

5.5-msec transition is that it is  $\Delta I=2$ , no change in parity, which would make it electric quadrupole. Then one would have to hypothesize a low-energy gamma in the spectrum, which was the half-life-determining transition.

#### CONCLUSIONS AND SUMMARY

From the bombardment of several common elements with 32-Mev protons, only one gamma emitter with half-life between 2 msec and 25 msec was found. On the basis of thresholds for neighboring activities and shapes of their excitation curves and that of the

new activity, this 5.5-msec half-life activity from  $p+Ta$  has been tentatively assigned as a metastable state of stable  $W^{180}$ . No sure assignment can be made to the transition concerning spin and parity.

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## Neutron Spectrum and Absolute Yield of a Plutonium-Beryllium Source\*

LEONA STEWART

*University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico*

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A Pu-Be neutron source has the advantages of high neutron yield, low gamma-ray intensity, and very long half-life. The neutron spectrum from such a source, composed of 13 g of plutonium and 7 g of beryllium as  $PuBe_{13}$  (density 3.7 g/cc), was measured by means of proton recoils in nuclear research emulsions. The data obtained from 2057 observations indicate the neutrons have a maximum energy of approximately 10.5 Mev with broad intensity maxima at 4.0, 7.2, and 9.7 Mev. The observed spectrum shows marked similarities with published data on neutrons from Po-Be and Ra-Be sources. A comparison is made between the present data and previously reported energy levels in the residual carbon nucleus. The absolute yield of neutrons above 0.5-Mev energy is  $1.2 \times 10^9$  neutrons/sec.

### I. INTRODUCTION

ALTHOUGH neutron sources employing the  $(\alpha, n)$  reaction have been in use for over two decades, few precise spectral measurements have been reported. Of the early work,<sup>1</sup> probably the most widely known are the data of Richards and of Demers obtained by proton recoil measurements in photographic plates. Both these measurements were made with the old Ilford half-tone emulsions which were characterized by low silver halide concentration and consequently poor energy resolution. In addition, the energy calibration of these emulsions was not accurately known. It is not surprising therefore to find some disagreement in these early measurements.

More recently, Whitmore and Baker,<sup>2</sup> Guier, Bertini,

and Roberts,<sup>3</sup> Gursky, Winnemore, and Cowan,<sup>4</sup> and Elliot, McGarry, and Faust<sup>5</sup> have used various methods<sup>6</sup> to obtain quite precise data on Po-Be sources by making use of the many refinements achieved during the past decade in neutron spectrum measurements. (For a discussion of these methods see Barschall, Rosen, Taschek, and Williams<sup>7</sup> and Rosen.<sup>8</sup>)

The most accurate work appearing in the literature on Ra-Be was done by Hill,<sup>9</sup> who made ionization chamber measurements employing a coincidence-anticoincidence arrangement which permitted high statistical accuracy as well as good resolution.

Although Po-Be is presently more widely used as a neutron source because of its availability, Pu-Be possesses many advantages over Po-Be and may be expected to replace it where neutron intensity is not the determining factor. Po-Be is a mixture, and the spectrum therefore varies with grain size. Even after

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<sup>1</sup> N. Feather, Proc. Roy. Soc. (London) **A142**, 3 (1933); P. Auger, J. phys. radium **4**, 749 (1933); J. R. Dunning, Phys. Rev. **45**, 586 (1934); M. Blau, J. phys. radium **5**, 61 (1934); T. W. Bonner and Mott-Smith, Phys. Rev. **46**, 258 (1934); G. Bernardini and D. Bocciarelli, Atti. accad. nazl. Lincei **24**, 132 (1936); G. Bernardini, *Kernphysik Vortrage*, Zurich, 1936 (unpublished); Perlman, Richards, and Speck, Atomic Energy Commission Report MDDC-39, 1943 (unpublished); H. T. Richards, Atomic Energy Commission Report MDDC-1504, 1944 (unpublished); P. Demers, Montreal Report MP-74 and MP-204, 1945 (unpublished).

<sup>2</sup> B. G. Whitmore and W. B. Baker, Phys. Rev. **78**, 799 (1950).

<sup>3</sup> Guier, Bertini, and Roberts, Phys. Rev. **85**, 426 (1952).

<sup>4</sup> Gursky, Winnemore, and Cowan, Phys. Rev. **91**, 209 (1953).

<sup>5</sup> Elliot, McGarry, and Faust, Phys. Rev. **93**, 1348 (1953).

<sup>6</sup> The first two measurements were made with photographic emulsions, the second with an ionization chamber, and the third with a coincidence spectrometer.

<sup>7</sup> Barschall, Rosen, Taschek, and Williams, Revs. Modern Phys. **24**, 1 (1952).

<sup>8</sup> L. Rosen, Nucleonics **11**, No. 7, 32 and No. 8, 38 (1953).

