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Fissionability of nuclides in the thorium region at excitation energies to 100 MeV

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We have measured seven and compiled from the literature seventeen excitation functions for spallation residues from the interaction of 10–100 MeV protons on ^{232}Th. Calculations of these excitation functions have been made using the theoretical framework of the pre-equilibrium exciton model of nuclear reactions to which has been added a fission option. The fit of the theory to the experimental data is excellent, lending confidence to our treatment of the competition between fission and particle emission. After showing and discussing the comparison of the calculated and experimental results, we present representative graphs of the variations in fissionability with Z^2/A and with the excitation energy.

[NUCLEAR REACTIONS ^{232}Th(p,xn), $x=1$, 3, 5–7; (p,pxn), $x=1$, 4–6; $(p,2pxn)$, $x=3$–7; $(p,3pxn)$, $x=5$–7. $E=10$–100 MeV; measured $\sigma(E)$, calculated $\sigma(E)$, Γ_f, Γ_n, Γ_p, Γ_α.]

I. INTRODUCTION

Fissionability, defined as the probability that a nucleus of given Z and A fissions, has been a topic of considerable interest to nuclear scientists throughout the four decades since the discovery of fission. With the coming of high energy charged particle beams, the energy to which the nucleus was excited also became a parameter in the determination of fissionability; even today, the question of whether the fissionability is, or is not, excitation energy dependent, remains controversial. The crucial competition between emission of neutrons (or charged particles) and fission as a means of de-excitation has been studied by large numbers of workers for a quarter century in an effort to gain insight into the elusive parameter, fissionability.

The first studies in the early fifties were by Tewes and co-workers[1,2] and were of spallation yields from thorium irradiated by protons of energy up to 32 MeV; in the late fifties, Seaborg's group performed comprehensive studies of spallation residues from 50 MeV alpha particle bombardments of targets having $88 \leq Z \leq 98$.[3,4] None of these studies showed clear evidence for an excitation energy dependence of fissionability and the simplifying assumption generally was made that fissionability was largely, if not entirely, dependent only upon Z^2/A of the fissioning nucleus. A comprehensive review of the topic by Vandenbosch and Huizenga in 1958[5] suggested that for nuclei excited to less than 25 MeV the Z^2/A value is much more important to a determination of fissionability than the excitation energy. Nevertheless, the question remained an open one.

As higher energy proton beams became available, the question increased in interest because high excitation energies with consequently long evaporation chains left scientists studying the fission process not knowing the indentity of the nucleus actually dividing. The sharply increasing fission cross sections of such "nonfissionable" nuclei as Au and Bi with increasing proton energy into the hundreds and then thousands of MeV made an energy dependence of fissionability appear likely (although it is not absolutely necessary because of long de-excitation chains) at least for nuclei with low values of Z^2/A.

A direct measurement was made by Cheifetz *et al.*[6] of the number of pre- and post-fission neutrons evaporated in bismuth fission by resolving their angular distributions into forward peaked (pre-fission) and isotropic (post-fission evaporation) components. Most analyses of fissionability, however, continued to concentrate on fission-spallation competition.

In general, the approach has been to predict

spallation yields by a theoretical calculation without fission competition, then compare the theoretical estimates with experimental spallation cross section, ascribing the difference to fission of intermediate nuclei along the evaporation chain. It is obvious that accurate predictions of spallation residue cross sections are necessary and it is here that problems arose in the early work. Two major efforts which approached the problem from opposite directions should be noted.

Dostrovsky, Frankel, and Rabinowitz,[7] used the Bohr-Wheeler[8] formulation of the fission width in conjunction with a Monte Carlo type evaporation calculation to compare the total fission cross section calculated for protons incident upon ^{238}U at energies up to 460 MeV with experiment. Their agreement with experiment was quite good using this formulation. However, the charge, mass, and excitation energy of the spectrum of residual nuclei from the fast cascade were parameters in the calculation; hence the detailed predictions of the model were treated with reservation. Also the choice of highly fissionable uranium and high energy protons made the calculation less sensitive to variation in the parameters than might otherwise have been hoped.

The opposite approach, calculating the spallation residues, comparing them to experiment, and ascribing the "missing" cross section to fission was first suggested and carried out by Huizenga in 1958.[9] Lindner and Turkevich using the newly available Metropolis *et al.* Monte Carlo calculation of the fast cascade then examined the results for 340 MeV protons on Th and U.[10] These authors all attempted to extract information about that energy dependence of the fission-evaporation competition which best fit the experimental data. Unfortunately, the reliability of early intranuclear cascade calculations, particularly for "simple" reactions, prevented a definitive answer to the problem. The authors concluded that they achieved as good a fit with no dependence on excitation energy as with, and concluded therefore that they had no evidence to dispute the conclusions of Vandenbosch and Huizenga.[5] Pate and Poskanzer,[11] performing a similar analysis on higher energy proton induced uranium fission, showed conclusively that problems with the intranuclear cascade calculation precluded this type of detailed analysis of fissionability.

More recent contributions have not clarified this matter. For example, Suk and Moore,[12] in their analysis of $^{232}Th(p, 6n)$ and $(p, 7n)$ excitation functions suggest that an energy independent Γ_n/Γ_f suffices to explain their experimental results. On the other hand, recent analyses of fission induced by π^- (at rest) absorbed by nuclei ranging from the rare earth region to uranium are based on a Dostrovsky-type approach[13, 14, 15] but the different authors do not agree on the value of parameters like the ratio a_f/a_n (see Sec. III D) which greatly influence nuclear fissility.

In the past few years however, pre-equilibrium calculations have proven highly successful in predictions of spallation yields.[16, 17] Specifically, we have calculated the excitation functions of approximately 50 nuclides produced in the spallation of targets with $A \simeq 90$ and 200[18, 19] using the exciton model developed by Gadioli and co-workers[17] at energies up to 100 MeV. In this paper, we extend those calculations to include the competition of fission with particle emission in the relaxation of excited nuclei. We report on studies of thorium fission making use of experimental data from this laboratory, reported herein, and the extensive work of the Orsay group of Lefort and others in the past two decades. This system was chosen both because of the availability of experimental data, and because thorium represents a midway choice between highly fissionable nuclei with $Z \geq 92$ for which spallation residues have very low cross sections, and elements having high fission barriers ($Z \leq 83$) where fission plays a minor role.

