Spallation of Uranium and Thorium Nuclei with Bev-Energy Protons*

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Cross sections have been measured for the production of isotopes of uranium, protactinium, thorium, and actinium in the irradiation of U^{238}, U^{235}, and Th^{232} with 0.68- and 1.8-Bev protons. In addition, some yields were determined at other bombarding energies ranging up to 6.2 Bev. Calculations of the cross sections at 1.8-Bev bombarding energy were made, based on recent Monte Carlo calculations of the knock-on phase of the interaction, combined with published systematics of nuclear evaporation, and several assumptions as to fission-evaporation competition. Even without fission competition the calculated yields are considerably lower than the experimental ones, indicating a failure of the model for the knock-on phase of the reaction to predict sufficient probability for simple processes with low deposition energy. In view of the better agreement with experiment that previous workers have obtained with this type of calculation for 0.34-Bev protons on uranium, it is suggested that the present discrepancy may be due to an overemphasis of meson processes in the proliferation of the knock-on cascade in the calculation.

INTRODUCTION

HIGH-ENERGY nuclear reactions may be considered to take place in two steps[1,2]: a knock-on cascade in which a distribution of excited nuclei is produced, and subsequent de-excitation by nuclear evaporation with fission as a competing process. The extent to which fission competes with evaporation has been the subject of several studies. Experimental data concerning fission-evaporation competition in heavy nuclei excited to 8–35 Mev have recently been summarized by Vandenbosch and Huizenga.[3] Experimental studies of the competition at higher energies include those with thorium bombarded with protons of energies up to 100 Mev[4] and the study by Lindner and Osborne[5] of the spallation products from the bombardment of uranium and thorium with protons of up to 340 Mev. A number of analyses of the experimental data, particularly those of Lindner and Osborne,[5] have been published. Among the earlier treatments was that of Batzel[6] who concluded, from studies using a very simple model for the knock-on cascade and the available data for the ratio of fission to neutron evaporation in systems excited to about 10 Mev, that the fission-evaporation competition was essentially independent of excitation energy. On the other hand, an alternate analysis of the same data[5] by Shamov,[7] also by means of a simple, but different model, for the knock-on cascade led to the conclusion that fission competition was unimportant until the nucleus had been almost completely de-excited by evaporation. In more recent years, more accurate analysis of the data has become possible since detailed calculations via the Monte Carlo technique of the knock-on phase of high-energy nuclear reactions have been published. The work of Metropolis *et al.*[8] is the most sophisticated treatment published so far. The agreement between experimental spallation cross sections and the results of these calculations, when combined with a simple model for the evaporation process, has been good, although the calculations generally underestimate the cross sections for the (p,pn) and $(p,2p)$ reactions. The predictions of this calculation[8] as to the distribution in charge, mass, and excitation energy of the products from the interaction of heavy nuclei with high-energy protons has recently been employed by Lindner and Turkevich[9] in a further and more detailed analysis of the Lindner and Osborne data. The analysis employed data on low-energy fission-evaporation competition from a recent summary,[3] several assumptions as to the variation of the competition with energy, and a simple neutron evaporation model. The conclusions, in agreement with the conclusions of Batzel,[6] were that the competition between fission and evaporation is independent of excitation energy up to about 100 Mev.

The results of Metropolis *et al.*[8] have also been used as the starting point for further calculations by Dostrovsky, Fraenkel, and Rabinowitz.[10] Fission probability as a function of nuclear type and excitation energy was derived on the basis of the Bohr-Wheeler model,

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rather than from low-energy experimental data. The results obtained are compared with experiment later in this paper.

The present paper presents experimental information on the spallation reactions of U^{238}, U^{235}, and Th^{232} with protons at higher bombarding energies. It also extends the calculations to these higher bombarding energies by means of a model similar to that used by Lindner and Turkevich,[9] but with the inclusion of charged-particle evaporation.

EXPERIMENTAL

Bombardments were begun at the Brookhaven National Laboratory Cosmotron at proton energies of 1.0, 2.0, and 3.0 Bev, and were continued at the Lawrence Radiation Laboratory at Berkeley, at the 184-in. synchrocyclotron with 680-Mev protons and at the Bevatron with 1.8- and 6.2-Bev protons.

The target foils used were 0.002-in. thick natural uranium, 0.001-in. enriched uranium (93.2% U^{235}, 1.0% U^{234}, 0.5% U^{236}, 5.3% U^{238}), and 0.002-in. thorium. The beam intensity was determined by the activation of a 0.003-in. aluminum monitor foil intercepting the same flux as the target foil. The $Al^{27}(p,3pn)Na^{24}$ cross section was taken to be 11.0 mb at 680 Mev[11] and 10.5 mb for 1.0 Bev and above.[12–15] The recoil loss of spallation products from both the target and monitor foils was expected to be less than 2%.

