

## Fission of $U^{233}$ with 14.8-MeV Neutrons\*

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We have measured the yields of 25 products in the mass range from 66 to 175 from fission of  $U^{233}$  with 14.8-MeV neutrons. Each fission yield was measured in an absolute way, the number of atoms formed of each product being measured by absolute  $\beta$ - and  $\gamma$ -counting techniques. The number of fissions occurring was calculated from the target mass, the fission cross section, and the total neutron fluence. The neutron fluence in the target was monitored by the  $(n, 2n)$  reaction on yttrium foils. Total chain yields were estimated by correcting for the effects of nuclear charge dispersion in fission. The contribution of target-impurity activation to the measured fission yields is generally small.

The yields of products on the wings of the mass-yield distribution are consistent with a Gaussian function, which is similar to those found previously for  $U^{235}$  and  $U^{238}$  fission. The yields of a number of unmeasured products on the wings have been estimated and tabulated using the Gaussian curve.

The yields of a sufficient number of products in the peak and valley regions were measured so that a rough outline of the high-yield portion of the mass distribution could be made. The area under each half of the mass-yield curve was about 1.08, or 8% high. This discrepancy may be due to insufficient knowledge of the mass-yield curve, the contribution of fission induced by non-14.8-MeV neutrons, and/or errors in the cross sections that were used for the monitor reaction and the  $U^{233}$  fission reaction.

We observe a very low peak-to-valley ratio of about 3.1, which is smaller than found for fission of other nuclides with 14.8-MeV neutrons. Our yields in the valley region are about 70% higher than those measured previously by others. We feel that this is probably due to the contribution of low-energy neutron fission in the earlier work, coupled with their normalization to unit fragment yield. A thermal-neutron contaminant of 0.3% in the target region could have caused the observed discrepancy.

The fission yield of the shielded product  $Tb^{160}$  was measured as  $(3.2 \pm 0.8) \times 10^{-7}$ . Using the total chain yield for mass 160 calculated from our Gaussian distribution function, we obtain  $(4.3 \pm 1.2) \times 10^{-3}$  for the independent fractional chain yield. This value was used to estimate the  $Z_p$  function for  $U^{233}$  fission in order to calculate total chain yields from observed fission yields.

### INTRODUCTION

There are only two previous reports on the mass-yield distribution of products from fission of  $U^{233}$  with 14-MeV neutrons.<sup>1,2</sup> The distribution of high-yield products in the peak and valley regions has been fairly well defined and found to be similar to that for the fission of  $U^{235}$  with 14-MeV neutrons, except perhaps in the mass-95 to -105 region.<sup>2</sup> This earlier work was done with low 14-MeV neutron flux densities and small amounts of  $U^{233}$  target material. The resulting low counting rates limit the accuracy of the measurements, and preclude the measurement of products formed in low yield. The work by Bonyushkin *et al.*, was done by separating and purifying the individual products from the dissolved  $U^{233}$  ( $U_3O_8$ ) sample.<sup>1</sup> Borden and Kuroda used a recoil technique in which the recoiling fission products from a thin  $U^{233}$  source were caught on aluminum catcher foils, which were then dissolved for analysis.<sup>2</sup>

We have made use of the high-intensity source of

14-MeV neutrons available here at the insulated-core-transformer (ICT) accelerator to measure the yields of a number of products from 14.8-MeV fission of  $U^{233}$  in the mass range from 66 to 175. Our main interest is in the products on the wings of the mass-yield curve that are formed in very low yield, but we have also measured the yields of some products in the peak and valley regions to delineate further the over-all mass-yield curve. This work is an extension of some earlier measurements reported for the 14.8-MeV neutron fission of  $Th^{232}$ ,  $U^{235}$ , and  $U^{238}$ .<sup>3</sup> We have emphasized the measurement of the very low-yield products in the hope of developing a general method of estimating the yields of other such products that have not been measured.

