Measurement of the (n,H^3) Cross Section in Nitrogen and Its Relationship to the Tritium Production in the Atmosphere*

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Nitrogen gas is irradiated by fast neutrons from a U^{235} fission plate and tritium is produced. The tritium is identified and counted by two different methods: (1) The gas is put into a cloud chamber where the H^3 electrons are identified by their range and counted; (2) hydrogen is separated from nitrogen by its passage through palladium and is then counted in a Geiger counter. The average cross section for fission neutrons with energy sufficient to make the reaction proceed [4.4 Mev is $(11\pm2)\times10^{-27}$ cm^2].

This cross section combined with cosmic-ray neutron data gives an H^3 production rate of between 0.10/cm^2 sec and 0.20/cm^2 sec averaged over the earth. Cosmic-ray stars eject H^3 at a rate estimated between 0.30/cm^2 sec and 0.70/cm^2 sec. These two processes maintain a world reservoir of 50 to 110 million curies of H^3. This H^3 production leads to a mean escape time of He^3 from the atmosphere of about 5 million years. This is consistent with a temperature at the base of the exosphere of 1500°K.

INTRODUCTION

RECENTLY Norwegian lake water[1] and atmospheric hydrogen[2] have been found to contain measurable quantities of H^3. The lake water is found to have of the order of 1 part tritium to 10^{18} parts hydrogen, while the atmospheric hydrogen has of the order of one part in 10^{14} hydrogen. Appreciable tritium production is necessary to maintain the H^3 concentration in water. It has been proposed[3] that the fast neutron reaction

$$(1)\quad n+N^{14}=C^{12}+H^3 \qquad (1)$$

may be the major production mechanism for natural tritium. This reaction has been reported in the laboratory.[4] The nature of the laboratory experiment was such that it was difficult to estimate the cross section. The reported value was 10^{-26} cm^2 to about a factor of 5 for neutrons from a thick beryllium target bombarded by 16-Mev deuterons.

This cross section has been measured here to an accuracy of better than 25 percent for fission neutrons. H^3 production in the atmosphere from reaction (1) is estimated to be about one-third that resulting from the direct ejection of H^3 in cosmic-ray stars. This H^3 production yields an average H^3/H^1 ratio of 2 to 5×10^{-18} in precipitation and an average H^3/H^1 ratio of 1 to 2×10^{-20} in ocean water.

Experimental Procedure and Results

A stainless steel container of 180-cc volume is filled to 1500-psi pressure with nitrogen and hydrogen or nitrogen, hydrogen, and ammonia. Most irradiations were done with a 50–50 mixture of nitrogen and hydrogen. The container is covered with a $\frac{1}{32}$-in. cadmium sheet to eliminate slow neutrons. This container is placed in the fission plate facility of the Brookhaven reactor.[5,6] The gas is irradiated for four or five days; the container is not very radioactive upon removal so no special handling precautions are necessary. The gas is emptied directly from the container into an expansion cloud chamber of 10-liter volume or into a flask connected to a palladium thimble and counter filling system.

In the normal cloud-chamber filling procedure, the cloud chamber is first evacuated. Alcohol and tank hydrogen are added to an initial pressure of 37.5 cm Hg; then the irradiated mixture is let into a chamber pressure of 115.5 cm, which is just over $1\frac{1}{2}$ atmospheres. After a few expansions, the chamber operates with well-defined tracks. There are about five tritium electrons in each photograph; less than one hundred photographs are necessary for good statistics. Independent track counts made by D. Schwarzer and the author agreed much better than the statistical accuracy. The background of slow electrons that might be confused with tritium electrons is about one per photograph. The background is determined before and after each run by filling the container in exactly the same way as with the irradiated sample and using this gas as the chamber filling. If any tritium were left in the walls of the container, it would show itself in the second background. Any tritium originally in the container walls would show itself in the first background. Table I summarizes the cloud-chamber data.

The only runs in which tritium is not noticed are the irradiations with pure hydrogen and with no U^{235} plate. In the 119-hour irradiation, the mixture is let into the cloud chamber by bubbling through a water tower. Titration of the water with HCl showed 99 percent of the NH_3 to be trapped in the water. In the 140-hour

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[6] The author wishes to thank Mr. W. Kato for helping him carry out his early irradiations.

