

(*n,2n*) Cross Section of Na²³ at 14.1 Mev*

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An experimental determination of this cross section was made on six individual samples and involved the use of 4π counting, low background counting, neutron monitoring, and sample thickness corrections. The value obtained is 13.8 ± 2.2 millibarns and the error quoted is twice the standard deviation obtained from the six samples. The experimental work is described in detail and a discussion of the possible errors is given.

I. INTRODUCTION

THIS investigation describes the determination of the absolute cross section for the reaction $\text{Na}^{23}(n,2n)\text{Na}^{22}$ for neutrons at 14.1 Mev and supplements previous work^{1,2} on activation cross sections for 14-Mev neutrons. The number of (*n,2n*) events was measured by counting the positrons from Na^{22} (2.6-yr half-life; β^+ 0.542 Mev; γ 1.272 Mev). The measurement is limited in accuracy by the low value of the cross section and the long half-life of Na^{22} .

The 14-Mev neutron flux from a Cockcroft-Walton machine was monitored by Al foils which undergo the reaction $\text{Al}^{27}(n,\alpha)\text{Na}^{24}$. Absolute positron counting involved the counting of weightless Na^{22} samples in a 4π counter to establish the effective geometry of a second low background counter; and the experimental measurement of a self-absorption correction for the thick sodium chloride samples actually prepared from fast-neutron-irradiated targets.

II. EXPERIMENTAL**(a) Neutron Source**

Neutrons were obtained from the Los Alamos Cockcroft-Walton machine by bombarding a thick tritium target³ with 240-keV monatomic deuterium ions. The intensity of the neutron source was determined by counting α particles from the *d*-T reaction. The neutrons emitted at 90° with respect to the incident beam were used in the present investigation and have an energy of 14.1 Mev. However, because of the finite size of the Na foils which were irradiated, the incident neutron energy had a total spread of 13.9 to 14.3 Mev, most of the incident neutrons being 14.1 Mev. The foils consisted of NaF mixed with polythene and rolled into sheets approximately 15 mils thick.⁴ Sample foils of area 1.3 cm² were punched from these sheets by means of a special punch.

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¹ E. B. Paul and R. L. Clarke, *Can. J. Phys.* **31**, 267 (1953).

² Stuart G. Forbes, *Phys. Rev.* **88**, 1309 (1952).

³ Graves, Rodriguez, Goldblatt, and Meyer, *Rev. Sci. Instr.* **20**, 579 (1949).

⁴ These foils were prepared by Pierre Hartshorne and Marvin R. Murphy of the Chemistry and Metallurgy Division of the Los Alamos Scientific Laboratory.

(b) Irradiations

The NaF foils were sandwiched between two 5-mil Al monitor foils of identical area and were placed 2 cm from the tritium target. Al foils were also placed at 10- and 20-cm positions from the target in the same alignment. The Al foils undergo the $\text{Al}^{27}(n,\alpha)\text{Na}^{24}$ reaction with high-energy neutrons. Since at distances of 10 and 20 cm the inverse square law is found to hold, it is possible to calculate the flux (neutrons per cm²) at these distances. By comparing the counting rates of Na^{24} made in these two distant foils with that of the foils sandwiching the NaF, the flux irradiating the NaF is found.

(c) Preparation of Na²² Samples for Counting

After the Na foils were irradiated, they were ignited at 600°C to remove the polythene. The remaining NaF was dissolved in water and converted to NaClO_4 by fuming to dryness with concentrated HClO_4 . The NaClO_4 was then dissolved in *n*-butanol and the solution heated. NaCl was precipitated by the addition of *n*-butanol which had been saturated with HCl gas.⁵ The NaCl precipitate was filtered onto a tared No. 50 Whatman Filter circle ($\frac{7}{8}$ -in. diameter) on a ground-off Hirsch funnel attached to a filter chimney. (This technique gives a sample of uniform thickness and well-defined area of 2.9 cm².) The sample was dried at 110°C, weighed, mounted, and counted.

(d) The Counter

The instrument used for counting the Na^{22} samples is a very low background methane proportional counter. The background of this counter during this investigation was 3.8 counts per minute. This counter was calibrated for the absolute counting of Na^{22} in the following manner. Samples from a high specific activity Na^{22} solution (<4 μg total solids per μC) obtained from Oak Ridge were accurately measured out and evaporated to dryness on thin Zapon films suitable for 4π counting. The technique for mounting these samples and the reliability of the 4π counter had been checked previously.⁶ From the 4π count of the samples the

⁵ H. H. Willard and G. F. Smith, *J. Am. Chem. Soc.* **44**, 2816 (1922).

⁶ The author acknowledges his indebtedness to J. P. Balagna who developed the mounting technique and spent much time in establishing the reliability of the 4π counter.

TABLE I. Summary of data.

