Helium-Ion-Induced Fission Cross Sections of U^{233} and U^{238} and the Nuclear Radii of Heavy Elements*

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New radiochemical data of the total fission cross sections have been obtained for U^{233} and U^{238}, which can be used to derive nuclear radii for these isotopes. This and certain accurate previous results on U^{235} and natural bismuth can be interpreted in terms of an r_0 value of 1.41×10^{-13} cm, when $R_\alpha=2.19\times10^{-13}$ cm, using the Weisskopf square-well nuclear model. Further, the nuclear radii obtained by this analysis are also in good agreement with the radii obtained by others from alpha-particle scattering data with a similar model. A diffuse-potential nuclear model proposed by Igo shows agreement with experiment in the lower energy range. The agreement, however, is not observed with data for higher energies. Certain features of the U^{233} fission curves are also discussed.

INTRODUCTION

WITH the recent increased interest in nuclear radii generated by the accurate electron scattering data,[1] it is of interest to obtain comparable interaction data by other means for comparison. This communication is concerned with the experimental determination of accurate total reaction cross-section data for certain selected heavy-element isotopes where, because of the predominance of fission, the possibility of missing any reaction with a large cross section is minimized. Two nuclear models can be tested at the present time, that featuring the Weisskopf square-well potential[2,3] and the diffuse potential proposed by Igo.[4] Values of nuclear radii so obtained are in good agreement with alpha-particle scattering data.[4–6]

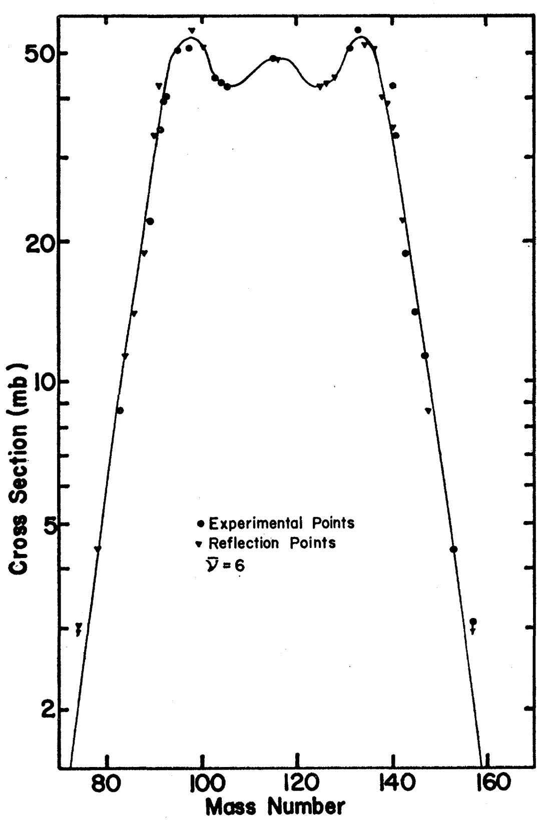

FIG. 1. Fission mass yield curve for 40.5-Mev helium ions on U^{233}.

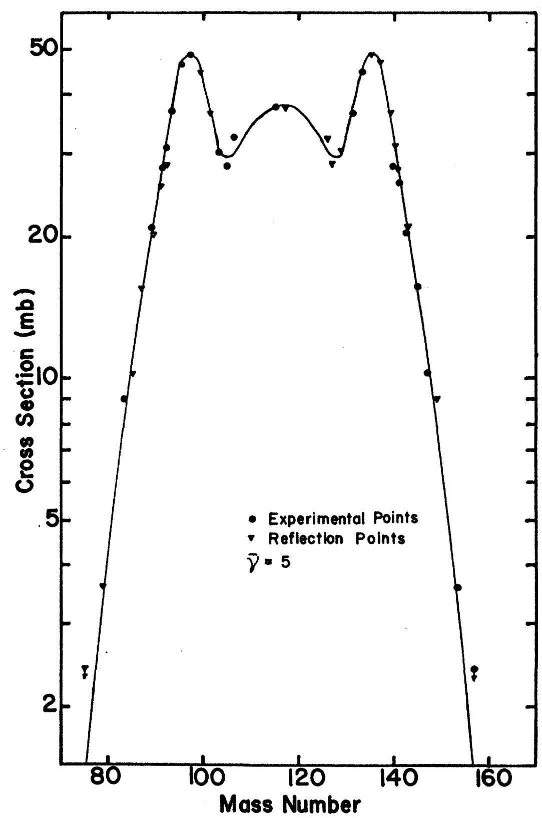

FIG. 2. Fission mass yield curve for 34.5-Mev helium ions on U^{233}.

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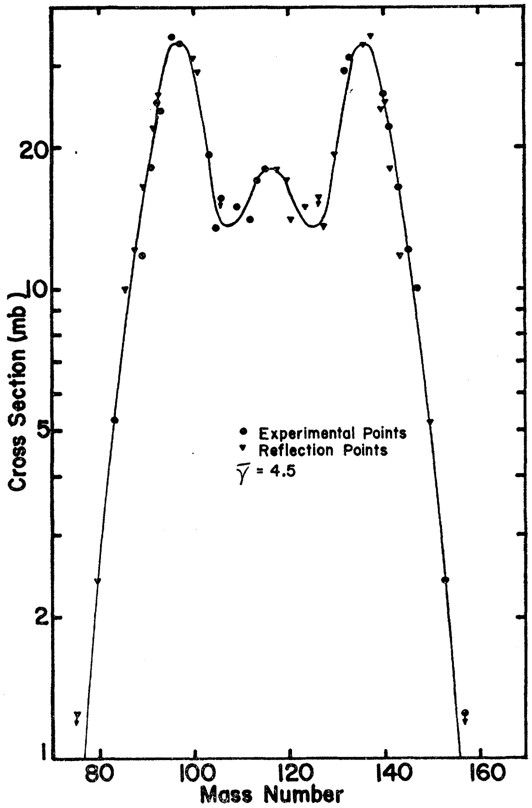

FIG. 3. Fission mass yield curve for 29.0-Mev helium ions on U^{233}.

