the proton spectrum, while background due to prompt fission neutrons might very well be present in this spectrum. The latter type of background would, however, cause the mass-distribution curve to resemble that of binary fission. Hence, this type of background cannot account for the different behavior of the proton fission mass distribution.

The anomalous behavior of proton-accompanied fission has been previously pointed out by Raisbeck and Thomas[7] in connection with the energy spectrum and angular distribution of these particles. The results of our experiment, therefore, indicate that the different behavior of proton-accompanied fission is also observed in connection with the properties of the fission fragments. It should, however, be added that the behavior of the total fragment kinetic energy distribution and of the average fragment kinetic energy as a function of fragment mass are in proton fission similar to the other fission modes studied here. Proton-accompanied fission is perhaps at this stage the least understood mode of light-particle fission and further experimental work would be useful to the understanding of this process.

ACKNOWLEDGMENTS

The authors are very thankful to Professor Zeev Fraenkel for his very helpful advice and criticism. One of the authors (S.K.) is grateful to the Guggenheim Foundation for a grant.

PHYSICAL REVIEW VOLUME 182, NUMBER 4 20 JUNE 1969

Low-Yield Products from Fission of Th^{232}, U^{235}, and U^{238} with 14.8-MeV Neutrons*

D. R. Nethaway, B. Mendoza, and T. E. Voss

Lawrence Radiation Laboratory, University of California, Livermore, California 94550

(Received 12 August 1968; revised manuscript received 18 February 1969)

We have measured the fission yields of a number of products from 14.8-MeV neutron fission of Th^{232}, U^{235}, and U^{238}. The fission products chosen are all on the wings of the mass-yield curves and are formed in very low yield. They extend from Ni^{66} to Zn^{72} and from Sm^{153} to Er^{172}. The amount formed of each product was determined by absolute β and γ counting techniques. The number of fissions in each target was calculated from the target mass, the fission cross section, and the neutron flux. The neutron flux was measured by means of the $Y^{89}(n, 2n)Y^{88}$ reaction with Y_2O_3 monitor foils. The results show that, within experimental uncertainty, the wings of the mass-yield curves are consistent with Gaussian functions. These Gaussian curves allow interpolation and prediction of fission yields of unmeasured products. The widths of the mass-yield curves for U^{235} and U^{238} are almost the same, while that of Th^{232} is significantly narrower. The centers of the Gaussian distributions are shifted to higher mass numbers than would be predicted from the average total neutron emission in fission. The effect of target impurities on the measured fission yields was shown to be generally small. An attempt was made to examine the effect of nuclear charge distribution on the mass yields. This effect, which would cause the observed fission yields to be less than the total mass yield, is probably significant only for the yields of masses 166 and 172. As a check on our experimental method we also remeasured the fission yields of three products near the peaks of the mass-yield curves. Our results are consistent with those reported before.

INTRODUCTION

The mass-yield curves for the fission of Th^{232}, U^{235}, and U^{238} induced by 14-MeV neutrons have been characterized fairly well in the areas of high fission yield. Much fewer experimental data have been reported for products formed in low yield. This has been due mainly to the relatively weak sources of 14-MeV neutrons that are available, compared, for example, to sources of thermal or reactor neutrons. The amount of experimental data obtained for 14-MeV neutron fission is still large compared to that obtained for fast neutron fission at other energies. The deuterium-tritium fusion reaction $(d+t\rightarrow n+\alpha)$ provides a unique source of monoenergetic neutrons with energy about 14 MeV.

For U^{235} and U^{238} the data taken at 14 MeV indicate the usual double-humped asymmetric mass-yield distribution.[1–4] For 14-MeV neutron fission of Th^{232} this asymmetric distribution is modified by a small central peak due to symmetric fission.[5–8]

Very few data exist for the products on the wings of the mass-yield curves. For Th^{232} no yields have been reported below mass 83 or above mass 157. For U^{235}

* Work performed under the auspices of the U.S. Atomic Energy Commission.

[1] S. Katcoff, Nucleonics **18**, 201 (1960).
[2] D. G. Vallis and A. O. Thomas, Atomic Weapons Research Establishment Report No. AWRE-O-58-61, 1962 (unpublished).
[3] M. P. Menon and P. K. Kuroda, J. Inorg. Nucl. Chem. **26**, 401 (1964).
[4] R. H. James, G. R. Martin, and D. J. Silvester, Radiochimica Acta **3**, 76 (1964).
[5] K. M. Broom, Phys. Rev. **133**, B874 (1964).
[6] S. J. Lyle, G. R. Martin, and J. E. Whitley, Radiochimica Acta **3**, 80 (1964).
[7] R. Ganapathy and P. K. Kuroda, J. Inorg. Nucl. Chem. **28**, 2071 (1966).
[8] T. Mo and M. N. Rao, J. Inorg. Nucl. Chem. **30**, 345 (1968).

the measured yields range from mass 66 to mass 156, and for U^{238} they range from mass 83 to mass 156.

The new insulated core transformer (ICT) neutron generator at this Laboratory provides a higher-intensity source of 14-MeV neutrons than has been available before. We have made use of this high flux to measure some of the fission products that are formed in very low yields. Our measurements extend from mass 66 to 72 and from mass 153 to 172. We were able to measure fission yields as low as 2×10^{-7}. In addition, we measured the fission yields of a few products on the peaks of the mass-yield curve to allow a better comparison to be made between our measurements and those of others.

