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# SPALLATION-FISSION COMPETITION IN HEAVY-ELEMENT REACTIONS: $Th^{232}$ + $He^{4}$ AND $U^{233}$ + d[*]

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## ABSTRACT

Cross sections and excitation functions have been determined for spallation and fission products from bombardments of $Th^{232}$ with helium ions (15 to 46 Mev) and $U^{233}$ with deuterons (9 to 24 Mev). This work extends a series of investigations of charged particle ($\alpha$, d, and p) induced reactions in heavy elements ($z \geq 88$). Radiochemical methods were employed to isolate products corresponding to the following spallation reactions: neutron emission, ($\alpha$,4n), ($\alpha$,5n), (d,n), (d,2n), and (d,3n); emission of one proton and neutrons ($\alpha$,p), ($\alpha$,pn), ($\alpha$,p2n), and ($\alpha$,p3n); and emission of two protons and neutrons, ($\alpha$,2p), ($\alpha$,2pn), and ($\alpha$,$\alpha$n), and (d,$\alpha$n). In addition, the following fission products were isolated from one or more bombardments: $Zn^{72}$, $Ge^{77}$, $As^{77}$, $Br^{82,83}$, $Rb^{86}$, $Sr^{89,91}$, $Y^{93}$, $Zn^{95,97}$, $Nb^{96}$, $Mo^{99}$, $Ru^{103,105,106}$, $Pd^{109,112}$, $Ag^{111}$, $Cd^{115,115m,117}$, $I^{130,131,133}$, $Cs^{136}$, $Ba^{139,140}$, $La^{140}$, $Ce^{141,143,144}$, $Nd^{147}$, $Eu^{157}$, and $Gd^{159}$.

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The results show that fission is the predominant reaction at all energies for $Th^{232}$ and to an even greater extent for $U^{233}$. The data for the surviving spallation products are consistent with several mechanisms of reaction, including compound-nucleus formation and evaporation, direct interactions between nucleons of the incoming helium ion or deuteron and nucleons of the nucleus, and a combination of these types of processes (direct interaction followed by evaporation). In general, the results confirm and extend previously established concepts.

The neutron-emission spallation reactions as well as fission are best explained as proceeding through compound-nucleus formation. The shapes and magnitudes of $(\alpha,4n)$, $(d,2n)$, and $(d,3n)$ excitation functions correlate well with a compound-nucleus treatment modified to include fission competition. According to this treatment, ratios of neutron to total-reaction level width, $\Gamma_n/\Sigma_i\Gamma_i$, are 0.49 for $U^{236-233}$ [from $Th^{232}$ $(\alpha,4n)$], 0.17 for $Np^{235-234}$ [from $U^{237}$ $(\alpha,2n)$], and 0.20 for $Np^{235-233}$ [from $U^{233}$ $(d,3n)$]. In addition the total-reaction excitation functions (consisting mostly of the fission excitation functions) are consistent with theoretical cross sections for compound-nucleus formation calculated with a nuclear radius parameter $r_o = 1.5 \times 10^{-13} A^{1/3}$.

The fission mass-yield curves are similar to those found for other heavy target isotopes (for elements from thorium to plutonium). The minimum in the curves in the region of mass 120 tends to disappear as helium-ion or deuteron energy is increased.

The $(\alpha,pxn)$, $(\alpha,2pxn)$, $(\alpha,\alpha n)$, $(d,n)$, and $(d,\alpha n)$ products are attributed to direct interactions, with complex particles emitted in preference to a series of protons and neutrons. Thus $(\alpha,d)$, $(\alpha,t)$, and $(\alpha,tn)$ mechanisms would account for most of the $(\alpha,pn)$, $(\alpha,p2n)$, and $(\alpha,p3n)$ products, respectively. In the case of the $(\alpha,t)$ and $(\alpha,tn)$ reactions, analysis of the ratio $\frac{\sigma(\alpha,tn)}{\sigma(\alpha,t)}$ leads one to the conclusion that with 35-Mev helium ions only 9% of outgoing tritons leave the residual nucleus with sufficient energy to evaporate a neutron or undergo fission, and with 44-Mev helium ions only 20% do so. The $(d,n)$ product probably results from the stripping reaction.

# SPALLATION-FISSION COMPETITION IN HEAVY-ELEMENT REACTIONS: $Th^{232} + He^{4}$ AND $U^{233} + d$

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## I. INTRODUCTION

This paper extends a series of radiochemical investigations of excitation functions for spallation and fission reactions induced in heavy elements ($Z \geq 88$) by medium-charged particles.[1-6]

A number of studies[1-3,5-10] in the medium-energy range (5 to 50 Mev) have involved compound systems[11] of high total charge (between Z = 94 and Z = 100), in which fission accounts for more than 90% of the total cross section observed. Other studies[12-15] have involved compound systems of lower total charge ($Z \leq 90$), in which the percentage of the reactions proceeding by fission is small ( < 10%). This paper considers compound systems in an intermediate region, where the contribution from fission and spallation should be more nearly equal. Some investigations on fission[16-23] and spallation[23-26] in this region have previously been reported.

Both $Th^{232}$ and $U^{233}$ provide a further opportunity for studying spallation products that survive the fission reaction, whether by successful competition in the compound-nucleus—evaporation chain or by avoiding the competition in direct interaction.[1] Thorium-232 presents an ideal case for studying reactions of the ($\alpha$,pxn) and ($\alpha$,2pxn) type,[27] which may contain contributions from ($\alpha$,d), ($\alpha$,t), ($\alpha$,$He^3$) and other direct interactions. Uranium-233 presents a convenient case for the exploration of deuteron-induced reactions in the heavy-element region, for comparison with the large amount of data available from helium-ion—reactions.

It is also of interest to compare the fission mass-yield distribution in this intermediate region[28] with the results obtained for compound systems of higher Z[1,5,7] and those of lower Z.[14,15]

## II. EXPERIMENTAL PROCEDURES[29]

Uniform 1-mil thorium metal foils were used for most of the thorium bombardments. A few of the thorium targets as well as all the $U^{233}$ targets (96% $U^{233}$, 3% $U^{238}$, < 1% $U^{234}$) were thin uniform deposits (~500 $\mu g/cm^2$) of the hydrated thorium or uranium oxide electrodeposited[1] on aluminum. The amount of $Th^{232}$ or $U^{233}$ was determined by alpha counting of the resulting thin deposits. For thorium, corrections were applied to account for the contribution from alpha-particle-emitting daughters of the $Th^{232}$.[31]

The targets were bombarded in a water-cooled microtarget holder which also served as a Faraday cup for beam-intensity measurements. The helium-ion beam (48.0 ± .5 Mev) for the deuteron beam (24.0 ± .5 Mev) of the Berkeley 60-inch cyclotron was used. Aluminum or platinum foils were used to degrade the helium-ion or deuteron beam to the desired energy.[33]

For the thorium foil bombardments the large amounts of activity produced allowed separate aliquots of the dissolver to be used for the various fractions analyzed. In the other cases the carriers and tracers were all present during the dissolution and a sequential separation was performed. For the thorium bombardments the protactinium fraction was purified by extraction with diisopropyl ketone (DIPK) and elution on an anion-exchange column.[31] The thorium fraction was extracted into thenoyltrifluoroacetone (TTA) re-extracted into 3 M $HNO_3$, precipitated as the iodate, and passed through an anion-exchange column. The uranium fraction was purified by an anion-exchange-column elution. For the $U^{233}$ bombardments the chemical separation of the neptunium fraction was essentially that described by Vandenbosch et al.[5] The protactinium fraction was separated from the neptunium by carrying neptunium(IV) fluoride on $LaF_3$. The protactinium was purified further by extraction into DIPK from hydrochloric acid solution.

The fission products were purified by techniques adopted from those described by Meinke[34] and Lindner.[35]

The counting rates of alpha-particle-emitting spallation products were measured by use of a multichannel alpha-pulse-height analyzer. The counting rates of spallation products which decay by orbital electron capture were determined with a methane-flow windowless proportional counter. The counting efficiency of this counter for $Np^{234}$ was measured as 63 ± 2% by milking the daughter $U^{234}$ and determining its alpha-disintegration rate. The counting efficiencies for $Np^{233}$ and $Np^{232}$ were estimated to be 80 ± 20%.[5,30] The counting rates of the protactinium, thorium, and fission-product samples were determined by using end-window "amperex" Geiger counter tunes. Appropriate correction factors[30,31] were applied to obtain disintegration rates from the measured counting rates.

## III. RESULTS

### A. Spallation

The measured spallation cross sections[27] for helium-ion-induced reactions in $Th^{232}$ are listed in Table I. The limits of error are estimated. The excitation functions constructed from these data are shown in Figs. 1 and 2.

