Alpha-induced reactions on fissile nuclei at energies to 50 MeV

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An existing model of alpha-particle-induced reactions has been extended to include fission competition. Forty-six excitation functions from nine target nuclei at incident energies to 50 MeV have been calculated and compared to experimental data taken from the literature of the last 35 years. In particular, it is shown that specific modes of interaction of the alpha particle are responsible for the features of individual excitation functions. A simple fission barrier characterized only by its height is shown to suffice in reproducing the fission-evaporation competition.

I. INTRODUCTION

In the 1950s, when alpha-particle beams up to 50 MeV became available at the Crocker Laboratory in Berkeley, the Seaborg research group carried out a number of experimental studies of the fissionability of nuclides in the Th-Pu mass region.[1–3] These involved the measurement of production cross sections of the spallation residues produced in competition with fission. At the time, the state of knowledge of the mechanisms involved in the interaction of alpha particles with heavy nuclides precluded any calculation of the fissionability of a given nuclide at a given excitation energy. Only average values of the fission width Γ_f over a range of energies, and often over one or more nuclides encountered during the deexcitation, were extracted. All but forgotten in the intervening years, however, has been the large number of excitation functions measured from eight different targets.

As reaction models became more sophisticated, Alexander *et al.*[4–6] remeasured the (α,xn) excitation functions from ^{237}Np (Refs. 4 and 5) and from ^{233,234,235}U (Ref. 6) and attempted to fit them, including the fission competition, with the hybrid model.[7] Although moderately successful, problems arose mainly because these calculations did not account for several α-particle-target nucleus interaction modes, including inelastic scattering and binary breakup.

At nearly the same time, Gadioli *et al.*[8] made use of the exciton model for proton-induced reactions to successfully reproduce a range of spallation excitation functions from a ^{232}Th target, including the fission competition in the decay of the intermediate excited nuclei. Somewhat later, the exciton model was extended[9] to consider alpha-induced reactions on nonfissile nuclei. This work was greatly enhanced by the then newly available data on breakup mechanisms in alpha-nucleus reactions performed largely by Holmgren, Chang, and co-workers[10–12] at Maryland. This extended model, known as OMEGA,[9] included five distinct preequilibrium mechanisms (in addition to compound nucleus formation), the inelastic scatter of the alpha from the nucleus, pickup reactions forming ^{5}He and ^{5}Li,[13] binary fragmentation to a variety of combinations,[10–12] dissolution of the alpha into four nucleons, and alpha-nucleon collisions.[14]

The combination of these three factors, the availability of many experimentally measured spallation functions from a variety of fissionable nuclei, the successful inclusion of fission as a deexcitation mode in proton-induced reactions, and the extension of the exciton model to include complex modes of alpha-nucleus interaction, suggested strongly that an attempt should be made to calculate the spallation-fission competition in heavy nuclides produced in alpha-particle-induced reactions.

In succeeding sections, we first examine the wealth of experimental data in the literature, and apply and discuss such corrections as may be needed in the light of forty years of study of the radioactive decay modes, branching ratios, and half-lives, and of adjustments to the quoted alpha-particle energies in the light of the remeasurement of some cross sections and updated range-energy formulations.

There follows a discussion of the OMEGA model, in particular as it is affected by the evaluation of the relative contributions of various alpha-nucleus interaction modes, a description of the calculations performed, and a comparison of the theoretical predictions with the experimental data.

A total of 46 excitation functions on nine target nuclei, ^{232}Th, ^{233,234,235,238}U, ^{237}Np, and ^{238,239,242}Pu at α-particle energies from 20–50 MeV have been considered. There are no freely varying parameters in this work; even values of the total reaction cross section and relative importance of differing reaction mechanisms were chosen *a priori* on the basis of previous work. All parameters used were fixed for all reaction channels at all energies on all nine targets.

II. EVALUATION OF EXPERIMENTAL DATA

The data used in this work are those of Seaborg and co-workers,[1–3] Alexander and co-workers,[4–6] Gunnink and Cobble,[15] Bethune *et al.*,[16] and Colby *et al.*[17] These data are collected in Table I. For the most part they are

TABLE I. Corrected experimental excitation functions used in the comparison to the theoretical calculations. The first column gives the incident α-particle energy (MeV), the remaining the cross sections (mb). The estimated percentage errors are also indicated.

^{232}Th (Ref. 3) MeV	$(\alpha,4n)$ ^{232}U	(α,p) ^{235}Pa	(α,pn) ^{234}Pa	$(\alpha,p2n)$ ^{233}Pa	$(\alpha,p3n)$ ^{232}Pa	$(\alpha,2pn)$ ^{233}Th	$(\alpha,\alpha n)$ ^{231}Th
Error in data is 10% except <2 mb=20%.							
15.7				0.21	0.11		
20.4				0.21	0.02		
22.1				0.41	0.02		
24.0				0.31			
26.2		1.12	0.61	3.30		0.52	0.84
28.1		0.65	0.41	2.40	0.08		3.59
30.3		3.47	5.24	8.50	0.34	0.59	2.29
32.3				10.00	0.25		19.94
35.4	18.40	2.96	16.11	14.00	0.93	2.78	15.95
35.9	19.53					0.34	15.95
36.9	17.99	5.51	21.15	17.00	1.80	1.82	27.92
38.6	28.68					1.91	14.96
39.8	56.23	3.06	22.15	21.00	2.30		36.89
40.5	56.03		21.15	18.00			30.91
44.1	32.48		19.13	24.00	4.10	4.02	34.90
45.0	35.67		18.13	22.00	5.20		41.87
45.0	34.34						48.85

^{233}U (Ref. 2) MeV	(α,n) ^{236}Pu	$(\alpha,2n)$ ^{235}Pu	$(\alpha,3n)$ ^{234}Pu	$(\alpha,4n)$ ^{233}Pu	(α,p) ^{236}Np	(α,pn) ^{235}Np	$(\alpha,p2n)$ ^{234}Np	$(\alpha,p3n)$ ^{233}Np	(α,f)
Error	10%	20%	20%	50%	22%	50%	5%	25%	20%
19.5	0.19								
22.8	0.44	1.28			0.20	0.97	0.16		184
25.5	0.62	3.62	0.08		0.54	1.74	1.63		400
28.3	1.01	6.44	0.06						
28.8	0.68				0.64	3.38	5.04		
30.1			0.89		1.76	0.29	4.91		1060
31.3	1.07	3.35	0.38		0.65		3.52		
31.9					1.09	13.04	10.90		
33.8			0.95		0.59	2.42	5.20	0.22	
34.9	0.52	1.17			1.49	6.28	10.50		1270
36.4			0.66						
37.4			0.47		0.76	3.38	7.25	0.11	
38.7	0.55	0.92	0.53						
39.7			0.43						
40.1					0.41	4.44	10.40	1.20	
40.8	0.44	1.17	0.32		0.63	14.39	11.80	0.62	1430
42.5			0.19	0.28	0.72	2.51	9.40		
43.6					2.59	8.50	17.80	1.46	
44.2	0.77		0.25		0.76	18.06	19.90	1.78	1990
44.3			0.50	1.08	0.75		15.90	0.66	
46.1	0.83		0.44	1.18	0.31	20.58	19.60	1.14	
46.1		1.29	0.20	0.34					
46.1			0.15	0.36					

^{233}U (Ref. 17)	
Error	25–50 %
25.3	350
29.0	606
34.5	1090
40.5	1345

TABLE I. (*Continued*).

^{233}U (Ref. 6) MeV	(α,n) ^{236}Pu	$(\alpha,2n)$ ^{235}Pu	$(\alpha,3n)$ ^{234}Pu
Error	10%	10%	10%
21.2		0.21	
22.0	0.43		
22.5		0.51	
22.7		0.65	
23.5		1.43	
24.0	0.73	1.84	
24.3		3.42	
24.7		2.77	
25.5	1.05	5.04	
26.8	1.24	5.45	
28.9	1.22	2.69	0.05
30.0	1.22	3.13	
30.3			0.25
30.9	1.43		0.21
31.9	1.12	2.78	0.38
31.9			0.49
32.7	1.67		1.21
33.7			2.05
33.8			1.47
33.8	1.95	3.14	1.75
34.1			1.45
34.8	1.34		
35.1			1.42
35.6	1.80		1.07
36.9	2.01	3.11	1.44
37.1			0.98
37.4			1.06
37.4	1.15		0.74
38.2			0.93
39.0	1.33		0.48
39.0			0.74
39.1			0.90
39.5			0.67
40.4		1.86	0.66

^{234}U (Ref. 6) MeV	(α,n) ^{237}Pu	$(\alpha,2n)$ ^{236}Pu	$(\alpha,3n)$ ^{235}Pu	$(\alpha,4n)$ ^{234}Pu
Error	10%	10%	10%	15%
18.8	0.17			
20.1	0.40	0.17		
21.4	0.34	0.40		
21.9		1.24		
23.1		2.48		
24.2	1.86			
24.3		6.26		
25.5	1.58	10.22		
25.9	1.34	7.14		
28.0		9.39		
30.0	1.86	6.68		
30.0	2.18	8.51	0.19	
30.0		6.58		
30.3	2.24	6.30	0.28	
31.9	1.37	3.25		
32.0		6.95		
32.3			1.30	
33.1			1.93	

TABLE I. (*Continued*).

