PHYSICAL REVIEW VOLUME 172, NUMBER 4 20 AUGUST 1968

Fission of Iridium at Intermediate Excitation Energies*†

R. L. BRODZINSKI‡ AND J. W. COBBLE
Department of Chemistry, Purdue University, Lafayette, Indiana 47907

(Received 6 March 1968)

Cross sections have been determined radiochemically for 11 nuclides resulting from the fission induced in natural iridium ($Ir^{191,193}$) by 42.2-MeV He^4 ions. The mass distribution was found to be symmetric with a full width at half-maximum of 23 ± 2 mass units. The average number of prompt neutrons emitted per fission event was 2 ± 1, and the total fission cross section was found to be 45 ± 11 μb.

Total fission cross sections were determined at 13 different energies by a plastic-track technique. Analysis of the Γ_f/Γ_n ratio, which is a measure of the competition between fission and neutron emission, indicates the fission threshold to be 22.3 ± 0.7 MeV and the fission level-density parameter to be $a_f = A/10$. These results are compared with similar data from the fission of other nuclei.

INTRODUCTION

As studies of fission in the compound-nucleus energy range were expanded to include the elements lighter than thorium, the mass-yield distribution of fission fragments was found to change from a predominantly double-humped asymmetric mode[1–5] in the heavy elements ($Z\geq 90$), through fairly equal contributions of symmetric and asymmetric modes[6–9] in the intermediate elements ($84\leq Z\leq 89$), to the predominantly single-peaked symmetric distribution[8–15] in the light elements ($Z\leq 83$). Halpern[2] suggested in 1959 that the asymmetric mode of fission might again become important in the elements lighter than bismuth. Griffioen[16] considered the possibility that the ground-state deformations of the fissioning nucleus in the low-Z region might also enhance the asymmetric mode. The first data reported on the fission of gold[13] tended to substantiate this hypothesis because of the wide mass distribution of the fragments. Subsequent data on the fission of gold[14] and rhenium,[15] however, tended to refute this possibility.

It has been determined[10,17] that the fission excitation functions of the light nuclei rise sharply above the threshold and slowly level off as the excitation energy is increased. Since fission is an improbable reaction in the lighter elements, the ratio of the fission width to neutron-emission width is a very small number which increases rapidly as the excitation energy is increased. This ratio of Γ_f/Γ_n yields information about the fission barriers and level-density parameters. Data have been reported for the light elements in the closed-shell region (gold to bismuth), and recently Raisbeck and Cobble[17] gave results for thulium, lutetium, and rhenium which are in the region of deformed nuclei. This work showed the value of the fission level-density parameter a_f decreased from a value of $A/8$ for thulium and lutetium to a value of $A/20$ for natural rhenium. Since Burnett *et al.*[18] found a value of $a_f = A/11$ for the fission of gold, Raisbeck and Cobble had no satisfactory explanation for the anomalously low value of $A/20$ for rhenium, although there is a possibility that it results from the use of mixed isotopes.

The iridium system used in the present research was chosen because of its location between the deformed and closed-shell nuclei. Natural iridium also consists of a mixture of two isotopes of about the same percentage abundance as natural rhenium. Therefore, a valid comparison of the level-density parameters from the fission of rhenium and iridium can be made.

EXPERIMENTAL PROCEDURES

The iridium used in this research was 99.999+% pure[19] iridium metal sponge of natural isotopic abundance (37.3% Ir^{191}, 62.7% Ir^{193}). Cyclotron targets were prepared in thicknesses varying from 1 to 3 mg Ir/cm^2 by evaporation of the metal in an electron-beam vacuum evaporator onto substrates of pure silver and aluminum.

---

* This work was supported by the Atomic Energy Commission.

† From the Ph.D. thesis of Ronald L. Brodzinski, Purdue University, 1968 (unpublished).

‡ Present Address: Battelle Northwest Laboratory, Richland, Wash.

[1] E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1964), Vol. III.

[2] I. Halpern, Ann. Rev. Nucl. Sci. **9**, 245 (1959).

[3] I. R. Huizenga and R. Vandenbosch, in *Nuclear Reactions*, edited by P. M. Endt and P. B. Smith (North-Holland Publishing Co., Amsterdam, 1962), Vol. II, pp. 42–112.