### EXPERIMENTAL DETAILS

The experimental details are similar to those reported previously,<sup>3</sup> and will be described here only briefly. The 14.8-MeV neutron irradiations were performed here at the ICT neutron generator. The

neutrons are produced by the reaction of a beam of 400-keV deuterons striking a rotating titanium-tritide target.<sup>4</sup> The U<sup>233</sup> target was placed at 0° to the deuteron beam, where the neutron energy was 14.8 ± 0.3 MeV and the flux density about 6 × 10<sup>10</sup> cm<sup>-2</sup> sec<sup>-1</sup>. Continuous monitoring of the neutron flux density with a proton telescope counter allowed corrections to be made for the small variations in neutron yield.

The U<sup>233</sup> target assembly usually consisted of a 10-mil U<sup>233</sup> metal foil covered with 5-mil aluminum foils to catch recoil products. On both sides of the uranium-aluminum packet were 10-mil foils of yttrium metal to act as neutron fluence monitors. A 30-mil cadmium can was used to reduce the low-energy neutron background. The entire assembly was sealed in an aluminum can to prevent leakage of the radioactive material. The inside foils were all  $\frac{5}{8}$  in. in diameter.

The U<sup>233</sup> foils each weighed about 930 mg, and were notably clean and shiny. The isotopic assay was: U<sup>232</sup> 6 ppm, U<sup>233</sup> 97.5%, U<sup>234</sup> 1.1%, U<sup>235</sup> 250 ppm, U<sup>236</sup> 20 ppm, and U<sup>238</sup> 1.4%. A spectroscopic analysis (before conversion to the metal) showed

the presence of 2-ppm Cu, 10-ppm Fe, and very little else. The formation of activation products such as Zn<sup>65</sup> by the (*n*, 2*n*) reaction provides the most sensitive measure of the amount of certain impurities in the irradiated foil.

Nine separate irradiations were made varying in length from 2 to 12 h, and producing up to 3 × 10<sup>13</sup> fissions in the U<sup>233</sup> target. The target foil and aluminum guard foils were dissolved together after waiting 9 to 15 h for short-lived products to decay away. Two procedures were used for the initial separation of the desired products from the bulk of the uranium (and from the other α-active nuclides present). In one the target foils were dissolved in the presence of 10- to 20-mg amounts of carriers for each of the product elements. Each element was then separated from the bulk of the solution. In the second procedure small aliquots of the dissolved target solution were added to various carrier solutions for further processing.

Conventional radiochemical procedures were used for the purification of each of the product elements.<sup>5,6</sup> The individual rare-earth elements were separated on Dowex-50 ion-exchange columns us-

TABLE I. Radioactivity and counting-efficiency measurements.

Nuclide	Counter	Discriminator settings (keV)	Basis for counting efficiency	$E_\gamma(I_\gamma)^a$ ( $E_\gamma$ in keV)	Uncertainty in efficiency (%)
Ni <sup>66</sup>	β		β efficiency curve		10
Cu <sup>67</sup>	β		β curve and NaI PHA	184 (0.43)	10
Zn <sup>72</sup>	β		Ge PHA <sup>b</sup>	835 (0.96, Ga <sup>72</sup> )	3
Y <sup>88</sup>	γ	1600–1910	c	1836 (1.00)	3
Y <sup>93</sup>	β		β efficiency curve		10
Zr <sup>95</sup>	γ	600–900	Ge PHA	765 (0.99, Nb <sup>95</sup> )	4
Zr <sup>97</sup>	γ	600–900	Ge PHA	743 (0.94)	6
Mo <sup>99</sup>	γ	600–900	NaI and Ge PHA	739 (0.124)	9
Rh <sup>105</sup>	γ	200–400	NaI and Ge PHA	319 (0.19)	8
Ag <sup>111</sup>	γ	200–400	β efficiency curve		10
Pd <sup>112</sup>	γ	520–700	NaI and Ge PHA	617 (0.435, Ag <sup>112</sup> )	5
Cd <sup>115g</sup>	γ	50–580	NaI and Ge PHA	336 (0.50, In <sup>115m</sup> )	10
Cd <sup>115m</sup>	β		β efficiency curve		10
Te <sup>132</sup>	γ	400–1000	NaI and Ge PHA	668 (0.98, I <sup>132</sup> )	9
Ba <sup>140</sup>	γ	1100–1700	NaI and Ge PHA	1596 (0.96, La <sup>140</sup> )	4
Ce <sup>141</sup>	β		4π counter and Ge PHA	145 (0.49)	8
Ce <sup>143</sup>	γ	200–400	NaI and Ge PHA	293 (0.47)	5
Ce <sup>144</sup>	β		Ge PHA	134 (0.11)	10
Nd <sup>147</sup>	γ	470–650	NaI and Ge PHA	531 (0.132)	7
Sm <sup>153</sup>	β		β curve and NaI PHA	103 (0.28)	8
Gd <sup>159</sup>	β		β efficiency curve		10
Tb <sup>160</sup>	β		4π counter		5
Tb <sup>161</sup>	β		4π counter		5
Dy(Ho) <sup>166</sup>	β		4π counter		5
Er <sup>169</sup>	β		4π counter		5
Er(Tm) <sup>172</sup>	β		4π counter		5
Yb <sup>175</sup>	β		β efficiency curve		15