During preparation of this work, a calculation appeared by Delagrange, Fleury, and Alexander[20] utilizing the hybrid model developed by Blann[16] for 18–40 MeV alphas incident upon uranium for which four (α, xn) excitation functions were measured. A discussion of the results of both calculations are included in this work.

II. EXPERIMENTAL PROCEDURE

The target assembly consisted of three 3-mil thorium metal foils and three 2-mil copper foils from which the central Th and Cu foils were analyzed. Irradiations were performed in the internal beam of the McGill University Synchrocyclotron with an integrated beam intensity of about 0.1 μA h. The energy was determined by using the standard energy vs radius calibration for this machine. Reported energies have an uncertainty of ±2 MeV. The beam intensity was monitored using the $^{65}Cu(p, pn)\,^{64}Cu$ reaction.[21]

The thorium target was dissolved in concentrated HCl containing a few drops of HF, and thorium was radiochemically purified by successive precipitation of $Th(IO_3)_4$ and $Th(C_2O_4)_2$, followed by extraction from $Al(NO_3)_3/HNO_3$ into mesityl oxide.[22] The thorium was finally precipitated again as the oxalate, ignited to ThO_2, weighed, mounted, and gamma counted-in some cases by means of the growth of descendants down the alpha decay chains. Average yields were bout 65%.

TABLE I. Nuclear properties of products observed in this work.

Reaction product	Half-life	Characteristic gamma rays (keV)	Gamma ray abundance (per 100 decays)	Ref.
^{231}Th	25.52 h	163.1 165.0	0.159	26
^{228}Th	1.913 13 yr	238.6 (^{212}Pb) 241.0 (^{224}Ra)	48.8	27
^{227}Th	18.718 d	234.9 236.0	11.6	28
^{226}Th	30.9 min	242.0	0.866	29
^{225}Ra	14.8 d	439.7 (^{225}Ac)	21.000	30
^{224}Ra	3.66 d	238.6 (^{212}Pb) 241.0	48.8	27
^{223}Ra	11.434 d	154.2	5.59	28
^{64}Cu	12.82 h	511.0	38.6	31

Radium isotopes formed in $(p, 3pxn)$ reactions were isolated using a barium carrier and standard barium chemistry. The thorium was removed by precipitation of $Th(OH)_4$ and Ba/Ra was separated as $BaCl_2$ from HCl/ether reagent with several hydroxide scavenges. The barium was precipitated and counted as $BaCrO_4$ with an average yield of about 60%.[23]

Copper was purified by the identical procedure used routinely in this laboratory for determination of the ^{65}Cu(p, pn) monitor cross section.[24] The copper was separated from zinc by elution from a cation exchange column, reduced to Cu^+ and prepared for counting as CuSCN. The yields averaged 60%.

Counting of thorium and radium samples was carried out on 31 cm^3 and 41 cm^3 Ge(Li) detectors coupled with appropriate pulse height analyzers. The ^{64}Cu was dissolved in a standardized glass vial with conc. NH_4OH and the 511 keV annihilation radiation counted on a 3 in.$\times$ 3 in. NaI scintillation detector. All detectors were calibrated for energy and efficiency using standard IAEA sources with known disintegration rates.

The gamma ray peaks of interest were integrated and the decay curves least squares analyzed by the program EXPONENT.[25] The disintegration rates of the various nuclides at the end of irradiation were calculated applying corrections for detection efficiency, chemical yields, and gamma ray branch abundances. Absolute cross sections were then determined by comparison of the calculated disintegration rate of the nuclide of interest to the appropriate monitor cross sections. The nuclear

TABLE II. Reaction cross sections in millibarns for reactions measured in this work.

Proton energy (MeV)	^{231}Th	^{228}Th	^{227}Th	^{226}Th	^{225}Ra	^{224}Ra	^{223}Ra
13	2.9 ± 2.4						
18	45.8 ± 5.6						
20	96.5 ± 11.2						
30	112.3 ± 12.3						
40	94.1 ± 11.9	6.6 ± 4.2					
45	109.6 ± 13.3	36.6 ± 6.5	0.8 ± 0.5				
49	98.6 ± 11.2	36.8 ± 6.6	4.5 ± 1.2				
51	85.8 ± 16.3	35.9 ± 6.5	7.8 ± 1.3	1.6 ± 1.2	0.02 ± 0.01		
57	95.6 ± 12.3	58.2 ± 10.2	23.4 ± 2.9	3.8 ± 1.0	0.05 ± 0.03		
64			27.7 ± 3.0		0.19 ± 0.04		
70	100.0 ± 13.5	59.6 ± 6.5	39.8 ± 6.5	21.0 ± 2.7	0.23 ± 0.05		0.04 ± 0.03
75	88.5 ± 10.0	63.1 ± 9.0	39.4 ± 4.6	26.9 ± 2.6	0.33 ± 0.05	0.20 ± 0.17	0.29 ± 0.08
85					0.44 ± 0.08	0.53 ± 0.25	0.48 ± 0.12
90	69.9 ± 8.0	42.8 ± 7.4	32.2 ± 3.8	24.1 ± 2.4	0.45 ± 0.08	0.62 ± 0.22	0.49 ± 0.12

properties of nuclides measured in this work are listed in Table I and the reaction cross sections in Table II.

III. THEORETICAL CALCULATIONS

A. General

The calculation of the spallation products from 10–100 MeV proton irradiations of thorium is very similar to that described in detail in a previous paper[18] to which has been added the possibility of fission.[13] Except for a brief description of this option and one modification concerning the evaluation of α emission during the pre-equilibrium stage, we therefore do not discuss this calculation in detail. The exciton model[17] of pre-equilibrium emission was used to evaluate the initial stages of the interaction of proton and nucleus while an approach similar to that of Dostrovsky, Frankel, and Friedlander[32] was used for the equilibrium evaporation. The fission option was included only in the second (equilibrium) stage of the calculation because of the belief that it is unlikely that fission which involves a major collective motion of nucleons could occur on the time scale of the relaxation time of a nucleus whose excitation energy is concentrated among only a few nucleons.

B. The pre-equilibrium stage

As in our previous work on 10–100 MeV proton induced reactions,[18, 19] the initial configuration is, in most cases, assumed to be a two-particle–one-hole (2p1h) state. In a few cases, however, the incident proton may interact with a "preformed" α particle exciting 1p1α1αh states. The parameter ϕ, typically 5–10%, has been used to define the ratio of interactions with preformed alphas to those with individual nucleons. These initial states can decay either by particle emission or by exciton-exciton interactions (including interactions involving preformed α particles). Both in the case of the excited residual system after particle emission and in the case of the composite system in a more complex configuration, the competition between the two different decay modes continues. Eventually the nucleus reaches a state of statistical equilibrium which further decays by evaportation. The decay rates for exciton-exciton interaction and neutron and proton emission are given in Ref. 18; the level densities for neutrons and protons in the residual and composite nuclei [Eqs. (6) and (10) of Ref. 18] are evaluated with $\phi = 0$.

