High-Energy Proton Production of ^{3}H, ^{3}He, and ^{4}He in Light Targets*

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At 0.6 and 3 GeV, cross sections for the production of ^{3}H, ^{3}He, and ^{4}He in C and Si were determined, and for the production of ^{3}H and ^{3}He in oxygen. At 33 GeV only ^{3}H:^{3}He:^{4}He production ratios in C and Si could be established due to a fault in the monitoring. The ^{3}H/^{3}He yield of 0.85± 0.10 embraces the three target materials throughout the energy range investigated. On the other hand, the ^{4}He/^{3}He ratios varied with energy and with target material by up to a factor of 2. Most of the previous ^{3}H cross-section determinations in carbon are shown to be low by a factor of 2 or 3, apparently because of tritium diffusion from the thermoplastic materials used in the earlier studies. The majority of ^{3}H cross sections at 600 MeV in targets from helium to bismuth fit the equation $\sigma(A) = 30\exp(A/100)$, which should be beneficial in predicting unknown cross sections. A trend in the ^{4}He cross sections as a function of target mass was discernable, but the scatter of the data does not permit making a more detailed examination; no ^{3}He trend as a function of A is observed.

INTRODUCTION

The ^{3}H-^{3}He method of calculating cosmic-ray exposure ages of stone meteorites has been the most widely applied. Yet these calculations are hampered by the fact that tritium and ^{3}He cross sections are either poorly known or not known at all for the major elements comprising stone meteorites. In the past, the production ratio ^{3}H/^{3}He = 1, determined by high-energy protons on iron, was assumed to hold for the entire meteorite, although the iron group (Fe, Co, Ni) made up only about 10–25% of the atomic elemental composition. Oxygen (the major component), silicon, and magnesium generally account for about 80–90% of the meteorite and, hence, are the major contributors to ^{3}H and ^{3}He production.

Tritium and ^{3}He yields in silicon and magnesium by high-energy protons were measured by Goebel, Schultes, and Zähringer (GSZ).[1] On the basis of certain systematic trends in these and other cross sections, Kruger and Heymann[2] suggested that the spallation ^{3}H/^{3}He yield in oxygen could be as low as 0.5. If this were indeed the case, the assumed meteoritic ^{3}H/^{3}He spallation yield would be substantially lower than the adopted value, and radiation ages would have to be adjusted downward by as much as 30%.

In order to settle this issue more firmly, one of our objectives was to determine the ^{3}He cross sections and remeasure the ^{3}H cross sections in oxygen as a function of proton bombarding energy. Three energies representative of the crest of the galactic cosmic-ray energy spectrum, 0.6, 3.0, and 33 GeV, were chosen. We considered it beneficial to determine ^{3}H and ^{3}He cross sections in silicon also, since they had previously been measured at one energy only, and the experimenters had raised doubts about the tritium value. The interpretation of these results, as far as meteorite exposure ages are concerned, will be made in a subsequent publication.

From the point of view of nuclear structure, ^{12}C and ^{16}O nuclei are particularly interesting and potentially informative. Recently, considerable experimental evidence has led to the suggestion that these nuclei are composed of, respectively, three and four loosely joined α clusters rather than individual nucleons.[3] Attempts have been made to include α clustering in nuclear models, especially within the framework of the shell model.[4] There also exists the possibility of time-variant intranuclear subclustering of ^{3}H and ^{3}He particles. Investigation of the interaction between high-energy protons and light nuclei can yield information on the probability of a definite type of nuclear clustering and on the physical nature of such clusters. With this objective in mind, we have also included carbon as a target.

The irradiations were performed at the following facilities: 33 GeV at Brookhaven National Laboratory (BNL), 3 GeV at the Princeton-Pennsylvania Accelerator (PPA), and 0.6 GeV at the Space Radiation Effects Laboratory (SREL).

EXPERIMENTAL PROCEDURES

A. Targets

Elemental graphite and silicon were used for determining the tritium and helium-isotope production rates in C and Si. Water and Delrin[5] (a polymer of composition CH_2O) were chosen for the tritium determinations in oxygen, and quartz (SiO_2) was used for the helium-isotope measurements in

oxygen. Individual target disk thicknesses ranged from 0.25 to 0.37 g/cm^2 except for the 0.11-g/cm^2-thick Delrin targets; the liquid H_2O targets were 2.0 g/cm^2 thick.

The graphite, silicon, and quartz targets were precleaned by heating to 1000°C under vacuum until low pressures were achieved. The Delrin targets were pumped down under moderate heating.