^{234}U (Ref. 6) MeV	(α,n) ^{237}Pu	$(\alpha,2n)$ ^{236}Pu	$(\alpha,3n)$ ^{235}Pu	$(\alpha,4n)$ ^{234}Pu
Error	10%	10%	10%	15%
34.1	1.32	2.61	1.67	
35.2		4.88	5.93	
35.6		4.04	6.11	
36.9	3.09	9.22	5.54	
37.4	1.28		3.36	
40.0	1.58	5.84	3.77	0.63
40.4	1.38	2.03	3.24	0.51
40.4	1.30	3.94	1.87	0.37
42.3				0.78
43.1	0.89	2.78	1.28	
43.2				1.17
44.1				0.83

^{235}U (Ref. 2) MeV	(α,n) ^{238}Pu	$(\alpha,2n)$ ^{237}Pu	$(\alpha,3n)$ ^{236}Pu	$(\alpha,4n)$ ^{235}Pu	(α,p) ^{238}Np	$(\alpha,p2n)$ ^{236}Np	(α,f)
Error	15%	15%	15%	25%	15%	25%	25%
17.9	0.26						1.80
21.1	0.35	4.56			0.02		58
22.9	1.29	13.70			0.04	0.04	
24.5		0.00			1.02	0.09	
26.7	1.70	16.27	0.64		0.56	0.53	420
29.1		0.00	0.00		1.72	1.90	
29.4		8.55	4.67		1.44	2.27	
30.0	1.39	7.05	4.38		1.59	2.43	
32.0							780
33.6	2.10	0.00	9.10		2.10	4.48	1290
34.3		7.00	7.63			4.30	
36.7		0.00	3.87	0.17	1.94	6.04	1490
39.2	2.21	5.82	3.29	1.48	1.89	8.70	
42.6	2.46	4.94	2.35	2.36	1.96	10.95	1760
45.3	0.89	3.61	1.96	1.53	1.22	10.74	1840

^{235}U (Ref. 16)	25%
Error	
25.3	384
25.7	438
26.0	478
26.2	506
26.5	537
26.8	605
27.1	594
27.4	668
27.9	756

^{235}U (Ref. 17)	
Error	25–50 %
20.5	10
23.1	87
25.9	290
28.2	580
33.8	1030
39.9	1386

TABLE I. (*Continued*).

^{235}U (Ref. 6) MeV	(α,2n) ^{237}Pu	(α,3n) ^{236}Pu	(α,4n) ^{235}Pu
Error	8%	8%	20%
19.2	0.35		
20.4	0.33		
21.6	1.84		
21.9	3.13		
22.8	8.00		
24.0	17.00		
24.2	14.42		
24.3	11.12		
25.5	18.03		
25.5	21.42	0.17	
28.3	14.52	0.49	
29.5		1.95	
30.8	11.43	5.74	
32.7	13.91	7.31	
33.4	10.06		
33.7	5.93	9.21	
34.0	8.89		
34.0	7.70		
35.3	6.27	9.03	0.21
35.5	6.04		
35.6	6.45		0.07
36.0	5.93	7.15	0.18
36.5	3.27	5.46	
37.0	4.36	5.01	0.06
38.0	0.00	4.69	0.54
38.2	5.28	5.22	0.56
40.0	3.90	4.01	
40.0	4.52	3.89	0.90
41.9	3.40	3.63	
42.0		3.85	2.20
44.0		3.94	1.32
44.1	3.54	3.08	
46.0	2.51	2.50	
46.0		2.88	0.87
46.0		1.80	

^{238}U (Ref. 2) MeV	(α,pn) ^{240}Np	(α,p2n) ^{239}Np	(α,αn) ^{237}U
Error in data = 15%			
21.9	0.03	0.22	
24.5	1.23	1.08	0.60
26.4	1.34	9.30	1.50
32.0	1.90	9.20	8.20
33.3	4.02	9.50	7.90
37.6	6.70		
38.3	6.81	17.89	49.20
38.3		20.95	56.20
39.7			
41.2	7.04	21.67	
43.8			56.00
45.3	5.92	34.13	74.00

TABLE I. (*Continued*).

^{237}Np (Ref. 4) MeV	(α,n) ^{240}Am	$(\alpha,2n)$ ^{239}Am	$(\alpha,3n)$ ^{238}Am	$(\alpha,4n)$ ^{137}Am
Error in data = 20%				
19.5	0.39	0.10		
19.8				
20.0	0.55	0.28		
20.1	0.55			
20.8	0.86	0.83		
21.6	1.35	1.41		
22.0				
22.3	1.56	3.20		
22.7				
22.9	1.05	6.32		
25.0	2.44	16.91		
26.3	2.55	17.79		
27.2		23.59		
27.6	2.87	27.03		
27.9		26.15		
28.5		19.46		
28.7	2.66	26.05		
29.2	2.46	14.45	0.36	
30.3	2.86	13.76	1.80	
31.0			1.30	
31.3	3.06	11.11	3.00	
32.1			1.77	
32.6	2.63	11.99	2.33	
33.1			3.25	
33.7	2.57	8.06	6.12	
34.6	2.05	6.19	5.73	
36.5	1.72	6.06	6.35	
37.6	1.33	5.87	7.03	
39.2			7.73	0.14
40.0	1.40	6.48	3.95	
40.4			4.77	0.30
41.1	1.88	6.13	4.42	0.54
43.5	1.23	4.74	3.07	0.95
45.9	1.11	4.95	2.64	0.99

^{237}Np (Ref. 5)				
Error in data = 20%				
17.5				
18.0	0.02			
18.5	0.16			
19.0	0.34	0.03		
19.5	0.46	0.08		
20.0	0.44	0.14		
20.5	0.83	0.43		
21.0	1.08	0.96		
21.5	1.31	1.75		
22.0	1.04	2.02		
22.5	1.30	3.91		

^{238}Pu (Ref. 7) MeV	(α,n) ^{241}Cm	$(\alpha,2n)$ ^{240}Cm	(α,pn) ^{240}Am	$(\alpha,p2n)$ ^{239}Am
Error	25%	15%	10%	20%
24.5	4.31	15.00		
28.1	6.65	14.00	2.59	2.95
29.6		9.90	1.89	5.11

TABLE I. (*Continued*).

^{238}Pu (Ref. 7) MeV	(α,n) ^{241}Cm	(α,2n) ^{240}Cm	(α,pn) ^{240}Am	(α,p2n) ^{239}Am
Error	25%	15%	10%	20%
32.5	5.62	8.90	3.29	6.78
36.2	2.90	4.70	14.94	26.54
42.0	2.34	4.30	12.95	21.63
47.4	2.62	3.50	7.97	17.69

^{239}Pu (Ref. 1) MeV	(α,n) ^{242}Cm	(α,2n) ^{241}Cm	(α,3n) ^{240}Cm	(α,p) ^{242}Am	(α,p2n) ^{240}Am
Error	15%	25%	15%	25%	10%
19.4	1.11			0.03	
23.8	0.84	6.28		0.58	
26.1	1.11	9.18	1.20	0.72	0.30
26.9	1.31	11.24	0.86	0.29	
33.5	2.41	8.90	3.50	1.30	5.18
37.7	2.21	8.43	4.50	0.96	7.67
38.9	1.81	7.59	3.60	0.58	9.56
40.4	0.97	6.84	2.30	1.10	13.94
43.6	1.51	7.03	2.70		14.94
44.2	1.61	5.43	3.00		
45.9	2.61	3.94	2.10		12.95
47.5	0.82	4.31	1.90	0.56	16.93

^{242}Pu (Ref. 1) MeV	(α,2N) ^{244}Cm	(α,4n) ^{242}Cm
Error	15%	15%
22.9	103.62	
25.2	116.70	
26.4	70.42	
27.9	68.41	
32.3	30.18	1.81
38.5	24.14	8.64
43.3	15.09	8.34

reported as the authors reported them; errors are their estimates for statistical errors (only) which for the sake of simplicity we have generally rounded up to the nearest 5%. Discussion of the errors is in Sec. II D below. The data of Table I vary from those reported in the original work for the following reasons.

A. Decay schemes

Because the early results[1–3,15] were measured almost entirely by detecting gross alpha particles or gross beta particles, and because nearly all the nuclides measured decay effectively 100% by one or the other mode, there were no corrections necessary for unknown or incorrect branching ratios. In a few cases, the original data were reported "relative" to other nuclides because at the time the α (β) yields were unknown. Appropriate corrections have been applied using the compilation of the Table of Isotopes[18] to convert the reported results to absolute values. One excitation function, ^{237}Np (α, 3n) taken from Ref. 4, lists as the measured γ ray a transition which is not found in the Table of Isotopes. It may be that this merely represents a misprint in the table of the reference since the data are quite consistent with that of other (α,xn) excitation functions; however, as will be seen, the peak of this excitation function seems to be shifted from the expected value by several MeV.

B. Half-lives

Recent compilations of decay schemes[18] have changed many of the half-lives from those used in the early works.[1–3,15] Having measured an activity, the number of atoms produced in the irradiation—and therefore the reported cross sections—is directly proportional to the half-life used in the analysis. For the most part the recent half-lives differ from those used by only a few percent, the worst case being 5.7%, so the corrections made were small and normally within the originally reported statistical uncertainty.

