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PHOTO-NEUTRON SOURCES

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FOREWARD

This report was written to be part of a general treatise on neutron measurements. It is planned to publish the complete work at a later date, but it is believed that earlier distribution of this material through the National Research Council is desirable in order to take advantage of criticisms, comments, suggestions and new data. The author of this report will welcome such advice.

DISCLAIMER

PHOTO-NEUTRON SOURCES*

Introduction

A photon falling upon a nucleus can cause the ejection of a neutron if the energy of the photon is greater than the energy with which the neutron is bound in the nucleus. The most useful sources of photons are radioactive elements, which emit gamma rays whose energies are always less than 4 Mev. In most nuclei, the binding energy of the neutron is from 6 to 10 Mev. There are only two known nuclei with binding energies below 4 Mev -- Be^9 and D^2. Consequently, the photo-neutron sources which have been employed to date have used either beryllium or deuterium.

The main advantage of photo-neutron sources is that the emitted neutrons are monoenergetic. The chief disadvantages are the low yields of neutrons from such sources and the difficulties which are associated with the presence of high intensity gamma radiation.

High energy X-ray machines and betatrons are sources of energetic photons. Photo-neutron sources employing the photons produced by these machines are useful. However, as the energy spectra of the photons from such machines are continuous, such sources do not yield monoenergetic neutrons.

The Energy Equation

The photo-disintegration reactions in beryllium and deuterium are

$$ {}_4Be^9 + \gamma \rightarrow {}_4Be^8 + {}_0n^1 $$

$$ {}_1D^2 + \gamma \rightarrow {}_1H^1 + {}_0n^1 $$

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These reactions are endothermic; the thresholds are 1.63 Mev for beryllium (1) and 2.18 Mev for deuterium (2).

Due to the endothermic nature of these reactions, monoenergetic photons produce a monoenergetic group of neutrons. From the conservation of energy and momentum, the energy of the neutrons is given by

$$E_n = \frac{A - 1}{A} \left[E_\gamma - Q - \frac{E_\gamma^2}{1862\ (A - 1)} \right] + \delta \qquad (1)$$

where E_n is the energy of the neutrons in Mev, A is the mass number of the target nucleus, E_γ is the energy of the gamma rays in Mev, and Q is the threshold energy in Mev for the (γ,n) reaction in the nucleus of mass A. δ is a small spread in energy and is a function of the angle θ between the direction of the impinging gamma ray and the direction in which the neutron is emitted; it is given by

$$\delta \simeq E_\gamma \left[\frac{2(A - 1)\ (E_\gamma - Q)}{931 \times A^3} \right]^{\frac{1}{2}} \cos \theta \qquad (2)$$

If the gamma rays fall upon the target nucleus in an isotropic manner, there is an inherent spread in the energy of the emitted neutrons which is given by

$$\delta_{max} = 2E_\gamma \left[\frac{2(A - 1)\ (E_\gamma - Q)}{931 \times A^3} \right]^{\frac{1}{2}} \qquad (3)$$

This spread occurs with most sources which have been used.

The fractional spread δ_{max}/E_n decreases with increasing neutron energy; it is of the order of five to ten times greater in deuterium than in beryllium sources. For 100 kev neutrons, the fractional spread is about 4% in beryllium and 25% in deuterium. For neutrons from Be of energy greater than 100 kev, δ_{max} is generally negligible compared to other causes of energy spread.

One can avoid this inherent spread in energy by constructing the photo-neutron source so that the gamma-ray emitter subtends only a small solid angle at the target. However, such an arrangement greatly reduces the actual yield of photo-neutrons.

Gamma-Ray Emitter

For a gamma-ray emitter to be useful in a photo-neutron source, the energy of the gamma ray must exceed 1.63 Mev. The literature on the energies of gamma rays is probably still incomplete. Listed in Table I are thirteen artificially produced radioisotopes and two natural radioisotopes which are known to satisfy the energy requirement. Their half-lives, gamma-ray energies, and the nuclear reactions by which they can be produced are also listed. The mixture of radioisotopes produced in fission can also be used as a source of gamma rays to produce photo-neutrons (3). Table I does not include some extremely low intensity high energy gamma rays which were recently observed (4). With the exception of the natural radioisotopes and Sc^{44}, Zn^{63}, and Y^{88}, the isotopes listed in Table I can be produced by slow neutron capture and, therefore, can be manufactured in chain-reacting piles. The last column of Table I lists the neutron capture cross section for the production of the radioactive isotope (for its natural isotopic abundance).