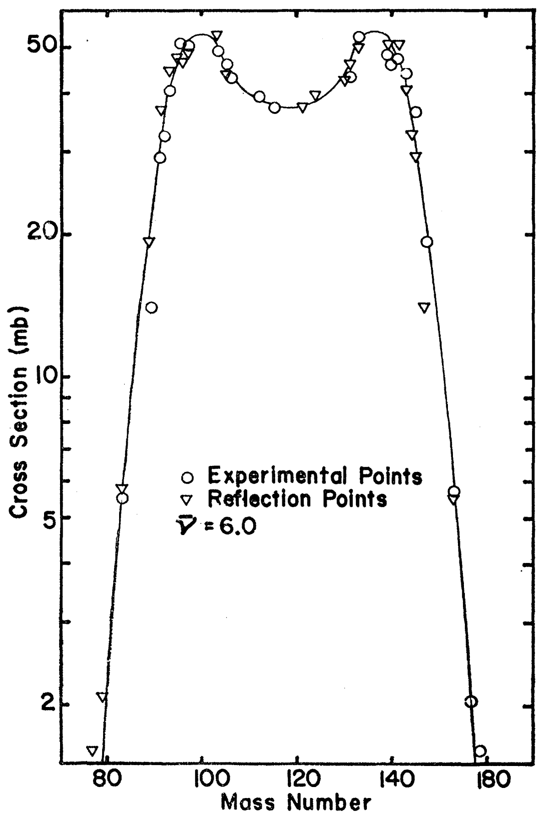

FIG. 5. Fission mass yield curve for 39.9-Mev helium ions on U^{238}.

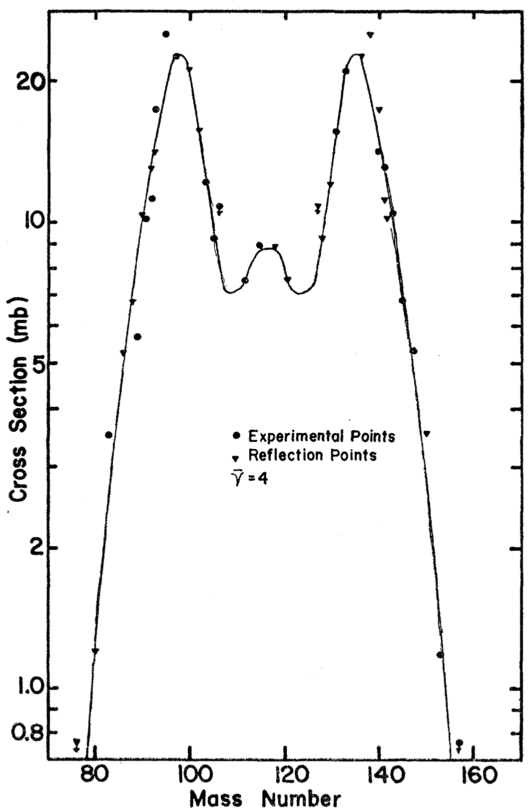

FIG. 4. Fission mass yield curve for 25.3-Mev helium ions on U^{233}.

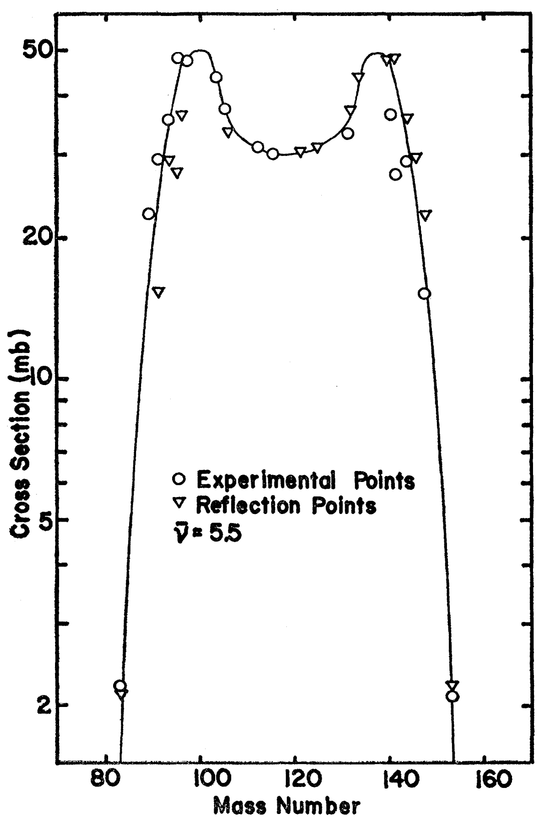

FIG. 6. Fission mass yield curve for 36.8-Mev helium ions on U^{238}.

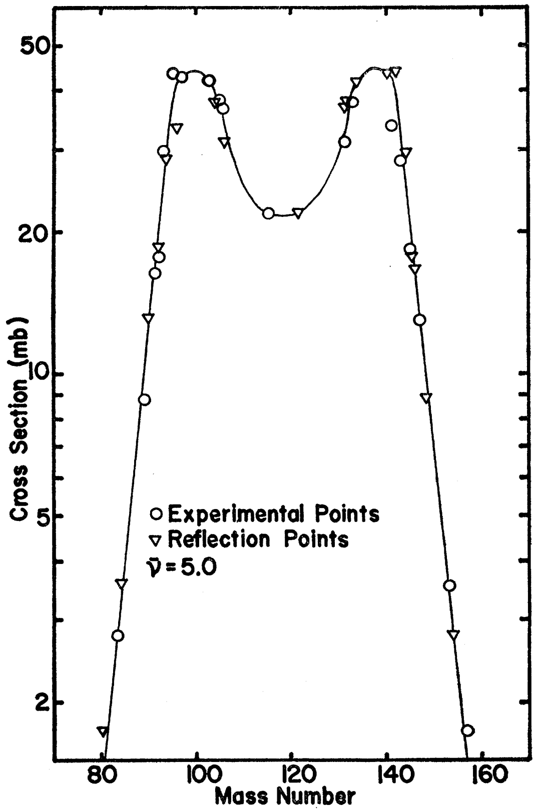

FIG. 7. Fission mass yield curve for 33.8-Mev helium ions on U^{238}.

