TRITIUM PRODUCTION IN METALS BY 120 TO 660-Mev PROTONS

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Results of an investigation of tritium production in various elements by 660-Mev protons are presented. Excitation functions are given for tritium production in the 120 to 660 Mev range.

EXPERIMENTAL METHOD

THE present investigation was undertaken to supplement the results of references 1 to 6 and to determine additional characteristics of tritium production in metals by protons.

Specimens of metals measuring $2\times6\times15$ mm were bombarded with protons in the internal beam of the synchrocyclotron of the Joint Institute for Nuclear Research. Four to six specimens were irradiated simultaneously while fastened in a massive aluminum holder. The proton beam was directed parallel to the 6-mm side. The bombarding energy was varied by changing the target radius in the synchrocylotron. Irradiation times were 2 to 5 min in an internal beam of 10^{11} to 10^{12} protons/sec.

The quantity of tritium in a bombarded target was determined by means of a special vacuum system which is shown diagramatically in Fig. 1. The apparatus consists of a tubular furnace 1 with a quartz tube 2 for melting the targets 3; a palladium thimble 4 with an electric heater 5 to separate the hydrogen-tritium mixture from the other gaseous reaction products; traps containing activated charcoal 6 cooled by liquid nitrogen (a trap was used to produce a pressure drop during passage of the active gas); and a Geiger counter 7 in a lead shield 8 (40 mm thick). The apparatus also included several reservoirs — for hydrogen 9, pure argon 10, alcohol 11, and a tank 12 for diluting the highly active mixture.

Bombarded targets were melted in a quartz tube of 140 cm^3 volume, in a hydrogen atmosphere* at 50 mm Hg. The melting time was 1.5 to 2 hours at 900 to 1050°C.† About 90% of the tritium was separated from the specimens. A second melting process under the same conditions gave 8 or 9% and a third process yielded 1 to 2% of the total tritium activity. A special check of the yield of the active mixture by activated charcoal showed that the charcoal yields practically all of the gas at room temperature.

The vacuum system enabled us to send into the counter from 0.057 to 0.1 of the hydrogen-tritium mixture at 1 to 6 mm Hg. The β-particle detector was a cylindrical glass counter of 60 cm^3 volume with a copper cathode. The counter was filled with alcohol vapor at 15 mm and argon at 95 to 100 mm in addition to the hydrogen-tritium mixture. Under these conditions, the Geiger counter had the following operating characteristics: a counter plateau from 100 to 150 v, a plateau slope not greater than 10% at 100 v, and a background of 80 to 100 pulses per minute. The tritium activity was measured at 1000 to 10000 pulses per minute. The efficiency of the counter for registering beta particles from tritium decay is estimated at about 90%.

The proton beam was monitored by a 20μ aluminum foil in which the specimens were wrapped during bombardment. The dependence of the yields of the $Al^{27}(p, 3pn)Na^{24}$ reaction on the proton energy was taken from references 7 and 8. After bombardment the aluminum foil was dissolved in 1 ml of HCl, after which the decay of activity was measured in the fraction 0.01 of the solution. The activity of the monitor was measured by an end-window counter with mica windows 3 mg/cm^2 thick. The detector recorded 18% of the entire activity of the monitor.

EXPERIMENTAL RESULTS

The table contains the cross sections of various elements for tritium production by 120 to 660 Mev protons. The values in the table are the averages of not less than 3 measurements, except that for magnesium, tin, gold and bismuth single

*The hydrogen used experimentally as a carrier first passed through the palladium thimble.

†The experiments were performed with elements whose melting points did not exceed 1100°C.

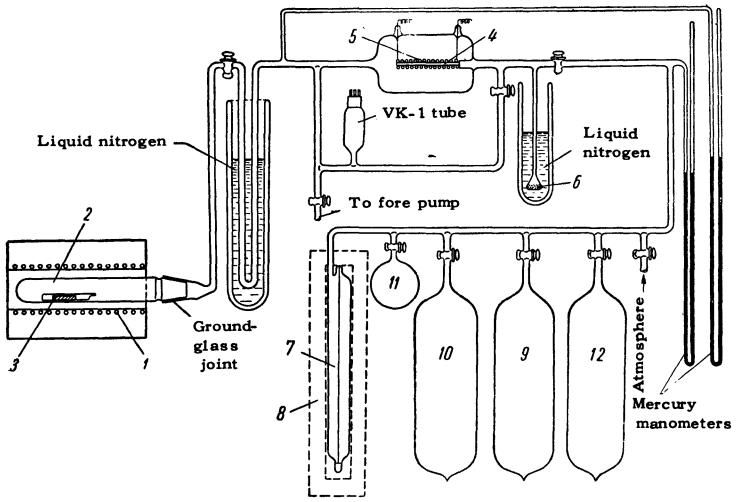

FIG. 1. Diagram of the vacuum system for determining the amount of tritium produced in various elements by high-energy protons.

measurements are sometimes given. The total error in determining each value is estimated at ± 30%. This is double the half-width of the curve of deviations from the average experimental values for all of the experiments; the curve was almost Gaussian in shape.

