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SPALLATION-FISSION COMPETITION IN HEAVIEST ELEMENTS: TRITON PRODUCTION

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ABSTRACT

Stacked foils of Au^{197}, Th^{232}, and U^{238} were bombarded, in a series of experiments, with 48-Mev helium ions, 24-Mev deuterons, and 32-Mev protons. Tritium from each foil was collected and then measured by a gas-counting technique. The qualitative results indicate that high-energy tritons are emitted in relatively large abundance from all targets and with each of the bombarding particles. Cross sections for triton production from fissionable U^{238} and Th^{232} and from non-fissionable Au^{197} are comparable. The integrated (α,t) cross sections thus determined for U^{238} and Th^{232} are nearly the same as the integrated cross sections for the "$(\alpha,p2n)$" reactions as determined by measuring the yield of the product heavy isotopes in radiochemical experiments. All of the facts are consistent with a picture of emission of high-energy tritons (whether due to a stripping, pick-up, or other mechanism) in which fissionable intermediate nuclei are formed mainly at levels of excitation below their fission thresholds.

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I. INTRODUCTION

Considerable information about nuclear reaction mechanisms can be obtained both from radiochemical yield studies of residual nuclei and from investigations of outgoing neutrons, protons, and other small particles. Initial studies of the present series[1-4] dealt with radiochemical yields while the present study is the first to measure the yield of an outgoing particle.

In the first paper of the series[1] radiochemically determined yields were reported for spallation products from helium-ion-induced reactions in plutonium isotopes. These yields were characterized by the similar magnitude of the (α,xn) and (α,pxn) type cross sections. The more recent radiochemical studies[2-4] of heavy-element nuclear reactions induced by helium ions of energies less than 50 Mev also have shown that certain cross sections, particularly that for the production of the $(\alpha,p2n)$ product, are too large relative to those for the production of the (α,xn) product to be explained on the basis of a compound-nucleus, evaporation picture in which charged-particle emission would be discriminated against. It has been observed that for U^{238},[2] U^{235},[3] U^{233},[3] and Th^{232} [4] the $(\alpha,p2n)$ reaction is, in general, the most prominent of all the (α,xn) and (α,pxn) reactions observed. These considerations led to the hypothesis that the large values of the $(\alpha,p2n)$ cross sections in the region of fissionable nuclei are due to direct interaction of helium ions and target nuclei in which are produced tritons of sufficient energy to often leave the residual nuclei in states below fission and particle-emission thresholds. If this is true, it is reasonable to expect comparable $(\alpha,p2n)$ type cross sections for the heavy fissionable and the somewhat lighter non-fissionable nuclides. The following work was undertaken to test this hypothesis, and includes work on the (d,t) and (p,t) reactions.

Only a few experiments on tritium emission have previously been described. Tritons from the α and d and p,t reactions in bombardments with projectile energies greater than 100 Mev have been observed.[5,6,7] In experiments with 600- and 2000-Mev protons Currie, Libby, and Wolfgang[8] collected tritium from a number of elements and measured its radioactivity. With lower-energy particles (< 25 Mev), tritium production from deuteron and proton bombardments has been the subject of experimental investigation in both the light-element[9,10] and heavy but non-fissionable element[11] regions. The stripping theory of Butler[12] has been applied by Newns[13] by considering the (d,t) process as the inverse of stripping, i.e., neutron pick-up.

II. EXPERIMENTAL

For the helium-ion and deuteron bombardments, stacked .05-mm foils of natural Au^{197} and isotopically pure U^{238}, and 0.1-mm foils of natural Th^{232} were subjected to the external beams of the 60-inch Crocker Laboratory cyclotron. The proton bombardments were carried out on the Berkeley linear accelerator using the same technique. In a later paper it will be shown that triton production by these three bombarding particles is quite general over the entire periodic table. For this reason it was impossible to vary the beam energy by imposing degrading foils in front of the target without introducing an extraneous source of tritons. Thus, only maximum-energy beams were available to the first foil in the stack (48-Mev helium ions, 24-Mev deuterons, and 32-Mev protons).

The tritium was extracted by heating the foils to 1200°C in a measured amount of hydrogen carrier followed by selective diffusion of hydrogen isotopes through a palladium thimble and introduction into a counter tube. This apparatus is essentially the same as used by Currie, Libby, and Wolfgang, who discussed the possible experimental errors inherent in the apparatus. Counting was done in the proportional region using a methane filling of 1 atmos pressure and a partial pressure of hydrogen of ~0.04 atmos. Counting rates varied from 2×10^2 to 1×10^5 c/m, representing a total yield of tritium from the target into the counter tube of 35 to 40%.

