

PRODUCTION OF TRITIUM IN LEAD AND ALUMINUM BY HIGH-ENERGY  
PROTONS, DEUTERONS, AND ALPHA PARTICLES

V. V. KUZNETSOV

Joint Institute for Nuclear Research

Submitted to JETP editor December 3, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 40, 1263-1269 (May, 1961)

Experimental data are presented on the production of tritium in lead and aluminum by 70 – 390 Mev deuterons and 140 – 750 Mev  $\alpha$  particles, and in zinc and cadmium by 750-Mev  $\alpha$  particles. The tritium yields from aluminum and lead targets of different thicknesses bombarded with 660-Mev protons are given.

EXPERIMENTAL PROCEDURE

TRITIUM production by high-energy protons has been investigated by many authors,<sup>1-9</sup> but relatively few investigations of tritium production by deuterons and  $\alpha$  particles have been published.<sup>5,10</sup> In the present work tritium production in metals by high-energy deuterons and  $\alpha$  particles is studied.

As in earlier work,<sup>7</sup> the targets were made of aluminum and lead with the dimensions (1.5 – 2)  $\times$  6  $\times$  (15 – 30) mm, and were bombarded by 70 – 390 Mev deuterons and 140 – 750 Mev  $\alpha$  particles in the internal beam of the synchrocyclotron of the Joint Institute for Nuclear Research.

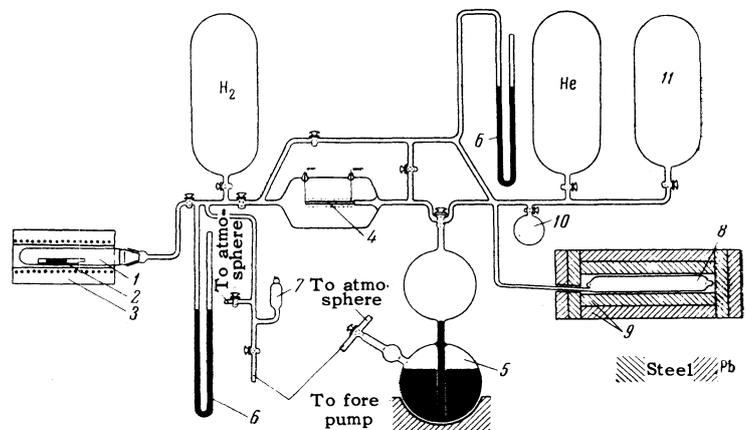
Three to six target samples were fastened simultaneously to a heavy aluminum holder. Heat was removed from the target through the holder, which was clamped tightly to the synchrocyclotron probe. When lead targets were bombarded the deuteron beam was reduced to one-half or one-third of the maximum,  $2 \times 10^{12}$  deuterons/sec. The intensity of the  $\alpha$ -particle beam was (4 – 5)  $\times 10^{10}$  particles/sec. The beam direction was parallel to the 6-mm side of the target. Different bombard-

ing energies were associated with the positioning of the targets in orbits of different radii within the vacuum chamber of the synchrocyclotron. Deuteron bombardment lasted from 2 to 5 min, while  $\alpha$  particle bombardment lasted from 5 to 20 min.

The amount of tritium in a bombarded target was determined by separating the tritium component from the target using the vacuum system represented in Fig. 1. This system consists of a quartz tube 1 for melting the targets, a palladium filter 4 with an electric heater, where a hydrogen-tritium mixture was separated from the other gaseous reaction products, a mercury pump 5 producing the required pressure drop when the active hydrogen-tritium mixture was admitted, and a Geiger counter shielded (9) by 35 mm of steel and 35 mm of lead. The system was also equipped with glass reservoirs for hydrogen, pure helium, and alcohol (10), and with an extra vessel 11 for dilution of the highly active mixture.

The targets were melted in the 200-cm<sup>3</sup> quartz tube in a hydrogen atmosphere at 40 – 100 mm Hg. Each melted target was kept in the hydrogen atmosphere at 850 – 950° C for 40 – 100 min, resulting in

FIG. 1. Diagram of vacuum system: 1 – quartz tube, 2 – target, 3 – furnace, 4 – palladium filter, 5 – vacuum pump, 6 – mercury manometer, 7 – VK-1 vacuum tube, 8 – MS-9 counter, 9 – shield, 10 – ethyl alcohol container, 11 – additional vessel.



