Measurement of the $O^{16}(n,p)N^{16}$ Cross Section at 14.7 Mev

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The $O^{16}(n,p)N^{16}$ cross section has been measured for 14.7-Mev neutrons. A recoil proton telescope was used to measure the neutron flux. Oxygen was incorporated in a liquid scintillator which was irradiated in the flux. The absolute activity of the resulting N^{16} was measured by 4π beta counting the solution with two photomultipliers in coincidence. A value of 39.2 ± 1.6 mb was obtained for the cross section.

INTRODUCTION

ACTIVATION cross sections for 14-Mev neutrons are usually measured by counting the beta-particle activity of the resulting daughter nuclei. Measurements of the absolute neutron flux accurate to three percent can be obtained by use of recoil proton telescopes[1] or associated particle counters.[2] All the published cross-section values have standard errors of at least five percent. The principal contribution to the final error arises from the determination of the beta counting efficiency. Probably the most reliable efficiencies are achieved with 4π proportional counters; however, quantities of one milligram or less must be used so that the beta absorption in the sample will be small and the efficiency very nearly one. In the present experiment 4π scintillation counting is employed, and the beta-counting efficiency is accurately determined for gram quantities.

Previous measurements[3,4] of the absolute value of the $O^{16}(n,p)$ cross section for a monoenergetic flux have published errors of 30–50%. As this reaction causes the principal radioactivity in the coolant of water-moderated reactors, an effort has been made to improve the precision.

The neutron generator available was a small accelerator with ion source grounded and target at -100 kev, capable of operating as a pulsed source only. This source had a yield of about 10^8 neutrons per second at its normal duty cycle.

DETERMINATION OF DETECTION EFFICIENCY FOR N^{16} ACTIVITY

N^{16} has the following beta-ray decay branches[5]:

Maximum beta energy of branch (Mev)	Branch percent
10.40	26 ± 2
4.26	69 ± 2
3.29	4.9 ± 0.4
1.53	1.0 ± 0.2

As the beta-ray energies are quite high, N^{16} is a favorable case for 4π counting. In our experiments, the oxygen was incorporated in a liquid scintillator having this composition:

Dioxane	383g	(375 ml)
Naphthalene	37.5g	
PPO	3.0g	
POPOP	0.010g	

This mixture is 32.9% oxygen by weight.

The only suitable holder that has no objectionable radioactivity from an irradiation in a 14-Mev flux is a hydrocarbon plastic. None of the plastics tried proved satisfactory because of solubility or transparency problems. Accordingly, we decided to irradiate the liquid in a stainless steel container. The liquid is then poured into a vycor cell between two matched EMI 6097S photomultipliers and is counted in coincidence. A calibration cell was filled with the solution plus Tl^{204} and served as a standard in adjusting amplifier and photomultiplier voltages. Tl^{204} is free of gamma rays and has a maximum beta energy of 0.764 Mev. The endpoint of its beta spectrum was used to calibrate a 100-channel pulse-height analyzer and the discriminators of the coincidence analyzer (Fig. 1). Most of the phototube pulses caused saturation pulses in the nonoverloading linear amplifiers at the gain settings used in the activation measurements. Calibrated attenuators were inserted between the preamplifiers and amplifiers so that the end point of the thallium spectrum was not saturated. For most measurements, each beta-channel dis-

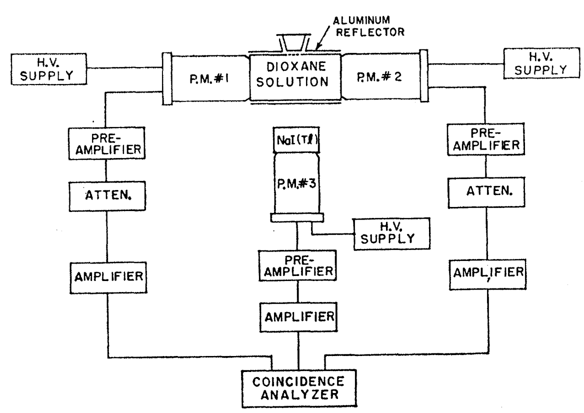

FIG. 1. 4π liquid scintillation instrumentation.