EXPERIMENTAL DETAILS

The 14-MeV neutron irradiations were performed at the ICT neutron generator at the Lawrence Radiation Laboratory at Livermore. The neutrons are produced by the reaction of deuterons on tritium in a rotating target assembly.[9] The deuteron accelerator is designed to deliver a 10-mA beam with energy up to 500 keV. The deuteron beam strikes a 6-in.-diam titanium tritide target rotating at 1100 rpm. The target is cooled by a water spray on the back side. The target rotation increases the effective area of tritium being heated, allowing better cooling and, therefore, high beam currents. To minimize the effects of scattered neutrons, the target area is located in a large, fairly empty room. The radiochemistry target was placed at 0° to the deuteron beam, and just outside the thin plastic water catch cage. At this position the neutron energy was 14.8 ± 0.3 MeV, and the flux $\sim6\times10^{10}$ cm^{-2} sec^{-1}. The neutron flux was monitored continuously during the irradiations with a proton telescope counter. This allowed a correction to be made for the small variations in flux.

The radiochemistry target generally consisted of a set of Th^{232}, U^{235}, and U^{238} foils. The U^{235} foils were covered with 1-mil U^{235} foils to reduce recoil losses. Each of the three targets was sealed in a thin plastic bag to avoid cross-contamination. At the front and back of the packet were 10-mil foils of Y_2O_3 pressed in plastic. Their function as a neutron flux monitor will be discussed later. Finally, a covering of 20-mil cadmium foil surrounded the entire package to reduce the low-energy neutron background. The target foils and monitor foils were all cut to the same size. The target foils varied in thickness from 5 to 20 mils and in weight from 1 to 15 g.

The uranium foils were made from enriched materials (93% U^{235} and 99.8% U^{238}, respectively), while the thorium foil was the natural material. The fission yields for U^{235} were corrected for the contribution from the fission of the U^{238} ($\sim$5% of the mass), using the measured U^{238} fission yields. Spectrographic analysis of the thorium showed the presence of 40 ppm each of Fe, Ni, Cu, and Zn, 5 ppm Tm, and 2 ppm Er. However, the most sensitive indication of impurities in all three targets proved to be their $(n, 2n)$ products, such as Ni^{57}, Cu^{64}, and Zn^{65}.

The irradiations were usually 8 h in length, producing up to 4×10^{12} fissions in the Th^{232}, 3×10^{13} fissions in the U^{235}, and 1.5×10^{13} fissions in the U^{238}. After the neutron irradiations, the target foils were dissolved in the presence of 10–20-mg amounts of carriers for each of the desired products. Each of the elements was then separated from the entire solution. The bulk of the thorium was first removed by passing through a Dowex-1 anion-exchange resin column in $8N$ HNO_3 solution, the thorium remaining fixed on the column. The uranium was removed by passing through a Dowex-50 cation-exchange resin column in a 10%-6N-HNO_3–90% tetrahydrofuran solution. The uranium was washed through the column; the other elements were then eluted with a dilute HCl solution. Each element was finally purified by standard radiochemical procedures.[10] The individual rare-earth elements were separated on Dowex-50 ion-exchange resin columns using ammonium lactate as the eluant. The Y_2O_3 (plastic) monitor foils were ignited to destroy the plastic, and then dissolved. Aliquots were taken to prepare the final Y_2O_3 samples. The nuclides Dy^{166} and Er^{172} were determined by separating and counting the Ho^{166} and Tm^{172} daughter activities. These separations were done on the Dowex-50 ion-exchange columns.

The samples were counted on gas-flow β proportional counters or NaI(Tl) γ counters. Details of these measurements are given in Table I. Several methods were used for determining the necessary counting efficiencies. These were (1) comparison with a 4π counter, (2) comparison with calibrated sodium iodide and germanium detectors used with pulse-height analyzers, (3) the use of calibrated standard solutions,[11] and (4) the use of an experimentally determined curve of β counting efficiency versus mean β energy.

The Y_2O_3 (plastic) neutron-flux monitor foils were calibrated at the ICT neutron source in separate experiments. Stacks of Al-Y_2O_3-Al foils covered with Cd foil were irradiated with neutrons of the same energy as that used in the fission measurements. The flux in the aluminum foils was calculated from the known (n, α) cross section[12] of 113 ± 5 mb and the measured Na^{24} disintegration rates. The aluminum foils were counted on a β proportional counter. The counting efficiency for Na^{24} in these foils was measured by 4π counting and by 4π β-γ coincidence counting. The two

[9] R. Booth, Lawrence Radiation Laboratory Report No. UCRL-70183, 1967 (unpublished).

[10] M. Lindner, Lawrence Radiation Laboratory Report No. UCRL-14258, 1965 (unpublished).

[11] Obtained from International Atomic Energy Agency, Vienna, Austria.

[12] J. Stehn, M. Goldberg, B. Magurno, and R. Wiener-Chasman, in, *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz (U.S. Government Printing Office, Washington 25, D.C., 1964), 2nd ed., Suppl. 2, Vol. 1, Z=1 to 20.

TABLE I. Details of radioactivity and counting efficiency measurements.

Nuclide	Counter[a]	Basis for counting efficiency	$E_\gamma(I_\gamma)$ [b]	Error in efficiency (%)
Ni^{66}	β	β-efficiency curve		10
Cu^{67}	β	β curve and NaI PHA	184 keV (0.43)	10
Zn^{72}	β	Ge PHA[c]	835 keV (0.955)	3
Y^{88}	γ	d		5
Y^{93}	β	β-efficiency curve		10
Ba^{140}	γ	NaI and Ge PHA	1596 keV (0.962)	5
Nd^{147}	γ	NaI and Ge PHA	531 keV (0.131)	7
Sm^{153}	β	β curve and NaI PHA	103 keV (0.28)	10
Gd^{159}	β	β-efficiency curve		10
Tb^{161}	β	4π counter		5
$Dy(Ho)^{166}$	β	4π counter		5
Er^{169}	β	4π counter		5
$Er(Tm)^{172}$	β	4π counter		5

[a] Lower and upper discriminator settings on the γ counters were 1600–1910 keV for Y^{88}, 1100–1700 keV for Ba^{140}, and 470–650 keV for Nd^{147}.
[b] Energy of γ ray (absolute intensity, photons/disintegration).
[c] By direct comparison with the 835-keV photon ($I_\gamma=1.00$) in a standard sample (Ref. 11) of Mn^{54}.
[d] Factor relating cpm $Y^{88}/g\ Y_2O_3$ to 14.8-MeV n/cm^2 obtained by comparison to the $Al^{27}(n,\alpha)Na^{24}$ reaction in separate experiments. See text for details.

methods gave good agreement, and the uncertainty in the Na^{24} efficiency is about 2%.