C. Incident energy of the alpha particles

All of the early work reported[1–3,15] was performed using stacked targets with beam degradation. Unfortunately, the exact range-energy formulation used in this work references a U.S. Government document we were unable to obtain. However, one of the recent works[16] remeasured some of the original data, concluding that the Seaborg results erred by 0.7 MeV at a reported incident energy of 25 MeV. With the assumptions that the extracted cyclotron beam energy was exactly known, and that range is proportional to velocity,[19] an equation was derived relating the original reported incident energy, E, to that used in this work, E_{new}:

$$E_{\text{new}} = 1.07446E - 0.5252E^{0.5} + 0.0642 \ .$$

Notwithstanding this reevaluation of the average beam energy striking a target, the stacked foil technique suffers from a spread of beam energies striking a given target in the stack. This problem is particularly difficult at the lower beam energies where there has been considerable degradation of the incident energy; Seaborg suggests a spread of ± 2 MeV at incident energies of 20 MeV. Without (unpublished) detailed knowledge of the stacks, no correction can be made for this effect. It should be noted that the effect is strongest on the steeply rising or falling portions of an excitation function and it must, therefore, be expected that the calculations of this work will lead to sharper peaks and valleys than reported in the experiments. This secondary effect of the energy degradation was noted when two groups, working twenty years apart, measured the same excitation functions[2,6] and found the steep rise from the threshold differing by 2–5 MeV while peak heights and the higher energy portions were in reasonable agreement.

D. Uncertainties in the data

As stated above, the errors of Table I reflect the original reported errors, generally rounded up to the next 5% interval. In only a few cases were systematic errors discussed by original authors, and these were not included in the reported tables of results. From those discussions, it may be surmised that the absolute values of Refs. 1–3 and 15 contain at least a 25% error, that errors of 30–50 % are likely in most results, and for cases where cross sections were <1 mb, an error of 100% is not unlikely. Such estimates are justified by the scatter of the data and by the inconsistency of the 1950s data[1–3,15] to some of the data from the work in the 1970s.[4–6,16]

III. THEORETICAL CALCULATIONS

The calculations performed were based on the theoretical approach discussed in Refs. 9, 14, and 20–22. We will not describe this theory again in detail, but simply recall that various interaction modes of the alpha particle with the nucleus are considered, of which the two most important are α-particle fragmentation into four nucleons, whose cross section will hereafter be indicated by σ_{4P} (P stands for particle), and the interaction of the α particle with single nucleons of the target. The total cross section for the processes originated by this interaction (α-particle inelastic scattering, and various fragmentation modes) will be indicated by $\sigma_{\alpha N}$. Minor contributions to the reaction cross section are binary fragmentation (σ_{BU}) and mean field inelastic scattering with excitation of collective levels and giant resonances (σ_{IS}). The only new ingredient of the present calculations is the inclusion of fission competition in the decay of the equilibrated excited nuclei that are produced at the end of the cascades of nucleon-nucleon interactions triggered by these various interactions of the α particle with the nucleons.

The validity of this theoretical approach has been tested by comparison of the results obtained with a large number of experimental data referring to excitation functions, emitted particle spectra, and recoil distributions of spallation fragments.[9,14,20–23] If one excludes the parameters necessary to evaluate the fission probability that will be discussed below, all the parameters utilized to evaluate the processes occurring during the deexcitation cascades are those used in our previous calculations, reported at the end of Sec. II of Ref. 22.

Most of our previous calculations referred to incident energies in excess of 50 MeV, while the excitation functions considered in this paper are measured at energies smaller than 50 MeV, and as slow as 20 MeV. Thus, the use of our theoretical approach requires the extrapolation of the cross sections of various α-nucleus interaction modes to energies smaller than those previously considered. This introduces an extra source of uncertainty in the theoretical predictions that is difficult to assess quantitatively, but which, in the worst case, could introduce a systematic error of the order of 30–50 % in the estimated cross sections.

Figure 1(a) reports, as a function of the α-particle energy, the values of the cross sections we used in the case of reactions induced on the U isotopes. The reaction cross section σ_R is taken from Kapoor *et al.*[24] The same total cross section was used to generate the reaction cross sections for the processes induced on ^{232}Th, ^{237}Np, and Pu isotopes with the procedure suggested by Friesleben and Huizenga[25] (the reaction cross section for target nuclei with Z and A not very different is the same when reported as a function of E_α / E_B, where E_α is the incident α-particle energy and E_B the interaction barrier as defined by these authors).

Figure 1(b) reports the percentages of cross sections corresponding to the formation of the compound nucleus, σ_{CN}, with mass equal to the sum of the target and the α-particle mass at the end of a series of α-nucleon interactions (without α-particle fragmentation), to α-particle fragmentation as it enters the nuclear field, σ_{4P} (this process also may lead to the formation of the compound nucleus, if in the course of the nucleon-nucleon cascades initiated by each of the four excited nucleons, no preequilibrium particles are emitted), to α-particle emission after one or more α-nucleon interactions, σ_α, and to fragmentation of the α-particle following one α-nucleon interaction, a process which leads to a five-particle, one-hole configuration, σ_{51}.

Fission is assumed to compete with particle and γ-ray emission only after formation of a fully equilibrated system which may be either the compound nucleus or may be the residual left after one or more preequilibrium emissions. The formulas used for the fission width and the ratio of the fission width to particle emission width are discussed in Ref. 8. We neglect the possibility of transmission through the barrier and we take account of the double humped nature of the barrier of most of the nuclei involved in the reactions simply by assuming that fission may occur only when the excitation energy of the nucleus is greater than the height of the higher barrier. This approximation is rather rough; on the other hand, the use of a more refined theory requires a great number of parameters which are unknown or poorly defined (barrier widths, heights of secondary barriers, and transmission coeffcients). This point is discussed below.

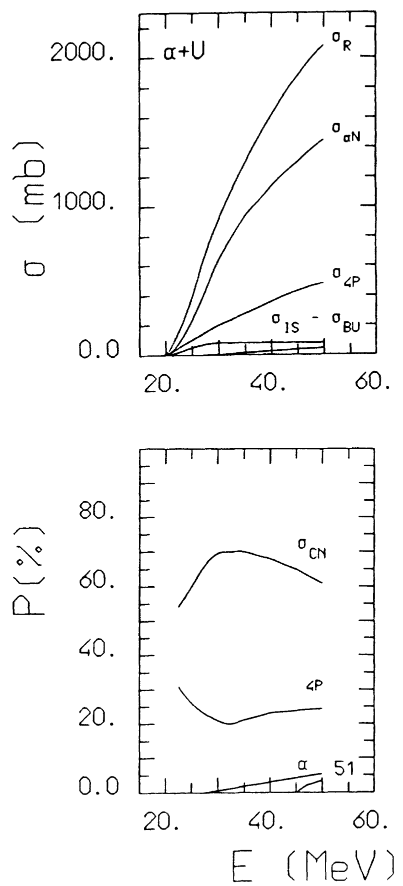

FIG. 1. In the upper part of the figure, the values of the reaction cross section and the cross sections corresponding to the different α-nucleus interactions in the case of uranium isotopes are reported. In the lower part are shown the percentages of cross sections leading to compound nucleus formation at the end of a series of α-nucleon interactions (CN), α-particle fragmentation to 4 nucleons (4*P*), α-particle inelastic scattering after one or more α-nucleon interactions (α, mean field inelastic scattering is not included), α-particle breakup after one α-nucleon interaction, leading to a five-particle, one-hole configuration (51).

Table II reports the fission barriers used in the calculations. They correspond (i) to the compound nuclei created in the various reactions, (ii) to the compound nuclei minus 1–5 neutrons, (iii) to the compound nuclei minus one proton and 1–4 neutrons, and (iv) to the compound nuclei minus one alpha particle (emitted in the preequilibrium cascade) and 1–4 neutrons.

Most of the calculated cross sections are highly sensitive to the fission barrier since fission is overwhelmingly the predominant mode of decay. Many of the values reported in the table have been obtained after several trials, by small adjustments of the values reported in the compilation of Dahlinger *et al.*;[26] the remaining values are those predicted by the droplet model with inclusion of shall corrections.[27] The table also reports the extremes of the experimental values reported by various authors (taken from Ref. 26). The measured fission barriers are characterized by an uncertainty which usually is 0.2–0.3 MeV, and in some cases as great as 0.4. From the table, it may be seen that most of the values used in the calculations are within the interval of the experimental values reported and even when outside this interval, are usually compatible, within the quoted uncertainties, of the smallest experimental value.

The only nuclides for which values smaller than experiment (smaller than the lowest acceptable value within the limits of accuracy reported) are used are ^{239}Pu and ^{234}Np. However, there is no doubt that the values used tend to be nearer the smallest value listed. This is understandable since, as mentioned above, in evaluating the fission probability, penetration through the barrier was not included. This approach leads to an underestimate of the barrier heights since due to the rapid increase with excitation energy of the level density above the saddle point, fission occurs in most of the cases slightly below the maximum of the barrier.