III. RESULTS

A. The (α,t) reaction.

The apparent cross sections (σ) in mb for the (α,t) reaction calculated on the basis of thin-target approximations are shown in Figure 1-A as a function of the mg/cm^2 of gold, thorium, and uranium as measured from the end of the stack upon which the beam was incident. The number of tritium atoms found in a foil, the number of particles incident on the stack, and the atoms per cm^2 in the foil were employed in each calculation. Since the helium-ion beam ranges are 460, 485, and 500 mg./cm^2 of gold, thorium, and uranium respectively, the maximum cross section consistently seems to be observed at depths in the target beyond the range of the helium ions. The integrated (α,t) cross sections determined for gold, thorium, and uranium are listed in Table I in units of millibarn-mm and tritons per incident helium ion.* The cross sections are compared with cross sections for the production of the corresponding residual nuclei determined by the radiochemical method in the cases of thorium[4] and uranium[2]. It will be noted that tritium-emission and residual-nuclei cross sections agree within 20% in both cases. Table I also shows that the integrated (α,t) cross section for gold agrees within 20% with the radiochemical yield values for the corresponding reaction for uranium and thorium.

B. The (d,t) and (p,t) reactions.

Figures 1B and 1C show plots for the yields of tritium in the (d,t) and (p,t) reactions. In these cases no radiochemical cross-section determinations are available for comparison. The deuteron beam is completely stopped by 930, 995, and 1000 mg./cm^2 of gold, thorium, and uranium respectively, and the proton beam by 2450, 2580, and 2660 mg./cm^2 of gold, thorium, and uranium respectively.

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* In general, the number of tritons per incident particle is

$$t_i = 6.02 \times 10^{-7} \frac{\beta}{\mathrm{A.\ W.}} \sum_{0}^{x(\mathrm{max.})} \sigma_x ,$$

where β is the target density in mg./cm^2-mm, A. W. is the atomic weight, and σ_x is the cross section in millibarns for foil x.

IV. DISCUSSION

The striking result that the apparent cross section for the yield of tritium in the (α,t) reaction reaches a peak in target foils that the beam does not reach, leads, on detailed examination of the data, to the conclusion that tritons with velocities comparable with those of the helium ions incident on the foil stack must be emitted in forward directions. The high energy of the tritons (20–30 Mev from an analysis of Fig. 1A) indicates that for a large share of interactions the residual heavy product must be left in the ground state or in a low-lying excited state, and hence little fission competition is possible for U^{238} and Th^{232}. This conclusion that these nuclei survive fission is given confirmation by the agreement seen in Table I between the cross sections for the production of the "$(\alpha,p2n)$" product determined by radiochemical methods which measure only the events surviving fission, and the tritium-production cross sections. The fact that the integrated cross sections for tritium production are somewhat higher than the corresponding $(\alpha,p2n$ or $t)$ cross sections determined radiochemically may indicate the presence of a spectrum of triton energies of which approximately 20% results in residual nuclei being left in states sufficiently excited to undergo fission. The differences as they stand, however, are such that it is possible that the two types of cross sections are actually equal. It should be noted that although it is believed that the tritium activity collected and the corresponding heavy fragments observed radiochemically in the main represent simply (α,t) reactions, the tritium actually may result from (α,t), (α,tn), and (α,tf) reactions and the "$(\alpha,p2n)$" products may result from (α,t), $(\alpha,p2n)$, and (α,dn) reactions. For this reason and also because of the fact that the experimental procedures are so different, one must exercise caution in interpreting the data.

A direct interaction mechanism such as stripping of a proton from the helium ion is suggested by experimental data presented here. If the residual nucleus is to be left in an energy state between ground and the fission threshold, the tritons must carry off an energy of between $(48 + Q)$ Mev and $(48 + Q - 5)$ Mev, or approximately $(35 - 30)$ Mev in the case of full-energy helium ions. If tritium production is assumed most predominant in the first few mils of foil, that is, with helium-ion energies greater than 30 Mev as shown by the radiochemical data, then the energy of the tritons found deep in the stacked foils can be calculated without too much uncertainty. The results show that these tritons, having an angular dependence strongly favored in the direction of the helium ion beam, have energies of 30 ± 5 Mev. Further, analysis of the data of Fig. 1-A shows that if the majority of tritons have energies between 25 - 35 Mev they are emitted within a cone of total included angle of 60°.