In our previous papers the energy distribution of the α's at each stage of the process was considered to be the one resulting from the statistical partition of the excitation energy among the α, the α hole and the remaining excitons assuming as equiprobable all the possible states. In our present approach, the α energy distribution is evaluated on the basis of nucleon-α scattering dynamics within the nucleus. A detailed discussion of this modification and of the parameters entering the calculation (which have been fixed by means of a detailed fit to α particle spectra on a number of nuclei ranging from Nb to Au at energies varying from ~ 20 to ~ 72 MeV) can be found elsewhere.[33] The reaction paths which include α emission always constitute a small contribution (at most a few percent at a proton energy of the order of 100 MeV) of the total absorption cross section. The calculated excitation functions of reactions like $(p, 2pxn)$ and $(p, 3pxn)$ reported later agree very well with those obtained with our previous code utilizing a value of $\phi \approx 0.07$.

The possible decays of the excited nucleus both during the pre-equilibrium and the evaporative stage are evaluated by means of a Monte Carlo approach. Multichance pre-equilibrium emission is allowed, just as in Ref. 18, but unlike the calculations of Delagrange, Fleury, and Alexander,[20] we have found a substantial probability of two nucleon pre-equilibrium emission, particularly at higher excitation energies (e.g., at $E_p \cong 100$ MeV, the average number of particles emitted per absorbed proton during the pre-equilibrium stage is ≈ 1.15). This finding confirms the suggestion of this possibility in their work.

C. The evaporative stage

The excited nucleus resulting from the pre-equilibrium cascade is then allowed to de-excite by emission of protons, neutrons, or alpha particles, or by fissioning. Except for the fission option described below the evaporation stage is exactly as described in detail in Ref. 18. This entails utilizing the analytical expressions of the Dostrovsky, Frankel, and Friedlander[32] procedure divided by the square of the residual excitation energy to account for the pre-exponential factor in the level density expression originally neglected by these authors. It is to be noted that this procedure removes the necessity of using unrealistic values of the level density parameter noted originally by the authors themselves.[32] This evaporation code does not require the necessity of normalizing yields to experimental values as was required, for example, in the Delagrange, Fleury, and Alexander,[20] work. The masses and binding energies, and the pairing energies, used in both the evaporation and fission calculations were, as before, taken from Wapstra and Gove[34] and Nemirovski and Adamchuck,[35] respectively.

D. The fissionability option

After equilibrium is reached, the excited nucleus at each stage of the evaporation chain is given the option of fissioning by computing a fission width, Γ_f, which has been taken as a function of Z, A, and the excitation energy of the nucleus. The emission of charged particles, protons, and alphas was not ignored in the calculation.

To compute the fission width we have followed the approach of Vandenbosch and Huizenga.[3] It is assumed that fission takes place above the barrier with a transmission coefficient equal to unity and below the barrier with a zero transmission coefficient. No account is taken of the shape of the fission barrier. This point is discussed below. The value of Γ_f may be evaluated[3]

$$\Gamma_f = \frac{1}{\overline{U}_s^{\,2}} \left[\frac{1}{2\pi\rho(E)} \int_0^{E-B_f-\Delta_s} \exp\{2[a_f(E-B_f-\Delta_s-K)]^{1/2}\}\, dK \right], \tag{1}$$

where $\overline{U}_s$ = average excitation energy at the saddle point, $\rho(E)$ = level density of the fissioning nucleus at excitation energy E, B_f = fission barrier from Ref. 13, Δ_s = pairing energy modified at the saddle point by less than 0.5 MeV from Ref. 35, as discussed in Ref. 13, a_f = level density parameter of the nucleus when deformed to the saddle configuration.

Equation (1) above may be integrated to yield

$$\Gamma_f = (\{2[a_f(E-B_f-\Delta_s)]^{1/2}-1\}\exp\{2[a_f(E-B_f-\Delta_s)]^{1/2}\})/[\overline{U}_s^{\,2}\times 4\pi a_f \times \rho(E)]. \tag{2}$$

The analogous expressions for Γ_n, Γ_p, and Γ_α are, where the subscript i may be n, p, or α,

$$\Gamma_i = \frac{\mu_i r_{0i}^{\,2} A^{2/3}(2s_i+1)}{\overline{U}_i^{\,2}\, 2\pi\hbar^2 a_i \rho(E)} \left(\frac{3}{4a_i}(x_i^{\,2}-2x_i+2) + \beta(x_i-1) - (E-B_i-C_i-\Delta_i) \right) e^{x_i}, \tag{3}$$

which except for the factor $1/\overline{U}_i^{\,2}$ coincides with the Dostrovsky, Frankel, and Friedlander expression.[32]

In the expression above, we define the following: μ_i = reduced mass of the residual nucleus A_i -emitted particle system, r_{0i} = radius parameter from Ref. 18, s_i = spin of the emitted particle i, $x_i = 2[a_i(E-B_i-C_i-\Delta_i)]^{1/2}$, $a_i = A_i/8$, B_i = binding energy of emitted particle i, from Ref. 34, $\beta = (2.12A_i^{-2/3}-0.050)/(0.76+2.2A_i^{-1/3})$ for neutrons, and $\beta = 0$ for protons and α particles, C_i = Coulomb barrier for the emission of particle i, Δ_i = pairing energy of the residual nucleus after the emission of particle i, from Ref. 35, $\overline{U}_i$ = average excitation energy of the residual nucleus after the emission of particle i.

In this work we will discuss Γ_f/Γ_n, the usual fissionability parameter. However, since the calculation also accounts for charged particle emission, we will also use the quantity P_f, which is the fractional probability that a nucleus fissions;

$$P_f = \frac{\Gamma_f}{\Gamma_n+\Gamma_p+\Gamma_\alpha+\Gamma_f}. \tag{4}$$

Several points should be specifically noted:

(a) The fission barrier is defined by a single parameter, its height. No account is taken of sub-barrier fission. The barrier is single peaked; no account is taken of structure. From a practical viewpoint, there simply is no experimental information about barrier structure for most of the nuclides encountered in the evaporation chain during, for example, the de-excitation of a ^{230}Pa nucleus excited to 60 MeV. Furthermore, the overall results of this calculation would not be strongly affected by this assumption in that typically excitation energies well above the fission barrier result from the pre-equilibrium stage. Only in the case of high Z^2/A, low excitation energy residual nuclei might the results be affected.