The cross sections for the spallation reactions induced in $U^{233}$ by deuterons are listed in Table II. The cross sections for the (d,αn) reaction are based on a value of 8% for the $\beta^-$ branching of $Pa^{230}$, [36] and were determined by observing the growth and decay of $U^{230}$ in the protactinium fraction. The quoted limits of error in the spallation cross sections do not include uncertainties in the counting efficiencies used. The $U^{233}$ excitation functions are shown in Fig. 3.

### B. Fission

The measured cross sections for the formation of various nuclides in helium-ion-induced fission of $Th^{232}$ are listed in Table III. Once again the limits of error are estimated. These cross sections were increased by a small amount to account for the mass-chain yields not represented by the measured fission product[37] (because of products of higher atomic number which do not decay to the observed fission product), in order to obtain total fission yields for each mass number (also given in Table III).

The fission mass-yield curves for $Th^{232}$ are shown in Fig. 4. Complementary fission-product cross sections were obtained by assuming that the average number of neutrons, $\nu$, emitted per fission is an integral number near the value given by the empirical formula[38]

$$\bar{\nu} = 2 + E_{ex}/8 ,$$

where $E_{ex}$ is the excitation energy of the compound nucleus.

The total fission cross sections indicated in Table III were obtained by summation under the smooth fission mass-yield curves of Fig. 4.

Table I. Spallation-product cross sections for $Th^{232}$+$He^{4}$ (millibarns)

| Reaction[a] | (α,4n) | (α,5n) | (α,p) | (α,pn) | (α,p2n) | (α,p3n) | (α,2p) | (α,2pn) | (α,αn) |
|---|---|---|---|---|---|---|---|---|---|
| Product | $U^{232}$ | $U^{231}$ | $Pa^{235}$ | $Pa^{234}$(UZ)[b] | $Pa^{233}$ | $Pa^{232}$ | $Th^{234}$ | $Th^{233}$ | $Th^{231}$ |
| Mode of decay | α | α,EC | $\beta^-$ | $\beta^-$ | $\beta^-$ | $\beta^-$ | $\beta^-$ | $\beta^-$ | $\beta^-$ |
| Half life | 70 y | 4.3 d | 23.7 m | 6.7 h | 27.0 d | 1.31 d | 24.1 d | 23.3 m | 25.6 h |
| Threshold(Mev) | 29.5 | 36.9 | 11.9 | 18.1[c] | 23.3[c] | 29.2[c] | 17.5 | 23.7[c] | 6.5 |
| $E_\alpha$ (Mev)[d] | | | | | | | | | |
| 15.0-18.0 | | | | | 0.21±.02 | 0.11±.002 | | | 3.6±1.2 |
| 19.9-22.4 | | | | | 0.21±.03 | 0.016±.003 | | 0.11±.06 | 0.93±.4 |
| 21.6-23.9 | | | <0.07 | | 0.41±.05 | 0.020±.006 | | <0.14 | <0.14 |
| 21.9-25.5 | | | | | (1.14±.2)[e] | (<0.013)[e] | | | |
| 23.6-25.9 | | | | | 0.31±.05 | | | | |
| 26.5 | | | | | (1.3±.4)[e] | | <2.1 | | |
| 25.7-28.1 | | | 1.14±.4 | 0.61±.20 | 3.3±.4 | | 0.16±.08 | 0.54±.27 | 0.84±.28 |
| 25.7-28.9 | <0.008 | | | | (6.21±1.3)[e] | | | | |
| 25.5-29.1 | | | | | (0.94±.6)[e] | (0.31±.2)[e] | | | |
| 29.4 | | | | (5.1±2)[e] | (9.0±2)[e] | | | | <0.1 |
| 27.6-29.9 | | | 0.64±.21 | 0.41±.04 | 2.4±.3 | 0.08±.02 | | | 3.6±1.2 |
| 29.9-31.8 | 0.11±.06 | | 3.4±0.6 | 5.2±.5 | 8.5±.9 | 0.34±.1 | 0.055±.028 | 0.62±.31 | 2.3±.8 |
| 32.3 | <30 | | (8.1±4.0)[e] | (3.4±1.2)[e] | (7.8±2.1)[e] | (<3)[b] | | | 3.5±1.2 |
| 30.6-33.4 | 1.0±.1 | | | | (7.1±1)[e] | (0.30±.15)[e] | <0.01 | | 1.5±.6 |
| 32.0-33.7 | 0.12±.01 | | 3.3±1.2 | 6.4±.6 | 10±1 | 0.25±.05 | <0.08 | 2.9±1.1 | |
| 35.9 | | | | | | | <0.2 | | 16±3 |
| 34.8-36.9 | 17.9±1.8 | | 2.9±.6 | 16±2 | 14±2 | 0.93±.2 | | 0.36±.19 | 16±2 |
| 34.9-37.7 | 15.6±1.6 | | | | (12±6)[e] | | | | 20±4 |
| 34.9-37.7 | 19.0±1.9 | | | | (<17)[e] | | | | 12±3 |
| 35.8-37.8 | 17.5±1.8 | | 5.4±2.0 | 21±2 | 17±2 | 1.8±.4 | | 1.9±1.4 | 28±7 |
| 38.9 | 27.9±2.8 | | | (10±3)[e] | (8.2±2)[e] | | | | 11±2 |
| 39.3-41.0 | 54.7±5.5 | | 3.0±1.5 | 22±2 | 21±2 | 2.3±.4 | | 2.0±.7 | 15±5 |
| 39.0-41.6 | 54.5±5.5 | | | 21±2 | 18±2 | | <6 | | 37±19 |
| 39.6-42.2 | | | | (17±8)[e] | (14±8)[e] | (<15)[b] | | | 31±6 |
| 43.8-45.4 | 31.6±3.2 | | <1.7 | 19±4 | 24±5 | 4.1±1.0 | | 4.2±1.5 | 35±9 |
| 43.4-45.9 | <100 | | | | | | <0.07 | | |
| 43.4-45.9 | 34.7±3.7 | | | | (<117)[e] | | | | 42±8 |
| 43.4-45.9 | 33.4±3.3 | 3.7±1 | | 18±2 | 22±3 | 5.2±1 | | | 49±14 |

a. See Reference 27.

b. Only one isomer determined; $UX_2$ yields not measured.

c. Thresholds for reactions in which d,t, or $He^3$ are emitted are lower than those listed by approximately 2.2, 8.7, and 7.7 Mev, respectively.

d. All energy values are uncertain by 0.5 Mev. A range of energies indicates a foil target, a single energy and electroplated target.

e. This cross section was measured through the use of a preliminary protactinium chemistry which did not include the anion-exchange column elution. It is consequently considered unreliable and is therefore not shown in Fig. 1.

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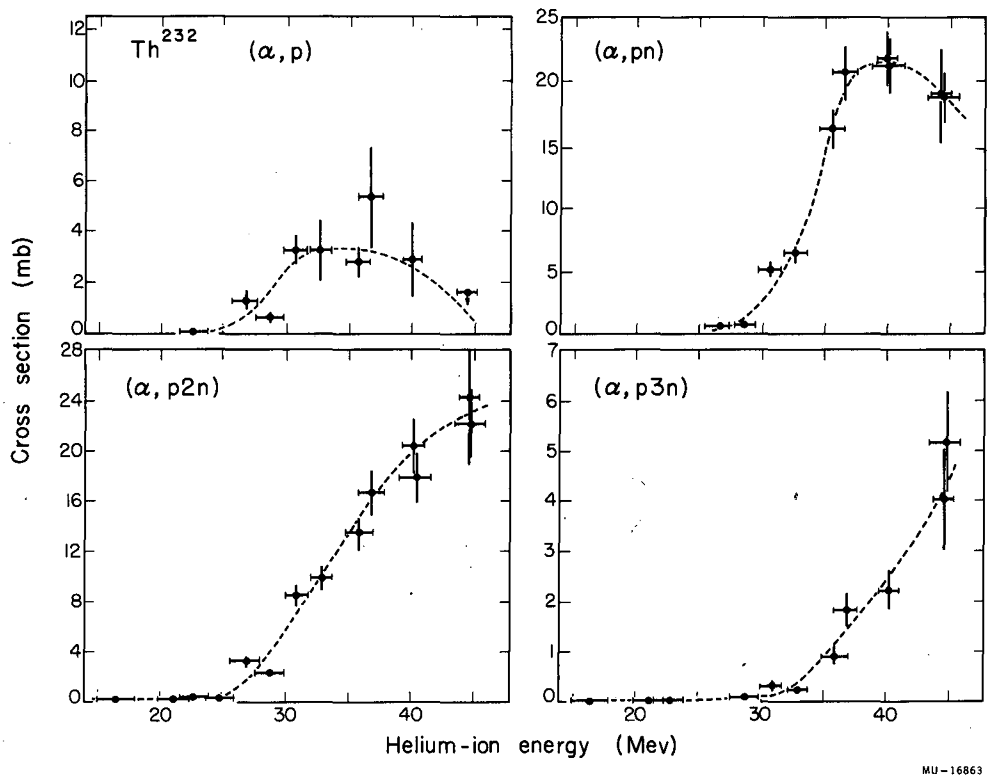


Fig. 1. Spallation excitation functions for the (α,pxn) products of $Th^{232}$ + $He^{4}$. All curves shown are empirical. Only one (α,pn) product isomer was measured.