It has been recognized that the predominant nuclear reaction in isotopes of elements with $Z=92$ or higher, bombarded with charged particles up to about 40 Mev of excitation energy, is fission. Spallation-fission competition has received considerable attention by Seaborg and his co-workers,[7–9] as well as others.[10–14] Since, in many cases, the spallation reactions contribute only from 5 to 10% of the total cross section, studies of the heavy elements provide a means for obtaining accurate total reaction cross sections.

A number of measurements of total reaction cross sections have been reported for heavy elements.[7–14] Unfortunately, these data were not obtained with sufficient accuracy to test anything but order-of-magnitude agreement with various nuclear models. The first exceptions were the data of Gunnink and Cobble[15] on the fission of U^{235} with 20–40-Mev helium ions. The agreement of the total cross sections for this reaction with those calculated from the Weisskopf square-well nuclear potential was good, indicating a r_0 somewhat greater than 1.5×10^{-13} cm. However, recently Igo[4,6] has proposed a diffuse nuclear potential to explain alpha-particle scattering data for targets over a wide range of nuclear charge, and which also appears to be consistent with the previous, less accurate, total reaction cross-section data.

TABLE I. Fission cross sections (mb) for helium ions on U^{233}. Each left-hand column lists the observed yield of each isotope. Each right-hand column lists the corrected cross section for the mass chain.[a]

Energy (Mev) / Isotope	40.5		34.5	
	σ	σ corr.	σ	σ corr.
Br^{83}	8.6±0.1 (2)	8.7	9.5±0.5 (2)	9.5
Sr^{89}	22.0±1.0 (2)	22.2	21.5±0.5 (2)	21.5
Sr^{91}	33.1±0.1 (3)	34.4	27.0±2.3 (3)	27.8
Sr^{92}	34.8	39.2	28.5±0.5 (2)	31.0
Y^{93}	38.8±0.4 (2)	40.0	35.9±1.0 (2)	36.6
Zr^{95}	49.6	50.6	46.0±0.5 (2)	46.4
Zr^{97}	46.0±2.5 (3)	51.7	45.9±1.0 (2)	48.5
Ru^{103}	44.0±2.8 (2)	44.0	30.0±1.0 (2)	30.0
Ru^{105}	35.0±1.2 (2)	43.1	22.3±0.3 (2)	27.1
Ru^{106}	40.0±3.2 (2)	42.5	31.0±0.9 (2)	32.3
Cd^{115}	42.1	48.5	33.2	37.5
Cd^{115m}	5.3		4.0	
I^{131}	32.9	51	26.2	36.4
I^{133}	13.5	56	14.7±0.2 (2)	44.5
Ba^{140}	12.0	42.8	11.0	28.2
Ce^{141}	16.3	33.4	19.3±0.7 (2)	25.6
Ce^{143}	12.2	18.8	15.5	20.2
Pr^{145}	10.3	13.9	13.0±0.2 (2)	15.7
Nd^{147}	9.4±0.4 (2)	11.4	9.0	10.3
Sm^{153}	2.8±0.1 (2)	4.4	2.7±0.3 (2)	3.6
Eu^{157}	1.0	3.1	1.1±0.05 (2)	2.4
Gd^{159}	0.55±0.05 (2)	1.3	0.55±0.05 (2)	1.0
Energy (Mev) / Isotope	29.0		25.3	
	σ	σ corr.	σ	σ corr.
Br^{83}	5.1±0.1 (2)	5.2	3.4±0.1 (2)	3.5
Sr^{89}	11.7±0.4 (2)	11.7	5.7±0.5 (2)	5.7
Sr^{91}	17.5±0.5 (3)	18.1	10.0±0.9 (3)	10.2
Sr^{92}	23.5±0.9 (2)	25.3	11.4±0.4 (2)	12.1
Y^{93}	23.3±0.3 (2)	23.8	17.0±0.4 (2)	17.3
Zr^{95}	34.3±0.5 (3)	34.6	25.0±1.3 (2)	25
Zr^{97}	31.0±1.9 (3)	33.2	21.0±1.9 (3)	22.4
Ru^{103}	19.2±0.3 (3)	19.2	12.1±0.1 (2)	12.1
Ru^{105}	11.0±0.9 (3)	13.4	7.6±0.1 (2)	9.3
Ru^{106}	15.0±0.5 (3)	15.6	10.4±0.6 (3)	10.8
Pd^{109}	15.0	15		
Pd^{112}	13.0±0.3 (2)	14.1	7.0	7.5
Ag^{113}	16.9	17.2		
Cd^{115}	16.0±0.9 (2)	18	8.0±0.5 (2)	8.9
Cd^{115m}	(1.6)		0.8	
I^{131}	19.2	29.3	11.7±0.5 (2)	15.6
I^{133}	12.0±1.3 (3)	31.0	9.0±0.5 (3)	21
Ba^{140}	11.4±0.4 (2)	26.0	7.3±0.5 (2)	14
Ce^{141}	15.5±1.1 (2)	22.0	12.2±1.0 (2)	13.1
Ce^{143}	13.4±0.6 (2)	16.5	8.7±0.1 (2)	10.4
Pr^{145}	10.4±0.1 (2)	12.1	6.0±0.1 (2)	6.8
Nd^{147}	8.9±0.2 (2)	10.0	4.9±0.3 (2)	5.3
Sm^{153}	1.9±0.05 (2)	2.4	1.0±0.10 (2)	1.2
Eu^{157}	0.67	1.26	0.45±0.03 (2)	0.76
Gd^{159}	0.27±0.05 (2)	0.44	0.20±0.02 (2)	0.30

[a] Where more than one bombardment was made at a given energy, the number of determinations is indicated in parentheses.