With the exception of water, all of the targets were irradiated under vacuum. Hydrogen gas at atmospheric pressure was placed above the H_2O targets. Separate target chambers were constructed for each of the irradiation sites, and each chamber contained from three to nine target disks of the same material. The total target thickness per chamber was in the neighborhood of 1 g/cm^2.

B. Proton Irradiations and Monitoring

The 33-GeV irradiation at BNL was only partially completed because of machine failure and just the graphite and silicon targets were bombarded. Unfortunately for this irradiation also, the proton beam was not well aligned through the target chambers and the monitoring was not reliable.

Two separate bombardments at 3 GeV and one at 600 MeV were performed at the PPA and SREL facilities, respectively. The beam spot locations were continually monitored by television via scintillators to assure good beam alignment. The proton currents were stable at approximately 0.03 μA at both facilities, and individual bombardments lasted between 40 and 70 min apiece. The maximum target thickness in the beam at any given time was 2.0 g/cm^2.

The proton beams were monitored by irradiating aluminum foils of 0.5-mil thickness and measuring the ^{24}Na and ^{18}F activities. The monitor cross sections adopted are those given by Cumming.[6] Three foils were taped over both the beam-entrance and beam-exit windows of each chamber, and the middle foils were used as the actival monitors. The ^{24}Na and ^{18}F activities were assayed in well-type NaI(Tl) scintillation counters at BNL following the method developed by Cumming.[7] For the PPA and SREL irradiations, counting commenced roughly 8 and 16 h, respectively, after the first targets were bombarded. In the case of ^{24}Na the counting statistics were excellent, while for ^{18}F they were good to poor, depending upon the elapsed time.

There was a twofold purpose in monitoring both upstream and downstream foils by radionuclides: (1) to provide a check on beam positioning and (2) to determine the effect of secondary particles. There was good agreement between the ^{24}Na activities in upstream and downstream foils in the individual targets from both the PPA and SREL irradiations. This indicates that the beams were well centered and passed entirely through the beam windows. For the 3-GeV irradiations the ^{18}F/^{24}Na ratios were nearly identical in upstream and downstream foils, indicating that secondary particles were of negligible effect.

C. Tritium Analysis

Tritium in the irradiated targets and background samples was converted to water and collected in the extraction line shown in Fig. 1. Water had to be added as a carrier in each extraction since the amount of tritium per irradiated target was less

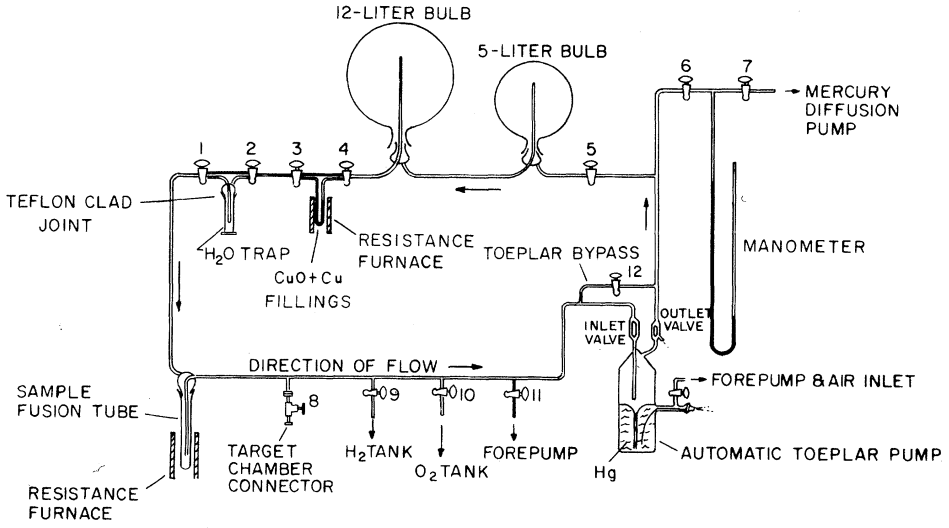

FIG. 1. Schematic diagram of the tritium extraction line.

than 10^{-6} cm^3 STP. The extraction procedure varied somewhat according to the sample material.

Graphite

The graphite targets were burned to carbon dioxide in an oxygen atmosphere and in the process tritium in the targets was oxidized to water. A sample would be placed in the quartz fusion tube and pure oxygen admitted to the line. While oxygen and water vapor were circulated through the system by action of the Toepler pump, the sample was heated to 900°C by a resistance furnace until it disappeared. The water was then collected in the H_2O trap and pipetted into counting vials.

