$\mathbf{P}^{31}(n, p)$ Si³¹ and Al²⁷ (n, α) Na²⁴ Cross Sections*

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The excitation functions and absolute cross sections for the $P^{31}(n,p)Si^{31}$ and $Al^{27}(n,\alpha)Na^{24}$ reactions have been measured by the activation method for neutron energies below 9.5 Mev and at 14.1 Mev. The excitation function for $P^{31}(n,p)S^{31}$ rises from 1.6 Mev to 5.5 Mev, is fairly constant at 0.14 barn to 9.5 Mev and considerably lower at 14 Mev. The Al²⁷ (n,α) Na²⁴ excitation curve rises from 6 to 9 Mev neutron energy and is greater at 14 Mev. Comparisons are made with earlier measurements of the $P^{31}(n,p)Si^{31}$ cross section and some discrepancies appear.

I. INTRODUCTION

CONTINUING investigation of differences in A fission neutron spectra from the principal thermally-fissile isotopes¹ makes use of a number of threshold reactions including $P^{31}(n,p)Si^{31}$ and $Al^{27}(n,\alpha)Na^{24}$. For interpretation of results, it was desirable to supplement the meager information concerning the excitation functions for these reactions. The $P^{31}(n, p)$ cross section has been measured between 1.9 and 3.6 Mev² and at 14 Mev,3 while only determinations at 14 Mev have been published on the Al²⁷(n,α) reaction.^{3,4}

Measurements at this laboratory of the $S^{32}(n,p)P^{32}$ cross section⁵ for neutron energies up to 9.5 Mev afforded an opportunity for the simultaneous determination of the $P^{31}(n,p)$ cross section. A single irradiation of high total neutron flux provided the basis for the $Al^{27}(n,\alpha)$ measurement. Subsequent calibration of counters makes it possible to report the $P^{31}(n, p)$ and $Al^{27}(n,\alpha)$ excitation functions on an absolute cross section scale. Unreported results of an earlier rough measurement of the $Al^{27}(n,\alpha)$ excitation function are included for completeness.

II. EXPERIMENTAL METHOD

Phosphorus Irradiations

Samples of phosphorus along with sulfur were fastened to the outside of an ionization chamber containing a U²³⁸ fission foil and irradiated in the forward direction with neutrons of various energies produced by the $D(d,n)He^3$ and $T(p,n)He^3$ reactions at the large Los Alamos Van de Graaff generator. The samples were about 5 cm from a 3-cm long gas target. Neutron fluxes were measured by observing the fission disintegration rate in the U²³⁸ foil and using the known fission cross section for U²³⁸. Details of source and sample arrangement as well as neutron energy and flux determinations are given by Allen *et al.*⁵ in their account of the $S^{32}(n, p)$

work. Two additional corrections were applied to the neutron flux values given by Allen: (1) a constant factor of 1.051 to correct for the difference in sample distance; and (2) a variable correction of +0.2 to +14% to correct for the anisotropy of the incident neutrons which results in a higher average intensity over the phosphorus sample than over the larger diameter U²³⁸ monitor foil. The phosphorus samples, 0.130 g disks and 1.07 cm in diameter, were punched from a sheet of polyethylene-phosphorus mixture (29.5 wt. % (CH₂)_n, 70.5 wt. % P, 0.094 cm thick) prepared by the plastics group of this laboratory.

Aluminum Irradiations

For the Al (n,α) measurement, a semicircular "2S" aluminum strip of radius 6.65 cm was centered about the gas target so that the midpoint of the strip was in the forward direction. The same fission chamber served as the neutron monitor and the absolute neutron flux in the forward direction was obtained as described by Allen et al.⁵ Earlier measurements⁶ of the angular distribution of neutrons from the D(d,n)He³ reaction (deuteron energy = 5.80 Mev) were used to get the relative neutron intensities. Six adjacent samples, 1.06 cm diameter and 0.165 cm thick, at angles up to 50° with the deuteron beam were punched from this strip after the irradiation.