TABLE I. Cloud-chamber data.

Chamber filling procedure	Irradiation time (hours)	Irradiated mixture	H^3 tracks per picture	Background before (tracks/pict)	Background after (tracks/pict)	Fission flux (averaged over container) neutrons/cm² sec	How flux obtained	Concentration of H^3 in chamber (no. of H^3/no. of atoms)	$\bar{\sigma}$ (millibarns)
Normal	67 with U^{235} plate	50% N_2 50% H_2	2.3 ±0.2	1.0 ±0.2	0.90±0.20	(1.0 ±0.3) ×10⁸	Estimated from pile power	(1.2±0.2) ×10⁻¹⁴	(10.7±4.0)
Normal	134 with U^{235} plate	50% N_2 50% H_2	5.3 ±0.4	1.05±0.10	1.10±0.20 (1.12±0.20)[a]	(1.4 ±0.2) ×10⁸	Thermal flux	(4.0±0.4) ×10⁻¹⁴	(12.5±2.5)
Normal	138 with U^{235} plate	50% N_2 50% H_2	5.6 ±0.4	0.70±0.17	(0.80±0.15)	(1.75±0.2) ×10⁸	Thermal flux	(4.3±0.4) ×10⁻¹⁴	(11.6±2.0)
						(1.6 ±0.2) ×10⁸	Sulfur activation		
N_2 added first to chamber	200 with U^{235} plate	100% H_2	0.75±0.17	···	···	(1.3 ±0.2) ×10⁸	Thermal flux	0	
Normal	122 without U^{235} plate	50% N_2 50% H_2	1.07±0.13	0.98±0.15	1.09±0.15	5.5×10⁵	Sulfur activation		
						0	Magnesium activation		
						2.0×10⁸	Thermal flux		
NH_3	119 with U^{235} plate	50% N_2	3.97±0.15	1.09±0.15	1.0 ±0.2	(1.9 ±0.3) ×10⁸	Thermal flux	(2.8±0.2) ×10⁻¹⁴	(8.5±2.0) +NH_3 loss
Trapped out		48.5% H_2 1.5% NH_3				(1.6 ±0.2) ×10⁸	Sulfur activation		
NH_3	140 with U^{235} plate	50% N_2	5.37±0.39	···	···	(1.9 ±0.3) ×10⁸	Sulfur activation	(4.1±0.4) ×10⁻¹⁴	(9.3±2.0) +NH_3 loss
Trapped out		48.5% H_2 1.5% NH_3				(1.8±0.3) ×10⁸	Magnesium activation		
1.3-cm D_2 added to background filling	···	···	8.15±0.55	0.95±0.10	1.05±0.10	···	···	(6.6±0.5) ×10⁻¹⁴	

[a] In this background run, the container is filled with hydrogen to 1500 psi and heated at 100°C for 24 hours.

irradiation, the mixture passes first through a liquid air trap and then through a water tower into the chamber. Titration showed 99 percent of the NH_3 to be taken out in the liquid air trap. This trapped NH_3 is later decomposed over hot Mg, passed through palladium, and put into a Geiger counter.

The cloud chamber is calibrated for tritium detection by introducing a small "known" amount of tritium either into the chamber or into the container. The "known" tritium is simply a measured amount of deuterium from a tank of deuterium whose tritium concentration has been measured to be $(3.6\pm0.1)\times10^{-12}$ by passing it through the palladium thimble into the stainless steel counter. When 7-mm pressure of this deuterium is added to a background chamber filling, (3.83±0.40) H^3 tracks/picture result. When 13 mm is added, (7.15±0.55) H^3 tracks/picture result. And finally, when 13.8 psi of this deuterium is added to the container with its background filling of 750 psi of N_2, 750 psi of H_2, and 22 psi of NH_3 and this mixture is passed through a water tower into the chamber, then (6.7±0.6) H^3 tracks/picture result. These three calibrations give one H^3 track/picture for D_2 to H_2 ratios of $(2.5\pm0.3)\times10^{-3}$, of $(2.5\pm0.2)\times10^{-3}$, and of $(2.7\pm0.3)\times10^{-3}$, respectively. Since the tank deuterium has a H^3 concentration of 3.6×10^{-12}, a H^3/H^1 ratio of 9.3×10^{-15} in the expansion chamber gives one H^3 track/picture. The hydrogen in the chamber normally consists of a combination of 32 cm of tank hydrogen with 39 cm of hydrogen from the container; therefore, the H^3/H^1 ratio for the container is (71/39) times the ratio for the cloud chamber. A column in Table I gives the H^3/H^1 ratio for the cloud chamber.

