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NEUTRON SPECTRA FROM CALIFORNIUM-252 WIRE SOURCES

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by

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ABSTRACT

Neutron spectra from four types of ^{252}Cf wire sources were measured with commercially available ^{3}He and ^{6}Li "sandwich" spectrometers. The sources all emitted essentially pure fission neutron spectra. The presence of non-radioactive cationic impurities at concentrations up to the accepted limit of four times the californium content produced insignificant amounts of additional neutrons. Spectra from the four source types were obtained between 0.5 and 4.7 MeV with the ^{3}He spectrometer and between 0.2 and 5.0 MeV with the ^{6}Li spectrometer with no detectable variation from the fission spectrum obtained from an electroplated source containing high-purity ^{252}Cf. The neutron spectrum from a wire source containing samarium and terbium did not differ significantly from the ^{252}Cf fission spectrum between 0.5 and 8.6 MeV as measured with the ^{3}He spectrometer.

The instrumentation and techniques for fissile neutron detection and counting are described. Spectrometer response to neutrons of different energies was verified by resolving the energy groups from a plutonium-beryllium neutron source.

CONTENTS

INTRODUCTION

The USAEC is providing sources of ^{252}Cf for various industrial, medical, and nuclear applications.[1] In support of this program, the Savannah River Laboratory (SRL) has developed a ^{252}Cf-bearing palladium metal wire that contains a uniform concentration of ^{252}Cf per unit length and minimizes wet chemistry techniques in source preparation.[2] The wire production process includes chemically plating ^{252}Cf oxalate with palladium, calcining the resulting powder to cermet powder, compressing and sintering the cermet powder into a high density pellet, and rolling the pellet into wire.

During early development work on the cermet, various carriers that would ensure complete precipitation of small amounts of ^{252}Cf oxalate were investigated. Samarium, cerium, and terbium were tried as carriers because their chemical properties were similar to californium. Terbium was selected for production use mainly because of its low cross section for thermal neutron capture.

The elemental composition of a typical production wire source for commercial use is palladium with relatively small amounts of ^{252}Cf, terbium, and oxygen. Small quantities of calcium, sodium, potassium, barium, magnesium, aluminum, copper, iron, zinc, and lead are present mainly from impurities in the ^{252}Cf feed solution. Alpha particles from ^{252}Cf induce (α,n) reactions in some of these impurity elements. The (α,n) yields for sodium, magnesium, and aluminum were reported in 1944.[3] The (α,n) yield from oxygen is well known,[4,5] and derives mainly from the ^{18}O isotope in natural oxygen. Calculations showed that $^{18}O(\alpha,n)^{21}Ne$ reactions would produce more neutrons than all other impurities combined. However, these neutrons were calculated to contribute less than 1% of the total neutrons from the wire source. Some of the reaction yields and Q-values are not well known so that calculations could give an erroneous idea of the total neutron yield from the impurities. Because of the uncertainty in the total neutron yield, neutron spectral measurements were undertaken to determine that the wire sources emitted an essentially pure fission neutron spectrum.

SOURCE TYPES

Four sources were prepared by different techniques. They contained varying impurities as listed in Table 1. Neutron spectra were obtained on all four sources.

Feed specifications for the ^{252}Cf require that total actinide and lanthanide impurities shall not exceed the californium content, and nonradioactive cations shall not exceed four times the californium content.[6] Major nonradioactive impurities are oxygen, sodium, calcium, potassium, barium, magnesium, aluminum, copper, iron, zinc, and lead, as determined by spark source mass spectrographic analysis of ^{252}Cf feed. Isotopic purity of the californium is typically 79% ^{252}Cf, 15% ^{250}Cf, 4% ^{251}Cf, and 2% ^{249}Cf.

