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**PRODUCTION OF TRITIUM AND HELIUM-3
BY PROTON BOMBARDMENT OF METALS;
SOLAR ORIGIN OF TERRESTRIAL TRITIUM**

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PRODUCTION OF TRITIUM AND HELIUM-3 BY PROTON
BOMBARDMENT OF METALS; SOLAR
ORIGIN OF TERRESTRIAL TRITIUM*

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November 8, 1965

ABSTRACT

Radiochemical analyses of recovered satellites have generated considerable interest in nuclear reactions initiated by relatively low-energy (< 150-MeV) solar protons. Of particular interest are the results obtained for the Discoverer XVII satellite, which was in orbit during the intense solar flare of November 12, 1960. The amounts of tritium and helium-3 found in portions of the recovered satellite appear to be one and two orders of magnitude too large, respectively, to be explained as products of reactions induced by solar protons. The fact that the helium-3 content was roughly ten times that of tritium is especially interesting, since proton-induced nuclear reactions were expected to produce roughly equal amounts of these two isotopes. To help explain these results, measurements of the cross sections for the production of tritium and helium-3, and especially of the ratio of these cross

*This work was done at UCLA.

sections, for proton energies in the vicinity of 30-50 MeV were desired. Accordingly, targets of aluminum and iron were bombarded with 35-45 MeV protons. Each target was then split, and the relative gross gamma activity of the two portions was determined. One portion of each target, together with blanks of the target materials, was then exposed to a thermal neutron flux of about 5×10^{19} n/cm². The tritium content of all the samples was determined by a system of vacuum extraction and gas counting. The results indicate that the ratio $\sigma\text{He}^3/\sigma\text{H}^3$ for bombardment of both aluminum and iron with 35-45 MeV protons lies in the range 0.8-1.4.

INTRODUCTION

At about 1320 UT on November 12, 1960, a large flare was observed on the surface of the sun.^{1, 2, 3, 4} The intensity of the flare, as measured by the resulting neutron flux at the earth's surface, reached a maximum at about 2000 UT.

On the same day, November 12, at 2042 UT, the United States satellite Discoverer XVII was launched into a polar orbit around the earth. The satellite remained in orbit for fifty hours, and was retrieved in mid-air upon reentry. A number of groups expressed interest in the nature of the bombardment of solar protons to which the satellite had been exposed. Accordingly, portions of the satellite were made available for study.

Fireman, deFelice, and Tilles² examined the tritium content of several lead and stainless steel components of Discoverer XVII. Their results are shown in Table I. The values shown for the required tritium production cross section are based on an integrated proton flux of 10^9 /cm². This number is derived from a number of additional studies of Discoverer XVII materials,^{1, 5, 6}

and from information from the Explorer VII satellite,⁷ which was also in orbit at this time. The proton flux number is only an approximate one, with rather uncertain limits of accuracy, but it does appear that the required cross sections are higher than one could expect for (p, t) reactions on iron and lead in the 100-MeV region by perhaps a factor of ten.

Schaeffer and Zahringer³ looked at He³ in aluminum and lead components from Discoverer XVII. The results of their work are shown in Table II. Here again the values for the required He³ production cross section are based on the rather uncertain 10^9 protons/cm², the values are higher than one would expect for (p, He³) reaction on Al and Fe in the 100-MeV region by at least a factor of one hundred.

It is rather difficult to make a direct comparison between the tritium values from Fireman, deFelice and Tilles and the He³ number from Schaeffer and Zahringer because of the different analytical techniques and the different samples studied by the two groups. It does appear, however, that He³ was at least ten times as abundant in Discoverer XVII as was tritium. Although very little information is available as to the ratio of the cross sections of (p, t) and (p, He³) reactions, especially at energies below a few hundred MeV, it was generally felt that the cross sections should be roughly equal.

