MLM-1422

NEUTRON ENERGY SPECTRA OF ^{239}Pu -Be, ^{238}Pu -F,
AND ^{238}Pu - ^{18}O (α , n) SOURCES

MASTER

M. E. Anderson

AEC Research and Development REPORT

MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO COMPANY

M O U N D L A B O R A T O R Y

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S. GOVERNMENT CONTRACT NO. AT-33-1-GEN-53

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

MLM-1422
TID-4500
UC-34 Physics

**NEUTRON ENERGY SPECTRA OF ^{239}Pu -Be, ^{238}Pu -F,
AND ^{238}Pu - ^{18}O (α , n) SOURCES***

M. E. Anderson

Date: February 4, 1967

Issued: October 31, 1967

*Paper presented in part at the 1966 Winter Meeting of the American Nuclear Society in Pittsburgh, Pa.

MONSANTO RESEARCH CORPORATION

A Subsidiary of Monsanto Company

MOUND LABORATORY

Miamisburg, Ohio

operated for

UNITED STATES ATOMIC ENERGY COMMISSION

U S GOVERNMENT CONTRACT NO AT 33-1-GEN 53

TABLE OF CONTENTS

	Page
Summary	3
Introduction	4
Data Reduction	5
Neutron Energy Spectra	
Spectrum of a ^{239}Pu -Be(α ,n) Source	7
Spectrum of a ^{238}Pu -F(α ,n) Source	8
Spectrum of a ^{238}Pu - ^{18}O (α ,n) Source	9
Conclusions	11
References	12

SUMMARY

The energy spectra of isotopic neutron sources are being determined with a single stilbene crystal, fast neutron spectrometer. Pulse shape discrimination is used to separate pulses originating from neutron interaction and those originating from gamma interactions. Spectra were obtained for (α ,n) sources containing plutonium-239 with beryllium, plutonium-238 with fluorine, and plutonium-238 with oxygen-18.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission.

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

INTRODUCTION

At Mound Laboratory a single stilbene crystal, fast neutron spectrometer has been used for determining energy spectra of isotopic neutron sources. In such a spectrometer, neutrons are detected by measurements on protons which have been elastically scattered in the crystal. For a beam of monoenergetic neutrons, there will be a distribution of pulse heights varying from (essentially) zero up to a maximum corresponding to the energy of the incident neutrons. Pulse shape discrimination¹ is used to separate pulses originating from neutron interactions and those originating from gamma interactions. Figure 1 is an example of some pulse height data for a ^{239}Pu -Be source. It is necessary to unfold these data to obtain the desired neutron energy spectrum.

Figure 1. Pulse Height Data for Neutron Energy Spectrum of Source M-591.

DATA REDUCTION

Basically, there are two ways of unfolding these data. One is to use a set of experimentally determined response functions obtained by exposing the crystal to beams of monoenergetic neutrons.^{2,3} The other way is to infer the crystal response in a less direct manner. We use the latter method, primarily because we do not have an accelerator to produce the needed monoenergetic neutron beams. However, results show that this does not seem to be a serious disadvantage.

Most of the neutrons of interest have energies less than 10 Mev. This, combined with our small crystal size (1 cm radius x 1 cm height), which limits the numbers of neutrons which scatter twice to only a few percent,⁴ results in most of the light pulses from neutron interactions being those from elastically scattered protons.

Other investigators^{4,5} have shown that the light output for protons scattered at an angle of 0° is essentially proportional to the proton energy raised to a power, with the exponent being between 1.3 and 1.5. A response function assuming this relation between the light output and proton energy is shown as the top curve in Figure 2 for the case of perfect energy resolution. (Had the light output been linearly proportional to proton energy, such a curve would be a horizontal line on this graph at a relative intensity of 1.0.)

Figure 2. Response Functions for Monoenergetic Neutrons Showing the Effect of the Dependence of the Directions of Travel of Protons within the Crystal.

