A Study of the $H^3(d, n)He^4$ Reaction*

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The nuclear reaction $H^{3}(d, n)He^{4}$ has been studied for deuteron energies from 10 kev up to 1732 kev. The yield curve exhibits a maximum at a deuteron energy of 109 kev, where the total cross section is $5.1\pm0.1 \times 10^{-24}$ cm². The experimental data accurately fit a single level Breit-Wigner resonance curve for deuteron energies below 500 kev. Above this energy the experimental cross section is larger than predicted by a single level in the compound He⁵ nucleus.

INTRODUCTION

THE large cross section of the reaction $H^3(d, n)He^4$ makes it interesting both from the standpoint of its use as a neutron source and because the magnitude of the cross section is essentially the maximum possible predicted by nuclear resonance theory. The published data indicate a maximum cross section at around 200 kev triton energy.¹⁻³ Allen and Poole give the maximum cross section as 5.0 barns.

The purpose of the present work was to make accurate measurements of the cross section over a wide range of particle energies and to compare results with predictions of a single level resonance theory.

EXPERIMENTAL APPARATUS AND PROCEDURE

In the deuteron energy range from 10.3 to 167 kev the Rice Institute Cockroft-Walton machine was used as the accelerator. The energy of the particles was determined by measuring the voltage of the set with a calibrated milliammeter and a calibrated high resistance column of 208.2 megohms. Resistor connections were protected from corona by eliminating sharp edges and by shielding with Lucite. At 180-kev set energy the measured peak to peak ac ripple was 6.5 kev or 3.6 percent, while at 100 kev the ripple was 1.6 kev or 1.6 percent.

The beam from the acceleration tube was deflected about 10° with an electromagnet in order to separate the different mass spots. Apertures of 5 mm diameter were used to define the beam entering and leaving the magnetic field. The ratios of the mass 2, 3, and 4 spots were approximately 1:2:15, respectively, other mass spots being extremely small. Under these conditions the mass 2 beam was about 10 percent to 30 percent molecular hydrogen as determined from the measured cross section using all three beams. Cross section measurements up to 110 kev could be made with the mass 3 beam.

Arrangement of the target chamber is shown in Fig. 1. Targets were mounted so that the resolved beam struck the target at an angle of 20° to the normal of the target surface. The target support extended through the target chamber wall and was insulated with Teflon so that beam current to the target could be measured. The charge striking the target was measured with a beamcurrent integrator of the type described by Watt.⁴ Secondary electrons knocked out of the target were repelled back into it by a potential of -300 volt with respect to ground applied to the target chamber walls; the target itself operated at +40 to +80 volts from ground potential because of the beam integrator. The resulting field between the target chamber and the target was such as to draw to the target secondary electrons knocked out of the analyzing magnet exit aperture. By inserting another electrode (B) between the exit aperture and the target at a negative potential of 600 volts, secondary electron currents either to or from the target were stopped.

In traveling the 35 cm between analyzing magnet and target, some of the beam particles became uncharged and were not counted by the integrator. In order to take account of this effect a large magnetron magnet was placed near the exit aperture so that all charged particles in the beam were deflected away from the target. Disintegrations observed with this arrangement were due to uncharged particles striking the target. This procedure was always executed in every run from 10 kev to 100 kev. The magnitude of the neutral beam correction was 2 to 3 percent at 20 key, and about 0.5 percent at 100 key, while at the lowest energy of 10 kev the correction varied widely from 5 to 30 percent. The neutral beam depended on the vacuum in the system, being lower with lower pressure. Operating pressures were in the range of 1.0- to 2.0×10^{-5} -mm Hg.

A liquid air trap was placed near the target in the target chamber in order to reduce carbon formation on the targets. Since the trap would hold liquid air only about two hours, the targets were always removed overnight and at any other time the trap could not be kept cold continuously. No decreases in the yields were observed after 5000 microcoulombs hit the targets. Beam currents were limited to less than 1 μ amp.

Alpha-particles from the reaction were counted in a

^{*} Supported by the AEC. A preliminary report of the work has been given at the Denver meeting (June, 1952) of the American Physical Society.

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¹ D. L. Allen and M. J. Poole, Proc. Roy. Soc. (London) A204, 488 (1950).

