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PROTON RECOIL MEASUREMENTS OF THE PuBe SOURCE NEUTRON SPECTRA

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ABSTRACT

The spectrum of a 5 Ci PuBe neutron source has been measured from 40 keV to 800 keV. The measurement was made using a spherical proportional counter containing 4 atmospheres of hydrogen. Pulses due to gamma-ray interactions in the chamber were distinguished by rise time analysis using a two-parameter pulse-height analyzer. The proton recoil spectrum was analyzed using a code written by Benjamin which accounts for contributions to the proton recoil spectrum from neutrons of energy greater than that to be analyzed.

The proportional counter measurement is combined with a recent liquid scintillator measurement to give a complete spectrum. Good agreement exists between the two measurements in the region of overlap from 500 to 800 keV. The proportional counter data show that the flux is fairly smooth and flat in the region from 40 keV to 800 keV.

The present measurements indicate a neutron yield below 1 MeV for this source of 20.2 ± 3.9 percent.

INTRODUCTION

For many applications of radioactive neutron sources it is necessary to have an accurate knowledge of the neutron spectrum as well as the source strength. The use of large water bath techniques has made it possible to determine source strengths to an accuracy of 3 percent or better but the neutron spectrum is not nearly so well known. Considerable past work with emulsions and organic scintillators has resulted in fairly well established spectra at energies greater than about 1 MeV but below this energy the spectra of many radioactive sources have not been measured.

In the present work a 5 Curie (Ci) PuBe source spectrum is measured from 800 keV down to 40 keV using a spherical proportional counter (1). This measurement is combined with a recent liquid scintillator measurement (2) to give a complete spectrum. The primary difficulties in making a proportional counter measurement of the low energy part of the $Be(\alpha,n)$ spectrum are caused by the large number of high energy neutrons emitted by these sources, and by the gamma-ray background.

EXPERIMENT

A spherical proportional counter designed by Benjamin et.al., (1) was used in the measurements. The counter was filled with 4 atmospheres of hydrogen and a trace of helium-3 and had an inside diameter of 3.94 cm. The counter wall was .051 cm thick stainless steel. A lead shield about .64 cm thick, and an outer cadmium cover about .075 cm thick surrounded the counter.

The measurements were made with the counter mounted on a light frame 1.8 meters above the floor of a room 4 meters high, and 6 meters square. The PuBe source was supported with its axis vertical at 15 cm from the counter by a .62 cm diameter threaded iron rod.

A block diagram of the electronics is shown in figure 1. To separate protons from electron recoils, a two-parameter analyzer was driven by an identification signal as well as by a linear signal. To preserve the linear response for large pulses in hydrogen, which may take up to 6μ sec to rise, the preamplifier decay time was made 130μ sec, and a single 7μ sec RC integration and differentiation time constant was chosen in the linear amplifier. To obtain an identification pulse separating fast-rising proton recoils from electrons, the preamplifier output was R-C clipped with a short time constant in the second linear amplifier. This pulse was used to drive a fast stretcher and a slow stretcher of about 5μ sec duration. The slow stretcher pulse was adjusted to reach its peak at its end. Finally, the identification pulse was delayed 4.75μ sec. These precautions ensured that the two-parameter analyzer would always

convert the full voltage of the linear signal.

Careful measurements were made of the analyzer linearity and pulse height zero. Amplifier gains were measured and absolute gas gains were obtained using the 764 keV helium-3 peak. A plot of ln A/V versus ln V gave a straight line over a range of gas gain from A=8 to A=250.

Measurements were made with gas gains of from 45 to 180. The resolution of the helium-3 peak was about 10 percent full width at half maximum (FWHM).

THE 5 CURIE PuBe SOURCE

This source is a right cylinder, outer diameter 3.33 cm, and length 6.91 cm. It contains 79.88 gms of plutonium and 39.30 gms of beryllium intimately mixed and melted in a tantalum cup and doubly encapsulated in stainless steel.

The source emission rate was $9.50 \pm .29$ neutrons per second in September of 1962. The isotopic fraction of plutonium-241 was .74 percent (3). This corresponds to an initial rate of increase of source strength due to the formation of americium-241 of 2.1 percent per year.