It appears, then, that the He³ and tritium found in Discoverer XVII are incompatible in absolute and particularly in relative amount with a mechanism of production by proton bombardment of the satellite. Either the assumption of roughly equal cross sections for (p, t) and (p, He³) reactions at low proton energies (< 150 MeV) is considerably in error, or else part or all of the He³ and tritium in Discoverer XVII resulted from some process other than proton-induced

nuclear reactions in the satellite material. Possible diffusion loss of tritium from the satellite should be carefully examined to see whether it could constitute a significant factor.

Possible alternative mechanisms for the production of the observed tritium and He^3 will be presented at the end of this discussion.

To help clarify the Discoverer XVII data, we decided to attempt to measure the relative cross sections for production of tritium and He^3 in proton bombardment of aluminum and iron. We felt that it would be desirable to measure both the tritium and the He^3 in a single target so that the relative cross sections for the two reactions would be independent of the number of protons incident on the targets.

EXPERIMENTAL

Three aluminum targets, each 160 mg/cm^2 thick, and two iron targets, each 780 mg/cm^2 , were bombarded in the external proton beam of the 88-inch cyclotron at the Lawrence Radiation Laboratory in Berkeley. The external proton beam initially consisted of 45 (± 1) MeV protons. The second aluminum and iron targets were preceded by a 1/16-inch aluminum degrader foil and the third aluminum target was preceded by a 1/8-inch aluminum degrader. These degraders reduced the mean proton energies to 40 and 35 MeV, respectively. The targets were mounted in a Faraday cup assembly, and the time-integrated current from this holder, corrected for current leakage through the ionized cooling water, gave an approximate measure of the total proton flux through each target. Each target was bombarded by about 3×10^{17} protons (about 15 μA hours)

The bombarded targets were divided into two sections and the relative gross gamma activities of these two were determined. It was assumed that the distribution of gross gamma activity across the target would correspond closely to the distribution of tritium and He^3 . This was shown to be the case for tritium, and undoubtedly is also true for He^3 .

One portion of each target was then exposed to the high thermal neutron flux of the shuttle facility of the General Electric Test Reactor. The neutron flux was monitored by small, measured amounts of cobalt which were irradiated with the target portions. A separate irradiation of similar cobalt flux monitor, in which some of the monitors were shielded with 20 mils of cadmium foil, gave a measure of the ratio of epithermal (> 0.4 eV) to thermal flux at the shuttle position. This ratio, plus the effective cross section for the activation of Co^{59} to Co^{60} , corrected to a Maxwell distribution about the 50°C temperature of the GETR pool, was used to calculate the integrated thermal neutron flux to which the targets were exposed. The thermal neutron flux, in turn, was used to calculate the degree of conversion of the He^3 in the targets into H^3 . About 20 percent of the He^3 in the targets was converted into H^3 .

Each target portion was finally analyzed for tritium. The targets were held at 1000°C for one hour under an atmosphere of carrier hydrogen. After cooling, aliquots of this hydrogen were admitted into a large, center-wire type Geiger counter.^{8, 9} Argon and ethylene were added and the samples were counted.

The tritium in that portion of each target which was not irradiated with thermal neutrons was produced solely by (p, t) reactions on the target material during the proton bombardments. The tritium in the neutron-irradiated portion of each target was produced in three ways: by (p, t) reactions on the target material during the proton bombardments, by (n, t) reactions on impurities in the

target material (notably Li^6) during the neutron irradiations, and by (n, p) reactions on the proton-produced He^3 during the neutron irradiation.

The amount of (p, t)-produced tritium in each irradiated target portion was determined by the amount of tritium in the corresponding un-irradiated portion and the relative gross gamma activities of the two. The amount of tritium from (n, t) reaction on impurities was evaluated by irradiating blanks of the target material at the same time as the targets themselves, and relating the amount of tritium produced in these blanks to that produced in the targets as a result of impurities in the target material. The rest of the tritium in the irradiated target portions was taken to be the product of (n, p) reactions on He^3 ; these numbers, together with the independently determined degree of conversion of He^3 into H^3 in the neutron irradiation, yielded values for the amounts of He^3 produced in those target portions by proton bombardment.