² E. Bretcher and A. P. French, Phys. Rev. **75**, 1154 (1949). ³ Taschek, Hemmendinger, and Jarvis, Phys. Rev. **75**, 1464 (1949).

⁴ B. E. Watt, Rev. Sci. Instr. 17, 334, 338 (1946).

proportional counter at 90° with respect to the incident beam. The counting volume was separated from the vacuum system by a mica window of 1.5-mg/cm² thickness. The aperture defining the solid angle subtended by the counter was accurately one centimeter in diameter and was placed 7.66 cm from the center of the target, therefore subtending a solid angle of 0.0136 steradian. The counter was filled to a pressure of $\frac{1}{4}$ atmosphere with a gas mixture of argon plus 5 percent CO₂. With this mica window and filling, the alpha-particles just crossed the counter and hit the opposite wall. Pulses from the counter were fed through a cathode follower pre-amplifier to a linear amplifier and a scale of 64. The alpha-particle pulses from the amplifier were 50-60-volts high, and there were no smaller pulses greater than 10 volts. Data were obtained with a bias of 35 volts.

A background count was taken frequently during the runs by reducing the analyzing magnet current to zero, which removed the beam from the target but left all other parts of the set operating. The background counting rate was less than one count per minute, while counting rates of the order of a few thousand per minute were obtained from the reaction at all but the lowest energies.

The Rice Institute Van de Graaff accelerator was used to accelerate deuterons at the higher energy range of 134 to 1732 kev. The energy of the beam was measured with a 90° analyzing magnet, which was calibrated by the 873.5-kev resonance of the $F^{19}(p, \alpha\gamma)O^{16}$ reaction. From 134 to 496 kev, measurements were made with the molecular deuterium beam; from 481 to 1732 kev, the atomic beam was used.

For the molecular beam data, a "long neutron counter" about 4 cm away from the target and at 90° with respect to the beam counted neutrons from the reaction. The counting rate was normalized to the absolute Cockroft-Walton data in the overlapping energy range.

Above about 800 kev, appreciable background neutrons were observed from the $C^{12}(d, n)C^{13}$ reaction. In order to count only high energy neutrons from the $H^{3}(d, n)He^{4}$ reaction an anthracene crystal on a 5819 photomultiplier tube was used as the neutron counter for bombarding energies above 481 kev. The anthracene crystal, which was $1\frac{1}{2}$ in. in diameter and $1\frac{1}{2}$ -in. long, was placed about 12 cm from the target at 90° to the beam. The neutron pulses cut off at about 75 volts bias at the output of the amplifier; the operating bias for taking data was 50 volts. Such a high bias was used in order to exclude counts from γ -radiation up to 5 Mev. A small correction for the variation of counter efficiency with neutron energy was required. The maximum calculated neutron energy variation at 90° was 500 kev or 3.5 percent. The cross section for neutronproton collision was assumed constant, and only the increased number of recoil protons recorded above the 50-volt bias has been considered in computing the cor-



Fro. 1. Target and counter arrangement with the Cockroft-Walton. After being deflected 10° with the analyzing magnet, the beam passed through magnet exit aperture A, through the larger opening in secondary electron guard electrode B to the target T. The alpha-particles from the reaction passed through the mica window F into the counter C. The counter solid angle was defined by a brass knife-edge aperture D. The liquid air trap was placed about 4 cm from the target.

rection. The correction was 10 percent when the bombarding energy changed from 481 to 1732 key.

TARGETS

Targets of about 10 μ g/cm² of zirconium were used at low energies; in such a layer a 50-kev deuteron loses 1 kev and so the energy of the deuteron is well defined throughout the thin layer. Targets were prepared in a manner similar to that described previously,⁵ although certain changes in technique had to be made to get good very thin targets. Since zirconium films of predictable thickness were desired, only a small, definite amount of zirconium was placed on the evaporation filament and all the zirconium was evaporated with a fixed geometry. The mass of the zirconium deposit on the targets could in such a way be determined to about 20 percent without weighing each target. To prevent all of the small amount of zirconium from being used as a getter of gas in the evaporation system, another filament was used to evaporate a larger quantity of zirconium immediately before making the regular evaporation onto the targets.