[4] L. J. Colby, Jr., Mary LaSalle Shoaf, and J. W. Cobble, Phys. Rev. **121**, 1415 (1961).

[5] W. M. Gibson, Ph.D. thesis, University of California Radiation Laboratory Report No. UCRL-3493, 1956 (unpublished).

[6] R. C. Jensen and A. W. Fairhall, Phys. Rev. **109**, 942 (1958).

[7] R. C. Jensen and A. W. Fairhall, Phys. Rev. **118**, 771 (1960).

[8] H. C. Britt, H. E. Wegner, and J. C. Gursky, Phys. Rev. **129**, 2239 (1963).

[9] J. P. Unik and J. R. Huizenga, Phys. Rev. **134**, B90 (1964).

[10] A. W. Fairhall, Phys. Rev. **102**, B35 (1956).

[11] T. T. Sugihara, J. Roesmer, and J. W. Meadows, Jr., Phys. Rev. **121**, 1179 (1961).

[12] R. Vandenbosch and J. R. Huizenga, Phys. Rev. **127**, 212 (1962).

[13] E. F. Neuzil and A. W. Fairhall, Phys. Rev. **129**, 2705 (1963).

[14] F. L. Lisman, H. W. Brandhorst, and J. W. Cobble, Phys. Rev. **140**, B863 (1965).

[15] C. Menninga and J. W. Cobble, Phys. Rev. **153**, 1294 (1967).

[16] R. D. Griffioen, Ph.D. thesis, Purdue University, 1960 (unpublished).

[17] G. M. Raisbeck and J. W. Cobble, Phys. Rev. **153**, 1270 (1967).

[18] D. S. Burnett, R. C. Gatti, F. Plasil, P. B. Price, W. J. Swiatecki, and S. G. Thompson, Phys. Rev. **134**, B952 (1964).

[19] Obtained from the United Mineral and Chemical Corp., New York.

The silver backing foils were prepared by evaporation of 99.9999% pure silver shot[19] in a resistance heated vacuum evaporator onto 3-mil-thick Lexan polycarbonate plastic strips.[20] The Lexan was subsequently dissolved in dichloromethane. The 1-mil-thick aluminum foils[19] were rolled from 99.999% pure aluminum. In the radiochemical studies of fission fragments a catcher foil of the same material as the backing foil was placed over the targets. Due to the extremely slow rate of dissolution of iridium metal, only this backward (to the direction of the beam) catcher foil was dissolved in the fission-product separation chemistry, and appropriate corrections were applied for the recoil collection efficiency.[21] This procedure eliminates spurious activities from spallation reactions on impurities in the iridium.

The helium-ion irradiations in this research were carried out on the Argonne National Laboratory 60-in. cyclotron. In the bombardments in which chemical separation of fission fragments were made, a beam energy of 46.4 MeV and current of ~7 μA for 5 h were used. The excitation function bombardments were carried out at selected energies obtained by remotely inserting suitable aluminum degraders into the beam path; the current was held at approximately 1 μA.

The individual fission products were separated and purified by suitable modifications[21] of various radiochemical procedures[22,23] and counted in low-background anticoincidence shielded β-counting systems having backgrounds of 0.1 to 0.2 counts/min. The resulting decay curves were resolved by a least-squares analysis program on an IBM 7094 computer. Absolute fission cross sections were determined for zinc 72, arsenic 77, bromine 83, strontium 91, yttrium 93, zirconium 97, molybdenum 99, ruthenium 105, palladium 109, palladium 112, and cadmium 115. These isotopic cross sections were corrected for charge distribution by the "constant-charge-ratio" postulate which has been used successfully in these laboratories[14] for the fission of gold. The best smooth curve which represented the experimental and reflected points was obtained when the average number of prompt neutrons, $\bar{\nu}$, emitted per fission event was set at 2±1.