The data for triton production by deuteron projectiles appear reasonable in the light of a mechanism by which the deuteron "picks up" a neutron as it passes near the target nucleus. Harvey[11] has studied the excited states of the residual nuclei produced from this reaction in Au^{197} and found the production of nuclei in their ground state quite prevalent. Table I shows that the cross sections for triton production in (d,t) reactions are actually considerably larger in Th^{232} and U^{238} than in Au^{197} despite the fissionability of the former two. The nature of the experiments is such that the high energy of the tritons is not as obvious in deuteron bombardments, since the range of the deuterons is greater than the range of the maximum-energy tritons so that no tritium is found in foils beyond the deuteron range.

Cross sections for triton production in (p,t) reactions are similar in all three of the nuclides although the integrated cross sections are slightly larger in the case of Th^{232} and U^{238}. Thus, the magnitudes of the cross sections again reflect a lack of fission competition with the (p,t) reaction for Th^{232} and U^{238}. This reaction is a possible example of a pick-up of two nucleons (double pick-up). Cohen[10] has studied the (p,t) reaction in beryllium and iron and considers it to be a direct interaction process. Again the range of the tritons is less than that of the protons, and it is not possible to determine the range of the tritons by the present simple experimental techniques.

Thus, it has been established in a qualitative way that high-energy tritium emission is occurring to an important extent in heavy fissionable and non-fissionable isotopes; that most, if not all, of the production of "$(\alpha,p2n)$" products is accounted for by the (α,t) process; and that tritium emission reactions proceed at these energies through a mechanism not involving formation of a compound nucleus. The (α,t) reaction probably proceeds by a stripping mechanism with a high-energy triton emergent in the forward direction. On the basis of their large production cross sections the tritons from the (d,t) and (p,t) reactions would also appear to be generated by a mechanism circumventing compound-nucleus formation, and probably represent examples of "pick-up" reactions. It is interesting to note that cross-section values for the helium-ion, deuteron, and proton reactions are in the order $(\alpha,t) < (p,t) < (d,t)$, which is the generally accepted order of increasing ease of the processes, helium-ion stripping, double neutron pick-up, single neutron pick-up.

At the present time, independent determinations of the range and angular distribution of tritons produced in these various reactions are under investigation in this laboratory and will be reported shortly.

This project was performed under the auspices of the U.S. Atomic Energy Commission. Appreciation is expressed to the crews of the 60-inch cyclotron and the linear accelerator for their assistance.

Table I. Integrated Triton Production Cross Sections

Reaction	Isotope	Integral σ mb-mm	Integral σ Tritons/incident particle	Radiochemical value
(α,t)	Au^{197}	2.17	$1.28 \pm 0.11 \times 10^{-5}$	--
	Th^{232}	5.15	$1.56 \pm 0.13 \times 10^{-5}$	1.27×10^{-5} *
	U^{238}	2.60	$1.23 \pm 0.09 \times 10^{-5}$	1.08×10^{-5} **
(d,t)	Au^{197}	8.55	$5.04 \pm 0.45 \times 10^{-5}$	--
	Th^{232}	37.4	$11.4 \pm 1.0 \times 10^{-5}$	--
	U^{238}	20.0	$9.45 \pm 0.75 \times 10^{-5}$	--
(p,t)	Au^{197}	6.84	$4.04 \pm 0.52 \times 10^{-5}$	--
	Th^{232}	24.2	$6.15 \pm 0.70 \times 10^{-5}$	--
	U^{238}	13.7	$5.81 \pm 0.62 \times 10^{-5}$	--

* Assuming a counting efficiency of 100%.