TABLE 1. WIRE SOURCES

Source	Fabrication Process	Palladium Content, g	Carrier Content, g			^{252}Cf Content, μg[a]	Size, cm
			Sm	Tb	Ce		
Alloy 1	Chem. Plating plus Melting[b]	1.45	0.0003	0.003	-	175	0.1 x 0.1 x 10.7
1005-C	Admix	1.0	-	-	0.0126	805	0.08 x 0.08 x 11
1014-C	Chem. Plating	1.0	0.003	-	-	607	0.08 x 0.08 x 14.6
1021-C	Chem. Plating	1.0	-	0.003	-	25.8	0.1 x 0.1 x 8.6

a. Small samples of the first three types that contained between 5 and 20 μg ^{252}Cf were used in the spectrometer measurements.

b. Portions of several cermet wires were melted at 1600°C in a reducing atmosphere to demonstrate that an alloy could be made.

EXPERIMENTAL EQUIPMENT

The neutron spectra were measured with commercially available ^{6}Li and ^{3}He spectrometers and associated instrumentation as shown in Figure 1. Each spectrometer contained two matched surface-barrier detectors in a face-to-face or sandwich geometry plus a quantity of neutron-sensitive material, either ^{6}LiF or ^{3}He. The technical specifications and operating characteristics for the ^{3}He and ^{6}Li spectrometer were given in a previous report.[7]

The charged particle pairs that are produced by neutrons incident on the sensitive material are detected simultaneously in the two detectors. The resulting pulses are amplified in two separate channels. Each channel has double shaping modes with 0.2-μsec time constants in the multimode amplifiers. The two output pulses from the multimode amplifiers are summed to produce a pulse with amplitude proportional to the energy of the incident neutron plus the reaction Q-value (4.78 MeV for ^{6}Li and 0.76 MeV for ^{3}He events).* A biased amplifier expands the portion of interest in the total energy spectrum, and a pulse stretcher ensures compatibility with the input requirements of the multichannel analyzer.

The output pulses from the multimode amplifiers are also fed to crossover pickoff circuits that provide timing pulses to the fast coincidence unit. When the two timing pulses occur within the coincidence resolving time (adjusted to 110 nanoseconds for this work), a coincidence output signal is generated, inverted, and used to gate the multichannel analyzer and record the event. The fast coincidence unit eliminates signals from unwanted nuclear events (such as ^{3}He recoils) but does not eliminate coincidence background events (such as gamma rays or

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* The nuclear reactions are: $^{6}Li + {}^{1}n \rightarrow {}^{3}H + {}^{4}He + 4.78$ MeV

$^{3}He + {}^{1}n \rightarrow {}^{1}H + {}^{3}H + 0.76$ MeV

The sum pulse from the ^{6}Li spectrometer is the alpha pulse from one surface barrier detector added to the corresponding triton pulse from the opposite detector, or vice versa. For the ^{3}He spectrometer, the sum pulse is the proton pulse from one detector added to the corresponding triton pulse from the opposite detector.

energetic protons passing through both detectors). Coincidence background events are removed by making two source measurements: one measurement with the neutron-sensitive spectrometer and a second measurement under identical conditions with a background spectrometer. The background obtained by the second measurement is subtracted, channel-by-channel, from the counts obtained by the first measurement.