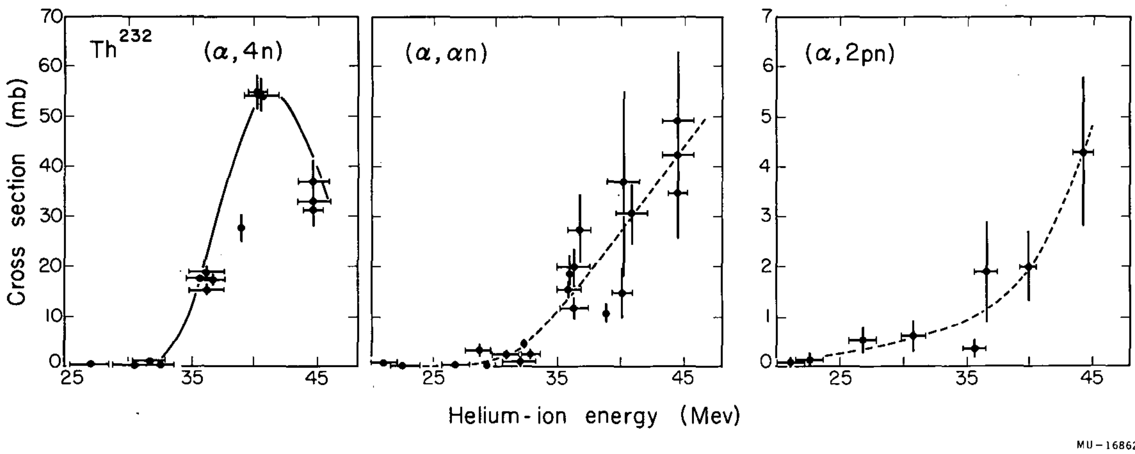


Fig. 2. Spallation excitation functions for the (α,4n) and (α,2pxn) products of $Th^{232}$ + $He^4$. The solid curve is theoretical; the dashed curves are empirical.

Table II. Spallation-product cross sections for $U^{233}$+d (millibarns)

| Reaction | (d,n) | (d,2n) | (d,3n) | (d,αn) |
|---|---|---|---|---|
| Product | $Np^{234}$ | $Np^{233}$ | $Np^{232}$ | $Pa^{230}$ |
| Mode of decay | E.C. | E.C. | E.C. | 8% $\beta^-$, 92% E.C. |
| Half life | 4.4 d | 35 min | 13 min | 17.7 d |
| Proportional-counter efficiency used[a] | 63 ± 2 | 80 ± 20 | 80 ± 20 | |
| Threshold (Mev) | -1.9 | 4.1 | 11.7 | -7.2 |
| Deuteron energy (Mev)[b] | | | | |
| 9.0 | <1.2 | 0.42 ± .06 | | |
| 12.1 | 10.5 ± 1.6 | 9.4 ± 1.9 | | |
| 14.0 | 10.7 ± 1.6 | 10.1 ± 1.3 | | 0.08 ± .03 |
| 15.4 | 13.2 ± 2.0 | 8.7 ± 1.2 | <2.3 | 0.10 ± .04 |
| 19.6 | 11.0 ± 1.7 | 3.7 ± 1.3 | 9.7 ± 2.0 | 0.91 ± .36 |
| 21.5 | 13.8 ± 2.1 | 4.36 ± .59 | 10.9 ± 2.2 | |
| 23.4 | 12.6 ± 1.9 | 5.75 ± .78 | 8.2 ± 1.7 | 1.86 ± .74 |

a. Value for $Np^{234}$ measured; others were estimated.

b. All energy values are uncertain by 0.5 Mev.

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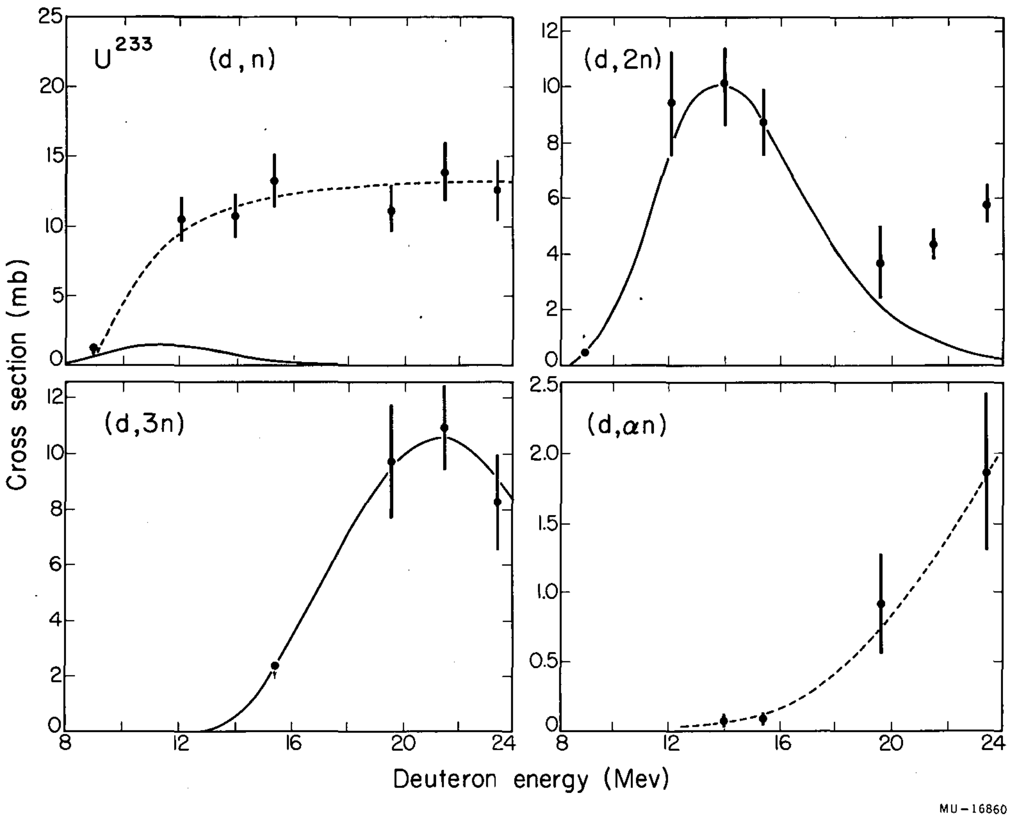


Fig. 3. Spallation-product excitation functions for $U^{233}$ + d. The solid curves are theoretical; the dashed curves are empirical.

Table III. Fission cross sections for $Th^{232}+\alpha$ (millibarns)