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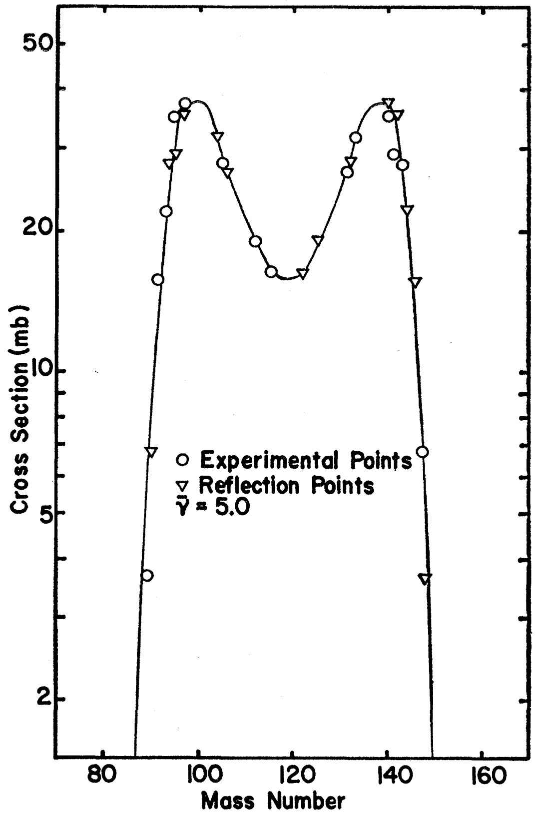

FIG. 8. Fission mass yield curve for 31.0-Mev helium ions on U^{238}.

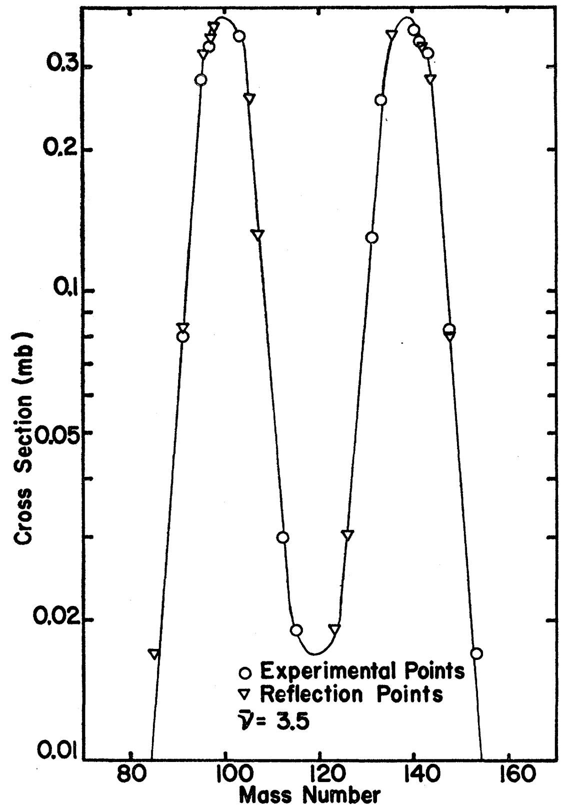

FIG. 10. Fission mass yield curve for 19.8-Mev helium ions on U^{238}.

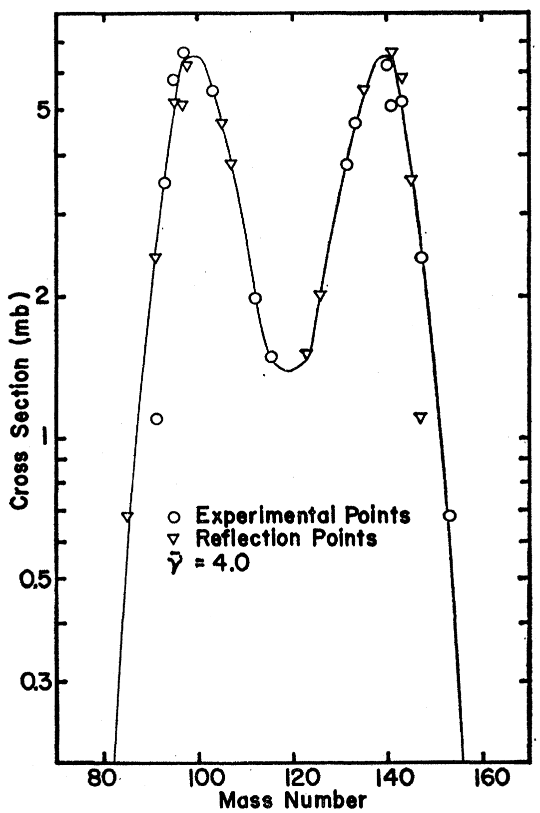

FIG. 9. Fission mass yield curve for 24.1-Mev helium ions on U^{238}.