The completeness of the tritium oxidation to water was checked several times in the following manner. Half of a pulverized irradiated target was burned out and collected by the above-mentioned procedure. The remaining half was also oxidized by the same method but, before collecting, about 0.5-cm Hg of hydrogen gas was added to the line, circulated for several hours, and then oxidized to H_2O in the CuO furnace at 700°C. Any target tritium not oxidized in the graphite-burning process should have exchanged with the hydrogen gas to form HT and been oxidized to HTO in the CuO furnace. No detectable difference in the activities of the portions was observed.

Delrin

Delrin melts at 170°C and is oxidized to CO_2 and H_2O in the presence of oxygen at temperatures above 300°C. A simple metal chamber was constructed for this purpose. After insertion of the sample, sufficient oxygen was admitted and the chamber was shut by a high-temperature vacuum valve. The chamber was then placed for several hours into a laboratory oven preheated to 350°C. Following combustion the chamber was attached to the extraction line and the water collected in the trap.

Silicon

Because of silicon's melting point of 1430°C, these targets could not be directly melted by the resistance furnace in the sample fusion tube. However, aluminum, among others, alloys with silicon at lower temperatures by virtue of its lower melting point.

The silicon targets were pulverized and approximately 1 g of target was placed into a ceramic crucible. On this was placed about 2 g of pure aluminum foil. This ratio of aluminum to silicon provides for an alloy-melt temperature of 850–900°C. The crucible was inserted into the fusion tube and partially capped with a ceramic lid. About 0.5-cm Hg of hydrogen gas was admitted to the line and circulated for several hours while the crucible temperature was maintained at 1000°C. Afterwards the hydrogen was converted to H_2O in the CuO furnace.

H_2O

Fireman and Rowland[8] discovered that about 10% of the tritium recovered from their irradiated water targets was in the form of HT gas. Accordingly, we also collected the gas above the water, oxidized it in the CuO furnace, and counted it separately. Afterwards the irradiated water samples were pipetted directly into counting vials.

Approximately 3 ml of tritiated-water sample was collected from each extraction. From this, four $\frac{1}{2}$-ml aliquots of sample were pipetted into separate low-activity counting vials after discarding the first pipetteful. To each vial was added 10 ml of "Bray's mixture" cocktail,[9] a solvent widely used for the counting of aqueous solutions. The cocktail mixture was added to the samples just prior to counting in order to guard against deterioration.

The tritium activities were measured in a Nuclear Chicago model-724 liquid scintillation counter with counter background of about 20 counts per minute (cpm) and tritium counting efficiency of about 13%. Sample activities normally fell in the range 200–500 cpm, and counting statistics of better than ±1% were achieved. Calibration standards in the same range of activity were prepared from a commercially-obtained tritiated-water sample.

D. Helium Analysis

The mass spectrometer used in this study was a Nuclide Corporation model 4.5-60 RSS with electron-bombardment ion source, a Reynolds-type instrument[10] with high sensitivity for noble-gas analysis. The ion current is measured by a nine-stage electron multiplier operated in conjunction with a vibrating-reed electrometer.

The helium extraction line, shown in Fig. 2, was constructed entirely of stainless steel, except for the supplementary quartz furnace used to heat samples above 1200°C. The purpose of the all metal construction was to suppress atmospheric-helium diffusion into the line, which occurs in all glass systems and is a sensitive function of temperature.

Sample heating was provided by a 15-kW rf-induction heater. The sample furnace temperature was measured by a thermocouple inserted into a cavity in the bottom of the furnace and also by optical pyrometer readings of the furnace surface.

Targets placed into the stainless-steel furnace

were slowly heated to 1200°C and maintained at this temperature for 30 min. The gas was exposed initially only to a liquid-nitrogen-cooled trap and later to a similarly cooled charcoal trap and a getter (36% Ti, 64% Zr) heated to 800°C. The getter was then slowly cooled to room temperature in the presence of the gas.

Neither graphite, silicon, nor quartz will melt at 1200°C. Two methods were used to determine whether all of the helium was released from the targets at this temperature. First, several targets of each type were stepwise heated in the temperature range 800–1200°C. For graphite about 88% of both helium isotopes was released at 800°C, 10% more in heating from 800 to 1000°C, 1–2% in going from 1000 to 1100°C, and no additional helium was released from 1100 to 1200°C. For the silicon targets, about 60% of the helium diffused out at 800°C and the remaining 40% in elevating the temperature to 1000°C. Beyond that, no additional helium was released. As far as the quartz targets were concerned, no ^{3}He at all was detected upon heating to 1200°C.