A second sensitive parameter in the calculated cross sections is R ($=a_f/a_n$), the ratio of the level density parameters of the fissioning nucleus at saddle point and the neutron residual. In these calculations we took $R=1.09$ in the cases of fission induced on ^{232}Th and U isotopes as previously determined,[8] and $R=1.11$ in the case of reactions induced on ^{237}Np and the Pu isotopes.

As in our previous calculations the computation technique adopted as a Monte Carlo approach.[8,9,20–23] In most of the cases, the calculated values of the excitation functions are characterized by small statistical uncertainty and one can draw through them, without ambiguity, a continuous line; in the case of a few, corresponding to very small cross sections, the statistical uncertainty affecting the theoretical estimates is so large that this cannot be done. In these cases, the theoretical estimates are reported with the corresponding statistical error.

IV. COMPARISON BETWEEN EXPERIMENT AND THEORY

The comparisons of the calculations to the experimental data have been grouped according to the types of reaction and are reported in Figs. 2–12. The reason for doing so is that, according to the theory, apart from minor fluctuations due to differing binding energies and fission barriers, the contributions to the excitation functions of the various preequilibrium events, including modes of breakup, inelastic scatter, etc., are similar at the same energy for all target nuclei considered.

A. (α,p) and (α,n) excitation function (Figs. 2 and 3)

The excitation function for these reactions sensitively depends on the value of σ_{4P} since, in this approach, this is the predominant interaction mode which contributes to the emission of a single fast nucleon; a second small component comes from σ_{51}, dissolution of the alpha leading to a five-particle, one-hole configuration. These cross sections, arising entirely from preequilibrium events, do not test the fission barrier of the compound nucleus and only marginally that of the residual nucleus. The agreement between theory and experiment is reasonably good in all cases, and, except in the case of ^{232}Th, within the accuracy one may expect *a priori* for the theoretical calculation.

B. ($\alpha,2n$) and ($\alpha,3n$) excitation functions (Figs. 4 and 5)

The contributions to the calculated excitation functions of two (three) neutron evaporations from a compound nucleus (the dominant component in the maxima following the reaction threshold) and of processes in which at least one neutron is emitted in the preequilibrium phase (dominant in the long flat tail at higher energies) are immediately evident. The agreement is rather good, within the limits of accuracy already given, without indication of any systematic disagreement, except in the case of ^{237}Np and, to a lesser extent, in the case of ^{234}U where, apparently, the experimental excitation functions are displaced by 2.5 to 5 MeV toward the higher energies. The magnitude of both the maxima and tail are well reproduced, the first evidence that the fission-neutron evaporation competition is well accounted for in these calculations. It is also worth mentioning that it was in the experimental data for the ^{237}Np ($\alpha,3n$) reaction that the authors refer to a γ ray which is nonexistent in the Table of Isotopes.[4]

C. ($\alpha,4n$) excitation functions (Fig. 6)

The dominant contribution, in this energy range, is 4 neutron evaporation from the compound system. In the case of ^{233}U, ^{234}U, and ^{237}Np, since the production cross section is very small, the theoretical estimates (made by a Monte Carlo technique, as described above) are characterized by rather large statistical errors and are given in the figure by the open circles with associated, purely sta-

TABLE II. The table gives the height in MeV of the fission barriers $B(Z,N)$ used in the calculations, reported as a function of Z and N. Below each value, in parentheses, the experimental values are given (Ref. 26). The values marked with an asterisk are the droplet model estimates corrected for shall effects (Ref. 27).

Z / N	Cm 96	Am 95	Pu 94	Np 93	U 92	Pa 91	Th 90	Ac 89
150	6.00 (5.7–6.2)	5.90 (5.6–6.1)	5.40 (5.4–5.6)	4.73*	4.89*	5.05*	5.21*	5.42*
149	6.40 (6.2–6.4)	6.20 (6.2–6.4)	6.10 (5.7–6.1)	4.74*	4.92*	5.09*	5.28*	5.46*
148	5.60 (5.8–6.3)	6.00 (5.8–6.3)	5.60 (5.6–6.1)	4.74*	6.00* (5.7–6.4)	5.10*	5.32*	5.52*
147	5.50 (5.8–6.4)	6.40 (6.3–6.5)	6.30 (5.8–6.3)	4.73*	6.60* (6.3–6.6)	3.89*	5.34*	5.55*
146	5.40 (5.8–6.3)	5.70 (5.7–6.4)	5.80 (5.6–6.7)	5.90 (5.7–6.3)	5.90 (5.7–6.1)	4.25*	5.35*	5.57*
145	5.64* (6.0–6.3)	6.05 (6.3–6.5)	5.60 (6.2–6.4)	6.00 (6.0–6.2)	6.40 (6.1–6.4)	4.69*	5.35*	5.57*
144	5.70	5.30 (5.5–6.3)	5.30 (5.5–6.2)	5.70 (5.7–6.2)	5.70 (5.6–6.1)	4.92*	6.50 (6.1–6.5)	5.56*
143	5.70	5.70 (5.3–5.4)	5.20 (5.3–5.9)	5.70 (5.7–5.8)	5.90 (5.9–6.1)	5.40	6.40 6.3)	4.97*
142	5.18*	4.80 (4.8)	5.00 (4.5–5.0)	5.60 (5.5–6.0)	5.60 (5.6–6.2)	6.00 (6.0–6.2)	6.20 (5.8–6.2)	5.08*
141	5.10*	5.44*	5.00 (4.6–5.1)	4.37 (5.3–5.6)	5.70 (<5.8)	6.10 (6.1–6.6)	6.20 (6.1–6.5)	5.61*
140	4.54*	4.96*	5.80 (5.8)	5.40 5.0–(5.4)	5.50 (5.2–5.6)	5.90 (5.9–6.0)	6.50 (6.5–7.0)	5.78*

tistical, error bars. The comparison of theory and experimental data does not indicate any systematic disagreement except, again, in the cases of ^{234}U and ^{237}Np where, as before, a shift in incident energy between the two seems to exist.

D. (α,pn) excitation functions (Fig. 7)

The (pxn) exit channels, in the case of these heavy nuclei, are predominantly produced in breakup processes of the incident alpha particle. Our predictions of the cross sections for these channels might be systematically wrong by a factor of 1.5 to 2 for all channels due to the difficulty of giving a reliable estimate of the total cross sections for the breakup interaction mode at such low energies. In the absence of direct experimental information the total cross section for α-particle breakup, σ_{BU}, was deduced by interpolating linearly from the thresholds expected for the different breakup processes to the values obtained at 80 MeV by an appropriate interpolation and/or extrapolation of the experimental cross sections measured by Koontz *et al.*[28] and Wu *et al.*[11] on various nuclei. To do this, the values measured for d, t, and ^{3}He channels on Al, Ni, and Zr at 80 MeV were extrapolated to Bi using

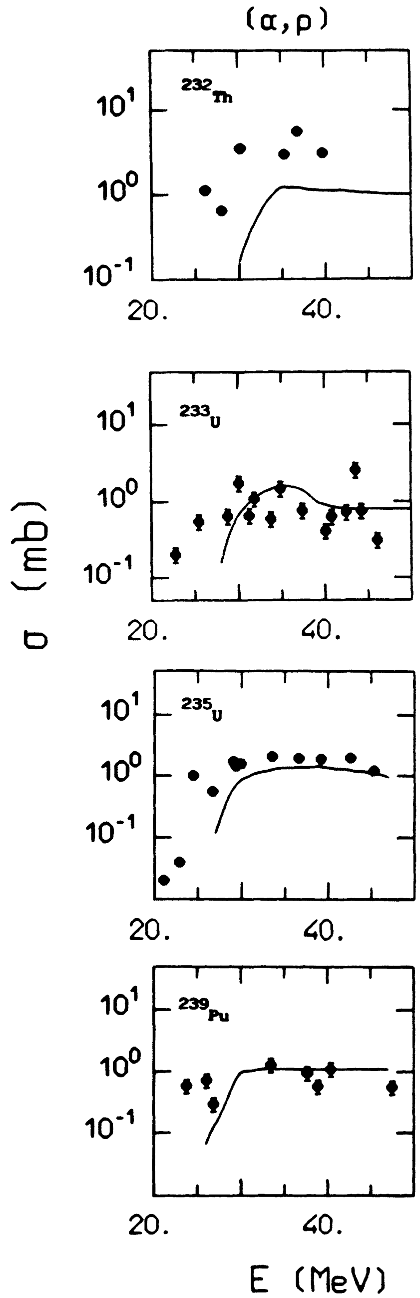

FIG. 2. Comparison between experimental and theoretical (full lines) excitation functions for the reaction (α,p). The experimental data are from the following references: ^{232}Th (Ref. 3); ^{233}U and ^{235}U (Ref. 2); ^{239}Pu (Ref. 1).