The Y^{88} produced in the Y_2O_3 foils by the $(n, 2n)$ reaction was measured in a γ counter (see Table I). This reaction has an energy threshold of 11.6 MeV and is not sensitive to low-energy neutrons. From these measurements we obtain a factor relating the Y^{88} counting rate per gram of Y_2O_3 to the 14.8-MeV neutron dose. The estimated uncertainty in this factor is $\pm5\%$. Flux measurements using the Y_2O_3 foils were compared with those obtained from proton-telescope counter measurements and found to agree within a few percent.

FISSION YIELD MEASUREMENTS

Results

The results of the fission yield measurements are summarized in Table II. The results are the averages of about three determinations in each case for the low-yield products. For Ba^{140} two measurements were made, and for Y^{93} and Nd^{147}, only one. The number of fissions occurring in each target foil in each irradiation was calculated from the product of the number of target atoms, the 14.8-MeV fission cross section for the target nuclide, and the integrated 14.8-MeV neutron flux. The fission yields were then calculated as the total atoms produced of each product divided by the number of fissions.

The neutron fluxes in the target foils were obtained by interpolating between the fluxes determined by the $(n, 2n)$ reaction on the two Y_2O_3 monitor foils. The fission cross sections[13] used are 0.391 b for Th^{232}, 2.24 b for U^{235}, and 1.24 b for U^{238}. The uncertainty in these cross sections is a few percent. Coupling this uncertainty with that of the flux measurements leads to a total uncertainty in the number of fissions of about 6%. The errors given for our results in Table II reflect only the uncertainty in the counting efficiencies, the counting statistics, and the agreement between different experiments. They do not include the 6% uncertainty in the number of fissions, which is essentially a constant uncertainty in all of these measurements.

The results given in Table II for U^{235} fission have been corrected for the contribution of the 5% U^{238} in the foils using the measured U^{238} fission yields. These corrections were about 1% for the light-mass products and about 4% for the heavy-mass products. The effect of low-energy neutrons has been neglected due to the combination of generally lower fission cross sections and much lower fission yields at lower neutron energies, although for the few peak yields that we have measured, low-energy fission could be significant.

The range in uranium of fission products from 14.5-MeV neutron fission of U^{238} has been measured by Desai and Menon.[14] The value of the range varies with mass number, and the average is about 10 mg/cm². We have extrapolated their results to the range of mass numbers of interest here, and find that for the very light fragments the range is about 10 mg/cm², and for the very heavy fragments the range is about 7 mg/cm².

[13] W. Hart, United Kingdom Atomic Energy Authority Report No. AHSB(S)R124, 1967 (unpublished).
[14] R. D. Desai and M. P. Menon, Phys. Rev. **150**, 1027 (1966).

TABLE II. Fission yields of Th^{232}, U^{235}, and U^{238} with 14.8 neutrons.

Product nuclide	Half-life (days)	Th^{232} Fission: Fission yield[a]	Th^{232} Fission: Previous measurement	U^{235} Fission: Fission yield[a]	U^{235} Fission: Previous measurement	U^{238} Fission: Fission yield[a]	U^{238} Fission: Previous measurement
Ni^{66}	2.29	$(1.31\pm0.13)\times10^{-6}$		$(2.8\pm0.3)\times10^{-6}$	$(4.0\pm0.4)\times10^{-6}$ [b]	$(8.5\pm0.9)\times10^{-7}$	
Cu^{67}	2.56	$(2.6\pm0.6)\times10^{-6}$		$(6.5\pm0.9)\times10^{-6}$		$(1.4\pm0.4)\times10^{-6}$	
Zn^{72}	1.94	$(7.0\pm0.6)\times10^{-5}$		$(6.3\pm0.3)\times10^{-5}$	$(7.8\pm0.8)\times10^{-5}$ [b]; 3.0×10^{-5} [c]	$(3.0\pm0.4)\times10^{-5}$	
Y^{93}	0.427	0.053 ± 0.005	0.058 ± 0.004[d]	0.054 ± 0.005		0.044 ± 0.004	0.041 ± 0.001[e]
Ba^{140}	12.80	0.058 ± 0.002	0.060 ± 0.004[g]	0.0425 ± 0.0017	0.051[f]; 0.044[h]	0.0446 ± 0.0018	0.045 ± 0.002[i]
Nd^{147}	11.04	0.0181 ± 0.0013	0.017 ± 0.001[j]	0.0164 ± 0.0011	0.020 [e]	0.0220 ± 0.0015	0.020 ± 0.001[k]
Sm^{153}	1.94	$(8.6\pm0.9)\times10^{-4}$	$(8.5\pm1.0)\times10^{-4}$ [j]	$(2.2\pm0.2)\times10^{-3}$	2.4×10^{-3} [e]	$(4.2\pm0.4)\times10^{-3}$	$(3.9\pm0.2)\times10^{-3}$ [k]
Gd^{159}	0.773	$(4.4\pm0.4)\times10^{-5}$		$(1.27\pm0.13)\times10^{-4}$		$(2.6\pm0.3)\times10^{-4}$	
Tb^{161}	6.96	$(1.06\pm0.06)\times10^{-5}$		$(5.6\pm0.4)\times10^{-5}$	6.0×10^{-5} [e]	$(8.9\pm0.5)\times10^{-5}$	
Dy^{166}	3.40	$(2.9\pm0.2)\times10^{-7}$		$(2.8\pm0.2)\times10^{-5}$		$(6.3\pm0.6)\times10^{-6}$	
(Ho^{166})	(1.116)						
Er^{169}	9.5	$(2.3\pm0.8)\times10^{-7}$		$(8.0\pm0.6)\times10^{-7}$		$(1.29\pm0.09)\times10^{-6}$	
Er^{172}	2.08			$(1.8\pm0.2)\times10^{-7}$		$(2.1\pm0.7)\times10^{-7}$	
(Tm^{172})	(2.65)						