In filling the zirconium layer with tritium, care was taken to keep the target temperature from becoming too high. According to Gulbransen and Andrew⁶ a temperature of 300° C is sufficient to cause hydrogen to go into zirconium at a fast rate, and the hydrogen is actually driven out again in a vacuum if the tempera-

⁵ A. B. Lillie and J. P. Conner, Rev. Sci. Instr. 22, 210 (1951). ⁶ E. A. Gulbransen and K. F. Andrew, J. Metals 1, 515 (1949).



FIG. 2. Gas target arrangement for calibrating tritium targets with the Van de Graaff. The analyzed beam passed through collimator A, through the secondary electron guard electrode B, through the foil F into the gas target. Fluorescent ZnS was placed on the metal body around the foil to make sure none of the collimated beam was lost. Connections to A and B were made by colloidal graphite lines painted on the inside of the glass tube G. The back plate of the chamber could be removed for insertion of targets. Neutrons were detected by a "long neutron counter" placed 4 cm from the gas chamber at 0° to the beam.

ture rises above about 450° C. The temperature of the targets was always kept below that necessary for the targets to have any red color as viewed in a darkened room.

The uniformity of the targets was checked in three different ways. Kodak type 103a-0 spectroscopic plates were exposed to the activity of the tritium in the targets and the developed plates served as a good indicator of uniformity. Targets were also placed in an alpha-ray electroscope, and the activity of different parts of the targets were measured. Tests were also made by making the deuteron beam strike different parts of the target. No target was used which had sharp changes in activity along the surface.

To take account of possible absorption of tritium by backing materials during the filling process, measurements were made on the amount of tritium on the bare metal of the back side of the targets with the alpha-ray electroscope and with the Cockroft-Walton set. All tungsten backed targets had essentially zero tritium on the back side. Aluminum and beryllium backed targets had some tritium primarily as a thin adsorbed layer. An excitation curve taken on the back side indicated the thickness of the layer was less than 10 kev at a deuteron energy of 110 kev. The amount of tritium on the back side of one beryllium backed target was 1 percent of that in the zirconium layer, on the other 3 percent, and on the aluminum backed target 3 percent. Another good reason for filling the targets at a low temperature was that the amount of tritium adsorbed on the backing material was greatly decreased.

To determine the cross section, the number of atoms/ cm² of tritium in the targets had to be measured. In order to have a low neutron background, a different nuclear reaction $H^{3}(p, n)$ He³ was used to produce neutrons from the tritium-zirconium targets and from a tritium gas target whose pressure, temperature, and purity could be measured. Protons were accelerated by the Van de Graaff to an energy of 1.4 Mev and entered the gas target chamber, shown in Fig. 2, through an aluminum foil 1.26-mg/cm² thick. The path length of the beam in the gas was 1.57 cm, the bulge of the foil being taken into account in computing the exact number of atoms/cm² of gas presented to the beam. Tritiumzirconium targets were mounted behind the foil and the chamber filled with ordinary hydrogen gas when taking a counting rate on the targets. The diameter of the beam used for calibration purposes was 5 mm. Neutrons from the reaction were counted by a "long neutron counter" about 5 cm from the target chamber and at 0° to the incident beam. With a fixed geometry the number of tritium atoms/cm² in a tritium-zirconium target was equal to the ratio of counting rates for gas and T-Zr targets times the number of atoms/cm² of gas.

The pressure of tritium gas in the target chamber was measured with a mercury manometer and a traveling microscope. Mercury was used rather than oil to prevent exchange of tritium and hydrogen. The measured values of the pressure were very consistent, having less than one percent spread in the values obtained by reading the manometer several times. Counting rates were always taken at two different pressures. The actual background was measured by putting a blank of the backing material into the target chamber and bombarding with protons. The background was negligible for the tungsten backed targets, 2 percent for the aluminum backed targets, and was 42 percent for one beryllium backed target. This high background is believed to be due to the neutrons from beryllium plus alpha-particles, where the alpha-particles are due to the Be⁹(p, α)Li⁶ reaction. Four sets of calibrations were made, the first being made with tritium gas whose purity was measured afterward and found to be 49-percent tritium. The last three measurements were made with gas which was 99percent tritium at the start and 88-percent tritium after the last calibration as determined by a Nier-type mass spectrometer. The tritium concentration was assumed to change linearly with use.