Target material	Energy of bombarding deuterons, Mev	$N_T/N_{Na}$	$\sigma_T$	No. of runs	Target material	Energy of bombarding $\alpha$ particles, Mev	$N_T/N_{Na}$	$\sigma_T$	No. of runs
Al	70	$0.57 \pm 0.10$	$12.5 \pm 3.1$	6	Al	140	$1.48 \pm 0.16$	$44.7 \pm 9.1$	8
Pb	70	$1.85 \pm 0.44$	$40.8 \pm 12.5$	6	Pb	140	$3.27 \pm 0.41$	$98.1 \pm 21.2$	13
Al	150	$1.33 \pm 0.05$	$29.2 \pm 4.7$	6	Al	300	$2.10 \pm 0.12$	$52.5 \pm 9.2$	6
Pb	150	$3.60 \pm 0.13$	$79.2 \pm 12.8$	6	Pb	300	$3.76 \pm 0.74$	$94.0 \pm 25.4$	8
Al	270	$1.35 \pm 0.05$	$29.7 \pm 4.6^*$	6	Al	540	$2.62 \pm 0.42$	$52.4 \pm 9.6^*$	6
Pb	270	$3.78 \pm 0.22$	$83.6 \pm 14.6^*$	5	Pb	540	$4.88 \pm 1.04$	$97.6 \pm 27.0^*$	10
Al	390	$1.73 \pm 0.06$	$38.1 \pm 6.0^*$	9	Al	750	$2.71 \pm 0.17$	$48.8 \pm 8.6^*$	6
Pb	390	$3.97 \pm 0.91$	$87.3 \pm 15.9^*$	12	Zn	750	$4.40 \pm 0.45$	$79.2 \pm 16.0^*$	2
					Cd	750	$4.80 \pm 1.30$	$86.4 \pm 27.2^*$	2
					Pb	750	$10.82 \pm 0.75$	$194.8 \pm 35.4^*$	14

\*Cross sections  $\sigma_T$  for tritium production were obtained by using extrapolations of data on  $Na^{24}$  production in aluminum<sup>11,12</sup> by high-energy deuterons and  $\alpha$  particles.

the removal of  $\sim 90\%$  of the tritium; this was checked by remelting under the same conditions. The vacuum system made available for measurement 4 to 63% of the hydrogen-tritium mixture produced by melting of the targets. This mixture produced a pressure of 1 to 25 mm Hg in the counter.

Beta particles from tritium decay were registered by a cylindrical  $\sim 200 \text{ cm}^3$  counter with a copper cathode. The counter contained, in addition to the hydrogen-tritium mixture, a working mixture of ethyl alcohol at 15 mm and helium at 95–100 mm. The counter was operated in the Geiger region, in most instances with the following characteristics: 100–150 v plateau, plateau slope not greater than 10% at 100 v, and intrinsic background 130–200 pulses/min. Under higher hydrogen pressure ( $> 15 \text{ mm}$ ) the counter characteristics were adversely affected:  $\sim 50$ –80 v plateau, and 15–20% slope at 100 v. The measured tritium activity amounted to 500 to 10 000 pulses/min. The efficiency of  $\beta$ -particle registration from tritium decay is estimated at about 90%.

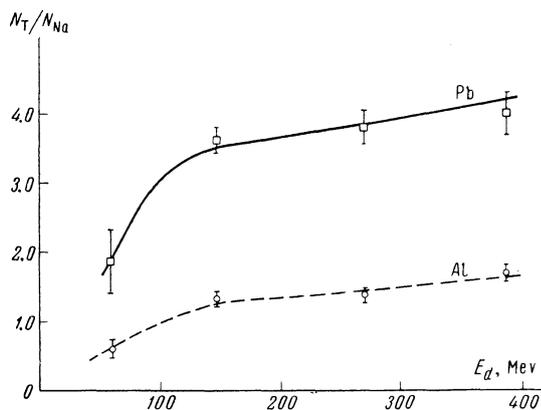


FIG. 2. Relative yields of tritium from aluminum and lead vs deuteron energy.

