The He³+He³ Reactions

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A study has been made in the energy range up to 800 kev of He³ capture on He³ and the reactions that accompany this capture. The proton energy spectrum was found to confirm an earlier observation that the capture is followed by two modes of disintegration, viz., into two protons and an α particle, and into a proton and Li⁵. Improved resolution and statistics give no evidence of well-defined groups of protons of energy less than the Li⁵ ground-state group. The total reaction cross section was observed to rise monotonically in a manner suggesting partial waves of l greater than one. At 200 kev the cross section is at least 2.5 microbarns, substantiating the fact that the capture of He³ on He³ is the dominant reaction terminating the p-p cycle for stellar production of energy.

INTRODUCTION

R ESULTS have been published to indicate that when He³ captures He³ the following processes occur:1,2

> $He^{3}+He^{3}\rightarrow Be^{6*}\rightarrow Li^{5}+p+11$ Mev (1)

> > $He^{4}+2p+12.83$ Mev. (2)

The characteristic features of the reactions, such as cross section vs energy, energy spectrum of particles, etc., might reasonably be expected to resemble the corresponding features of the $H^{3}(t; n, n)He^{4}$ and $H^{3}(He^{3}; n, p)He^{4}$ reactions. Results from the studies of the latter two reactions have been published. The $H^{3}(t; n, n)He^{4}$ reaction gives inconclusive evidence of the dineutron.^{3,4} The neutron spectrum shows^{4,5} beyond much doubt a mode of decay of He⁶ in which He⁵ is left in its ground state. The presence of a group of neutrons of energy less than the He⁵ ground state group but having an approximately equal energy spread is not unequivocal.^{4,5} Certainly the cross section vs energy does not give any very concrete information on the high-lying states in He^{6.6}

The cross section vs energy characteristic of the $H^{3}(He^{3}; n, p)He^{4}$ reaction resembles that of the $H^{3}(t; n, n)$ He⁴ reaction.^{6,7} The proton energy spectrum from $H^{3}(He^{3}; n, p)He^{4}$ shows a group of protons corresponding to the mode of Li⁶ decay which leaves He⁵ in its ground state.^{7,2} At the same time, a careful examination of the proton energy spectrum does not show evidence for any group of protons of energy less than the He⁵ ground-state group. If such a group exists, it is either much less probable than the ground state group, or of much greater spread in energy, or both.

It is reasonable to suppose that additional informa-

⁶ Agnew, Leland, Argo, Crews, Hemmindinger, Scott, and Taschek, Phys. Rev. 84, 262 (1951). ⁷ C. D. Moak, Phys. Rev. 92, 383 (1954).

tion on Li⁵ could be obtained from studying the proton energy spectrum from the decay of Be⁶ formed by capture of He³ on He³. This spectrum was the first objective of the present experiment. The second objective was to place limits on the cross section for reactions (1) and (2) in the energy range 100-kev to 800-kev He³ bombarding energy. The capture of He³ by He³ has been suggested as the probable termination of the proton-proton cycle in stellar production of energy.8,9

EXPERIMENTAL TECHNIQUE

The results, which are presented in this paper, were obtained using a He³ target made by the bombardment technique and a NaI crystal spectrometer identical to the one discussed in reference 7. The additional information, beyond that already published on the reaction, results from the use of pure He³. The accelerator, ion source, He³ gas-handling equipment, and target are the same as those employed in previous studies with the He³ ion beam.^{7,10,11}

The target arrangement, which is shown in Fig. 1, has been shown before, excepting that in the present instance provision had to be made for water-cooling, as indicated. The necessity for water-cooling arises principally from the fact that helium build-up in the target does not appear to take place if the targettemperature becomes too great. The target was built up in a 2-mil foil of clean aluminum. The appearance of such a foil after He³ bombardment is noteworthy in relation to the question of He³ distribution in the target. Whereas aluminum bombarded with protons will show at most a very slight coloration where the beam has struck, aluminum bombarded by He³ has a decidedly flaky appearance after bombardment, suggesting a rupturing of the aluminum and loss of aluminum surface layers. Such a process, if it takes place, would tend to make for uniformity of concentration of He³ with depth. Assuming that He³ does not diffuse in cold

¹ Good, Kunz, and Moak, Phys. Rev. 83, 845 (1951).