The fission excitation function was determined by a fission-track technique in which the fission fragments from a thin target recoil into a 3-mil-thick strip of Lexan polycarbonate plastic.[20] The characteristics of this plastic as a heavy-ion detector have been reviewed,[24] and the physical arrangement of the apparatus is described elsewhere in detail.[17,25] After bombardment the plastic detector strips were etched in a 6 *M* NaOH solution at 60°C for 60 min, rinsed with water, and blotted dry. These strips were mounted on a microscope slide, and the tracks counted with a Leitz Ortholux micrometer stage microscope. A photomicrography of some typical tracks from the fission of iridium is shown in Fig. 1. The number of fission tracks in a chosen area of the plastic strip detector were compared to the number of tracks in the same geometry in the plastic strip from a Re^{185} target irradiated under similar conditions. The desired cross section was then calculated by a comparison to the absolute rhenium fission cross section.[25]

In the case of the lighter elements the fission width can be related to the total fission cross section. Neutron emission is by far the most probable reaction under the conditions of the present study, and the neutron width was related to the total reaction cross section. The ratio of $\Gamma_f/\Gamma_n \approx \sigma_f/\sigma_r$ was calculated from the measured fission cross sections and the total reaction cross sections which were estimated according to the method of Huizenga and Igo.[26] The theoretical energy dependence of the ratio of the fission width to neutron-emission width used in the present analysis is essentially that sug-

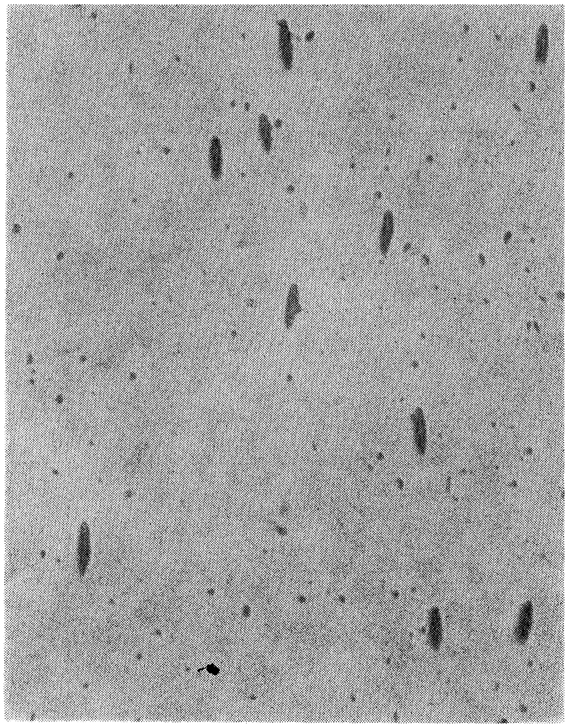

FIG. 1. Photomicrograph of fission-fragment tracks from the helium-ion-induced fission of iridium. Each track is about 10μ long.

---

[20] Obtained from the General Electric Corp., Pittsfield, Mass.

[21] R. L. Brodzinski, Ph.D. thesis, Purdue University, 1968 (unpublished).

[22] C. Menninga, Ph.D. thesis, Purdue University, 1966 (unpublished).

[23] F. L. Lisman, Ph.D. thesis, Purdue University, 1965 (unpublished).

[24] R. L. Fleisher, P. B. Price, and R. M. Walker, Ann. Rev. Nucl. Sci. **15**, 1 (1965).

[25] G. M. Raisbeck, Ph.D. thesis, Purdue University, 1966 (unpublished).

[26] J. R. Huizenga and G. J. Igo, Nucl. Phys. **29**, 462 (1962).

TABLE I. Fission-product cross sections for the fission of iridium by 42.2-MeV helium ions.

Nuclide	Uncorrected cross section (nb)	Corrected cross section[a] $\bar{\nu}=2$ (nb)
Zn^{72}	33.0±3.2	33.0±3.2
As^{77}	253±32	253±32
Br^{83}	1410±140	1410±140
Sr^{91}	3270±330	3270±330
Y^{93}	3860±370	3860±370
Zr^{97}	4120±390	4160±390
Mo^{99}	3200±310	3200±310
Ru^{105}	3030±330	3030±330
Pd^{109}	1810±180	1810±180
Pd^{112}	836±85	862±88
Cd^{115}	815±85	815±85

[a] Corrected for independent yields using the CCR hypothesis (see text).