The deuteron and  $\alpha$  particle intensities were monitored by aluminum foils, as described in reference 7. The dependences of the yields from the reactions  $Al^{27}(d, \alpha p)Na^{24}$  and  $Al^{27}(\alpha, \alpha 2pn)Na^{24}$  on bombarding energy were taken from the literature.<sup>11,12</sup> The cross sections for  $Na^{24}$  production in aluminum by deuterons above 200 Mev and  $\alpha$  particles above 400 Mev were determined by extrapolating the  $Na^{24}$  yields to higher energies. The cross section for  $Na^{24}$  production in aluminum by 270-Mev and 390-Mev deuterons is estimated at  $\sim 22 \text{ mb}$ .  $\sim 20 \text{ mb}$  and  $\sim 18 \text{ mb}$  are obtained for 540-Mev and 750-Mev  $\alpha$  particles, respectively.

Beta particles from  $Na^{24}$  decay were registered by a counter with a quartz end window ( $< 5 \text{ mg/cm}^2$ ). The detecting equipment registered  $\sim 18\%$  of the entire activity of a sample.

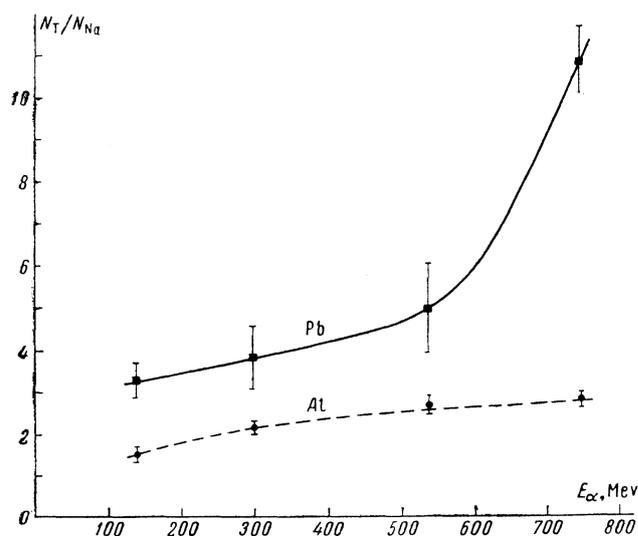


FIG. 3. Relative yields of tritium from aluminum and lead vs  $\alpha$ -particle energy.

## EXPERIMENTAL RESULTS

The table gives the relative yields  $N_T/N_{Na}$  and the cross sections  $\sigma_T$  for tritium production\* in aluminum and lead bombarded with 70–390 Mev deuterons and 140–750 Mev  $\alpha$  particles. The relative yields of tritium from zinc and cadmium bombarded by 750-Mev  $\alpha$  particles are also given. The relative yields are the averages of at least five independent measurements, except for zinc and cadmium, in which cases only two measurements were averaged. Determination of the cross sections  $\sigma_T$  took into account the systematic experimental errors and the errors in determining the cross section for  $Na^{24}$  production in aluminum.

The probable experimental error was estimated by constructing a histogram of the departures of relative tritium yields from the average in all runs. The error, which was the half-width of the near-Gaussian distribution, was under 15%.

Figures 2 and 3 show the relative yields of tritium from aluminum and lead vs deuteron and  $\alpha$ -particle energies. The relative yields from aluminum do not increase much with the bombarding energy. In the case of lead the increase is much more pronounced for both deuterons and  $\alpha$  particles.

Figure 4 shows the dependence of tritium production by 750-Mev  $\alpha$  particles on atomic number. The yield increases with the atomic number just as in the case of bombardment by 450-Mev and 660-Mev protons.<sup>7</sup> With 750-Mev  $\alpha$  particles the tritium yield from lead is about four times greater than that from aluminum; corresponding values of  $A$  differ by a factor of about 8.

As a basis for deciding on target thicknesses the tritium yields were determined from 60–1500  $\mu$  lead targets and 500–2000  $\mu$  aluminum targets bombarded by an internal 660-Mev proton beam. Three 6 mm  $\times$  20 mm targets were fastened to the aluminum holder at one time. The beam path was parallel to the 6-mm side of the targets. Lead targets were covered with 2-mm aluminum. In this way the number of protons passing through the lead targets was reduced to less than one-tenth of the maximum intensity ( $5 \times 10^{12}$  protons/sec), and the proton flux was more uniformly distributed throughout the target thickness.

\*The cross section for tritium production was determined from the formula  $\sigma_T = \sigma_{Na} N_T/N_{Na}$ , obtaining  $N$  from  $N = N_0 e^{\lambda t_2} / (1 - e^{-\lambda t_1})$ , where  $t_1$  is the duration of target bombardment,  $t_2$  is the length of time from the determination of bombardment to the start of activity measurement,  $N_0$  is the activity per gram-atom of the target material at the time  $t_2$ , and  $\lambda$  is the decay constant.