(b) No account is taken of angular momentum effects either in the calculation of Γ_i or Γ_f. As we discuss in Ref. 18, it is difficult to define spin distribution when the excitation energy is shared between few excitons, and therefore even more difficult to establish such a distribution for the nucleus reached at the end of the equilibrium cascade. Isomer ratios calculated from intranuclear cascade codes in which account is taken of angular momentum have not been notably successful, for example. It is also less likely that angular momentum effects seriously alter the results of proton induced reactions, although it is an effect which might be expected to play a relatively larger role in fission induced by moderate energy alpha particles[20] or heavy ions.

(c) There is little definitive information available regarding the values of a_f except that they are always greater than a_n due to deformity in the saddle nucleus. Evidence that they are not equal is derived from detailed analyses of fissionability at low energy.[3] Unfortunately, various authors have, or have not, included a variety of pairing energy

corrections (as we have done) in an evaluation of the energy available in the fissioning system; this makes direct comparison of results impossible. In this work we have treated the two (related) problems of pairing energy and level density as follows[13]:

For an odd-odd fissioning nucleus, E, the excitation energy available for fission used in the equation above is simply the excitation energy of the system.

For an odd A system, E is reduced by the pairing energy of the even neutron (proton) and further reduced by 0.25 MeV. Typically, for $Z = 90$, this reduction amounts to 1.14 MeV; $N = 140$ (e.g., ^{231}Pa), the reduction is 1.01 MeV.

For an even-even system, E is reduced by the neutron and proton pairing energies, and a further (2×0.25 MeV). Thus for ^{224}Ra, the reduction is 2.17 MeV.

The ratio of a_f/a_n was then treated as a parameter in the calculation and due to the multistep fission possibility in these reactions the results for, say, the $(p, 7n)$ reaction are extremely sensitive to it. Here the advantage of having a broad range of experimental data is quite evident. Both the simple reactions with one chance of fission such as the (p,n) and the very complex reactions must be simultaneously fit. However, it must be recognized that because of the interconnection of the pairing energy and level density parameters, it is not possible to state that these represent a unique (and therefore presumably correct) set of choices. The sensitivity of these calculations to the parameter a_f/a_n will be indicated later on, when discussing the comparison of experimental data with theoretical calculations.

IV. DISCUSSION

A. Comparison of the experimental data and the exciton model calculations

Comparisons between the experimental data of several authors (Refs. 1, 2, 12, 36–42, this work) and the excitation functions calculated using the model described above are shown in Figs. 1–5. In Figs. 4 and 5, the curves are labeled by the number of particles emitted without regard for mechanism; portions of the $(p, 2pxn)$ excitation functions actually result from $[p, \alpha(x-2)n]$ reactions. Because the experimental data are derived from several works, there is considerable scatter in the excitation functions; no attempt has been made to draw a "best" line through them. For the sake of clarity there are no errors shown either in the magnitude of the cross section or representing uncertainty in the beam energy, except for those cross sections measured in this work. Some "editing" has been done when a point or points clearly fall well off an otherwise well established excitation function. In each calculated figure, the open circles and solid line represent our calculated excitation function. Because of the "Monte Carlo" approach to the calculation, it must be recognized that there is also a statistical error associated with the theoretical results. This is particularly the case for the $(p, 2pxn)$ and $(p, 3pxn)$ reactions where cross sections as low as a few tenths of a millibarn have been measured and calculated. These may represent events occurring with a frequency as low as 1 in 5000.

1. The (p,xn) reactions (Fig. 1)

Excitation functions calculated assuming $a_f/a_n = 1.05$ are shown as the solid line. In the cases of $(p, 3n)$ and $(p, 5n)$ the effect of varying this parameter to 1.10 (dashed line) is also shown.

For the (p,n), $(p, 6n)$, and $(p, 7n)$ reactions, the fits to the experimental data are excellent

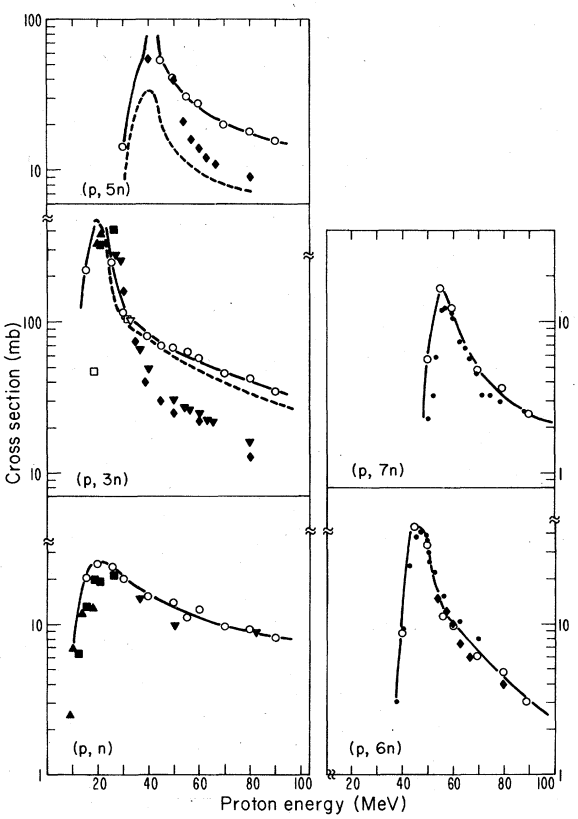

FIG. 1. Excitation functions for ^{232}Th(p,xn) reactions. Data are taken from ▲, Ref. 1; ■, Ref. 2; ▼ Refs. 36, 37; ◇, Ref. 38; ●, Ref. 12; open circles and the solid line are calculated values for $a_f/a_n = 1.05$. The dashed lines show the result of increasing a_f/a_n to 1.10.

for $a_f/a_n = 1.05$. The (p,n) results are rather insensitive to variations of this ratio; however, in the case of $(p,6n)$ and $(p,7n)$ reactions an increase of a_f/a_n by 5% on the average reduced the calculated excitation functions by, respectively, a factor of 10 and 20, and a decrease of a_f/a_n by the same amount increases them by, respectively, a factor of 3.5 and 5. In the case of the $(p,3n)$ reaction, although the tail of the excitation function is poorly reproduced in both cases, a better overall agreement is obtained by employing $a_f/a_n \sim 1.1$, while in the $(p,5n)$ reaction a value intermediate between 1.05 and 1.1 is indicated. If we take into account that fission competes with evaporation much more effectively at the end of the decay chains when Z^2/A is substantially increased, and therefore that the fissioning nuclei are substantially different in the $(p,3n)$ and $(p,7n)$ reactions, the above results suggest that a_f/a_n might vary, from 1.1 to 1.05, in going from the higher mass to the lighter mass Pa isotopes.