gested by Huizenga and Vandenbosch[3]:

$$\Gamma_f/\Gamma_n = K_0 \frac{a_n[2a_f^{1/2}(E-E_f)^{1/2}-1]}{4A^{2/3}a_f(E-B_n)} \times \exp[2a_f^{1/2}(E-E_f)^{1/2}-2a_n^{1/2}(E-B_n)^{1/2}], \quad (1)$$

where a_n and a_f are the level-density parameters for neutron emission and fission, respectively, E is the excitation energy, E_f is the fission barrier, B_n is the neutron binding energy, A is the mass number of the compound nucleus, and K_0 is a constant. The experimental values of Γ_f/Γ_n were analyzed in terms of Eq. (1) using a least-squares fitting procedure,[27] by (a) holding one of the three variable parameters a_f, a_n, and E_f constant and varying the other two, and (b) by "floating" all three parameters. The best values for the variables were chosen on the basis of the sum of the squares of the deviations, ψ^2.

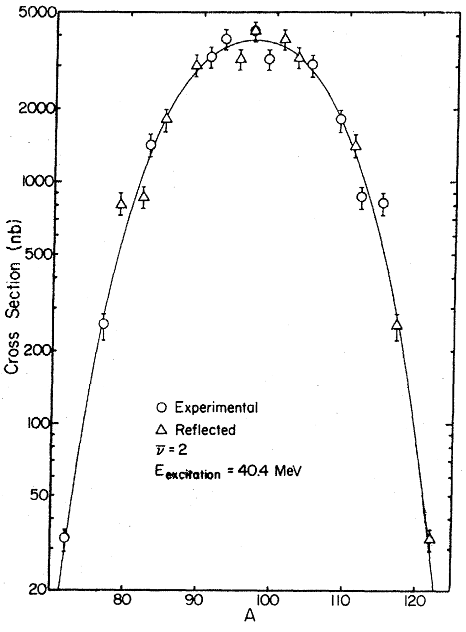

FIG. 2. Fission mass-yield curve for natural iridium bombarded by 42.2-MeV helium ions.

EXPERIMENTAL RESULTS

The concentration of heavy-element impurity in the target material was determined by an activation analysis procedure described elsewhere.[25] The data indicated a value of only one atom part per billion as an upper limit for a heavy-element impurity (Th or U) in the iridium targets. This value was sufficiently low so that corrections to the data were not necessary. The isotopic cross sections and corrections to isobaric yields are summarized in Table I. The limits of error for each nuclide were determined from an over-all analysis of the errors arising from uncertainties in the beam energy and current, the target thickness, the counter and collection efficiencies, the chemical yield, and the standard deviations indicated from the computer analysis of the decay curves. The resulting mass-yield distribution is given in Fig. 2. The full width at half-maximum (FWHM) of this distribution is 23±2 mass units, and the integrated total fission cross section is 45±11 μb.

TABLE II. Fission cross sections for natural iridium.

Excitation energy E_x (MeV)	Total reaction cross section σ_R(b)	Measured fission cross section[a] σ_f(b)	Γ_f/Γ_n $\approx\sigma_f/\sigma_R$
44.6	1.976	$1.045\pm0.020\times10^{-4}$	5.29×10^{-5}
43.3	1.940	$7.23\pm0.17\times10^{-5}$	3.73×10^{-5}
41.4	1.877	$4.03\pm0.13\times10^{-5}$	2.15×10^{-5}
40.2	1.836	$2.744\pm0.072\times10^{-5}$	1.50×10^{-5}
38.8	1.780	$1.817\pm0.046\times10^{-5}$	1.02×10^{-5}
37.4	1.725	$1.102\pm0.030\times10^{-5}$	6.39×10^{-6}
36.2	1.677	$6.57\pm0.18\times10^{-6}$	3.92×10^{-6}
35.1	1.621	$3.54\pm0.10\times10^{-6}$	2.18×10^{-6}
33.6	1.544	$1.611\pm0.044\times10^{-6}$	1.04×10^{-6}
32.4	1.482	$7.55\pm0.24\times10^{-7}$	5.09×10^{-7}
31.2	1.406	$3.93\pm0.14\times10^{-7}$	2.80×10^{-7}
30.0	1.326	$1.514\pm0.081\times10^{-7}$	1.14×10^{-7}
28.7	1.240	$5.54\pm0.38\times10^{-8}$	4.47×10^{-8}

[a] For computational purposes, one more significant figure is recorded than is justified from known errors in the beam energies.