The second method consisted of further heating to 1600°C in the quartz furnace those targets which had already been heated to 1200°C. (The silicon targets melted below this temperature.) No additional helium was observed from any of the targets at 1600°C.

Before any of the irradiated targets themsleves were measured, the target chambers were connected to the extraction line and the gases in them released to the mass spectrometer. ^{4}He was present in all of the chambers but, due to the absence of ^{3}He (except in the case of quartz), was resolved to be atmospheric helium which had diffused into the evacuated chambers.

RESULTS

Approximately the anticipated amounts of irradiation-produced ^{3}He were found in the gas phase of the quartz-target chambers and none in the targets themselves. The ^{3}He cross sections in oxygen at 0.6 and 3 GeV are calculated on the assumption that this gas-phase ^{3}He represents all of the isotope produced in the irradiations. The cross sections thus derived are comparable to the ^{3}He cross sections in carbon and silicon and consistent with the ^{3}H cross sections in oxygen at the same energies. The ^{4}He detected in the quartz targets is believed to be atmospheric in origin; that is, ^{4}He which permeated the quartz after the bombardment during the time the targets were exposed to air.

The tritium and helium cross sections obtained in this investigation are given in Table I, together with the production ratios ^{3}H/^{3}He, ^{4}He/^{3}He, and ^{4}He/(^{3}H + ^{3}He). Because of the uncertainty in the 33-GeV beam-monitoring measurements, at this energy we show only the production ratios.

The cross sections of Table I are average values from several measurements. With the exception of H_2O and SiO_2, at least three targets of the same material were exposed at each energy. Variations in the tritium or helium contents of individual targets from the same exposure were small, generally less than 10%. There was no uniform trend of helium or tritium content in the targets as a function of depth.

For carbon and oxygen targets, the ^{3}H, ^{3}He, and ^{4}He cross sections increase by roughly 10% as the bombarding energy is raised from 0.6 to 3 GeV. Judging from the production ratios at 33 GeV in carbon, it appears as though there should be little or no change in the cross sections between 3 and

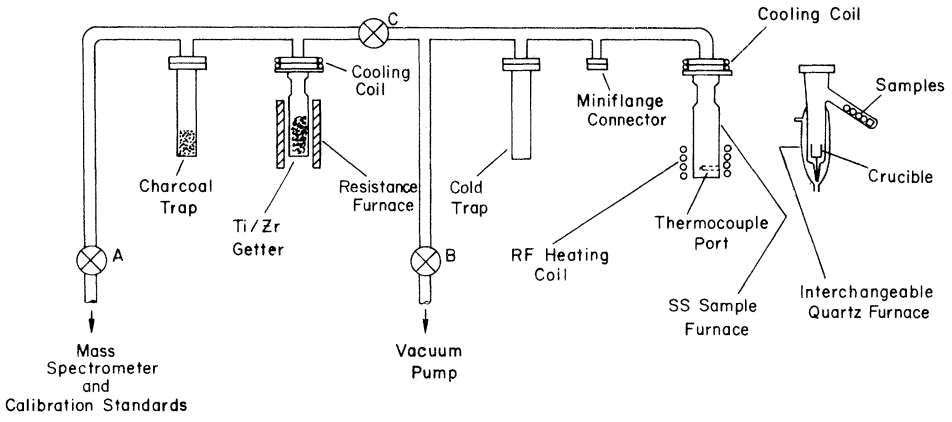

FIG. 2. Schematic diagram of the helium extraction line.

33 GeV in carbon and oxygen.

For silicon, there is a substantial increase in the cross sections at 3 GeV over those measured at 0.6 GeV. Whereas it appears that the ^{4}He cross section may be leveling off at about 3 GeV, according to the production ratios at 3 and 33 GeV, the ^{3}H and ^{3}He cross sections are still growing above 3 GeV. Mekhedov and Mekhedov[11] noted a similar increase for tritium production in aluminum from 1 to 30 GeV.

The most determinative factors in calculating ^{3}H-^{3}He meteorite exposure ages are the ^{3}H/^{3}He yields in oxygen and silicon. We find that this ratio is very nearly constant in the three target materials studied for incident protons of 0.6–33 GeV, and has an average value of about 0.85. This is in contrast to the estimated value of 0.5 in carbon and oxygen suggested by Kruger and Heymann,[2] but is closer to the value of 1.0 found in iron by earlier investigators.[12]