These results should not be interpreted as due to an *energy* dependence of the ratio a_f/a_n. The main reason is that fission occurs at fairly similar energies in the case of the different reactions. Let us consider, e.g., the case of $(p,5n)$ and $(p,7n)$ reactions at proton energies of, respectively, 40 and 100 MeV. In the first case, fission-evaporation competition is begun at an excitation energy of approximately 45 MeV (at the maximum of the excitation function the probability of previous pre-equilibrium emissions is negligible). In the second case we expect that during the pre-equilibrium phase ~ 1.5 particles have been emitted carrying away ~ 52 MeV of kinetic energy and ~ 9 MeV of binding energy. Then, just as before, fission-evaporation competition is initiated at an excitation energy of approximately 45 MeV.

This conclusion is strengthened by an analysis of the total Th fission cross section. At energies lower than $E_p = 35$ MeV (see Fig. 2) the fission cross section is very satisfactorily fitted by assuming $a_f/a_n = 1.1$; on the other hand the calculation based on $a_f/a_n = 1.05$ underestimates the fission cross section by an amount varying from 60% to 20% going from 15 to 32 MeV incident proton energy. At high incident proton energy where the mass of the fissioning nucleus is much lower, the fission cross section is better reproduced by assuming $a_f/a_n = 1.05$. In fact at $E_p \cong 100$ MeV, σ_f shows very little variation with proton energy and its value is approximately 870 mb. The values calculated assuming $a_f/a_n = 1.05$ and 1.1 are, respectively, 900 mb and 1070 mb.

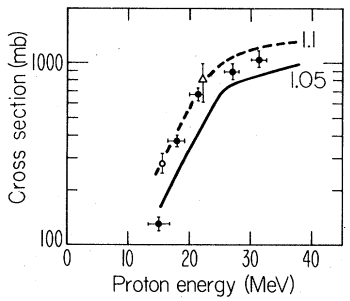

FIG. 2. The total fission cross section for low energy protons on ^{232}Th. Data are taken from ●, Ref. 1; O, Ref. 41; Δ, Ref. 42. Values calculated taking $a_f/a_n = 1.05$ and 1.1 are shown as solid and dashed lines, respectively.

For purposes of this work and the discussion of fissionability a point of great importance is the fit to the $(p,6n)$ and $(p,7n)$ data. The compound nucleus formed with 55–65 MeV of excitation energy must de-excite to the $(p,7n)$ product by sequential emission of seven neutrons and in each case, in competition with the fission process. At higher energies, e.g., 100 MeV, the predominant mechanism is expected to be the emission of ~ 1.5 neutrons in the pre-equilibrium stage with absorption of the proton and deposition of 45–50 MeV. Neutron evaporation to the product nucleus follows. In both cases, there is competition between fission and neutron emission over 5 or 6 (Z,A) pairs and over a wide range of excitation energy. Any error in the formulation of fissionability will be magnified in the cross section results by successive application of Γ_f/Γ_n five or six times. It is felt that achievement of such a fit without adjustable parameters to any $(p,6n)$ or $(p,7n)$ reaction would be surprising. To do so in a calculation in which fission plays a competing role is particularly gratifying.

2. *The (p,pxn) reactions (Fig. 3)*

Comparison of the calculations of the (p,pxn) excitation functions (made by using $a_f/a_n = 1.05$) with the experimental data are shown in Fig. 3. These experimental results were measured entirely in this work except for the two data points of Lefort, Simonoff, and Tarrago, (Refs. 36, 37) at 82 MeV. The fits of the calculation to the experimental data are excellent. The discrepancy in the increase of the (p,pn) cross section is similar to that seen and discussed previously[18,19] and has been attributed to the (p,d) reaction which is beyond the scope of the model used in this calculation. The calculated (p,pxn) excitation functions are relatively insensitive to variations of the a_f/a_n ratio. This is due to the fact that the proton is usually emitted during the pre-equilibrium phase. The Z^2/A of the fissioning nuclei is, as a consequence, noticeably reduced and fission competition becomes less effective. A further consequence of the reduced fis-

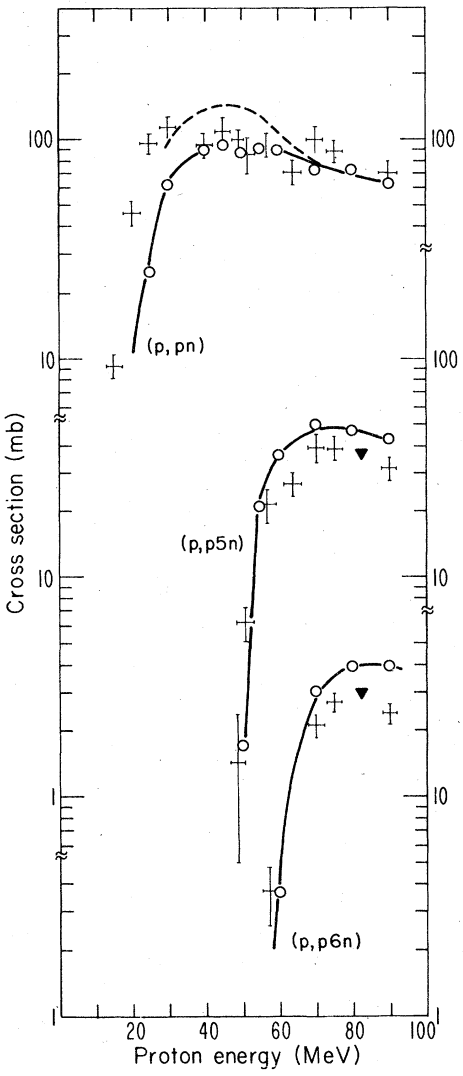

FIG. 3. Excitation functions for ^{232}Th(p,pxn) reactions. Data are from this work, and ▼, Refs. 36, 37; open circles and the solid line are our calculated values. The dashed portion of the (p,pn) curve includes the contribution from the (p,d) reaction, estimated from Ref. 19.

sionability of Th isotopes relative to the Pa ones is the high value of the ratio $\sigma(p,p6n)/\sigma(p,7n)$ (≈ 10 at $E_p \approx 90$ MeV). For a heavy but not highly fissionable nucleus like ^{209}Bi, the same ratio is essentially unity.[43] The calculation without any parameter variation satisfactorily reproduced this finding thus increasing our confidence in the substantial correctness of our procedure.