[1] S. J. Bame, E. Haddad, J. E. Perry, Jr., and R. K. Smith, Rev. Sci. Instr. **28**, 997 (1957).
[2] H. H. Barschall, L. Rosen, and R. F. Taschek, Revs. Modern Phys. **24**, 1 (1953).
[3] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).
[4] H. C. Martin, Phys. Rev. **93**, 493 (1954).
[5] D. E. Alburger, A. Gallmann, and D. H. Wilkinson, Phys. Rev. **116**, 939 (1959).

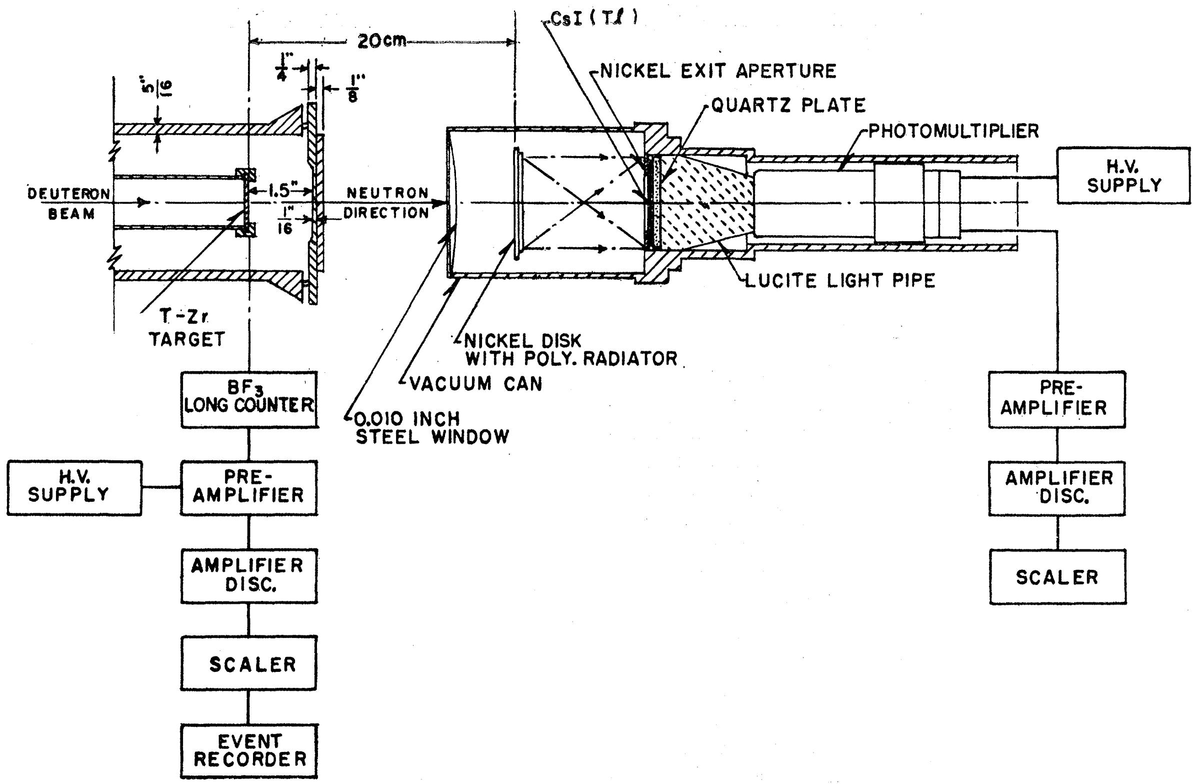

FIG. 2. Neutron flux measurement and monitoring instrumentation.

criminator was set to count betas of energy greater than 20 kev, based on the thallium calibration. Because of possible poisoning of the liquid by the thallium and the nonlinearity of liquid scintillators in the low-energy range, this cutoff may not be precise.

The beta channels were put in coincidence to eliminate individual photomultiplier noise and normally 0.25-microsecond resolution was employed. The phototubes were placed in a light-tight box which had a compartment beneath the assemblage for dry ice. Vycor cells, 3.9 cm in interior diameter and either 2 or 4 cm in width, were placed in optical contact with the phototube faces. Several irradiations of the solution were made in the highest flux obtainable with the source, and pulse-height spectra were obtained for both cells. Assuming the number of pulses per channel below 20 kev is the same as the number per channel between 20 and 30 kev, 0.5% of the pulses fall below 20 kev for both cells, or the beta efficiency is 99.5%.

