Tritium and Neutron Production by 2.2-Bev Protons on Nitrogen and Oxygen*

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The cross sections for tritium production by 2.2-Bev protons on nitrogen and oxygen are measured to be 28±4 and 33±4 mb, respectively. The cross sections for neutron production by 2.2-Bev protons are measured to be 1000±400 mb for nitrogen and 650±250 mb for oxygen. These results indicate that cosmic-ray primaries produce approximately 0.05 tritons/cm² sec and 1–2 neutrons/cm² sec in the first nuclear interaction with the atmosphere.

I. INTRODUCTION AND SUMMARY

NEUTRONS and light elements are produced in the high-energy nuclear interactions of protons on oxygen and nitrogen. The yields of these reactions are of interest both for the study of high-energy reactions and because of their occurrence in the interactions of cosmic radiation with the atmosphere and with meteorites. The cross sections for the (p,T) reactions on nitrogen and oxygen with 2.2-Bev protons accelerated in the Brookhaven cosmotron have been measured to be 28±4 and 33±4 mb, respectively. The cross sections are approximately 10 percent of the geometric cross sections for these elements, indicating triton emission to be a common event in high-energy interactions.

The neutron production cross sections have been measured to be 1000±400 mb for nitrogen and 650±250 mb for oxygen corresponding to about 3 and 2 neutrons per interaction, respectively. The neutron yields are obtained from measurements in the vicinity of 90° to the incident beam and therefore are largely evaporation neutrons.

The primary cosmic radiation is composed of protons and heavier nuclei in proportions varying with latitude, with energies of several Bev/nucleon.[1] The number of primary nucleons[2] is estimated as 0.6/cm² sec over the earth's surface. With the approximation that the tritium production cross section is the same for each primary nucleon, the number of tritons produced in the first interaction with the atmosphere is 0.05/cm² sec.

Additional tritium will be produced by high energy secondaries and by evaporation neutrons. The tritium produced by high-energy secondaries is estimated to be about 0.05/cm² sec on the basis of the attenuation of tritium with depth in area targets and from the attenuation of star-production[3] in the atmosphere. The natural tritium production by evaporation neutrons in the atmosphere has been estimated to be about 0.1/cm² sec.[4] This totals approximately 0.2/cm² sec and may be compared with the value of (0.12±0.04)/cm² sec obtained from the tritium content of natural waters.[5]

II. EXPERIMENTAL DISCUSSION

A. Targets

The target materials for oxygen and nitrogen were water (actually 0.1N HNO_3 for passivity with aluminum) and liquid ammonia. These were contained in aluminum cylinders 4.4 cm in diameter, 6.3 and 10.1 cm in length, and 0.12 cm wall thickness; and were irradiated in the internal beam of the cosmotron. Three cylinders in series (total length 22.7 cm) were used with the water; two (total length 16.4 cm) with the liquid ammonia.

B. Tritium Assay

The water cylinders were opened in an atmosphere of carrier hydrogen, which was then pumped off in such a manner that the hydrogen bubbled through the water. This hydrogen was then purified through a palladium thimble, and counted in a 2.4-liter counter previously described.[4] The counting was done in a low-level counting facility[6] which reduced the background to 30 counts/min. Table I gives the tritium results for this hydrogen. About 10 percent of the tritium produced in the water cylinders was in the molecular hydrogen.

The water from the cylinders was distilled and converted to hydrogen over magnesium at 600°C. The tritium activity was then determined by visual counting of the characteristic tritium tracks in a hydrogen-filled diffusion chamber, as previously described.[7] Table I also gives the results for the tritium produced as water.

The aluminum containers were equipped with pressure fittings and filled with liquid ammonia for the NH_3 run. The irradiated ammonia was expanded directly into the Mg furnace at 600°C where it was converted to Mg_3N_2 and H_2. The exit gas passed

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[1] *Progress in Cosmic-Ray Physics*, edited J. G. Wilson (North Holland Publishing Company, Amsterdam, 1952).
[2] From the data of Winkler, Stix, Dwight, and Sabin, Phys. Rev. **79**, 656 (1950).
[3] B. Rossi, Revs. Modern Phys. **91**, 922 (1953).
[4] E. L. Fireman, Phys. Rev. **91**, 922 (1953).
[5] S. Kaufman and W. F. Libby, Phys. Rev. **93**, 1337 (1954).
[6] The authors wish to thank Dr. Raymond Davis for the use of his low-level counting facility.
[7] E. L. Fireman and D. Schwarzer, Phys. Rev. **94**, 386 (1954).

TABLE I. Summary of experimental data.