<sup>a</sup> Energy of γ ray (absolute intensity, photons per disintegration).

<sup>b</sup> By direct comparison with the 835-keV photon ( $I_\gamma=1.00$ ) in a standard sample (Ref. 9) of Mn<sup>54</sup>.

<sup>c</sup> By direct counting of a standard sample (Ref. 9) of Y<sup>88</sup>.

ing ammonium lactate as the eluant. Extra pre-column purification steps were necessary to remove all of the radioactive uranium daughter nuclides from the rare-earth fraction. The lowest-yield rare-earth nuclides ( $Tb^{160}$  and  $Yb^{175}$ ) were further purified by a second elution from an ion-exchange column.

The neutron fluence in the target was measured by the  $Y^{88}$  produced by the  $(n, 2n)$  reaction on the yttrium monitor foils.<sup>3</sup> We have used a cross section of  $1.02 \pm 0.05$  b for the  $(n, 2n)$  reaction at 14.8 MeV.<sup>7,8</sup> The threshold energy for this reaction is about 11.6 MeV.

The final samples were counted on gas-flow  $\beta$ -proportional counters or NaI(Tl)  $\gamma$  counters. The details of these measurements are given in Table I. The counting efficiencies are based on several methods: (1) comparison with a  $4\pi$  counter, (2) comparison with calibrated sodium iodide and ger-

manium detectors used with pulse-height analyzers (PHA), (3) the use of calibrated standard solutions,<sup>9</sup> and (4) the use of an experimentally determined curve of  $\beta$ -counting efficiency versus mean  $\beta$  energy. The counting efficiencies used here are the same as those used in Ref. 3, so that the results are directly comparable. Because counting rates were generally high except for the lowest-yield products ( $Tb^{160}$  and  $Yb^{175}$ ), the radioactive decay could be followed sufficiently long to verify the sample purity.

#### FISSION YIELD MEASUREMENTS

##### Results

The results of the  $U^{233}$  fission yield measurements are summarized in Table II. Each result is the average of about five separate determinations. The number of fissions in each irradiation was cal-

Table II. Yields of products from fission of  $U^{233}$  with 14.8-MeV neutrons.