[a] Experimental errors given here do not include a systematic 6% uncertainty in the measurement of the number of fissions. This additional uncertainty should be included when considering absolute fission yields; it was omitted here to allow a more meaningful comparison between relative fission yields.

[b] Reference 2.

[c] Calculated from data in H. Hicks, H. Levy, W. Nervik, P. Stevenson, J. Niday, and J. Armstrong, Phys. Rev. **128**, 700 (1962).

[d] Average of results from Refs. 6 and M. Thein, M. Rao, and P. Kuroda, J. Inorg. Nucl. Chem. **30**, 1145 (1968).

[e] Reference 4.

[f] Average of results from H. Hicks, H. Levy, W. Nervik, P. Stevenson, J. Niday, and J. Armstrong, Phys. Rev. **128**, 700 (1962) and G. P. Ford and J. S. Gilmore, Los Alamos Scientific Laboratory Report No. LA-1997, 1956 (unpublished).

[g] Reference 5.

[h] Calculated from A. Protopopov, G. Tolmachev, V. Ushatskii, R. Venediktova, I. Krisyuk, L. Rodionova, and G. Iakovleva, At. Energ. (USSR) **5**, 130 (1958), assuming mass-99 yield 0.051.

[i] Average of results from Ref. 4 and G. P. Ford and J. S. Gilmore, Los Alamos Scientific Laboratory Report No. LA-1997, 1956 (unpublished); A. Protopopov, G. Tolmachev, V. Ushatskii, R. Venediktova, I. Krisyuk, L. Rodionova, and G. Iakovleva, At. Energ. (USSR) **5**, 130 (1958); K. M. Broom, Phys. Rev. **126**, 627 (1962); and J. G. Cuninghame, J. Inorg. Nucl. Chem. **5**, 1 (1957).

[j] M. Thein, M. Rao, and P. Kuroda, J. Inorg. Nucl. Chem. **30**, 1145 (1968).

[k] J. G. Cuninghame, J. Inorg. Nucl. Chem. **5**, 1 (1957).

We will assume similar values for the range of fission fragments in thorium.

The thicknesses of our target foils varied from 150–300 mg/cm^2 of thorium and 240–900 mg/cm^2 of uranium. Due to the scatter in the data, it was difficult to detect any correlation of yield with target thickness. We have assumed that the recoil loss of fission products from Th^{232} and U^{238} fission is negligible. This assumption was unnecessary in the case of U^{235} fission, as 1-mil U^{235} foils were conveniently available, and were used as guard foils covering the U^{235} target foil.

The yields of the three products (Y^{93}, Ba^{130}, and Nd^{147}) located near the peak of the mass-yield curves were measured so that a comparison could be made between our results and measurements of others. An examination of Table II reveals that our results are generally consistent within experimental uncertainty with the earlier work.

Charge-Distribution Effects

The effects of nuclear charge distribution must be considered before assuming that measured fission yields of particular nuclides represent the total chain yields for those mass numbers. Independent formation of products further on in the isobaric decay chain can cause measured fission yields to be much less than the total mass yield. Because of the position of most long-lived fission products in their isobaric chains, this effect generally increases with increasing excitation energy. Unfortunately, not quite enough is known about charge distribution in fast-neutron fission to enable accurate estimates of independent fission yields to be made.

Coryell *et al.*[15] have proposed a method of predicting unmeasured independent yields by comparison with the rather well-studied case of thermal-neutron fission of U^{235}. We will use a modification[16] of their method to predict the most probable charge Z_p for the mass chains under study here:

$$\Delta Z_p = 0.5(Z_c-92) - 0.19(A_c-236) + 0.023(E^*-6.4), \quad \text{light fragment}$$

$$\Delta Z_p = 0.5(Z_c-92) - 0.19(A_c-236) + 0.047(E^*-6.4), \quad \text{heavy fragment}$$

where ΔZ_p is the change in Z_p for a particular mass number in going from thermal-neutron fission of U^{235} to the fission process in question, and Z_c, A_c, and E^* are the charge, mass, and excitation energy, respectively, of the compound nucleus. Wolfsberg[17] has also proposed an extension of the Coryell method.

For 14.8-MeV neutron fission, the above equations reduce to $\Delta Z_p(L)=0.12$, $\Delta Z_p(H)=0.20$ (Th^{232}); $\Delta Z_p(L)=0.34$, $\Delta Z_p(H)=0.69$ (U^{235}); and $\Delta Z_p(L)=0.11$, $\Delta Z_p(H)=0.24$ (U^{238}). The uncertainty in these values is unknown, but large. The values of ΔZ_p can be used with an assumed Gaussian charge dispersion curve[18,19] and measured or estimated values of Z_p for thermal-neutron fission of U^{235}, used as a reference state (Z_p^{ref}), to calculate the desired independent yields. One can readily see that ΔZ_p (and hence the correction to be made to the measured fission yields) is largest for the case of U^{235}.