Target cylinder	Analyzed sample	Measured by[a]	Counts/min	(T/H) $\times 10^{15}$	$\times(10^{-10})$ Number of tritons	$\times(10^{-12})$ Proton through target	Tritium cross section σ(mb)	Total target contents	Neutrons from contents	Neutron cross section σ(mb)
Front H_2O 75gH_2O	330 cc H_2 14 g H_2O	G.C. D.C.	... 19.5±0.9	158±15 5.6±0.3	0.28 2.8	4.9	31			
Middle H_2O 100 g H_2O	270 cc H_2 14 g H_2O	G.C. D.C.	... 17.9±0.8	287±15 5.2±0.2	0.42 3.5	4.1	35	250 g H_2O	(1.6±0.6)$\times 10^{12}$	650±250
Rear H_2O 75 g H_2O	310 cc H_2 14 g H_2O	G.C. D.C.	... 13.8±0.7	138±15 3.9±0.2	0.23 2.0	3.3	32			
Front NH_3 82 g NH_3	9.0 g NH_3	D.C.	16.8±0.8	4.8±0.2	4.1	6.4	29	133 g NH_3	(2.1±0.8)$\times 10^{12}$	1000±400
Rear NH_3 51 g NH_3	9.0 g NH_3	D.C.	13.3±0.7	3.8±0.2	2.6	5.6	27			
Calibration H_2O	14 g H_2O	D.C.	17.5±0.8	5.0						

[a] G.C. = Geiger Counter; D.C. = Diffusion Chamber.

through a water column and cold trap before going into the diffusion chamber where it was counted. The chemical conversion was shown to be more than 99 percent complete by titrating the water column and by passing a sample of the chamber gas through palladium. The tritium activity is given in Table I.

C. Neutron Detection

The neutrons from the target were detected and measured by the activation of a series of indium foils imbedded in a cadmium-covered paraffin block. The In^{116m} yield as a function of paraffin depth is a measure of the neutron intensity and average energy. The paraffin was 12 in.×12 in.×18 in. with a 12 in.×12 in. side facing the target. The detector was 90° to the beam direction for the water run, and at 60°, 90°, and 120° for the ammonia run. The detector arrangements were calibrated by standard Ra-Be sources at the cosmotron positions and in a large empty room.

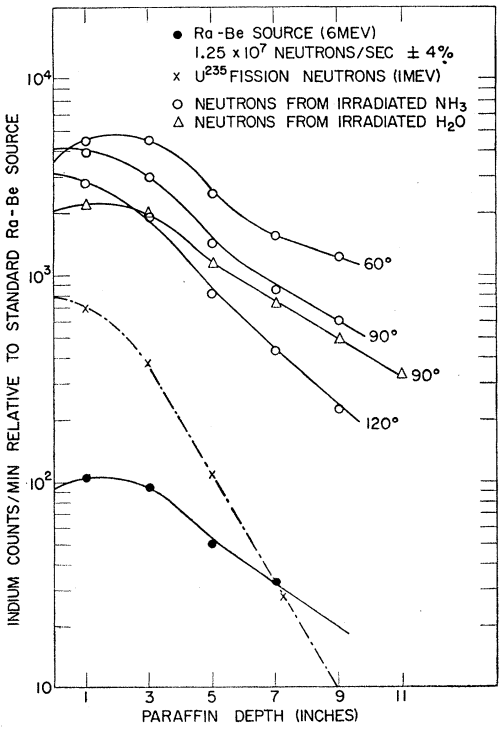

FIG. 1. Indium activation in Cd-covered paraffin.

Figure 1 gives the In activation results for the water, ammonia, a Ra-Be calibration source, and a U^{235} fission neutron calibration source. The In activity falls much more rapidly with depth for the U^{235} neutrons (1 Mev) than for the Ra-Be neutrons (6 Mev). The neutrons at 90° both for the water and ammonia are similar to the Ra-Be neutrons. The neutrons at 60° to the beam direction are of slightly higher average energy and the neutrons at 120° are of slightly lower energy than the Ra-Be neutrons. The NH_3 curves show the neutrons favor the forward direction. The angular distribution is consistent with neutrons being isotropic in a reference frame moving with a velocity of about $\frac{1}{3}$ that of the emitted neutrons.