Product nuclide	Half-life (days)	Measured fission yield <sup>a</sup>	Estimated total chain yield <sup>b</sup>	Previous measurement <sup>c</sup>
Ni <sup>66</sup>	2.29	$(7.7 \pm 0.8) \times 10^{-6}$		
Cu <sup>67</sup>	2.56	$(1.8 \pm 0.2) \times 10^{-5}$		
Zn <sup>72</sup>	2.45	$(1.46 \pm 0.06) \times 10^{-4}$	$1.49 \times 10^{-4}$	
Y <sup>93</sup>	0.427	$0.060 \pm 0.006$		
Zr <sup>95</sup>	65.0	$0.056 \pm 0.002$		
Zr <sup>97</sup>	0.701	$0.052 \pm 0.003$	0.054	
Mo <sup>99</sup>	2.75	$0.041 \pm 0.004$		$0.036 \pm 0.002$
Rh <sup>105</sup>	1.48	$0.022 \pm 0.002$		$0.016 \pm 0.002^d$
Ag <sup>111</sup>	7.4	$0.0185 \pm 0.0019$		$0.0121 \pm 0.0015$
Pd <sup>112</sup>	0.875	$0.0190 \pm 0.0011$	0.0193	$0.0108 \pm 0.0010$
Cd <sup>115g</sup>	2.21	$0.0170 \pm 0.0016^e$		$0.0103 \pm 0.0013^d$
Cd <sup>115m</sup>	43.			
Te <sup>132</sup>	3.24	$0.035 \pm 0.004$	0.039	$0.040 \pm 0.003^d$
Ba <sup>140</sup>	12.80	$0.043 \pm 0.002$	0.044	0.056
Ce <sup>141</sup>	32.6	$0.045 \pm 0.004$		$0.050 \pm 0.005^f$
Ce <sup>143</sup>	1.39	$0.036 \pm 0.002$		
Ce <sup>144</sup>	284.	$0.026 \pm 0.003$		
Nd <sup>147</sup>	11.04	$0.0129 \pm 0.0009$		
Sm <sup>153</sup>	1.94	$(1.56 \pm 0.13) \times 10^{-3}$		
Gd <sup>159</sup>	0.773	$(1.16 \pm 0.12) \times 10^{-4}$		
Tb <sup>160</sup>	72.1	$(3.2 \pm 0.8) \times 10^{-7}^g$		
Tb <sup>161</sup>	6.96	$(5.0 \pm 0.3) \times 10^{-5}$		
Dy <sup>166</sup>	3.40	$(2.6 \pm 0.3) \times 10^{-6}$	$2.65 \times 10^{-6}$	
Er <sup>169</sup>	9.5	$(9.1 \pm 0.6) \times 10^{-7}$		
Er <sup>172</sup>	2.08	$(1.95 \pm 0.15) \times 10^{-7}$	$2.1 \times 10^{-7}$	
Yb <sup>175</sup>	4.2	$(2.1 \pm 0.3) \times 10^{-8}$		

<sup>a</sup>The experimental standard deviations given here do not include a systematic 6% uncertainty in the calculation 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.

<sup>b</sup>Values of the estimated total chain yield are given where they differ appreciably from the measured fission yield.

<sup>c</sup>From Ref. 2, unless otherwise specified. Their reported errors probably do not include any systematic error in the  $Ba^{140}$  yield, which was used as a reference standard.

<sup>d</sup>These yields were measured as  $Ru^{105}$ ,  $Ag^{115}$ , and  $I^{132}$ , respectively.

<sup>e</sup>The measured  $Cd^{115m}$ -to- $Cd^{115g}$  ratio is 0.081.

<sup>f</sup>From Ref. 1.

<sup>g</sup>Independent fission yield.

culated from the product of the number of target atoms of each uranium isotope, the 14.8-MeV fission cross sections, and the 14.8-MeV neutron fluence. Corrections were made for the contribution of the  $U^{234}$ ,  $U^{235}$ , and  $U^{238}$  fission using measured or estimated fission yields for these isotopes. The  $U^{233}$  fission yields were then calculated as the total atoms produced of each product from  $U^{233}$  fission divided by the number of  $U^{233}$  fissions.

The integrated neutron fluence in the target foil was obtained by averaging the results from the yttrium monitor foils in front and back of the uranium. The front-to-back ratio was usually about 1.19 so that there was only a small error in the interpolation. The fission cross section used for  $U^{233}$  at 14.8 MeV is  $2.40 \pm 0.06$  b.<sup>10</sup> The total uncertainty in the number of fissions is about 6%. The errors assigned to the results in Table II include only the uncertainty in the counting efficiency, the counting statistics, and the agreement between replicate measurements. They do not include the 6% uncertainty in the number of fissions; this uncertainty value is a constant for all of the measurements.