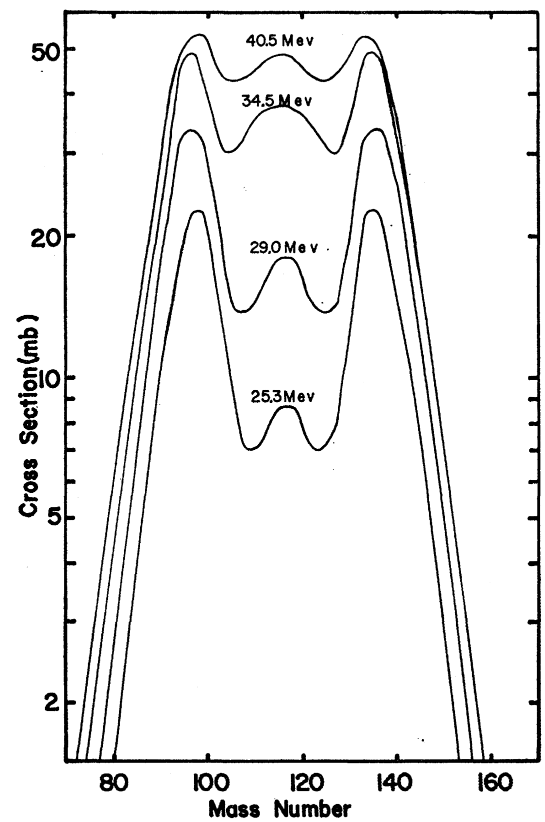

FIG. 11. Composite fission mass yield curves for medium-energy helium ions on U^{233}.

TABLE II. Fission cross sections (mb) for helium ions on U^{238}. Each left-hand column lists the observed yield for each isotope. Each right-hand column lists the corrected cross section for the mass chain.[a]

Isotope \ Energy (Mev)	39.9 σ	39.9 σ corr.	36.8 σ	36.8 σ corr.
Zn^{72}	0.54	0.54		
Br^{83}	5.3±0.5 (3)	5.52	2.2	2.27
Sr^{89}	14 ±1.0 (2)	14.0	22.5	22.5
Sr^{91}	29 ±2.0 (3)	29.3	29.4±0.9 (2)	29.5
Sr^{92}	32.1±1.0 (2)	32.7		
Y^{93}	40.4±1.3 (2)	40.8	35.6±1.4 (2)	35.9
Zr^{95}	50.5±1.8 (4)	51.0	48.6±2.6 (2)	48.6
Zr^{97}	49.6±2.4 (5)	50.6	47.2±0.4 (2)	47.6
Ru^{103}	49.7±3.0 (3)	49.7	43.8	43.8
Ru^{105}	37.6±1.3 (2)	46.0	37.5	37.5
Ru^{106}	42.3±0.3 (2)	43.1		
Pd^{112}	39.6	39.6	30.4±2.1 (2)	31.0
$Cd^{115,115m}$	37.3±0.8 (2)	37.3	30.4	30.4
I^{131}	40.9±1.1 (3)	43.5	31.6±1.6 (2)	33.3
I^{133}	39.4	53.2		
Ba^{139}	43.9	48.2		
Ba^{140}	37.5±2.5 (3)	46.3	31.1±0.6 (2)	36.6
Ce^{141}	47.2±0.3 (2)	47.2	27.2±1.7 (2)	27.5
Ce^{143}	43.0±0.5 (2)	44.8	28.2±0.4 (2)	29.0
Pr^{145}	35.7±0.6 (2)	36.8		
Nd^{147}	19.0±0.9 (2)	19.4	15.1±0.4 (2)	15.3
Sm^{153}	5.5±0.8 (4)	5.73	2.1±0.2 (2)	2.14
Eu^{157}	1.8±0.2 (2)	2.09		
Gd^{159}	1.5±0.1 (2)	1.63	1.0	1.06

Isotope \ Energy (Mev)	33.8 σ	33.8 σ corr.	31.0 σ	31.0 σ corr.
Br^{83}	2.7±0.3 (2)	2.78		
Sr^{89}	8.8±0.9 (2)	8.8	3.66	3.66
Sr^{91}	16.5±0.7 (2)	16.5	15.6	15.6
Sr^{92}	17.7±1.1 (2)	17.8		
Y^{93}	29.7±1.6 (2)	29.8	22.0±0.2 (2)	22.0
Zr^{95}	43.5±2.8 (2)	43.5	35.1±1.5 (3)	35.1
Zr^{97}	42.8±1.9 (2)	43.2	37.2±1.3 (3)	37.5
Ru^{103}	42.0±0.7 (2)	42.0		
Ru^{105}	31.8±1.3 (2)	38.4	27.7	27.7
Ru^{106}	36.4±0.4 (2)	36.6		
Pd^{112}			19.0±0.5 (2)	19.2
$Cd^{115,115m}$	22	22	16.3	16.3
I^{131}	29.1±0.6 (2)	30.3	25.4	26.4
I^{133}	31.4±0.5 (2)	37.8	26.3±1.7 (3)	31.7
Ba^{140}			30.8±0.9 (2)	35.0
Ce^{141}	33.4	33.4	29.2	29.2
Ce^{143}	27.8	28.4	27.3±0.1 (2)	27.9
Pr^{145}	17.9±1.9 (2)	18.3		
Nd^{147}	13.0±1.0 (2)	13.1	6.74±0.1 (2)	6.80
Sm^{153}	3.5±0.5 (2)	3.57	0.40	0.41
Eu^{157}	1.6±0.5 (2)	1.75		
Gd^{159}	1.0±0.3 (2)	1.04		

Isotope \ Energy (Mev)	24.1 σ	24.1 σ corr.	19.8 σ	19.8 σ corrected for neutron background	19.8 σ corr.
Sr^{91}	1.1±0.1 (2)	1.1	0.24	0.08	0.08
Y^{93}	3.5	3.5			
Zr^{95}	5.8±0.1 (2)	5.8	0.53	0.28	0.28
Zr^{97}	6.6	6.6	0.63	0.33	0.33
Ru^{103}	5.48	5.48	0.40	0.35	0.35
Ru^{105}	4.74	4.74			
Pd^{112}	2.0±0.1 (2)	2.0	0.057	0.03	0.03
$Cd^{115,115m}$	1.48	1.48	0.099	0.019	0.019
I^{131}	3.71±0.4	3.79	0.276	0.13	0.132
I^{133}	4.1±0.1	4.71	0.388	0.23	0.256
Ba^{140}	5.7±0.1	6.2	0.641	0.341	0.359
Ce^{141}	5.1	5.1	0.67	0.34	0.34
Ce^{143}	5.2	5.2	0.50	0.32	0.32
Nd^{147}	2.43±0.15 (2)	2.43	0.166	0.082	0.082
Sm^{153}	0.68	0.68	0.028	0.017	0.017

[a] Where more than one bombardment was made at a given energy, the number of determinations is indicated in parentheses.