In making the target calibration the beam struck the targets normally to the surface; in the Cockroft-Walton set the targets were tilted so that the beam struck at an angle of 20° with respect to the normal. The effect of the tilt is to increase the target thickness presented to the beam by 6.0 percent.

From the mass/cm² of zirconium on the targets, the thickness in kev of the tritium-zirconium layer has been calculated from results of Warshaw.⁷ One-half the thickness has been subtracted from the observed incident deuteron energy to get the average energy of the beam in the target. This correction amounts to only 0.2 kev at 10 kev-deuteron energy, and so a small error in the energy loss is relatively unimportant.

At very low energy some appreciable scattering of the deuterons may take place in the tritium-zirconium layer. A scattered deuteron would travel through a greater thickness of tritium and would lead to too high a value for the measured cross section.

The mean square net scattering angle for multiple scattering was calculated⁸ taking into account the fact that the target was tilted at 20° to the normal. This correction amounts to an increase of path length in the target of 2.0 percent at 10 kev and 0.9 percent at 15 kev.

A possible objection to using the tritium-zirconium targets is that some of the incident deuterons might be scattered more than 90° in the backing material, pass through the tritium layer twice, and, if they emerge still charged, not be counted by the current integrator. Both effects would increase the experimentally determined value of the cross section. At the lowest energies, where the backscattering effect would be important, the stopping powers of all atoms are roughly equal. The backscattering effect should then depend on the Z of the scattering material in the same way as Rutherford scattering cross section does, that is, as Z^2 . In order to measure experimentally what the backscattering might be, the tritium-zirconium films were prepared on tungsten, aluminum, and beryllium backings polished to a good mirror surface. The plot in Fig. 3 of data

TABLE I. The differential cross section (σ) at 90° versus the bombarding deuteron energy at the center of the target (*E*). Where measurements were made with more than one target at a particular energy, the average value is given, and the targets involved are indicated.

E kev	σ barns per steradian	Al	Target Be – 1	s Be —2	$E_{ m kev}$	σ barns per steradian	Al	Target Be —1	s Be — 2
10.3 15.4 20.7 25.8 31.0 34.5 36.3 41.5	0.00013 0.00135 0.00491 0.0125 0.0233 0.0348 0.0401 0.0593	xxxxx xx	××× × ×	*****	72.8 76.2 78.0 83.3 90.3 93.7 97.1 104	0.260 0.279 0.284 0.330 0.351 0.384 0.391 0.404	× × × ×	×× × ××	xxxxx xx
46.7 48.5 52.0 55.4 57.1 62.3 67.5 69.3	0.0845 0.0939 0.118 0.141 0.141 0.183 0.224 0.238	× ×	×××× ×	*****	109 111 115 125 136 146 156 167	0.406 0.405 0.401 0.385 0.360 0.329 0.300 0.272	* *****	×××× × ×	******

⁷ S. D. Warshaw, Phys. Rev. 76, 1759 (1949).

⁸ E. Fermi, Nuclear Physics (University of Chicago_Press Chicago, Illinois, 1950), p. 37.



FIG. 3. Effects of target backing on measurements of the $H^{3}(d, n)He^{4}$ cross section. Crosses indicate tungsten backed Zr-T targets, open circles aluminum backed targets, and solid circles, beryllium backed targets.

obtained from the three kinds of targets indicates the results. A tungsten backing is seen to give about 40 percent larger cross section at 15-kev energy than a target with a beryllium backing. If the backscattering goes as Z^2 then aluminum should give about 1.3 percent effect and beryllium 0.1 percent. The experimental data show no appreciable difference between beryllium and aluminum backings, a result which is consistent with backscattering which varies as Z^2 .

RESULTS

The variation with energy of the differential cross section at 90° is given in Table I and Table II. Results in Table I were obtained with the Cockroft-Walton and represent data from one aluminum backed target and two beryllium backed targets. The targets were $10-\mu g/cm^2$ thick. The statistical probable error of the values from each target was about 1 percent except for the points at 10.3 and 15.4 kev. Table II gives the

TABLE II. Cross section data at higher energies. The differential cross section (σ) at 90° as a function of the deuteron energy (*E*) at the center of the target.