#### Charge-Distribution Corrections

Column 4 of Table II gives the estimated total chain yields that are appreciably greater than the measured yields. In order to make these corrections it was necessary to estimate the independent fractional chain yield of the next higher- $Z$  element in each mass chain. We started with the recent set of  $Z_p$  values for thermal-neutron fission of  $U^{235}$  by Wahl, Rouse, and Williams.<sup>11</sup> This  $Z_p$  reference function was extrapolated to masses 66 and 175 by comparison of  $Z_A - Z_p$  values in neighboring mass regions. Values of  $Z_A$ , the most stable charge, were taken from the compilation of Hillman.<sup>12</sup> The extrapolation of the  $Z_p$  function is admittedly very uncertain. However, the corrections to the measured yields proved to be small in these regions, except for mass 172.

The estimation of the change in  $Z_p$  ( $\Delta Z_p$ ) for each mass number in the change from thermal-neutron fission of  $U^{235}$  to 14.8-MeV neutron fission of  $U^{233}$  is an important but uncertain step. A value for  $\Delta Z_p$  can be estimated by the method of Wolfsberg<sup>13</sup> to be 0.77. Until now there have not been any measurements of independent fission yields reported for 14.8-MeV fission of  $U^{233}$ . However, we have measured the independent fission yield of  $Tb^{160}$  (see Table II, and a later section of this paper), and a value of  $\Delta Z_p = 0.56 \pm 0.05$  has been calculated from this one measurement.

We have used an average value of  $\Delta Z_p = 0.6$  for all mass chains to calculate values of  $Z_p$  for 14.8-

MeV fission of  $U^{233}$ . These values were used together with a Gaussian charge-dispersion curve with  $\sigma = 0.56$ <sup>11</sup> to calculate the independent fractional chain yields of the next-higher- $Z$  element for each mass chain. The measured fission yields are close to the estimated total chain yields in each case, except for  $Te^{132}$  and  $Er^{172}$  where the corrections were about 10%. The uncertainty in the reference  $Z_p$  function and in the value of  $\Delta Z_p$  has a negligible effect in most cases.

#### Effect of Target Impurities

A rather important source of error in the measurement of very low fission yields is the formation of the product nuclide by 14-MeV neutron reactions on target impurities. It was shown in a previous paper<sup>3</sup> that the amounts of many trace impurities are indicated by the production of other nuclides not formed in fission. For example, the fission products  $Ni^{66}$  and  $Cu^{67}$  can be formed by fast-neutron reactions on zinc impurity in the uranium target. However, the amount of zinc present can be measured by the amount of  $Zn^{65}$  produced uniquely by the  $(n, 2n)$  reaction.

The principal reactions on impurities that we are concerned with are the following:  $Zn^{70}(n, n\alpha)Ni^{66}$ ,  $Zn^{67}(n, p)Cu^{67}$ ,  $Gd^{160}(n, 2n)Gd^{159}$ ,  $Dy^{160}(n, p)Tb^{160}$ , several reactions on Er, Tm, and Yb to form  $Er^{169}$ , and several on Yb, Lu, and Hf to form  $Yb^{175}$ . Small amounts of  $Ni^{57}$  and  $Cu^{64}$  were found in decay curves from a few of the irradiations, but there was no sign of any  $Zn^{65}$ . The  $Tm^{172}$  and  $Yb^{175}$  samples always had long-lived components in their decay, but they could not definitely be identified as belonging to  $Tm^{170}$ ,  $Tm^{171}$ , or  $Yb^{169}$ . In general, the  $U^{233}$  foils appear to have been sufficiently pure for the purposes of these measurements.

#### Independent Yield of $Tb^{160}$

As noted previously, no other independent yield measurements have been reported for 14-MeV neutron fission of  $U^{233}$ . In two of our experiments we made a special effort to obtain very pure terbium samples, in order that the very low independent yield of  $Tb^{160}$  could be measured. Double passes through cation-exchange columns were made to insure a complete separation from the likely contaminant  $Y^{91}$ . The independent fission yield is given in Table II as  $(3.2 \pm 0.8) \times 10^{-7}$ . The total chain yield for mass 160 is estimated in the next section to be  $7.5 \times 10^{-5}$ ; therefore, the independent fractional chain yield is  $(4.3 \pm 1.2) \times 10^{-3}$ .