Those sources with half-lives shorter than one hour are generally not very useful, and, unless there is a radioisotope producing machine nearby, those with half-lives shorter than one day will be difficult to use.

In order that a photo-neutron source emit a monoenergetic group of neutrons, the radioisotope must emit only one gamma ray with an energy greater than 1.63 Mev if beryllium is the target, or greater than 2.185 Mev if deuterium is the target. For example, MsTh in equilibrium with its decay products is not a monoenergetic source of neutrons with a beryllium target, but it is with a deuterium target.

Energy of Neutrons

Using the energies of the gamma rays listed in Table I, and the Q values for Be and D, one can estimate from Eq. 1 the energies of the photo-neutrons from various sources. The second column of Table II lists these energies.

It is rather difficult in practice to obtain exactly these theoretical values in sources which have a high enough yield of neutrons to be useful. To get an appreciable yield, one must employ a source of finite size. Some of the neutrons suffer collisions before escaping from the source, and these collisions cause a decrement in the energy of the escaping neutrons. Column three of Table II lists observed energies of the neutrons emitted from sources employed by some workers in this field.

Many techniques are available for determining the energy of the neutrons emitted by sources. Probably the best methods are those which measure the energy spectrum by means of examining the recoil protons, such as the photographic plate technique or the cloud chamber technique of Hughes and Eggler (14).

Hughes and Eggler measured the range of the protons produced by head-on collisions of neutrons. The range, when converted to proton energy, gives the neutron energy directly, since all the energy of the neutron goes to the proton in a head-on

TABLE I

Useful Gamma-Ray Emitters

Radio-isotope	Half-life (5)	Gamma-Ray Energy* (Mev)	Nuclear Reactions for Producing Isotope (5)	Cross section for (n,γ) Reaction (barns)
F^{20}	12 s	2.2 (5)	F(d,p), F(n,γ), Na(n,α),	0.01 (18)
Na^{24}	14.8 h	2.76 (6)	Mg (n,p), Mg(d,α), Na(n,p), Na(n,γ), Al(n,α)	0.41 (19)
Al^{28}	2.4 m	1.8 (5, 7)	p(n,d), Mg(α,p), Mg(α,n), Al(d,p), Al(n,γ), Si(n,p)	0.23 (18)
Cl^{38}	37 m	2.15 (8) (1.65 ?)	Cl(d,p), Cl(n,γ), K(n,α)	0.15 (18)
Sc^{44}	4.1 h	1.80 (5)	Sc(γ,n), Sc(n,2n), K(α,n), Ca(d,n)	
Mn^{56}	2.59 h	1.81, 2.13, 2.7 (9, 10)**	Fe(n,p), Ce(n,α), Cr(α,p), Mn(n,γ), Mn(d,p), Fe(d,α)	10.7 (18)
Zn^{63}	38 m	1.9 (11)	Cu(p,n), Ni(α,n), Zn(n,2n), Zn(γ,n)	
Ga^{72}	14.1 h	1.87, 2.21, 2.51 (12)	Ga(d,p), Ga(n,γ), Ge(n,p)	1.3 (18)
As^{76}	26.8 h	2.2, 1.8 (13)	Ge(p,n), Sc(d,α), As(d,p), As(n,γ), Br(n,α)	4.6 (18)
Y^{88}	105 d	1.9, 2.8 (5)	Sr(p,n), Sr(d,2n), Y(n,2n)	
In^{116}	54 m	1.8, 2.1 (14)**	In(n,γ), Cd(p,n), In(d,p)	150. (18)
Sb^{124}	60 d	1.70, (2.06 ?) (15) 1.67 (9)**	Sb(d,p), Sb(n,γ)	1.1 (18)
La^{140}	40 h	2.50 (9)**	La(d,p), La(n,γ), U(n,f)	7.0 (19)
MsTh in equilibrium with its decay products	6.7 y	1.80, 2.20, 2.62 (16)	Natural	
Ra in equilibrium with its decay products	1590 y	1.69, 1.75, 1.82 2.09, 2.20, 2.42 (17)	Natural	

*Only those gamma-rays with energies greater than 1.63 Mev are listed.

**References 9 and 14 were actually determinations of photo-neutron energies from which gamma-ray energies were estimated.