The elements uranium, protactinium, thorium, and actinium were separated chemically from the irradiated targets. Outlines of the chemical procedures and methods of sample preparation are given in Appendix I. Three methods of radiometric assay were employed: alpha counting in a windowless 2π proportional counter or ion chamber (U^{228}, Pa^{227}, Th^{226}, Th^{225}), alpha pulse-height analysis with a Frisch grid ion chamber (U^{232}, U^{230}, U^{229}, Pa^{229}, Pa^{228}, Pa^{227}, Th^{228}, Th^{227}, Ac^{226}, Ac^{225}, Ac^{224}), and beta counting with an end-window proportional counter (U^{237}, Pa^{237}, Pa^{235}, Pa^{234}, Pa^{233}, Pa^{232}, Th^{234}, Th^{233}, Th^{231}). In addition, some nuclides (Np^{238}, Np^{236}, Ac^{228}) were assayed by alpha-pulse analysis of a long-lived daughter activity. Beta counting was performed through an aluminum foil (~ 8 mg/cm^2) to absorb alpha particles, and the efficiency for this system was determined with samples calibrated by 4π counting[16] or with beta activities in equilibrium with long-lived alpha-active parents. The counting efficiencies of the alpha counters were determined by standards calibrated by 4π counting to be 50–53% depending on α energy. The counting efficiency of the gridded ion chambers for resolved alpha groups was determined in the same way to be 45–49%.

Chemical yields of elements other than that of the target were determined by the addition to the solution in which the target was dissolved of known amounts of long-lived alpha-active isotopes of the elements to be separated. The nuclides used for this purpose were Np^{237}, Pa^{231}, Th^{230}, and Ac^{227}. In the case of Ac^{227} a small correction was necessary for the amount of this isotope produced during the bombardment. The correction was estimated by assuming the Ac^{227} cross section to be the mean of the Ac^{226} and Ac^{228} cross sections.

For several of the elements studied, the presence of certain naturally occurring isotopes in the target foil had to be taken into account. The appropriate correction was determined by assay in an unirradiated foil or by calculation from the known composition of the foil. These corrections were necessary for Th^{231} and Th^{234} in natural uranium, Th^{228}, Th^{230}, Th^{231}, Th^{234}, and U^{232} in enriched uranium and Th^{228} and Th^{230} in thorium.

The measured cross sections are presented in Table I. These represent independent yields unless otherwise indicated, and the U^{235} yields have been corrected for the 5.3% U^{238} content. Half-lives and decay branching ratios given by Strominger *et al.*[17] were used. However, the beta activity of protactinium samples separated from bombarded U^{238} targets showed no evidence of a 39-minute half-life as reported[18] for Pa^{237} although it was expected that the 39-minute activity would have been formed with a substantial yield were the assignment to Pa^{237} correct. The 10-minute activity that was observed was assumed[17] to be due to Pa^{237}.

The errors shown are generally the mean deviations of duplicate determinations. When the duplicates showed fortuitously good agreement or when a duplicate determination was not made, an estimate based on the compounding of the errors of measurement was employed. Generally this type of estimate was of the same order as that obtained from the agreement of duplicate determinations.

In Fig. 1, a contour plot on a table of isotopes is pre-

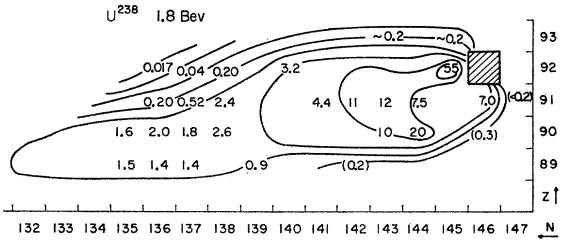

FIG. 1. Cross sections in millibarns for production of the indicated isotopes from U^{238} bombarded with 1.8-Bev protons. The target is indicated by the shaded box. Contour lines are drawn a factor of three apart.

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TABLE I. Cross sections in millibarns for production of the indicated isotopes with protons.