E kev	σ barns per steradian	$E \\ kev$	σ barns per steradian	$E \\ \mathrm{kev}$	σ barns per steradian
134	0.363	302	0.0989	774	0.0230
143	0.339	329	0.0875	864	0.0200
153	0.309	357	0.0766	957	0.0183
162	0.276	402	0.0655	1055	0.0166
173	0.260	448	0.0547	1156	0.0143
183	0.245	481	0.0506	1263	0.0125
205	0.198	496	0.0488	1374	0.0117
227	0.167	533	0.0434	1489	0.0111
251	0.142	607	0.0349	1608	0.0099
276	0.118	689	0.0278	1732	0.0090

data at higher energies as obtained with the Van de Graaff accelerator. A zirconium-tritium target which was 40 μ g/cm² was used in the energy interval 134 to 496 kev and a thicker target of 200 μ g/cm² in the higher interval of 481–1732 kev. These zirconium targets were evaporated onto a silver and a tungsten backing, respectively, so that there would be no neutrons or γ -rays from the backing material at the higher energies which were used.

Below 130-kev deuteron energy, the angular distribution of the reaction is symmetrical in the center-of-mass system.¹ Therefore, the total cross section may be deter-



FIG. 4. Theoretical fit to the $H^{3}(d, n)He^{4}$ data. The solid line is the calculated curve, while the circles are the experimental points.

mined from measurements at 90°. Several targets have been used to determine the total cross section at 109kev deuteron energy. The average value is 5.1 barns; the individual values obtained for the different targets are given in Table III. Probable errors in the solid angle, the number of atoms/cm² in the target, and the number of incident deuterons are estimated to be about 1 percent each. The probable error in the number of counts is less than $\frac{1}{2}$ percent. Thus, the over-all probable error is about 2 percent or 0.1 barn.

An attempt has been made to fit the experimental data with a single level, Breit-Wigner, formula. It has been found that the fit obtained is very much improved by using the more exact formula⁹

$$\sigma = \frac{2J+1}{(2s+1)(2i+1)} \frac{\pi}{k^2} \frac{\Gamma_d \Gamma_n}{(E_\lambda + \Delta_\lambda - E)^2 + \frac{1}{4}(\Gamma_d + \Gamma_n)^2}$$

where J is the angular momentum of the compound nucleus, s and i are the spins of the incident particle and initial nucleus, respectively, k is the wave number of relative motion, Γ_d and Γ_n are deuteron and neutron

TABLE III. Values of the total cross section obtained with different targets at 109 kev deuteron energy.

Target backing	$T-Zr$ thickness $\mu g/cm^2$	Total	cross section, barns
Al Al Al Al Al Be	30 10 10 10 10 10 10 10		5.20 4.96 5.00 5.10 5.13 4.99 5.29
		Average	5.10 ± 0.03

widths, respectively, E_{λ} is a constant, Δ_{λ} is the level shift, and E is the center-of-mass energy. The width Γ equals $2kP\gamma^2$, where P is the penetrability and γ^2 is the reduced width of the particle. The level shift Δ_{λ} is given by Thomas¹⁰ to be $\gamma_d{}^2g/a$, where a is the radius of the interaction and g is a function of the Coulomb wave functions. The penetrability of the deuteron and the values of g have been computed from tables of the Coulomb wave functions.¹¹

The maximum possible value of the resonance term is unity and occurs if $(E_{\lambda}+\Delta_{\lambda}-E)=0$ and $\Gamma_d=\Gamma_n$. A value of $\frac{3}{2}$ for *J*, the angular momentum of the intermediate excited He⁵ nucleus, is required to obtain a sufficiently large value for the cross section from the formula. Deuterons with l=0 are assumed because of the low bombarding energies. Conservation of total angular momentum and parity then requires that the intermediate He⁵ nucleus have even parity and that the neutrons given off have l=2.

The calculated fit together with the experimental

¹⁰ R. G. Thomas, Phys. Rev. 81, 148 (1951).
 ¹¹ Bloch, Hull, Broyles, Bouricius, Freeman, and Breit, Revs. Modern Phys. 23, 147 (1951).