Results from the 3-GeV irradiation of Delrin (CH_2O) require a special interpretation. Three of the nine Delrin targets were analyzed for their tritium activity, and it was found to be abnormally low. According to the Delrin results alone, one would calculate tritium-production cross sections in both carbon and oxygen of about 12 mb. This is in contrast to the values of 35 and 36 mb obtained from graphite and H_2O targets, respectively, at the same energy. It is apparent that about $\frac{2}{3}$ of the tritium produced in the Delrin had disappeared. Recoil loss could not be a factor since the overall thickness of the Delrin targets was the same as for the graphite. The tritium extraction mechanisms were similar, with both Delrin and graphite being burned to CO_2 and H_2O at high temperature in the presence of pure oxygen. It is likely that the Delrin tritium loss took place during bombardment via diffusion from the warm targets. The Delrin did not melt during the irradiation, but appreciable heating was evidenced by discoloration; the creamy-white material was charred brown by the proton beam. In the presence of the organic target material and the radiation environment, the tritium may have formed a gaseous hydrocarbon which then diffused out of the targets.

ACCURACY OF MEASUREMENTS

The amounts of ^{3}H, ^{3}He, and ^{4}He found in a given target may be different from that produced in the primary interactions for three reasons: secondary production, diffusion loss, and recoil loss.

Secondary protons and neutrons produced in the targets may themselves interact to form ^{3}H, ^{3}He, and ^{4}He. Secondary production of tritium was investigated by Currie, Libby, and Wolfgang[13] by irradiating targets of varying thicknesses with 2.05-GeV protons. The authors found that no correction for secondary production was necessary for targets less than 2 g/cm^2 thick. Fireman and Rowland[8] found that secondary production of tritium by 2.2-GeV proton irradiation of a thick (22-g/cm^2) H_2O target was also negligible. While secondary ^{3}H, ^{3}He, and ^{4}He production is likely taking place to some extent even in target thicknesses of 1 g/cm^2, the effect probably makes little contribution in the targets of this investigation.

Helium diffused completely out of the quartz targets but no ^{3}He was found in any of the other target chambers. The latter is strong evidence against helium diffusion in all but the quartz targets. Diffusion loss of tritium from the Delrin targets seemingly accounts for their low values, and tritium was found in the gas phase above the H_2O targets; however, tritium diffusion from the remaining targets is much less likely. Diffusion loss of tritium from beryllium and aluminum targets was shown to be negligible in a controlled experiment by Wolfgang and Libby.[14] Conversely, Fechtig, Festag, and Schultes[15] observed substantial tritium

TABLE I. Measured tritium and helium cross sections and production ratios at proton-bombarding energies of 0.6, 3.0, and 33 GeV.

Target	Proton energy (GeV)	Cross section ^{4}He (mb)	^{3}He (mb)	^{3}H (mb)	^{3}H/^{3}He	^{4}He/^{3}He	^{4}He/(^{3}H+^{3}He)
C	0.6	202±24	39±5	32±4	0.82±0.8	5.2±0.5	2.8±0.3
	3.0	207±25	43±5	35±4	0.81±0.8	4.8±0.5	2.7±0.3
	33				0.85±0.9	4.7±0.5	2.6±0.3
O	0.6		38±5	34±4	0.89±0.9		
	3.0		40±5	36±4	0.90±0.9		
Si	0.6	331±40	42±5	36±4	0.86±0.9	7.9±0.8	4.2±0.4
	3.0	365±44	70±8	58±7	0.83±0.8	5.2±0.5	2.9±0.3
	33				0.81±0.8	4.4±0.4	2.4±0.3

diffusion from an iron meteorite at room temperature, whereas GSZ[1] noted that iron degassed in a vacuum prior to proton exposure retains tritium much better than ordinary iron. Except for Delrin and H_2O, all of the targets in this investigation were thoroughly degassed before irradiation. Although the evidence is ambiguous, it is likely that little tritium loss by diffusion occurred except in the cases already noted.

The problem of recoil loss of light products from thin target bombardments is complicated, and a proper treatment requires knowledge of the energy and angular distributions of the product nuclides. For the present we shall consider only the experimental evidence.