In several of the runs, samples of the irradiated gas or of the chamber filling are stored in 1-liter glass flasks. The hydrogen is later taken out of these samples by its passage through palladium and put into a counter; 1.0 to 1.5 cm of butene is added to the counter. This filling has fairly good counting characteristics with a Geiger threshold of 1150 volts for 5 cm of H_2 and 1500 volts for 15 cm of H_2. The counter is a stainless steel tube of $29\frac{1}{2}$-inch inside length and $2\frac{9}{16}$-inch inside diameter with a 0.005-inch tungsten wire; the length of exposed wire is 29 inches. This counter is placed in a low-level counting unit,[7] consisting of anticoincidence counters and shielding. This unit records the counts with and without the anticoincidence counts subtracted. Table II summarizes the counter data. The background rate in the counter is 400 counts/minute with just the shielding and 30 counts/minute with the shielding and anticoincidence counters combined. There is no difference in background with argon or hydrogen counter fillings. Since the 400 counts/minute is largely composed of minimum ionizing cosmic rays, the efficiency of the hydrogen butene filling is unity.

The palladium thimble has to be outgassed before each run since its hydrogen retention is almost 1/5 liter. Successive rows in Table II for the same sample give

[7] This unit was designed by Raymond Davis. The author wishes to thank Dr. Davis for its use.

TABLE II. Geiger counter data.

Gas mixture before Pd	Gas and press.[a] in counter	Counts/min without anticoincidence (volts above threshold) 25 volts	50 volts	75 volts	Counts/min with anticoincidence (volts above threshold) 25 volts	50 volts	75 volts	H^3 counts/min per cm H_2	H^3/H^1 ratio
	2.2 cm A	406±9	416±9	419±9	26.2±2.4	29.8±1.5	34.6±2.6		
	10.4 cm A	405±9	407±5	419±9	24.6±2.2	31.6±1.4	37.6±2.7		
50% N_2 tank	5.5 cm H_2	390±7	405±9	409±9	23.0±1.7	29.6±2.4	33.0±2.6		
	9.6 cm H_2	384±9	391±9	402±9	23.9±2.5	30.2±2.5	34.0±2.8		
50% H_2 tank	15.4 cm H_2	410±9	412±9	422±9	29.2±2.5	37.2±2.7	40.6±2.8		
Cloud-chamber calibration filling with 1.3 cm of D_2	7.2 cm H_2	454±9	454±9	474±10	93.2±4	88.8±4	94.6±4	8.7	5.2×10^{-14}
	6.0 cm H_2	412±9	440±9	439±9	84.2±4	79.2±4	78.2±4	8.4	5.1×10^{-14}
	6.4 cm H_2	427±9	445±9	466±9	85.6±4	87.0±4	100.5±5	9.5	5.7×10^{-14}
	4.9 cm H_2	444±9	454±9	460±10	74.0±4	78.8±4	78.8±4	9.6	5.8×10^{-14}
134-hour irradiation sample from cloud chamber	6.5 cm H_2	442±10	435±10	432±10	66.0±4	65.0±4	64.5±4	5.4	3.2×10^{-14}
	7.8 cm H_2	451±10	454±10	455±10	74.0±4	76.8±4	78.0±4	5.9	3.5×10^{-14}
140-hour irradiation mixture from container	2.9 cm H_2	424±7	436±7	459±7	49.0±3.1	54.0±3.2	65.5±3.5	9.0	5.4×10^{-14}
Decomposed NH_3 from 140-hour irradiation	4.9 cm H_2	485±10	500±10	490±10	104.4±4	104.9±4	111.6±5	15.7	9.5×10^{-14}
140-hour irradiation sample from cloud chamber	5.6 cm H_2	438±10	438±10	439±10	60.3±4	67.0±4	68.0±4	6.2	3.7×10^{-14}
	5.3 cm H_2	432±10	437±10	447±10	62.0±4	65.6±4	72.0±4	6.7	4.1×10^{-14}