| Nuclide | Energy (Mev) 15.6 - 19.6 σ meas. | 15.6 - 19.6 σ mass | 19.8 σ meas. | 19.8 σ mass | 25.7 - 28.9 σ meas. | 25.7 - 28.9 σ mass | 34.9 - 37.7 σ meas. | 34.9 - 37.7 σ mass | 39.6 - 42.2 σ meas. | 39.6 - 42.2 σ mass | 43.4 - 45.9 σ meas. | 43.4 - 45.9 σ mass |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $Zn^{72}$ | | | | | 0.036±.009 | 0.038±.010 | 0.14±.03 | 0.15±.04 | | | 0.34±.07 | 0.37±.09 |
| $Ge^{77}$ | | | | | 0.083±.03 | | 0.040±.02 | | | | 0.58±.14 | 0.58±0.14[b] |
| $As^{77}$[a] | | | | | 0.12±.05 | | 0.37±.11 | | | | ~0 | |
| Total mass 77 | | | | | 0.20±.10 | 0.20±.10 | 0.41±.13 | 0.41±.13 | | | 0.58±.14 | |
| $Se^{82}$[a] | | | | | 0.034±.01 | | 0.30±.006 | | | | | |
| $Br^{83}$ | | | | | | | 10±5 | 10±5 | | | | |
| $Rb^{86}$[a] | | | | | 1.2±.5 | | 0.034±.009 | | | | 0.099±.025 | |
| $Sr^{89}$ | 0.051±.010 | 0.051±.010 | 0.84±.2 | 0.84±.2 | 38±19 | 38±19 | 30±10 | 30±10 | | | 27±6 | 27±6 |
| $Sr^{91}$ | 0.049±.01 | 0.049±.01 | 0.96±.32 | 0.97±.32 | 66±33 | b | 34±7 | 35±8 | | | 30±8 | 30±8[b] |
| $Y^{93}$ | | | | | 7.4±3.7 | 7.4±3.7 | | | | | | |
| $Zr^{95}$ | 0.11±.04 | 0.11±.04 | 1.1±.2 | 1.1±.2 | 7.4±3.7 | b | 65±13 | b | 8.3±1.7 | 8.4±1.7 | 91±18 | 91±18[b] |
| $Zr^{97}$ | 0.058±.02 | 0.059±.02 | 1.1±.2 | 1.1±.2 | 25±5 | 25±5 | 42±8 | 45±9 | 7.7±1.9 | 8.2±2.1 | 73±18 | 76±20 |
| $Nb^{96}$[a] | | | | | | | | | | | 0.45±.15 | |
| $Mo^{99}$ | | | | | 28±6 | 28±6 | 40±8 | 41±9 | | | 56±11 | 56±11 |
| $Ru^{103}$[c] | | | | | 11±2 | 11±2 | 15±4 | 15±4 | | | 24±5 | 24±5 |
| $Ru^{105}$[c] | | | | | | | | | | | 23±8 | 24±8 |
| $Ru^{106}$[c] | | | | | 8.6±1.7 | 8.7±1.8 | 16±3 | 17±4 | | | 31±6 | 32±6 |
| $Pd^{109}$ | | | | | 8.2±2.1 | 8.2±2.1 | 17±3 | 17±3 | | | 38±8 | 38±8 |
| $Pd^{112}$ | | | | | 8.6±2.1 | 8.8±2.2 | 20±4 | 21±4 | | | 33±7 | 36±7 |
| $Ag^{111}$ | | | | | 11±2 | 11±2 | 27±5 | 27±5 | | | 63±13 | 63±13 |
| $Cd^{115}$ | 0.0047±.003 | | 0.30±.2 | | 12±4 | | 24±5 | | | | 54±27 | |
| $Cd^{115m}$ | | | | | 0.79±.2 | | 1.4±.3 | | | | 7.8±3.9 | |
| $Cd^{115}$ (total) | 0.0052±.004[d] | 0.0052±.004 | 0.32±.31[d] | 0.32±.31 | 13±4 | 13±4 | 25±5 | 25±5 | | | 62±31 | 62±33 |
| $I^{130}$[a] | | | | | | | | | | | | |
| $I^{131}$ | | | | | 16±4 | 17±5 | 22±6 | 26±6 | | | | |
| $I^{133}$ | | | | | 26±7 | 32±9 | 34±14 | 52±25 | | | | |
| $Cs^{136}$[a] | | | | | 2.8±.6 | | 7.1±1.4 | | | | 17±4 | |
| $Ba^{139}$ | | | 0.77±.3 | 0.85±.3 | | | | | 42±12 | 50±16 | | |
| $Ba^{140}$ | 0.037±.019 | 0.042±.021 | 0.82±.2 | 0.96±.2 | 25±6 | | 30±6 | 42±8 | 37±7 | 52±11 | 30±8 | |
| $La^{140}$[a] | | | | | 1.7±.5 | | | | | | 3.7±1 | |
| Total mass 140 | | | | | 27±7 | 28±7 | | | | | 34±9 | 38±10 |
| $Ce^{141}$ | | | | | | | | | | | 55±11 | 58±13 |
| $Ce^{143}$ | | | | | | | | | | | 57±14 | 57±14[b] |
| $Ce^{144}$ | | | | | | | | | | | 27±5 | 35±8 |
| $Nd^{147}$ | | | | | | | | | | | 11±4 | 12±5 |
| Total fission cross section | 0.93±.24 mb | | | | 590±150 mb | | 930±240 mb | | | | 1660±410 mb | |

a. Shielded nuclide (one whose precursor in the $\beta^-$ decay chain is either stable or long-lived).

b. In these cases the cross section for formation of the daughter was measured and found negligible.

c. Not shown in Fig. 4 because of isotopic-exchange difficulties which make ruthenium cross sections unreliable.

d. Contribution from $Cd^{115m}$ estimated.

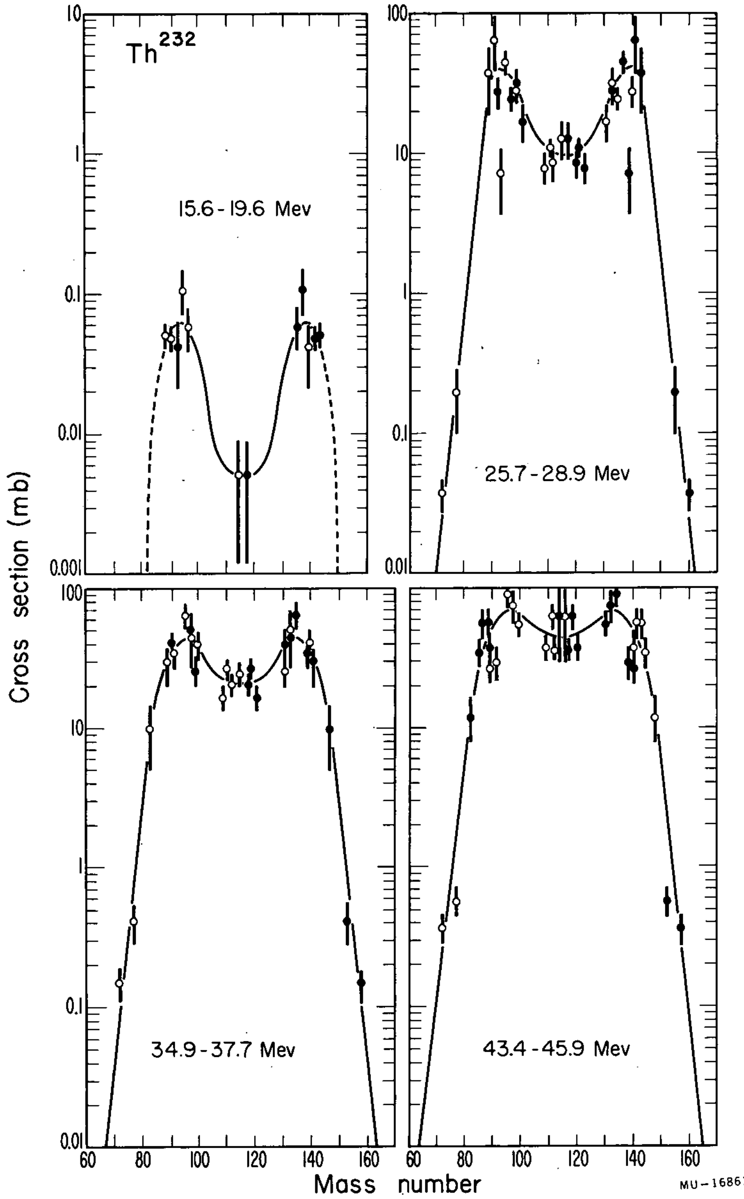


Fig. 4. Fissión mass-yield curves for $Th^{232} + He^{4}$. Points: ○, corrected cross sections; ●, cross sections for complementary (reflected) fission products obtained with the assumption of 3 to 7 neutrons emitted [15.6 - 19.6 Mev (3), 25.7 - 28.9 Mev (4), 34.9 - 37.7 Mev (6), 43.4 - 45.9 Mev (7)].

The measured and corrected fission-product cross sections for $U^{233}$ are listed in Table IV and shown in Fig. 5. Very fast chemical procedures were required to separate and characterize the short-lived neptunium isotopes produced in the bombardments, particularly at the highest energies. Consequently, some of the fission-product decay chains were broken before sufficient time had elapsed for all the short-lived predecessors of some of the fission products to decay. Corrections for this effect were applied where possible. The limits of error are estimated to be about 20 to 30%.