Target:	U^{238}					U^{235}		Th^{232}	
Energy (Bev):	0.68	1.0	1.8	3.0	6.2	0.68	1.8	0.68	1.8
Np^{238}	0.25±0.04								
Np^{236} (22 hr)	0.26±0.03								
U^{237}	63±10		55±10						
U^{232}	4.7±0.7		3.2±0.7			25±6			
U^{230}	0.23±0.04		0.20±0.04			1.3±0.3	6.3±1.3		
U^{229}	0.05±0.01		0.040±0.008			0.10±0.01	0.06±0.01		
U^{228}	0.024±0.005		0.017±0.004			0.017±0.003			
Pa^{237}	13±2		7.0±0.5	10±1	8±1				
Pa^{235a}	17±2		7.5±0.5	8.6±0.5	8.0±0.5	⩽0.2	⩽0.2		
Pa^{234} (6.6 hr)	22±4		12±2	12±2	12±2	23±4	20±4		
Pa^{233}	19±5		11±4	11±4	13±4	23±4	18±3		
Pa^{232}	6.5±0.8		4.4±0.5	3.3±0.5	4.6±0.5	6.6±0.8	4.3±0.6		
Pa^{229}	4.9±0.7		2.4±0.6	2.4±0.6	3.0±0.6	4.2±0.5	1.4±0.2	2.3±0.3	2.4±0.3
Pa^{228}	1.0±0.1		0.52±0.08	0.64±0.06	0.60±0.06	1.4±0.4	0.28±0.04	0.8±0.2	0.6±0.1
Pa^{227}	0.39±0.07		0.20±0.04	0.17±0.04	0.15±0.04	0.42±0.08	0.05±0.01	0.26±0.04	0.22±0.04
Th^{234}	5±3		20±10	6±2					
Th^{233}	7.5±1.0		10±2		9±2	0.5±0.2	0.3±0.2		
Th^{231a}	3±1			7.4±1.5		8±2	10±2	120±20	100±10
Th^{228}	2.7±0.2		2.6±0.2		2.5±0.2	1.5±0.5		25±4	
Th^{227}	2.3±0.2	2.2±0.2	1.8±0.2	1.9±0.2	2.1±0.2	1.7±0.2	1.9±0.2	15±3	15±2
Th^{226}	2.3±0.2	2.4±0.2	2.0±0.2		2.0±0.2	1.8±0.3	2.2±0.2	14±2	9.0±1.0
Th^{225}	1.5±0.2		1.6±0.2			1.2±0.4	1.8±0.2	12±2	6.5±1.0
Ac^{228}	1.1±0.2		0.9±0.2			0.48±0.10	0.24±0.06	5.3±1.5	8.0±0.7
Ac^{226}	1.5±0.2		1.4±0.2			0.83±0.08	1.0±0.2	5.5±1.4	7.3±1.5
Ac^{225}	1.6±0.2		1.4±0.1			1.1±0.2	1.1±0.2	5.0±0.8	8.2±1.0
Ac^{224}	2.1±0.2		1.5±0.1			1.5±0.2	1.2±0.2	5.4±0.7	5.4±1.0

[a] β^- cumulative yield.

sented for the yields from U^{238} bombarded with 1.8-Bev protons. The yields with a "∼" sign are actually from 0.68-Bev data and those in parentheses from a U^{235} target. The contours were drawn to be an estimated factor of three in cross section apart. The general mass and charge distribution of spallation products of a target bombarded with Bev-energy protons is evident. Also obvious is the strong effect of fission in sharply reducing the yields at high Z^2/A, indicated by the crowding of the contour lines in the upper left part of the figure.

CALCULATIONS

In common with the calculations of Lindner and Turkevich,[9] the present analysis used as a starting point the results of the calculations of Metropolis *et al.*[8] for the charge, mass, and excitation energy of the products of about 750 individual intra-nuclear cascades for the interaction of 1.8-Bev protons with U^{238} nuclei.[19] In the calculations for the reactions of U^{235} and Th^{232} with 1.8-Bev protons, the assumption has been made that the distribution of Z, A, and excitation energy would be similar to that for U^{238} but shifted in Z and A by an amount corresponding to the difference in the charges and masses of the target nuclei concerned. A program was written for the Brookhaven National Laboratory Merlin computer to perform an analysis of the de-excitation of the nuclei resulting from the Metropolis calculations. It used the results of unpublished calculations[19] due to Dostrovsky *et al.*[20] as to the relative probability for the emission of neutrons, singly-charged and doubly-charged particles, as a function of the excitation energy and neutron deficiency or excess of the evaporating nucleus. The average de-excitation per emitted nucleon was derived from the calculations of Dostrovsky *et al.*[20] as a function of deposition energy. The actual de-excitation per emitted nucleon was varied in a linear manner for successive evaporation events so as to preserve this average and to give a value of 8 Mev per nucleon near zero excitation.

Several modes of fission-evaporation competition were considered: (a) no fission competition with evaporation, (b) fission competition only at excitation energies above 100 Mev, (c) fission competition at all times. In all cases where fission competition was included, the probability for fission relative to that for evaporation was taken from the summary of low-energy data of Vandenbosch and Huizenga.[3] The exact input parameters are described in Appendix II. The calculation was in no sense a full statistical analysis, since for example, average de-excitation per nucleon emitted was used rather than the appropriate distribution in energy predicted by evaporation theory. It was also limited by the small number of cascades followed in the Monte Carlo calculation[8] of the knock-on phase of the interaction, which led to poor statistical accuracy for these results. A procedure[21] was adopted to add one to the number of

[19] Data kindly made available to the authors by Dr. G. Friedlander.