#### DISCUSSION

##### Mass-Yield Curve

All of the experimental data for products with

fission yields greater than 0.1% are plotted as a mass-yield curve in Fig. 1. A smooth curve has been drawn through our data points and their reflected points. It was assumed that the average mass number of the fissioning nucleus was 229.7. It is obvious that we are in serious disagreement with the previous work, especially in the valley region. This matter will be discussed later.

The total yield under each half of our mass-yield curve is about 1.08, whereas it should be 1.00. This discrepancy may arise from three possible causes. We have measured the yields of a very limited number of high-yield products – 28 (including reflected points) out of a total of 66 with over 1% fission yield. We may have missed some fine-structure in the curve, and overestimated the yields.

The second possibility is that our method of monitoring the total neutron fluence may have caused us to underestimate the number of fissions. The monitor reaction  $Y^{89}(n,2n)Y^{88}$  has an excitation function that is different from that for the  $U^{233}$  fission reaction. Scattered or other low-energy neutrons can cause fission, but not the  $(n,2n)$  reaction

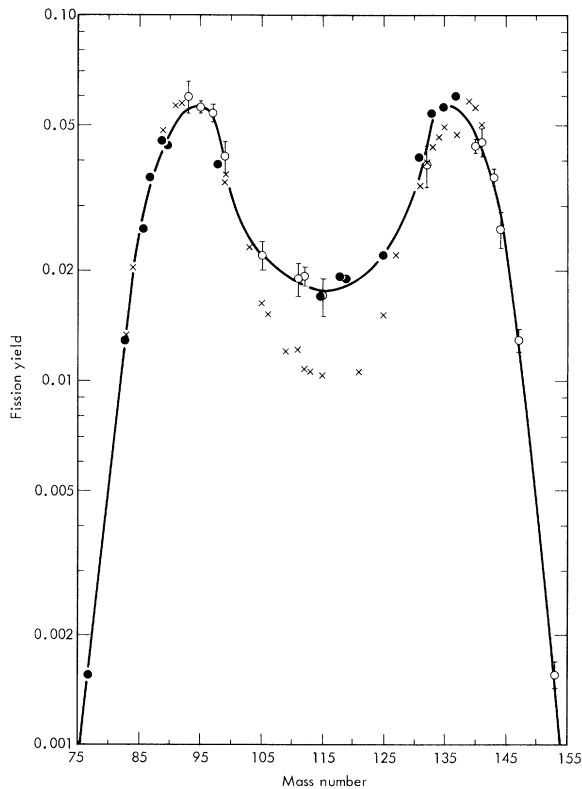


FIG. 1. Mass-yield curve for fission of  $U^{233}$  with 14.8-MeV neutrons. Results from this work are indicated by open circles with error bars; mirror points are closed circles. Results from Refs. 1 and 2 are indicated by crosses, with no mirror points indicated.

on yttrium. The  $U^{233}$  foils were covered with 30 mil of Cd to reduce the amount of thermal-neutron fission.

The third possibility is that the cross sections that we have used for the yttrium  $(n,2n)$  reaction and the  $U^{233}$  fission reaction are in error. We indicated that the total uncertainty in the cross sections was about 6%. The 8% discrepancy in the area under our mass-yield curve may be due to any combination of these three factors. In particular, an arbitrary 8% reduction of all of our measured yields is not at all appropriate at this time. If fission induced by lower-energy neutrons is the cause of the 8% error in area, then high-yield products will be affected much more than the low-yield products because of their steeply rising excitation functions. The uncertainty in drawing the mass-yield curve must also be considered.