TABLE II

Energies of Photo-Neutrons

Source	Theoretical Estimate of Neutron Energy (Mev)	Observed Neutron Energy (Mev)
F^{20} + Be	0.50	----
Na^{24} + Be	1.00	0.83 (9)†
Na^{24} + D	0.29	0.22 (9)†
Al^{28} + Be	0.15	----
Cl^{38} + Be	0.46 (0.0187)	----
Sc^{44} + Be	0.15	----
Mn^{56} + Be	0.15, 0.45,* 0.95*	0.15, 0.30 (14)
Mn^{56} + D	0.26	0.22 (9)†
Ga^{72} + Be	0.78	----
Ga^{72} + D	0.16	0.13 (9)†
As^{76} + Be	0.28, 1.44**	----
As^{76} + D	0.50	----
Y^{88} + Be	0.24, 1.04**	0.22 (20) 0.16 (21)
Y^{88} + D	0.31	---- (22)
In^{116} + Be	0.15, 0.46	>0.15, 0.30 (14)
Sb^{124} + Be	0.060 [0.035]	0.025 (9, 21)† 0.035 (14)
La^{140} + Be	0.76	0.62 (9)†
La^{140} + D	0.16	0.13 (9) 0.15 (21)
MsTh + Be	0.15, 0.88	----
MsTh + D	0.22	0.195 (21)

Ra + Be	0.052, 0.11, 0.17, 0.41, 0.50, 0.70	----
Ra D	0.006, 0.12	0.12 (20)

*More than 80% of the neutrons from the Mn + Be source seem to be in the lowest energy group.
**The number of neutrons in the high energy group should be of the order of 1% or less since these gamma rays seem to be due to "forbidden" transitions.
†Sources are shown in Figure 1.

collision. In this way, they were able to measure the spread in the neutron energy, and were also able to determine the relative number of neutrons in different energy groups where more than one energy group was present. Hughes and Eggler (14) found that the sources described in reference 9 and shown in Figure 1 had a spread in energy of about 25% of the mean energy of the neutrons. The spread observed in these sources was attributed to the slowing down of the neutrons within the sources.

Another technique for measuring neutron energies is to use a proton-recoil proportional counter or ionization chamber (23, 24). A discussion of this method is given by Barschall and Bethe (25). An application of the proton-recoil counter for determining a neutron spectrum is described by Barschall, et al (26).

A "mean energy" can be determined easily by measuring the hydrogen scattering cross section of the neutrons (9). The scattering cross section for neutrons between 10 kev and 1000 kev varies rapidly as a function of energy (27). By making a transmission measurement on a hydrogenous material, and by determining the transmission due to the other elements present in the substance, one can readily determine the cross section of hydrogen for neutrons from a source. The "mean energy" of the neutrons can then be estimated by referring to Bohm and Richman's curve (27) of neutron scattering cross section versus energy.

Another method that has been employed to determine an average energy is that of slowing down the neutrons in a water bath and measuring the distribution of the resonance neutrons (20). The slope of the distribution beyond the maximum depends on the initial energy of the neutrons.

Yields of Photo-Neutrons

Since various workers use sources of different sizes and shapes, and since the yield of neutrons from a source depends upon the thickness of the target material, the gamma-ray intensity, and the geometry, it is best to discuss the yield of neutrons from an idealized source. Idealized sources will be considered to be composed of a 1 curie source placed at a distance of 1 cm from either one gram of beryllium metal or one gram of heavy water. These will be referred to as "standard sources."