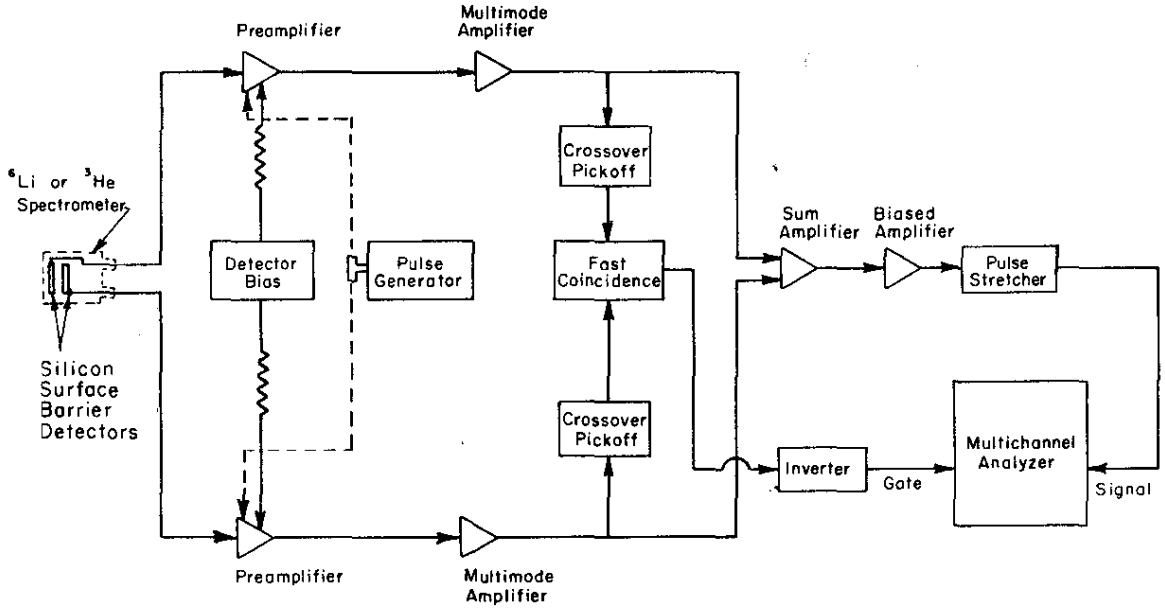

FIG. 1 SPECTROMETER INSTRUMENTATION

CALIBRATION

The spectrometers were calibrated with the peak arising from ^{6}Li plus thermal neutrons (4.78 MeV) and with the peak arising from ^{3}He plus thermal neutrons (0.76 MeV) by determining the linearity of response of the electronic system with an ORTEC* Precision Pulse Generator, and then moving the ^{6}Li or ^{3}He thermal peak over the analyzer scale in discrete steps by changing the electronic gain. The system was then adjusted to analyze and record neutron energies over the desired energy interval.

Actual thermal peak data (as displayed on the readout oscilloscope of the multichannel analyzer) is shown in Figure 2. This figure also shows the calibration technique. The peak on the left is that from ^{6}Li thermal neutrons. The low energy fall-off from this peak was biased out in the electronic system. On the right is the ^{6}Li thermal peak obtained by doubling the electronic gain. The energy difference between these two peaks is then 4.78 MeV, and the energy calibration as keV/channel is simply obtained by dividing 4.78 MeV by the total number of analyzer channels between the two peaks. The ^{3}He spectrometer was similarly calibrated. This calibration technique has been used in neutron spectroscopic work at SRL since 1965[8] and automatically compensates for charged particle energy loss in the ^{6}LiF coating or in the ^{3}He gas. The energy loss in the coating or gas is assumed to be the same for neutrons of all energies.

An additional calibration check is made by momentarily switching out the circuit coincidence requirement and observing the individual alpha peak at 2.05 Mev and the triton peak at 2.73 MeV as well as the sum peak at 4.78 MeV from thermal neutron reactions in ^{6}Li. One similarly observes the individual triton peak at 0.190 MeV and the proton peak at 0.570 MeV as well as the sum peak at 0.76 MeV from thermal neutron reactions in ^{3}He. These techniques have also been used at SRL since 1965. The ^{6}Li technique was recently used by Yang in his investigation of a ^{6}Li sandwich spectrometer.[9]

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* ORTEC Incorporated, Oak Ridge, Tenn.