Measured values of Z_p^{ref} have been reported[20] for only two of the mass chains that we are considering here, 93 and 140. In addition, the cumulative fractional chain yields of Kr^{93} and Xe^{140} have been reported[17] for 14.6-MeV neutron fission of U^{235} and U^{238}. For all other mass numbers we have resorted to some form of charge-distribution systematics. A set of calculated Z_p^{ref} values has been conveniently tabulated by Crouch.[21] We have used his values over the mass range 72 to 161 with the understanding that their validity is questionable for those products that we are actually interested in. Beyond this mass range even less is known about the Z_p^{ref} function and the shape of the charge dispersion curve. In the mass ranges 72 to 76 and 157 to 161 the average values of $Z_A - Z_p^{\mathrm{ref}}$ are about 3.6 and about 2.5, respectively. These values have been used to obtain estimates of Z_p^{ref} further out on the wings of the mass-yield curve. Values of Z_A, the most stable chage for a given mass number, were taken from the compilation of Hillman.[22]

The independent fractional-chain-yield calculations are summarized in Table III. It must be emphasized that they are only crude guesses. They are included merely to illustrate that, for example, the measured fission yields of Dy^{166} and Er^{172} may not be too close to the total chain yields. Actually, in the case of Dy^{166}, it will be shown in our discussion that these estimates may not be too far off.

Effect of Target Impurities

One of the most important sources of error in the measurement of such low-fission yields is the formation of the product nuclide by neutron-induced reactions on target impurities. The presence and amount of impurities can usually be determined by sensitive spectro-

---

[15] C. Coryell, M. Kaplan, and R. Fink, Can. J. Chem. **39**, 646 (1961).

[16] D. R. Nethaway and H. B. Levy, Phys. Rev. **139**, B1505 (1965).

[17] K. Wolfsberg, Phys. Rev. **137**, B929 (1965).

[18] We have used the Gaussian curve

$$y = \exp\left[-\frac{1}{2}\left(\frac{Z-Z_p}{\sigma}\right)\right] \Big/ \sigma(2\pi)^{1/2},$$

with $\sigma=0.59$. Values of this function have been conveniently tabulated by K. Wolfsberg, Los Alamos Scientific Laboratory Report No. LA-3169, 1965 (unpublished).

[19] A. Wahl, R. Ferguson, D. Nethaway, D. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).

[20] A. E. Norris and A. C. Wahl, Phys. Rev. **146**, 926 (1966).

[21] E. A. Crouch, United Kingdom Atomic Energy Authority Report No. AERE-R 5488, 1967 (unpublished).

[22] M. Hillman, Brookhaven National Laboratory Report No. BNL-846, 1964 (unpublished).

TABLE III. Summary of independent-yield calculations.

Mass number	Z_p^{ref}	Reference	Element	Calculated independent-fractional-chain yield Th^{232}	U^{235}	U^{238}
66	~26.1	a	Cu	10^{-5}	10^{-4}	10^{-5}
67	~26.4	a	Zn	10^{-8}	10^{-6}	10^{-8}
72	28.33	21	Ga	6×10^{-5}	1×10^{-3}	5×10^{-5}
93	37.29±0.04	20	Zr	4×10^{-5}	8×10^{-4}	4×10^{-5}
140	54.34±0.03	20	La	4×10^{-4}	7×10^{-3} [b]	6×10^{-4}
147	57.72	21	Pm	6×10^{-6}	2×10^{-4}	8×10^{-6}
153	60.15	21	Eu	1×10^{-4}	2×10^{-4}	2×10^{-4}
159	62.64	21	Tb	3×10^{-3}	2×10^{-2}	3×10^{-3}
161	63.47	21	Dy	1×10^{-3}	1×10^{-2}	1×10^{-3}
166	~65.2	a	Ho	3×10^{-2}	0.15	3×10^{-2}
169	~66.7	a	Tm	3×10^{-3}	3×10^{-2}	3×10^{-3}
172	~67.4	a	Tm	6×10^{-2}	0.24	6×10^{-2}

[a] Estimated from Z_A value; noting that, for very light fragments, $Z_A - Z_p$ is about 3.6, and for very heavy fragments, $Z_A - Z_p$ is about 2.5. These values lead to very crude guesses for the independent yields; they are listed here merely to illustrate that the measured fission yields of Dy^{166} and Er^{172} may not be too close to the total chain yields.

[b] Measured value from Ref. 16.

scopic analysis of the sample material or, in some cases, by measuring other fast-neutron reaction products that are not formed in fission. The latter method has the virtue that the analysis is performed on the actual piece of material on which the fission-yield measurements are being made, and in some cases is much more sensitive. A similar procedure has been used by Bramlitt and Fink,[23] for example.

The principal reactions on impurities that we must concern ourselves with are the following: $Zn^{70}(n, n\alpha)Ni^{66}$, $Zn^{67}(n, p)Cu^{67}$, and several reactions on Er, Tm, and Yb leading to Er^{169}. Other reactions can be neglected because of their small ratio of isotopic cross section to fission yield.