² Almqvist, Allen, Dewan, and Pepper, Phys. Rev. 91, 1022 (1953).

³ Los Alamos Scientific Laboratory, Phys. Rev. 79, 238 (1950). ⁴ Allen, Almqvist, Dewan, Pepper, and Sanders, Phys. Rev. 82, 262 (1951).

⁵ W. T. Leland and H. M. Agnew, Phys. Rev. 82, 559 (1951).

 ⁸ W. A. Fowler, Phys. Rev. 81, 655 (1951).
 ⁹ E. Schatzman, Compt. rend. 232, 1740 (1951).
 ¹⁰ Kunz, Moak, and Good, Phys. Rev. 91, 676 (1953).

¹¹ W. E. Kunz, thesis (to be published).



FIG. 1. Target arrangement.

aluminum, the bombarding technique permits making a target whose thickness is known in terms of kilovolts range at one specified energy, *viz.*, the buildup bombarding energy. What is not known is the He³ distribution with thickness from the maximum depth of the target to the surface.

When a 40 μ a beam, 2 mm in diameter, of He³⁺ at 360 kev is placed upon a clean aluminum foil, a counting rate immediately appears, which energy analysis shows to be 14.72-Mev protons from the $He^{3}(d,p)He^{4}$ reaction. Because the maximum energy release from reactions (1) and (2) is 12.86 Mev, protons from these reactions do not appear beyond 11 Mev. The energy resolution of the NaI crystal used was a few percent in the energy range 11 to 15 Mev, and, hence, the 14.72-Mev contamination protons, and their recoil α particles as well, could easily be removed by the use of two pulse-height discriminators. In order to do this one pulse-height discriminator was set to count all pulses larger than, say, 3 Mev; the other pulse-height discriminator was set to count all pulses larger than 12 Mev. The difference between these two pulseheight counting rates will hereafter be called the "integral He³+He³ rate." This "integral He³+He³ rate" did not include the entire proton spectrum from the reactions being studied, but it did exclude both protons and α particles from the contamination He³(d, p)He⁴ reaction. Employing a 40 μ a beam of about 0.1 cm² at 360 kev, this "integral He³+He³ rate" was initially zero. However, it rose under the conditions just specified to an equilibrium value of several thousand counts/min in about thirty minutes.

The proton energy spectrum and the cross section with energy were both obtained in the same target apparatus with the NaI spectrometer at 90° to the beam. In determining the proton spectrum the "integral He^3+He^3 rate" served to monitor the number of He^3+He^3 disintegrations against which the relative number of protons in various energy intervals were compared. The measured cross section was for all protons in the "integral He^3+He^3 rate" range of energies.

For the study of the proton spectrum a maximum counting rate is desired. The maximum counting rate would be achieved by bombarding with as large a current density as possible at as high voltage as possible. Actually since many hours of continuous counting were required by the single-channel differential pulse-height selector used to measure the pulse spectrum, less than maximum currents and voltage were employed. The 40 μ a of He³⁺ at 360 kev, quoted previously, could easily be maintained for long periods, and, hence, represents the conditions under which the spectrum was obtained.

For obtaining the total reaction cross section, Kunz's method¹¹ of measuring a relative cross section was employed. That is to say, the "integral He³+He³ rate" per microcoulomb against He³ bombarding energy was compared with the He³(d, p)He⁴ rate per microcoulomb against deuteron bombarding energy on the same He³ target. Since dE/dx for helium is uncertain in the energy range up to 800 kev, it is particularly desirable that the target be thin. This requirement of a thin target was not very well met because the lowest convenient energy available for target buildup at sizeable beams was 84 kev.