Table IV. Fission cross sections for $U^{233}+d$ (millibarns)[a]

| | Deuteron energy (Mev)[b] | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 9.0 | | 12.1 | | 14.0 | | 15.4 | | 19.6 | | 21.5 | | 23.4 | |
| Isotope | σ meas. | σ mass | σ meas. | σ mass | σ meas. | σ mass | σ meas. | σ mass | σ meas. | σ mass | σ meas. | σ mass | σ meas. | σ mass |
| $Sr^{89}$ | 4.95 | 4.64 | 14.4 | 14.6 | 18.9 | 19.1 | 26.4 | 26.7 | 46.5 | 48.2 | 34.6 | 35.1 | 42.7 | 43.3 |
| $Sr^{91}$ | 3.90 | 4.01 | 34.1 | 34.8 | 18.7 | 19.2 | 31.0 | 32.0 | 36.9 | 38.0 | 52.5 | 54.6 | 44.3 | 46.2 |
| $Zr^{95}$ | 6.45 | 6.55 | | | 44.8 | 45.6 | | | 58.5 | 59.7 | 61.6 | 63.0 | 54.2 | 55.5 |
| $Zr^{97}$ | 4.59 | 4.80 | 25.6 | 26.8 | 39.3 | 41.1 | | | 49.8 | 52.4 | 61.8 | 66.0 | 53.5 | 57.2 |
| $Ru^{105}$ | 5.03 | 5.11 | | | | | | | | | | | | |
| $Pd^{109}$ | | | | | | | | | | | 46.7 | 47.5 | | |
| $Pd^{112}$ | | | | | | | | | | | 52.9 | 56.9 | | |
| $Cd^{115}$ | 1.22 | 1.24 | 8.43 | 8.57 | 14.7 | 15.0 | 22.9 | 23.4 | 44.0 | 45.0 | 47.3 | 48.6 | 63.9 | 65.7 |
| $Cd^{115m}$[c] | | | 1.55 | 1.58 | 1.97 | 2.0 | 3.92 | 4.00 | 5.58 | 5.70 | 8.5 | 8.7 | 4.8 | 4.9 |
| $Cd^{115}+Cd^{115m}$ | 1.34 | 1.36 | 9.98 | 10.2 | 16.1 | 17.0 | 26.8 | 27.4 | 49.6 | 50.7 | 55.8 | 57.3 | 68.7 | 70.6 |
| $Cd^{117}$ | 1.70 | 1.78 | 12.1 | 12.5 | 19.5 | 20.4 | 21.9 | 23.2 | 41.8 | 44.2 | 50.0 | 53.8 | 59.0 | 63.5 |
| $Ba^{139}$ | 4.26 | 4.82 | 20.4 | 23.1 | | | 39.3 | 46.2 | 49.2 | 57.9 | 46.5 | 58.1 | 34.0 | 45.3 |
| $Ba^{140}$ | 3.50 | 4.27 | 17.1 | 20.9 | 21.8 | 26.6 | 29.2 | 41.7 | 37.5 | 53.6 | 35.5 | 53.0 | 31.5 | 47.0 |
| $Nd^{147}$ | | | | | | | | | | | | | 12.2 | 13.3 |
| $Eu^{157}$ | | | | | | | | | | | | | 0.76 | 1.03 |
| $Gd^{159}$ | | | | | | | | | | | | | 0.54 | 0.70 |
| Total fission cross section | 125 ± 34 | | 605 ± 163 | | 857 ± 231 | | 1093 ± 295 | | 1502 ± 406 | | 1687 ± 456 | | 1861 ± 503 | |

a. ± 20 to 30%.

b. ± 0.5 Mev.

c. Upper limit.

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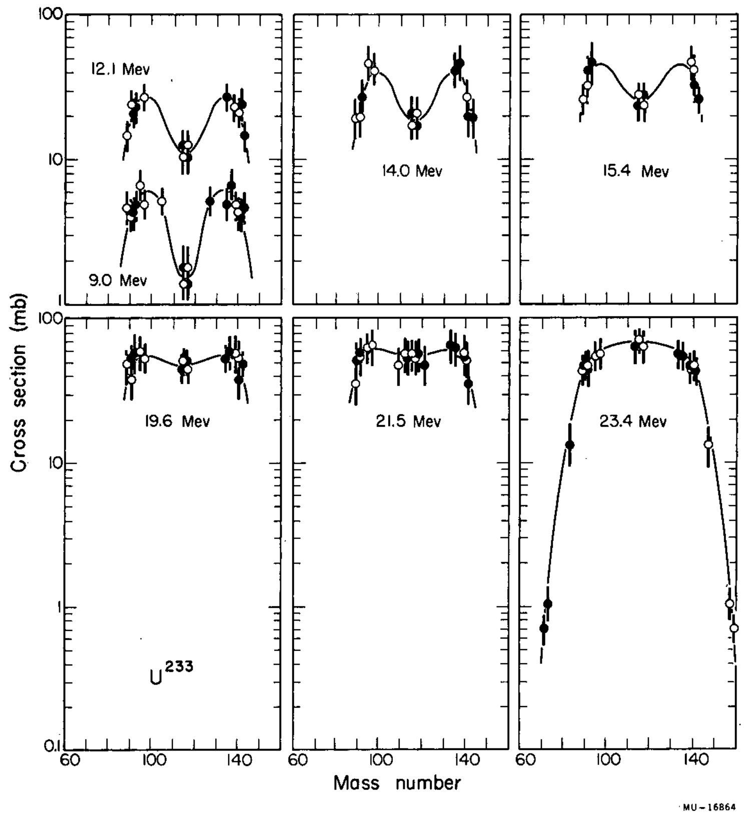


Fig. 5. Fission mass-yield curves for $U^{233}$ + d. Points: ○, corrected cross sections; ●, cross sections for complementary fission products obtained with the assumption of 3 to 5 neutrons emitted [9.0 Mev (3), 12.1 Mev (3), 14.0 Mev (3), 15.4 Mev (4), 19.6 Mev (4), 21.5 Mev (5), 23.4 Mev (5)].

## IV. DISCUSSION

Previous studies[1-5] of spallation-fission competition in the heaviest elements have indicated that the ($\alpha$,2n), ($\alpha$,3n), and ($\alpha$,4n) reaction products arise principally from de-excitation of the compound nucleus and are drastically reduced by the competition of the fission reaction, while certain other reactions such as the ($\alpha$,p2n) appear to be due to non-compound-nucleus processes and are relatively unaffected by fission competition. The results of this study are discussed in terms of the same general concepts.

### A. Compound-Nucleus Spallation Reactions

The general features of the excitation functions for the reactions involving only neutron emission are similar to those noted for other heavy elements.[1-3,5-10,22-26]

Let us first consider the shape of the excitation functions for reactions involving only neutron emission. The solid curves on the graphs of the excitation functions for the ($\alpha$,4n) reaction on $Th^{232}$ in Fig. 2 and the (d,n), (d,2n), and (d,3n) reactions on $U^{233}$ in Fig. 3 were calculated according to the modified Jackson compound-nucleus model described by Vandenbosch et al.[5] using a nuclear temperature of 1.35 Mev (in agreement with previous studies).[5] The fit of the shape (the normalization is discussed below) of the theoretical curves to the experimentally determined excitation functions for the ($\alpha$,4n) and the (d,3n) reactions indicates that these reactions do indeed proceed by a compound-nucleus mechanism. The excitation function for the (d,2n) reaction indicates that in the region of maximum cross section the reaction proceeds predominantly by a compound-nucleus mechanism. At higher energies an increasing direct-interaction contribution is suggested by the fact that the observed cross sections are higher than calculated.

The very large reduction in the magnitude of the cross sections for the ($\alpha$,4n), (d,2n), and (d,3n) reactions observed in this study, as compared with the cross sections for heavy nuclei that are nonfissionable,[12,13] shows the effect of fission competition and also suggests that these reactions proceed primarily by compound-nucleus de-excitation. The magnitude of the maximum cross section for $(\alpha,4n)$ reactions on various nuclides has been used as a sensitive measure of the fissionability of those nuclides.[39] In order to determine quantitatively the degree of fission competition we define (following Vandenbosch et al.[5]) the neutron branching ratio $G_n$ as

$$G_n = \frac{\Gamma_n}{\Sigma_i \Gamma_i}$$

where $\Gamma_n$ is the probability (level width) for neutron emission and $\Sigma_i\Gamma_i$ contains terms for all possible modes of decay for the compound nucleus. In practice $\Sigma_i \Gamma_i$ is assumed to contain only $\Gamma_n$ and $\Gamma_f$ terms, where $\Gamma_f$ is the level width for fission. A geometric mean value $\overline{G}_n$ is obtained from the normalization factor necessary to fit the magnitude of the theoretical curves based on the midified Jackson model[5] to the experimental cross sections. The values of $\overline{G}_n$ used to normalize the $(\alpha,4n)$, (d,2n), and (d,3n) curves are 0.49, 0.17, and 0.20 respectively.[40] Within the limits of experimental error, the average neutron-branching ratios of the neptunium and uranium isotopes produced as intermediates in this study are consistent with the considerations of Vandenbosch and Seaborg.[39]

B. Fission

Figures 6 and 7 show the total fission yields from helium-ion reactions on $Th^{232}$ and deuteron reactions on $U^{233}$. These were determined by summing under the curves of Figs. 4 and 5. The dashed lines above the points represent the total cross sections and include the measured and estimated[41] spallation cross sections in addition to the measured fission cross sections.