The tritium production cross sections σ_{H3} of aluminum and lead at 120 Mev are almost identical. Considerable increase of σ_{H3} for heavy elements is observed as the proton energy is increased. In the 300 to 500 Mev range the increase of the cross section is slowed down, but above 550 Mev it increases sharply. At 660 Mev the σ_{H3} of lead and bismuth is almost twice as large as on the plateau (300 to 500 Mev) and reaches 10% of the geometric cross section. This relation is clear for lead and bismuth. For aluminum and magnesium the relation is almost linear. The values for tin lie between the values for lead and aluminum.

The dependence of σ_{H3} on atomic weight at 660 Mev is shown in Fig. 2, where the dashed line shows the dependence of the excitation energy on atomic number at 450 Mev. The figure also shows the cross sections which we obtained experimentally at 450 Mev or by means of interpolation of the energy relations (triangles), and gives results taken from reference 1 (open circles). There is good agreement between our results and reference 1 for aluminum and lead at 450 Mev. Figure 2 is marked by a steady increase of σ_{H3} with A. At 660 Mev σ_{H3} of aluminum and bismuth increases ~3 times as A increases ~8 times.

DISCUSSION

It is interesting to observe how the tritium production cross section depends on proton energy at still higher energies. Fig. 3 gives data on tritium production in aluminum and lead by fast protons; the data were obtained in the present work or from the literature.[1,4,6] Experimental values

Target material	Proton energy, Mev	Average cross section	Number of experiments	Target material	Proton energy, Mev	Average cross section	Number of experiments
Magnesium	660	43 ± 13	2	Tin	550	69 ± 21	1
Aluminum	660	46 ± 14	17	Lead	550	87 ± 26	3
Copper	660	74 ± 22	6	Bismuth	550	75 ± 22	1
Zinc	660	67 ± 20	7	Aluminum	500	37 ± 11	5
Silver	660	76 ± 23	4	Magnesium	450	30 ± 9	1
Cadmium	660	95 ± 28	3	Aluminum	450	24 ± 7	3
Tin	660	122 ± 37	3	Lead	450	91 ± 27	3
Antimony	660	88 ± 26	2	Magnesium	300	19 ± 6	2
Gold	660	139 ± 41	1	Aluminum	300	25 ± 7	3
Lead	660	186 ± 56	11	Tin	300	38 ± 11	2
Bismuth	660	167 ± 50	4	Lead	300	73 ± 22	4
Aluminum	600	44 ± 13	2	Bismuth	300	73 ± 22	1
Tin	600	86 ± 26	1	Aluminum	200	18 ±6	3
Lead	600	157 ± 47	3	Aluminum	120	16 ± 5	3
Magnesium	550	26 ± 8	2	Lead	120	17 ± 5	3
Aluminum	550	33 ± 10	6				

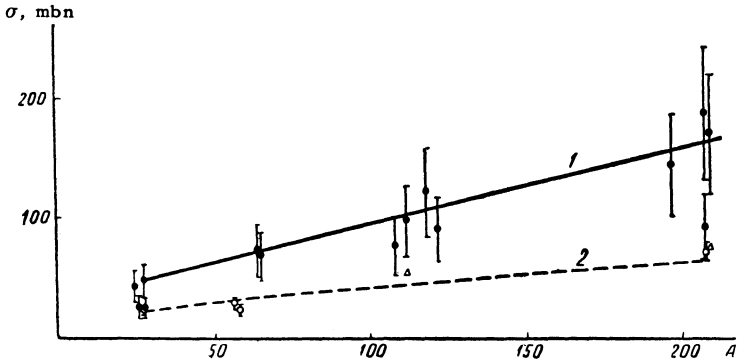

FIG. 2. Dependence of the tritium production cross section on the atomic weight of the target material at 1) 660 Mev and 2) 450 Mev.

taken from the literature are indicated in Fig. 3 by open circles and squares. The crosses denote cross sections in iron for the proton energies 0.16, 1.0, 3.0 and 6.2 Bev (reference 4) and 50 to 170 Mev.[6] The figure shows that the observed growth of σ_{H3} in lead above 550 Mev is continued. For 2.05 Bev σ_{H3} exceeds the cross section at 300 to 500 Mev by a factor of more than 5 and reaches 25% of the geometric cross section. In the case of aluminum σ_{H3} is almost unchanged up to 2.05 Bev. σ_{H3} increases steadily in iron with proton energy, but the curve for iron is closer to the curve for aluminum than for lead. Only for the energy region from 0.1 to 0.17 Bev is σ_{H3} in iron below the value for aluminum. Thus with increasing proton energy the heavy elements show a relatively greater rise of the tritium production cross section than the light elements.