Even if our yields were to be reduced by 8%, they would still be 60% higher in the valley region (mass 110 to 120) than those reported previously.<sup>1,2</sup> The best explanation of this difference is that there was a significant amount of low-energy fission that perturbed the results of the earlier work. Low-energy fission is characterized by a high peak-to-valley ratio. This high ratio would tend to depress

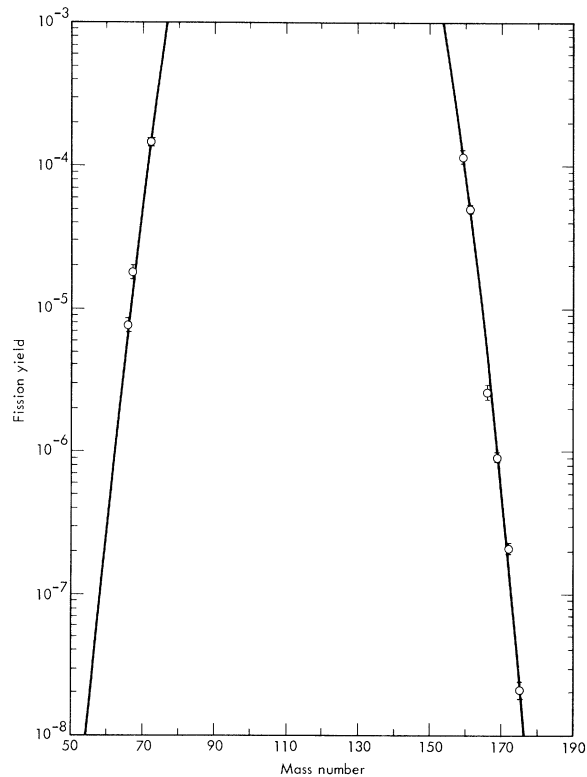


FIG. 2. Low-yield products from fission of  $U^{233}$  with 14.8-MeV neutrons. All results are from this paper. The Gaussian curve is described in Table III.

TABLE III. Gaussian fits to wings of mass-yield curves for 14.8-MeV neutron fission of  $\text{Th}^{232}$ ,  $\text{U}^{233}$ ,  $\text{U}^{235}$ , and  $\text{U}^{238}$ . Results for  $\text{Th}^{232}$ ,  $\text{U}^{235}$ , and  $\text{U}^{238}$  are from Ref. 3. The Gaussian curve used is

$$\text{yield} = \text{area} \times \exp\left\{-\frac{1}{2}[(A - A_0)/\sigma]^2\right\} / \sigma(2\pi)^{1/2}.$$

Target nuclide	Center mass number	Area	$\sigma$
$\text{Th}^{232}$	114.9 $\pm$ 0.1	450 $\pm$ 130	8.52 $\pm$ 0.08
$\text{U}^{233}$	115.2 $\pm$ 0.1	55 $\pm$ 16	9.86 $\pm$ 0.13
$\text{U}^{235}$	116.2 $\pm$ 0.1	86 $\pm$ 24	9.47 $\pm$ 0.12
$\text{U}^{238}$	117.7 $\pm$ 0.1	145 $\pm$ 25	9.21 $\pm$ 0.07

the valley yields when the over-all mass-yield curve is normalized to 200%. No mention was made in either Refs. 1 or 2 of any effort to reduce the intensity of low-energy neutrons in the  $\text{U}^{233}$  targets. Since the thermal-neutron fission cross section of  $\text{U}^{233}$  is 528 b, a neutron spectrum with 0.3% thermal neutrons would cause the valley yields to be reduced by 60%. It should be clear that it is very important to reduce the amount of low-energy fission as much as possible when measuring high-yield products from 14-MeV neutron fission.

#### Low-Yield Products

In Fig. 2 we have plotted the yields of the products that are formed in very low yield against the

mass number. The upper portion of the mass-yield curve has been omitted for clarity. A Gaussian curve has been fitted to the wings of the mass-yield curve and is also shown in Fig. 2. A least-squares procedure was used in which the data were weighted by the reciprocal of the square of their standard deviations. The Gaussian curve fits the  $\text{U}^{233}$  data rather well; the average deviation of the 10 points from the calculated curve is  $\pm 15\%$ . This same procedure was used for the products from fission of  $\text{Th}^{232}$ ,  $\text{U}^{235}$ , and  $\text{U}^{238}$  in Ref. 3. The parameters of all four Gaussian curves are summarized in Table III. In general, there is not too much one can say about this comparison, other than that the curve for  $\text{Th}^{232}$  is significantly narrower than for the uranium isotopes.