In order to test the two models further, the helium-ion-induced fission of two more isotopes of uranium has been studied. The new data do not confirm the older fission-spallation studies, and are not in complete agreement with the Igo-model cross sections.

EXPERIMENTAL PROCEDURE

The experimental procedure has been patterned after the methods previously discussed.[15] Briefly these consist of electroplating the hydrated oxides of the various isotopes of uranium onto small aluminum disks; the deposits are tested for uniformity and assayed by alpha counting. The disks are covered with a thin aluminum foil which serves as a collector for any fission products which recoil out of the target. The target is assembled just behind any further aluminum foils that are required to degrade the cyclotron helium ion beam to the desired energy. After bombardment, the target and cover foil are completely dissolved in the presence of added carriers of the fission products to be assayed.

Natural uranium was used for the U^{238} bombardments and considered to be 100% U^{238}. The U^{233} was supplied by the University of California Radiation Laboratory. An alpha-spectrum analysis showed only 2.5% alpha active impurity in the sample. This is, therefore, an

TABLE III. Total fission cross-section data for helium ion bombardments on U^{233}, U^{235},[a] and U^{238}.

Isotope	Energy Mev	Total fission σ_f (mb)	Total spallation $\sigma_{\Sigma\,\text{spall}}$ (mb)	Total cross section σ_T (mb)
U^{233}	40.5	1345	22	1367
U^{233}	34.5	1090	17	1107
U^{233}	29.0	606	10	616
U^{233}	25.3	350	4	354
U^{235}	39.9	1386	20	1400
U^{235}	33.8	1030	20	1050
U^{235}	28.2	580	20	600
U^{235}	25.9	290	16	306
U^{235}	23.1	87	8	95
U^{235}	20.5	10	2	12
U^{238}	39.9	1317	97	1414
U^{238}	36.8	1127	96	1223
U^{238}	33.8	970	95	1065
U^{238}	31.0	800	95	895
U^{238}	24.2	168	55.4	223
U^{238}	19.8	5.4	6.6	12

[a] See reference 15.

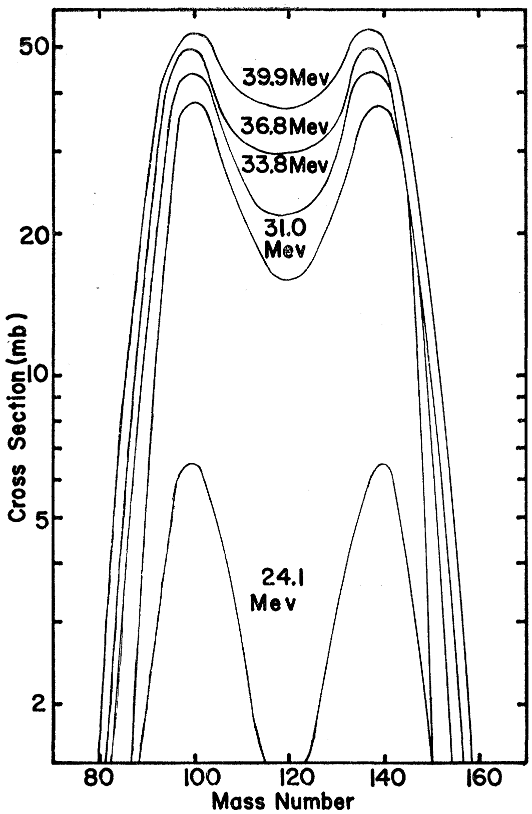

FIG. 12. Composite fission mass yield curves for medium-energy helium ions on U^{238}.

indication of the isotopic purity of the material. It was considered to be 100% U^{233}.

The counting procedures and standardizations have been discussed in detail elsewhere.[16–18] The bombardments were carried out on the Argonne cyclotron and the Crocker cyclotron at the University of California. The range-energy relations determined for protons[19] were used to calculate the degraded beam energies. The initial beam energy determined at the respective cyclotrons was consistently measured to ± 0.5 Mev. The use of two different cyclotrons indicated that certain systematic errors, e.g., beam current, beam energy, total current integration, etc., were within the error limits quoted. The most significant improvements over the procedures developed in similar research lies in the great amount of attention paid to absolute beta and gamma counting,[16,17] and in the use of chemical procedures which insure complete exchange of the fission product activities and their inactive carriers.[20]

Further, it can be shown that the chemical compounds used in many previous fission product determinations for chemical yield determinations are now known to be unreproducible in composition and unstable when dried by heating. The reported decay scheme for one key isotope, Ru^{105}, is known to have some serious error. Preliminary information obtained in our laboratories and others has indicated that the 0.726-Mev gamma ray represents only 79% of the total disintegrations.[21,22]

EXPERIMENTAL RESULTS

The fission product formation cross sections from U^{233} and U^{238} are summarized in Tables I and II, respectively. In those cases where multiplicate bombardments were made at the same energy, the number of determinations is indicated parenthetically. The effect of the neutron background present around the accelerators was determined by separate experiments and subtracted out of the data for the lower energy U^{238} bombardments. The cross sections are corrected for the value for that part of the mass chain which was not directly determined using the correlations based on