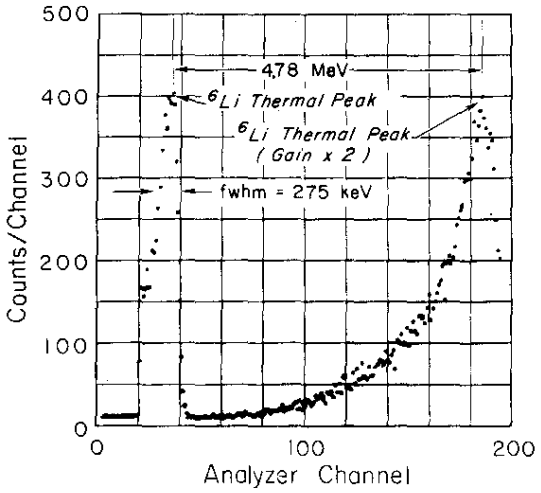


FIG. 2 CALIBRATION WITH ^6Li THERMAL PEAK

MEASUREMENT TECHNIQUE

During source measurements, the spectrometers were covered with 0.076 cm of cadmium to shield them from thermal neutrons, and enclosed in a 1.3-cm-thick lead housing to shield them from gamma radiation. This assembly was placed inside a thin-walled aluminum housing in which the temperature was controlled at 30° ±0.5°C. The source to be measured was positioned at the appropriate distance from the spectrometer housing to prevent radiation damage to the detectors and overloading of the electronic system. The spectrometer housing and source were centrally located inside a shielded facility at least 1.5 meters away from any large object that could scatter neutrons.

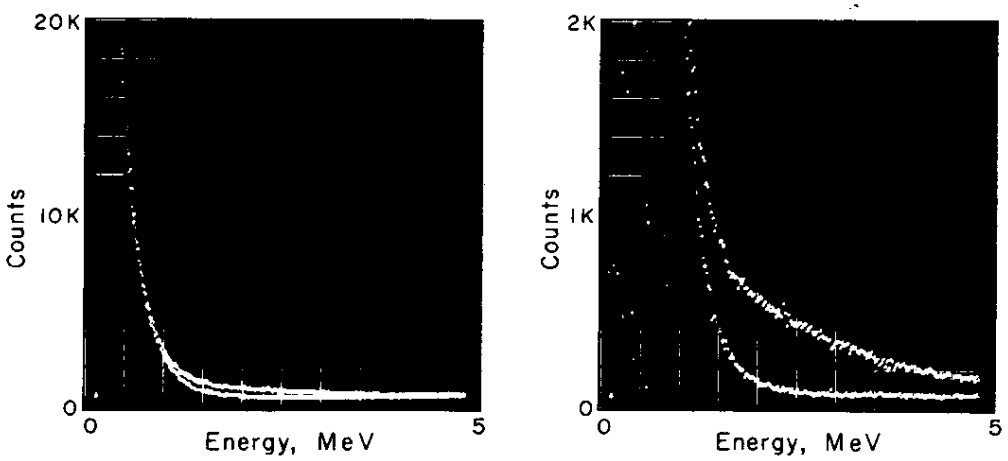
Typical measurement times were 48 to 72 hours for the ^{6}Li spectrometer (with an efficiency of $\approx 10^{-6}$ in the MeV range) and 12 to 24 hours for the ^{3}He spectrometer (with an efficiency of $\approx 10^{-5}$ over the same range).

The neutron data in each case were accumulated in 200 channels of a 400-channel analyzer. The remaining 200 channels were used periodically to check the instrumentation for drift with the pulse generator and, if necessary, to make small gain adjustments.

DATA ANALYSIS AND SPECTRA

Actual data obtained with the ^3He spectrometer and Alloy 1 source were displayed on the readout oscilloscope of the multichannel analyzer (Figure 3). The signal plus background data were accumulated with the spectrometer filled with ^3He to 1200-cm-Hg, and the background data were accumulated with the spectrometer evacuated to 5 μm. The background data were subtracted from the signal plus background data to obtain the uncorrected signal. The energy range for these data was 0.52 to 4.77 MeV over 200 analyzer channels with a calibration of 21.8 keV/channel.

The energy interval for data analysis was selected within the energy resolution of the spectrometer for reliably determining the shape of the spectrum and wide enough to contain enough counts for an acceptable statistical spread in the resulting data. The energy interval in this case was 5 analyzer channels or 109 keV, and the energy resolution of the spectrometer was about 200 keV with the 1200-cm-Hg ^3He filling. The total of background-corrected counts in each energy interval was corrected with the $^3He(n,\rho)$ cross section value at the midpoint of the interval. The resulting data for the corrected spectrum were generally plotted as a histogram of normalized counts per energy interval versus neutron energy.