The fission cross sections measured by the plastic-track technique are given in Table II using a weighted average Q value of -1.8 MeV for the mixed isotopes of iridium to determine the excitation energy. The errors are largely based on the statistical accuracy of the number of tracks observed. Figure 3 is a plot of the resulting excitation function.

The results of the least-squares fitting procedure of the theoretical Γ_f/Γ_n function to the experimental data are summarized in Table III. Nine different trials were made where all three variable parameters were "floated" in one trial, the value of the fission threshold E_f was held constant (italicized in Table III) in four trials, and

---

[27] R. H. Moore and R. K. Ziegler, Los Alamos Scientific Laboratory Report No. LA-2367, 1960 (unpublished).

the value of the level-density parameter at the saddle point a_f was held constant (italicized in Table III) at the values of $a_f=A/8$, $A/10$, $A/12$, and $A/20$ in the last four trials. The best over-all correlation gave a value of 22.3±0.7 MeV for the fission threshold and yielded a value for the level-density parameter of $a_f=A/10$, with a ratio of $a_f/a_n=1.18$. Figure 4 shows the "best-fit" curve to the data and the extreme case of setting $a_f=A/20$.

DISCUSSION

The value of 23±2 mass units measured for the FWHM of the symmetric mass-yield distribution is quite similar to the experimental results[8,9,14,15] obtained for the fission of nuclides from rhenium to bismuth at similar excitation energies. This value is also in good agreement with the theoretical calculations of Nix and Swiatecki.[28] The effect of using mixed isotopes has been investigated by Raisbeck and Cobble[17]; their data indicate that the use of a weighted average for all mass-dependent parameters for the mixed isotopes yield results which are consistent with those of the separated isotopes. Using this procedure the mass-yield curve for natural iridium was resolved into two components corresponding to the fragment yields from the individual isotopes. It was found that both curves had a (FWHM) of 23 mass units, with an average number of 2±1 prompt neutrons emitted, and that the iridium 191 accounted for approximately 63% of the total fission cross section in natural iridium.

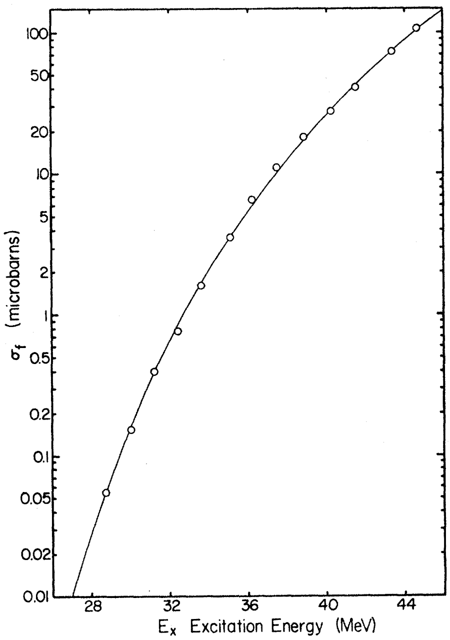

FIG. 3. Measured fission cross sections of the helium-ion-induced fission of natural iridium.

TABLE III. Least-squares fitting of the theoretical Γ_f/Γ_n function for the fission excitation function of iridium.[a]

Barrier E_f(MeV)	Level-density parameters a_f	a_n	a_f/a_n	Sum of squares ψ^2
22.3±0.7	19.3 ±3.5	16.3	1.18	0.0086
22.0	20.7 ±0.3	17.5	1.18	0.0087
24.0	12.5 ±0.2	10.5	1.19	0.16
26.0	7.21±0.27	6.19	1.17	0.073
28.0	3.33±0.38	3.24	1.30	0.533
21.4±0.1	*24.0*	20.4	1.18	0.0097
22.3±0.1	*19.2*	16.2	1.18	0.0086
23.0±0.1	*16.0*	13.5	1.19	0.0097
25.0±0.1	*9.67*	8.14	1.18	0.032

[a] Values in italics were fixed in obtaining each set of parameters.

It has been observed[1,2] that $\log\Gamma_f/\Gamma_n$ is an approximately linear function of Z^2/A at constant excitation energy. Such an analysis is given for 18 nuclides in Fig. 5 at an excitation energy of 40 MeV. The data[17,18,29–31] start to deviate from the simple relationship at radium 226, which is also the point at which measurable amounts of asymmetric fission become evident.