3. *The (p,2pxn) reactions (Fig. 4)*

Before discussing the comparison between theory and experiment it must be mentioned that the $(p,2p3n)$ excitation function appears to have a threshold which is essentially the same as that for

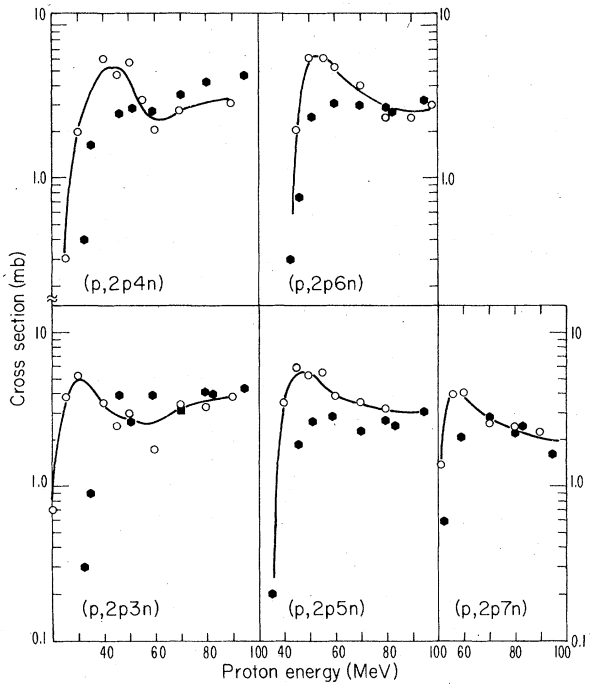

FIG. 4. Excitation functions for ^{232}Th$(p,2pxn)$ reactions. Data are taken from Refs. 39 and 40; open circles are our calculated values.

the $(p,2p4n)$ and appears to peak at, if anything, a higher energy. In view of the emission of one extra neutron in the $(p,2p4n)$ reaction, it is not surprising that a calculation cannot reproduce at the same time the $(p,2p3n)$ and the $(p,2p4n)$ excitation functions in the 30–50 MeV incident proton energy range. The $(p,2pxn)$ reactions involve the emission of alpha particles as well as the emission of individual nucleons. The agreement between the calculated ($a_f/a_n = 1.05$) and the measured excitation functions is not very good in the first peak region which arises from emission of alpha particles. A much more satisfactory fit would have been obtained by reducing the α particle density ϕ_α which enters the theoretical calculation of nucleon-α scattering within the nucleus, from the adopted value of 0.19ρ (Ref. 33) (ρ is the nuclear density) to 0.13ρ. We have not done this because the new value would noticeably differ from the one obtained by an extended fit of (p,α) spectra on nuclei spanning the periodic table and, at the same time, would worsen the excellent fit obtained in the case of $(p,3pxn)$ reactions (see next paragraph).

At large energies, when the emission of individual particles becomes predominant, we got an excellent fit which adds considerable support to the approach to fissionability we have taken. For example, the $(p,2p6n)$ cross section at 95 MeV incident energy involves excitation energies of 100

MeV and eight successive particle evaporation steps each with a fission competition. The results, we feel, are excellent and for completeness we include them here even though the fit to the experimental $(p, \alpha xn)$ cross sections at the lower energies is not extremely good.

Even going to the still more complex $(p, 3pxn)$ calculations, the results are excellent.

4. The (p, 3pxn) reactions (Fig. 5)

The fits to these excitation functions which over this energy range correspond to $[p, 1p(x-2)n1\alpha]$ reactions are extraordinarily good especially in view of the low cross sections and the Monte Carlo approach in the calculation. As many as 80 000 cascades were run at high energies to calculate these reactions which occur only once in 5000 events. The ratio a_f/a_n was taken equal to 1.05.

B. Fissionability

In summary, we conclude from the fits to the excitation functions that there is certainly justification for our feeling that this treatment of fissionability is valid. It remains to examine more closely the results to the calculation of a variation in the parameters, and to ensure that the data available have adequately tested the assumptions.

1. The dependence of fissionability of Z^2/A

The quantity Z^2/A is the classical "fissility" parameter. Errors which would be introduced into the calculation because of an incorrect dependence of the fissionability on Z^2/A would be easily observable because of the sequential nature of the evaporation chain leading to the final product, and because certain sets of experimental data specifically test the fissionability in certain regions of Z^2/A. For example, the (p, xn) reactions test the dependence of fissionability on Z^2/A for the large values of

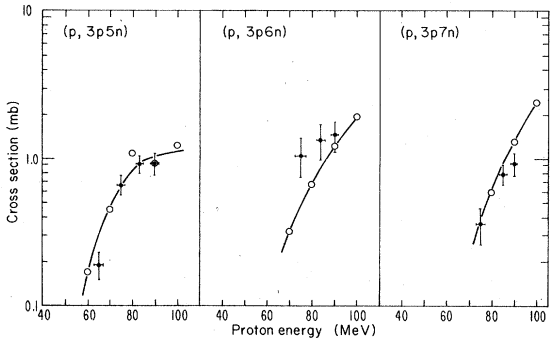

FIG. 5. Excitation functions for ^{232}Th$(p, 3pxn)$ reactions. Data are taken from this work; open circles are our calculated values.

parameter without ever encountering a low value at at any excitation energy. If Γ_f/Γ_n were too high for these isotopes of protactinium, all of the excitation functions would be sharply underestimated because of the "loss" of spallation residues to fission. This would particularly be the case near the peaks since the mechanism of compound nucleus formation and sequential evaporation of the requisite number of neutrons is almost certainly the mechanism of production of these nuclei. If on the other hand Γ_f/Γ_n were too low, the same rationale leads to the prediction of large overestimates in the peaks of the excitation functions. As noted previously, any such problem is magnified with the number of neutrons evaporated. Since the best fits, indeed nearly perfect fits, are found for the $(p, 6n)$ and $(p, 7n)$ reactions, it must be concluded that the fissionability dependence on Z^2/A is reasonable, at least for the more fissile nuclei.

