Tritium Production by 6-Bev Protons

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Tritium-formation cross sections for 6-Bev protons on various targets between C and Pb are reported. No significant variations in tritium-production cross sections are observed between 2 Bev and 6 Bev. The problem of tritium production in nature is discussed briefly.

INTRODUCTION

IN addition to furthering knowledge of high-energy nuclear reactions which eject light fragments, tritium formation by protons of cosmic-ray energies may shed light upon "natural" tritium abundance on the earth as well as upon tritium production—and "age"—for meteorites. In a previous paper[1] the author reported the cross sections for tritium formation in a number of nuclei between carbon and lead induced by 450-Mev and 2-Bev protons. Since cosmic-ray protons extend to still higher energies, it was desirable to extend the accelerator studies; this paper includes results obtained with 6-Bev protons.

Previous results which have been modified and pertinent results of others have been included in the discussion.

EXPERIMENTAL DETAILS

Targets

Targets, with areas of about 2 cm×2 cm and thicknesses varying from 0.41 g/cm² to 4.8 g/cm², included pure metals as well as organic materials. Included with the target material itself were monitoring foils consisting of 0.003 in. or 0.005 in. Al and usually 1 mg/cm² (∼0.00002 in.) or 0.002 in. Au. In most cases the ends of the targets were milled flat at the beam end.

Chemistry

The procedures for the quantitative determination of tritium were essentially the same as previously reported.[1] That is, metal targets (Al, Fe, Sn, Pb) were heated to 950°C in an atmosphere of nonradioactive hydrogen in order to allow exchange with the contained tritium; after cooling, the hydrogen was pumped through palladium (to eliminate other radioactive gases) and into a Geiger-Müller counter. Two modifications were made in the previous procedure: (1) a quartz tube was used for the extraction in place of the Alundum crucible and porcelain tube, (2) 66 cm-Hg of *Q*-gas[2] was used as the counting gas in place of 1.5 cm-Hg of ethylene plus 3.5 cm-Hg of argon.

As a check on the extraction procedure for iron (diffusion at 950°C in the solid), a portion of sample No. 53 was analyzed for residual tritium by induction-melting in the presence of carrier hydrogen. No additional activity was observed.

In order to measure tritium-production cross sections in nitrogen and oxygen, organic targets were used. Thus, it was also necessary to measure the production from carbon independently. Lucite ($C_5O_2H_8$) and melamine ($C_3N_6H_6$)[3] were substituted for oxalic acid ($C_2O_4H_2$) and hexamethylene tetramine ($C_6N_4H_{12}$) used previously. Polyethylene (CH_2) was used for the independent carbon target. As before, the organic targets were burned quantitatively in dry oxygen to form water which was then quantitatively reduced (over zinc at 450°C) to hydrogen which was counted in a Geiger-Müller counter.

Monitors

Proton counting was accomplished by means of the monitoring activities: F^{18} and Na^{24} from Al. Tritium cross sections at 0.120 Bev and at 0.450 Bev were based upon $Al(p,3pn)Na^{24}$ cross sections of 10 mb[4] and 10.8 mb,[5] respectively. The previous results at 2 Bev were based upon an $Al(p,3pn)Na^{24}$ cross section of 9.0 mb. This number has been revised in the meantime to 10.8 mb[6,7]; the 2-Bev results have been revised accordingly (see Table I).

Monitoring at 6 Bev was based upon F^{18} from the aluminum foils. The cross section employed was 7.4 mb.[8] The $Al(p,\text{spall})F^{18}$ was preferred to the $Al(p,3pn)$-Na^{24} monitor because low-energy secondary neutrons may induce the reaction, $Al(n,\alpha)Na^{24}$.[9]

[1] Currie, Libby, and Wolfgang, Phys. Rev. **101**, 1557 (1956).
[2] Available from Nuclear-Chicago Corporation; consists of 98.7% helium plus 1.3% butane. Experiments showed the efficiency for counting tritium was the same as with the ethylene-argon mixture. The *Q*-gas was much more convenient since it could be used directly from the cylinder without purification and mixing.
[3] The author is grateful to Dr. R. L. Wolfgang for suggesting melamine as a target material.
[4] Hicks, Stevensen, and Nervik, Phys. Rev. **102**, 139 (1956).
[5] L. Marquez, Phys. Rev. **86**, 405 (1952).
[6] R. L. Wolfgang and G. Friedlander, Phys. Rev. **96**, 190 (1954), and **98**, 1871 (1955).
[7] Cumming, Swartz, and Friedlander, Bull. Am. Phys. Soc. Ser. II, **1**, 225 (1956).
[8] A. Caretto *et al.*, Phys. Rev. **110**, 1130 (1958).
[9] The F^{18} reaction may be even more accurate for calculating the tritium cross sections, since it should be affected by secondary neutrons and protons somewhat similarly to the tritium-producing reactions (see Fig. 1).