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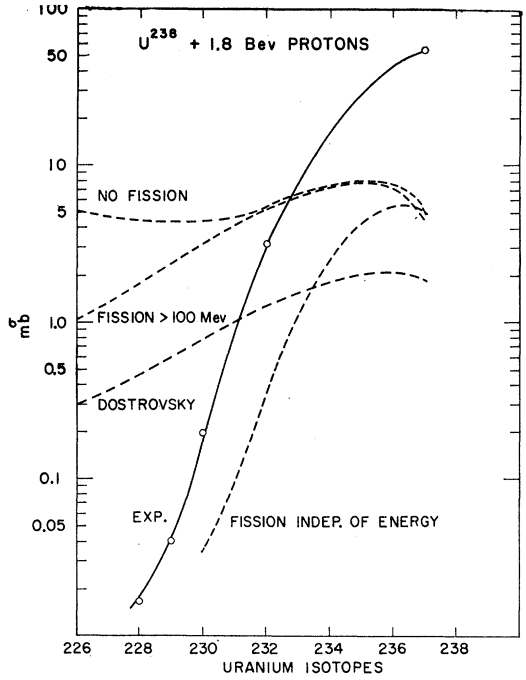

FIG. 2. The cross sections for the production of uranium isotopes from U^{238} bombarded with 1.8-Bev protons compared with those calculated with various assumptions as to fission-evaporation competition.

cascades that would lead to a certain product and then renormalize so that the total reaction cross section remained the same. This had the effect of smoothing the data and making the calculated curves upper limits in the regions where the knock-on calculations had the smallest statistical accuracy.

DISCUSSION

The present study, while performed at higher bombarding energies than that of Lindner and Osborne,[5] deals with the same product nuclides from the same targets, probably formed from knock-on cascades with similar deposition energies, namely of less than 100 Mev. Thus the conclusions of previous workers, such as Lindner and Turkevich,[9] that the fission-evaporation competition is independent of excitation energy up to about 100 Mev should apply to the interpretation of the present data.

In Fig. 2 experimental yields of uranium nuclides, from a U^{238} target plus 1.8-Bev protons, are compared with the present calculations. The use of a constant de-excitation per emitted nucleon of 10 Mev instead of the more detailed assumption described above was found not to affect appreciably the calculated yields. The curve labeled Dostrovsky comes from some unpublished results of the calculation described by Dostrovsky, Fraenkel, and Rabinowitz,[10] which used a Bohr-Wheeler formula for the variation of fission probability with excitation energies. Qualitatively this formula has the effect of making the fissile nuclei under consideration here less fissionable at high excitation energies. One concludes from Fig. 2 that a fission-evaporation competition independent of excitation energy is the only formula among those tried that reproduces even the general trend of the yields with mass number. However, even in this case one observes a discrepancy in the magnitude of the yields, not only for the (p,pn) product (as observed by previous workers)[9,22,23] but also for mass numbers up to ten units below that of the target. This is in sharp contrast to the excellent agreement with experiment obtained by Lindner and Turkevich[9] at 0.34 Bev for yields of the same nuclei from the same target.

In Figs. 3 to 6 experimental yields from U^{235}, U^{238}, and Th^{232} targets plus 1.8-Bev protons are presented according to reaction type. [The designation of a reaction (p,pxn), for example, is intended to describe a product-target relationship rather than a reaction mechanism.] Also presented are yield data calculated under the assumption of fission-evaporation competition being independent of energy. It must again be pointed out that the calculated yields are subject to large uncertainties, since the number of cascades followed in the calculations[8] of the knock-on phase was small. The resulting poor statistical accuracy of the cross sections from which these curves were drawn make them reliable only as to their gross features.

In Fig. 3, where the (p,pxn) reaction cross sections

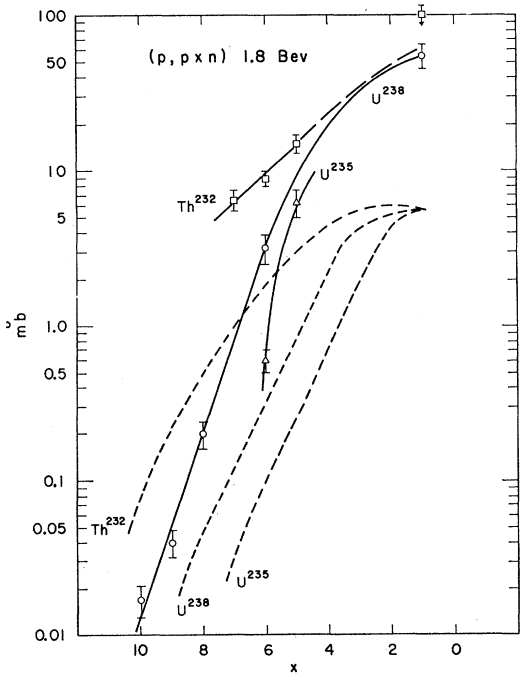

FIG. 3. Cross sections at 1.8 Bev for the reactions (p,pxn) as a function of x for the indicated targets. Broken lines calculated.