[a] 1.0-cm to 1.5-cm butene added to counter.

results for successive runs through palladium from the same flask. These increase by about 10 percent in going from the first to the last counter filling which is indicative of some isotopic fractionation. About 25 percent of the original hydrogen is left in the flask after the last counter filling. The isotopic fractionation tends to make the counter result somewhat smaller than the chamber result. The 1.3-cm deuterium calibration gives 5.1 to 5.8×10^{-14} for the H^3/H^1 ratio in the counter compared to $(6.6\pm0.5)\times10^{-14}$ in the chamber. The 134-hour irradiation gives 3.2 to 3.5×10^{-14} in the counter compared to $(4.0\pm0.4)\times10^{-14}$ for H^3/H^1 in the chamber. The 140-hour irradiation gives 3.7 to 4.1×10^{-14} in the counter compared to $(4.1\pm0.4)\times10^{-14}$ in the chamber.

To test the possibility that some tritium produced during the irradiation might form ammonia,[8] irradiations were carried out with a mixture of 50 percent N_2, 48.5 percent H_2, and 1.5 percent NH_3. The NH_3 is trapped out in a liquid air trap and decomposed over hot Mg. This hydrogen (see Table II) has a H^3/H^1 ratio of 9.5×10^{-14} compared to 5.4×10^{-14} for the H^3/H^1 ratio of the hydrogen gas carrier. This means that when 5 percent of the hydrogen carrier is in the form of NH_3, about 10 percent of the tritium is in the NH_3. One concludes that less than 10 percent of the activity is lost because of ammonia formation when there is no ammonia carrier.

The fission flux is determined in several ways. The thermal flux at the U^{235} plate is measured by activating sodium and comparing its activation with that obtained in a standard pile where the neutron flux is known. Cadmium difference measurements are made in both cases. From this measurement of the thermal flux, a straightforward calculation gives the fission flux and its distribution over the container. The fission flux is also measured directly by activating sulfur, using the $S^{32}(n, p)P^{32}$ reaction, which has a 14-day half-life and a 1.0-Mev threshold; its cross section for fission neutrons is 50 millibarns. The fission flux is also measured by activating magnesium by the $Mg^{24}(n, p)Na^{24}$ reaction which has a 14.8-hour half-life and 4.7-Mev threshold; its cross section for fission neutrons is 1.0 millibarn. The sulfur activation is used because its long half-life averages out fluctuations in pile power during the irradiation. The magnesium reaction is used because its threshold is very close to that for tritium production in nitrogen. The pile power also gives an indication of the flux. A column in Table I gives the fission flux averaged over the container; the adjacent column tells how it was obtained. The term "thermal flux" indicates that the fission flux is determined by the sodium activation method. The different methods for determining flux agree quite well except for the run without the U^{235} plate where, of course, they should not agree.

The relation between cross section, flux, and tritium produced is

$$N_T/N_H = (N_N/N_H)tF\int_{4.4\text{ Mev}}^{\infty}\sigma(E)n(E)dE, \quad (2)$$

where N_T/N_H is the tritium to hydrogen ratio, N_N/N_H is (0.55) the nitrogen to hydrogen ratio; t is the irradiation time in seconds, F is the fission flux averaged over the container, $\sigma(E)$ is the cross section as a function of energy, and $n(E)dE$ is the energy distribution of U^{235}

[8] This possibility was suggested to the author by Dr. L. Friedman.