The earlier cursory investigation of the $Al(n,\alpha)$ excitation function made use of individual hollow cvlinders about an inch in diameter placed at various angles to the deuteron beam in a flux of neutrons from the D(d,n)He³ reaction (deuteron energy=4.15 Mev). The angular distribution of neutrons was determined with a long counter, and samples were subsequently counted on a thin-walled Geiger-Müller counter.

Beta Counting

For the present work, counting was done with two insertion-type, methane-flow proportional counters of the same design. The beta activity of identical 0.020 cm thick foils of \tilde{U}^{238} was used as a standard for the counting systems throughout the experiment. Sensitivities of the two systems for standard foil and sample activities were

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¹ J. A. Grundl and J. J. Neuer, Bull. Am. Phys. Soc. Ser. II, 1, 95 (1956).

⁶ (1950).
² R. Ricamo, Nuovo cimento 8, 383 (1951).
⁸ S. G. Forbes, Phys. Rev. 88, 1309 (1952).
⁴ E. B. Paul and R. L. Clark, Can. J. Phys. 31, 267 (1955).
⁵ Allen, Biggers, Prestwood, and Smith, Phys. Rev. 107, 1363 (1957).

⁶ J. E. Perry and R. K. Smith (private communication).

Source reaction	$E_{n^{\mathbf{a}}}$ (Mev)	C ^b Initial counting rate (counts/min)	$\sigma_f(\mathrm{U^{238}})$ barns	$\left(\frac{10^{8} \text{ neutrons}}{\text{cm}^{2} \text{ min}}\right)$	$\sigma_{n,p}$ (millibarns)	د ا Absolute uncertainty (millibarns)	€2 Relative uncertainty (millibarns)
$\mathrm{T}(p,n)\mathrm{He^3}$	$\begin{array}{c} 1.59 \pm 0.08 \\ 1.98 \pm 0.07 \\ 2.08 \pm 0.07 \\ 2.19 \pm 0.07 \\ 2.57 \pm 0.06 \\ 2.92 \pm 0.06 \\ 3.56 \pm 0.05 \\ 4.05 \pm 0.05 \\ 4.54 \pm 0.05 \\ 4.82 \pm 0.06 \\ 5.33 \pm 0.05 \\ 5.69 \pm 0.05 \end{array}$	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{c} 0.40\\ 0.55\\ 0.56\\ 0.56\\ 0.56\\ 0.56\\ 0.56\\ 0.56\\ 0.56\\ 0.565\\ 0.565\\ 0.57\\ 0.57\\ 0.59\end{array}$	5.67 10.4 11.9 13.2 14.7 15.4 12.6 11.8 11.1 7.87 7.81 4.65	$\begin{array}{c} 1.31\\ 11.00\\ 6.50\\ 10.80\\ 44.7\\ 80.6\\ 96.2\\ 95.9\\ 106.3\\ 134.3\\ 134.6\\ 136.9\\ \end{array}$	$\begin{array}{c} \pm \ 0.14 \\ 1.0 \\ 0.65 \\ 1.0 \\ 4.5 \\ 7.5 \\ 9 \\ 9 \\ 10 \\ 13 \\ 13 \\ 13 \end{array}$	$\begin{array}{c} \pm \ 0.13 \\ 0.45 \\ 0.30 \\ 0.45 \\ 1.8 \\ 3.0 \\ 4.0 \\ 4.0 \\ 4.0 \\ 5.0 \\ 5.0 \\ 5.5 \end{array}$
D(d,n)He ³	$\begin{array}{c} 6.11 {\pm} 0.12 \\ 6.61 {\pm} 0.11 \\ 7.11 {\pm} 0.11 \\ 7.61 {\pm} 0.10 \\ 8.09 {\pm} 0.09 \\ 8.58 {\pm} 0.09 \\ 9.07 {\pm} 0.09 \\ 9.57 {\pm} 0.10 \end{array}$	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{c} 0.665\\ 0.865\\ 0.975\\ 1.01\\ 1.02\\ 1.02\\ 1.01\\ 1.01\\ 1.01\\ \end{array}$	5.17 7.03 8.32 9.40 10.0 8.05 13.5 11.4	145.9 148.1 136.4 136.7 144.2 138.9 143.4 142.1	14 14 13 13 14 13 14 14	8.5 9.0 8.0 9.0 8.5 9 12
$T(d,n)He^4$	14.1 ± 0.04	2672 ± 24	•••	6.94	85.5	7	5.0

TABLE I. Summary of $P^{31}(n, p)$ results.