In Fig. 3 the solid curves give the dependence of the variation of the probability of tritium emission from lead and aluminum as calculated by evaporation theory in the 120 to 500 Mev proton energy range. The calculated curves were adjusted to experimental results so as to provide coincidence of the greatest possible number of points. The calculation was performed by means of the formulas given by Hagedorn and Mackay,[9] using the excitation energies given in reference 10. (For lead we used the excitation energy calculated for bismuth.) The relation which gives the temperature of a nucleus at a given excitation energy was taken from reference 11. The effective barrier for tritium emission was $V'_{H^3} = V_0/2 = 8.1$ Mev for lead and $V_{H^3} = V_0/2 = 2.3$ Mev for aluminum. We see that the evaporation theory gives a relation between σ_{H^3} and proton energy which is about the same as the experimental result.

The relative probability of tritium emission, $\gamma_{H^3}/\gamma_{tot}$, where γ_{tot} is the total evaporation probability of neutrons, protons, alpha particles etc. ($\gamma_{tot} = \gamma_n + \gamma_p + \gamma_d + \gamma_{H^3} + \gamma_{He^3} + \gamma_{He^4}$), also agrees with the experimental ratio $\sigma_{H^3}/\sigma_{geom}$ for 120- and 500-Mev protons. For aluminum $\gamma_{H^3}/\gamma_{tot}$ was 10^{-2} and 3×10^{-2}, whereas $\sigma_{H^3}/\sigma_{geom}$ is 2.5×10^{-2} (neglecting the transparency of the nucleus). For lead $\gamma_{H^3}/\gamma_{tot}$ was 3×10^{-3} and 1.1×10^{-2} with the same proton energies. $\sigma_{H^3}/\sigma_{geom}$ (neglecting transparency) was 8×10^{-3} and 4×10^{-2}, respectively. Thus the relative probability of tritium emission varies in the same direction as the experimental ratio $\sigma_{H^3}/\sigma_{geom}$, differing by a factor of not more than 3. This discrepancy is not significant in view of the approximate character of the calculations.

The agreement of the proton-energy dependence of σ_{H^3} (Fig. 3) and of $\sigma_{H^3}/\sigma_{geom}$ with evaporation theory indicates that in our experiments we recorded mainly tritium nuclei which

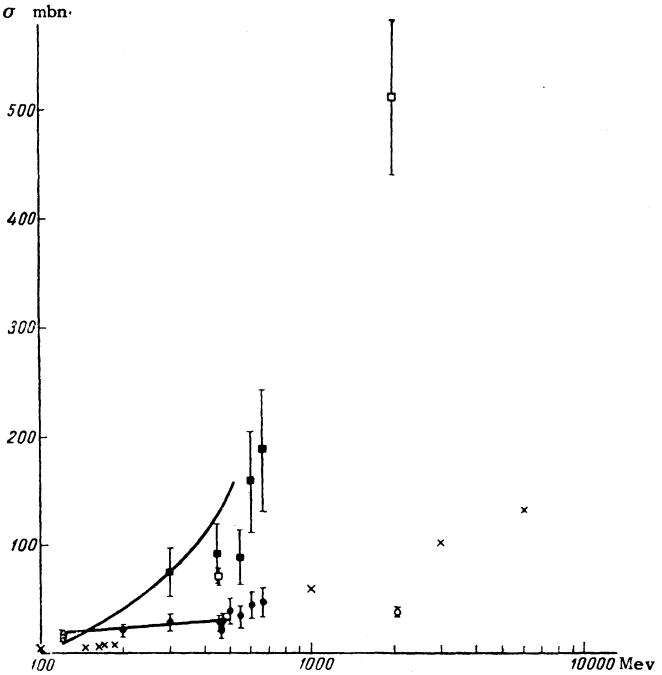

FIG. 3. Dependence of the tritium production cross section of aluminum, lead and iron on proton energy. ■, □ – Pb; ●, ○ – Al; × – Fe; solid curve – calculation by evaporation theory.

had been evaporated. This conclusion is also supported by the shapes of the curves for the excitation energy and production cross section σ_{H^3} in different elements with 450-Mev protons (Fig. 2).

For proton energies above 500 Mev evaporation theory gives a probability of tritium emission which is notably above the experimental cross sections, although the ratio $\gamma_{H^3}/\gamma_{tot}$ remains approximately unchanged. If we assume that theory agrees with experiment in the 120 to 500 Mev energy range, then for 2 Bev the probability of tritium emission exceeds the experimental value by ~ 7 times in aluminum and ~ 13 times in lead. These excessive calculated values are evidently due to the fact that at such high proton energies the evaporation theory cannot be applied.

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