The source strength in September 1970 is estimated to be 13.6 percent greater than in 1962. The relative emission rate of this source as a function of direction was measured with a long counter, the center of the front fact of which was at 100 cm from the center of the source. The emission rate in the direction perpendicular to the source axis was measured to be $1.067 \pm .020$ times as great as the emission rate averaged over the total solid angle. The calculated neutron flux at one meter from the source perpendicular to the axis is then

$$\phi = \frac{9.50 \times 1.067 \times 1.136 \times 10^6}{4\pi (100)^2} = 91.6 \text{ n/cm}^2\text{sec}$$

DATA ANALYSIS

The maximum pulse size that can be produced by a gamma-ray in this chamber corresponds to that of about a 100 keV proton, so that there is no gamma-ray interference above this energy. Consequently, the proton recoil data analysis is straightforward. The Spec 4 code of Benjamin (4) was used. This code requires a neutron spectrum shape

above the highest energy to be analyzed. The required shape was obtained from the data of reference 2. The code generates a proton recoil response shape for neutrons above the energy to be analyzed, using analytic response functions. It then normalizes this shape to the data, and strips the differentiated remainder.

Below about 100 keV proton energy, gamma events occur in the chamber. A two-dimensional plot of counts from a cobalt-60 source is shown in figure 2. The pulses from a 40 keV electron travel a total distance equivalent to the diameter of this chamber, and may require as long as 6μ sec to rise. Consequently, at a given energy the clipping circuit places most of the pulses near the energy axis.

Figure 3 shows data from the PuBe source. A 100 keV proton in the counter travels only about a millimeter, so that all of the 100 keV proton pulses should rise within the clipping time of the identification circuit. These pulses appear as a peak in the identification channel. Since the identification pulse size for these pulses is proportional to the energy, this peak occurs in a diagonal line on the plot (5).

Unfortunately, not all the pulses on this plot can be clearly ascribed to the proton-recoil peak, or to gamma-rays. Above 100 keV, where the gamma-ray induced pulses are gone, there is a flat continuum of pulses which have smaller identification pulses than those in the peak. It is believed that these pulses are due to higher energy protons that lose only a fraction of their energy in the gas before colliding with the wall. At 2.5 MeV for example, a proton will lose 100 keV while travelling nearly 2 cm. in the gas.

Figure 4 shows the spectrum of pulses from the source surrounded by a spherical shell of tungsten, which sharply reduces the fraction of high-energy neutrons in the spectrum. The calculated fraction of the flux above 2 MeV in the tungsten leakage spectrum is only 17 percent compared with 70 percent for the bare source. The number of counts not on the diagonal have been much reduced. This non-peak fraction is 35 percent at 100 keV for the bare source, and only 4 percent for the tungsten leakage spectrum.

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It is possible to subtract an arbitrary fraction of a gamma-ray spectrum from a mixed spectrum so as to obtain a smooth continuation below 100 keV of the distribution above 40 keV, so that the subtraction is from the non-peak counts.

Figure 5 shows the total counts measured for the 5 Ci PuBe source. Two slightly different gamma fractions are subtracted below 100 keV. The non-peak contribution to the spectrum for these subtractions is shown also. A modified version of Spec 4 was used to calculate the fraction of recoils from neutrons of energy greater than 1.5 MeV. This is also shown in figure 5. Figure 6 shows similar results for the tungsten leakage spectrum, where the non-peak fraction is much lower.

In both fluxes, the fraction of non-peak counts is not a constant with energy. For a fixed clipping time, there will be some energy of proton-recoil below which all the recoils will be in the group along the diagonal. The rise time for these very small pulses will be within the clipping time. Since the absolute number of recoils from high energy events increases only about 10 percent from 100 keV to 10 keV, the number of counts occurring off the diagonal should, at most, rise no more than this.

The uncertainty in the gamma-ray subtraction above 40 keV is of the order of the difference between the two subtractions shown in figures 5 and 6. The approximate upper limit of the counts in the non-peak fraction is the upper curve shown. If one subtracts more than enough to give the lower curve, portions of the identification channel region become negative.