20K Full Scale to Show Complete Spectra. Bottom Curve Is Background

Same Spectra at 2K Full Scale to Enhance High Energy Region

FIG. 3 ^3He SPECTROMETER DATA FROM ALLOY 1 SOURCE

The Alloy 1 source was studied in more detail than the other sources because it contained both terbium and samarium. Additional spectra were accumulated with the ^{3}He spectrometer adjusted to cover an energy range from 1.25 to 8.67 MeV over 200 analyzer channels with a calibration of 39.1 keV/channel. A plot of corrected data was obtained over the two energy ranges (Figure 4). The neutron spectrum from the wire source containing samarium and terbium did not differ significantly from the ^{252}Cf fission spectrum[10] between 0.5 and 8.6 MeV. From the data obtained and assuming that (α,n) neutrons would mainly occur in the 1 to 3 MeV energy range, a variation of about 1.5% from the fission neutron spectrum was detectable with the ^{3}He spectrometer. The ^{3}He spectrometer was used for this definitive measurement because of its relatively high efficiency. The maximum drift observed during the measurements amounted to two channels, or about 40 keV for the lower energy range and 80 keV for the higher energy range.

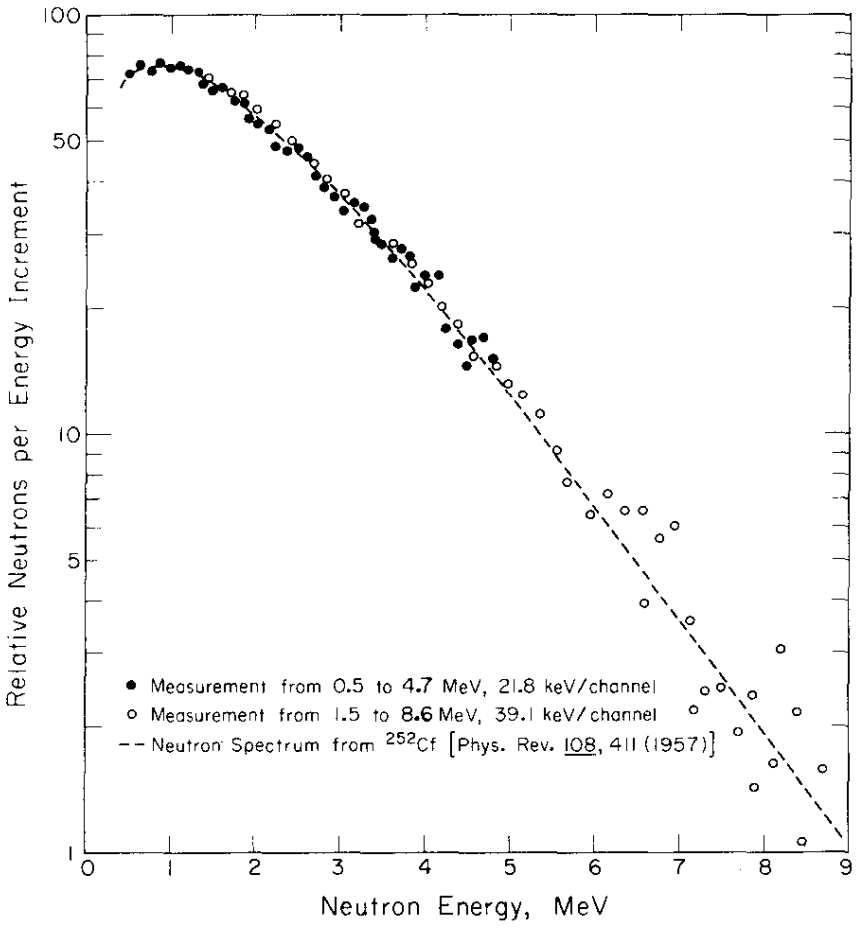
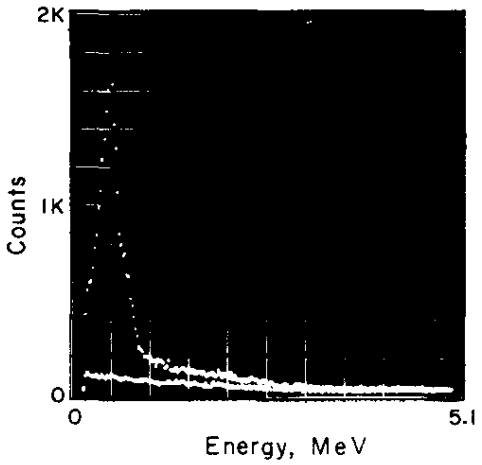