The solid curves in Figs. 6 and 7 show the theoretical compound-nucleus-formation cross sections of Blatt and Weisskopf[42] (in Fig. 6) and Shapiro[43] (in Fig. 7) for nuclear radius parameters $r_o = 1.3 \times 10^{-13}$ cm. and $r_o = 1.5 \times 10^{-13}$ cm, where the nuclear radius is given by $R = r_o A^{1/3}$. It can be seen that the best fit to the experimental data is given in both cases by $r_o \sim 1.5 \times 10^{-13}$ cm. This is in agreement with the results of studies on heavier nuclides.[1,5,30] For deuterons on $U^{233}$ the experimental points at the lower energies fall above the theoretical curves. This is well outside the experimental error, and is not noted in cases in which helium nuclei are the

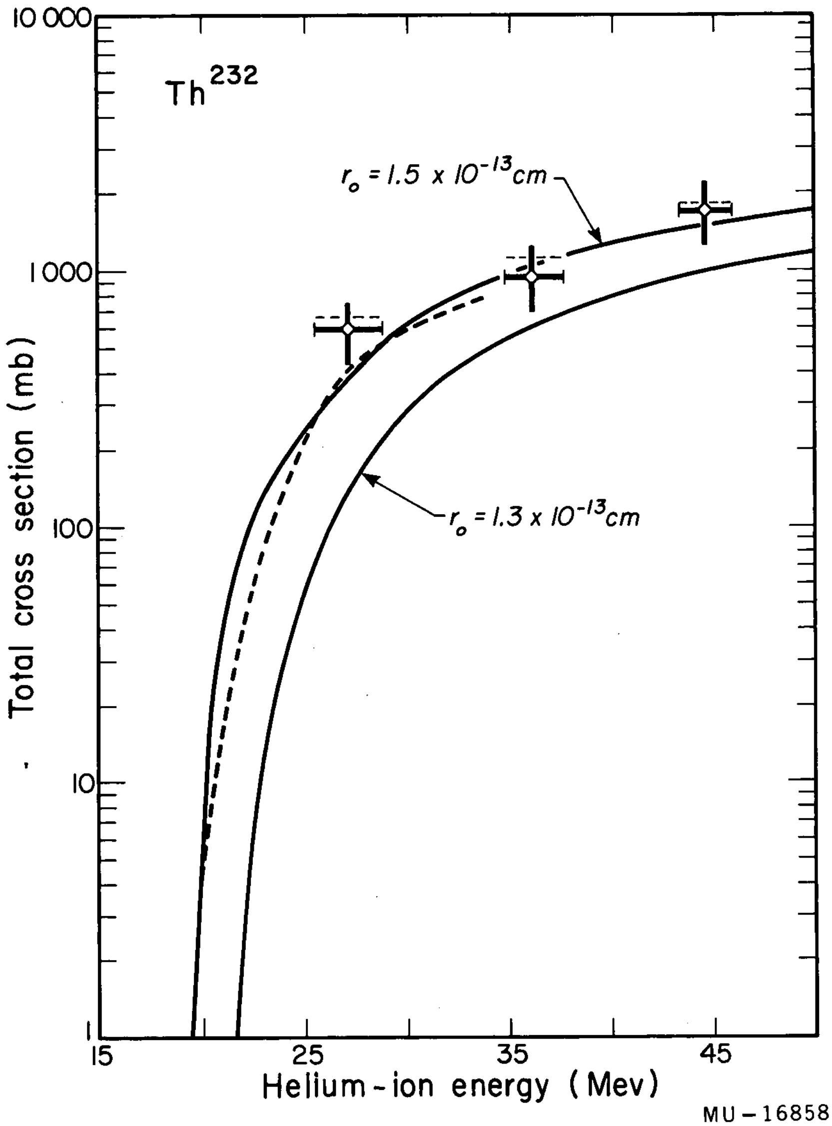


Fig. 6. Total reaction excitation function for $Th^{232}$ + $He^4$. Diamonds represent total fission cross section and dashed bars represent empirical total reaction cross sections. The dashed curve represents ionization-chamber measurements of total fission cross section from Ref. 16. The solid curves are theoretical.

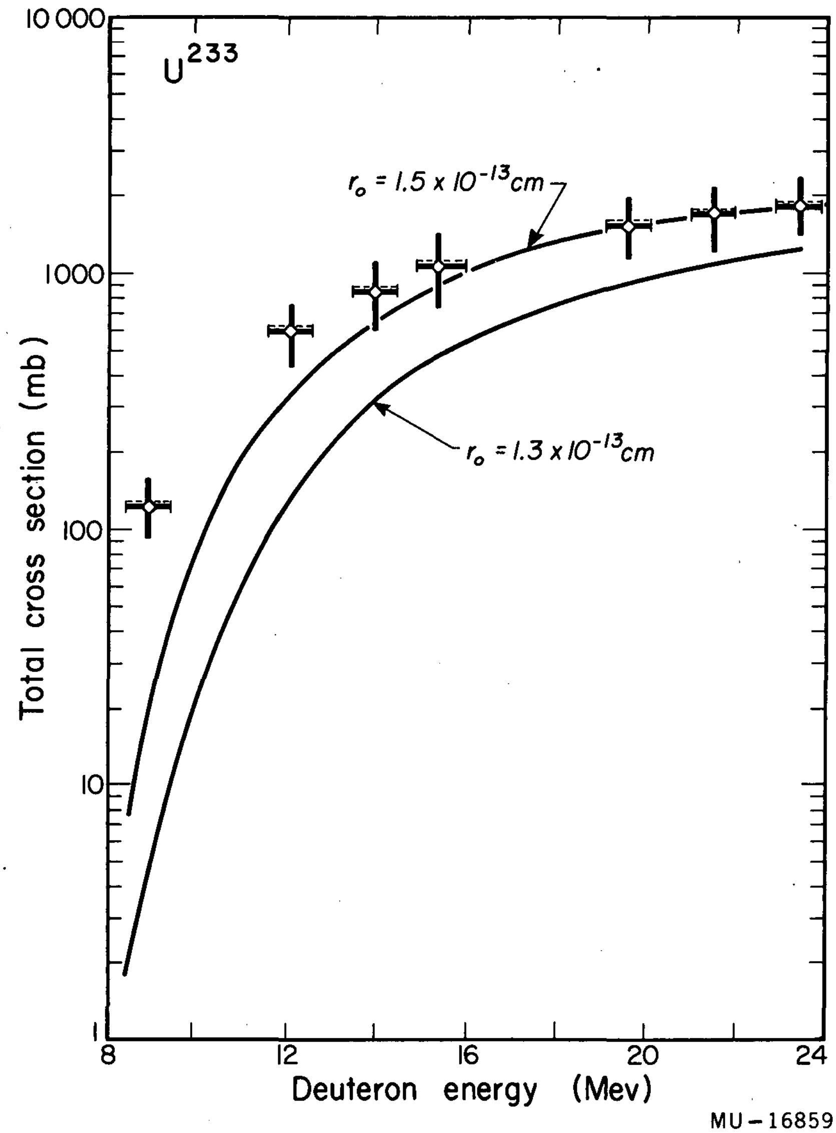


Fig. 7. Total reaction excitation function for $U^{233}$ + d. Diamonds represent total fission cross sections and dashed bars (above diamonds) represent empirical total reaction cross sections. The solid curves are theoretical.

incident particles. This deviation is probably due to the contribution from (d,p) stripping[20,21,25] followed by fission at the lower energies.

Although the compound systems studied here are intermediate between the very heavy region, where fission is predominent, and the lighter region, where fission is a minor contributor, it is apparent from Figs. 6 and 7 that fission still accounts for most of the geometric cross section. In fact, for deuterons on $U^{233}$ the fission branching ratio ($\Gamma_f/\Gamma_t \cong 1 - G_n$) is even higher than for some heavier nuclides.[5,39] In thorium the average fission branching ratio is lower than observed for most heavier nuclides by about 0.2. Even here, however, fission accounts for most of the total cross section.

The fission mass-yield curves from this study show the same general trends as were observed in helium-ion bombardments on uranium[5] and plutonium isotopes.[1] Predominantly asymmetric fission occurs at the lowest energies and the degree of asymmetry decreases rapidly as the energy of the incident particle is increased. The asymmetry characteristics of $U^{233}$ fission are similar to those of many of the heavier nuclides.[1,5,30] It is perhaps significant that $Th^{232}$, on the other hand -- a less fissionable nuclide -- appears to undergo more asymmetric fission for a given excitation energy. The fission-yield curves in Fig. 4 are similar in shape to those by Newton[17] for 38.5-Mev helium-ion bombardment of $Th^{232}$, but the cross sections are higher (for comparable energies) by a factor of about two. However, Newton's measurement of the cyclotron current was probably uncertain by a factor of at least two.[44] No indication of the unusual characteristics noted for the fission mass-yield curves of $Ra^{226}$ [15] and $Bi^{209}$ [14] was apparent in these studies, but it should be noted that the sensitivity of the measurements is not high, since the determination of detailed fission yields was not a primary aim of these experiments.