Sample	Wt. of NaCl mg	I neutron/cm ²	Net counts/min at counting time	Net counts/min corrected to irradiation time	Disintegrations/min, per mg Na $-dN/dt$	$\sigma_{n,2n}$ millibarns
1	57.4	1.865×10^{12}	1.60	2.04	0.331	13.35
			1.29	1.66		
2	81.0	4.078×10^{12}	1.35	1.78	0.693	12.79
			3.92	4.79		
			3.75	4.62		
3	75.9	4.247×10^{12}	3.71	4.69	0.761	13.48
			4.39	4.70		
			4.59	4.96		
4	34.2	7.816×10^{12}	4.73	5.24	1.305	12.56
			4.91	5.13		
			4.73	4.98		
5	36.2	3.111×10^{12}	4.52	4.89	0.630	15.24
			2.54	2.67		
			2.18	2.31		
6	51.9	3.805×10^{12}	2.35	2.56	0.771	15.25
			3.65	3.93		
			3.53	3.83		
			3.76	4.19		
			Average		13.78	
			Total spread		19.5%	
			Standard deviation		7.9%	

disintegrations per minute per ml were determined. One ml of the Na²² solution was then added to 12.0 ml of a standardized NaCl solution containing 10.0 mg of Na per ml. The resulting solution was made up to 25 ml and thoroughly mixed. Six aliquots of varying size were then taken, evaporated to dryness, and, as described under Sec. II(c), filtered, weighed, and mounted. The samples were then counted on the low background counter, and from the weight of NaCl in each sample the number of disintegrations of Na²² were calculated. A curve was obtained giving the weight of NaCl *vs* ratio of counts/min to disintegrations/min.

III. RESULTS

For the calculations of the cross section of the Na²³(*n,2n*)Na²² reaction, the following fundamental equations are required:

$$-dN/dt = \lambda N, \quad (1)$$

$$N = I\sigma n, \quad (2)$$

where $-dN/dt$ = disintegrations per minute per mg of irradiated Na; λ = decay constant of Na²² ($= 5.076 \times 10^{-7} \text{ min}^{-1}$);⁷ N = atoms of Na²² per mg of irradiated Na; I (flux) = neutrons per cm²; σ = cross section in millibarns (10^{-27} cm^2); n = atoms of Na²³ per mg of irradiated Na ($= 2.619 \times 10^{19}$). By combining Eqs. (1) and (2), one obtains

$$-dN/dt = \lambda I\sigma n. \quad (3)$$

Separate irradiations were made on six different NaF foils. After allowing the Na²⁴ formed by the (*n,γ*) reaction to decay, the foils were treated and mounted as previously described. The samples were then counted on the low background counter over a period of 3 months. Alternate one-hour sample and background counts were made until each sample had been counted a total of 10 hours. The counting process

⁷ L. J. Laslett, Phys. Rev. **76**, 858 (1949).

was repeated twice, thus giving a total of 30 hours of counting for each sample. A chi-square analysis⁸ of all the backgrounds during this period indicated that the counter was behaving properly. By means of the curve obtained giving the weight of NaCl *vs* ratio of counts/min to dis/min, each sample count was converted to disintegrations per minute. The value of n and λ are known. I was calculated for each sample from the α -monitor count and the Al foil measurements. Then by substitution in Eq. (3) of the appropriate numbers, six values for the cross section were obtained.

A summary of the data is shown in Table I.

IV. DISCUSSION

The errors in this measurement may come from the following sources: (1) contamination of samples; (2) calibration of low background counter with 4π counting; (3) the α monitor of the Cockcroft-Walton machine; (4) counting statistics of the samples; (5) using the Al²⁷(*n,α*)Na²⁴ reaction to monitor an (*n,2n*) reaction.

(1) Every precaution was taken to avoid any contamination, but one cannot say definitely that any sample is not contaminated. The internal consistency of the counting over a 3-months period at least eliminates the possibility of short-lived contamination. The chemistry performed and the fact that fast neutrons were used lead to a low probability for contamination.

(2) The 4π counter was checked carefully against our own standards as well as standards furnished by the Bureau of Standards and was shown to be accurate to ± 1 percent on both P³² and Na²² standards.

(3) The α monitor of the Cockcroft-Walton machine is believed to be accurate to ± 4 percent. During this investigation the α monitor was changed and recalibrated. The results obtained after this change showed no consistent variation in the cross-section value.

(4) The standard deviation of each 10-hour count for these samples ranged from approximately $\pm 3\frac{1}{2}$ percent for the higher counting samples to ± 8 percent for the lowest. The cross section calculated in Table I is based on an average of three such counts for each sample.

(5) The plot of energy of the neutrons from the Cockcroft-Walton machine *versus* laboratory angle is very linear in the region used in this investigation. Also in this region the Al²⁷(*n,α*)Na²⁴ excitation function was found to be quite linear. Several (*n,2n*) excitation functions on elements with high thresholds (10 to 12 Mev) have shown linearity in the 14-Mev region. The threshold for the Na²³(*n,2n*)Na²² reaction is 12.57 ± 0.21 Mev.⁹ One can conclude that the use of the Al²⁷(*n,α*)Na²⁴ reaction to monitor the Na²³(*n,2n*)Na²² reaction would not lead to a very large error.