This work further substantiates[17] that the expression given in Eq. (1) adequately describes the energy dependence of Γ_f/Γ_n in the light elements. In the present research the normal neutron binding energy was used for

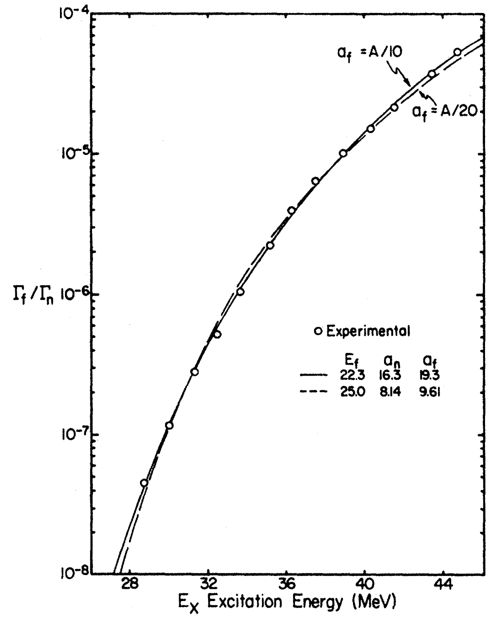

FIG. 4. Theoretical fitting of the Γ_f/Γ_n function for the helium-ion-induced fission of $Ir^{191,193}$, assuming values of $A/10$ and $A/20$ for the level-density parameter.

---

[28] J. R. Nix and W. J. Swiatecki, Nucl. Phys. 71, 1 (1965).

[29] R. Vandenbosch, and J. R. Huizenga, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (Pergamon Press, Ltd., London, 1960), Vol. 15, paper P/688.

[30] J. E. Gindler, G. L. Bate, and J. R. Huizenga, Phys. Rev. 136, B1333 (1964).

[31] A. Khodai-Joopari, R. C. Gatti, and S. G. Thompson, University of California Radiation Laboratory Report No. UCRL-11828, 1965 (unpublished).

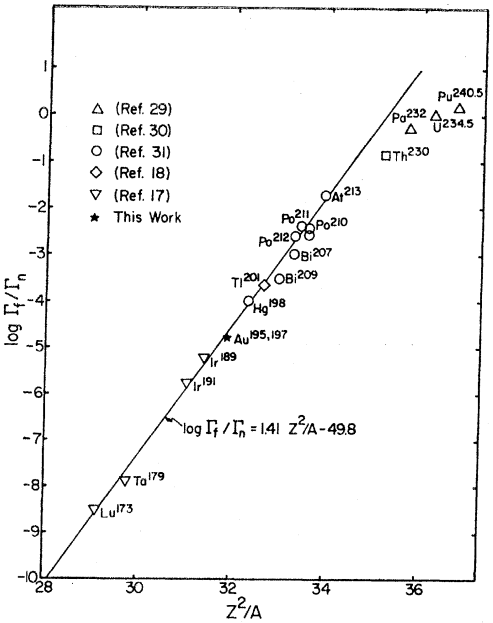

FIG. 5. Fission to neutron-emission widths as a function of Z^2/A at an excitation energy of 40 MeV. The symbols refer to the compound nucleus. Logarithms are to the base 10.

fitting this theoretical Γ_f/Γ_n function to the data rather than an effective binding energy which takes into account pairing and shell corrections.

The "best" value of $a_f = A/10$ determined for the iridium system is significantly different from the value of $a_f = A/20$ reported for the rhenium system.[17] Since iridium lies between rhenium and the closed-shell nuclei, it is apparent that the low value of a_f for rhenium is not due to its proximity to the closed-shell region. Further, since iridium and rhenium have similar natural isotopic abundances, the anomalously low value for rhenium cannot be ascribed to the use of mixed isotopes.

Sikkeland[32] has concluded from the results of his heavy-ion bombardments on elements from cesium to bismuth that the ratio of a_f/a_n was a constant value of 1.22 ± 0.05, independent of the nature of the target nucleus. The ratio of $a_f/a_n = 1.18 \pm 0.01$ determined in this research, when compared to the values for thulium,[17] lutetium,[17] rhenium,[17] and gold[18] of 1.08, 1.11, 1.17, and 1.35, respectively, clearly demonstrates a steady increase of the function with increasing Z of the target nucleus.

