DETERMINATION OF (d, α) REACTION CROSS-SECTIONS*

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Abstract—Absolute cross-sections were determined for several (d, α) reactions. Thin targets were bombarded by a beam of 7·8 MeV deuterons which were collected in a Faraday cup and measured by a current integrator. Product nuclei were separated chemically from extraneous activities. Absolute disintegration rates were determined by 4π β-counting. Cross-section values and estimated standard deviations are reported for formation of the following nuclei: ^{22}Na, ^{24}Na, ^{32}P, ^{46}Sc, 68 min ^{104}Ag, ^{112}Ag and ^{111}Ag.

WHILE many relative cross-section values have been reported in the literature, few absolute cross-sections have been measured.[1,2] In particular reactions which are characterized by small cross-sections, such as the (d, α), have not been the subject of a systematic study.[3] Since measurable yields of the (d, α) reaction for elements of Z up to about 60 can be obtained with the high currents of 7·8 MeV deuterons of the University of Michigan cyclotron, this reaction was chosen for a detailed study. Furthermore, modern 4π β-counting techniques[4] eliminated the uncertainty in counting the product nuclei thereby permitting the determination of cross-section values of higher accuracy than had been hitherto possible. One such absolute value at a well-defined energy permits normalization of relatively precise excitation functions obtained elsewhere. For example, several excitation functions for the ^{24}Mg(d, α)^{22}Na reaction have been reported but absolute values for a given energy vary from 180 ± 27, or 147, to 9 ± 3 mbarns. A value of 151 mbarns, good to within ±4 per cent, as determined in this work, establishes an absolute scale for these precise relative stacked foil values.

EXPERIMENTAL[5]

A. Bombardment arrangement

The 42 in. University of Michigan cyclotron provides 7·8 MeV deuterons with deflected external beam currents as high as 100 μA. This deflected beam is magnetically focused and is collimated several times before striking the target which is placed just beyond the focal plane. The transmitted beam is caught in a Faraday cup placed behind the target. The cup is a brass cylinder, $11\frac{3}{4}$ in. long

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[1] O. U. ANDERS and W. W. MEINKE. Document number 4999, American Documentation Institute, Library of Congress, Washington, D.C. (Revised June, 1956).

[2] N. JARMIE and J. D. SEAGRAVE, Los Alamos Scientific Laboratory Unclassified Report LA-2014 (March 1956).

[3] J. P. BLASER, F. BOEHM, P. MARMIER and P. SCHERRER, *Helv. Phys. Acta* **24**, 441 (1951).

[4] B. D. PATE and L. YAFFE, *Canad. J. Chem.* **33**, 610 (1955).

[5] The apparatus and procedures outlined in this section and the experimental results given later are described in considerably more detail in the Ph.D thesis of K. L. HALL, University of Michigan, 1955, and *AECU*-3126 (September 1955).

by 2½ in. inside diameter and stands on Teflon supports. The target might have been placed inside rather than in front of the cup, but this would have complicated target handling. Panofsky and Phillips[6] found no difference between the two arrangements.

Secondary electrons produced when the beam is stopped in the cup must be prevented from escaping. Similarly, electrons produced when the deuteron beam strikes the target must be prevented from entering the Faraday cup. To accomplish this a negative suppressing potential of at least 200 V was applied to an insulated brass ring mounted in front of the cup.

B. Beam measurement

The current collected by the Faraday cup was integrated with a modified form of Watts' integrator circuit.[7] The output powered a digital counter which gave a record of the time variation of the beam. The integrator was calibrated by feeding in a known current from a high voltage power supply and observing the counting rate with the input condenser shorted out. The difference between

TABLE 1.—Data on cyclotron targets

	Mg	S	Ti	Cd
Melting point (°C)	651	113	1800	321
Boiling point (°C)	1110	445	3000	767
Purity (%)	"High purity"	99·98	99·99	"Spec-Pure"
Supplier	Dow	Stauffer	Foote	Johnson-Matthey
Preparation method	Evaporated	Poured	Foil	Evaporated
Substrate	Mylar	Polystyrene	—	Aluminium
Thickness (mg/cm²)	0·5–1·1	2·9	29	1·9

this (maximum) current and the average value when the condenser was in the circuit was less than 0·5 per cent. The connecting cable and Faraday cup were included in the calibration. Calibration constants were 39 ± 4 and $9\cdot9 \pm 0\cdot1$ counts/min/μA for two input condensers. These values did not change during the two month period of bombardment work. Day to day variations were accounted for by secondary calibrations.