FIG. 4 NEUTRON SPECTRUM OF ALLOY 1 SOURCE WITH ^{3}He SPECTROMETER

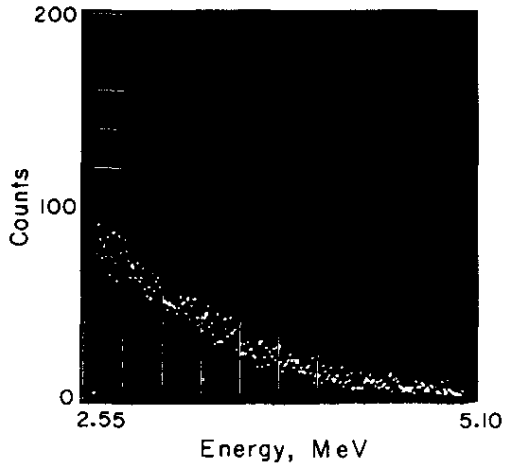
Data were obtained during a 96 hour run with the ^{6}Li spectrometer and Alloy 1 source, and displayed on the readout oscilloscope of the multichannel analyzer (Figure 5). The energy range for these data was 0.20 to 5.08 MeV over 200 analyzer channels with a calibration of 27.9 keV/channel. The energy interval for data analyses was five analyzer channels or 140 keV, and the energy resolution of the spectrometer was about 300 keV. Note that relatively few background counts were obtained over this energy interval. The low energy peak in Figure 5 is due primarily to the resonance in the $^{6}Li(n,t)$ cross section that occurs around 250 keV.[11] The total of background-corrected counts in each energy interval was corrected with the $^{6}Li(n,t)$ cross section value at the midpoint of the interval to obtain the histogram shown in Figure 6. The maximum drift observed in this case was three channels, or about 80 keV.

Both spectrometers were used in measuring the other wire sources listed in Table 1. The spectra obtained were essentially the same as from Alloy 1. An electroplated source containing high-purity ^{252}Cf was also measured with the ^{3}He spectrometer and exhibited the same spectrum as the four wire sources.

Data were collected during a 71 hour run with the ^{3}He spectrometer on a 1-curie Pu-Be source (Figure 7). The spectrometer was adjusted to cover the energy range from 1.25 to 8.67 MeV over 200 channels as previously described. The intensity maxima at 3.2, 4.7, and 7.7 MeV as well as the characteristic dip at 6.1 MeV are apparent in the uncorrected data. The corrected data points were normalized to the Pu-Be spectra that Anderson and Bond[12] obtained by a different technique (Figure 8). The corrected data provided additional calibration points and indicated that the spectrometer responded properly to neutrons of different energies in the measurement range of 1.25 to 8.67 MeV.



2K Full Scale to Show Complete Spectra. Bottom Curve Is Background



Upper 100 Channels of Signal at 200 Counts Full Scale to Enhance High Energy Region