<sup>9</sup> D. L. Hill, Atomic Energy Commission Report AECD-1945, 1947 (unpublished).

the source is fabricated, the extent of mixing changes due to large heating effects from the polonium decay. These effects are very pronounced in compact sources of high yields. Furthermore, the short half-life of polonium (138.2 days) necessitates time-dependent corrections and eventually makes it necessary for the source to be discarded. Pu-Be, on the other hand, is an intermetallic compound, and the spectrum consequently does not depend upon grain size; in addition, its relatively long half-life ( $2.2 \times 10^4$  years), assures constancy with time. Pu-Be may even be preferable to Ra-Be where cost is a serious factor and in experiments where gamma rays constitute a liability. In view of the above comparison, a careful determination of the neutron spectrum of Pu-Be is of interest.

## II. EXPERIMENTAL PROCEDURE

### A. Method

Iford C2 plates were placed approximately 17 cm from a Pu-Be source with the longitudinal axis of the plate parallel to that of the cylindrical source. The source and plates were supported by lightweight nylon thread fixed taut by weights, and the whole assembly was suspended in the center of a canvas structure where the nearest scattering material consisted of the small iron supports more than 7 ft from the center.

In general two plates were exposed at the same time. They were wrapped in two layers of 0.005-in. black paper with the emulsion surfaces separated by a thin platinum strip, the purpose of which was to prevent recoil protons starting in one emulsion from passing into the other. Background plates underwent the same history as the data plates except for the exposure to the neutron source. Both 200- and 400- $\mu$  plates were exposed for approximately one week. These were developed immediately after exposure according to the procedures outlined in reference 8.

The analysis of the plates was carried out by measuring the projected ranges of the proton recoils which proceeded in a direction away from the source within a half-angle of 12 deg. These ranges were then converted to true range either by making use of the average angle of acceptance or (in the case of 60 percent of the tracks) by measuring the horizontal and dip angles and converting to true range for each individual event using an IBM calculator. The emulsion thickness was measured before and after processing in order to obtain an "apparent" shrinkage factor<sup>8</sup> for determining the dip angle in the unprocessed emulsion.

### B. Reduction of the Data

In performing neutron flux measurements using photographic emulsions, it is necessary to evaluate quantitatively a rather imposing number of factors which enter into an absolute determination of the flux of neutrons as a function of energy incident on the detector. The time-integrated flux in the energy interval,

$E_n$ , which projects a number of protons,  $N_p(E_p)$ , in the emulsion is given by:

$$F(E_n) = \frac{4\pi}{n_0 t A \Omega_{\text{lab}} 4 \cos^2 \theta} \frac{N_p(E_p) P_c(E_p) T_c(E_n)}{\sigma_{n-p}(E_n)}, \quad (1)$$

where  $F(E_n)$  = number of neutrons/cm<sup>2</sup>,  $n_0$  = number of hydrogen atoms/cc,  $t$  = thickness of emulsion in cm,  $A$  = area of emulsion analyzed in cm<sup>2</sup>,  $\Omega_{\text{lab}}$  = solid angle of acceptance of tracks in emulsion,  $N_p(E_p)$  = number of protons measured in the energy interval  $E_p$  corresponding to the neutron energy interval  $E_n$ , where for a given collision  $E_p = E_n \cos^2 \theta$ ,  $\theta$  being the angle between the directions of the incident neutron and the projected proton,  $P_c(E_p)$  = probability correction for tracks leaving the emulsion,  $T_c(E_n)$  = attenuation correction for neutrons traversing an average distance in the emulsion and/or glass backing, and  $\sigma_{n-p}(E_n) = n-p$  total scattering cross section at energy  $E_n$ .

The range-energy relations for protons in nuclear emulsions have been established<sup>10-13</sup> to an accuracy of better than 2 percent. The number of hydrogen atoms present in the dry emulsion is known from the manufacturer's specifications. When the emulsion is not used in a vacuum, however, this quantity is a function of relative humidity. Recently, Webb<sup>14</sup> performed a series of measurements from which it is possible to calculate the correction to be applied to the dry emulsion. (Webb's data check very closely a similar analysis performed at Los Alamos by A. R. Ronzio.) The  $n-p$  scattering cross section is known to be isotropic up to 14 Mev and has been accurately determined by transmission measurements<sup>15</sup> up to this energy.

The probability corrections, which must be applied for tracks leaving the emulsion, have heretofore been calculated on the assumption that the tracks proceed in a straight line,<sup>16</sup> that is, do not suffer scattering. This assumption is not adequate at energies greater than 4 Mev and leads to a systematic error which increases with energy. Accurate corrections have recently been determined at Los Alamos which take multiple scattering into account.<sup>8</sup> The evaluation of  $\Omega_{\text{lab}}$  implies that measurement criteria must be established which give precise delineation of the rectangular pyramid of acceptance of the proton recoil tracks in the unprocessed emulsion. These measurement criteria were  $\pm 11$  deg for both the horizontal and dip angles.