The presence of zinc impurity in the target material will lead to the formation of Cu^{64} by the (n, p) reaction and Zn^{65} by the $(n, 2n)$ reaction. Cu^{64} can also be made by reactions on copper impurity so that it alone does not necessarily measure the amount of zinc. We occasionally did find a small amount of a long-lived component in the zinc decay curves; however, attempts to show that it was Zn^{65} failed because of the low level. We did find Cu^{64} in all of the copper decay curves. Assuming that the (n, p) cross-section ratio[24] of Zn^{64} to Zn^{67} is 5, that all of the Cu^{64} came from zinc, and correcting for the zinc isotopic abundances, we can use the observed Cu^{64} to calculate the amount of Cu^{67} formed. For Th^{232} fission the correction to the fission yield was $\leq 7\%$, for U^{235} it was $\leq 1\%$, and for U^{238} it was $\leq 2\%$. Since it is not known how much of the Cu^{64} came from zinc impurity, we will not apply any correction to the Cu^{67} results, but simply note that the correction is probably small. The amount of Cu^{64} observed in the Th^{232} fission is consistent with either 50-ppm zinc impurity or 13-ppm copper impurity. Ni^{66} can also be formed from zinc, but the Zn^{70} abundance and the $(n, n\alpha)$ cross section[24] are both so small that the yield is negligible.

There are a number of impurity reactions that lead to the formation of Er^{169}. All we have done is note that their effect is probably negligible, except in the case of Th^{232} fission, based on a comparison of the fission yields of masses 166, 169, and 172. This will be shown more clearly in the next section. Fortunately, the products Dy^{66} and Er^{172} can only be formed from impurities by $(n, n\alpha)$ reactions, which have a very low cross section (<0.05 mb).[23]

DISCUSSION

We have attempted to correlate all of the data for products formed in low yields by fitting Gaussian curves to the wings of the mass-yield curves. This procedure admittedly fails to account for the peak yields and valley region, but does prove to be a reasonable method for intercomparing the yields of products on the two wings. Gaussian curves were fitted to the Th^{232}, U^{235}, and U^{238} data for products with yields less than about 1%, and are shown in Figs. 1–3. A least-squares procedure was used in which the data were weighted by the reciprocal of the square of their standard deviations.

The calculated Gaussian curves generally represent the experimental yield distribution fairly well. In the Th^{232} fission, the Er^{169} yield was not used in determining

[23] E. T. Bramlitt and R. W. Fink, Phys. Rev. **131**, 2649 (1963).

[24] M. Goldberg, S. Mughabghab, B. Magurno, and V. May, in, *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz (U.S. Government Printing Office, Washington 25, D.C., 1966), 2nd ed., Suppl. 2, Vol. 2A, Z=21 to 40.

the Gaussian curve. It is shown later that the measured yield may be high due to target impurities. Other measured yields that are not in satisfactory agreement with the calculated curves, but were used to determine the curves, include Eu^{156} from Th^{232} fission (high by 92%) and Er^{172} from U^{235} fission (high by 70%). For Th^{232} the average deviation of ten measurements from the curve is ±16%; for 12 measurements for U^{235} it is ±12%, and for 11 measurements for U^{238} it is ±12%.

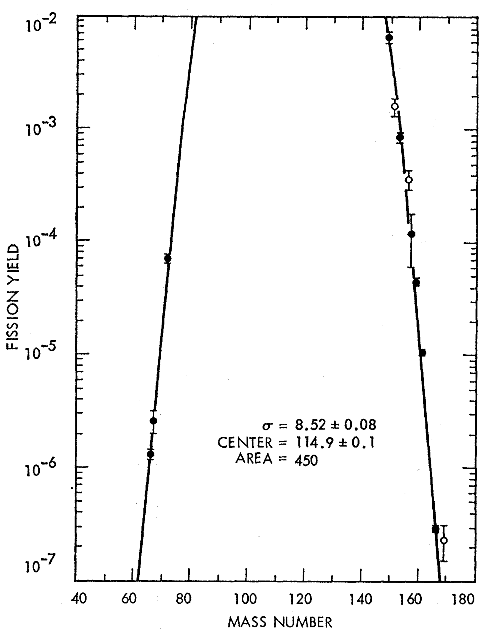

Fig. 1. Low-yield products from fission of Th^{232} with 14.8-MeV neutrons. The data point at mass 169 was not used for determination of the Gaussian curve.

The widths of the Gaussian curves for U^{235} and U^{238} are almost the same within experimental error (σ= 9.47±0.12 and 9.21±0.07), while that for Th^{232} is definitely smaller (σ=8.52±0.08). The calculated centers of the Gaussian curves are at 114.94±0.11 (Th^{232}), 116.24±0.11 (U^{235}), and 117.68±0.09 (U^{238}). The average number of neutrons emitted per fission can be calculated from the mass centers, and are 3.13±0.22 (Th^{232}), 3.52±0.23 (U^{235}), and 3.65±0.18 (U^{238}). These are significantly lower than the measured value of about 4.4 for 14-MeV neutron fission.[25]

These Gaussian curves provide a good method of estimating unmeasured yields on the wings of the mass-yield curves. For this purpose we have summarized in Table IV the calculated and measured yields for masses 64 to 78 and 158 to 172. This table includes values for the three target nuclides we have measured. Very few data exist for low-yield products from 14.8-MeV fission of other target nuclides, so that it is rather difficult to estimate the yields of such products. However, one could assume that they also follow a Gaussian distribution, that the area under the curve is about 115, and that the value of σ is about 9.3. The latter assumption should be reasonable for other easily fissionable targets such as U^{233} and Pu^{239}. The centers of the mass distributions can be estimated from the mass of the com-

[25] F. L. Fillmore, J. Nucl. Energy **22**, 79 (1968).