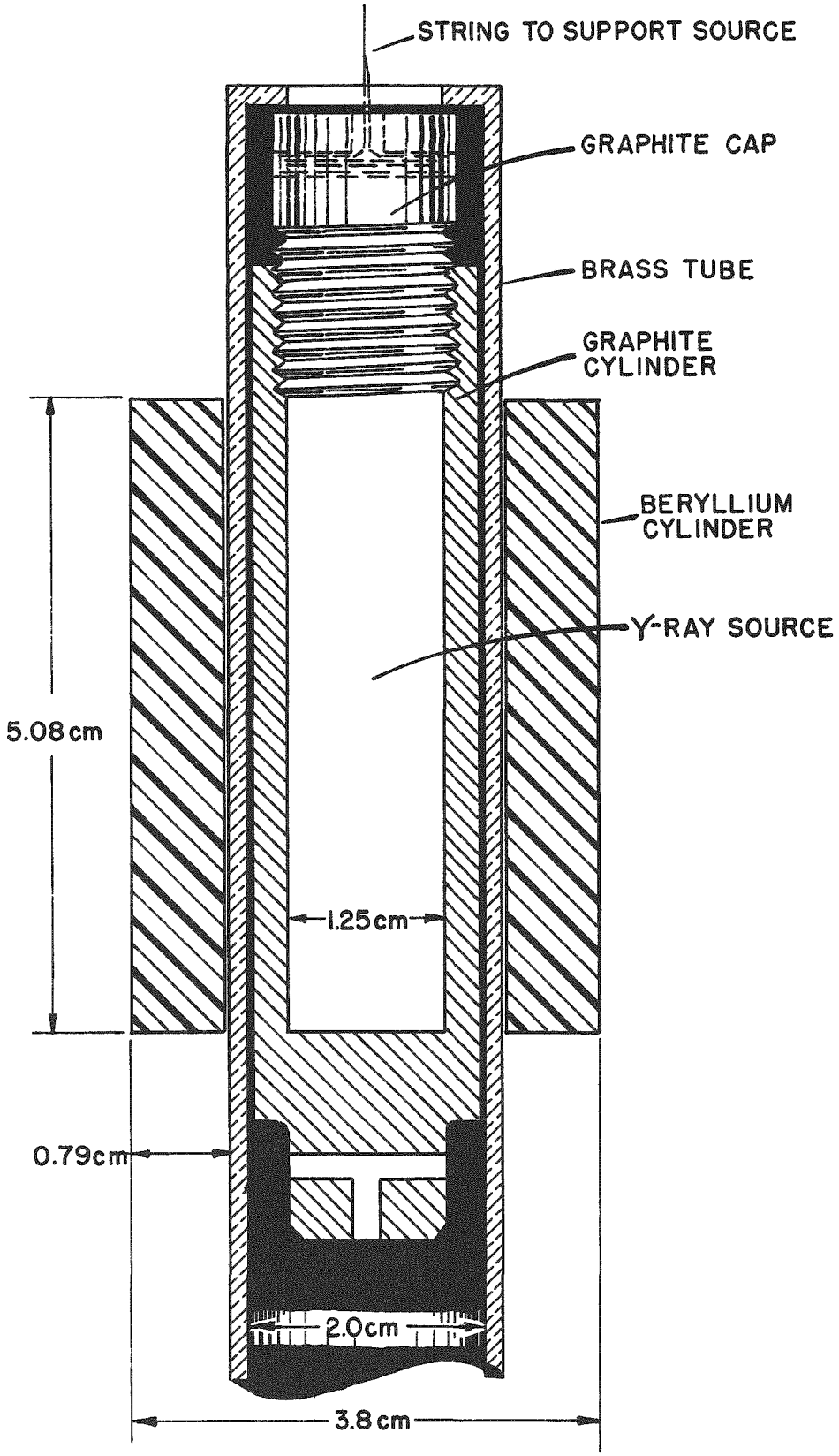

STRING TO SUPPORT SOURCE
GRAPHITE CAP
BRASS TUBE
GRAPHITE CYLINDER
BERYLLIUM CYLINDER
γ-RAY SOURCE
5.08 cm
1.25 cm
0.79 cm
2.0 cm
3.8 cm

FIGURE 1

The yields of neutrons from various "standard sources" are listed in Table III. It will be observed that the yield of photo-neutrons varies over a wide range. These large variations are due to two factors:

1. The disintegration schemes of the radioisotopes are generally not simple; consequently, less than one photon (of high energy) is emitted per disintegration.

2. The cross sections for the photo-disintegration of beryllium and deuterium are functions of the photon energy.

The disintegration schemes are known for only a few of the radioisotopes employed. The third column of Table III lists the number of photons per disintegration where there is some information available. It is to be noted that the number of photons per disintegration from some of the isotopes is only of the order of a few percent. In these cases, the disintegration scheme introduces the most important factor leading to a poor yield of photo-neutrons.

For the sources listed in Table III, the variation of the photo-disintegration cross section is not greater than a factor of ten. A detailed discussion of the theoretical energy dependence of the deuterium cross section, $\sigma_D(\gamma,n)$, has been given by Bethe and Bacher (28). According to this theory, $\sigma_D(\gamma,n)$ is a monotonically increasing function of the energy from threshold to about 4.4 Mev, where it reaches a broad maximum. It falls off more slowly at higher energy, and is still of the order of 10^{-28} cm^2 at 100 Mev. The most recent published calculation of $\sigma_D(\gamma,n)$ was performed at only one energy (29). These calculations have been extended to several other energies by Myers and Moskowski (30); their results are shown in Figure 2. According to Rarita and Schwinger, $\sigma_D(\gamma,n)$ should be 15.3×10^{-28} cm^2 at 2.62 Mev. This is larger than Bethe and Bacher's value by a factor of 1.5. The most recent experimental measurements (31,32,33,34) have been in fair agreement with Rarita and Schwinger's calculations, and are indicated in Figure 2. For the gamma rays emitted by the radioisotopes listed in Table III, it is estimated that $\sigma_D(\gamma,n)$ lies between 1×10^{-27} and 2×10^{-27} cm^2.

