzer. The multichannel analyzer recorded only those nuclear events in which the triton from a given reaction was absorbed in one detector while the corresponding proton was absorbed in the opposite detector. To eliminate signals from other nuclear events the coincidence analyzer provided an output signal only when the input signals from the two detectors occurred within 180 nanoseconds. The output signal from the two coincident input signals was used to open the gate of the multichannel analyzer so as to record the energy of the neutron that participated in the two-way coincidence.

Checkout and Calibration

The spectrometer, which was filled with air when received from the vendor, was initially evacuated to 5 microns and checked for its response to neutrons from a bare Pu-Be source (1 curie) and to gamma radiation from millicurie sources of ^{137}Cs and ^{60}Co . A large number of low-energy counts were observed with the neutron source, and a smaller number with the gamma sources. Several counts were observed with the neutron source in the higher energy range and were probably due to neutron-induced reactions in the silicon.⁽²⁷⁾ All of the high-energy counts and most of the low-energy counts were eliminated with the coincidence analyzer.

The spectrometer was then filled to a pressure of 75 psi with ^3He from the tank supplied by the vendor and was exposed to thermalized neutrons from a paraffin-moderated, Pu-Be source. After the detectors were tested individually without coincidence to determine that they had the same energy response, they were tested together with coincidence to determine the energy resolution of the summed thermal peak. The data obtained are shown in Figure 9.