The deuteron energy has been measured by Bach,[8] the results of two separate measurements being 7·89 and 7·74 MeV. At 7·8 MeV the focusing magnet dispersion was 6·5 keV/mm. The ½ in. collimating slit placed just before the focal plane gave an energy spread of 83 keV for a point source, although the actual spread was greater because of the finite source slit at the deflector. The beam energy was degraded by about 0·08, 0·21, 1·6, and 0·08 MeV respectively in passing through the magnesium, sulphur, titanium, and cadmium targets.

C. Target preparation

The targets were supported on thin plastic substrates placed over a 2¼ in. diameter hole in an aluminium frame. Usually the area covered by the target matched the hole so that deuterons striking the target would not be stopped in the frame. This procedure prevented interference due to (n, p) reactions caused by secondary neutrons.

Pertinent data for the various targets used are presented in Table 1. Thin magnesium and cadmium targets were evaporated onto their substrates in a high-vacuum metal evaporator. Sulphur targets were prepared by pouring sulphur vapour onto the polystyrene substrate.[9] A second layer of polystyrene was sealed over the target to prevent the sulphur from evaporating when struck by the deuteron beam. Titanium was used as the 0·0026 in. commercial foil.

The uniformity of the thin targets was established by determining the weight and area of several

[6] W. K. H. Panofsky and R. Phillips, Phys. Rev. 74, 1732 (1948).
[7] R. J. Watts, Rev. Sci. Instrum. 22, 356 (1951).
[8] D. R. Bach, Ph.D. Thesis, University of Michigan (1955).
[9] E. Nielsen and A. Weinstein, Brookhaven National Laboratory Unclassified Report BNL-259 (S-19) (September 1953).

pieces cut from a typical target. In the evaporated targets no variation was observed within the limits of detection (0·4 per cent standard deviation). The poured sulphur targets showed a variation of less than 2·2 per cent. The titanium foils were uniform to within about 5 per cent when measured with a micrometer.

D. Chemical separations

For the most part the (d, α) reaction products were chemically separated from other activities without addition of an inert carrier. The purpose of such carrier-free separations was to eliminate self-absorption in the absolute β-counting. For the same reason, care was exercised to prevent contamination of the sample with inert material.

The chemical yield was determined by adding a known amount of a long-lived tracer isotopic with the (d, α) product. After the short-lived components had decayed, the tracer activity was measured and the chemical yield calculated. Two bombardments were necessary to determine absolute cross-sections when the tracer isotope was also a product of the reaction studied. For the first bombardment no tracer was added and the yield of the tracer isotope was measured relative to some other (d, α) activity produced. The tracer was then added in the second bombardment and the amount of tracer recovered in the chemical procedure was calculated, the amount produced by the nuclear reaction being known.

The target substrate was always dissolved and processed with the target element. This was essential because some product nuclei were recoiled beyond the target foil. It was experimentally determined that at least 0·0005 in. of a plastic substrate was needed to catch all the recoils; in practice 0·001 in. was used. A further experiment in which a thin ($\sim$100 μg/cm^2) target of cadmium metal mounted on thin ($\sim$65 μg/cm^2) Zapon with 0·00025 in. Mylar films in front and back was bombarded with the 7·8 MeV deuterons showed no detectable backward recoil and hence recoil catcher films have been used only behind the target[10] in the following bombardments.