The detected neutrons are produced not only in the water and liquid ammonia but also in the aluminum containers and by secondary reactions in material in the vicinity of the target. On the basis of an irradiation with empty containers and consideration of geometry and weight, the additional neutrons were estimated to be (35±15) percent of the total. The uncertainty in this correction and in the angular distribution of the neutrons is reflected in the large error assigned to the neutron yields which were (2.1±0.8)$\times 10^{12}$ for the nitrogen and (1.6±0.6)$\times 10^{12}$ for the oxygen. The resulting neutron cross sections are 1000±400 mb for nitrogen and 650±250 for oxygen (see Table I).

D. Proton Monitors

The proton flux was measured by Na^{24} production in 0.003-in. aluminum monitor foils inserted between

each cylinder as well as front and back. For the NH_3 run, 0.001-in. gold foils were also used with each aluminum foil. The cylinders were equipped with a $\frac{1}{8}$-in. lip of Lucite projecting $\frac{1}{4}$ in. from the target toward the beam, which intercepted the beam first and scattered it more or less uniformly over the circular cross section of the aluminum containers on the next traversal.

The average number of protons through each cylinder for the NH_3 run was calculated from the average of the gold foil activity at the front and rear of each section. Because the aluminum foils are contaminated to some extent by Na^{24} from the (n,α) reaction, the proton intensity for the water run was calculated by correcting the aluminum foil monitors for contamination. The correction factor, 0.89, was that calculated from the ratio of gold to aluminum activities in the NH_3 run. The average number of protons through each cylinder is given in Table I.

The monitors detect the reactions $Al(p,3pn)Na^{24}$ and $Au(p,15p34n)Tb^{149}$. All cross sections have been calculated on the basis of 9.0 mb for the former reaction with 2.2-Bev protons[8]; the quoted errors do not include the error in this measurement, but represent the error in the cross sections relative to it. The alpha-emitting Tb^{149} is the only activity detected in the gold foil with an alpha-counter after the first two hours. This reaction has a threshold of 0.6 Bev and a cross section of 1.3 mb for 2.2-Bev protons.[9]

E. Secondary Reactions

The use of thick targets, while greatly increasing neutron production and eliminating recoil loss from the target, introduces the possibility of tritium production by secondary reaction. The decrease of tritium activity with target depth parallels the decrease in proton intensity with target depth, and the expected decrease on the basis of geometric cross sections (Table I and Fig. 2), indicating that triton production by high-energy secondaries is negligible, since the secondaries would result in an excess of tritium in the rear cylinder.

The possibility of contamination by low-energy secondaries produced in evaporation processes can be eliminated by comparison of the activities observed on gold and aluminum foil. The most troublesome low-energy secondary reaction for producing tritium is the (n,T) reaction on N^{14}, which has a threshold of 4.4 Mev and a cross section of 11 ± 2 mb for fission neutrons above this energy.[4] Since the (n,α) reaction on Al producing Na^{24} has a threshold of 2.7 Mev and a cross section of 110 ± 20 mb for 14-Mev neutrons,[10] it is much more sensitive to neutron secondaries than the $N(n,T)$ reaction. The contamination of the Al foil by this secondary reaction can be calculated from the comparative activities of the gold and aluminum foils, and amounts to less than 20 percent, indicating that secondary neutrons produce only about 1 percent of the tritium in the NH_3 run. The other secondary neutron and proton reactions have much higher thresholds and should be much less significant.

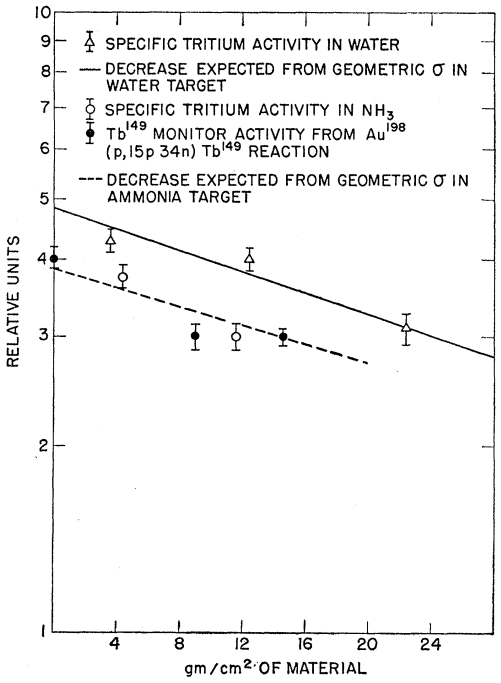

FIG. 2. Tritium and monitor activities *vs* target depth.

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[9] R. B. Duffield and N. Sugarman, Phys. Rev. **94**, 776 (1954).
[10] *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040, Supplement 2 (Technical Information Division, Department of Commerce, Washington, D. C., 1953). Data from Los Alamos and Chalk River.