⁹ E. P. Wigner and L. Eisenbud, Phys. Rev. 72, 29 (1947).

data are presented in Fig. 4. The parameters are $a=5.0\times10^{-13}$ cm, $E_{\lambda}=-464$ kev, $\gamma_d{}^2=10^{-9}$ kev cm, and $\gamma_n{}^2=2.8\times10^{-11}$ kev cm. The quality of the fit is not sensitive to the value of *a* which is used if the other parameters are suitably changed. Total cross-section values were obtained from the measured differential cross section by assuming a symmetrical angular distribution in the center-of-mass system. At the higher energies, where the calculated curve falls below the experimental points, the angular distribution is probably not exactly symmetrical. Deviations from a one level formula might be expected at the highest energies because of higher levels in He⁵, the compound nucleus. Also at higher energies the stripping process¹² must

¹² S. T. Butler, Proc. Roy. Soc. (London) A208, 599 (1951); S. T. Butler and J. L. Symonds, Phys. Rev. 83, 858 (1951). account for some of the effects since the results of Allred¹³ and others at 10 Mev can be almost entirely explained by stripping.

An alternative explanation of the cross-section curve for the $H^3(d, n)He^4$ reaction is that proposed by Flowers.¹⁴ His theory does not make use of an isolated state of the compound nucleus. An experimental study of the inverse process, the scattering of high energy neutrons in He⁴, would serve as a means of testing the application of the two theories to this reaction. If an isolated compound state of *He⁵ is important in the H³(d, n)He⁴ reaction, an anomaly in the scattering cross section of 20–21 Mev neutrons would be expected.

¹³ J. C. Allred, Phys. Rev. 84, 695 (1951); Brolley, Fowler, and Stovall, Phys. Rev. 82, 502 (1951).
 ¹⁴ B. H. Flowers, Proc. Roy. Soc. (London) A204, 503 (1950).

PHYSICAL REVIEW

VOLUME 88, NUMBER 3

NOVEMBER 1, 1952

Cross Section and Angular Distribution of the $He^{3}(d, p)He^{4}$ Nuclear Reaction*

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The cross section for the reaction $\text{He}^3(d, p)\text{He}^4$ has been studied for deuteron energies from 188 kev to 1597 kev. The yield of protons at 0° to the deuteron beam exhibits a maximum at 400-kev deuteron energy. At this energy the angular distribution of the disintegration particles is spherically symmetrical and the total cross section is 0.69×10^{-24} cm². At higher energies there is a preferential emission of protons at 0°. At 1600 kev the ratio of the number of protons at 0° to that at 90° is 1.08. The variation of total cross section with bombarding energy agrees with a calculated single level resonance formula from 188 kev to 600 kev. At higher energies the theoretical values give too small a cross section.

INTRODUCTION

THE reaction $He^{3}(d, p)He^{4}$ is quite similar to the reaction¹ $H^{3}(d, n)He^{4}$ which has a very large cross section at low deuteron energies. Consequently a large cross section at a low bombarding energy might be expected for this reaction. The double nuclear charge on the He³ nucleus, however, would decrease the penetrability of the deuterons into this nucleus as compared to a H³ nucleus. Previous experiments² on this reaction which were carried out by slowing down He³ particles from a cyclotron and sending these into deuterium gas, indicate a large cross section at low bombarding energies. The purpose of the present experiments was to study this reaction with deuterons of a well-defined energy from a Van de Graaff accelerator.

EXPERIMENTAL APPARATUS AND PROCEDURE

Deuterons were accelerated by the Rice Van de Graaff accelerator and passed through a 90° analyzing magnet before reaching the He³ gas target holder. A

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ⁱ Conner, Bonner, and Smith, Phys. Rev. 88, 468 (1952).

² Baker, Holloway, King, and Schreiber, Atomic Energy Commission_Declassified Report. AECD 2189 (1948) (unpublished). diagram of the gas target holder is shown in Fig. 1. The beam of deuterons was collimated by aperture A



FIG. 1. The gas target holder showing the collimating aperture A, the electron repelling electrode B, the thin aluminum foil F, and the gas chamber C.

^{*} Supported by the AEC. A preliminary report of this work was given at the New York Meeting (January, 1952) of the American Physical Society.