Currie, Libby, and Wolfgang[13] measured tritium cross section as a function of depth in targets from carbon to lead at proton energies of 0.450 and 2.05 GeV. They concluded that most of the tritons are emitted with energies below 15 MeV and that corrections for recoil loss in targets of 1 g/cm^2 are negligible. Kuznetsov[16] irradiated varying thicknesses of aluminum and lead targets with 660-MeV protons and found that for aluminum thicknesses between 135 and 500 mg/cm^2, the tritium yield is constant. The tritium yield from the lead targets indicated that the triton kinetic energy does not generally exceed 30–40 MeV. Zhdanov and Fedotov[17, 18] analyzed the energy and angular distributions of up to 72-MeV α particles emitted from carbon targets bombarded by 660-MeV protons. The mean α-particle energy is about 7 MeV (our estimate) and the distribution is notably peaked in the forward direction. Dudkin *et al.*[19] present semiempirical energy distributions of ^{3}H, ^{3}He, and ^{4}He particles from oxygen bombarded by 660-MeV protons, and we calculate mean energies of about 12, 10, and 8 MeV, respectively. The result of the investigations would indicate that, for bombarding energies below about 2 GeV, recoil losses from our targets are relatively unimportant. For greater energies, however, there is evidence that recoil losses may not be negligible. Sizable numbers of very high energetic ^{3}He and α particles from the 28-GeV proton bombardment of BeO was observed by Wang.[20] The energy and angular distributions of mass-3 and mass-4 nuclei produced in the interaction of 5.5-GeV protons with silver and uranium have been reported by a group from Berkeley.[21, 22] Although the targets in the latter investigation are much heavier than those of this paper, the general trends observed should be similar. The authors find that the cross sections for the production of very energetic ^{3}He nuclei is significantly greater than that for very energetic ^{4}He nuclei, and that the energy spectra for ^{3}H and ^{3}He are similar, although their specific energy losses will be quite different. The essential point is that recoil losses may be large at the higher bombarding energies, and that any recoil losses will specifically affect the ^{3}H/^{3}He and ^{3}He/^{4}He ratios. The monitoring error (using the two nuclides) was placed at ±6%. The tritium standard to which all tritium activities are compared had a quoted error of ±3%. Errors in the helium calibration system are known to be on the order of ±1% from previous work with this system. An additional error of ±10% is included to account for uncertainties in helium and tritium extractions, recoil and diffusion losses. It is possible that this figure is underestimated. The overall error then is ±12% for the cross sections and ±10% for the ratios, the latter being unaffected by monitoring errors.

COMPARISON WITH EARLIER MEASUREMENTS

Table II lists all published results of tritium, ^{3}He, and ^{4}He cross-section determinations in carbon, oxygen, and silicon for proton-bombardment energies above 100 MeV. In silicon, the helium cross sections are approximately 10–20% below our values, and the tritium cross section at 600 MeV is about one half of our result. The discrepancy in the helium results is not understood but it is systematic and may be due to differences in calibration techniques. GSZ[1] mention that incomplete tritium extraction might account for their low yield in silicon.

Our results of the tritium cross sections in oxygen are in good agreement with previous determinations. The only other ^{3}He cross-section determination in oxygen is that by Dudkin *et al.*[19] at 600 MeV, which the authors state is simply a rough estimate.

Zhdanov and Fedotov[17] studied the disintegration of carbon nuclei by 660-MeV protons. They recorded 0–72-MeV α-particle tracks from individual reactions in nuclear emulsions and arrive at a ^{4}He cross section in carbon of 250 ± 30 mb. This value is obtained by integrating over the energy and angular distributions of emitted α particles and concurs with our results within the limits of experimental error.

The tritium results from the irradiation of carbon are indeed puzzling. All of the earlier measurements are lower than those obtained in this investigation by a factor of 2 or 3. Partly in order to recheck our results, the graphite irradiation at 3 GeV was performed twice. Yet there was no variation in the results of tritium production from carbon. It is very unlikely that this discrepancy can be due to any systematic error in monitoring, tritium extraction, or counting, since the previous measurements were carried

TABLE II. Comparison of cross sections with previous measurements.