The reactions in which charged particles are emitted lead to spallation residues of lower values of Z^2/A. Again those reactions having a large number of particles emitted are the most valid test of the theory due to the multiplication of the effect of errors through the sequential evaporation. Looking at our fits to the experimental data, we find that the most complex reactions are reproduced best and, of greater importance, there appears to be no systematic over- or underestimate of the cross sections as would be expected if there were an error in the formulation of the fissionability parameter.

That there is a dependence of fissionability on Z^2/A (even if the exact form of the dependence is not agreed upon) is, however, well known. We feel that the more interesting point is the dependence on the excitation energy of the fissionability.

2. The dependence of fissionability on excitation energy

As mentioned in the Introduction, this has been a point of debate for many years. Part of the reason for the continued controversy is that no single set of excitation functions adequately tests the theory; rather one must look at the overall set of data to examine the effect of an excitation energy independent fissionability function.

We have taken fissionability to be a function of excitation energy. If, rather, it were to be some "average constant value" then in the case of fissionability increasing with energy the most observable effects would be a general overestimate of the peaks in the very complex reactions involving large deposition energy and an underestimate of peaks for very simple reactions at low incident energies. This would be due in most cases to a lower fissionability at high excitation energies and a

higher fissionability at lower energies. The reverse should be true if fissionability is a decreasing function of energy. Nevertheless, it might be argued that the sequential evaporation down a long de-excitation chain might lead to compensating effects. However, it is doubtful that the effects would exactly balance and such a postulate necessarily fails in the case of low deposition energy events, e.g., the (p,n) and (p,pn) reactions.

We feel that the excellent fits to the high deposition energy events $(p,6n)$, $(p,7n)$, $[p,p(4-6)n]$, and $[p,3p(5-7)n]$ militate very strongly against such an approach at high energies and the very good fits to the two simplest reactions (p,n) and (p,pn) do the same at the lowest energies. Indeed in the whole set of seventeen excitation functions there are only two "regions" of bad fits, the evaporative alpha peaks in the $[p,2p(3-6)n]$ data and the high energy tails of $(p,3n)$ and $(p,5n)$. Ignoring the former as it is clearly independent of the fission option, we must consider the latter. It is not at all clear why the tails of these excitation functions are not well reproduced, but as we have suggested above, there is a possibility that some of the Pa isotopes might be better represented by a larger value of the parameter a_f/a_n. It is also possible that these discrepancies are the result of the breadth of the collection of data which we have used to construct these excitation functions.

Taken as a complete set, there appear to be no systematic deviations from experimental data, e.g., peaks or tails being overestimated or underestimated, simple low deposition energy or complex high deposition energy excitation functions being miscalculated, or highly fissile or less fissile nuclides being in substantial error. This is the strength of a large body of experimental data. It is difficult to imagine even a pair of compensating errors let alone a single one which would not lead to some systematic discrepancies in this large a body of data.

During the course of this work, the major work of Delagrange, Fleury, and Alexander (DFA)[20] was published and it would appear reasonable that a comparison of the two approaches should be made.

The foremost point is that the DFA calculation of fissionability, based on their earlier publication[44] and described in detail therein, is a highly complex one with many parameters to be evaluated. That calculation, far more sophisticated than the one described here, treats the shapes and heights of both the first (ground state) and second (isomeric) fission barriers as parameters whose values have been determined in low energy fission studies and evaluated elsewhere.[45] Its principal range of validity is for energies lower than that treated in this paper. Angular momentum effects and fission from specific deformed spin states are also taken into account. These effects are of far greater consequence in low energy alpha particle induced fission that it is in this work. Plutonium is much more fissionable than the thorium target we have studied, and as discussed in the Introduction, the spallation residues are fewer, more difficult to measure experimentally, and produced with lower cross section.

Coupling their fissionability calculation to the pre-equilibrium hybrid model and the evaporation program ALICE, both developed by Blann (see Ref. 16), DFA fitted the excitation functions for (α,xn) reactions. They conclude, at variance with our result, that "fits of pre-equilibrium model calculations to the tails of the excitation functions require significantly different parameters from those in the literature for other systems." While taking into account that the fissioning nuclei they considered are different from ours, we think that this conclusion should not be considered as a statement implying a general failure of pre-equilibrium evaporation calculations of fissilities in the

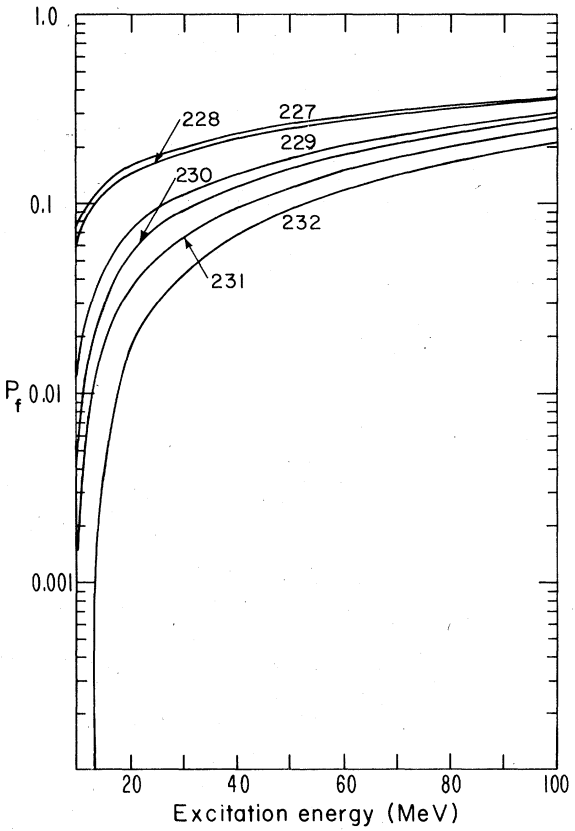

FIG. 6. The probability of fission, P_f, calculated as a function of excitation energy, E^*, for a series of isotopes of thorium. Curves are labeled by mass number, A. The value of a_f/a_n was taken equal to 1.05.