Since the mean free path of neutrons in emulsion and/or glass backing is quite small, and since the

<sup>10</sup> Lattes, Fowler, and Cier, *Nature* **159**, 301 (1947).

<sup>11</sup> J. Rotblat, *Nature* **167**, 550 (1951).

<sup>12</sup> Bradner, Smith, Barkas, and Bishop, *Phys. Rev.* **77**, 462 (1950).

<sup>13</sup> J. J. Wilkins, Atomic Energy Research Establishment, Harwell Report AERE-GR 664, 1951 (unpublished).

<sup>14</sup> J. H. Webb (private communication).

<sup>15</sup> *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (Technical Information Division, Department of Commerce, Washington, D. C., 1952).

<sup>16</sup> H. T. Richards, *Phys. Rev.* **59**, 796 (1941).

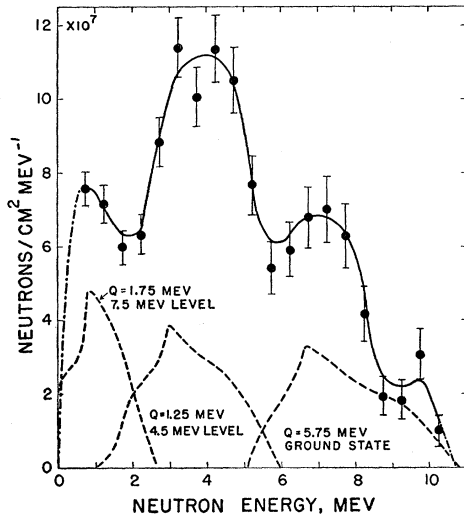


FIG. 1. Energy distribution of the neutrons from Pu-Be.

average distance from the center of the swath read to the edge of the plate was considerable (this distance varied from 0.75 to 3.85 cm), it was necessary to apply an attenuation correction<sup>17</sup> which, on the average, amounted to about 23 percent of the neutron flux. (For a detailed discussion of the difficulties involved and the procedures used in performing absolute flux measurements see reference 8.)

In the analysis of the background plates, in which a volume of emulsion was scanned equal to that analyzed in the exposed plates, no tracks were observed which satisfied the imposed angular criteria. A single track observed in this volume would have indicated an integrated flux of background neutrons of the order of  $10^5$  neutrons/cm<sup>2</sup>.

In order to calculate the neutron flux as a function of energy, the data for each of three exposures were tabulated separately and Eq. (1) was used. The source strength is then given by:

$$Qt = 4\pi R^2 F_n, \quad (2)$$

where  $Qt$  is the number of neutrons emitted in  $4\pi$  solid angle in time  $t$ ,  $R$  is the distance between source and detector, and  $F_n$  is the number of neutrons/cm<sup>2</sup> incident on the detector. The final energy distribution is shown in Fig. 1.

The average value of  $Q$  obtained in this manner from three sets of data was  $1.22 \times 10^6$  neutrons/sec as compared with the source calibration<sup>18</sup> of  $1.24 \times 10^6$ . It is seen from Fig. 1 that neutrons below 0.5 Mev were not detected. If one extrapolates the data in this region, it is reasonable to account for about 5 percent of the neutron flux, which would then give a value for  $Q$  of  $1.28 \times 10^6$ .

<sup>17</sup> This correction was inadvertently omitted in the initial report [Phys. Rev. 92, 1095 (1953)].

<sup>18</sup> A. H. Spano (private communication).

Consideration of the errors in this experiment indicates that the above agreement must be considered fortuitous. The errors shown in Fig. 1 are statistical standard deviations. Other errors encountered in determining the results are:

$\sigma_{n-p}(E_n)$	2 percent	$\Omega_{\text{lab}}$	10 percent
$P_c(E_p)$	4 percent	$t$	5 percent
$T_c(E_n)$	5 percent	Geometry	2 percent
$n_0$	5 percent		

It should be pointed out, however, that the errors in  $n_0$ ,  $\Omega_{\text{lab}}$ ,  $t$ , and the experimental geometry do not affect the relative neutron distribution.

### III. DISCUSSION OF RESULTS

The energy distribution shown in Fig. 1 was obtained from observation of 2057 tracks by two microscopists. The results, tabulated independently, were in agreement within statistical uncertainties. From energy and momentum considerations, the upper limit of this spectrum should be approximately 10.6 Mev. The longest track recorded corresponds to a neutron energy of 10.2 Mev. This discrepancy is not unreasonable if one considers the relatively small number of neutrons with energies greater than 10 Mev. The effective energy,  $E_{\text{eff}}$ , of the neutrons from these results, is given by

$$E_{\text{eff}} = \frac{\int_0^{10.5 \text{ Mev}} E_n F_n(E_n) dE_n}{\int_0^{10.5 \text{ Mev}} F_n(E_n) dE_n} = 4.4 \text{ Mev.} \quad (3)$$

The average energy,  $E_{\text{Av}}$ , defined by

$$\int_0^{E_{\text{Av}}} F_n(E_n) dE_n = \frac{1}{2} \int_0^{10.5 \text{ Mev}} F_n(E_n) dE_n \quad (4)$$

is 4.2 Mev.