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are plotted, it is seen that the large discrepancy between the calculated and the experimental yields is common to all three targets. However, the general trend and ordering of the curves according to target leads one to believe that qualitatively fission competition may have been introduced correctly, and that the main part of the discrepancy is in the calculation for the knock-on phase.

In Fig. 5, the $(p,3p2n)$ product yield seems anomalously high when compared to those of the other $(p,3pxn)$ products and one might be tempted to ascribe this to high yield from a $(p,p\alpha)$ reaction, with a single-collision mechanism analogous to the (p,pn) and $(p,2p)$ reactions. However, when the yield of this product is compared with the yields of products with atomic number one higher in Fig. 2, the anomaly is not as pronounced. It does not seem that there is clear evidence at present of a $(p,p\alpha)$ reaction with anomalously high cross section. (The large error on this cross section is due to the large correction for the natural amount of Th^{234} in the U^{238} target. The cross section at 1.8 Bev is large compared with the values at the other bombarding energies.)

Figure 6 is the only one in which the calculated yields are larger than the experimental ones. This is even more surprising when it is considered that Lindner and Turkevich[9] observed just the opposite. That is, they obtained good agreement between calculated and experimental yields for (p,pxn) and $(p,2pxn)$ reactions but found that the calculated $(p,3pxn)$ cross sections fell below the experimental ones and the $(p,4pxn)$ cross sections fell still further below. They pointed out that the discrepancy became worse at the lower bombarding energies and attributed it to the emission of charged aggregates in the knock-on phase of the interaction, in reactions like the $(p,p\alpha)$ mentioned above. The procedure described in the last section of adding one to the calculated number of knock-on cascades leading to a certain nuclide would have its greatest effect for those isotopes furthest from the target. The lowest of the three calculated curves in Fig. 6 was obtained without using this procedure and it is seen that the discrepancy with experiment is reduced, but not so much as to bring the calculated yields below the experimental ones. The corresponding effect in Figs. 3–5 would have been less severe but generally in the direction of making the observed discrepancies bigger, rather than smaller. The present calculation differs from that of Lindner and Turkevich,[9] since it took into account charged particle evaporation. Lindner and Turkevich estimate that the effect of proton evaporation on their $(p,3pxn)$ yields was small. In the present calculation, it is estimated that the introduction in the analysis of the evaporation of singly and doubly charged particles has raised by about a factor of two the $(p,4pxn)$ yields shown in Fig. 6. It is concluded that the difficulty mentioned by Lindner and Turkevich, of being unable to account for the yields of isotopes far from the target, is not apparent in this work.

With the help of Fig. 1, cross sections for unmeasured nuclides were estimated and summed to obtain the mass-yield curve shown in Fig. 7 as a histogram. While

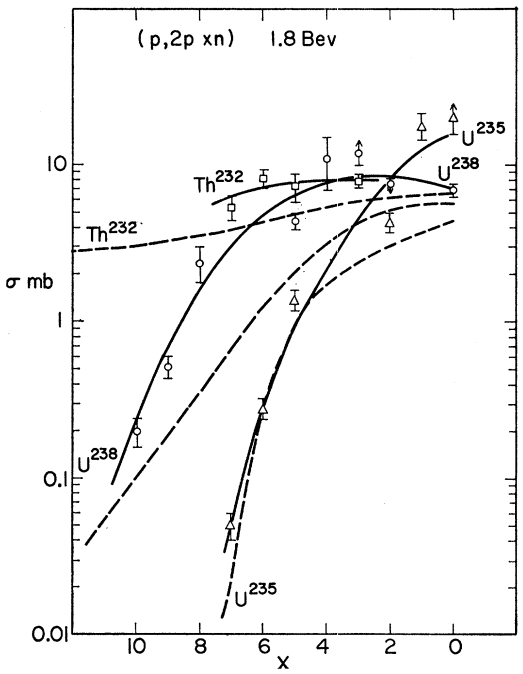

FIG. 4. Cross sections at 1.8 Bev for the reactions $(p,2pxn)$ as a function of x for the indicated targets. Broken lines calculated.