RESULTS AND DISCUSSION

The solid line of figure 7 shows the neutron flux spectrum reduced to 1 meter distance for the 5 Ci PuBe source as presently measured by the 4 atmosphere hydrogen proportional counter. The curve drawn through the data is the weighted averages of two runs each above and below about 100 keV. The hatched area represents the estimated error. The analysis is not inconsistent with a nearly featureless spectrum. But the analyses of runs taken on different days show the source spectra features reproduced here. Below 70 keV, the large uncertainty in the spectrum is caused by the uncertainty in the gamma fraction subtracted from the non-peak data. Because the non-

peak proton recoil contribution is expected to decrease with decreasing neutron energy, the upper limit of the spectrum shown is believed to be near the upper limit for the spectrum in this region. The lower limit is not so clearly defined.

The integral of the flux from 0 to 800 keV with a flat 30 n/cm^2 sec MeV below 40 keV is 15.6 ± 2.7 n/cm^2 sec. The total flux at one meter is calculated to be 91.6 ± 3.7 n/cm^2 sec. The data of reference 2, therefore, have been normalized to include 76.0 n/cm^2 sec between 800 keV and 11 MeV. The fraction of the neutron spectrum below 1 MeV is then 20.2 ± 3.9 percent.

The spectrum of reference 2 is very similar to that of Anderson and Bond (6) who measured the spectrum of a similar PuBe source with nuclear emulsions. Their measured spectrum extrapolated smoothly to zero neutron energy indicated about 18 percent below 1 MeV. St. Romain, et.al., (7) using Bonner spheres to estimate neutron yields found about 17 percent of the spectrum of a 1 Ci PuBe source below 1 MeV. They inferred a peak in the neutron spectrum at 0.3 MeV due to inelastic scattering of alpha particles from beryllium. The excited beryllium nucleus was postulated to decay by neutron emission. Shook, et.al., (8) measured the age of neutrons in various media and using a similar 5 Ci PuBe source, estimated that 21 percent of the neutrons were emitted below 1 MeV. They used the Anderson and Bond spectrum above 1 MeV.

In conclusion, the neutron spectrum of a 5 Ci PuBe source measured from 40 keV to 800 keV is generally flat and smooth. The measurement indicates there are about 20.2 ± 3.9 percent of the neutrons emitted below 1 MeV, which is consistent with other measurements in magnitude but not in shape. The method of estimation of the contribution of gamma-rays to the recoil spectrum below neutron energies of 100 keV is the principal source of uncertainty in the present proton-recoil measurements which have large fractions of the proton recoils caused by high-energy neutrons.

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PROPORTIONAL COUNTER

PREAMPLIFIER

H.V. SUPPLY

AMPLIFIER RC = 0.2 μsec

PULSE STRETCHER

DELAY AMPLIFIER

IDENTIFICATION PULSE

AMPLIFIER RC = 7 μsec

ENERGY PULSE

TWO PARAMETER ANALYZER

Figure 1. - Block diagram of instrumentation.