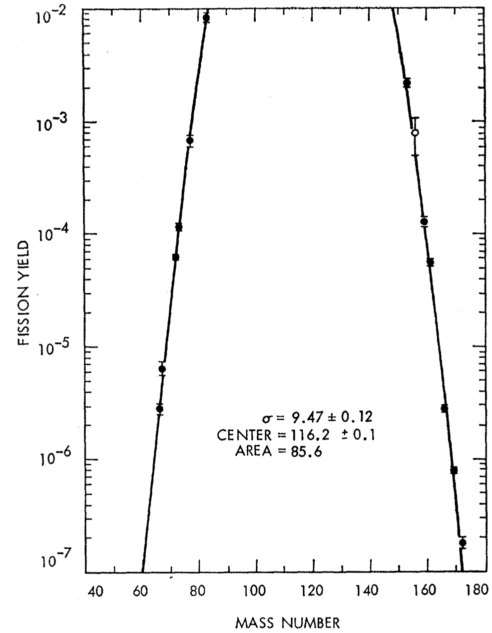

Fig. 2. Low-yield products from fission of U^{235} with 14.8-MeV neutrons.

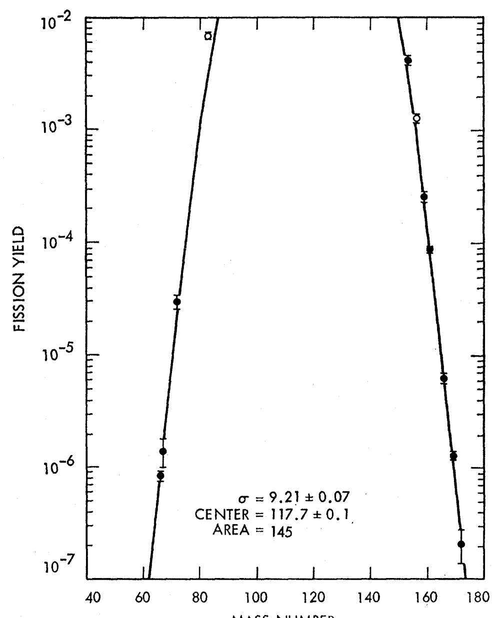

Fig. 3. Low-yield products from fission of U^{238} with 14.8-MeV neutrons.

TABLE IV. Calculated fission yields for 14.8-MeV fission of Th^{232}, U^{235}, and U^{238}.

	Th^{232}		U^{235}		U^{238}	
Mass	Calculated[a]	Measured[b]	Calculated[a]	Measured[b]	Calculated[a]	Measured[b]
64	3.6×10^{-7}		$9.\times10^{-7}$		2.6×10^{-7}	
65	$7.\times10^{-7}$		1.6×10^{-6}		$5.\times10^{-7}$	
66	1.4×10^{-6}	1.3×10^{-6}	2.8×10^{-6}	2.8×10^{-6}	$9.\times10^{-7}$	8.5×10^{-7}
67	2.8×10^{-6}	2.6×10^{-6}	$5.\times10^{-6}$	6.5×10^{-6}	1.7×10^{-6}	1.4×10^{-6}
68	$5.\times10^{-6}$		$9.\times10^{-6}$		3.0×10^{-6}	
69	1.0×10^{-5}		1.4×10^{-5}		$5.\times10^{-6}$	
70	1.9×10^{-5}		2.4×10^{-5}		$9.\times10^{-6}$	
71	3.5×10^{-5}		4×10^{-5}		1.6×10^{-5}	
72	$6.\times10^{-5}$	7.0×10^{-5}	7×10^{-5}	6.3×10^{-5}	2.8×10^{-5}	3.0×10^{-5}
73	1.1×10^{-4}		1.1×10^{-4}	1.2×10^{-4} [c]	4.8×10^{-5}	
74	2.0×10^{-4}		1.7×10^{-4}		$8.\times10^{-5}$	
75	3.5×10^{-4}		2.7×10^{-4}		1.4×10^{-4}	
76	6×10^{-4}		$4.\times10^{-4}$		2.2×10^{-4}	
77	1.0×10^{-3}		$7.\times10^{-4}$	6.8×10^{-4} [c]	3.6×10^{-4}	
78	1.7×10^{-3}		1.0×10^{-3}		5.8×10^{-4}	
158	$6.\times10^{-5}$		2.1×10^{-4}		4.3×10^{-4}	
159	3.2×10^{-5}	4.4×10^{-5}	1.3×10^{-4}	1.3×10^{-4}	2.6×10^{-4}	2.6×10^{-4}
160	1.7×10^{-5}		$8.\times10^{-5}$		1.6×10^{-4}	
161	$9.\times10^{-6}$	1.11×0^{-5}	$5.\times10^{-5}$	5.6×10^{-5}	1.0×10^{-5}	8.9×10^{-5}
162	$5.\times10^{-6}$		3.1×10^{-5}		$6.\times10^{-5}$	
163	2.5×10^{-6}		1.8×10^{-5}		3.4×10^{-5}	
164	1.3×10^{-6}		1.1×10^{-5}		2.0×10^{-5}	
165	7.3×0^{-7}		$6.\times10^{-6}$		1.1×10^{-5}	
166	3.3×10^{-7}	2.9×10^{-7}	3.6×10^{-6}	2.8×10^{-6}	$7.\times10^{-6}$	6.3×10^{-6}
167	1.6×10^{-7}		2.1×10^{-6}		3.7×10^{-6}	
168	$8.\times10^{-8}$		1.2×10^{-6}		2.0×10^{-6}	
169	3.7×10^{-8}	2.3×10^{-7}	$7.\times10^{-7}$	8.0×10^{-7}	1.1×10^{-6}	1.3×10^{-6}
170	1.7×10^{-8}		3.6×10^{-7}		$6.\times10^{-7}$	
171	$8.\times10^{-9}$		2.0×10^{-7}		3.3×10^{-7}	
172	3.7×10^{-9}		1.11×0^{-7}	1.8×10^{-7}	1.7×10^{-7}	2.1×10^{-7}

[a] Fission yields calculated from Gaussian curves discussed in the text (see Figs. 1–3).
[b] Measured values are those reported in this paper unless otherwise noted.
[c] Reference 2.

pound nucleus, with a neutron loss of about 3.6. We have taken the average of our results for U^{235} and U^{238}.