A clean aluminum foil was introduced into the target apparatus and bombarded with a 40 μ a beam at 84 kev until equilibrium "integral He³+He³ rate" was achieved. Three precautions were taken to minimize uncertainties due to target nonuniformity across the area of the target. The beam was made to fill the collimator hole, the beam was carefully positioned over the last collimator hole by means of a quartz viewer, and the count rate per microcoulomb at 800 kev was taken alternately with the count rate per microcoulomb at the other energies. The 800 key was obtained from the He³⁺⁺ component of the He³ ion beam. Current measurement employed the same equipment and precautions as previously employed for the $H^2(He^3, p)He^4$ and $H^{3}(He^{3},d)He^{4}$ and $H^{3}(He^{3}; n,p)He^{4}$ cross-section measurements. For the purpose of assuring that the He³ beam did not disturb the He³ target, the currents subsequent to target buildup were kept small and the

total number of particles used to measure cross sections was an insignificant fraction of those used to make the target.

Following the determination of the "integral He³+He³ rate" per microcoulomb with energy, the He³ ion source and gas supply were quickly removed and a deuterium ion source and gas supply introduced. Using the deuteron beam, the He³(d, p)He⁴ rate per microcoulomb was obtained for deuterons up to 400-kev or 600-kev equivalent He³ energy. The ratio of the "integral He³+He³ rate" per microcoulomb to the He³(d,p)He⁴ rate per microcoulomb at the same He³ bombarding energy is numerically equal to the ratio of corresponding differential cross sections. From the fact that the $\operatorname{He}^{3}(d,p)\operatorname{He}^{4}$ cross section is known to a few percent, the cross section for reactions (1) and (2) can be obtained.

EXPERIMENTAL RESULTS

The energy spectrum of the protons from the reactions being studied is shown in Fig. 2. Comparing the energy spectrum of the $\text{He}^3(\text{He}^3; p, p)\text{He}^4$ protons with the energy spectrum of the $H^3(He^3; p,n)He^4$ protons,^{2,7} a conspicuous difference appears in the relative intensity of the Li⁵, He⁵ ground-state proton groups. In the case of the $H^{3}(He^{3}; p, n)He^{4}$ reaction there is one proton in the three-body breakup process whose energy distribution appears with the group of He⁵ ground-state protons. In the case of the He³(He³; p, p)He⁴ reaction there are two protons in the three-body breakup process whose energy distributions appear with the group of Li⁵ ground-state protons. In consideration of the number of protons in the three-body breakups, the proton spectra of the $H^3(He^3; p, n)He^4$ and $\operatorname{He}^{3}(\operatorname{He}^{3}; p, p)\operatorname{He}^{4}$ reactions appear consistent with there being for both the reactions approximately the same ratio of two-body to three-body decays. The proton energy spectra from both reactions give no evidence for groups of protons of energy lower than the Li⁵, He⁵ ground-state groups. The energy of the proton group in Fig. 2 is insufficiently well-determined to improve upon the present precision of the mass of Li⁵. The reasons for a large uncertainty in the energy of the





FIG. 3. He³+He³ total reaction cross section assuming isotropy of disintegration products.

proton group are that the proton energy spectrum from the three-body breakup process is not known and that the crystal employed to obtain the spectrum had only a single calibration point at 14.72 Mev.

The cross section of reactions (1) and (2) vs bombarding energy obtained with a target whose thickness at 84 kev was 84 kev is shown in Fig. 3. The interpretation of this yield curve is subject to the uncertainty of how the loss of energy of He³ ions varies with energy in the energy range 100 kev to 800 kev, and in addition to the uncertainty of how the He³ is distributed in depth. With regard to the distribution of He³ with depth, the three simplest assumptions are that all the He³ atoms lie at the end of bombarding energy range, that they all lie on the surface, or that they are uniformly distributed with depth between these two limits. The following argument will show that the last of the above simplest assumptions is most nearly correct. The He³ atoms cannot all lie at a depth corresponding to the



FIG. 4. Comparison of thin target and built-up target yields with bombarding energy of the reaction $\operatorname{He}^{3}(d,p)\operatorname{He}^{4}$.