Gamma rays follow the beta decays in all but the 10.4-Mev beta branch. As each gamma is practically simultaneous with the preceding beta, no double counting occurs and efficiency of counting disintegrations is increased slightly.

A sodium iodide detector 2 in. in diameter and 1.0 in. thick mounted on a Dumont 6292 photomultiplier was placed adjacent to the cell to count gammas from the N^{16} decay. The principal gammas have an energy of 6.14 Mev and are associated with the 4.26-Mev beta branch. Assuming only one disintegration branch having a single efficiency, ϵ_B, in the cell and a single efficiency in the NaI detector, the beta efficiency is given by

$$\epsilon_B = C_{B\gamma}/C_\gamma, \tag{1}$$

where $C_{B\gamma}$ is the beta-gamma coincidence rate and C_γ is the gamma detector rate. Actually the beta efficiency decreases for disintegrations near the boundaries since 20-kev energy loss is required, and the gamma detector weights portions of the cell close to boundaries more heavily than the interior; as a result ϵ_B is slightly underestimated. The cells were filled with irradiated solution and counted for repetitive periods of 15 seconds on and 5 seconds off. Since the N^{16} half-life is 7.352 sec,[6] the first interval contains most of the disintegrations and the remaining intervals can be used for background subtraction. The NaI crystal was not completely shielded from source neutrons so only the second interval was used for background to minimize the activity which becomes apparent after long bombardments. The two beta channels were put in coincidence with the gamma channel, and a resolution of one microsecond was used for the coincidence count. For the small cell, an efficiency of 99.1% was obtained and for the large cell, 99.3% from the accumulation of several measurements. The

[6] J. O. Elliot and F. C. Young, Nuclear Sci. and Eng. 5, 55 (1959).

coincidence background is small and the main error arises from the NaI background. Other processes, such as bremsstrahlung from energetic beta rays, may contribute to the NaI counting rate. These change the gamma efficiency and the relative contribution of the different beta branches to the beta efficiency. Since the systematic errors give a result lower than the true cell efficiency, the results may be regarded as lower bounds to the efficiency. Accordingly, an efficiency of 99.5% was adopted for both cells at the 20-kev level and the error should be less than 0.5%.

MEASUREMENT OF NEUTRON FLUX

The neutron flux was determined at the position of cell irradiation by use of a simplified proton recoil telescope of the Los Alamos design.[7] The tables given by Bame *et al.*[1] were used to obtain the telescope efficiency following a method by Johnson.[8] The tabulation makes use of the known *n-p* scattering cross section, the radiator composition, and the solid angle of the totally absorbing detector. Figure 2 is a sketch of the experimental arrangement used to calibrate the BF_3 long counter which was utilized as a flux monitor during the cell irradiation. A second BF_3 long counter, not shown, located at a greater distance from the target was used as an auxiliary monitor.

The source of neutrons was a pulsed neutron generator in which deuterons from an ion source at ground potential impinged on a 0.010-in.×1.0-in. diameter T-Zr target at −100 kv. The target had a thickness of less than 120 μg/cm^2 zirconium which corresponds to a deuteron energy loss of 19 kev. The mean energy of the resulting neutron beam is 14.7 Mev and the spread ±0.1 Mev. There is a vacuum insulating gap of 1.5 in. between the stainless steel end plate and the target. A 0.071-in. Pb sheet was attached to the end plate to prevent low-energy x rays originating in the generator structure from reaching the telescope.

The recoil telescope was positioned on the target axis at a distance forward of target such that the polyethylene radiator was located at the midpoint of the intended cell irradiation. The long counter monitor shown in Fig. 2 was positioned approximately 119 cm from the target center at right angles to the target axis. The auxiliary long counter was positioned approximately 232 cm from the target at 115 degrees to the beam direction.