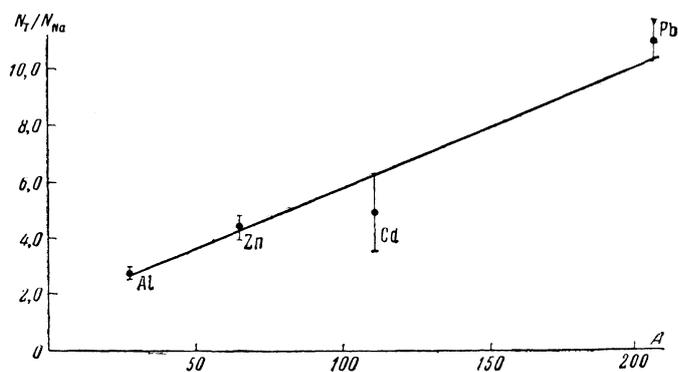


FIG. 4. Relative yield of tritium produced by 750-Mev  $\alpha$ -particles vs atomic weight of target.

Figure 5 shows the experimental tritium yields from aluminum and lead targets of different thicknesses. The tritium yield is practically constant for lead thicknesses greater than 800  $\mu$  and aluminum thicknesses greater than 500  $\mu$ . For lead thicknesses less than 800  $\mu$  the tritium yield is observed to decrease.

The triton energy spectrum can be determined indirectly from the given experimental setup with a lead target. For this purpose calculated curves of tritium yields from lead targets of different thicknesses are plotted in the figure. The method of calculation is similar to that described in reference 13 for the emission of lithium fragments from lead foils of different thicknesses bombarded with protons. In the present work it was assumed that the experimentally observed tritium was formed only by evaporation and that the angular distribution of the tritons is isotropic. The triton energy spectrum was obtained from the formula

$$P(E) = (E - V) \tau^{-2} e^{-(E-V)/\tau},$$

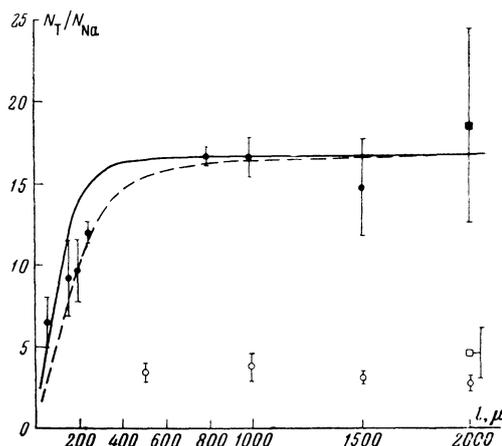


FIG. 5. Tritium yields from different thicknesses  $l$  of aluminum (open circles) and lead (filled circles) targets. The small squares represent data from reference 7. The solid curve was calculated from evaporation theory for  $\tau = 3.5$  Mev and  $V = 7$  Mev, and the dashed curve for  $\tau = 5$  Mev and  $V = 7$  Mev.

in Heisenberg's evaporation theory.<sup>14</sup> The parameters  $\tau$  and  $V$ , representing the temperature of the nucleus and the Coulomb barrier, respectively, were selected to satisfy the experimental results.

The two curves shown in Fig. 5 were calculated with two sets of parameters:  $\tau = 3.5$  Mev,  $V = 7$  Mev and  $\tau = 5$  Mev,  $V = 7$  Mev. The second set is in better agreement with experiment.

The mean temperature  $\tau = 5$  Mev is higher than that predicted by the existing evaporation theory ( $\tau_{\text{evap}} \approx 3$  Mev). This indicates that evaporation is not the only mechanism for tritium production by high-energy protons. Wade et al.<sup>5</sup> observed some anisotropy of triton emission favoring the forward direction in proton, deuteron, and  $\alpha$ -particle bombardments. Lefort et al.<sup>8</sup> observed  $\sim 10\%$  fast tritons unaccounted for by the existing evaporation theory, in addition to the bulk of the tritons, which are associated with evaporation. The experimental tritium yields from lead targets of different thicknesses show that the triton kinetic energy does not generally exceed 30–40 Mev. Our experimental arrangement permits an additional contribution from cascade neutrons and neutrons of "evaporation" origin from both the target and screening aluminum. The additional triton yield from interactions of evaporation and cascade neutrons with the aluminum screening of the targets was investigated separately. A stack of 14 identical 400- $\mu$  aluminum foils was irradiated with 400-Mev deuterons for 10 min in the internal synchrocyclotron beam. Foil activity decreased progressively with depth in the stack. This experiment was intended to determine the systematic increase of  $N_T/N_{Na}$  from the first to the last foil. The failure to observe this increase indicates that less than 10% of the tritium results from interactions of cascade and evaporation neutrons with the aluminum.