neutrons. Measurements[9,10] up to 17 Mev have given $n(E)dE$ the form

$$n(E)dE = (2/\pi e)^{\frac{1}{2}} e^{-E} \sinh[(2E)^{\frac{1}{2}}]dE, \quad (3)$$

where E is in Mev. According to Eq. (3), 8.5 percent of the fission neutrons have energy above 4.4 Mev. If σ is taken outside the integral and called $\bar{\sigma}$, the average cross section for fission neutrons above 4.4 Mev, then relation (3) has the form

$$N_T/N_H = N_N/N_H l F \bar{\sigma}(0.085). \quad (4)$$

The last column in Table I is obtained by substituting the measured quantities into relation (4). In the last two irradiations 10 percent need be added to the cross section to account for the tritium trapped in the NH_3. The average of the five runs give (11 ± 2) millibarns for $\bar{\sigma}$.

If the Coulomb factor for tritium escape from C^{12} is included in the cross section, it has the form

$$\bar{\sigma}' = \sigma_0 \exp(-Z_1 Z_2 e^2/\hbar v_T) = \sigma_0 \exp[-1.51/(E-4.4)^{\frac{1}{2}}], \quad (5)$$

where E is the energy of the incident neutron in Mev. The substitution of relations (5) and (3) into (2) gives

$$\bar{\sigma}' = (46\pm9) \exp[-1.51/(E-4.4)^{\frac{1}{2}}] \text{ millibarns}. \quad (6)$$

The cross sections $\bar{\sigma}$ and $\bar{\sigma}'$ give the same result for the nitrogen tritium production by fission neutrons and, as we shall see in the next section, approximately the same result for tritium production by cosmic-ray neutrons.

Tritium Production in the Atmosphere

Neutrons in the atmosphere originate from cosmic-ray stars. The rate of neutron production in the atmosphere is the sum of the C^{14} production, the escape of neutrons into space, and the neutron loss by (n, α) and other reactions in air. The mechanism of star production need not be considered here. We shall be concerned only with neutrons having less than 20-Mev energy.

The C^{14} production rate[11] averaged over the earth is 2.23/cm² sec. The number of neutrons that escape into space can be obtained directly from Yuan's measurement[12] of the slow neutrons as a function of altitude. A plot of Yuan's cadmium-difference measurement as a function of atmospheric pressure shows a broad maximum at 10-cm Hg pressure followed by an exponential decrease for pressures greater than 20 cm. This exponential follows the star-production curve which continues exponentially to the top of the atmosphere. The difference in area between the exponential to the top of the atmosphere and the measured slow neutron curve gives the number of neutrons that escape into space. The reason is quite simple. If neutrons were ejected from stars with less than 0.4 ev, then the cadmium difference measurement as a function of altitude would be parallel to the star-production curve. More detailed considerations[13–15] based on the solution of the diffusion equation would give the same answer. The difference in area between the curves equals the area under the slow neutron curve. The number of neutrons that escape, therefore, equals the number of neutrons of energy less than 0.4 ev that produce C^{14}. According to Ladenburg,[16] half of the C^{14} production is by neutrons of less than 0.4 ev; therefore, the neutron escape rate is 1.1/cm² sec. The number of neutrons lost by reactions other than (n, p) in air is estimated to be less than 1.0/cm² sec. The rate of neutron production averaged over the earth is

$$P_n = 2.2+1.1+1.0 = 4.3 \text{ neutrons/cm}^2 \text{ sec}. \quad (7)$$

The number of scattering collisions required to reduce the energy of a neutron from E to 4.4 Mev is

$$\nu = \ln(E/4.4)/\rho, \quad (8)$$

where ρ is the average logarithmic energy decrease per collision; ρ is 0.130 for air. The probability of producing H^3 in a collision is $0.80\,\sigma/\sigma_s$, where σ is the cross section for reaction (1); σ_s is the scattering cross section (1.5 barns); and 0.80 is the nitrogen concentration of air. The fraction of neutrons which make ν collisions without producing H^3 is