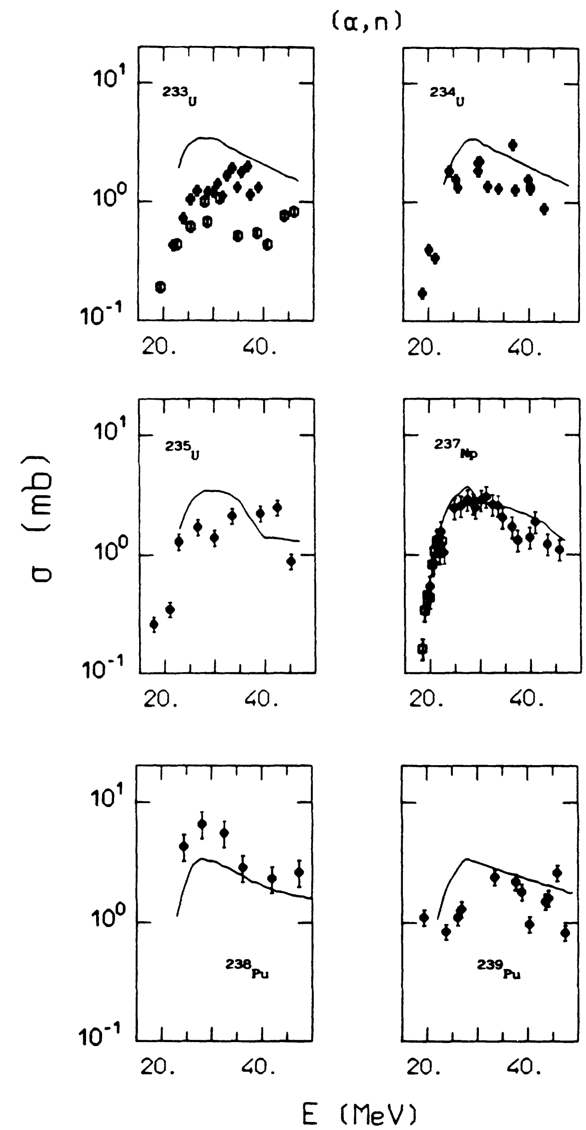

FIG. 3. Comparison between experimental and theoretical (full lines) excitation functions for the reaction (α,n). The experimental data are from the following references: ^{233}U (Ref. 2), open squares, and (Ref. 6), black dots; ^{234}U (Ref. 6); ^{235}U (Ref. 2); ^{237}Np (Ref. 4), black dots, and (Ref. 5), open squares; ^{238}Pu and ^{239}Pu (Ref. 1).

the same ratio between the corresponding cross sections found at 160 MeV for the reactions induced on Bi and these nuclei, and then, from Bi to the target nuclei of this work, using an $A^{1/3}$ dependence. This quite laborious procedure with extrapolations over both energy and target A is the reason for caution in estimating the reliability of the values given. The energy distributions of the various breakup particles, necessary to estimate the complementary energy given to the target nucleus, were estimated by the Serber approximation as suggested by Mutsuoka *et al.*[29] This approximation may lead to considerable error due to the distortions of the incident and outgoing particle wave functions expected at such low energies.

Two different contributions to the (α,pn) excitation functions are discernible in the calculated excitation functions (see in particular the case of ^{238}U). The contribution from α-particle breakup leading to one deuteron emission is dominant up to 35–40 MeV; thereafter the contribution from α-particle fragmentation becomes equally important. Except for the case of ^{232}Th, the agreement with the data is quite satisfactory, within the limits of accuracy expected.

E. $(\alpha,p2n)$ excitation functions (Fig. 8)

According to our calculations, the dominant contribution to these reactions from the threshold up to about 30–35 MeV is from α-particle breakup leading to triton emission; thereafter the contribution from α-particle breakup leading to deuteron emission becomes predominant. At the higher energies, α-particle fragmentation to four particles also provides a sizable contribution. The shape of the experimental excitation functions is reason-

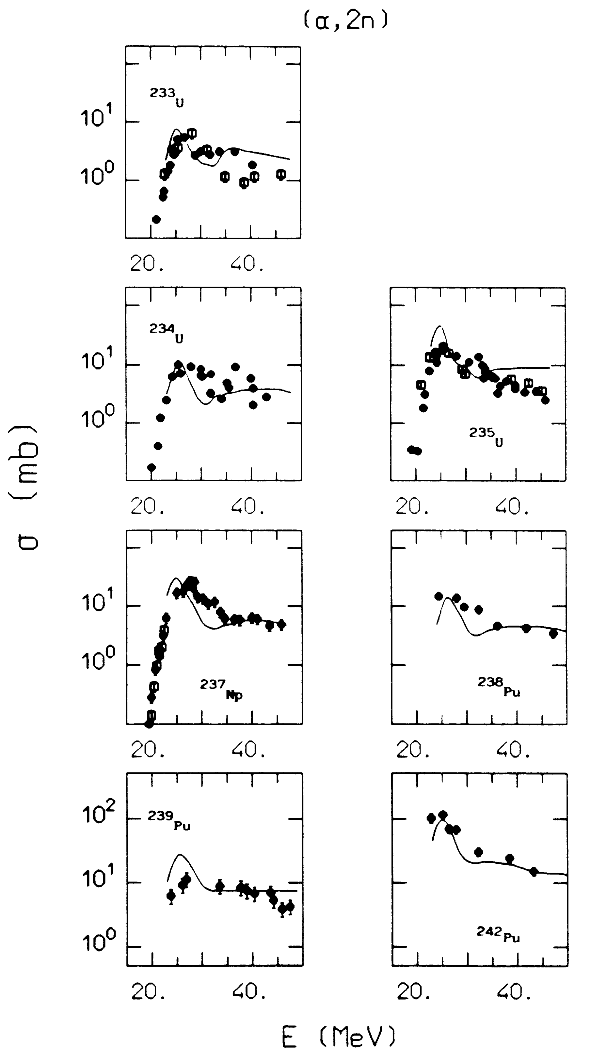

FIG. 4. Comparison between experimental and theoretical (full lines) excitation functions for the reaction $(\alpha,2n)$. The experimental data are from the following references: ^{233}U (Ref. 2), open squares, and (Ref. 6), black dots; ^{234}U (Ref. 6); ^{235}U (Ref. 2), open squares, and (Ref. 6), black dots; ^{237}Np (Ref. 4), black dots, and (Ref. 5) open squares; ^{238}Pu, ^{239}Pu, and ^{242}Pu (Ref. 1).

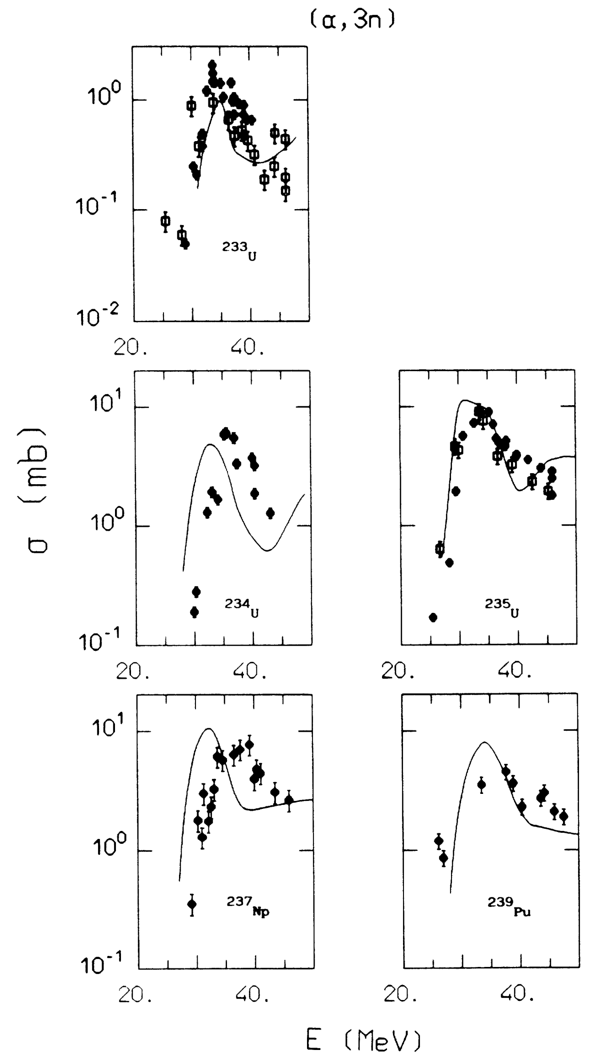

FIG. 5. Comparison between experimental and theoretical (full lines) excitation functions for the reaction $(\alpha,3n)$. The experimental data are from the following references: ^{233}U (Ref. 2), open squares, and (Ref. 6), black dots; ^{234}U (Ref. 6); ^{235}U (Ref. 2), open squares, and (Ref. 6), black dots; ^{237}Np (Ref. 4); ^{239}Pu (Ref. 1).

ably well reproduced in all the cases and the agreement between the calculated and experimental absolute cross sections is in all cases within the limits of accuracy expected.

F. $(\alpha,p3n)$ excitation functions (Fig. 9)

Similar to the $(\alpha,p2n)$ excitation functions, the dominant contribution from the threshold up to 40–45 MeV is from α-particle breakup leading to triton emission; thereafter the contribution of breakup to a deuteron becomes predominant. At the higher energies, also in this case, α-particle fragmentation provides a sizable contribution. In the case of ^{232}Th, the agreement between the data and the theory is reasonably good. In the case of ^{233}U, the experimental values scatter rather considerably, preventing any definite conclusions from being reached.