A comparison with published data on Po-Be and Ra-Be shows marked similarities, especially when the differences in the energies of the incident alpha particles are taken into account. Whitmore and Baker<sup>2</sup> establish intensity maxima from Po-Be at 3.2, 4.8, 7.7, and probably 1.2 Mev; doubtful maxima are also suggested at 5.7 and 9.7 Mev. The present experiment is indicative of a single broad maximum from 3 to 5 Mev, a minimum between 5.5 and 6.0 Mev, another maximum at 7.5 Mev, and perhaps still another at 9.6 Mev. It is interesting to note that Richards' data<sup>1</sup> (Po-Be) show a minimum around 6 Mev, relatively good agreement with the present data at higher energies, but lack of agreement below 5 Mev.

The data of Gursky, Winnemore, and Cowan<sup>4</sup> on Po-Be (only neutrons above 2.5 Mev were detected) again agree with reference 2 except for the reported maximum at 7.7 Mev. The work of Elliot, McGarry,

and Faust<sup>6</sup> appears to be in disagreement with the present results as well as other recent measurements on Po-Be in that their data show maxima displaced by  $\frac{1}{2}$  to 1 Mev and also a high-energy tail which exceeds the maximum energy possible.

The data (Po-Be) of Guier, Bertini, and Roberts<sup>8</sup> are not comparable since a thin target was used, giving rise to a discrete range of alpha-particle energies. (Demers' data<sup>2</sup> on Po-Be show maxima only at 5.2 and 10.1 Mev.) Hill's data on Ra-Be show fair agreement with reference 2 at the higher energies except for an apparent minimum around 6 Mev which also shows up in the experiment reported here.

Many workers<sup>19</sup> have reported various energy levels in the residual nucleus from the  $\text{Be}^9(\alpha, n)\text{C}^{12}$  reaction. A calculation was made to determine the "expected distribution" on the basis of levels in  $\text{C}^{12}$  at 4.5 and 7.5 Mev. In order to carry out the calculations one must take into account the excitation function for the  $(\alpha, n)$  reaction in beryllium. An alpha particle of a given incident energy will liberate neutrons of varying energies and the maximum and minimum energies are easily calculated from the dynamics of the collision. Therefore, the relative number of neutrons for a given energy interval was calculated by numerical integration from

$$F(E_n)dE_n = dE_n \int_{E_{\alpha_1}}^{E_{\alpha_2}} \frac{f(E_\alpha)}{R(E_n)} dE_\alpha, \quad (5)$$

where  $(E_{\alpha_1}$  to  $E_{\alpha_2})$  is the range of energies of the alpha particles corresponding to the emitted neutron energies  $E_n$  to  $E_n + dE_n$ ,  $R(E_n)$  is the energy range over which

<sup>19</sup> F. Ajzenberg and T. Lauritsen, *Revs. Modern Phys.* **24**, 321 (1952).

the neutrons are distributed, and  $f(E_\alpha)$  is the excitation function for the  $(\alpha, n)$  reaction in beryllium reported by Halpern.<sup>20</sup> The results are shown underlying the experimental data in Fig. 1.

It should be noted that if the angular distribution of the emitted neutrons is anisotropic, as it may well be, these calculations would give little indication of the true spectrum. These data also assume that the transition probabilities are equal for all the possible excited states of  $\text{C}^{12*} \rightarrow \text{C}^{12*} + n$ . In order to understand the small maximum between 9 and 10 Mev, where the leaving of  $\text{C}^{12}$  in the ground state is the only likely possibility, a predominately forward distribution would be a reasonable assumption. An approximate calculation using a nuclear radius of  $2 \times 10^{-13}$  cm indicates an interaction with one or two units of angular momentum for the most energetic alpha particles. Bradford and Bennett<sup>21</sup> also suggest the possibility of different angular distributions for the two groups of neutrons observed when beryllium is bombarded with 1.4-Mev alphas. Due to the lack of knowledge concerning the angular distributions, no attempt is made to fix specific energy levels in  $\text{C}^{12}$ . The statistical accuracy of this experiment would also discourage such a detailed analysis.

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<sup>20</sup> I. Halpern, *Phys. Rev.* **76**, 248 (1949).

<sup>21</sup> C. E. Bradford and W. E. Bennett, *Phys. Rev.* **78**, 302 (1950).