Calibration of the monitor long counter was accomplished with the neutron generator producing 5-microsecond neutron bursts at a repetition rate of 800 per second. The narrow burst width and high repetition rate were utilized with this generator in order to minimize pulse pile-up in the telescope preamplifier. The proton recoil spectrum obtained with the aid of a 100-channel pulse-height analyzer is shown in Fig. 3. Since the telescope is constructed so that either the polyethylene radiator or its nickel backing can be positioned to face the CsI scintillation detector, the background radiation may be determined and subtracted from the obtained spectrum. A variation in the telescope pulse-height discriminator level between 30 and 50% of the maximum pulse height did not affect the ratio of telescope to long counter counts after background subtraction.

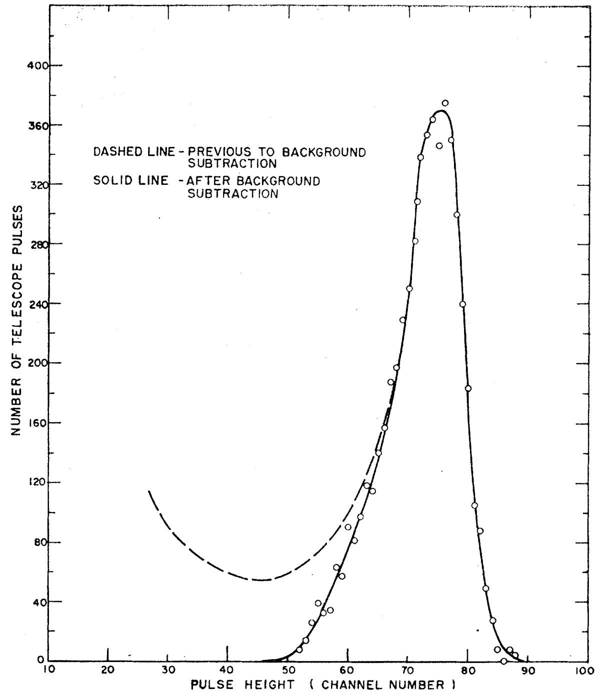

FIG. 3. Simplified telescope flux measurement.

PROCEDURE

The stainless steel cell containing the dioxane solution to be irradiated was positioned on the target axis so that its midpoint coincided with the midpoint of the telescope polyethylene radiator used in the flux calibration. Each irradiation lasted 30 seconds during which the event recorder was used to obtain the differential time distribution of the long counter monitor counts. This distribution is necessary in order to account properly for the N^{16} activation and decay during the irradiation because of the variation in the neutron yield from the generator used in this experiment. A timed oscillograph was used to record every 400th count from the long counter monitor.

Following irradiation, the stainless steel cell was quickly transported to the 4π beta counting instrumentation as shown in Fig. 1. Here the dioxane solution was poured into an empty Vycor cell already in place between the photomultipliers which were housed in the

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[7] S. J. Bame, E. Haddad, J. E. Perry, Jr., and R. K. Smith, Rev. Sci. Instr. **29**, 652 (1958).

[8] C. H. Johnson, *Fast Neutron Physics, Part I: Techniques* (Interscience Publishers, Inc., New York, 1960), Chap. II.C, p. 247.

light-tight box. After the box lid was secured, the photomultiplier high-voltage was turned on and beta counting was initiated. A switching system provided an accurate measure of both the irradiation time and the interval between the end of irradiation and onset of counting. The coincidence pulse output of the beta counting system was counted by a multichannel time analyzer with four second channel widths. Background was measured after the N^{16} activity was negligible. Usually two half-lives elapsed before counting commenced.

ATTENUATION OF NEUTRONS IN CELL MATERIALS AND GEOMETRIC CORRECTIONS

The stainless steel holders used in the solution irradiations had a 20-cm curvature in the long (9.36-cm) dimension but the cross section was rectangular, 4.72×0.64 cm for the small cell and 4.72×1.10 cm for the large cell. As the 14.7-Mev neutron flux traverses the solution, it is attenuated and somewhat degraded in energy. The N^{16} activity as a function of depth for a spherical cell is given by

$$A=(K/r^2)e^{-\alpha(r-r_0)}, \quad (2)$$

where r_0 is the inner radius of the solution. Actually the attentuation factor should be measured with an oxygen detector; however, the $Cu^{63}(n,2n)Cu^{62}$ cross section has a threshold and energy response similar to oxygen, and "poor" geometry cross sections measured with it for 14-Mev neutrons[9] can be used. These cross sections agree well with measurements using proton recoil detectors biased at 12.6 Mev.