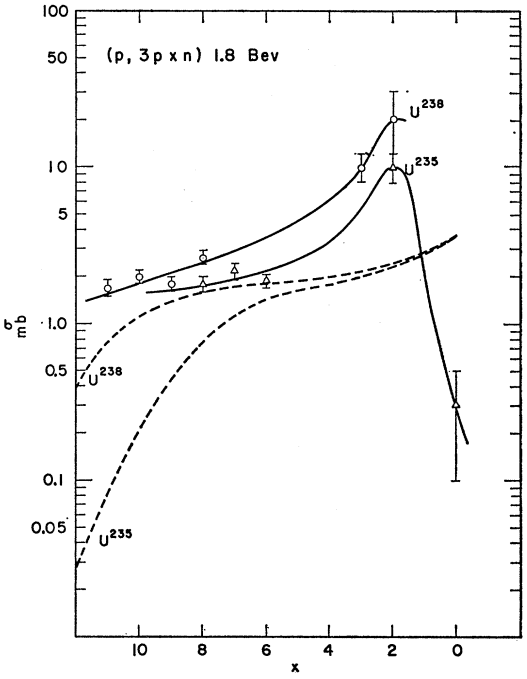

FIG. 5. Cross sections at 1.8 Bev for the reactions $(p,3pxn)$ as a function of x for the indicated targets. Broken lines calculated.

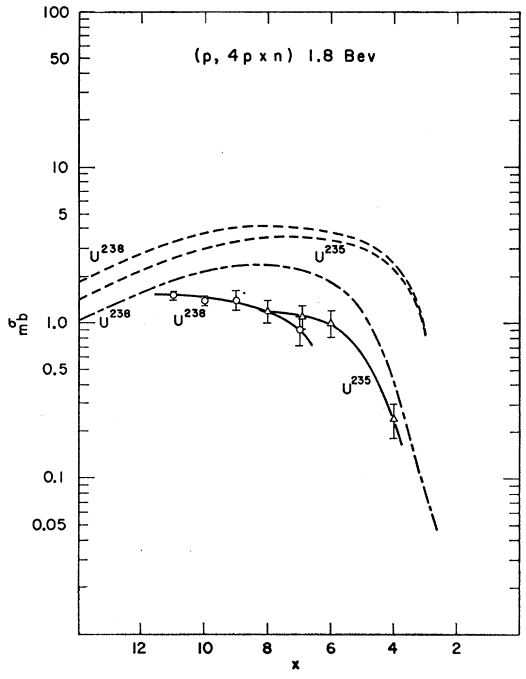

FIG. 6. Cross sections at 1.8 Bev for the reactions $(p,4pxn)$ as a function of x for the indicated targets. Broken lines calculated. See text for explanation of lower broken line.

there is considerable uncertainty in interpolating unmeasured yields it is thought that the existence of two maxima in the curve is real. The lower heavy curve in the figure was calculated assuming fission-evaporation competition independent of excitation energy, and the upper heavy curve assuming no fission competition at all. It is seen that both curves, even the one assuming no fission in the calculation, fall significantly below the experimental data. This can probably be interpreted as a failure of the calculations of the knock-on phase[8] to predict sufficient probability for simple processes leading to low deposition energy. This is in contrast to the good agreement (except for p,pn and $p,2p$ reactions) obtained between this type of calculation and experiments with 0.38-Bev protons on bismuth,[24] 0.36-Bev protons on copper,[1] the experiments mentioned above with 0.34-Bev protons on uranium,[9] and even those with 1.8-Bev protons on copper.[1,25] However, recently, Winsberg[26] has found that this type of calculation predicts low yields for (p,pxn) and $(p,2pxn)$ reactions with 2-Bev protons on iodine and indium. Thus it appears that the apparent failure in the calculation of the knock-on phase is greatest at the highest bombarding energy for the targets of highest atomic number. It has been a common assumption[8,9,22,23] that introduction of a diffuse nuclear surface[27] into the knock-on calculations, in place of the sharp discontinuity used, would lead to better average agreement with measured yields of (p,pn) and $(p,2p)$ reactions. However, it is still necessary to explain why the discrepancy for uranium between calculation and experiment for reactions with ΔA greater than one exists at 1.8-Bev bombarding energy but not at 0.34.