E. Absolute β-counting

The disintegration rate of the samples were determined by 4π counting methods. The advantages of this technique for absolute measurements have been described by several authors.[4,11–14] Problems of decay scheme, particle scattering, secondary radiation, and self-absorption are largely eliminated. With this technique, β-rays can be counted absolutely to within about 1 per cent provided the electrical field of the counter and the counting circuit are properly adjusted. Only in the case of electron capture branching[15] or delayed states with lifetimes comparable to the resolving time of the counter are there any major discrepancies between the counting rate and disintegration rate. With a 250 cm^3 chamber, samples as low as 10 disintegrations/min can be assayed.

In this work absolute decay curves of samples containing several isotopes were obtained directly with a Borkowski type[16] 4π counter. Methane was the counting gas used. The counters were operated in the proportional region (about 4500 V for pure methane) so that high counting rates could be measured without large coincidence losses. The dead time, measured by following the decay of ^{116}In over 13 half-lives, was 7·9 μsec (a correction of 1·3 per cent at 100,000 c/m). The 4π chamber was placed in a copper box for electrical shielding to reduce the input capacitance and hence the resolving time and the attenuation of pulses entering the amplifier. When 2 in. of lead surrounded the copper box, the background of the counter was about 100 counts/min.

Samples were prepared on thin Zapon films whose thickness was determined by the absorption of ^{63}Ni β-rays.[17] The radioactive deposit was usually covered with another layer of film to prevent contaminating the counter.

Film absorption corrections may be required if a significant fraction of the electrons from the low energy portion of the β-ray spectrum are absorbed in the Zapon film. While several methods

[10] S. C. Fung, Ph.D. Thesis UCRL-1465 (1951).
[11] F. G. Houtermans, L. Meyer–Schützmeister and D. H. Vincent, Z. Physik **134**, 1 (1952).
[12] L. Meyer–Schützmeister and D. H. Vincent, Z. Physik **134**, 9 (1952).
[13] R. Cohen, Ann. Physik (12th series) **7**, 185 (1952).
[14] J. L. Putman, Brit. J. Radiol. **23**, 46 (1950).
[15] M. H. MacGregor, Ph.D. Thesis, University of Michigan (1953).
[16] C. J. Borkowski and T. H. Handley, ORNL-1056 (1951).
[17] B. D. Pate and L. Yaffe, Canad. J. Chem. **33**, 15 (1955).

for making these corrections have been reported[11–13,15,18–22] the extrapolation method[21] was adopted for use here. The counting rates of samples on films of varying thicknesses, made from multiple Zapon layers, were extrapolated to zero thickness. This involved extrapolation over a thickness of about 15 μg/cm^2 corresponding to a range of about 2 keV electrons.[23] The samples were counted several times during decay to detect changes in film absorption as different components decayed.

Even in carrier-free separations, self-absorption may be significant due to inactive foreign material entrained in the active deposit. A lower tolerance limit for such material was experimentally determined to be ~0·5 μg of material for 0·3 MeV β-rays.

When a non-conducting plastic film is used as the source mount, a small field-free space will exist at the centre of the chamber and very weak β-rays may not escape this region to produce a pulse. This situation may be remedied by applying a thin metallic conducting layer to the film, or by reducing the aperture in the aluminium sample plate to a size where the effect is negligible. MANN and SELIGER[24] found that a low-field correction is necessary for the 0·3 MeV ^{60}Co β-ray but not for the 1·7 MeV ^{32}P β-ray when an 0·8 in. aperture is used. BORKOWSKI[16] reported that no conducting coating is necessary if the aperture is 0·5 in. or less in diameter. In this work, a 0·5 in. diameter aperture was used, and excellent agreement was obtained with the National Bureau of Standards' standardization of ^{32}P. When the existence of a voltage plateau was taken as a criterion[4] of proper functioning, the counter used here was found to work equally well with or without a gold coating for the isotopes measured. Therefore, no conducting layer was used for routine 4π counting.

Discrimination curves[18] were obtained for three voltages and three gains each time the absolute β-activity was determined. This was repeated as the sample decayed and the radiations changed in the isotopic mixture. The slope was usually zero over the flat portion of the discrimination curves.