The following atom densities and cross sections were used for the scintillator:

Element	Atom density (cm^{-3})	Cross section (barns)
Hydrogen	5.48×10^{22}	0.54
Carbon	3.02×10^{22}	0.76
Oxygen	1.28×10^{22}	0.87
$\alpha=\Sigma N_i\sigma_i=0.637\ cm^{-1}$		

Integration of Eq (2) over the actual volume of the holders and division by the unshielded activity at 20 cm gives a reduction factor of 0.982 for the small holder and 0.962 for the large holder.

The long-counter calibrations were made with 0.180-cm lead in front of the telescope, reducing the flux 1.5% and the 0.16-cm thick stainless steel wall reduced the flux in the holders 1.9%.

RESULTS AND DISCUSSION

Four sets of measurements of the induced N^{16} activities at 20 cm from the target were made for each cell. The large cell held about 48 g of solution and the small cell 24 g. Normally around 40 000 counts were recorded on the long-counter tape during the thirty-second irradiation. As the flux was not constant, the contribution to the N^{16} activity from one-second intervals was calculated. The decays were consistent with the published half-life of 7.352±0.009 seconds,[6] which was used in reducing the data. The small cell yielded a value of 39.05 mb and the large cell 39.35 mb for the $O^{16}(n,p)$ cross section. For each cell the data has a relative accuracy of about one percent from the N^{16} counting statistics.

The final standard error is based on the following individual errors:

(1) Absolute accuracy of recoil proton telescope	3.0%
(2) Reproducibility of telescope to long-counter ratios with different targets	2.5%
(3) Counting statistics	1.0%
(4) Efficiency of beta counting	0.5%
(5) Errors in attenuation factors	0.5%

These yield a final standard error of 4.1%. As a result, the data from both cells yield a value of 39.2±1.6 mb for the $O^{16}(n,p)$ cross section at 14.7 Mev.

Previous values of the cross section are 49±25 mb by Paul and Clarke[3] at 14.5 Mev, and 89±30 mb at 14.0 Mev by Martin.[4] The latter result is from a relative measurement of the beta yield from an irradiated CuO sample, and no correction was made for the difference in self-absorption of the positrons (β max=2.9 Mev) from the resulting Cu^{62} activity as compared with the higher energy betas from N^{16} in the thick sample.

As a check on the method, the $Cu^{63}(n,2n)$ cross section was measured. Because of the attrition of the thin targets, a thick T-Zr target was used. Difficulty was experienced in finding a good scintillator loaded with copper. A mixture of 201 g dioxane, 14 g napthalene, 1 g PPO, and 0.967 g $Cu(NO_3)_2\cdot 3H_2O$ in 5 g H_2O gives about half the pulse height of the original solution for the 624-kev beta line from Cs. Cu^{62} is a positron emitter and the 0.51-Mev annihilation radiation may be employed in the beta-gamma coincidence technique. Two measurements of the beta efficiency gave values of $\epsilon_B=0.99$ and >1.0 for beta discriminator settings of approximately 30 kev. An efficiency of 0.99 was adopted which should have an error of less than two percent.

At this concentration, it was necessary to make irradiations at 10 cm with the large holder. The data were reduced using a half-life of 9.80±0.02 min. This value was determined from an irradiation of a copper foil which was counted in a 2π flow proportional counter. The solution irradiations gave consistent results. After suitable geometric corrections were made, two measurements gave an average value of 518 mb for the total activation cross section. The $N^{14}(n,2n)$ cross section has a value of 5±1 mb at 14 Mev and the resulting positron emitter has a half-life of 10 min and is part of the total activation cross section. As a result, $\sigma_{Cu}+2\sigma_N=518$ mb and $\sigma_{Cu}=508$ mb. If a correction factor of 1.043 is used for K capture,[10] the value becomes 530 mb. This value

[9] D. D. Phillips, R. W. Davis, and E. R. Graves, Phys. Rev. **88**, 600 (1952).