The photo-disintegration cross section of beryllium, $\sigma_{Be}(\gamma,n)$, has most recently been estimated on a theoretical basis by Guth and Mullin (35). The available experimental evidence [Russell, et al. (32)] indicates that $\sigma_{Be}(\gamma,n)$ passes through a maximum in the first 100 kev above the threshold energy, then passes through a minimum, and is rising again at about 2.5 Mev. Except for the 1.67 Mev gamma ray from Sb^{124}, and for the Ra + Be source (38), $\sigma_{Be}(\gamma,n)$ seems to be less than 1×10^{-27} cm^2 for the sources listed in Table III.

Some fairly reliable figures which may be of use are ratios of $\sigma_{Be}(\gamma,n)$ to $\sigma_D(\gamma,n)$ listed in Table IV.

For sources which are 1 cm thick or greater, the attenuation in the gamma-ray intensity due to the Compton effect must be taken into account. The mean free path for the Compton effect in heavy water is about 25 cm for the gamma rays available for photo-neutron sources; in beryllium metal it is about 10 cm. The mean free path will be much shorter in the gamma-ray source material, and will be dependent upon the material and its density. The Compton effect is of the order of 10^3 times more probable than the photo-disintegration process; therefore, even with an infinite thickness of target material one can expect only something of the order of one neutron for every thousand gamma rays.

TABLE III

Yields of Neutrons from Photo-Neutron Sources

Source	Neutrons per Second per Curie (1 gm of target at 1 cm)	Photons per Disintegration
Na^{24} + Be	14×10^4 (32) 12×10^4 (36)	1.0 (6, 40)
Na^{24} + D_2O	29×10^4 (32) 24×10^4 (36) 27×10^4 (33)	1.0 (6, 40)
Mn^{56} + Be	2.9×10^4 (32)	0.25(1.81 Mev), 0.15 (2.13 Mev) (41, 42)
Mn^{56} + D_2O	0.31×10^4 (32)	~0.01 (2.7 Mev) (32)
Ga^{72} + Be	5.9×10^4 (32) 3.7×10^4 (36)	0.26(2.51 Mev), 0.33(2.21Mev) 0.08, (1.87) (12)
Ga^{72} + D_2O	6.9×10^4 (32) 4.6×10^4 (36)	0.26(2.51 Mev), 0.33(2.21 Mev) 0.88, (1.87) (12)
Y^{88} + Be	$10. \times 10^4$ (33)	1.0 (1.9 Mev) (43)
Y^{88} + D_2O	0.3×10^4 (22)	~0.01 (2.8 Mev) (22)
In^{116} + Be	0.82×10^4 (32)	?
Sb^{124} + Be	19×10^4 (32)	~0.5 (44)
La^{140} + Be	0.23×10^4 (32) 0.34×10^4 (36)	~0.04 (45)
La^{140} + D_2O	0.68×10^4 (32) 0.97×10^4 (36)	0.04 (45)
MsTh + Be	3.5×10^4 (31)*	(1.0)* (46)
MsTh + D_2O	9.5×10^4 (31)*	(1.0)* (46)
Ra + Be	1.2×10^4 (37, 38)	0.0224(1.69 Mev)0.143(1.75 Mev) 0.024(1.82 Mev)0.022(2.09 Mev) 0.059(2.2 Mev) 0.025(2.42 Mev) (17)
Ra + D_2O	0.1×10^4 (39)	0.059 (2.2 Mev) 0.025 (2.42 Mev) (17)

*Although the number of photons per disintegration from Th C'' is unity, when a MsTh source is used, the neutron yield values are reduced by the branching ratio of Th C → Th C'', a factor of 0.35. This factor is included in the values given in column 2.