## C. Direct Interactions

High-energy nuclear reactions are generally interpreted in terms of a two-stage process in which the first stage is a "cascade" involving direct interactions between the incoming particle and individual particles in the nucleus. The second stage consists of evaporation of nucleons from the residual nucleus in a manner similar to that of a compound-nucleus reaction. It is of interest to examine the question whether a two-step process of this sort can occur in the energy region considered here. Previous papers in the present series have already given considerable evidence that both direct-interaction and evaporation processes do indeed take place.

The fact that cross sections for the (α,2p) reaction on $Th^{232}$ are extremely small below 50 Mev leads one to hypothesize that direct interactions in which two particles of any type are emitted do not occur with appreciable probability. This is consistent with Monte Carlo calculations, which indicate that below about 100 Mev absolute values of the cross sections for multiple-particle cascades are small,[45] while direct interactions in which only one particle is ejected have appreciable probability. These Monte Carlo calculations were done on the basis of protons impinging on $U^{238}$, but the systems studied in the work reported herein should behave similarly so far as general trends are concerned.

The mechanism for emission of charged particles is probably a cascade or direct ejection and not evaporation of protons (or other charged particles). This is suggested by comparing the cross sections for reactions involving charged-particle emission with the cross sections for reactions involving only neutron emission. In particular the excitation function for the (α,p) reaction (in Fig. 1) is comparable to that calculated for the (α,n) reaction (on the basis of the modified Jackson model mentioned above[5]) below 25 Mev, and much greater above 25 Mev. If the proton were evaporated from a compound nucleus the cross section for the (α,p) reaction would be affected by the Coulomb barrier and would be much smaller than the cross section for the (α,n) reaction over the entire energy range. We may therefore reasonably assume that the (α,pxn) reactions observed in this work proceed by single-particle ejections followed (in some cases) by evaporation of neutrons.

The observed yield of the (α,p2n) product can be explained by the ejection of tritons.[4] However, as will be shown later, a small contribution from (α,dn) reactions at the highest energies is probable.

Most of the (α,pn) reaction probably proceeds by deuteron ejection, which leaves the nucleus with insufficient energy either to emit a neutron or to undergo fission. This conclusion is based on the fact that yields for the (α,pn) reaction change gradually from one element to another[46] without any discontinuity between the heavy-element region in which fission is the predominant reaction and the slightly lower region where fission is not important. If the mechanism were largely proton emission followed by neutron evaporation, the cross sections for the $(\alpha,pn)$ reaction would decrease suddenly where fission begins to compete favorably with evaporation. The decrease in the excitation function for formation of the $(\alpha,pn)$ product at the highest energies indicates that some of the residual nuclei from deuteron emission are highly enough excited to be removed by fission or by evaporation of a neutron. With neutron evaporation this process would make some small contribution to the observed cross section for formation of the $(\alpha,p2n)$ reaction product.

If the $(\alpha,pxn)$ products result from mechanisms in which a proton, deuteron, or triton is ejected followed by neutron evaporation, the following expressions for the cross sections hold:

$$\sigma\ (\alpha,p) = \begin{cases} \sigma^*\ (\alpha,p) & \text{for } E_\alpha + Q\ (\alpha,p) < E_a^1 , \\ \sigma^*\ (\alpha,p) \displaystyle\int_0^{E_a^1} N\ (E_p)\ d\ E_{ex}^1 & \text{for } E_\alpha + Q\ (\alpha,p) > E_a^1 ; \end{cases} \tag{1}$$

$$\sigma\ (\alpha,pn) = \sigma^*\ (\alpha,p)\ G_n^1 \int_{B_n^1}^{E_\alpha + Q\ (\alpha,p)} N\ (E_p)\ P\ (E_{ex}^1,1)\ d\ E_{ex}^1$$

$$+ \begin{cases} \sigma^*\ (\alpha,d) & \text{for } E_\alpha + Q\ (\alpha,d) < B_n^2 , \\ \sigma^*\ (\alpha,d) \displaystyle\int_0^{B_n^2} N\ (E_d)\ d\ E_{ex}^2 & \text{for } E_\alpha + Q\ (\alpha,d) > B_n^2 ; \end{cases} \tag{2}$$

$$\sigma\ (\alpha,p2n) = \sigma^*\ (\alpha,p)\ G_n^1\ G_n^2 \int_{B_{2n}^1}^{E_\alpha + Q\ (\alpha,p)} N\ (E_p)\ P\ (E_{ex}^1,2)\ d\ E_{ex}^1$$

$$+\ \sigma^*\ (\alpha,d)\ G_n^2 \int_{B_n^2}^{E_\alpha + Q\ (\alpha,d)} N\ (E_d)\ P\ (E_{ex}^2,1)\ d\ E_{ex}^2 \qquad (3)$$

$$+ \begin{cases} \sigma^*\ (\alpha,t) & \text{for } E_\alpha + Q\ (\alpha,t) < E_a^3\ , \\ \sigma^*\ (\alpha,t) \displaystyle\int_o^{E_a^3} N\ (E_t)\ d\ E_{ex}^3 & \text{for } E_\alpha + Q\ (\alpha,t) > E_a^2\ ; \end{cases}$$

$$\sigma\ (\alpha,p3n) = \sigma^*\ (\alpha,p)\ G_n^1\ G_n^2\ G_n^3 \int_{B_{3n}^1}^{E_\alpha + Q\ (\alpha,p)} N\ (E_p)\ P\ (E_{ex}^1,3)\ d\ E_{ex}^1$$

$$+\ \sigma^*\ (\alpha,d)\ G_n^2\ G_n^3 \int_{B_{2n}^2}^{E_\alpha + Q\ (\alpha,d)} N\ (E_d)\ P\ (E_{ex}^2,2)\ d\ E_{ex}^2 \qquad (4)$$

$$+\ \sigma^*\ (\alpha,t)\ G_n^3 \int_{B_n^3}^{E_\alpha + Q\ (\alpha,t)} N\ (E_t)\ P\ (E_{ex}^3,1)\ d\ E_{ex}^3\ ,$$

where $\sigma^*\ (\alpha,p)$, $\sigma^*\ (\alpha,d)$, and $\sigma^*\ (\alpha,t)$ are the cross sections for the primary ejection of protons, deuterons, and tritons respectively, where $E^i_{ex}$ is the excitation energy of the residual nucleus $Pa^{236-i}$ after the cascade,

$E^i_a$ is the activation energy for fission[39] of $Pa^{236-i}$,

$N(E_p)$, $N(E_d)$, and $N(E_t)$ are the probabilities that the protons, deuterons, and tritons respectively will be emitted with energy E (normalized so that $\int_o^{E_\alpha + Q} N \, d \, E_{ex} = 1$,

$E_p = E_\alpha + Q(\alpha,p) - E^1_{ex}$,

$E_d = E_\alpha + Q(\alpha,d) - E^2_{ex}$,

$E_t = E_\alpha + Q(\alpha,t) - E^3_{ex}$,

$B^i_{jn}$ is the binding energy of the last j neutrons in $Pa^{236-i}$,

$G^i_n$ is the ratio $\Gamma_n/\Gamma_t$ for $Pa^{236-i}$ (assumed independent of excitation energy,

$P(E^i_{ex},j)$ is the probability of evaporating exactly j neutrons from a $Pa^{236-i}$ nucleus with initial excitation energy $E^i_{ex}$

[$p(E^i_{ex},j)$ can be calculated by the modified Jackson method[47]],

$Q(\alpha,p) = -11.66$ Mev,[48]

$Q(\alpha,d) = -15.51$ Mev,[48]

$Q(\alpha,t) = -14.42$ Mev,[48]

$E_\alpha$ is the energy of the incident helium ion in the center-of-mass system.

In order to compare these expressions rigorously with experimental data, we need, but do not have, information on the cross sections for the primary ejection processes and the spectra of outgoing particles. However, some information on the energy spectra of ejected tritons can be obtained by the use of Eqs. (3) and (4) with the following approximations:

(a) Contributions to the observed cross section for the $(\alpha,p2n)$ product from sources other than direct triton ejection can be neglected. The results of Wade et al.[4] support this assumption.