We have, then, determined the amounts of proton-produced He^3 and H^3 in the targets, and the ratio of these values is equal to the ratio of the production cross sections.

RESULTS

The values obtained for the ratio of the (p, He^3) and (p, H^3) cross sections are shown in Table III. Several corrections have been applied to these numbers. All of the targets were corrected for recoil loss of tritium and He^3 during the proton bombardments. The magnitude of this correction was somewhat uncertain because the initial recoil energy of the tritons or He^3 particles is not known. If the reactions were "pick-up" processes, the light product particles would carry off most of the available energy. On the other hand, if the reactions were "compound nucleus" or "evaporation" processes, the light particles would

leave with relatively low kinetic energies, perhaps just enough to overcome the Coulomb barrier. That work which has been done on recoil energies in (p, t) and (p, He³) reactions¹⁰⁻¹³ seems to indicate that the light particles are emitted essentially isotropically, and with fairly low kinetic energy, especially for low Z targets. For the value of the correction, recoil energies of 10 and 15 MeV were assumed for the tritons and He³ particles, respectively. Diffusion loss of tritium or He³ from the targets between proton bombardment and tritium analysis was also considered. The only case where such loss was significant was that of tritium in iron targets. The magnitude of the correction for this loss is strongly dependent upon the "temperature history" of the tritium-containing iron targets. Uncertainties in this "temperature history" resulted in considerable uncertainties in the rather large correction factors, and consequently also in the value for the ratio of the He³ and tritium production cross sections in iron.

The values for the tritium production cross section are based on values for the proton fluxes as measured by the Faraday cup target holder. The uncertainty involved in the flux measurements is incorporated in the stated deviations for those values.

DISCUSSION

The primary conclusion to be drawn from our results is that proton bombardment of aluminum and iron in the energy region 35-45 MeV produces He³ and tritium with approximately equal cross sections. Our values are compared with those of previous studies^{12, 14-18} in Fig. 1, and appear to be not inconsistent. It seems that the ratio of these two cross sections is remarkably constant over a three-order-of-magnitude change in proton energy.

It is, therefore, quite certain that the tritium and particularly the He^3 found in Discoverer XVII were not produced by proton bombardment of the satellite.

Lal¹⁹ has indicated that stripping of solar alpha particles may have contributed much of the tritium found in Discoverer XVII, but this mechanism too would be expected to yield roughly equal amounts of both nuclides.

Several authors^{2, 4} have suggested that tritium and He^3 could be produced in a solar flare on the solar surface by reactions between protons and He^4 . The reactions involved, together with their threshold energies and cross sections,²⁰ are shown in Table IV. The ratio of H^3 to He^3 production at energies just above threshold is of the order of 0.1-0.2, a value in reasonable agreement with the amounts of those two species in Discoverer XVII. Flamm, Lingenfelter, MacDonald and Libby⁴ have shown that the composition of the November 12, 1960 solar flare particles can be accounted for by invoking $p + \text{He}^4$ reactions in the solar flare, and have gone on to show that such mechanisms may account for about 20 percent of terrestrial tritium and perhaps 70-80 percent of terrestrial He^3 .