The Gaussian curves that we have obtained have been used to check on the importance of two sources of error in these measurements; charge-distribution effects and target impurities. In a previous section we noted that the measured yields of Dy^{166} and Er^{172} may not represent the total chain yields of masses 166 and 172, due to independent formation of Ho^{166} and Tm^{172}. It was shown that such an effect should be most important for these two products. An examination of Table IV reveals that the measured Dy^{166} yields are consistently lower than those given by the smooth curve. The difference is greatest for U^{235} (about 22% low), as expected. The effect is not as clear in the case of Er^{172}, due to the larger experimental errors.

In the section on target impurities, it was shown that Cu^{67} and Er^{169} were the only products measured that could also be made from target impurities in appreciable amounts. The measured yields of Cu^{67} and Er^{169} appear to be quite consistent with the Gaussian curves, except for the case of Er^{169} from Th^{232} fission. Here, the yield of Er^{169} is high by a factor of 6 from that obtained from the Gaussian curve. We feel that most of this discrepancy is due to a combination of Er, Tm, and Yb target impurities. Part of it could also be due to radioactive impurities in the low-counting Er samples.

These two sources of error operate in opposite directions, one tending to lower an observed yield, the other tending to raise it. An error in one measurement changes the Gaussian curve that we have fitted, using that value, thus obscuring errors in other measurements. Fortunately, the two sources of error are probably not important for any one product. The combined effect is then a general increase in scatter from the smooth Gaussian curve. As noted before, the average deviation from the curve is about 15%, which is not much greater than the average accuracy of the individual results.

SUMMARY

We have measured the fission yields of a number of products formed in low yield from 14.8-MeV neutron fission of Th^{232}, U^{235}, and U^{238}. The results show that, within experimental uncertainty, the wings of the mass-yield curves are consistent with Gaussian functions. These Gaussian curves allow interpolation and prediction of fission yields of unmeasured products, and a handy reference table has been included for this purpose. It was shown that the effect of target impurities on the measured fission yield is generally small and that the effect of nuclear charge dispersion on the mass yields is probably negligible, except for the cases of Ho^{166} and Er^{172}. The widths of the mass-yield curves for U^{235} and U^{238} are almost the same, while that of Th^{232} is significantly narrower.

ACKNOWLEDGMENTS

We would like to thank Roy Cedarlund and the operating crew of the Livermore ICT for their excellent cooperation in arranging and providing the 14-MeV neutron irradiations. We would also like to acknowledge the cooperation of Ruth Anderson, Ray Gunnink, and Jesse Meadows in performing many γ-ray pulse-height analyses and providing detector calibrations that we used for determination of absolute counting efficiencies. We also thank Lila Onstott for her help with the radioactivity measurements.

PHYSICAL REVIEW VOLUME 182, NUMBER 4 20 JUNE 1969

γ-γ Directional Correlations in the Decay of ^{192}Ir†

M. Y. KHAN,* L. D. WYLY, C. H. BRADEN, AND E. T. PATRONIS, JR.
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332

(Received 17 January 1969)

γ-γ directional correlations have been measured for the following cascades in the decay of 74-day ^{192}Ir: 468-417 keV, $A_2=-0.12\pm0.03$, $A_4=0.11\pm0.06$, $|\delta|$ (417 keV) >8; 588-612 keV, $A_2=0.09\pm0.03$; 604-316 keV, $A_2=-0.49\pm0.03$, δ(604 keV) ≈2; 308-612 keV, $A_2=-0.10\pm0.02$, δ(308 keV) $=-8\pm2$; 588-296 keV, $A_2=0.00\pm0.03$, δ(296 keV) >4 or <-20; 484-206 keV, $A_2=-0.28\pm0.03$, δ(484 keV) $=10(+10, -3)$; 374-206 keV, $A_2=0.10\pm0.03$. The results (1) support spin assignments of 3 and 4 for the 921- and 1200-keV levels, respectively, in ^{192}Pt, (2) are consistent with spin 4 for the 580-keV level in ^{192}Os, and (3) indicate spin 3 for the 690-keV level in ^{192}Os.

INTRODUCTION

THE nuclides ^{192}Pt and ^{192}Os lie in the transition region between nuclei with a spherical equilibrium shape and nuclei which show a well-defined series of rotational levels. The spins and parities of many levels and the multipolarities of many transitions have been assigned on the basis of γ-γ directional correlation experiments and conversion coefficient measurements.[1–7] In Fig. 1 the principal features of the level schemes are

† Supported in part by a grant from the National Science Foundation.

* U.S. A.I.D./Pakistan participant on leave from the University of Peshawar.

[1] L. L. Baggerly, P. Marmier, F. Boehm, and J. W. M. Dumond, Phys. Rev. **100**, 1364 (1955).
[2] W. H. Kelly and M. L. Wiedenbeck, Phys. Rev. **102**, 1130 (1956).
[3] V. Shiel, L. D. Wyly, and C. H. Braden, Phys. Rev. **105**, 1521 (1957).
[4] V. M. Kelman, R. Ya. Metaskhvarishvili, V. A. Romanov, and V. V. Tuchkevich, Nucl. Phys. **4**, 240 (1957).
[5] L. Simons, E. Spring, and G. Wendt, Nucl. Phys. **31**, 452 (1962).
[6] M. W. Johns and M. Kawamura, Nucl. Phys. **61**, 385 (1965).
[7] L. Schellenberg and J. Kern, Helv. Phys. Acta **39**, 420 (1966).