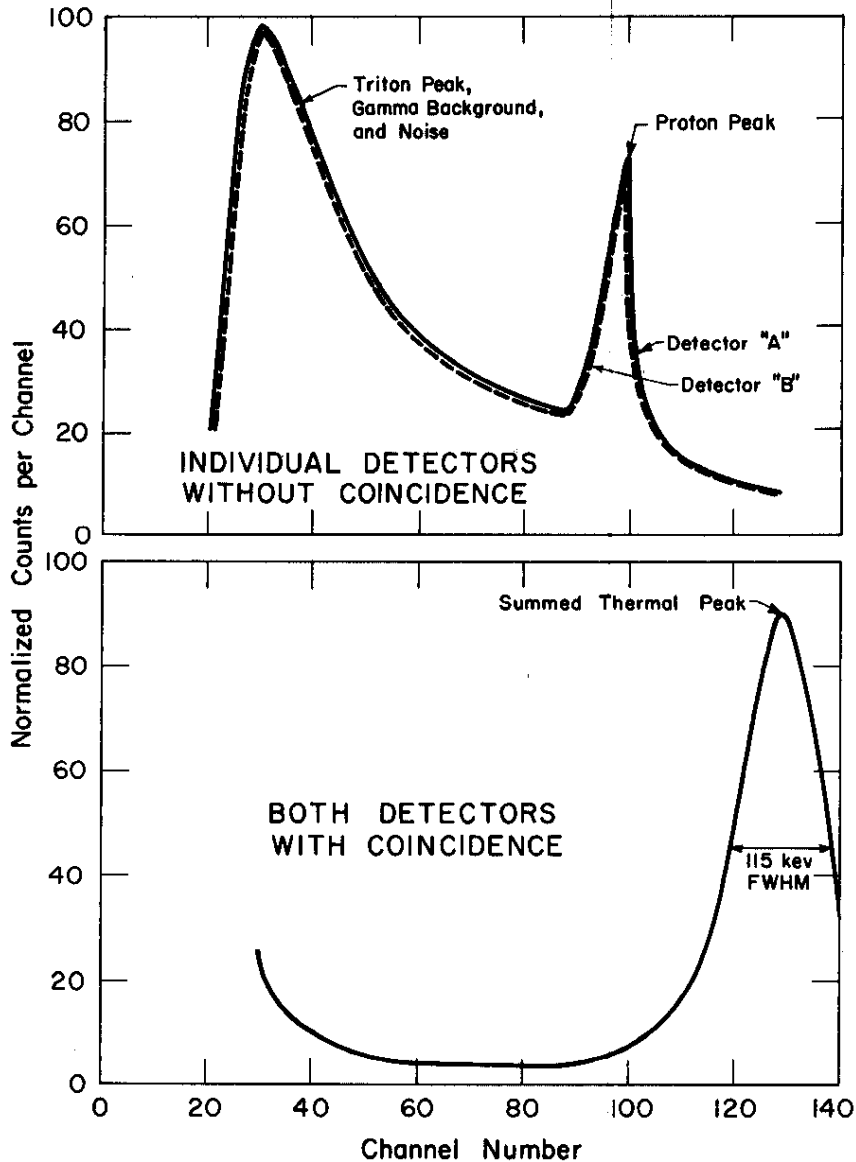


FIG. 9 DETECTOR RESPONSE TO THERMAL NEUTRONS

Primary calibration of the system was accomplished by detecting neutrons produced by the $d(d,n)^3\text{He}$ reaction in a neutron generator. The $d(d,n)^3\text{He}$ reaction was obtained by spraying a defocused deuteron beam over a relatively large area of stainless steel target and then bombarding a small area of the target with a focused deuteron beam. The neutron energy varied as a function of angle from the target, being 2.80 Mev at 0° and 2.20 Mev at 135° .⁽²⁸⁾ These two energy groups are resolved by the detector and are shown in Figure 10. The larger FWHM value for the 0° energy group was probably caused by the presence of cooling water between the target and the ^3He spectrometer at the 0° position.

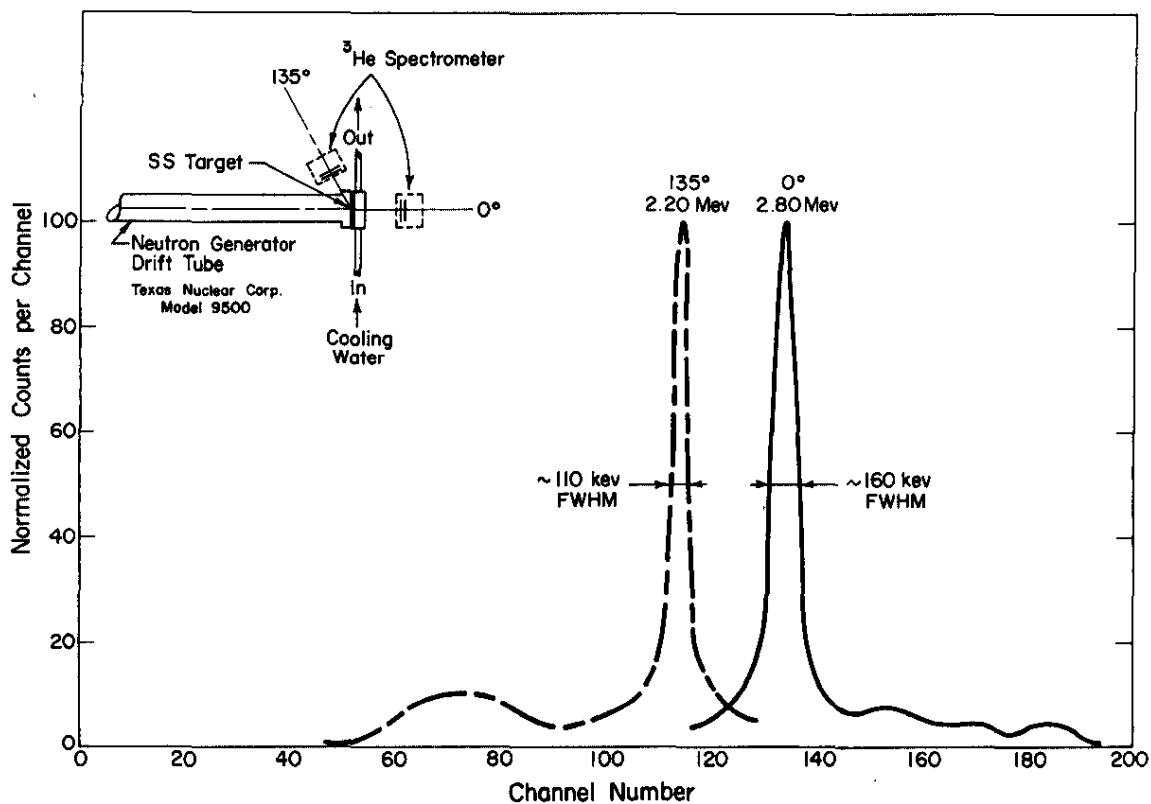


FIG. 10 CALIBRATION DATA FROM NEUTRON GENERATOR

The overall linearity of the electronic system was determined with a precision pulse generator that sent identical pulses into the inputs of both preamplifiers. The gain of each preamplifier-amplifier channel was first adjusted so that the amplitudes of the pulses going from each channel into the sum network were equal. The amplitude of the output pulses from the pulse generator was then varied to produce the response curve shown at the top of Figure 11. This curve was taken with the amplifier gain control in the x4 position. Figure 11 also shows the effect of changing the amplifier gain control to either the x2 or x1 position, with the same pulse generator settings that were used with