⁸ A. G. Worthing and J. Geffner, *Treatment of Experimental Data* (John Wiley and Sons, Inc., New York, 1943), pp. 183-187.

⁹ Sherr, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).

In view of the discussion above, the error quoted for the cross section is twice the standard deviation obtained for the six measurements, namely 13.8 ± 2.2 millibarns.

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Angular Distributions and Yields of Neutrons from (p, n) Reactions

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Angular distributions of neutrons from (p, n) reactions on Mg, Al, Cu, Mo, Ag, Ta, Au, Th, and U were measured for 23-Mev protons incident on thick targets. All angular distributions show peaks in the forward direction; except for Mg and Al, the $0^\circ/180^\circ$ intensity ratios vary from 5 to 15 for ~ 13 -Mev neutrons, and from 1.3 to 5 for ~ 8 -Mev neutrons. The Mg and Al high-energy data show minima and secondary maxima. Yield determinations indicate that the number of neutrons per nuclear reaction increases with atomic number from about 0.25 for Mg and Al to about 2.0 for most heavy elements and more than 4 for Th and U (probably due to fission). Temperatures of the neutron energy distributions are estimated; for the heavy elements at the backward angles, they are about 0.85 Mev at 3-Mev neutron energy, and about 1.9 Mev at 10-Mev neutron energy. It is concluded that direct interactions are of considerable importance in these reactions.

It has generally been assumed that nuclear reactions induced by neutrons and protons in the 10- to 25-Mev energy region proceed by a compound nucleus interaction.^{1,2} This assumption was originally based on theoretical predictions of very short mean free paths in nuclear matter for nucleons of this energy, but experimental supporting evidence was soon forthcoming. Measurements of energy spectra of emitted neutrons³ and protons⁴ showed them to be approximately Maxwellian. Excitation functions for $(n, 2n)$ and $(\alpha, 2n)$ reactions confirmed this.⁵ Excitation functions for proton-induced reactions on Cu^{63} and alpha-induced reactions on Ni^{60} were found⁶ to be remarkably similar when plotted against the excitation energy of the compound nucleus, which is the same in the two cases. All of these facts were considered to be more or less complete confirmation of the compound nucleus hypothesis.

Recently, however, accumulated evidence indicates that mean free paths in nuclear matter are considerably longer than had previously been suspected.⁷ It has been pointed out that the deviations from Maxwellian shape in the energy distributions of emitted particles are in

the opposite direction from what is demanded by even very general considerations from compound nucleus theory.⁸ It was found that (n, p) reaction cross sections in heavy elements are considerably larger than expected from compound nucleus interactions⁹; this was tentatively explained¹⁰ by assuming that a small fraction of the reactions (~ 10 percent) take place by a direct interaction. Very convincing evidence for direct interactions was obtained by Eisberg and Igo¹¹ in their measurements of angular distributions and energy distributions from inelastic scattering of 32-Mev protons by various heavy elements.

In this paper these matters are further studied by measurement of angular distributions of neutrons from (p, n) reactions induced by ~ 20 -Mev protons. In the process, data on neutron yields were automatically obtained, so that they are also presented.

EXPERIMENTAL

Angular distributions of neutrons from (p, n) reactions were measured by means of the internal, circulating, 23-Mev proton beam of the ORNL 86-inch cyclotron. The most feasible method of detecting neutrons in an experiment of this type is by activation detectors (sometimes called "threshold detectors"). This method has frequently been criticized as being potentially subject to large errors, but a rather extensive study indicated that, in these particular experiments at

¹ N. Bohr, *Nature* **137**, 344 (1936).

² J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

³ P. C. Gugelot, *Phys. Rev.* **81**, 51 (1951); P. H. Stetson and C. Goodman, *Phys. Rev.* **82**, 69 (1951); B. G. Whitmore and G. E. Dennis, *Phys. Rev.* **84**, 296 (1951); E. R. Graves and L. Rosen, *Phys. Rev.* **89**, 343 (1953).

⁴ P. C. Gugelot, *Phys. Rev.* **93**, 425 (1954).

⁵ Brolley, Fowler, and Schlacks, *Phys. Rev.* **88**, 618 (1953); H. C. Martin and B. C. Diven, *Phys. Rev.* **86**, 565 (1952); D. J. Tendam and H. L. Bradt, *Phys. Rev.* **72**, 1118 (1947); Bleuler, Stebbins, and Tendam, *Phys. Rev.* **90**, 460 (1953).

⁶ S. N. Ghoshal, *Phys. Rev.* **80**, 939 (1950).

⁷ Feshbach, Porter, and Weisskopf, *Phys. Rev.* **90**, 166 (1953).

⁸ B. L. Cohen, *Phys. Rev.* **92**, 1245 (1953).

⁹ E. B. Paul and R. L. Clarke, *Can. J. Phys.* **31**, 267 (1953).

¹⁰ H. McManus and W. T. Sharp, *Phys. Rev.* **87**, 188 (1952).

¹¹ R. M. Eisberg and G. Igo, *Phys. Rev.* **93**, 1039 (1954).