$$f = (1-0.80\sigma/\sigma_s)^{\nu}. \quad (9)$$

If E is 10 Mev, ν is 6.3. And if 11 millibarns is used for σ, then f is 0.963 which means that 3.7 percent of the neutrons produce H^3. If σ is taken as $44 \exp[-1.51/(E-4.4)^{\frac{1}{2}}]$ millibarns, then 4.7 percent of the neutrons produce tritium. A lower initial neutron energy allows a larger fraction of neutrons to escape H^3 production. If the neutron energy distribution given by Bagge[17] is used instead of 10 Mev, then the tritium production is 40 percent smaller. An initial neutron energy of 10 Mev combined with the solution of the diffusion equation is consistent with the slow neutron data;[14] higher initial neutron energies are not. The tritium production by reaction (1) is between 3 and 5 percent of P_n or between 0.10 and 0.20/cm² sec averaged over the earth.

How does this production rate compare with H^3 production by other processes? A number of other processes will be considered, but only one of them gives a greater rate than reaction (1). It is the direct ejection of H^3 in cosmic-ray stars. The number of neutrons from

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stars approximately equals the number of singly charged particles from stars. Measurements[18] on the ratio of protons to deuterons to tritons have been made for cosmic-ray stars in emulsions. A measurement[19] of this ratio has also been made for the bombardment of Be and C by 330-Mev protons. The highest percentage of tritons is found for particles from cosmic-ray stars in the 15–50-Mev range. Here[20] the proton-deuteron-triton ratio is 3–2–1, which means that 16 percent of the singly charged particles are tritons. The cyclotron measurement[19] gives for C a $H^3/H^1+H^2+H^3$ ratio of 12 percent. If the percentage of tritons among the singly charged particles from air stars is taken to be between 7 percent and 16 percent, then the resulting H^3 production rate is between 0.30 and 0.70/cm² sec. The slow neutron reaction on Li^6 in the ocean gives less than 10^{-6} tritons/cm² sec; the capture of neutrons in deuterium gives less than 5×10^{-9} triton/cm² sec; the reactions of spontaneous fission neutrons on terrestrial material give less than 10^{-11} triton/cm² sec. The relatively low number of cosmic-ray primaries (about 0.10/cm² sec) and their large energy make any appreciable contribution by tritons in the primaries themselves unlikely. However, the possibility of the accretion of tritium from space cannot be completely ruled out.

The two main processes, direct ejection from stars and reaction (1), give a total of between 0.4 and 0.9 triton/cm² sec. This production takes place largely at altitudes in the atmosphere between 0.5- and 30-cm pressure. The tritium will be oxidized, swept down by rain, and then diluted in the ocean. The yearly precipitation[21] over the oceans is 110 cm and over the continents is 66 cm. The ocean area is 71 percent of the earth surface; about 25 percent of the precipitation over the continents flows into the ocean. Therefore, the precipitation per cm² per year going into the ocean is $[110(0.71)+66(0.25)(0.29)]=84$ cm. On this basis rain should have on the average a H^3/H^1 ratio between 2 and 5×10^{-18}, in agreement with what has been found for Norwegian lake water.[1] This H^3 in rain goes into the ocean where it decays into He^3. The average H^3/H^1 ratio for ocean water is on this basis between 1 and 2×10^{-20}.

During the course of time He^3 builds up in the atmosphere both by H^3 decays and by the direct ejection of He^3 from stars. The He^3 star rate is approximately equal to that of H^3. Therefore, the He^3 production rate is between 0.7 and 1.5/cm² sec. This is also the escape rate of He^3 from the earth. Since there are 10^9 moles of He^3 in the atmosphere,[22] the mean escape time of He^3 is between 3 and 6 million years.

The escape of He^3 and of He^4 is determined by the temperature at the base of the exosphere. Estimates[23] of the escape of He^4, based on the natural α decay in rocks and their erosion, give a mean escape time of He^4 of about 3×10^8 years. The above escape times for He^3 and He^4 are consistent with a temperature of 1500°K at the base of the exosphere.[24] If this temperature were as low as 1000°K and the He^3 escape time 3 million years, then He^4 cannot escape. On the other hand, if the escape is caused by high temperature in the exosphere for short periods of time (for example, temperatures above 2800°K), then the escape time of He^4 would be too close to that of He^3 to be consistent with the erosion of rocks.

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