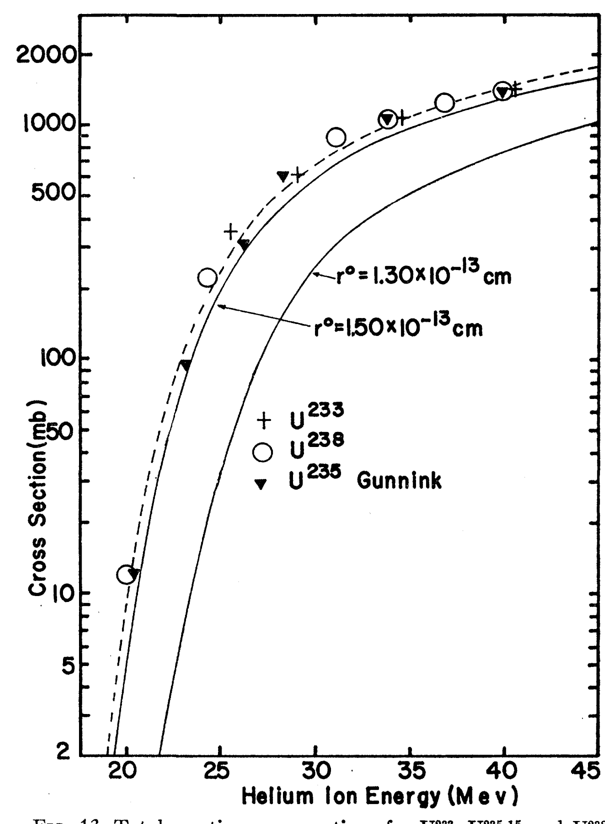

FIG. 13. Total reaction cross sections for U^{233}, U^{235},[16] and U^{238} at various helium ion energies and a comparison with compound nucleus theory assuming the square-well potential (Blatt and Weisskopf[2]).

[16] L. J. Colby, Jr., and J. W. Cobble, Anal. Chem. **31**, 798 (1959).

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[18] R. Gunnink and J. W. Cobble, Atomic Energy Commission Report AECU-4340, 1959 (unpublished).

[19] H. Bichsel, R. F. Mozley, and W. A. Aron, Phys. Rev. **105**, 1788 (1957).

[20] For the details of the radiochemical procedures, the reader is referred to the original theses by L. J. Colby and M. L. Shoaf.

[21] H. W. Brandhorst, Jr., and J. W. Cobble (private communication).

[22] B. Saraf, P. Harihar, and R. Jambunathan, Phys. Rev. Letters **4**, 387 (1960).

the constant-charge-ratio (C.C.R.) rule discussed elsewhere.[15,23]

The data are plotted in the customary manner as mass-yield curves in Figs. 1–10, and as composite plots for comparison in Figs. 11 and 12. From the scatter of points about the smooth curves, it is probable that the integrated fission cross sections at many energies are accurate to at least ±10%.

Certain features of the U^{233} fission curves are interesting, particularly the appearance of the "triple-hump," which is characteristic of some of the lighter elements.[24–27] Careful examination of the data upon which this unusual feature rests lead us to believe that the effect is clearly real. Further, if the effect is ignored and a smooth curve is drawn through the valley region, the area under the mass yield curve, i.e., total fission cross section, is significantly increased. The total fission cross sections so obtained then do not agree with those for the other uranium isotopes at comparable excitation energies. In this respect, it is interesting to note that some indication of this same type of phenomenon is also present in thorium irradiated with fast neutrons[25] and even in the previous U^{235} helium-ion-induced fission obtained in these laboratories.[15]

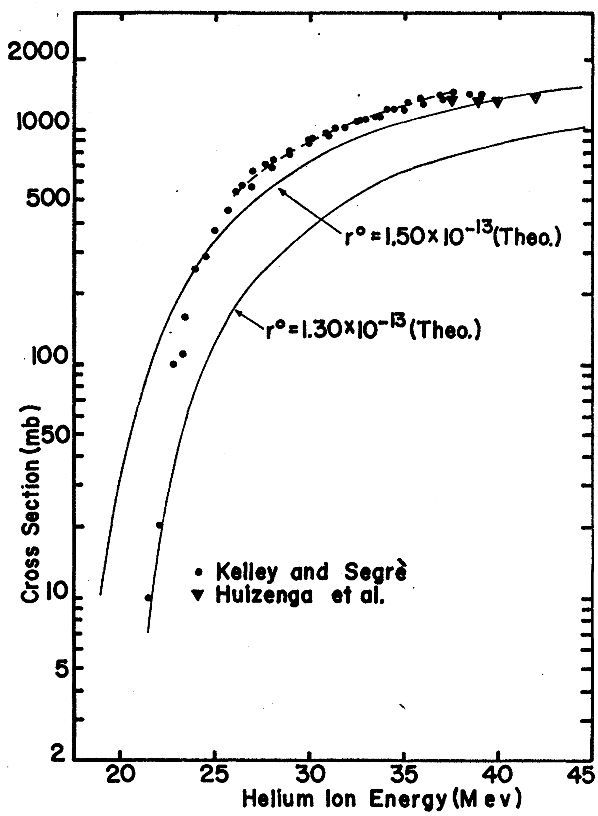

FIG. 14. Total cross-section data for the (α,xn) reaction on bismuth (Kelly and Segrè[13] and Huizenga *et al.*[10]) for various helium ion energies.