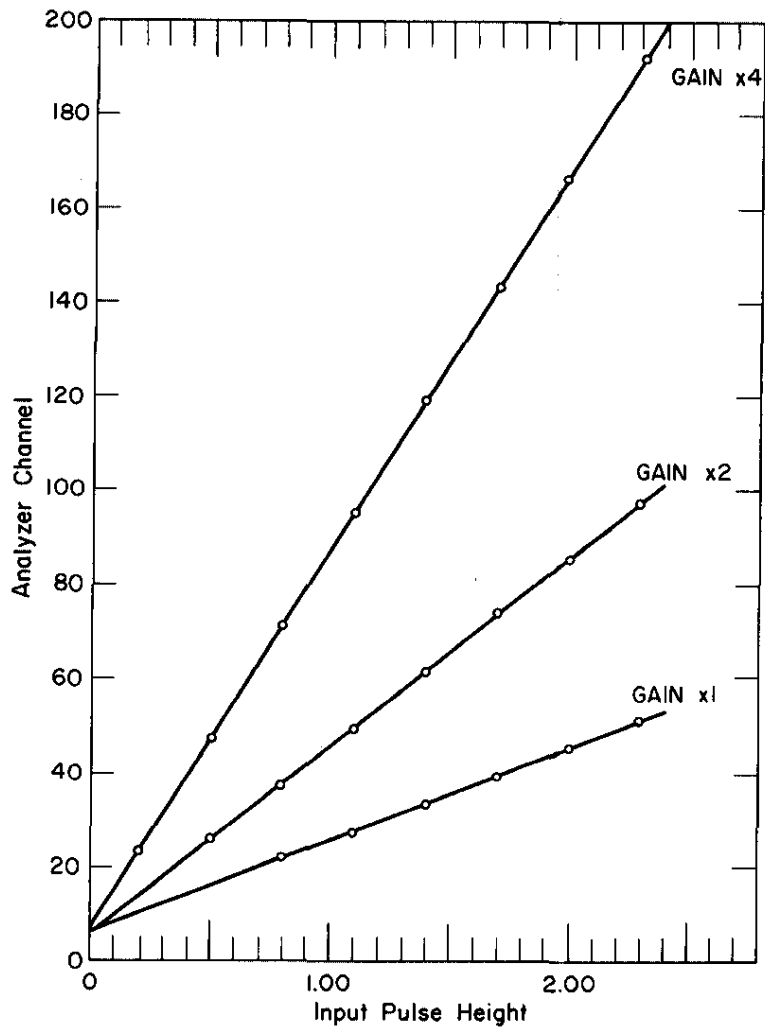


FIG. 11 ELECTRONIC SYSTEM RESPONSE TO PULSE GENERATOR INPUT

the gain control in the x4 position. The linearity was also checked by exposing the spectrometer to thermalized neutrons and obtaining the summed thermal peak for a given setting of the amplifier gain control. Figure 12 shows how the thermal peak position on the analyzer scale was then changed by changing the amplifier gain. Once the primary calibration of the system was accomplished, it could be changed by known amounts by simply changing the amplifier gain.

The main disadvantage of the ^3He spectrometer is its low efficiency. During the calibration of the system with the neutron generator, the efficiency for detecting 2.20-Mev neutrons was found to be about 5×10^{-6} . The efficiency should vary with neutron energy in much the same way that the $^3\text{He}(n,p)$ cross section varies with neutron energy.