We, as well as others,^{3,5,6,7} have observed that the maximum light output for a given neutron energy is a function of the orientation of the stilbene crystal to the neutron flux. (For our crystal the extremes in light output as a function of crystal orientation differ by about 18%.) A consequence of this dependence of the light output on crystal orientation is that the light output for a given proton energy is a function of the direction of travel of the proton within the crystal. This directional dependence affects the pulse height distribution and is illustrated by the bottom line in Figure 2. This lower response function is basically the one being used for unfolding the pulse height data.

Corrections for second scattering within the crystal and for protons leaving the crystal before expending all of their energies are made by using the approximate formula given by Broek and Anderson.³

NEUTRON ENERGY SPECTRA

SPECTRUM OF A $^{239}\text{Pu}\text{-Be}(\alpha,n)$ SOURCE

The first spectrum, that of a $^{239}\text{Pu}\text{-Be}$ source, is shown in Figure 3. Shown also in this figure are the results of an emulsion experiment (crosses) for this same source which was performed a few years ago.⁸ (All error bars shown in this and other figures are plus or minus one standard deviation.) It was stated earlier that the light output for protons scattered at 0° is approximately proportional to the proton energy raised to a power, with the power variously reported to be from 1.3 to 1.5. By adjusting b in the relationship $L \propto E^b$ until the best match between the stilbene and emulsion data was obtained, the value of b was found to be about 1.38.

Figure 3. Neutron Energy Spectrum of $^{239}\text{Pu}\text{-Be}(\alpha,n)$ Source M-591 Containing 80 g of Plutonium.

Some details about these data should be pointed out. (For purposes of plotting, these two sets of data were normalized so that the areas under both curves above an energy of 1.5 Mev are the same.) The statistical uncertainty (not to be confused with absolute uncertainty) of each stilbene data point varies from about 1-2% near the low-energy end to about 10% near 11 Mev. Note the intensity maximum near 6.75 Mev. This maximum was also seen in the emulsion data. However, when the emulsion experiment was done, there was not enough confidence in the data to cause one to believe there were two maxima between 6 and 8 Mev; so when the smooth curve was drawn through the points only one maximum was shown in this region. Two peaks in this region have also been seen by Sayers.⁹ Additional maxima in the region from 1.0 to 2.5 Mev are suggested by the stilbene data. The scatter of these data points is more than would be expected from statistical uncertainties, however, experimental uncertainty is not ruled out. Maxima in this region could result when the residual carbon nucleus from the $^9\text{Be}(\alpha,n)^{12}\text{C}$ reaction is left in its 7.66-Mev state.⁸

This spectrum is for a source containing 80 g of plutonium and is not necessarily the spectrum one would get for a ^{239}Pu -Be source of another size.⁸ Also, americium-241 is growing into this source¹⁰ from the decay of plutonium-241. Presently about 8% of the total neutron emission results from americium alpha particles while these alpha particles only accounted for about 2% of the emission at the time of the emulsion experiment. The minima in a ^{241}Am -Be spectrum are not as pronounced as that of a ^{239}Pu -Be source because of the difference in alpha particle energies.¹¹ Some of the differences between the stilbene data and the emulsion data may not be simply experimental errors but may be due to real differences in the spectra of this source obtained several years apart.

SPECTRUM OF A ^{238}Pu -F(α ,n) SOURCE

Another spectrum being studied is that of a ^{238}Pu -F source. Some data for this spectrum are shown in Figure 4. The theoretical maximum neutron energy for the $^{19}\text{F}(\alpha,n)^{22}\text{Na}$ reaction with 5.5-Mev alpha particles from plutonium-238 is 3.2 Mev. There are neutrons with energies greater than 3.2 Mev from spontaneous fission of plutonium-238. Neutrons resulting when sodium-22 is left in the ground state or one of the first three excited states account for most of the spectrum shown. Theoretically sodium-22 may be left in excited states up to and including 2.57 Mev. The character of the spectrum for energies less than those shown in these data would depend on what fraction of the sodium-22 is left in these higher states.