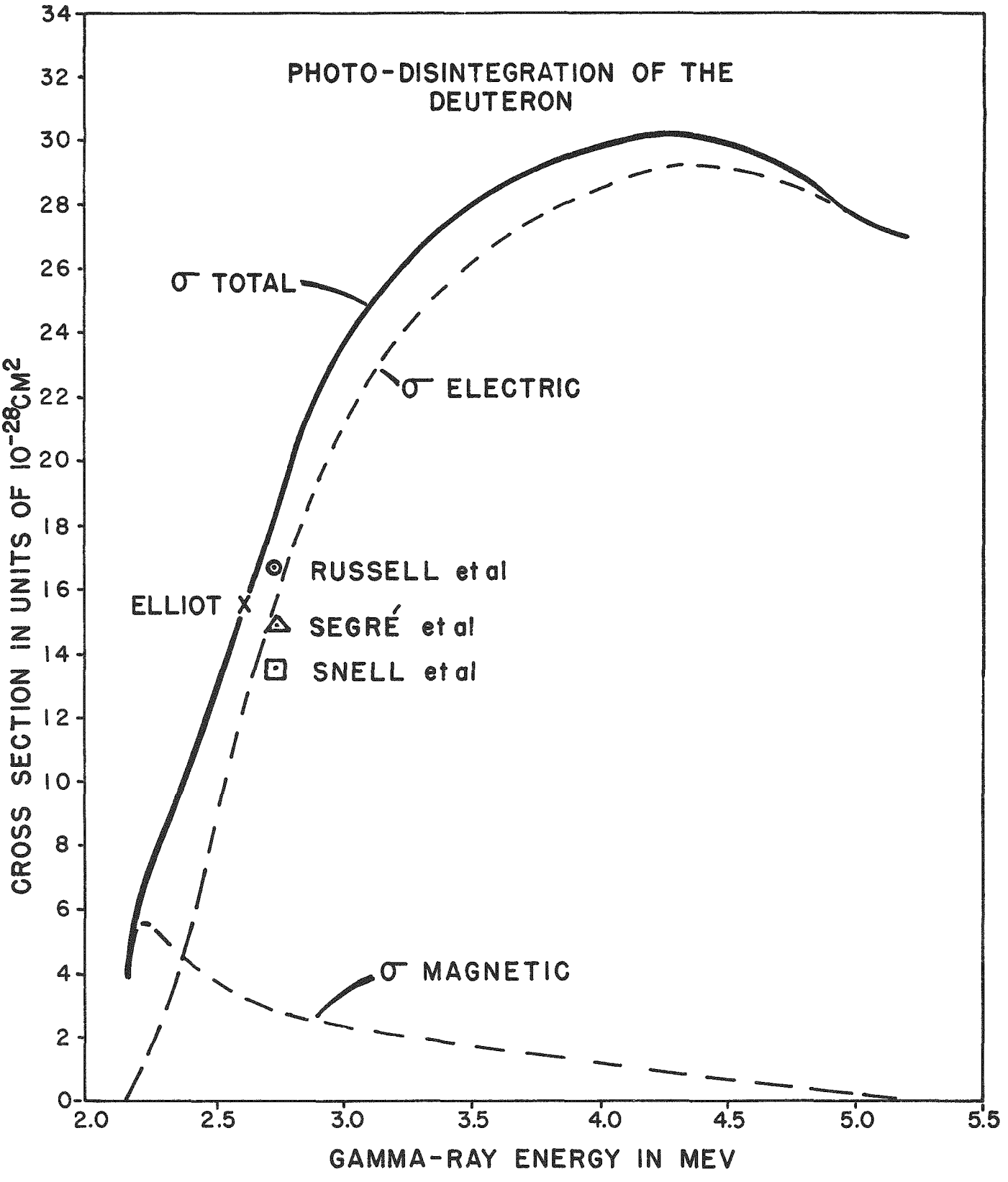


PHOTO-DISINTEGRATION OF THE DEUTERON
σ TOTAL
σ ELECTRIC
σ MAGNETIC
RUSSELL et al
SEGRÉ et al
SNELL et al
ELLIOT X
CROSS SECTION IN UNITS OF 10⁻²⁸CM²
GAMMA-RAY ENERGY IN MEV
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
32
34
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5

FIGURE 2

TABLE IV

Energy (Mev)	$\frac{\sigma_{Be}(\gamma, n)}{\sigma_{D}(\gamma, n)}$	Reference
2.50	0.30	32
2.62	0.35	47
2.76	0.43	32