G. $(\alpha,2pn)$ excitation function (Fig. 10)

There is only one excitation function of this type, that induced on ^{232}Th. According to the theory, the only contribution is from α-particle breakup leading to the emission of a ^{3}He. Unfortunately—since these experimental

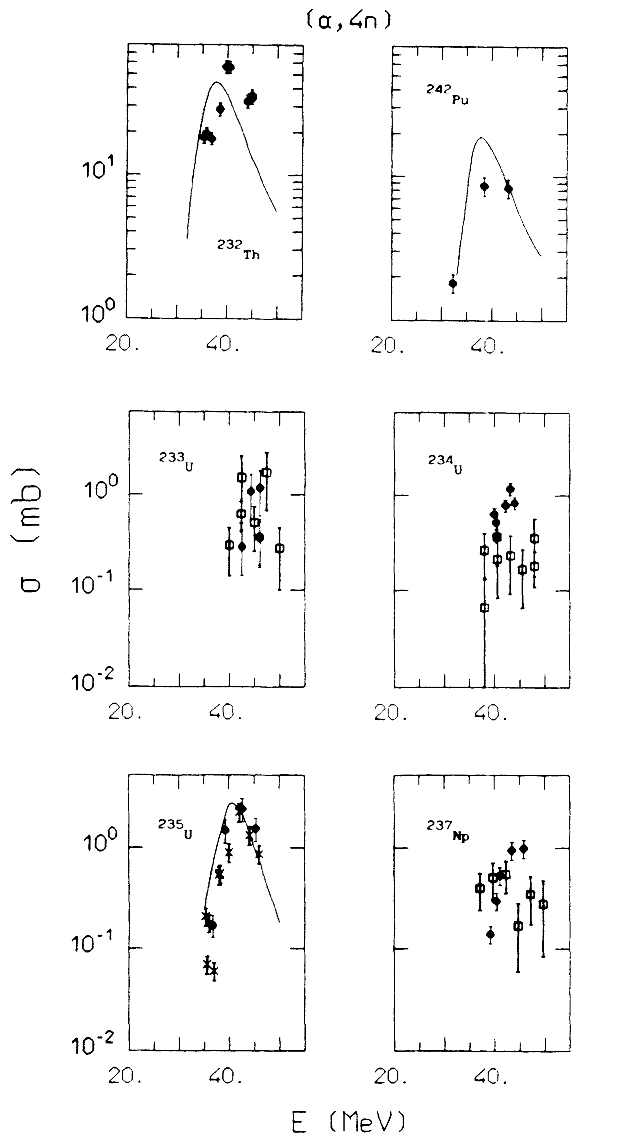

FIG. 6. Comparison between experimental and theoretical (full lines, and in the case of ^{233}U, ^{234}U, and ^{237}Np, open squares with error bars) excitation functions for the reaction $(\alpha,4n)$. The experimental data are from the following references: ^{232}Th (Ref. 3); ^{233}U (Ref. 2); ^{234}U (Ref. 6); ^{235}U (Ref. 2), black dots, and (Ref. 6), crosses; ^{237}Np (Ref. 4); ^{242}Pu (Ref. 1).

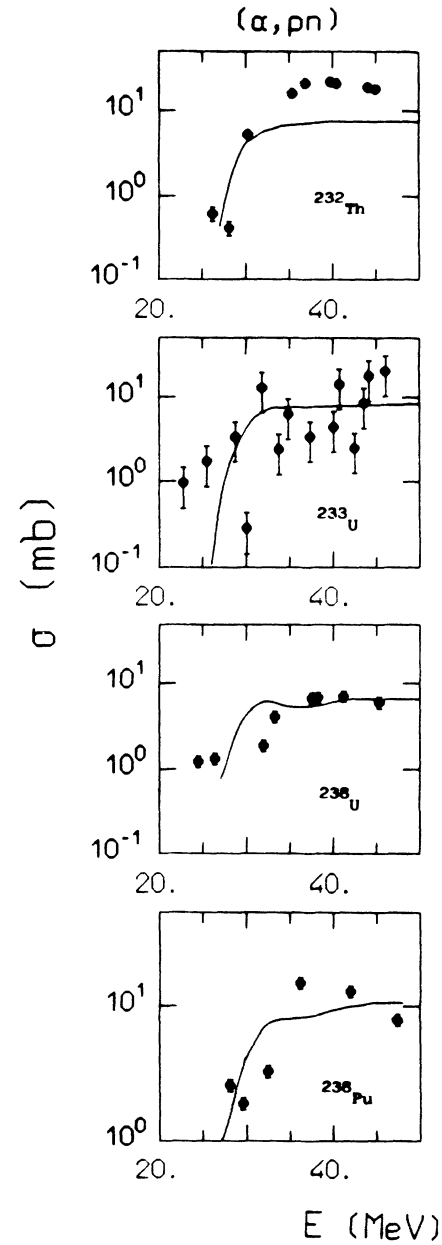

FIG. 7. Comparison between experimental and theoretical (full lines) excitation functions for the reaction (α,pn). The experimental data are from the following references: ^{232}Th (Ref. 3); ^{233}U and ^{238}U (Ref. 2); ^{238}Pu (Ref. 1).

data would provide a fine test of our evaluation of this breakup probability—the experimental values scatter rather considerably, preventing us from reaching any definite conclusions, although it is noteworthy that the agreement between theory and calculation is within the limits of expected accuracy.

H. (α,an) excitation functions (Fig. 11)

The dominant contribution to the (αn) exit channel comes from the mean field inelastic scatter of the incident α particle, with a contribution from α-nucleon scatter. An accurate calculation of the first process by coupled channel calculations is well outside the scope of the present work and the cross section values adopted for this process were taken from a previous analysis of spectra and excitation functions which we performed.[22] It was deduced by assuming this process to be the dominant contribution to the experimental inelastic α-particle spectrum up to about the isoscalar giant quadrupole resonance. Due to this very approximate procedure, we do not expect reliable theoretical estimates of these excitation functions below about 35 MeV. The calculated cross sections above this energy display a linear dependence on the incident α-particle energy, in good agreement with the experimental data.

I. (α,f) excitation function (Fig. 12)

The calculated fission cross sections for ^{233}U and ^{235}U (full line, the differences between the calculated values for the two nuclei are indistinguishable in the drawing), reproduce very satisfactorily the experimental data. The only value which considerably differs from the theoretical estimate is that for ^{233}U at 44.3 MeV; in this case, the reported experimental fission cross section was greater than the value adopted in this work for the total reaction cross section (1880 mb),[24] so one cannot hope to find closer agreement by any parameter adjustment.

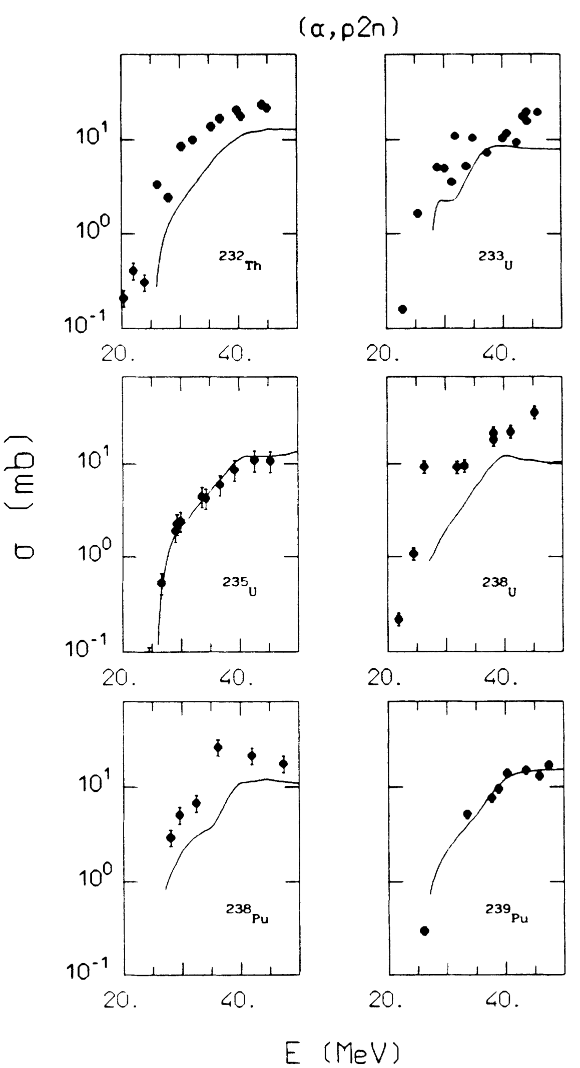

FIG. 8. Comparison between experimental and theoretical (full lines) excitation functions for the reaction (α,$p2n$). The experimental data are from the following references: ^{232}Th (Ref. 3); ^{233}U, ^{235}U, and ^{238}U (Ref. 2); ^{238}Pu and ^{239}Pu (Ref. 1).