IDENTIFICATION PULSE HEIGHT

RECOIL ENERGY (keV)

```
       1    2   3    4   5   6   7   8   9  10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32
 1 131C72
 2 185217
 3 181174
 4  85942    1
 5  48743   13
 6  336C6  149   5    1
 7  24248  495  48
 8  18533  795 196   14   1       1

 9  13715  889 331   63   6
10  10824  965 425  125  17   5
11   8425  993 440  250  60  18   3
12   6477 1040 538  256  95  32   2   1
13   5195 1024 556  250 116  50  18   2
14   4C53 1007 585  330 139  60  23   8
15   3474  987 605  348 200  75  30  12   3  1
16   2679  929 603  330 207  98  45  14   7  2  1

17   2211  912 603  383 207 108  44  20   9  3  1
18   175C  867 600  344 250 117  59  27   5  2
19   133C  801 609  370 238 114  63  29  13  3  1
20   1C94  741 537  384 218 130  64  35  17  6  5  1
21    758  664 526  329 209 128  82  24  21 11  1     2
22    577  563 472  362 234 138  77  32  22 11  3  1  1
23    4C4  511 448  340 192 141  76  44  25  5  6  1
24    3C9  466 430  269 209 117  78  50  31 10  5  1

25    234  334 371  326 198 124  74  38  25 11  5  2  1
26    176  293 333  274 199 110  74  37  16 11  7  2  1
27     91  222 236  224 191 127  65  41  21 11  8  4  1
28     75  198 228  197 160 100  55  38  16 11  7  3
29     55  152 196  169 132 106  58  24  18  9  4  1  1
30     21  101 145  149 132  78  65  29  25 16  3  1  1                               1
31     18   75 143  152 114  74  51  35  17 11  3  4  3  1
32     1C   66 115  110 110  75  44  27  19  7  5  5  1  2  1

33      8   40  87   96  90  66  49  31  19  7  7  6  3  1  1
34      1   28  63   68  80  60  41  21   9  9  5  3  2  1
35      1   22  45   6C  65  54  39  22  10  9  4  1  1  1
36          12  36   57  49  42  34  24   7  8  7  1     2  1
37      1   10  24   40  29  38  17  16  15  8  4  3  1
38      1    2  14   27  30  26  27  23   9 10  1  2  1
39           4  12   14  21  25  23   7  11  7  5  2  2  2  2
40                5  17  26  23  20   5   8  3  1  1  1

41           2   4   15   9  13  20   8  15  5  5  1  1
42               5   8  10  13   9   8  10  4  2  1  1
43      1        3   6  12   5   8   3   8  4  1  1  1     1  1
44               1   6   9  10   6   8   6  1  6     2
45               2   5   6   9   5   6   7  3  2  1        1     1
46                   3   3   6   6   4   2  3  2  1              1  1
47                   2   1   2   3   2   1  2  2  1                 1  1
48               3   3   1   4   2   2   3  3  1

49                       1   5   4   2   2  3     1
50                       4   1   3   2   1  1
51                           1   1   3   1  2  2     1
52               2           2   1   2   2  2  1
53                       1   2   3   1   2  2     1
54                       2       2       1  1
55                               2          1
56                   1   1   1                      1

57                               1   2      1  1  1
58                       1   1   1
59                               1             1  1     1
60                               1
61
62                           1                  1
63
64                                              1
```

Figure 2. - Proportional counter data, cobalt-60 source.

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IDENTIFICATION PULSE HEIGHT

RECOIL ENERGY (keV)

Figure 3. - Proportional counter data, 5 Ci Pu-Be source.

IDENTIFICATION PULSE HEIGHT

RECOIL ENERGY (keV)

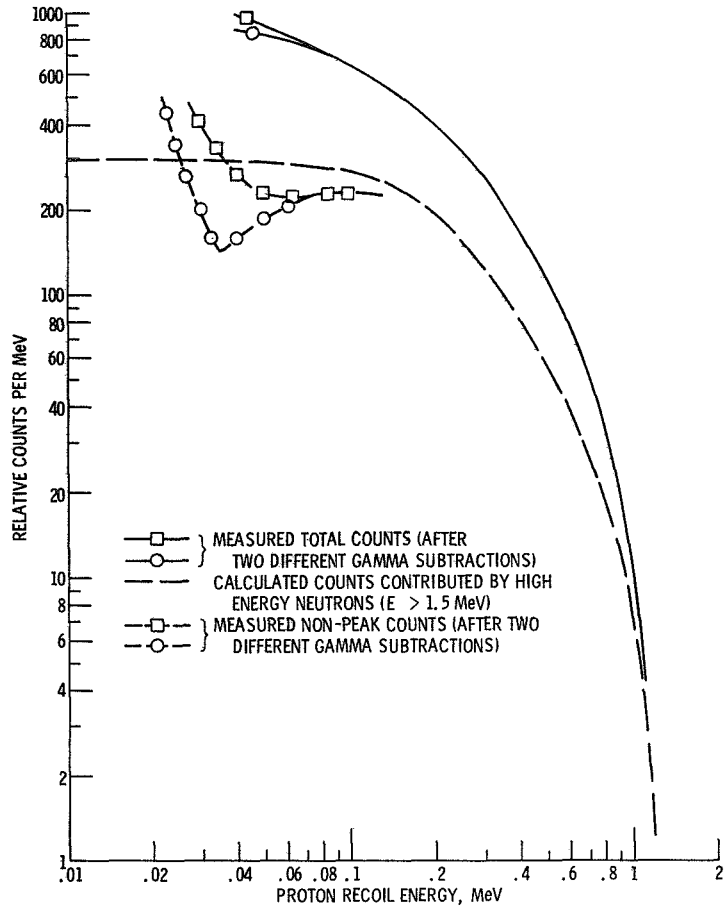Figure 4. - Proportional counter data, 5 Ci-Pu-Be source moderated by tungsten.