Reaction	Proton energy (MeV)	Cross section (mb)	Target material	Reference
$C+p \rightarrow {}^3H$	150	6.5 ± 1		a
	225	7.0 ± 1.1 [b]	$(CH_2)_n$	c
	300	6.4 ± 0.8 [b]	$(CH_2)_n$	c
	400	8.2 ± 1.0 [b]	$(CH_2)_n$	c
	450	7.2 ± 0.5 [b]	$(CH_2)_n$	d
	450	7.4 ± 1.1	$(CH)_n$	e
	550	10.1 ± 1.5	$(CH)_n$	e
	600	32 ± 4	Graphite	Present work
	660	10.6 ± 1.7	$(CH)_n$	e
	730	7.4 ± 1.2 [b]	$(CH_2)_n$	c
	2050	14.8 ± 1.2	$(CH)_n$	d
	3000	35 ± 4	Graphite	Present work
	5700	18	$(CH_2)_n$	f
	6200	17 ± 3 [b]	$(CH_2)_n$	g
$C+p \rightarrow {}^4He$	70	200 ± 60		h
	600	202 ± 24	Graphite	Present work
	660	>250 ± 30	Diamond	i
	3000	207 ± 25	Graphite	Present work
$O+p \rightarrow {}^3H$	225	13 ± 4.5	$(C_5O_2H_8)_n$	c
	300	19 ± 6	$(C_5O_2H_8)_n$	c
	400	36 ± 7	$Al(C_3H_5O_3)_3$	c
	450	37 ± 5 [b]	$C_2H_4O_2$	d
	600	34 ± 4	H_2O	Present work
	660	40 ± 12	O_2	j
	2050	32 ± 4 [b]	$C_2H_4O_2$	d
	2200	35 ± 4 [b]	H_2O	k
	3000	36 ± 4	H_2O	Present work
	6200	33 ± 6 [b]	$(C_5O_2H_8)_n$	g
$O+p \rightarrow {}^3He$	600	38 ± 5	SiO_2	Present work
	660	66 ± 13	O_2	j
	3000	40 ± 5	SiO_2	Present work
$Si+p \rightarrow {}^3H$	600	18 ± 3	Si	l
	600	36 ± 4	Si	Present work
	3000	58 ± 7	Si	Present work
$Si+p \rightarrow {}^3He$	600	34 ± 3	Si	l
	600	42 ± 5	Si	Present work
	2200	56 ± 11	Si	l
	3000	70 ± 8	Si	Present work
$Si+p \rightarrow {}^4He$	600	302 ± 30	Si	l
	600	331 ± 40	Si	Present work
	2200	330 ± 66	Si	l
	3000	365 ± 44	Si	Present work

[a] C. Brun, M. Lefort, and X. Tarrago, J. Phys. 23, 167 (1962).

[b] Renormalized to more recent monitor cross sections by J. Audouze, M. Epherre, and H. Reeves, in *High Energy Nuclear Reactions in Astrophysics*, edited by B. S. P. Shen (Benjamin, New York, 1967).

[c] M. Honda and D. Lal, Phys. Rev. 118, 1618 (1960).

[d] Reference 13.

[e] V. N. Mekhedov, Yadern. Fiz. 5, 34 (1967) [transl.: Soviet J. Nucl. Phys. 5, 24 (1967)].

[f] P. A. Benioff, UCRL Report No. UCRL 8780, 1959 (unpublished).

[g] L. A. Currie, Phys. Rev. 114, 878 (1959).

[h] C. Jacquot, thesis, University of Strasbourg, 1965 (unpublished).

[i] Reference 17.

[j] Reference 19.

[k] Reference 8.

[l] Reference 1.

out by five separate laboratories (discounting the 150-MeV irradiation as we are unfamiliar with the target material used). It is also improbable that recoil loss of tritons was a factor since the target thicknesses of the earlier investigations were in the range 0.5–3.2 g/cm^2. The only conspicuous major difference between our experiment and the previous ones was in the choice of target material. The older irradiations were all made on the thermoplastics polyethylene, $(CH_2)_n$, and polystyrene, $(CH)_n$. We are of the opinion that the earlier measurements are in error because a large part of the tritium produced in the irradiation of the thermoplastics diffused out of the targets. Consider the following evidence.

^{11}C production by high-energy protons on carbon targets is a widely used beam monitor and, as such, has been studied in depth. In thin polyethylene and polystyrene targets, ^{11}C loss by diffusion is a well-documented phenomenon.[6] Corrections of up to 12% for ^{11}C diffusion loss (depending on target thickness, not on beam intensity or target temperature) have been applied when these materials are used. It appears from the review of Cumming[6] that the loss of ^{11}C is slightly higher in $(CH_2)_n$. Experiments on ^{11}C production in graphite targets indicate no diffusion loss.

By far the most telling piece of evidence comes from our own measurements of tritium production in graphite, water, and the thermoplastic material Delrin, $(CH_2O)_n$. The irradiation of these materials was performed almost simultaneously, the graphite and Delrin targets were oxidized and the tritiated water collected in the same system, and the tritium measurements were made with the same counting equipment. Yet there was 2–3 times as much tritium present in the graphite and water as in the Delrin. In fact, we have calculated a tritium-production cross section based on the Delrin results alone and find a value (12–14 mb) very similar to those determined in polyethylene and polystyrene from the earlier publications. It is difficult to reach any conclusion other than that tritium rather readily diffuses out of the particular thermoplastic materials used in these studies and that, consequently, the previous measurements made with these materials are in error.