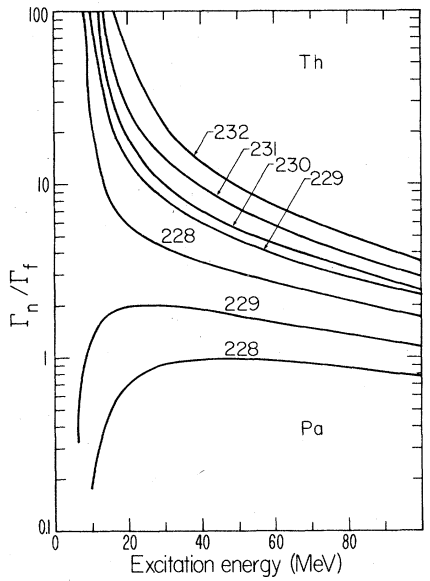

FIG. 7. The fission competition, Γ_n/Γ_f, calculated as a function of excitation energy, for isotopes of thorium and protactinium. Curves are labeled by mass number, A. The value of $a_f/a_n = 1.05$.

U-Th region. We feel that the results of the present study using a quite simple approach and few parameters offers great encouragement for future work in this field.

Our calculation simply incorporates a fissionability option into an already existing and, we believe, well proven approach to the calculation of spallation induced by 10–100 MeV protons.[18, 19] We examine now the predictions of our approach to fissionability and present and discuss a few of them. Interested readers may use the equations in the theory portion of this work to calculate complete tables.

In Fig. 6, we show the probability of fission of each of the thorium isotopes as a function of its excitation energy ($a_f/a_n = 1.05$ for all considered nuclei). To avoid misunderstanding, the probability that ^{232}Th, excited to 100 MeV, fissions is 21.5%. The other 78.5% may be partly accounted for by, for example, neutron emission leading to a ^{231}Th nucleus excited to the order of 90 MeV. The probability of it fissioning is about 24%. The *total* probability that a ^{232}Th nucleus having $E^* = 100$ MeV will fission at some time before reaching a final spallation residue must be calculated considering all possible evaporation sequences and the various nuclides produced at their respective excitation energies. Curves may also be derived for elements having Z other than 90.

In Fig. 7, we show the curves of Γ_n/Γ_f for the thorium isotopes of interest in this work. All show the decrease with increasing excitation energy characteristic of nuclides whose fission barrier is

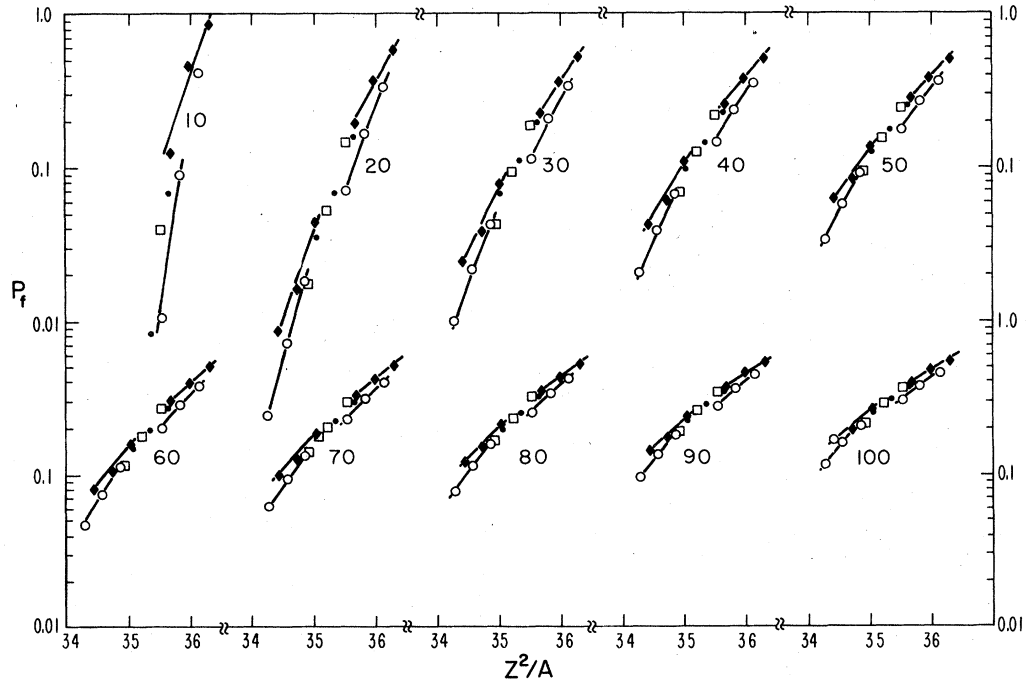

FIG. 8. The probability of fission, P_f, calculated as a function of the fissionability parameter, Z^2/A, for ten excitation energies (labeled). ◆, odd Z, odd N; O, odd Z, even N; ●, even Z, odd N; □, even Z, even N. The value of $a_f/a_n = 1.05$.

greater than the neutron binding energy. For comparison, the curves for ^{229,228}Pa whose fission barrier is less than the neutron binding energy is also shown. This behavior is in agreement with the suggestions of Huizenga and Vandenbosch[3] that the shape of the curves depends on the relative magnitudes of these quantities.

The dependence of P_f on Z^2/A is shown in Fig. 8 where several things are noteworthy. First, there is the strong Z^2/A dependence which is certainly to be expected, but second, and perhaps more important, its effect becomes less pronounced at higher and higher excitation energies. This appears to us to be reasonable in that in any competitive processes involving barriers that are small relative to the total energy available, selectivity is greatly reduced. The lines drawn, merely to guide the eye, connect odd-odd and odd-even nuclides and illustrate the magnitude of the effect of pairing energies on the fission process. It should be particularly noted that the relatively small pairing energy has a very strong effect on fissionability at low excitation energies again underlining the critical effect of the competition between fission and particle emission when near the barrier. This is completely in accord with the findings of DFA.[20]

V. CONCLUSIONS

We have fit quite successfully seventeen excitation functions for spallation residues in 10–100 MeV proton fission of ^{232}Th with an approach to fissionability based on Bohr theory and the equations of Vandenbosch and Huizenga. We have done so assuming fission, a highly collective process, does not compete in the fast pre-equilibrium step, and does have a functional dependence on E^* along with the "traditional" dependence on Z^2/A.

Because of the sensitivity of the calculation to the single free parameter, a_f/a_n, we feel that a value of 1.05 is appropriate (except perhaps for the high mass Pa isotopes) in this Z and A region at all energies up to 100 MeV after corrections for the pairing energy. And finally, we find that for these moderately fissionable nuclei we can reproduce a broad range of experimental data with a theory which is a straight-forward extension of the existing exciton model of nuclear reactions.

ACKNOWLEDGMENTS

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