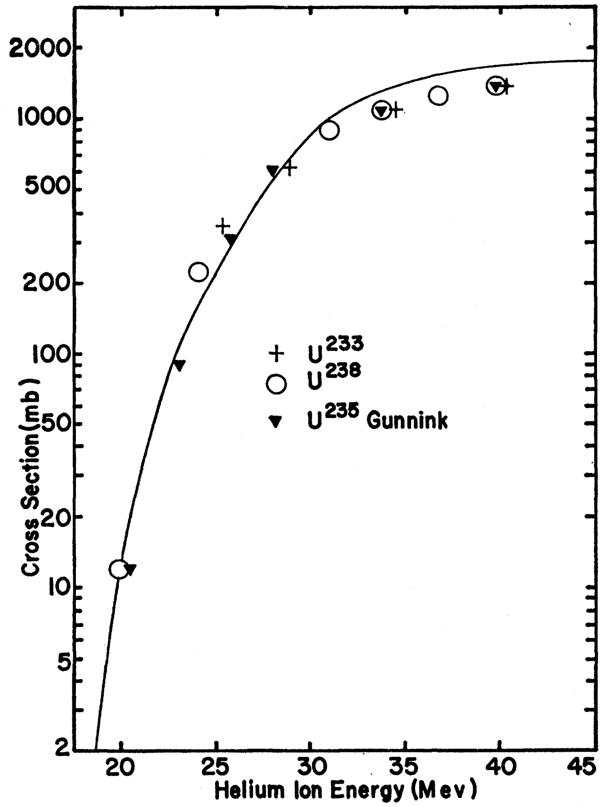

FIG. 15. Total reaction cross sections for U^{233}, U^{235},[15] and U^{238} at various helium ion energies and a comparison with compound nucleus theory assuming the diffuse potential of Igo.[5]

Spallation data were not collected in the present study. Since the contribution to the total cross section from spallation products varies from a few to as large as 50% for 19.8-Mev helium ions on U^{238}, the data of Seaborg *et al.*[8] and Huizenga *et al.*[14] have been added in to obtain the total reaction cross sections as summarized in Table III. Although the errors in some of these data are large, the total error so introduced in the total cross sections is in all cases within the quoted experimental accuracy of the fission data. For convenience, the previous data from this laboratory on U^{235} are included in this summary.

DISCUSSION

There are at present two different sets of excitation functions derived from theory with which to compare the experimental reaction cross sections. The first is based upon the model and calculations by Weisskopf,[2] who assumed a square-well nuclear potential (hereafter referred to as the sharp-cutoff model). In effect, one determines a total interaction distance, $R=r_0A^{\frac{1}{3}}+R_\alpha$, from comparison of the experimental and calculated

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[24] A. W. Fairhall and R. C. Jensen, Phys. Rev. **109**, 942 (1958).
[25] R. A. Nobles and R. B. Leachman, Nuclear Phys. **5**, 211 (1958).
[26] R. D. Griffioen and J. W. Cobble, Phys. Rev. (to be published).
[27] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).

excitation functions. Weisskopf has calculated the latter for r_0 values of 1.3×10^{-13} cm and 1.5×10^{-13} cm, based upon an alpha-particle radius, R_α, of 1.20×10^{-13} cm. Figure 13 shows such a comparison for the three uranium isotopes. The experimental data are in excellent agreement with the calculated curves over a wide range of energies for an interaction distance of $R=10.8\times10^{-13}$ cm or $r_0=1.54\times10^{-13}$ cm using $R_\alpha=1.20\times10^{-13}$ cm.

There is only one other set of experimental data, known to the authors, which is of comparable accuracy and with which one may make comparison, and that is excitation functions of helium ions on bismuth.[10,13] However, the present interpretation of these data is somewhat different; the situation is summarized in Fig. 14. As noted by the authors cited, the (α,n) reaction could not be determined, and the experimental excitation function falls below the theoretical Weisskopf curve at the lower energies. At the higher energies, all of the (α,xn) reaction products have been measured, but it is probable that some of the reactions involving proton emission are now becoming important, and their cross sections have not been determined. It is only over the middle part of the excitation function, therefore, where $r_0=1.54\times10^{-13}$ cm that good agreement is obtained with the total cross sections described earlier at a higher atomic number.

In view of the apparent agreement in this mass region, it would be of interest to see if the nuclear radius parameter, r_0, could be used with some generality throughout the rest of the periodic table. Unfortunately, there are only sparse data available in a few regions of Z, and even in these cases there is no assurance that all of the important spallation products have been determined. However, an analysis of alpha-particle scattering data based upon a sharp-cutoff model has been made by Kerlee *et al.*[4] and independent values of both r_0 and R_α have been obtained. If the alpha-particle radius obtained from the scattering data is also used to derive a new value of the r_0 from the present reaction data, r_0 becomes 1.38×10^{-13} cm, which value may be compared to the r_0 value from scattering experiment of 1.41×10^{-13} cm. The agreement is excellent, although still slightly outside of the experimental errors involved. It is gratifying that the two essentially different types of experiments are consistent to within such narrow limits. It is perhaps worthwhile to note that fixing the total reaction cross section to within ten percent determines r_0 to within much less error (assuming the calculated excitation functions are precise).

The importance of the above comparison is clearly to indicate that the nuclear radius obtained by reaction data, even if the model can correctly be used in this situation, is clearly dependent upon an assumed value for the radius of the alpha particle.

The other available theoretical treatment with which comparison can be made is that based upon the optical model with a diffuse nuclear potential. Calculations on this model have been used successfully by Igo[6] to correlate data for the scattering of alpha particles through much wider angles than is possible by the sharp cutoff model. Using the same parameters as were obtained from analysis of the scattering data, Igo has recently derived total reaction cross sections[5] as a function of energy. His comparison of the previous fission-spallation data with this model gave agreement to within an order of magnitude. Our analysis of the data obtained from the present research is summarized in Fig. 15. It now seems apparent that the low-energy regions of the excitation function are in good agreement with curves calculated from the diffuse potential model, but that the latter predicts too high cross sections at the higher energies, with values considerably outside the experimental errors involved.

The situation could perhaps be improved somewhat by a change of parameters in the diffuse nuclear potential, particularly the radius parameter value, $r_0=1.17\times10^{-13}$ cm. However, it is not at all clear that such a change would then be consistent with the scattering data. This point can only be settled by some further trial-and-error calculations and adjustments of the parameters in the proposed diffuse potential.

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