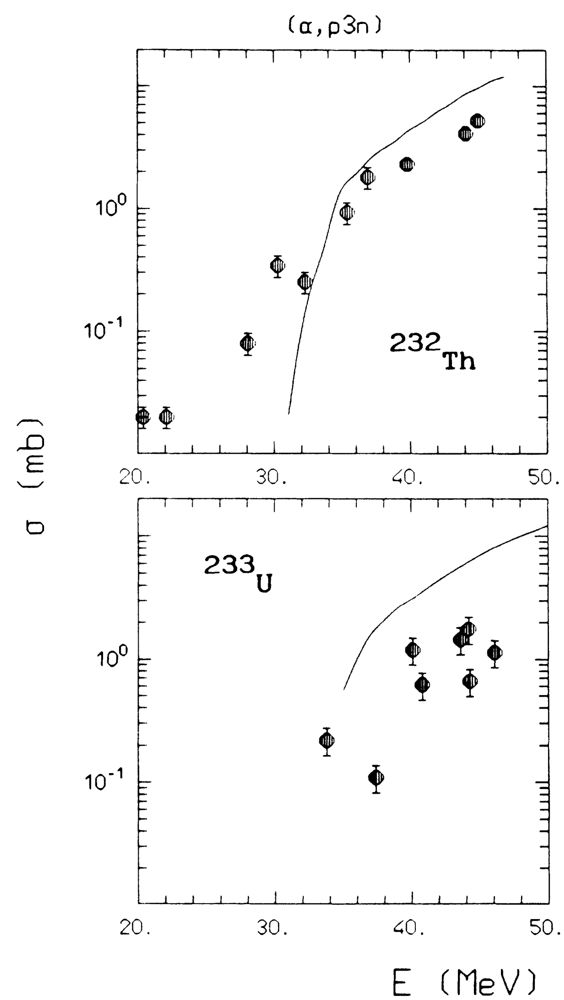

FIG. 9. Comparison between experimental and theoretical (full lines) excitation functions for the reaction (α,$p3n$). The experimental data are from the following references: ^{232}Th (Ref. 3); ^{233}U (Ref. 2).

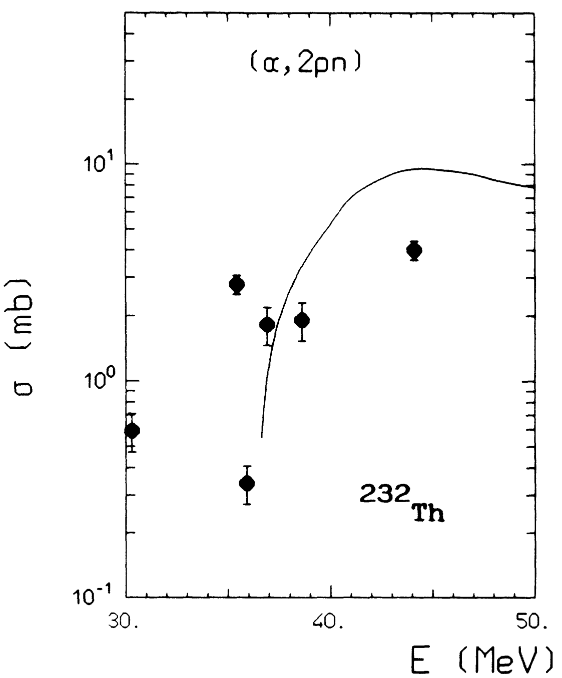

FIG. 10. Comparison between experimental and theoretical (full line) excitation functions for the reaction ^{232}Th $(\alpha,2pn)$. The experimental data are from Ref. 3.

V. DISCUSSION

A. The fission competition

At incident alpha-particle energies below 50 MeV, formation of a compound nucleus is the predominant event whether or not the alpha particle breaks up during the intranuclear cascade. The deexcitation of this compound nucleus, regardless of the target identity, or of the specific value of the excitation energy, leads with a probability ranging from 85% to 99% to fission. Thus direct calculation of the fission cross section followed by comparison to the experimental values is largely meaningless because any reasonable set of fission barriers will yield similar results. Indeed the scatter in the experimental results on σ_f, including their quoted uncertainties, is considerably greater than the total cross section for all nonfission events up to 30 MeV, and quite comparable in magnitude at all energies.

Therefore, computation of the fission cross section is not in itself a sensitive test of the fission competition in these experiments. However, examination of the nonfission excitation functions of reactions proceeding almost entirely via compound nucleus formation and neutron evaporation does provide a sensitive test. The (α,xn) cross sections are all small, accounting together for approximately 5% of the reaction cross section. The stepwise evaporation of, for example, three neutrons, tests the fission-evaporation competition three times with the final calculated cross section carrying a third power dependence on the ratios Γ_f/Γ_n encountered along the chain.

With seven target nuclei and eighteen $[\alpha,(2\text{–}4)n]$ excitation functions having been considered (Figs. 3–6), it may be inferred that the approach taken to the calculation of Γ_f/Γ_n, including the simple approximation to the fission barrier, seems to reproduce the fission competition in a satisfactory manner. The magnitude of all excitation functions are quite reasonable, even in the case of $(\alpha,4n)$ reactions which, typically, peak at a few mb (out of 1000–2000 between 30 and 50 MeV). In particular, the agreement is quite comparable to that found in the works of Delagrange *et al.*[6,30] and Gilat *et al.*[31] who used a highly parametrized, double-humped, fission barrier.

At first, this must be considered surprising as one might expect the more sophisticated treatment to yield far more accurate results. However, it is pointed out in Ref. 6 that at excitation energies above 13 MeV the fission probability is largely determined by the height of the second (lower) fission barrier, and this in turn is chosen "to fit the fission-evaporation competition." This indirectly supports the very different approach of this work; the "best-fit" heights of the lower barriers of Ref. 6

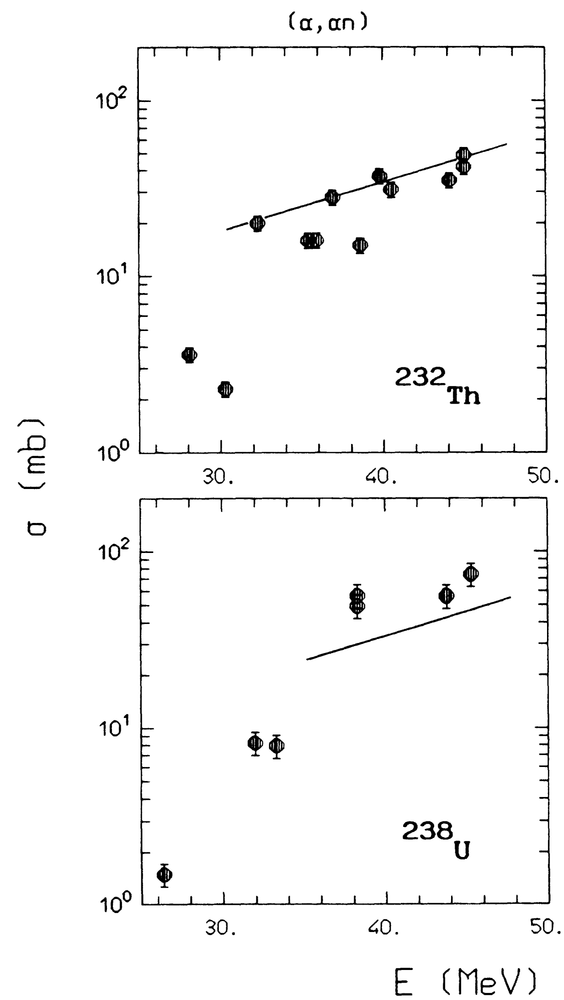

FIG. 11. Comparison between experimental and theoretical (full lines) excitation functions for the reaction $(\alpha,\alpha n)$. The experimental data are from the following references: ^{232}Th (Ref. 3); ^{238}U (Ref. 2).

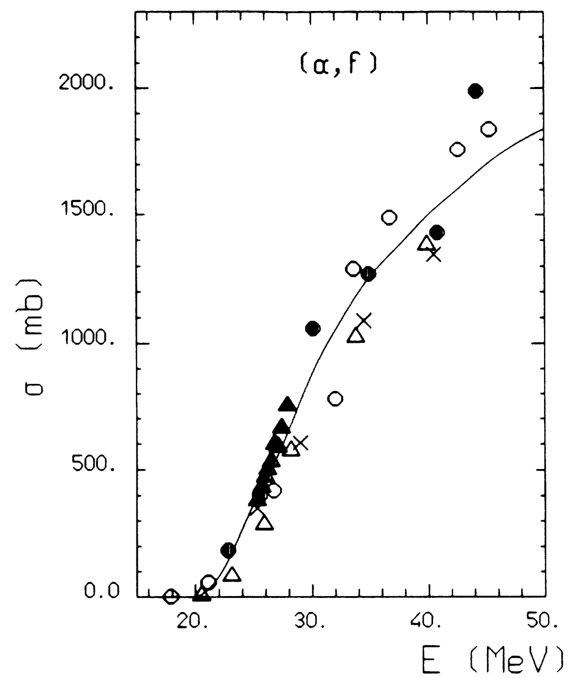

FIG. 12. Comparison between experimental and theoretical (full line) excitation functions for the reactions ^{233,235}U (α,f). The experimental data are from Ref. 2, black and open dots; Ref. 16, black triangles; Ref. 17, open triangles and crosses.

are in general agreement with our choice of fission barriers which tend to the low side of experimental values of the higher barrier when their uncertainties are included.