Considerations in the Manufacture of Photo-Neutron Sources

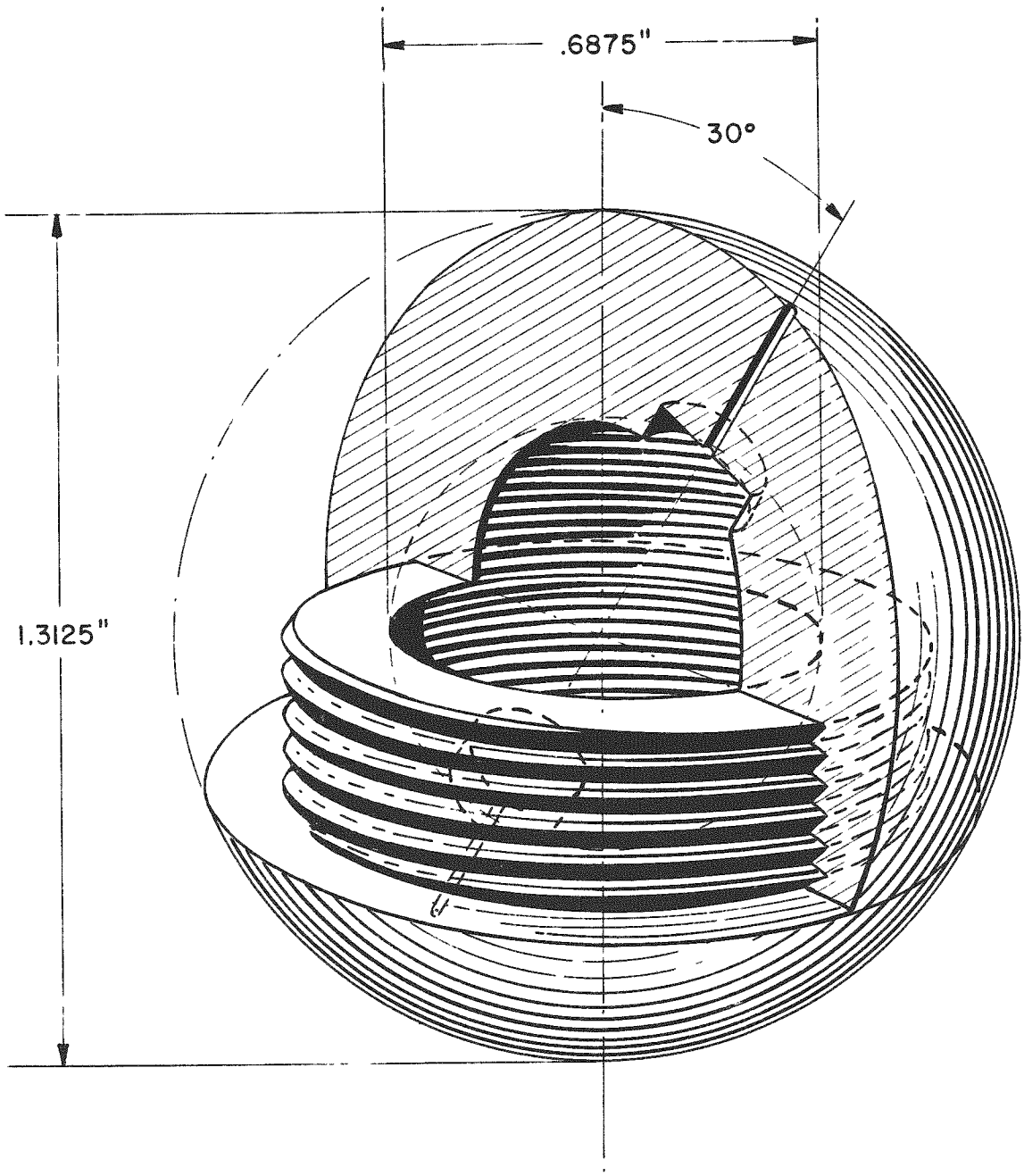
The need for a high yield of neutrons from a source will generally require the use of appreciable quantities of radioactivity and target material. Under these circumstances, the degradation in the energy of the neutrons due to slowing down within the source is likely to be much greater than the spread given by expression (3). If a monochromatic source is desired, it is generally best to use one as small as the intensity requirements of the experiment will permit.

In order to obtain a small source of neutrons, a photon emitter with as high a specific activity as possible is desirable. High specific activities can be obtained in radioactive sources by forming the desired radioisotope in another element and then performing a chemical separation. The proton, alpha, and fission reactions are useful for making sources in this way. The (n, ·) reactions in chain-reacting piles unfortunately produce radioisotopes of the elements being bombarded; unless a Szilard-Chalmers reaction can be developed, there is no way of increasing the specific activity by chemical techniques. Since chain-reacting piles are likely to be the best source of radioisotopes, the neutron cross sections for the formation of various isotopes are included in the last column of Table I. The attainable number of curies per gram depends directly upon these cross sections and the neutron flux, and inversely upon the atomic weight.

The physical size of sources is very important; the sources should have as high an atomic density as possible. Generally this is achieved by using the metallic form of an element; however, in the case of sodium, a higher atomic density is obtained in fused NaF than in metallic sodium. With a source of the order of a curie or more, there is the added advantage of ease of handling (and a reduction in the hazards of handling) if the source is in a metallic, pressed, or fused state.

