We now combine Eqs. (5) and (6) to obtain Eq. (4) subject only to the conditions which remain on Eq. (6). The results are readily extended to the case where the pth gamma ray is not common to both pairs but the n-p and m-q pairs are considered and $L_p=L_q$; $L_n=L_m$.

We have, thus, demonstrated the following theorem: In a multiple cascade the angular correlations between a gamma ray and each of two following or preceding gamma rays are equal when the following conditions are satisfied: (1) The two observed gamma rays not common to both pairs must have the same multipolarity and must result from basic transitions. (2) If the two observed gamma rays not common to both pairs have unobserved gamma rays between them, these unobserved gamma rays must come from basic transitions. (3) The transitions subject to the first two conditions must take place between levels with spins in monotonic sequence.

There are no restrictions on the multipolarity and mixing of unobserved gamma rays which are between the observed members of both pairs or of a gamma ray observed as a common member of each pair. Also, there are no restrictions on the spins of levels which are involved only in these unrestricted transitions.

The application of the theorem to the three-gamma-ray cascade of Pb^{204m} is of interest. The last two gamma rays of the cascade have been identified as $E2$ transitions. The obvious 4–2–0 spin assignment for the lower levels of the cascade will then result in identical angular correlation functions for the first two and the first and third gamma rays if both $E2$ transitions are unmixed. As the observed anisotropies are significantly different[4] and the transition to the ground (spin 0) state cannot be mixed, the second gamma ray must be a mixed ($E2+M3$) transition, or an alternate spin scheme is required.

We wish to thank Professor F. Coester for the use of his notes and for helpful suggestions.

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Excitation Functions for Some Proton-Induced Reactions of Thorium*

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The excitation functions for the (p,f), (p,n), and $(p,3n)$ reactions on thorium have been determined for proton energies up to 32 Mev.

INTRODUCTION

PREVIOUS work on the (p,f), (p,n), and $(p,3n)$ reactions on thorium has indicated that the (p,xn) processes account for much of the total proton cross section at all energies up to 20 Mev.[1]

However, as yet, no absolute cross sections have been published for these reactions, although some theoretical predictions have been made.

The work reported here was undertaken to ascertain the absolute cross sections of the (p,n) and $(p,3n)$ reactions at various energies up to 32 Mev and to allow estimates of the (p,f) cross section to be made over this energy range.

PROCEDURE

A single bombardment of a stack of 2-mil thorium foils was made on the University of California Radiation Laboratory linear accelerator. The energy of the protons as a function of depth in the stack was calculated from previously determined range-energy relationships. The irradiation was carried out over a twelve-hour period with a proton current averaging 10^{-2} microampere. The beam was integrated with a standard electrometer assembly available for routine use with the linear accelerator.

The thorium foils were dissolved in a mixture of hydrochloric and hydrofluoric acids; a few hundred counts of Pa^{231} tracer were added to each sample, as were 10 mg of zirconium carrier. The chemistry used to separate and purify the zirconium and protactinium fractions has been outlined in the appendix.

The proton-induced fission cross sections were calculated from the yields of Zr^{97}, since the fission yields of Zr^{97} have previously been determined for thorium which has been irradiated with protons up to 20-Mev energy.[1]

The ZrO_2 counting samples were slurried into one-inch-diameter aluminum "hats" and counted, using a standard Geiger tube (a thin-window, argon-filled, chlorine-quenched Amperex 100 C tube) and scaling circuit. Since the self-scattering, self-absorption corrections for the Zr^{97}-Nb^{97} combination are known,[2] the absolute counting rates were determined for the Zr^{97}

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[1] H. A. Tewes and R. A. James, Phys. Rev. 88, 860 (1952).
[2] R. E. Batzel (private communication).

TABLE I. Measured cross sections for proton-induced reactions of thorium.

E_p (Mev)	Fission yield of Zr^{97} [a] (percent)	$\sigma(p,f)$ (mb)	$\sigma(p,n)$ (mb)	$\sigma(p,3n)$ (mb)
31.3±0.4	3.5±0.2	1080 ±70	28.6 ±3	106 ± 5
26.1±0.4	3.5±0.2	910 ±60	21.4 ±2.5	402 ±38
20.8±0.5	3.6±0.1	680 ±40	19.5 ±1.5	324 ±12
18.0±0.5	4.1±0.1$_5$	375 ±20	19.9 ±2.0	47 ± 5
14.9±0.6	4.2±0.1	130 ± 8	13.1 ±1.5	5.0± 1
12.1±0.7	4.3±0.2	26 ± 2	6.4 ±0.7	
9.6±0.9	4.1±0.2	9.2± 0.5	0.85±0.07	
7.7±1.0			0.30±0.04	
5.3±1.2			0.39±0.05	

[a] Zr^{97} fission yields for proton energies below 21 Mev are given in reference 1; those yields above 21 Mev are estimated.

samples mounted on one-inch-diameter platinum disks; the beta decay of Pa^{232} was followed on the same counter used for the Zr^{97} counting. The scattering and geometry corrections were made by preparing a weightless Pa^{232} sample on a thin zapon backing, and comparing its counting rate in a proportional counter having 4π geometry with its counting rate as determined (with platinum backing) on the Geiger tube used in this work.