REFERENCES

1. J. T. Wasson, J. Geophys. Res. 66, 2659 (1961).
2. E. L. Fireman, J. deFelice and D. Tilles, Phys. Rev. 123, 1935 (1961).
3. O. A. Schaeffer and J. Zahringer, Phys. Rev. Letters 8, 389 (1962).
4. E. Flamm, R. E. Lingenfelter, G. J. F. MacDonald, and W. F. Libby, Science 138, 48 (1962).
5. J. E. Keith and A. L. Turkevich, J. Geophys. Res. 67, 4525 (1962).
6. R. W. Stoenner and R. Davis, Jr., Bull. Am. Phys. Soc. 6, 277 (1961).
7. J. A. VanAllen, Bull. Am. Phys. Soc. 6, 276 (1961).
8. R. L. Wolfgang and W. F. Libby, Phys. Rev. 85, 437 (1952).
9. W. F. Libby and R. L. Heft, Phys. Rev. 100, 799 (1955).
10. J. Gonzalez-Vidal and W. H. Wade, Phys. Rev. 120, 1354 (1960).
11. B. L. Cohen and T. H. Handley, Phys. Rev. 93, 514 (1954).
12. L. E. Bailey, UCRL-3334 (1956).
13. L. Currie, Thesis, University of Chicago, 1955 (unpublished).
14. T. Kirsten, D. Kranowsky, and J. Zahringer, Geochim. et Cosmochim. Acta 27, 13 (1963).
15. G. R. Martin, S. J. Thompson, G. Wardle, and K. I. Mayne, Phil. Mag. 45, 410 (1954).
16. O. A. Schaeffer and J. Zahringer, Phys. Rev. 113, 674 (1959).
17. J. R. Arnold, M. Honda, and D. Lal, J. Geophys. Res. 66, 3519 (1961).
18. K. Goebel and J. Zahringer, Z. Naturforsch. 16a, 231 (1961).
19. D. Lal, G. Rajagopalan, and V. S. Venkatavaradan. To appear in the Proceedings of the International Conference on Cosmic Rays held in Jaipur, India, December 2-14, 1963.
20. A. F. Wickersham, Jr., Phys. Rev. 107, 1050 (1957).

Table I. Tritium and Argon-37 in Discoverer XVII.

Material (location)	Thickness (g/cm ²)	Tritium (10 ⁶ atoms/cm ²)	Required σ H ³ (mb)	Ar ³⁷ (10 ² atoms/cm ²)	Ratio H ³ to Ar ³⁷
Pb sheet (A) (surface)	2.0	3.2	550	---	---
Pb sheet (A) (center)	3.5	1.3	130	---	---
Pb sheet (B)	7.2	1.9	91	---	---
Pb sheet (C)	7.2	0.47	23	---	---
Pb sheet (C)	7.2	0.51	24	0.52	9800
Stainless steel case	1	0.54	50	1.89	2900
Stainless steel case	1	0.48	44	---	---
Pb sheet (unflown)	7.2	0.05	---	---	---

Table II. Helium-3 in Discoverer XVII.

Material	Thickness (g/cm ²)	Helium-3 (10 ⁶ atoms/cm ²)	Required σ He ³ (barns)
Aluminum Flange I	1.62	162	4.5
Adjacent Sheet I	0.27	16.2	2.7
Aluminum Flange II	1.62	405	11.2
Adjacent Sheet II	0.27	5.4	0.90
Lead	6.80	88.5	4.5

Table III. Corrected final values for σH^3 and the ratio $\sigma He^3/\sigma H^3$.

Target	Average proton energy (MeV)	σH^3 (mb)	$\sigma He^3/\sigma H^3$
Aluminum No. 1	44 ± 1	7.0 ± 1.3	0.8 ± 0.2
Aluminum No. 2	39 ± 1	5.9 ± 1.1	1.4 ± 0.3
Aluminum No. 3	34 ± 1	4.4 ± 0.8	1.2 ± 0.3
Iron No. 1	42 ± 1	8.6 ± 1.0	1.4 ± 0.7
Iron No. 2	36 ± 1	7.0 ± 0.8	0.8 ± 0.7

Table IV. Reactions of 28-MeV protons on Helium-4.

Reaction	Threshold energy (MeV)	Cross section (mb)
$\text{He}^4(\text{p}, 2\text{p})\text{H}^3$	25.0	8.9 ± 1.0
$\text{He}^4(\text{p}, \text{pn})\text{He}^3$	25.5	4.8 ± 1.3
$\text{He}^4(\text{p}, \text{d})\text{He}^3$	23	50

Fig. 1. The ratio of $\sigma \text{He}^3 / \sigma \text{H}^3$ for proton bombardment of aluminum and iron.

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