DISCUSSION

Tritium-Formation Cross Sections

The results of the recent cross-section measurements at 6 Bev as well as an older measurement at 0.120 Bev and the previous results at 2 Bev (corrected for the change in monitor cross section) are recorded in Table I. In Table II other available tritium-production cross sections in iron targets (and one copper target) have been listed. Finally, from the data in Tables I and II and from the data at 450 Mev (Table II of reference 1), partial excitation functions for the tritium formation by protons have been plotted (Fig. 1). Only C, N, O, Al, Fe, and Pb have been included in the graph since other targets had been observed at fewer than three bombarding energies.

TABLE I. Tritium-formation cross sections.

Thickness (g/cm²)	Number	Target	Cross section (mb)	Proton energy (Bev)
2.5	44	Al	8.0	0.120
		Polystyrene (C)	17	2.05[a]
		Hexamethylene tetramine (N)	30	
		Oxalic acid (O)	36	
		Mg	36	
		Al	44	
		Fe	64	
		Ni	90	
		Ag	160	
		Pb	610	
0.75	51	Polyethylene (C)	18 } 19[b]	5.7
0.45	56		20 }	6.2
1.3	54	Melamine (N)	35	
1.3	54	Lucite (O)	38	
0.41	46	Al	50	5.7
4.8	53	Fe	57	6.2
2.5	55	Sn	440	
2.6	52	Pb	450 } 480[b]	
0.45	56		510[c] }	

[a] Previous results (from Table II of reference 1) corrected for the subsequent change in monitor cross section.
[b] Arithmetic average.
[c] Result of a 25% correction for loss of tritium from the thin Pb target due to recoil (assuming an average recoil energy of 10 Mev).

As is apparent from Fig. 1, there is little or no variation in the tritium production cross sections for proton energies between 2 Bev and 6 Bev.[10] The slight variations which do appear are probably smaller than the experimental errors. No errors have been indicated explicitly, but the over-all errors are estimated to be about $\pm 20\%$. Statistical counting errors in no case exceeded $\pm 5\%$; uncertainties in target analysis (tritium extraction and purification) are approximately $\pm 5\%$; monitor cross sections are probably not to be trusted to better than $\pm 10\%$.

[10] A least-squares analysis of the variation of the (p,t) cross section with target mass number in the 2 Bev to 6 Bev region resulted in: $\sigma = 1.40A^{1.07}$ mb. Although there is no obvious justification for such an analytic function, the analysis may be useful as an indication of the importance of intranuclear secondary reactions.

TABLE II. Tritium cross sections in iron by other observers.

Thickness (g/cm²)	Cross section (mb)	Proton energy (Bev)
...	4[a]	0.050
50	7.2[b]	0.160
...	6.6[a]	0.177
183	65[b]	1.0
36	62[c]	2.2
183	88[b]	3.0
0.212	202[d]	5.7
163	110[b]	6.2

[a] K. Goebel, Nuclear Sci. Abstr. **12**, 6961 (1958).
[b] E. L. Fireman and J. Zähringer, Phys. Rev. **107**, 1695 (1957). (These cross sections refer to the proton-entry side of the target.)
[c] E. L. Fireman, Phys. Rev. **97**, 1303 (1955).
[d] Copper target (rather than iron): D. Barr, University of California Radiation Laboratory Report UCRL-3793, May, 1957 (unpublished), p. 58.

One apparent discrepancy seems worthy of mention. Namely, the tritium-production cross sections in Fe reported by Fireman (Table II) become somewhat larger than those reported by us at higher bombarding energies. The author believes that this may be due partially to the contribution of secondary particles to tritium formation in Fireman's relatively thick targets. (This contribution, of course, should be included for total tritium production in meteorites.) Such secondary reactions could take place even in the initial depths of the thick targets since the threshold for the reaction $Fe^{56}(n,t)Mn^{54}$ is but 12 Mev.