range of 84 kev because, if they did, the yield at 84 kev would be low corresponding to an actual bombarding energy of zero for a measured energy of 84 kev. The measured yield at 84 kev He³ energy, on the other hand, was actually high. From this fact, it is safe to conclude that the He³ did not all reside at a depth corresponding to the range at 84 kev. The information that the He³ did not all reside at the surface is contained in the shape of the $He^{3}(d,p)He^{4}$ yield curve obtained with the He^{3} target. Figure 4 shows the $He^{3}(d, p)He^{4}$ yield curve as obtained by deuteron bombardment of the He³ target used for the reactions under investigation. Figure 4 also shows the $\text{He}^{3}(d,p)\text{He}^{4}$ thin target yield.¹¹ Comparing the two yield curves in Fig. 4, it will be seen that the yield of the "buildup" He³ target falls more rapidly with energy than does the thin target yield. Hence, it follows that the He³ cannot all reside on the surface of the target.

It was pointed out earlier in the description of the experiment that the "build-up" He³ target gave evidence of continuous flaking away at the surface and that such a process would likely tend to make the target uniform with depth. In the following interpretation of the yield curve of Fig. 3, the He³ target will be assumed to be uniformly distributed to a depth of 84 kev for He³ of 84-kev energy.

Having made an assumption about the He³ target distribution with depth, it remains to attach a scale of cross sections to the yield curve of Fig. 3, and finally to correct each energy point for target thickness. The scale of cross sections is obtained as follows: for bombardment with deuterons of 400-kev or 600-kev equivalent He³ energy, the He³(d, p)He⁴ cross section is almost at its maximum and is varying slowly with energy. The ratio of the "integral He³+He³ rate" per microcoulomb to the He³(d,p)He⁴ rate per microcoulomb will, therefore, be insensitive to target thickness and equal to the ratio of respective differential cross sections. The ratio of cross sections obtained, assuming isotropy, together with the He³(d, p)He⁴ cross section of 695 millibarns at 600-kev equivalent He³ energy, serves to place the scale of cross sections shown on Fig. 3.

There is little information on the stopping of He in the energy range below 1 Mev with which to make a correction for target thickness. A correction is shown in Fig. 3 which is based upon the stopping of low-energy α particles in helium, air and argon.¹² This correction makes the assumption, based upon the helium-airargon data of reference 12, that

$$dE/dx = \text{constant } E^{0.35}$$
, $0 < E < 400 \text{ kev}$
 $dE/dx = (dE/dx) \text{ at } 400 \text{ kev}$, $E > 400 \text{ kev}$.

In calculating the energy loss at the various bombarding energies by means of the above assumptions, the average energy loss in the target has been taken to be 84 kev at 84-kev He³ energy. Since dE/dx is increasing with energy, the rate of energy loss in the target at 84 kev is greater than the 84 kev lost in coming to rest. This means that the energy corrections applied to the observed cross section with energy are under-estimated, resulting in a corrected curve, Fig. 3, which represents at each He³ energy a value of cross section which is too low.

At sufficiently low bombarding energies it is reasonable to suppose that the cross section should depend upon energy as

$$\frac{e^{\frac{2\pi Z_1 Z_2 e^2}{\hbar v}}}{E}.$$

This expression is the form at low bombarding energies of the more general expression^{13,14}

$$\lambda/(F_l^2+G_l^2),$$

where F and G are the regular and irregular Coulomb wave functions corresponding to orbital angular momentum lh. The observed cross section has been compared with $\lambda/(F_i^2+G_i^2)$ and below 350 kev there is agreement within the experimental uncertainty. At higher energies the cross section can only be fitted with the just mentioned more general expression by including

 ¹² P. K. Weyl, Phys. Rev. 91, 289 (1953).
 ¹³ Bloch, Hull, Broyles, Bouricius, Freeman, and Breit, Revs. Modern Phys. 23, 147 (1951).
 ¹⁴ J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* 2007

⁽John Wiley and Sons, Inc., New York, 1952), p. 395.

partial waves of angular momentum of at least two units of \hbar .