^a Energy spread includes effects of energy degradation in target plus angular spread of sample. ^b Uncertainty shown is the root-mean-square deviation of the set of initial counting rates.

determined so that counting results could be normalized. Differences in the over-all sensitivity of the two systems did not exceed 3%.

The 2.65-hr silicon activity in the phosphorus sample was followed for three half-lives in general, and weighted averages taken after conversion to initial counting rates. The 15.0-hr Na²⁴ activity in the aluminum sample was followed for about $1\frac{1}{2}$ half-lives and the data treated similarly. The possibility of perturbing long-lived activities was investigated by counting samples that were irradiated in a slightly degraded fission spectrum. In phosphorus, a long-lived activity amounting to less than 0.1% was attributed to the 14.3 day activity resulting from the P³¹(n,γ)P³² reaction. No long-lived activity was noted in aluminum.

Efficiencies of the beta counting systems were determined by counting phosphorus and aluminum samples irradiated in the degraded fission spectrum and then obtaining their absolute specific activities by means of thin-window methane-flow proportional counters of known sensitivity developed and operated by the radiochemical group of this laboratory. Extensive calibration work on the latter counters⁷ has related absolute counting efficiency to average beta energy and sample thickness over a limited range of sample sizes. In terms of these parameters, the efficiency appears to be independent of the sample material. The general procedure consisted of making a series of individual efficiency determinations based upon the 4π counting (100%) efficiency) of "weightless samples" of the beta-emitting atoms which were chemically separated from macrosamples. Each macro-sample was counted previously

on the thin-window counters. The efficiencies of the two counting systems used in this experiment were thus found to be 0.30 ± 0.015 for the phosphorus samples and 0.137 ± 0.007 for the aluminum.

III. P³¹(n,p) ACTIVATION RESULTS

If the observed counting rates are converted to the equilibrium disintegration rate A, then the cross section for a thin sample is given by $\sigma = A/(N\phi)$, where N is the number of atoms in the sample and ϕ is the incident neutron flux. Table I presents the most important observed quantities involved in this relation and the resulting cross-section values. The U²³⁸ fission cross-section values used in the neutron flux calculation⁸ are also included so that any subsequent information may be applied to these data. Because there is a significant difference between uncertainties assigned to relative response and absolute cross section, they are tabulated separately.

In this experiment the sources of uncertainties are manifold. The absolute uncertainty, however, is dominated by two factors—the U^{238} fission cross section which is used in the flux measurement, and the counter efficiency. Both of these have been set at 5% leading, along with other estimated errors, to an over-all uncertainty of about 9% in absolute cross section.

When one considers the data as an excitation function, the counter efficiency can be ignored, but the uncertainty is still dominated by the U^{238} fission cross section, although in the flat region of the cross section between 2 and 6 Mev it is felt that the relative un-

⁷ B. Bayhurst and R. Prestwood (unpublished).

 $^{^8}$ Smith, Henkel, and Nobles, Bull. Am. Phys. Soc. Ser. II, 2, 196 (1957); see also reference 5.



FIG. 1. $P^{a1}(n,p)S^{a1}$ cross section as a function of neutron energy. Energy resolution is indicated by the triangles.

certainty might be as low as 3%. Some lesser uncertainties now become significant and although in many cases they represent only rather rough guesses, they are listed below for comparison. The percentages given are the maximum, there being in some cases considerable variation between runs.

Sample weight and compositionAbsolute2%Relative1%Sample positioning $1\frac{1}{2}\%$ Neutron anisotropy correction1%Background neutrons (>6 Mev only) $1\frac{1}{2}\%$ Normalization to counting standard2%Initial counting rates (rms deviation)2%

There are two exceptions to the last entry at the lowest and highest energies where the deviations are 8% and 5%, respectively.