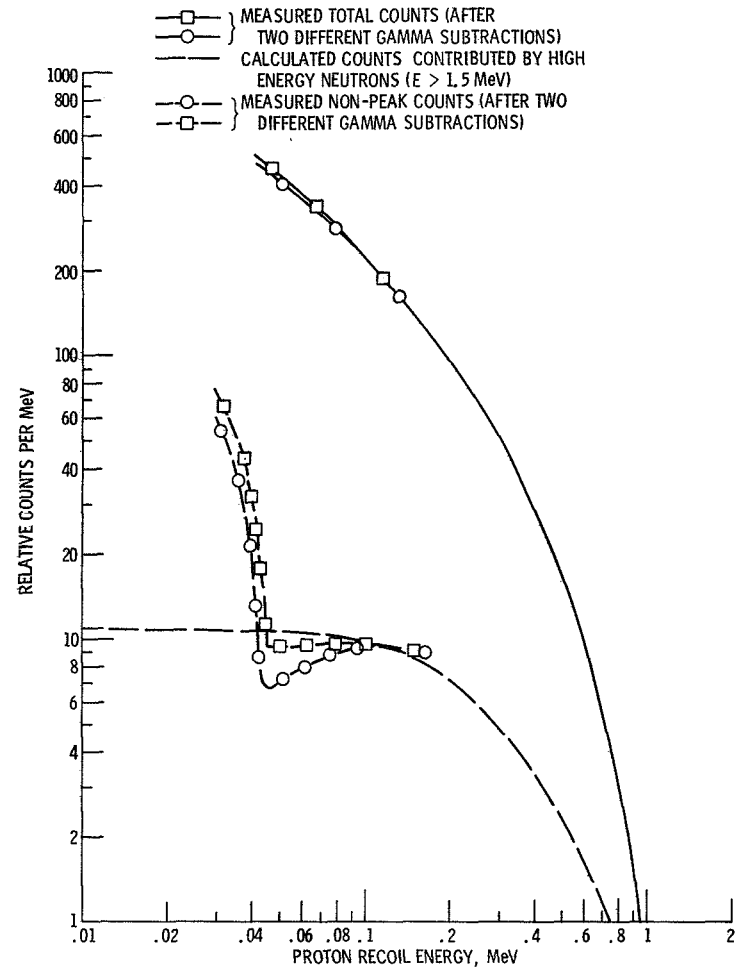Figure 5. - Comparison of measured with calculated count rates for 5-Ci Pu-Be source.

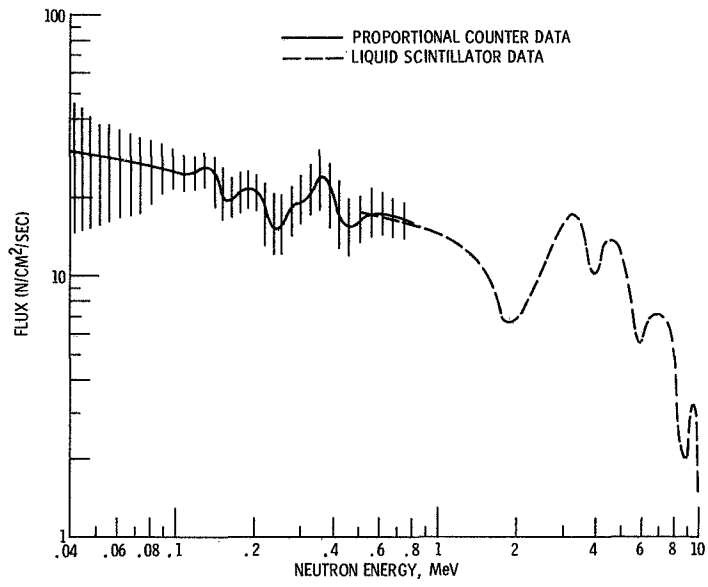Figure 6. - Comparison of measured and calculated count rates for tungsten leakage flux.

Figure 7. - Neutron flux at one meter from 5 Ci Pu-Be source.