RESULTS

A. Magnesium bombardments

Only the 2·60 year ^{22}Na and 14·93 hr ^{24}Na were observed as (d, α) products of magnesium. To determine the chemical yield, ^{22}Na tracer was added when the target was dissolved. Sodium was separated carrier-free by dissolving in a sulphuric acid–hydrogen peroxide mixture, fuming of the large excess of acid, neutralizing the remaining acid with magnesium, and adsorbing[25,26] the solution on Dowex-50 ion exchange resin in the H$^+$ form (obtained from Bio-Rad Laboratories, Berkeley, California). The sodium was eluted with 0·5 N HCl, and the eluant fractions were concentrated and transferred to the source mount for counting.[5]

There was no evidence of a slope in the plateau region of the discrimination curves of either ^{22}Na or ^{24}Na. The film absorption curves are also flat. The magnitude of the aliquoting error was shown to be about 2 per cent. The decay curve was resolved by least squares analysis.

Absolute cross-sections based on a single determination were:

$$\sigma(^{22}\text{Na}) = 151 \pm 6 \text{ mbarns at } 7\cdot8 \pm 0\cdot1 \text{ MeV,}$$

$$\sigma(^{24}\text{Na}) = 94 \pm 4 \text{ mbarns at } 7\cdot8 \pm 0\cdot1 \text{ MeV.}$$

[18] H. H. SELIGER and L. CAVALLO, J. Res. Nat. Bur. Stand. 47, 41 (1951).
[19] W. B. MANN and H. H. SELIGER, J. Res. Nat. Bur. Stand. 50, 197 (1953).
[20] R. C. HAWKINGS, W. F. MERRITT and J. H. CRAVEN, Recent Developments and Techniques in the Maintenance of Standards. Proceedings of a Symposium held at the National Physical Laboratory (May 1951).
[21] D. B. SMITH, AERE/I/R-1210 (1953).
[22] G. A. SAWYER, Ph.D. Thesis, University of Michigan (1950).
[23] R. O. LANE and D. J. ZAFFARANO, Phys. Rev. 94, 960 (1954).
[24] W. B. MANN and H. H. SELIGER, J. Res. Nat. Bur. Stand. 50, 197 (1953).
[25] J. BEUKENKAMP and W. RIEMAN, III, Analyt. Chem. 22, 582 (1950).
[26] V. J. LINNEBOM, J. Chem. Phys. 20, 1657 (1952).

The errors quoted are estimated standard deviations based on a consideration of the errors of each of the component measurements.[5] The ^{22}Na cross-section includes a correction for electron capture branching. This was based on an 8 per cent branching[27-29] and a 5 per cent electron capture counting efficiency giving an overall efficiency of 96 per cent for ^{22}Na. This value has been experimentally determined by SHERR and MILLER[28] for the BORKOWSKI 4π counter.

The above cross-section values agree closely with the 147 and 92 mbarn values reported by IRVINE and CLARKE.[30] BARTELL and SOFTKY[31] obtained 9 ± 3 mbarns for the ^{22}Na yield. Russian workers found the ^{22}Na cross-section to be 180 ± 27 mbarns.[32]

B. Sulphur bombardments

The 14·3 day ^{32}P was the only (d, α) product observed in the sulphur bombardments. This was isolated carrier-free by dissolving the polystyrene substrate in a sulphuric acid–hydrogen peroxide mixture, oxidizing the sulphur with a bromine–carbon tetrachloride mixture, neutralizing with sodium carbonate and chromatographing the resulting solution by a procedure of McISAAC and VOIGT.[33] The phosphorus was adsorbed on a column of Dowex-50 ion exchange resin upon which iron hydroxide had previously been precipitated. The activity was eluted with 0·125 N NaOH and the effluent was channeled directly through a second column of Dowex-50 resin in the hydrogen form to remove sodium ion.

The chemical yield was determined by adding 24·4 day ^{33}P in the dissolution step. Since the half-lives of ^{32}P and ^{33}P differ by a factor of only 1·7, the composite decay curves were analysed by a method outlined by WORTHING and GEFFNER[34] and applied by BILLER[35] and more recently by others[36,37] to decay curve resolution. In this method, the gross decay curve is written as a sum of two exponentials and rearranged to give

$$A e^{\lambda_2 t} = A_1{}^0 \exp (\lambda_2 - \lambda_1)t + A_2{}^0.$$

The left member is plotted against the exponential, and the slope and intercept give the initial activity of each component. In this way resolution into $\sim$2000 and $\sim$10,000 counts/min of ^{33}P and ^{32}P was accomplished. Discrimination and film absorption curves were flat for the ^{32}P samples.