** Assuming a counting efficiency of 70%.

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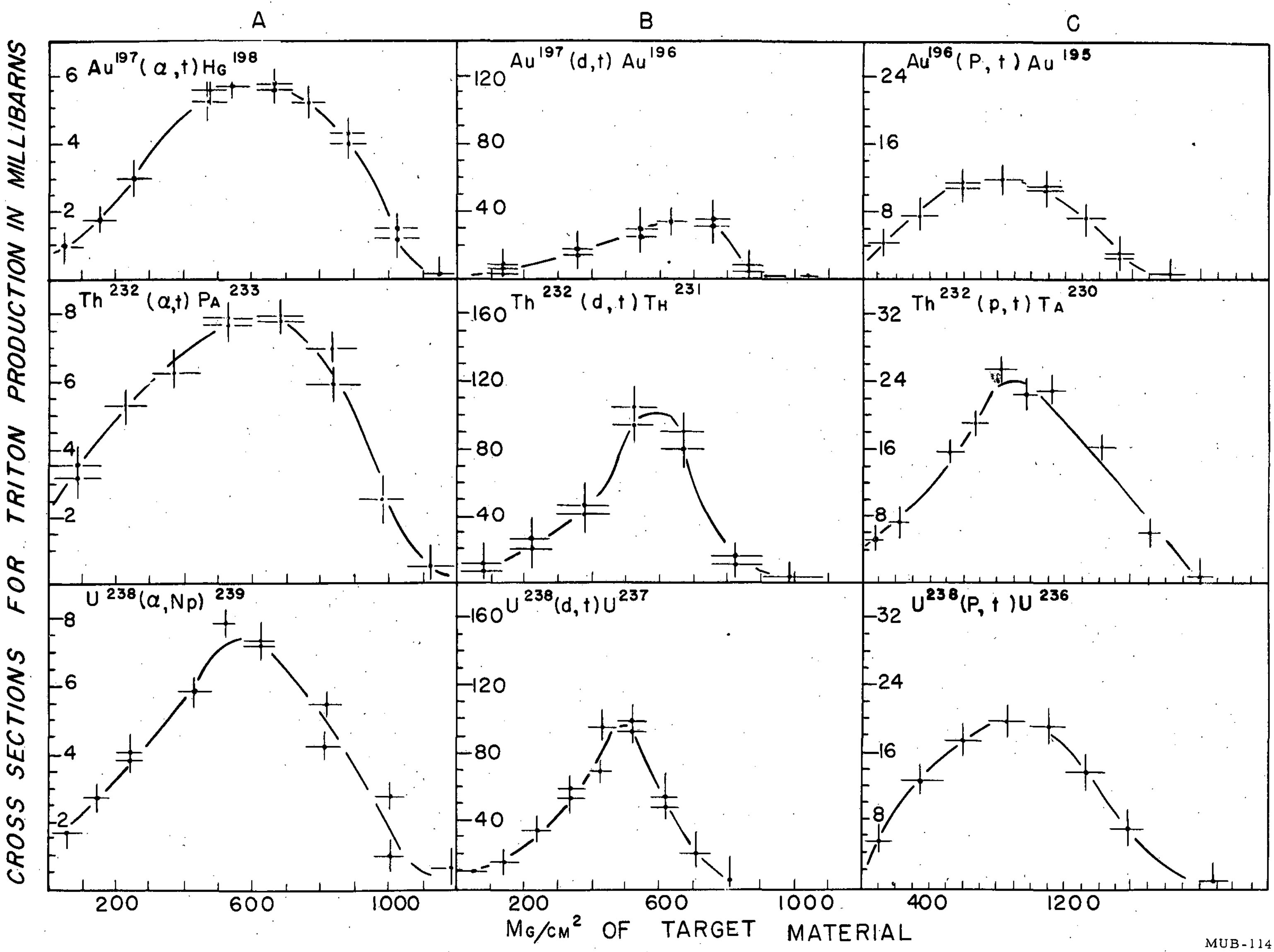
FIGURE CAPTION

Fig. 1. Cross section for triton production in Au^{197}, Th^{232}, and U^{238} induced by helium ions, deuterons, and protons. All abscissas are expressed in mg/cm^2 of target material with numerical scales as given below Sections 1A, 1B, and 1C. The ordinates are in units of millibarns with the individual numerical scales shown for each of the nine curves.

Fig. 1A, Cross sections for triton production induced by helium ions;
Fig. 1B, Cross sections for triton production induced by deuterons;
Fig. 1C, Cross sections for triton production induced by protons.


A
B
C
CROSS SECTIONS FOR TRITON PRODUCTION IN MILLIBARNS
Au197(α,t)Hg198
Au197(d,t)Au196
Au196(P,t)Au195
Th232(α,t)Pa233
Th232(d,t)Th231
Th232(p,t)Ta230
U238(α,Np)239
U238(d,t)U237
U238(P,t)U236
200
600
1000
400
1200
Mg/cm2 OF TARGET MATERIAL

MUB-114