Therefore, only in the final evaporation step, when the excitation energy $E^* < 13$ MeV, might one expect differences to be apparent between the current OMEGA model approach and that of Refs. 6 and 31. For statistical reasons, these differences would be most apparent in the magnitude of the peaks of the (α,xn) excitation functions. However, comparison with the earlier calculations of the fit to the experimental data shows no discernible trends. Thus, it must be concluded that while the highly parametrized (in heights, widths, and curvature) double-humped fission barriers are both more realistic and more versatile (for example, fission isomers may be calculated), there is no obvious improvement over the use of a single fission barrier characterized only by its height in determining the fission-evaporation competition.

B. Interaction modes of the alpha particle

The goal of this work was to examine the effects of the several α-nucleus interaction mechanisms on the spallation excitation functions. Explicitly included are four types of events in decreasing order of likelihood [Fig. 1(a)] (1) interactions in which the alpha undergoes interactions with individual nucleons, and may be reemitted; (2) events in which the alpha dissolves into four nucleons, either under the influence of the nuclear potential leading to a four-particle initial configuration, or following collision with a nucleon, producing a five-particle, one-hole initial configuration; (3) inelastic scatter of the alpha particle from the mean potential of the target nucleus; and (4) binary fragmentation of the alpha into various combinations.

1. α-nucleon interactions

At the low incident energies we are considering, the α-nucleon interactions normally lead to production of the compound nucleus and account for 60–70 % of all events. From 30 to 50 MeV of incident energy, the reemission of the alpha particle after one or more interactions with nucleons increases in probability to 6–7 % of all events. It is a necessary component of the $(\alpha,\alpha xn)$ excitation functions, accounting for their increase between 35 and 50 MeV. Figure 1(a) and 1(b) may be analyzed to show that at the highest energies this mechanism accounts for about one-half of the cross section in which an α particle is emitted, the other half coming from mechanism (3) discussed below. Unfortunately, little data exist (Fig. 11) which accurately define the extent of occurrence of this mechanism's contribution to the $(\alpha,\alpha xn)$ events. In that the mechanism is, however, one of the two prime components of compound nucleus production, and the general agreement of the magnitude of the peaks of the (α,xn) reactions is well reproduced, it may be assumed that this mechanistic component is evaluated with reasonable accuracy. This is significant in that it represents a fundamentally different approach from the previous calculations[6,31] in which the dissolution of the alpha particle into four nucleons was assumed. In fact most of the α-nucleon interactions lead to the formation of a compound nucleus without previous preequilibrium emissions. Only in a small number of cases does the α particle dissolve to four nucleons initiating an intranuclear cascade that may lead to preequilibrium emission.

The assumption that in all cases one produces a composite nucleus in a configuration characterized by a number of nucleons equal either to 4 or 6 led Alexander *et al.*[4,6] to overestimate the probability of emission of preequilibrium particles and, to compensate for that, to assume (in evaluating the decay rate for internal transitions) values of the average residual two body transition matrix element $\overline{|M|^2}$ substantially higher than those usually reported in literature. In our approach we do not need to change any of the parameters employed in previous analyses of the experimental data.[9,14,20–23]

A second difference between our calculations and those of Alexander *et al.*[4,6] is that in their calculations the tails of the $(\alpha,2n)$ and $(\alpha,3n)$ reactions are not well reproduced; thus Delagrange *et al.*[6] suggest that multiple preequilibrium nucleon emission must be invoked to account for the cross sections observed. In this work, the tails of these excitation functions are reproduced reasonably well (Figs. 3 and 4) without the need of multiple preequilibrium emissions that, according to our calculations, as discussed later, are very unlikely in this energy range.

2. Breakup of the alpha to four nucleons

The probability of breakup or dissolution of the α particle increases in the incident energy interval here considered and amounts to about 20–25 % of the reaction cross section. These events also lead to production of the compound nucleus with large probability. They comprise about one-third of compound nucleus production at the highest energies. There are two subcategories: breakup in the nuclear field, producing a 4p configuration, and breakup following a nucleonic collision, which produces a 5p-1h state. The 4p configuration is the primary component of the (α,p) and (α,xn) $(x=1-4)$ excitation functions in the high energy tails. Satisfactory reproduction of these excitation functions at the highest incident energies lends support to our estimate of the probability of occurrence of a 4p initial configuration shown in Fig. 1(b). It is worth noting that the probability of multiple preequilibrium emissions from 4p or 5p-1h states seems to be very unlikely. The calculated average number of preequilibrium neutrons increases from ≈ 0.023 at 22.5 MeV to ≈ 0.125 at 50 MeV, while for preequilibrium protons emitted the comparable values are zero below ≈ 27.5 MeV, increasing to ≈ 0.024 at 50 MeV. Clearly then, the probability of two preequilibrium emissions of fast particles is vanishingly small.

3. *Inelastic scatter of the α particle from the nucleus*

Accounting for about 80 mb of the reaction cross section at energies above 25 MeV, inelastic scatter of the alpha from the mean field of the nucleus mainly contributes to the $(\alpha,\alpha xn)$ reactions as one of two components, the other being the reemission of alphas as discussed in mechanism (1). At energies above 30 MeV, this second path increases from zero to a comparable cross section at 50 MeV. Too little data exist to seriously test the relative yields of these two mechanisms, particularly at lower incident energies. It may only be summarized that the increase in the calculated cross sections above 35 MeV arises from the latter mechanism, while those few events below ≈ 35 MeV (with large statistical uncertainty) arise purely from inelastic scatter.

4. *Binary fragmentation*

It has already been mentioned that the probability of emission of protons in the preequilibrium cascade is very small, occurring in less than 1 in 100 events at energies up to 40 MeV. Proton evaporation from compound systems is negligible due to the presence of the Coulomb barrier. Thus the cross section of effectively all reactions in which a proton is emitted is determined by the evaluation of binary breakup of the alpha particle into $n+{}^3$He, $d+d$, or $p+t$. Figures 7–10, comprising thirteen excitation functions, show the effect of the various modes of binary fragmentation. Because of the complexity of the interaction, it is worth reviewing the way it is included in this work.

It is assumed that the incident alpha particle divides in two with one fragment being emitted at forward angles with near beam velocity, while its complement is absorbed. Wu *et al.*[11] and Koontz *et al.*[28] found the energy distribution of such fragments to be approximately Gaussian centered on the beam velocity and having a variance increasing with beam energy. The complementary fragment (and its defined energy distribution) is then absorbed and initiates a preequilibrium cascade.

As discussed in Ref. 9 the breakup processes leading to emission of one single proton or neutron with the accompanying fragment being absorbed by the target nucleus is largely accounted for by the breakup mechanism to four particles discussed above. The extrapolations necessary from the experimental data to provide input to the present calculation in all the cases in which a deuteron, a triton, or a helion is emitted in the breakup process have been described in Sec. IV D above.

Even if the emission of a preequilibrium proton followed by neutron evaporation from fragmentation of the α to four particles provides a sizable contribution to the (α,pxn) reactions [especially to the (α,pn)], the breakup to two deuterons is the predominant mode of interaction in the (α,pn) reaction at energies up to 40 MeV and in the $[\alpha,p(2-3)n]$ tails at higher energies. This is significant because the first tests the amount of binary fragmentation to deuterons, while the second tests the energy spectrum of the emitted deuteron since absorption of its complement is responsible for the neutron evaporation.

Similarly, breakup to $p+t$ is the dominant contribution to the $[\alpha,p(2-3)n]$ reactions at the lower energies and suggests that our deduced yield of this fragmentation mode, however tenuous the extrapolation may be, is reasonably reproducing the experimental data.

Furthermore, the agreement between theory and experiment for the $(\alpha,2pn)$ reaction, given the large experimental fluctuations, is quite acceptable. Binary fragmentation to an $n+{}^3$He, with the ^{3}He being emitted in the forward direction at near beam velocity, is the only viable mechanism for this reaction. Once again, without a full treatment of the three fragmentation processes, no model calculation could account for this reaction.

It is fortunate that data exist for these reactions which clearly arise primarily from the processes of binary fragmentation, since it suggests strongly that such an approach may be workable for heavier incident beams. Treatment of the binary fragmentation of light heavy ions into two fragments, one behaving as a spectator with the complement initiating a preequilibrium cascade, has been used effectively in analyses of the interaction of ^{12}C with nuclei[32] and has even been extended to several other incident particles up to ^{20}Ne.[33] Several earlier works including our own[9,22–23] have used it in preequilibrium calculations.

VI. SUMMARY

The preequilibrium exciton model has been extended to include two features previously dealt with only separately, the competition of fission,[8] and reactions induced by α particles.[9] The necessity of incorporation of fragmentation modes for calculation of charged particle emission has been demonstrated; evaluation of each, despite a dearth of experimental data on such heavy target nuclei, has been carried out. The relative probabilities of the various interaction modes of the incident alpha have been evaluated and appear to reproduce rather well the excitation functions for all reactions, including those involving charged particle emission. For the first time, complete sets of excitation functions have been modeled for fissile nuclei. In particular, 46 excitation functions induced by alpha particles of up to 50 MeV from eight target nuclei have been calculated in a satisfactory manner. The model calculations are performed with a single set of input parameters, none of them varying with incident energy or target nucleus, and all evaluated *a priori* from previous work, existing experimental data, or extrapolations from them.

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