An attempt is made in the following paragraphs to bring out the main advantages and disadvantages of differently shaped sources.

1. Spherically shaped sources. The concentric sphere-type of source, in which a uniform thickness of target material surrounds a sphere of the radioisotope, makes the most efficient use of the available radioactivity. An example of such an arrangement is shown in Figure 3. The center sphere is (or contains) the gamma-ray emitter; the outer spherical shell is beryllium metal. Two holes are provided through the beryllium shell to permit the attachment of a wire for handling the source. Probably



CONCENTRIC SPHERE TYPE
OF
BERYLLIUM PHOTO-NEUTRON SOURCE

FIGURE 3

just as efficient is the source consisting of a homogeneous mixture of the beryllium or heavy water with the radioisotope. With heavy water, this is readily achieved by using a soluble compound of the radioisotope dissolved in the heavy water.

The greatest advantage to spherical sources is that one can be fairly certain that the neutrons are emitted with a spherically symmetrical distribution. In a great many experiments this is extremely valuable. Another advantage is that for the quantity of target material and source material, the highest possible yield is achieved in the smallest volume.

The only disadvantage to spherical sources arises in using large quantities of radioactivity (a curie or more) where gamma-ray source and target material cannot be assembled in advance of the irradiation of the gamma-ray source. In this case, a physiologically hazardous source may have to be manipulated. Associated with this difficulty is the fact that the target material generally cannot be separated from the gamma-ray source in order to determine the effects of the gamma rays alone on the experimental equipment.

In using a D_2O solution, the hazards of spillage are extremely serious, especially around large laboratories where it is desired to keep background to a minimum.

2. Cylindrically shaped sources. The concentric cylinder type of source, in which a tube of target material surrounds a cylinder of radioisotope, is not as efficient as the spherical source. The loss of efficiency results largely from those gamma rays which escape parallel to the cylinder axis without hitting any target material.

The main advantage of cylindrical sources is the ease with which they can be assembled and disassembled; these operations may thus be carried on at a safe distance from the source (9). An example of the concentric cylinder type of source is shown in Figure 1. The number of neutrons emitted by such sources is listed in Table I of reference 32. It is several times 10^6 for many of them.

A serious disadvantage of cylindrical sources is that the neutron distribution is not spherically symmetric. To a first approximation, if the source is large, the number of neutrons will be proportional to the surface area of the source seen at the detector. If this asymmetry is important in the experimental work, it must be measured.

For some types of work it is useful to employ a large cylinder of radioactive material together with a smaller cylinder of target material which has been formed with a hole along the axis. This type of source is useful where one can place the detector in the hole along the axis of the source.

3. Remote sources and the angular distribution of the emitted photo-neutrons. In some experiments, it is necessary or desirable to separate the gamma-ray emitter from the target material. With such arrangements, the yield of neutrons will be very low and will vary directly with the solid angle which the target material subtends at the gamma-ray source. In using such sources, one must also take into consideration any asymmetries in the angular distribution of the neutron production. This question was studied for deuterium by Chadwick and Goldhaber (48), Graham and Halban (49), and Lassen (50). The photo-magnetic part should be spherically symmetric, and the photoelectric part should obey a $\sin^2 \theta$ law. (See Figure 2).

Studies of the angular distribution in beryllium were made by Goloborodko and Rosenkewitch (51) using a radium source, and by Chadwick and Goldhaber (48) using a radon source. Their results indicated a spherically symmetrical distribution. Studies at other energies have still to be performed.

Remote sources can be made as monochromatic as desired, but at the expense of the neutron yield. It is impossible to discuss the details of all of the possible geometrical combinations of source and target materials. A description of one type of remote source is given by Gamertsfelder and Goldhaber (52). Whatever arrangement is employed, the solid angle factor is generally the most important one. The thickness of the target material has already been discussed. The thickness of the gamma-ray emitter is likely to be of importance in connection with the number of curies of activity obtainable in an artificial radioisotope. For radioisotopes produced by charged particle bombardment, only the surface layers will be penetrated by the bombarding particles. For radioisotopes produced by (n, γ) reactions, one must estimate the self-shielding of the sample if the sample is thicker than a few hundredths of the neutron mean free path.

For gamma-ray sources of the order of 10 gms/cm^2, corrections must be made for the loss in gamma-ray intensity due to the Compton effect within the emitter.

Although the photo-disintegration of beryllium and deuterium was discovered more than fifteen years ago (48), most values for neutron energies, yields, cross sections, and angular distributions are still rather poor. Much more quantitative work still remains to be done in this field.

* * *

Acknowledgement

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