GSZ[1] demonstrated that by plotting certain product nuclei as a function of target atomic weight for a given bombarding energy, one finds a generally smooth curve which will either increase, decrease, or remain relatively level. (GSZ[1] show graphs of ^{3}H, ^{3}He, ^{4}He, ^{21}Ne, and ^{38}Ar cross sections in dependence of A at 600-MeV proton energy). In Fig. 3 we have plotted all of the published tritium cross sections as a function of A in targets from ^{4}He to Bi, at or near the proton bombarding energy of 600 MeV. This energy was chosen because it offered the greatest number of data points. In this figure one observes a smooth trend of increasing σ with increasing atomic weight. The approximate equation for this trend is $\sigma(A) = 30e^{A/100}$. There appears to be a slight flattening of the curve in the higher-mass range. Three data points fall noticeably below the curve, one each for carbon, magnesium, and silicon. The Mg and Si values are known to be too low because of incomplete tritium extraction.[1] Thus, only the carbon value fails to conform to the trend

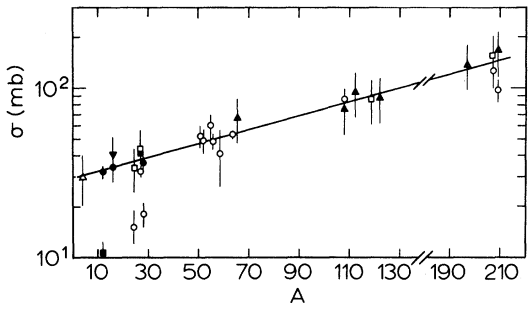

FIG. 3. Tritium cross sections as a function of target atomic weight at approximately 600-MeV proton bombarding energy. Symbols: ●, this work; ○, Goebel, Schultes, and Zähringer (Ref. 1); ■, Mekhedov, Yadern. Fiz. 5, 34 (1967) [transl.: Soviet J. Nucl. Phys. 5, 24 (1967)]; □, V. V. Kuznetsov and V. N. Mekjedov, Zh. Eksperim. i Teor. Fiz. 35, 587 (1960) [transl.: Soviet Phys. – JETP 8, 406 (1959)] at 600 MeV; ▲, Kuznetsov and Mekhedov, *ibid.* at 660 MeV; △, N. S. Kozodaev *et al.*, Zh. Eksperim. i Teor. Fiz. 38, 708 (1960) [transl.: Soviet Phys. – JETP 11, 511 (1960)]; ▼, Dudkin *et al.* (Ref. 19).

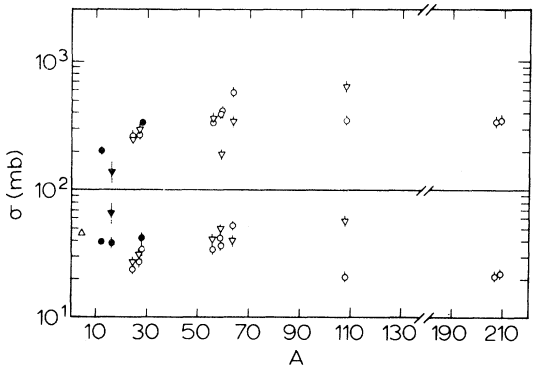

FIG. 4. ^{3}He (lower) and ^{4}He (upper) production cross sections as a function of target atomic weight at approximately 600-MeV proton bombarding energy. R. H. Bieri and W. Rutsch, Helv. Phys. Acta 35, 553 (1962) at 540 MeV. For explanation of symbols see Fig. 3.

established by more than 20 other elements.

At 3.0 GeV, the tritium and helium cross sections in C, O, and Si obtained in this investigation show the approximate $A^{2/3}$ increase predicted [23] by cascade and evaporation theories. The two cross-section formulas are not necessarily mutually inconsistent. At 600 MeV, cross sections for most high-energy products (generally corresponding to a large ΔA between target and product nuclides) are still increasing with increasing bombarding energy. Above a few GeV bombarding energy, the excitation functions are fairly level. This indicates that the nuclear mechanisms governing production rates are not identical for the two energy domains.

The tritium cross section in nitrogen at 450 MeV is 26 ± 4 mb.[13] Tritium cross section at 600 MeV are normally 10–20% higher than at 450 MeV, which would place nitrogen nicely on the curve in Fig. 3.

^{3}He and ^{4}He cross sections at about 600-MeV proton energy are shown as a function of target mass in Fig. 4. Disregarding the Pb and Bi data, there is a definite trend of increasing σ with increasing A for ^{4}He production. However, there is too large of a dispersion of data points to draw any reasonable curve. There does not seem to be any trend evident in the ^{3}He cross sections, even disregarding the few lowest values.

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