Figure 4. Neutron Energy Spectrum of a ^{238}Pu -F(α ,n) Source

No other spectrum of a ^{238}Pu -F source has been published, but the spectrum of a ^{210}Po -F source should not differ greatly since the alpha energies of plutonium-238 and polonium-210 differ by only 0.2 Mev. Figure 5 shows a comparison between our data and the emulsion data of Szilvasi, Geiger and Dixon.¹²

Figure 5. Neutron Energy Spectra of $^{19}\text{F}(\alpha, n)$ Sources.

SPECTRUM OF A $^{238}\text{Pu}-^{18}\text{O}(\alpha, n)$ SOURCE

The final spectrum is that of a $^{238}\text{Pu}-^{18}\text{O}$ source. These data are shown in Figure 6. The isotopic composition of the oxygen in this source is: oxygen-16 - 52.75%; oxygen-17 - 1.66%; and oxygen-18 - 45.61%. Not readily seen on this curve is the high-energy tail due to spontaneous fission of plutonium-238. The maximum near 2.3 MeV results from the product nucleus neon-21 being left in the ground or first excited state at 0.35 MeV.

Figure 6. Neutron Energy Spectrum of a $^{238}\text{Pu}-^{18}\text{O}(\alpha, n)$ Source.

The rise below 1.5 Mev is, at least partly, due to neon-21 being left in the second excited state at 1.73 Mev. It is also energetically possible for the neon-21 to be left in both the 2.80 and 3.73-Mev states, though the cross section for the latter would be very small, indeed. If the 2.80-Mev state were significantly populated, this would also help to explain the rise in intensity for low neutron energies. The statistical accuracy for these data is approximately 2%.

Again we have no spectrum with which to directly compare our results, but a ^{210}Po - ^{18}O spectrum was obtained by Khabakhpashev¹³ a few years ago using two scintillation detectors in a time-of-flight technique. This comparison is shown in Figure 7.

Figure 7. Neutron Energy Spectra of $^{18}\text{O}(\alpha,n)$ Sources.

CONCLUSIONS

In conclusion, this spectrometer system does give satisfactory results which compare well with spectra obtained with nuclear emulsions. For the sources discussed here, each of whose emission is greater than 10^6 neutrons/sec, reasonable spectra may be obtained with only one hour of counting time for each. The data shown in this report were obtained with counting times greater than one hour. Data reduction requires only a few minutes using an IBM 360 computer.

REFERENCES

1. F. A. Johnson, **Can. J. Phys.**, **41**, 793 (1963).
2. B. Brunfelter, J. Kockum and H. O. Zetterstrom, **Nucl. Instr. Methods**, **40**, 84 (1966).
3. V. V. Verbinski, J. C. Courtney, W. R. Burris and T. A. Love, "Proceedings of a Special Session on Fast Neutron Spectroscopy", **1964 Winter Meeting of the American Nuclear Society**, ANS-SD-2, 189 (1964).
4. H. W. Broek and C. E. Anderson, **Rev. Sci. Instr.**, **31**, 1063 (1960).
5. J. B. Marion and J. L. Fowler, (ed.), **Fast Neutron Physics**, Part I, New York, Interscience Publishers, Inc., 1960, p. 228.
6. K. Tsukada and S. Kikuchi, **Nucl. Instr. Methods**, **17**, 286 (1962).
7. M. McN. Wasson, AERE-R 4269 (1963) (Unpublished).
8. M. E. Anderson and W. H. Bond, Jr., **Nucl. Phys.**, **43**, 330 (1963).
9. A. Sayers, Columbia University, Private Communication (1964).
10. M. E. Anderson, **Trans. Am. Nucl. Soc.**, **7**, No. 2, 246 (1964).
11. K. W. Geiger and C. K. Hargrove, **Nucl. Phys.**, **53**, 204 (1964).
12. A. J. D. Szilvasi, K. W. Geiger and W. R. Dixon, **J. Nucl. Energy**, **11**, 131 (1960).
13. A. G. Khabakhpashev, **Soviet J. At. Energy, English Transl.**, **7**, No. 1, 591 (1960).