(b) The observed yield for the $(\alpha,p3n)$ product is due almost entirely to triton ejection followed by neutron evaporation. This is consistent with Assumption (a). Furthermore, the only other process that could contribute appreciably to the observed yield for this reaction is the $(\alpha,d2n)$ reaction, whose yield is even more drastically reduced by fission competition than is that of the $(\alpha,tn)$ process.

(c) The function $P(E^3_{ex},l)$ can be approximated by a constant value, independent of energy, over a fixed range of excitation energy. That this is a reasonably good approximation can be seen from Jackson's graph of a similar function.[47]

With the above approximations, in the region where the $(\alpha,tn)$ reaction is energetically permitted, we may write Eqs. (3) and (4) as follows:

$$\sigma\ (\alpha,t) = \sigma^*\ (\alpha,t) \int_0^{E^3_a} N\ (E_t)\ d\ E^3_{ex}\ , \tag{3'}$$

$$\sigma\ (\alpha,tn) = \sigma^*\ (\alpha,t)\ F \int_{B^3_n}^{E_\alpha + Q\ (\alpha,t)} N\ (E_t)\ d\ E^3_{ex}\ , \tag{4'}$$

where F is a combination of $G^3_n$ and the constant $P\ (E^3_{ex},l)$ assumed above. Its numerical value (based on the branching-ratio systematics of Lessler,[23] is 0.65. Combining Eqs. (3') and (4') we obtain

$$\frac{\sigma\ (\alpha,tn)}{\sigma\ (\alpha,t)} = 0.65\ \frac{\int_{B^3_n}^{E_\alpha + Q\ (\alpha,t)} N\ (E_t)\ d\ E^3_{ex}}{\int_0^{E^3_a} N\ (E_t)\ d\ E^3_{ex}}\ . \tag{5}$$

The $N\ (E_t)$ functions in Eq. (5) represent the emitted triton spectra for each reaction shown, therefore the integrals include all tritons emitted in each case. Measurement of the $\sigma\ (\alpha,tn)/\sigma\ (\alpha,t)$ ratio therefore allows one to calculate the fraction of the emitted tritons which are of high enough energy that the residual nucleus is left with insufficient excitation energy to evaporate a neutron or to undergo fission. The experimental values of $\sigma\ (\alpha,tn)/\sigma\ (\alpha,t)$ taken from the smooth curves of Fig. 1 are shown in Fig. 8. From Fig. 8 and Eq. (5) we calculate that at 35 Mev about 9% of the outgoing tritons are of sufficiently low energy that the residual nucleus is left with less than 6.5 Mev of excitation energy (i.e., triton energy $\nless$ 13.9 Mev), while at 44 Mev about 20% of the energing tritons leave the nucleus with more than 6.5 Mev of excitation energy (triton energy $<$ 22 Mev).

Since the cross sections for the $(\alpha,2p)$ reaction (see Fig. 2) are very small below 46 Mev, the cross section for the emission of two protons and a neutron is probably also very small. Thus the only plausible mechanism for the formation of the observed $(\alpha,2pn)$ product is direct emission of a $He^3$ nucleus. Possibly the mechanism for this reaction, as well as the $(\alpha,t)$ reaction, is one in which a single nucleon is stripped from the incident helium nucleus.

The $(\alpha,\alpha n)$ reaction[49] can be described by a mechanism involving inelastic scattering of the incident helium ion followed by neutron evaporation. It can also be described as a collision between the incident helium ion and a neutron, after which both escape from the nucleus. The latter process would be unique in this energy range ( $<$ 50 Mev), since it involves direct ejection of more than one particle. A determination of the excitation function for the $(\alpha,\alpha p)$ reaction would help to differentiate between the two possible mechanisms noted above. The cross sections for the (d,n) product (Fig. 3) are higher than those for the (d,2n) product over the entire energy range studied. This is the first time that this has been observed and illustrates in a striking manner both the fissionability of the $Np^{235*}$ compound system and the fact that the (d,n) reaction has little contribution from compound-nucleus processes. The magnitude of the compound-nucleus contribution is indicated by the theoretical curve of Fig. 3. The observed cross section is probably due almost entirely to a stripping mechanism which leaves the nucleus with insufficient energy to emit a neutron or to

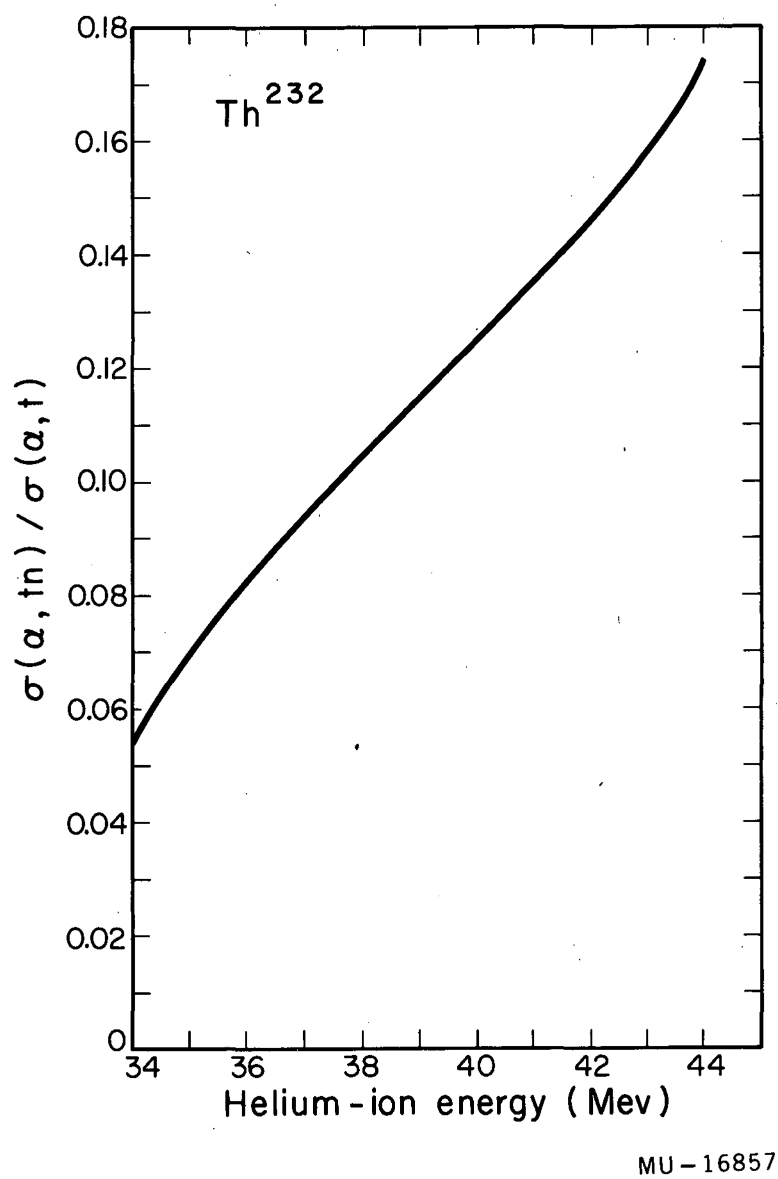


Fig. 8. Experimental ratio of the cross sections for the $(\alpha,tn)$ reaction to that for the $(\alpha,t)$ reaction for $Th^{232} + He^4$.

undergo fission. The shape and magnitude of the excitation function for this reaction are similar to those observed for the heaviest nonfissionable nuclides and to those predicted by Peaslee for a stripping reaction.[50] The high cross sections for the (d,2n) reaction at the highest energies studied (Fig. 3) are probably due to (d,n) stripping followed by evaporation of one neutron.

The mechanism for the (d,$\alpha$n) reaction on $U^{233}$ is not clear. The most plausible mechanism is an ejection of an alpha particle followed by evaporation of a neutron. The ejection may take place either through pickup of a neutron and a proton by the incident deuteron or by "knock-on" of an alpha particle with capture of the incident dauteron by the residual nucleus. Some support for the proposed double pickup process comes from the measurement of sizable cross sections for the (p,t) reaction on heavy nuclides by Wade et al.[4]

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27. In each case the cross section indicated is the cross section for formation of a particular product nuclide. Therefore, except for special cases [such as $(\alpha,\alpha n)$, $(d,\alpha n)$] in which the reaction indicated is the only one energetically possible, the reactions indicated should be regarded as general and do not imply the order or arrangement of the emitted particles.

For example, the indicated ($\alpha$,p2n) reaction could contain contributions from the ($\alpha$,p2n), ($\alpha$,dn), and ($\alpha$,t) reactions, since all lead to the same product.

28. See also the fission-yield curves in References 17 through 20, 22, and 23.

29. For details of the experimental procedures see References 30 and 31.

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