The cross section determined by Barr for 5.7-Bev protons on copper is still larger. Since the tritium-formation cross section does not appear to be increasing rapidly with mass number, one would expect the cross section in copper to be roughly comparable to that in iron. Also, since Barr's target was quite thin, secondary production of tritium should not be serious. The only conclusion that may be drawn is that additional studies

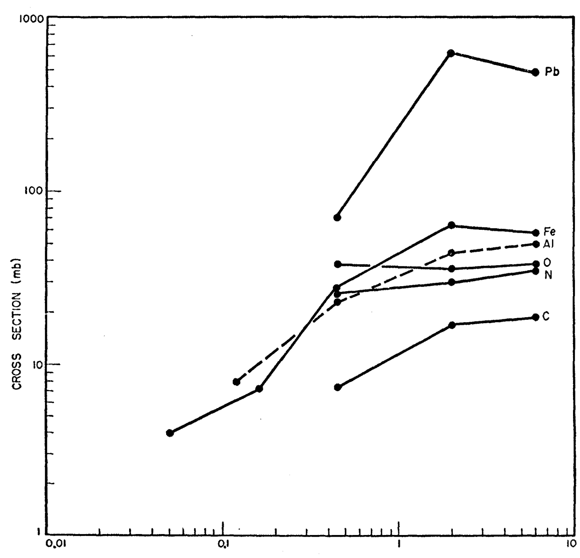

FIG. 1. Partial excitation functions for tritium formation by proton bombardment.

of both iron and copper targets are required to resolve these differences.

Natural Production of Tritium

Two problems involving the formation of tritium in high-energy nuclear reactions are of particular interest: the formation by cosmic rays in meteorites and in the atmosphere. Since the tritium-production cross sections are constant above 0.450 Bev for nitrogen and oxygen and constant above 2 Bev for iron, the discussion which appeared in reference 1 is essentially unaltered. Recent measurements of tritium in nature, however, have considerably altered the agreement between the observed tritium production in the atmosphere and that predicted on the basis of the cosmic-ray reactions.[11] The predicted tritium flux remains at about 0.14 t/cm^2-sec, whereas the observed flux may be $2.0\pm(50\%)$ t/cm^2-sec.[12] Possible sources for the discrepancy include: a higher flux of incident cosmic rays, additional tritium-producing reactions, influx of tritons with the cosmic rays, more serious secondary production of tritium in the atmosphere, and unsuspected loss of tritium from the targets.

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[11] H. V. Buttlar and W. F. Libby, J. Inorg. Nuclear Chem. **1**, 75 (1955); F. Begeman, Air Force Office of Scientific Research Report AFOSR-TR-58-41, December 31, 1957 (unpublished).

[12] F. Begeman and W. F. Libby, Geochim. et Cosmochim. Acta **12**, 277 (1957); H. Craig, Phys. Rev. **105**, 1125 (1957).

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Modified Analysis of Nucleon-Nucleon Scattering. I. Theory and p-p Scattering at 310 Mev*

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A modified method of analyzing nucleon-nucleon scattering is discussed and applied to proton-proton scattering experiments at 310 Mev. The modified scheme is based on an explicit inclusion in all higher angular-momentum states of the terms contributed by the one-pion exchange process. This procedure is suggested by Chew's conjecture that the singularities of the scattering amplitude in the $\cos\theta$ plane (θ being the scattering angle in the center-of-mass system) that are closest to the physical region are due to the one-pion exchange process and are given by the Born approximation. Or, alternatively, in terms of ranges, the one-pion exchange contribution has the longest range of the forces contributing to the nucleon-nucleon interaction and hence should be primarily responsible for the contributions to the scattering amplitude in the high angular-momentum states. Since the only parameter in the Born approximation is the pion-nucleon coupling constant, the modified scheme can also provide a determination of this coupling constant. The application of the modified scheme to p-p scattering at 310 Mev indicates that the first two of the five best solutions of the conventional phase-shift analysis are more satisfactory than the others for two reasons. Firstly, their goodness-of-fit parameters improve markedly when the higher angular-momentum contributions are added, whereas those of the others remain essentially unchanged. Secondly, as a function of the coupling constant, the goodness-of-fit parameters of the first two solutions show minima close to the accepted value of the coupling constant.

1. INTRODUCTION

IT has been suggested by one of us[1] (M.J.M.) that the conventional phase shift analysis of nucleon-nucleon scattering experiments be replaced by a modified scheme in which the contribution due to the exchange of one pion is explicitly included in the scattering amplitude. This approach was motivated by some conjectures of Chew[2] on the behavior of the scattering amplitudes in the nonphysical region of the complex $\cos\theta$ plane (θ being the scattering angle in the barycentric system). Chew argues that the singularities will be restricted to the real axis and that those closest to the physical region, $-1\leq\cos\theta\leq1$, will be two symmetrically situated poles associated with contributions to the scattering amplitude of one-pion inter-

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[1] Michael J. Moravcsik, University of California Radiation Laboratory Report UCRL-5317-T, August, 1958 (unpublished).

[2] Geoffrey F. Chew, Phys. Rev. **112**, 1380 (1958).