[10] S. Yasumi, J. Phys. Soc. Japan **12**, 443 (1957).

has a standard error within five percent but refers to an effective energy of about 14.6 Mev since a thick target was used. The solution used was chemically analyzed to determine the copper concentration. Based on weights of the ingredients, the copper concentration was 4.1% lower. Because of the observed tendency of the copper salt to lose weight upon weighing, the concentration from the chemical analysis was used. Other measured values of the cross section are 510 mb (±7%) by Forbes at 14.1 Mev,[11] 482 mb (±15%) by Paul and Clarke at 14.5 Mev,[3] and 556 mb (±5%) by Yasumi at 14.1 Mev.[10] Only the latter value includes the K-capture correction factor of 1.043.

With a steady 14-Mev source and grounded target, the neutron flux can be easily measured to a 2% if both a recoil proton telescope and associated particle counter are employed. The remaining errors are small and can be reduced to less than 1.0% (total) with a sufficiently strong source. As a result, activation cross sections accurate to 2–3% can be attained with the present technique.

[11] S. G. Forbes, Phys. Rev. **88**, 1309 (1952).

Note added in proof. J. M. Ferguson and W. E. Thompson, Phys. Rev. **118**, 228 (1960), have obtained a value of 507±45 mb for the $Cu(n,2n)$ cross section at 14.74 Mev. A 2.0% correction was made for K capture.

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Comparison of Po^{210} Alpha-Particle Energy with the $Li^7(p,n)Be^7$ Reaction Threshold Energy*

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Recent absolute measurements of the Po^{210} alpha-particle energy disagree with the older value used as the standard for many nuclear reaction energy measurements. A new comparison with the $Li^7(p,n)Be^7$ reaction threshold energy was made using the Notre Dame electrostatic accelerator and broad-range spectrograph. Four separate methods of comparison were used. In the first three the threshold was run and then protons or deuterons were scattered from appropriate targets so that the scattered group was recorded on the spectrograph plate near the alpha group from a source placed at the target position. First, the spectrograph and, second, the beam analyzer were used to compare particle momenta. Third, with both fields held constant after the threshold was run with the molecular beam, deuterons were scattered, giving particles of the same $B\rho$ as the alphas. In the fourth method several reaction energies that are precisely known in terms of the $Li^7(p,n)Be^7$ reaction threshold energy were measured in terms of the Po^{210} alpha-particle energy. These were the $Mg^{24}(d,d')Mg^{24*}$ reaction to the first excited state of Mg^{24} and the $N^{14}(d,p)N^{15}$ reaction leading to three excited states of N^{15}. The four measurements agree and give 5.3086±0.003 Mev for Po^{210} alpha-particle energy based on 1.8811 Mev for the $Li^7(p,n)Be^7$ reaction threshold energy.

I. INTRODUCTION

THE discrepancies between nuclear mass values obtained by mass spectroscopy and those derived from nuclear reaction energies are usually discussed in terms of a single energy standard for nuclear reaction energies. There are, however, two widely used standards: the $Li^7(p,n)Be^7$ reaction threshold energy and the energy of alpha particles emitted by Po^{210}. As will be discussed below, an earlier comparison of these two energies[1] was consistent with the early absolute determination of each. Recent absolute determinations of the Po^{210} alpha-particle energy,[2,3] however, indicate that the previously accepted value of this energy,[4] 5.2988 Mev, is low. Because of the importance of the Po^{210} alpha-particle energy to much of the pre-

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[1] W. J. Sturm and V. Johnson, Phys. Rev. **83**, 542 (1951).

[2] E. R. Collins, C. D. McKenzie, and C. A. Ramm, Proc. Roy. Soc. (London) **A216**, 216 (1953).

[3] F. A. White, F. M. Rourke, J. C. Sheffield, R. P. Schuman, J. R. Huizenga, Phys. Rev. **109**, 437 (1958).

[4] See for example E. N. Strait, D. M. Van Patter, W. W. Buechner, and A. Sperduto, Phys. Rev. **81**, 747 (1951); S. F. Zimmerman, thesis, Massachusetts Institute of Technology, 1955 (unpublished); S. Hinds and R. Middleton, Proc. Phys. Soc. (London) **74**, 196 (1959).