FIG. 5 ^{6}Li SPECTROMETER DATA FROM ALLOY 1 SOURCE

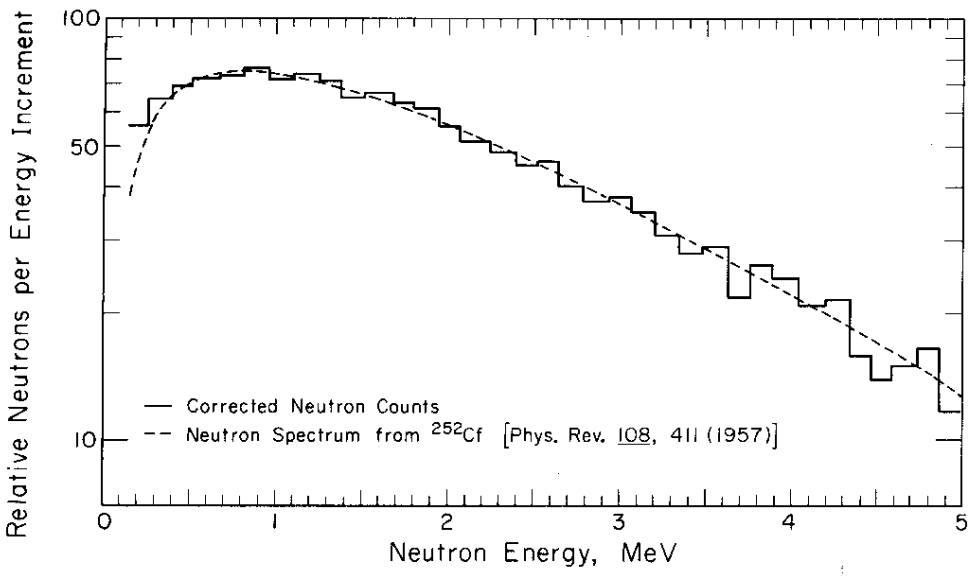
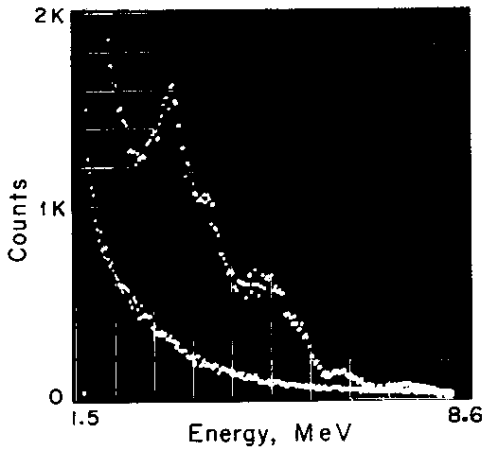


FIG. 6 NEUTRON SPECTRUM OF ALLOY 1 SOURCE WITH ^{6}Li SPECTROMETER



Bottom Curve Is ^{3}He Data
from Alloy 1 Source

FIG. 7 ^{3}He SPECTROMETER DATA FROM Pu-Be SOURCE

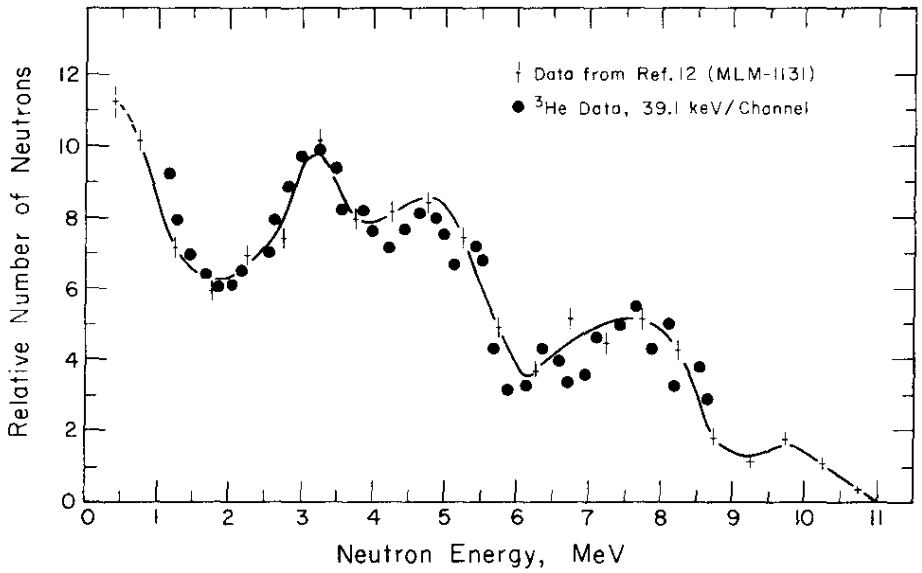


FIG. 8 NEUTRON SPECTRUM OF Pu-Be SOURCE WITH ^{3}He SPECTROMETER

CONCLUSION

Neutron production from (α,n) reactions is insignificant in the ^{252}Cf wire sources currently produced at SRL. The neutron counting techniques and instrumentation would have detected a variation of about 1.5% from the spectrum of a pure ^{252}Cf source.

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