Another way of looking at this discrepancy is the following. Knock-on calculations[8] predict that the probability of cascades of a given small ΔA, irrespective of deposition energy, goes through a maximum and decreases rapidly as the bombarding energy increases. Also the calculations predict that the spectra of deposition energies of particular cascade products are not very sensitive to bombarding energy. Thus, the calculations indicate much lower yields of simple spallation products at the higher bombarding energies, while experimentally the excitation functions do not decrease with increasing bombarding energy. Any modification of the cascade calculations that would raise the probability of small cascades at the higher bombarding energies, while not necessarily changing the present spectra of deposition energies of particular cascade products, would improve agreement with experiment. This type of modification also would, in effect, reduce the average deposition energy. Indeed the calculations of the knock-on phase[8] for uranium predict an increase of a factor of four in both the average deposition energy and number of cascade nucleons in going from 0.34- to 1.8-Bev bombarding energy. This is probably due to the increasing importance of mesons for the proliferation of the cascade. The corresponding ratio for a copper target is less, indicating less sensitivity to this effect. It was pointed out in the paper on the knock-on[8] calculations that the details of meson production and inter-

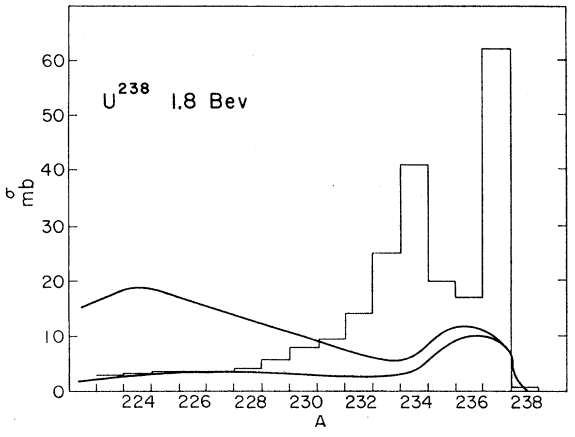

FIG. 7. Mass yield curves for the bombardment of U^{238} with 1.8-Bev protons. Histogram—from experimental data; upper curve—calculated with no fission competition; lower curve—calculated with fission-evaporation competition independent of excitation energy.

[24] E. T. Hunter and J. M. Miller, Phys. Rev. **115**, 1053 (1959).
[25] D. W. Barr [University of California Radiation Laboratory Report UCRL-3793, 1957 (unpublished)], using a simpler interpolation method, did not obtain as good agreement.
[26] I. Ladenbauer and L. Winsberg, Phys. Rev. **119**, 1368 (1960); B. R. Nethaway and L. Winsberg, *ibid.* **119**, 1375 (1960).
[27] R. Hofstadter, Ann. Rev. Nuclear Sci. **7**, 231 (1957).

actions were treated somewhat crudely, and it is suggested here that the importance of mesons in the proliferation of the cascade may have been overestimated, causing the discrepancy for uranium at 1.8 Bev but not at 0.34 Bev.

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APPENDIX I

Chemical Procedures

The anion and cation exchange resins used in the procedures below were Dowex-1 and Dowex-50, respectively. All TTA extractions were carried out with 1 to 2 ml volumes, in a 15-ml centrifuge cone, with emulsification by means of a transfer pipette. Usually the organic phase was evaporated directly on the sample plate, with a device for keeping the edge of the plate hotter than the center. The organic residue was then removed by igniting the plate at red heat. All samples were mounted on 0.010-in. platinum. Most of the following procedures were adapted from standard reference works.[28,29]

Uranium from Uranium

The uranium foil target was dissolved in concentrated HCl with the addition of hydrogen peroxide and adsorbed on a large anion column. After the column was washed with concentrated HCl, the protactinium activities were eluted with $3N$ HCl, and then the uranium was eluted with $1N$ HCl. The $1N$ HCl eluate was saturated with ammonium nitrate, and the uranium extracted into ether. The uranium was back-extracted into water and electrodeposited on platinum from $6M$ NH_4Cl solution at pH 5.

For faster separation in studies of short-lived activities, the ether extract was evaporated on a platinum sample plate and ignited.

Protactinium from Uranium

The protactinium eluate from above was made $6N$ in HCl and was extracted into a $0.25M$ TTA benzene solution. The organic phase was evaporated on the sample plate. For faster separations, the target solution in concentrated HCl was diluted to $6N$ and extracted directly.

For beta activity measurements in which zirconium fission products constituted an interference, the protactinium was separated from the column eluate via coprecipitation with MnO_2 or extraction into diisopropyl ketone prior to TTA extraction and mounting.

Protactinium from Thorium

The thorium metal target was dissolved in concentrated HCl with addition of $(NH_4)_2SiF_6$, and the acid diluted to $6N$. Extraction into a benzene solution of TTA and mounting of the organic phase was then accomplished as above.

Thorium from Uranium

The uranium target was dissolved and adsorbed on an anion column as above. The HCl washings were evaporated, and the thorium was adsorbed on a very small ($1\frac{1}{2}$ mm by 3 cm) cation column. The column was washed with $6N$ HCl and $6N$ HNO_3 and the thorium desorbed very slowly with $0.5M$ oxalic acid. The eluate was evaporated, the oxalic acid sublimated, and the residue taken up in $0.1M$ HNO_3. The thorium was then extracted into $0.2M$ TTA in benzene and the organic phase evaporated directly on the sample plate.