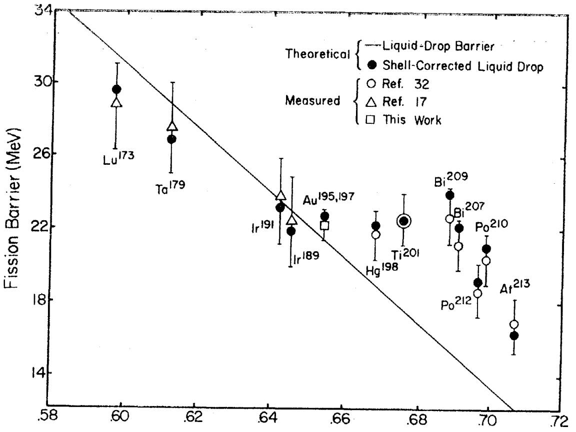

FIG. 6. Comparison of measured and predicted fission barriers for low-Z systems.

Experimental fission barrier energies or "thresholds" are plotted as a function of the "fissionability parameter" x in Fig. 6, along with the shell-corrected values[33] based on the simple liquid-drop model.[34] The fissionability parameter is defined[18] as the ratio of $(Z^2/A)/(Z^2/A)_{\text{critical}}$, where $(Z^2/A)_{\text{critical}} = 48.4$. The barrier energies given in Fig. 6 are consistent with a uniform value of the level-density parameter of $a_f = A/10$. The excellent agreement between the experimental points and the predicted points substantiates the shell-corrected liquid-drop model of Myers and Swiatecki.[33] It should be pointed out, however, that the experimental excitation function for the rhenium fission system clearly indicates a value of $a_f = A/20$. The threshold energy based on this value, $E_f = 26.6$ MeV, is not in agreement with the correlations of Fig. 6. In this respect, the rhenium system still remains somewhat of an anomaly.

ACKNOWLEDGMENTS

The authors wish to express their appreciation for the many helpful suggestions and assistance of M. Oselka and the staff at the Argonne National Laboratory cyclotron, and to Clarence Menninga, Grant M. Raisbeck, Hari Iyer, and Ken W. MacMurdo of this laboratory for their helpful comments and discussions.

---

[32] T. Sikkeland, Phys. Rev. **135**, B669 (1964).

[33] W. D. Myers and W. J. Swiatecki, University of California Radiation Laboratory Report No. UCRL-11980, 1965 (unpublished).

[34] S. Cohen and W. J. Swiatecki, Ann. Phys. (N. Y.) **22**, 406 (1963).

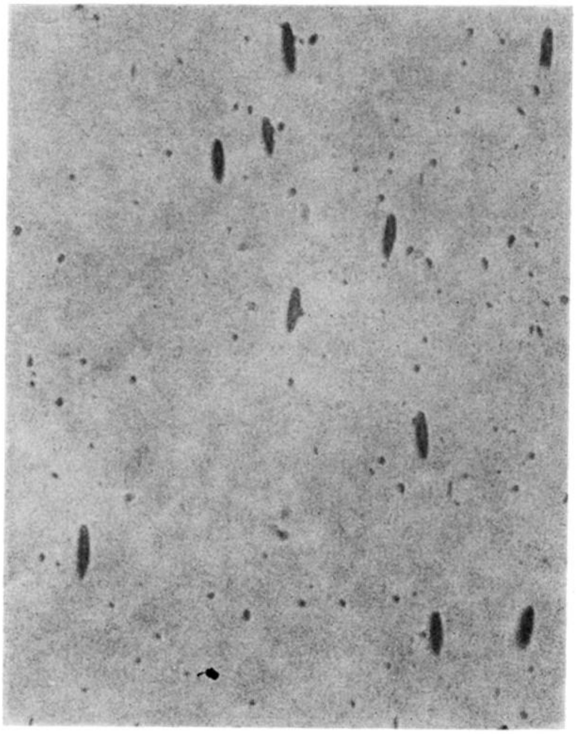

FIG. 1. Photomicrograph of fission-fragment tracks from the helium-ion-induced fission of iridium. Each track is about 10μ long.