The cross-section based on a single determination was found to be

$$\sigma(^{32}\mathrm{P}) = 300 \pm 200 \text{ mbarns at } 7 \cdot 7 + 0 \cdot 1 \text{ MeV.}$$

The error quoted is a rough estimate of the standard deviation, and is large because

[27] W. F. HORNYAK and T. COOR, *Phys. Rev.* **92**, 675 (1953).
[28] R. SHERR and R. H. MILLER, *Phys. Rev.* **93**, 1076 (1954).
[29] G. CHARPAK, *J. Phys. Radium* **16**, 62 (1955).
[30] J. W. IRVINE, JR. and E. T. CLARKE *J. Chem. Phys.* **16**, 686 (1948).
[31] F. O. BARTELL and S. SOFTKY, *Phys. Rev.* **84**, 463 (1951).
[32] N. A. VLASOV, S. P. KALININ, A. A. OGLOBLIN, V. M. PANKRAMOV, V. P. RUDAKOV, I. N. SERIKOV and V. A SIDOROV, *Soviet J. Atomic Energy* **2**, 169 (1957).
[33] L. D. McISAAC and A. VOIGT, Iowa State College Unclassified Report *ISC*-271 (1952).
[34] A. G. WORTHING and J. GEFFNER, *Treatment of Experimental Data* Chap. 2. John Wiley, New York (1943).
[35] W. F. BILLER, Ph.D. Thesis, *UCRL*-2067 (1952).
[36] E. C. FREILING and L. R. BUNNEY, *Nucleonics* **14**, No. 9, 112 (1956).
[37] D. H. PERKEL, *Nucleonics* **15**, No. 6, 103 (1957).

of several unknown errors, such as loss of sulphur during bombardment, volatilization during the sulphuric acid treatment, and variations in target thickness.

C. Titanium bombardments

Four (d, α) activities were produced when titanium was bombarded. Three have similar half-lives; ^{44}Sc, ^{47}Sc, ^{48}Sc with 2·4 day, 3·44 day, and 1·83 day half-lives, respectively, while the half-life of ^{46}Sc is 85 days. The latter was the isotope used in the chemical yield measurement. The titanium targets were dissolved in a sulphuric acid–hydrogen peroxide mixture and then treated with nitric acid. The scandium was separated carrier-free as a radiocolloid[38–40] by neutralizing with ammonium hydroxide to pH 8·5, adsorbing the resultant colloidal scandium on Whatman No. 50 filter paper while keeping the titanium in solution with excess hydrogen peroxide, and finally dissolving the adsorbed scandium with hydrochloric acid. Three cycles gave adequate decontamination from extraneous activities.

Discrimination curves exhibited a flat plateau but film absorption was not negligible. The decay curve was resolved into two components, the 85 day ^{46}Sc and a 2·8 day composite ^{44,47,48}Sc. Tracer ^{46}Sc, used in measuring the chemical yield, was referred to the composite 2·8 day activity. Two determinations gave cross-sections of 0·18 and 0·70 mbarns. As there was no reason to reject either, they were averaged to give:

$$\sigma(^{46}\text{Sc}) = 0\text{·}44 \pm 0\text{·}37 \text{ mbarn at } 7\text{·}0 \pm 0\text{·}8 \text{ MeV}$$

D. Cadmium bombardments

Four (d, α) products were identified in the silver fraction from cadmium bombardments. These were the 27 min and 68 min ^{104}Ag isomers, the 7·6 day ^{111}Ag, and the 3·2 hr ^{112}Ag. The silver was separated by dissolving the target in nitric acid, adding silver carrier, precipitating silver chloride, dissolving in ammonium hydroxide, scavenging with iron hydroxide and repeating this cycle three times. The chemical yield was determined gravimetrically.