Essentially as an independent counter-efficiency check, an additional measurement at 14.1 Mev was compared with the 90.6 ± 9 mb value given by Forbes.³ Neutron source, flux monitor and sample position were the same. The results differ by 6% which is within the estimated accuracy of each measurement.

All the phosphorus cross-section values are plotted in Fig. 1. Included are the results of Ricamo² which can be seen to differ significantly. Notably the present data are smaller at low energy and larger at high energy. The same qualitative difference appears between $S^{32}(n,p)$

$E_{n^{\mathbf{a}}}$ (Mev)	$\left(\frac{10^{3} \text{ neutrons}}{\text{cm}^{2} \text{ min}}\right)$	C ^b Initial counting rate (counts/min)	$\sigma_{n,\alpha}$ (millibarns)	د ا Absolute uncertainty (millibarns)	€2 Relative uncertainty (millibarns)
6.73 ± 0.60	0.32	46 ± 2	11.3	± 1.2	±0.9
7.45 ± 0.40	0.23	76 ± 3	25.5	± 3.3	± 2.3
8.09 ± 0.25	0.38	181 ± 3	37.6	± 4.9	± 2.8
8.58 ± 0.14	1.23	624 ± 4	39.7	± 6.0	± 3.5
8.90 ± 0.08	2.75	1675 ± 12	48.1	± 5.3	± 2.4
9.01 ± 0.04	3.68	2425 ± 13	51.3	± 4.6	± 1.5
14.1 ± 0.04	13.6	4149 ± 10	116	± 8.1	± 4.6

TABLE II. Summary of $Al^{27}(n,\alpha)$ results.

Energy spread includes effects of energy degradation in target plus angular spread of sample.
 Uncertainty shown is the root-mean-square deviation of the set of initial counting rates,



FIG. 2. Al²⁷ (n,α) Na²⁴ cross section as a function of neutron energy. Energy resolution is indicated by the triangles.

cross sections obtained by Ricamo,9 presumably under similar conditions, and later measurements by Huber and Hürlimann¹⁰ and by Allen et al.⁵ Since, however, the energies at which resonance peaks occur seem to coincide quite well, one questions the flux determinations of Ricamo, who obtained energy variation by placing samples at various angles to the deuteron beam and computing corresponding flux values from the work

of Hunter and Richards.¹¹ Since the latter workers used long counters to monitor neutron flux, some of the present discrepancy might be attributed to the then unknown details of long counter efficiency versus energy.

IV. $Al^{27}(n,\alpha)$ ACTIVATION RESULTS

Table II summarizes the results of the latest set of data for aluminum. Again the absolute and relative uncertainties are tabulated separately because of significant differences. In addition to the uncertainties discussed in Sec. III which apply here, there are others associated with both the relative and absolute positioning of samples as well as the neutron angular distribution measurements. These vary considerably and for some angles exceed 8% even in the relative case.

A measurement was again made at 14.1 Mev to compare with the value of 135 ± 9.5 mb obtained by Forbes. This time the results differ by 17% which just exceeds the sum of the estimated uncertainties. In Fig. 2, all the Al (n,α) cross-section values are plotted, the earlier data having been normalized to the later measurements at 7.45 Mev. Agreement at 6.73 Mev is reassuring.

A very recent measurement by Yasumi¹² gives the 14.1-Mev Al (n,α) cross section as 120 ± 14 mb, in excellent agreement with our value of 116 ± 8.1 mb.

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⁹ Luscher, Ricamo, Scherber, and Zunta, Helv. Phys. Acta 23, ¹⁰ P. Huber and T. Hürlimann, Helv. Phys. Acta 28, 33 (1955).

¹¹ C. H. Hunter and H. T. Richards, Phys. Rev. 76, 1445 (1949). ¹² S. Yasumi, J. Phys. Soc. Japan 12, 443 (1957).