The absolute alpha-decay rates of the protactinium samples were found by counting them in an ionization chamber with a known (52 percent) geometry; a 50-channel pulse analyzer was then used to determine the energy spectrum of the alpha particles, and hence, the fractions of the alpha particles having energies characteristic of different protactinium and uranium isotopes. The amount of Pa^{230} present was calculated from the decay rate of its daughter (U^{230}) which emits 5.85-Mev alpha particles; the chemical yield of Pa^{231} was calculated from the counting rate found for its ~5.0-Mev alpha particles.

RESULTS

The cross sections for the (p,f), (p,n), and $(p,3n)$ reactions at various proton energies are given in Table I; the fission yields for Zr^{97} which were used in the calculation of the (p,f) cross sections are also noted. The excitation functions for the three reactions under investigation are shown in Fig. 1.

Because the reactions $(p,2n)$, (p,pn), and $(p,p2n)$ were not studied, the total proton absorption cross section for thorium could not be calculated.

DISCUSSION

The excitation functions for the reactions which have been studied in this work do not check too closely the results which have been previously reported[1] (see Fig. 2); however, this may be due to the fact that in the earlier work, the incidence of (p,pn) and $(p,p2n)$ reactions were neglected in the calculations of the reported cross sections.

It is believed that the fission cross sections reported here for the lower proton energies may be too high; the large neutron flux present in the region of the thorium stack from which the "low-energy" foils were taken could well have produced an appreciable number of fissions. This conclusion is supported by the observation of appreciable amounts of the 27-day Pa^{233} [from the process $Th^{232}(n,\gamma)Th^{233}(\beta^-/23\text{ min})Pa^{233}$] in the protactinium fractions separated from several of the "low-energy" foils.

Batzel[3] has shown that for any heavy isotope, a correlation may be made between the (a,xn) cross sections and the (a,f) cross section, where a is some initiating particle. The relation can be expressed as

$$\Gamma_f/\Gamma_n = C,$$

where Γ_f/Γ_n may be defined (ignoring charged-particle emission) by the equation:

$$\frac{\Gamma_f}{\Gamma_n} = \frac{\sigma_f}{\sigma_n + 2\sigma_{2n} + 3\sigma_{3n} + \cdots + X\sigma_{xn}} = \frac{\sigma_f}{\sum_x X\sigma_{xn}}.$$

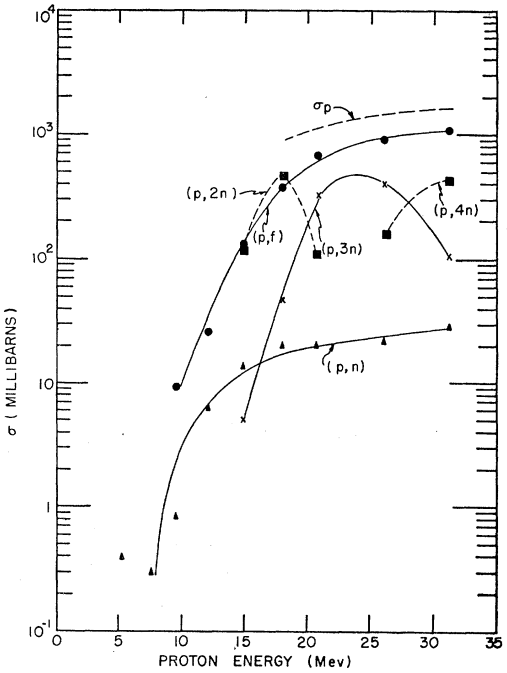

FIG. 1. Excitation functions for proton-induced reactions of Th^{232}.

It has been found that this ratio of the probability of fission to the probability of neutron emission is not dependent on the energy of the initiating particle, where this energy is great enough so that resonances in the compound nucleus are not important. However, the value of the ratio does vary for different isotopes, and is apparently a function of Z^2/A.

Since neither the σ_{2n} nor the σ_{4n} for protons on Th^{232} were determined in this work, an attempt was made to predict these cross sections from theoretical considerations.

[3] R. E. Batzel (private communication).

The total absorption cross section for Th^{232} was calculated from relations given by Blatt and Weiskopf,[4] using a radius for Th^{232} given by the relation, $R=1.5\times10^{-13}A^{\frac{1}{3}}$.

The total cross sections thus obtained are given in Table II; the calculations were not made for proton energies below 18 Mev since the values below this energy are extremely energy sensitive, and hence may be quantitatively misleading.