The discrimination curves were flat. The cross-sections for three silver isotopes based on a single determination were found to be

$$\sigma(68 \text{ min } ^{104}\text{Ag}) = 1\text{·}7 \pm 0\text{·}2 \text{ mbarn at } 7\text{·}8 \pm 0\text{·}1 \text{ MeV}$$

$$\sigma(^{112}\text{Ag}) = 0\text{·}49 \pm 0\text{·}02 \text{ mbarn at } 7\text{·}8 \pm 0\text{·}1 \text{ MeV}$$

$$\sigma(^{111}\text{Ag}) = 0\text{·}40 \pm 0\text{·}04 \text{ mbarn at } 7\text{·}8 \pm 0\text{·}1 \text{ MeV}.$$

The errors are estimated standard deviations based upon consideration of the individual errors in each of the factors, but the unknown self-absorption error was not included.

DISCUSSION

The cross-sections for deuterons of energy E are summarized in Table 2 together with the approximate reaction energy, Q, and approximate coulomb barriers of the target to deuterons (V_1) and of the product to α-particles (V_2). The low cross-section for the cadmium reactions reflects the high coulomb barriers relative to the energy

[38] J. R. Haskins, J. E. Duval, L. S. Chang and J. D. Kurbatov, *Phys. Rev.* **88**, 876 (1952).
[39] J. E. Duval and M. H. Kurbatov, *J. Amer. Chem. Soc.* **75**, 2246 (1953).
[40] I. D. Gile, W. M. Garrison and J. G. Hamilton, *J. Chem. Phys.* **18**, 1685 (1950).

of the incident particle. This bombardment energy exceeds the barrier for the other reactions listed. Unfortunately only very general trends are apparent from this table. The titanium cross-section appears to be quite low although there is no reason to disregard the value.

TABLE 2

Reaction	E (MeV)	σ (mbarns)	Q (MeV)	V_1 (MeV)	V_2 (MeV)
$^{24}Mg*(d, α)^{22}Na$	7·8 ± 0·1	94 ± 4	2·0	3·0	5·2
$^{26}Mg(d, α)^{24}Na$	7·8 ± 0·1	151 ± 6	2·9	3·0	5·1
$^{34}S(d, α)^{32}P$	7·7 ± 0·1	300 ± 200	5·0	3·7	6·6
$^{48}Ti*(d, α)^{46}Sc$	7·0 ± 0·8	0·44 ± 0·33	4·3	4·7	9·1
$^{106}Cd(d, α)^{104}Ag$	7·8 ± 0·1	1·7 ± 0·2	8·8	8·3	15·6
$^{113}Cd*(d, α)^{111}Ag$	7·8 ± 0·1	0·40 ± 0·04	9·8	8·2	15·3
$^{114}Cd(d, α)^{112}Ag$	7·8 ± 0·1	0·49 ± 0·02	7·5	8·2	15·3

* Possible (d, α n) contribution

Contributions to the (d, α) product yield from (d, α n) reactions are possible in the cases of ^{22}Na, ^{46}Sc and ^{111}Ag; Q values for the (d, α n) reactions are —5·4, —3·9 and 0·9 MeV, respectively. The (d, α n) products ^{33}P and ^{113}Ag were not detected experimentally in sulphur and cadmium bombardments and upper limits to the cross-section ratio for (d, α n) to (d, α) reactions were set at 0·1 and 0·05 respectively for these two elements. SCHOTT and MEINKE[41] similarly report the absence of (d, α n) reactions in zirconium at this energy. Considering finally the relative abundances of target isotopes for ^{22}Na, ^{46}Sc, and ^{111}Ag, it is felt that contribution of the (d, α n) reaction is probably minor.

Interference of (d, α 2n), (d, d p n), (d, 2p2n) and (d, 2d) with the reaction studied can be ruled out on the basis of insufficient deuteron energy to overcome the strongly negative Q values.

[41] G. L. SCHOTT and W. W. MEINKE, *Phys. Rev.* **89**, 1156 (1953).