No attempt has been made to compare the observed cross section with single level resonance theory because a sufficiently high bombarding energy was not achieved for good comparison and also because the level widths and level shifts have not been developed for three-body breakup.15

¹⁵ E. P. Wigner and L. Eisenbud, Phys. Rev. 72, 29 (1947).

From Fig. 3 the cross section for the reactions (1)and (2) at 200 kev is at least 2.5 microbarns. Fowler and Lauritsen¹⁶ found that for an assumed cross section of 4 microbarns, the He³(He³, 2p)He⁴ reaction is the dominant reaction terminating the p - p cycle in stellar production of energy. The essential correctness of their assumption seems established.

¹⁶ W. A. Fowler (private communication).

PHYSICAL REVIEW

VOLUME 94, NUMBER 1

APRIL 1, 1954

The Disintegration of Mo⁹⁹[†]

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The radiations of Mo⁹⁹ and Tc^{99m} have been reinvestigated by means of scintillation spectroscopy. Employing single-channel pulse-height analyzers in coincidence, it has been established that a triple cascade of gamma rays, 741 kev→41 kev→140 kev occurs. The 741-kev radiation is also coincident with a gamma ray at 181 key by an alternate branch of de-excitation which is equally probable. Radiation at 780 key is present but is non-coincident with other gamma rays. From these data, a disintegration scheme can be constructed. A gamma ray of energy 372 kev was also detected, and, although it appeared to have the proper half-period to be associated with Mo⁹⁹, was found to be not in immediate coincidence with beta rays or other gamma rays, suggesting that it might be related to an impurity.

INTRODUCTION

E ARLIER coincidence studies¹ showed the disinte-gration scheme of Mo⁹⁹ to be rather complex. Subsequent measurements²⁻⁶ have led to the conclusion that Mo⁹⁹ decays with the emission of two, or possibly three, groups of beta rays, and gamma rays having energies of 1.8, 40, 140, 142, 181, 367, 741, and 780 kev. A careful study^{5,6} of the disintegration of the 6-hour isomer of Tc⁹⁹ has revealed that it decays with cascade emission of the 1.8-kev and 140-kev quanta, and that the 142-kev gamma ray is the associated cross-over transition. The disintegration scheme of Mo99, as advanced by Medicus et al.,5 is based upon coincidence measurements between spectrometrically selected beta rays and gamma rays detected in an anthracene scintillation counter. However, the problem of the precise location in the scheme of the 181- and 367-kev gamma rays has remained unresolved. Accordingly, with the utilization of two single-channel pulse-height

analyzers in coincidence, the gamma-ray spectrum and the various cascade relationships have been reinvestigated.

THE MEASUREMENTS

For the purposes of the present investigations, a source of Mo^{99} was obtained when a quantity of MoO_3 was irradiated by slow neutrons in the Oak Ridge pile. Because no gamma rays other than those already reported were observed, extensive chemical purification was deemed unnecessary. On occasion, however, Tc^{99m} was separated from its parent element by the method of Coryell and Sugarman.⁷

In Fig. 1 is shown the pulse-height distribution generated by the gamma rays of Mo⁹⁹ in a crystal of thallium-activated sodium iodide which is three centimeters thick. In the case of the particular curve of Fig. 1, the radiation incident upon the detecting crystal had been filtered by a lead absorber of thickness about 2 g/cm^2 to reduce in intensity the 140-142 kev radiation relative to the harder gamma rays. The radiation at 78 kev arises from the emission from the absorber of the K line of lead, following photoelectric absorption of the intense 140-kev gamma ray. For the purpose of observing carefully the region of lower

[†] Assisted by the joint program of the U.S. Office of Naval

<sup>Research and the U. S. Atomic Energy Commission.
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¹ C. E. Mandeville and M. V. Scherb, Phys. Rev. 73, 848 (1948).</sup>

² Medicus, Maeder, and Schneider, Helv. Phys. Acta 22, 603 (1949).

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⁶ Mihelich, Goldhaber, and Wilson, Phys. Rev. 82, 972 (1951).

⁷ L. E. Glendenin in Radiochemical Studies: The Fission Products, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, New York, 1951), Paper No. 98, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, Part 5, Book 2.