## DISCUSSION OF RESULTS

Figures 2 and 3 and the table show that the tritium yield from aluminum is not observed to increase strongly with the bombarding energy. However, the yield from lead increases appreciably with deuteron energy, and even more strongly as  $\alpha$ -particle energy is increased from 540 to 750 Mev. In the latter case the effect apparently results from a considerable increase of the excitation energy due to nuclear capture of pions created in nucleon-nucleon collisions.

The mean excitation energies were calculated for the bombardment of both aluminum and lead

by deuterons and  $\alpha$  particles, using both the present data and the results given in reference 7. The formulas of Hagedorn and Macke were used,<sup>14</sup> showing direct dependence of tritium production probability on nuclear excitation energy. The combined data indicate that 70-Mev deuterons and 140-Mev  $\alpha$  particles striking a target nucleus contribute almost all their energy to nuclear excitation. Hence the mean excitation energy of aluminum and lead bombarded by 390-Mev deuterons is estimated at  $\sim 65$  Mev and  $\sim 110$  Mev, respectively; for 750-Mev  $\alpha$  particles the mean values are  $\sim 150$  Mev and  $\sim 180$  Mev, respectively.

An analysis of the tritium production cross sections in reference 7 at bombarding energies  $< 200$  Mev shows highest probability for  $\alpha$  particles, followed by deuterons and protons, in that order. A deuteron or  $\alpha$  particle impinging on a target nucleus must evidently be treated as two or four independent particles, respectively, which participate independently in cascade development within the nucleus; this leads to large energy transfer to the target nucleus and thus to a high probability of tritium production. It must not be forgotten, however, that in deuteron and  $\alpha$ -particle bombardments a possible contribution to triton production comes from neutron pickup by deuterons and proton stripping from  $\alpha$  particles in nuclear force fields.<sup>5,15</sup>

In conclusion the author wishes to thank M. Ya. Kuznetsova, V. N. Mekhedov, and V. A. Khalkin for their interest and several critical comments.

<sup>1</sup> Currie, Libby, and Wolfgang, *Phys. Rev.* **101**, 1557 (1956).

<sup>2</sup> E. L. Fireman and F. S. Rowland, *Phys. Rev.* **97**, 780 (1955).

<sup>3</sup> E. L. Fireman, *Phys. Rev.* **97**, 1303 (1955).

<sup>4</sup> E. L. Fireman and J. Zähringer, *Phys. Rev.* **107**, 1695 (1957).

<sup>5</sup> Wade, Gonzalez-Vidal, Glass, and Seaborg, *Phys. Rev.* **107**, 1311 (1957).

<sup>6</sup> K. Goebel, CERN 58-2 (1958).

<sup>7</sup> V. V. Kuznetsov and V. N. Mekhedov, *JETP* **35**, 587 (1958), *Soviet Phys. JETP* **8**, 406 (1959).

<sup>8</sup> Lefort, Simonoff, Tarrago, and Bibron, *J. phys. radium* **20**, 959 (1959).

<sup>9</sup> L. A. Currie, *Phys. Rev.* **114**, 878 (1959).

<sup>10</sup> J. L. Yntema, *Phys. Rev. Letters* **4**, 297 (1960).

<sup>11</sup> Batzel, Crane, and O'Kelley, *Phys. Rev.* **91**, 939 (1953).

<sup>12</sup> M. Lindner and R. N. Osborne, Phys. Rev. **91**, 342 (1953).

<sup>13</sup> Wang, Kuznetsov, Kuznetsova, Mekhedov, and Khalkin, JETP **39**, 527 (1960), Soviet Phys. JETP **12**, 370 (1961).

<sup>14</sup> W. Heisenberg, editor, Kosmische Strahlung (Springer Verlag, Berlin), 1953.

<sup>15</sup> J. B. Mead and B. L. Cohen, Phys. Rev. Letters **5**, 105 (1960).

Translated by I. Emin  
217