For a faster procedure, the solution in which the target was dissolved was passed directly through the small cation column. The thorium was eluted with oxalic acid directly on to a platinum plate on a hot plate and ignited. Since a small amount of uranium accompanied the thorium in this procedure the column was washed before elution with a U^{238} solution when a U^{235} target was used.

Thorium from Thorium

The target was dissolved in concentrated HNO_3 with the aid of $(NH_4)_2SiF_6$, the acid diluted to $8N$, and the thorium adsorbed on a large anion column. The column was washed with $8N$ HNO_3, and the thorium was eluted with $6N$ HCl. The solution was evaporated, neutralized, and extracted with a $0.2M$ benzene solution of Dagmar.[28] The organic phase was scrubbed twice with $0.1M$ acid and the thorium back extracted with $6N$ HCl. The solution was evaporated to dryness and the thorium vacuum vaporized onto a platinum plate (with low yield).

[28] E. K. Hyde, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 281.

[29] A. M. Poskanzer and B. M. Foreman, Jr., J. Inorg. & Nuclear Chem. **16**, 323 (1961).

For a faster procedure, the solution in which the target was dissolved was evaporated, neutralized, and extracted into a benzene solution of Dagmar. The organic phase was scrubbed with 0.1N acid. The benzene was evaporated off in a shallow boat, leaving an infinitely thick sample of thorium and Dagmar. From range-energy data for alpha particles, it was calculated that the alpha-counting efficiency of the 8-min Th^{225} was 1.10 times that of the 30-min Th^{226}.

Actinium from Uranium

The target was dissolved in HCl with the addition of H_2O_2, the solution evaporated, and passed through a cation column. The column was washed with 2N HNO_3 and the actinium eluted with 4N HNO_3. The elute was neutralized, buffered with ammonium acetate-acetic acid, and actinium extracted into 0.2M TTA in benzene. The organic phase was washed with distilled water, and the actinium back-extracted with $10^{-3}M$ acid. The aqueous phase was again buffered and the actinium reextracted. This time the organic phase was evaporated to prepare the sample plate. This procedure did not purify the sample from rare earths; no beta counting was done on the sample.

Actinium from Thorium

The 8N HNO_3 effulent from the anion column of the thorium-from-thorium procedure was evaporated. The rest of the procedure is the same as the actinium-from-uranium procedure, beginning at the cation-column step.

APPENDIX II

Merlin Calculations

From the published Monte Carlo calculations of Dostrovsky *et al.*[20] the average de-excitation per evaporated nucleon in the heavy-element region was taken as

$$D/\Delta A = 8+3.2(D/A)^{\frac{1}{2}},$$

where D is the deposition energy in Mev of the knock-on cascade product.

From the unpublished[19] calculations, of the same authors, for eleven different A, Z, deposition-energy combinations, it was concluded that, in the region of interest, the relative probability of emitting singly- and doubly-charged particles, Γ_{1+} and Γ_{2+}, to the total particle evaporation probability, Γ_s, could be represented by

$$\Gamma_{1+}/\Gamma_s = [5.5+0.65(Z-Z_A)]\times 10^{-4}E,$$
$$\Gamma_{2+}/\Gamma_s = [1+0.2(Z-Z_A)](1.5-0.001E)\times 10^{-4}E,$$

where E is the excitation energy in Mev when the particle is evaporated. The charge-value for beta stability, Z_A, was taken as $0.354A+9.1$. In order to take account qualitatively of the emission of deuterons and tritons in addition to protons, the de-excitation per singly-charged particle emitted was taken as 1.5 times the de-excitation per nucleon. The relative probabilities of fission and particle evaporation were obtained by fitting parallel lines of slope 0.130 to the data of Fig. 2 of the paper of Vandenbosch and Huizenga,[3] where $\log_{10}(\Gamma_n/\Gamma_f)$ is plotted vs mass number.

The calculation was carried out for each knock-on cascade product by first calculating the fractions undergoing fission and emitting each of the three types of particles. In turn, for each of the products of particle emission, the fractions of these going into each of the four branches was again calculated. The branching of the chains was followed until the residues were de-excited, and the remaining fractions were stored and summed according to Z and A. The process was repeated for each knock-on cascade product. Certain limits were built into the calculations, so that if any of the fractions along the chain fell below a certain value, usually set at 0.01, then this fraction was added into that of one of the more probable products at this branch point.

Because of the many average quantities used in the analysis, it was not expected that the excitation energy at the act of fission would be calculated very accurately. However, the number of chances at fission during the de-excitation process and, therefore, the A, Z distribution of final products, should not be strongly affected by the averaging processes used.