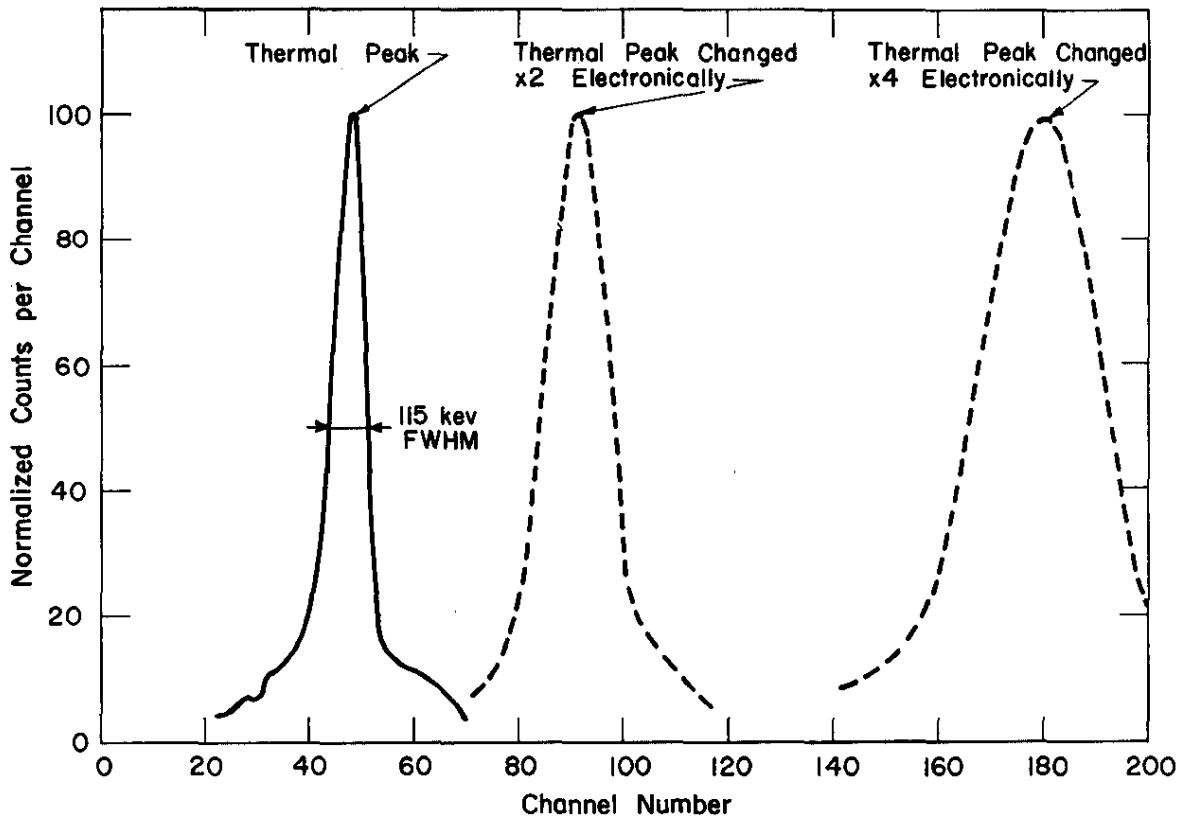


FIG. 12 EFFECT OF ELECTRONIC GAIN ON THERMAL PEAK POSITION

GAMMA SPECTROMETER

Figure 13 shows the instruments used to measure the energy spectrum of gamma rays from the spontaneous fission of ^{244}Cm . Most of the instruments used in the ^3He neutron spectrometer are also used in the gamma spectrometer. In addition to the neutron spectrometer instruments a scintillation detector is used to detect gamma rays. Pulses from the scintillation detector are fed through a conventional preamplifier and amplifier to the coincidence analyzer. The coincidence analyzer provides an output signal when the input neutron and gamma signals occur within 180 nanoseconds. The output signal opens the gate of the multichannel analyzer so as to record directly the energy of the gamma ray that participated in the coincidence.

The spectrometer was calibrated by bypassing the coincidence analyzer and determining the photopeaks of ^{137}Cs (0.662 Mev), ^{60}Co (1.173 and 1.333 Mev), and the composite photopeak of ^{231}Pa (0.313 Mev).

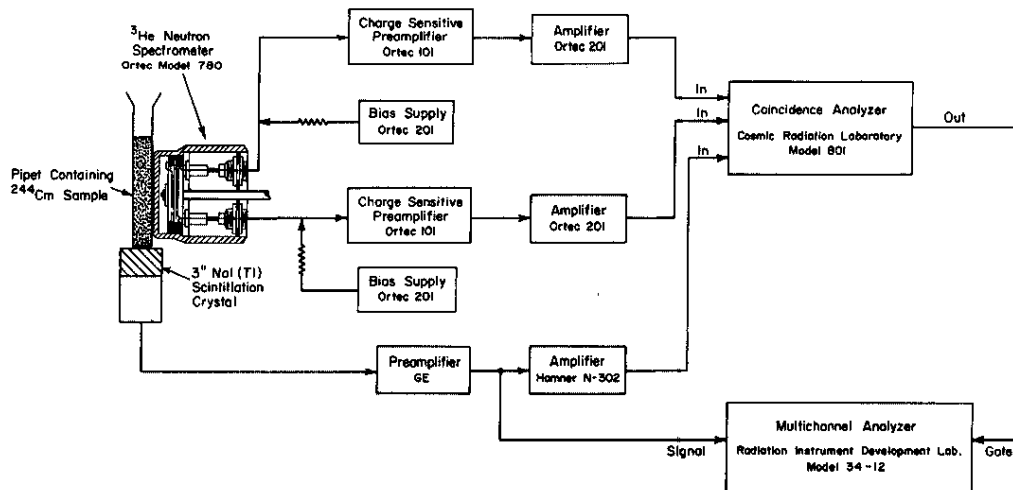


FIG. 13 INSTRUMENTS USED TO MEASURE GAMMA SPECTRA

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