In determining the values of σ_{4n}, it was assumed that only (p,f), $(p,3n)$, and $(p,4n)$ processes were important above 25 Mev; between 18 and 25 Mev, only the (p,f), $(p,2n)$, and $(p,3n)$ processes were considered in finding σ_{2n}.

In order to calculate a value of σ_{2n} for $E_p=14.9$ Mev, the ratio σ_{2n}/σ_n was found by using the treatment of Blatt and Weisskopf;[5] σ_{2n} was then obtained from the known value of σ_n. The calculated values for σ_{2n} and σ_{4n} are given in Table II and are plotted in Fig. 1.

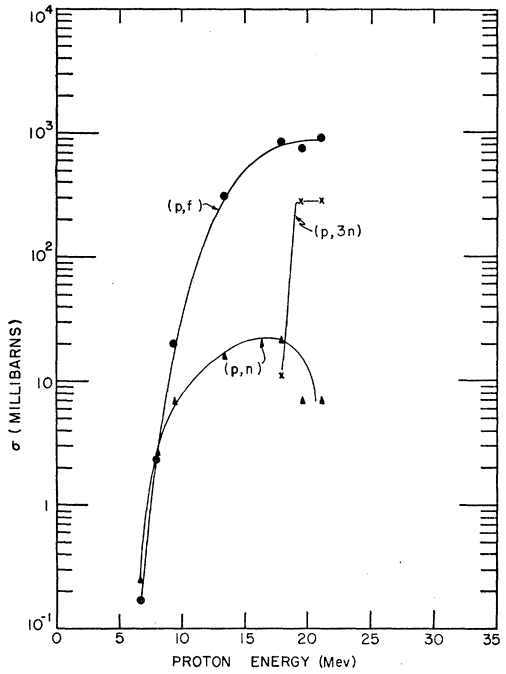

FIG. 2. Semitheoretical excitation functions for proton-induced reactions of Th^{232} (see reference 1, Fig. 9).

The calculated threshold for the $(p,3n)$ reaction is ~17.5 Mev; from neutron binding energies,[6] the threshold for the $(p,2n)$ reaction should be ~12 Mev. Consequently, at and below this energy, only the (p,f) and (p,n) reactions should occur.

[4] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 352.
[5] See reference 4, p. 379.
[6] G. T. Seaborg, "Nuclear Thermodynamics of the Heaviest Elements," Third Annual Phi Lambda Epsilon Lecture, Ohio State University, 1952 (unpublished).

TABLE II. Calculated cross sections for proton-induced reactions of thorium.

E_p (Mev)	σ_p (total) (mb)	$\sigma(p,2n)$ (mb)	$\sigma(p,4n)$ (mb)
31.3±0.4	1650		430
26.1±0.4	1500		160
20.8±0.5	1130	110	
18.0±0.5	910	470	
14.9±0.6		117	

TABLE III. Calculated values of the ratio Γ_f/Γ_n (probability of fission/probability of neutron emission).

E_p	Γ_f/Γ_n
31.3±0.4	0.52
26.1±0.4	0.49
20.8±0.5	0.56
18.0±0.5	0.34
14.9±0.6	0.49
12.1±0.7	4.3
9.6±0.9	9

On the basis of the foregoing treatment, the values for Γ_f/Γ_n were calculated; Table III is a tabulation of this ratio at different energies. It can be seen that Γ_f/Γ_n remains relatively constant at 0.5 for all energies above 14.9 Mev; the low value at 18 Mev may be attributed to the uncertainty of the total proton absorption cross section as calculated for this energy.

I wish to thank Dr. R. E. Batzel for many helpful discussions regarding the interpretation of the results obtained in this investigation. I should like to express my appreciation to Mr. G. H. Coleman for his help in making the chemical separations; also the cooperation of Mr. W. W. Olson and the linear accelerator operating staff is gratefully acknowledged.

APPENDIX

The chemical procedures used in the separation and purification of the zirconium and protactinium fractions have been slightly modified from those reported by Meinke.[7]

The thorium foils were dissolved in an HCl-HF mixture; the Pa was extracted into diisopropyl ketone (DIPK) from 6*M* HCl solution. The aqueous phase in this procedure was saved for Zr chemistry; the Pa was extracted into 2*M* $HClO_4$ from the DIPK, and subsequently re-extracted into 0.4*M* trifluorothenoylacetone (TTA) in benzene. It was then back extracted into 6*M* HCl (containing 0.1*M* HF) and plated on platinum.

The aqueous phase containing the Zr was made 3*M* in HCl and the Zr was extracted into 0.4*M* TTA in benzene. The TTA-benzene phase was washed and the organic phase was evaporated and ignited to give ZrO_2.

[7] W. W. Meinke, Atomic Energy Commission Report, AECD-2738 (UCRL-432) (unpublished).