The Gaussian curve that has been fitted to the experimental data for  $\text{U}^{233}$  fission provides a good method for estimating the yields of unmeasured products on the wings of the yield distribution. In Table IV we list the calculated yields for products with masses 56 to 80 and 152 to 176. The measured yields are also given for comparison.

#### Comparison of 14.8-MeV Fission of $\text{U}^{233}$ , $\text{U}^{235}$ , and $\text{U}^{238}$

The mass distribution in fission has now been studied in detail for three isotopes of uranium. In

TABLE IV. Total chain yields for fission of  $\text{U}^{233}$  with 14.8-MeV neutrons. Calculated yield is from Gaussian curve described in Table III. Measured values are from this paper.

Mass	Calculated yield	Measured yield	Mass	Calculated yield	Measured yield
56	$3.3 \times 10^{-8}$		152	$2.1 \times 10^{-3}$	
57	$5.9 \times 10^{-8}$		153	$1.5 \times 10^{-3}$	$(1.56 \pm 0.13) \times 10^{-3}$
58	$1.1 \times 10^{-7}$		154	$9.8 \times 10^{-4}$	
59	$1.9 \times 10^{-7}$		155	$6.6 \times 10^{-4}$	
60	$3.4 \times 10^{-7}$		156	$4.3 \times 10^{-4}$	
61	$6.0 \times 10^{-7}$		157	$2.8 \times 10^{-4}$	
62	$1.0 \times 10^{-6}$		158	$1.8 \times 10^{-4}$	
63	$1.8 \times 10^{-6}$		159	$1.2 \times 10^{-4}$	$(1.16 \pm 0.12) \times 10^{-4}$
64	$3.1 \times 10^{-6}$		160	$7.5 \times 10^{-5}$	
65	$5.1 \times 10^{-6}$		161	$4.7 \times 10^{-5}$	$(5.0 \pm 0.3) \times 10^{-5}$
66	$8.6 \times 10^{-6}$	$(7.7 \pm 0.8) \times 10^{-6}$	162	$2.9 \times 10^{-5}$	
67	$1.4 \times 10^{-5}$	$(1.8 \pm 0.2) \times 10^{-5}$	163	$1.8 \times 10^{-5}$	
68	$2.3 \times 10^{-5}$		164	$1.1 \times 10^{-5}$	
69	$3.7 \times 10^{-5}$		165	$6.6 \times 10^{-6}$	
70	$6.0 \times 10^{-5}$		166	$3.9 \times 10^{-6}$	$(2.65 \pm 0.3) \times 10^{-6}$
71	$9.5 \times 10^{-5}$		167	$2.3 \times 10^{-6}$	
72	$1.5 \times 10^{-4}$	$(1.49 \pm 0.06) \times 10^{-4}$	168	$1.4 \times 10^{-6}$	
73	$2.3 \times 10^{-4}$		169	$7.8 \times 10^{-7}$	$(9.1 \pm 0.6) \times 10^{-7}$
74	$3.5 \times 10^{-4}$		170	$4.5 \times 10^{-7}$	
75	$5.4 \times 10^{-4}$		171	$2.5 \times 10^{-7}$	
76	$8.1 \times 10^{-4}$		172	$1.4 \times 10^{-8}$	$(2.1 \pm 0.2) \times 10^{-7}$
77	$1.2 \times 10^{-3}$		173	$7.9 \times 10^{-8}$	
78	$1.8 \times 10^{-3}$		174	$4.3 \times 10^{-8}$	
79	$2.6 \times 10^{-3}$		175	$2.4 \times 10^{-8}$	$(2.1 \pm 0.3) \times 10^{-8}$
80	$3.7 \times 10^{-3}$		176	$1.3 \times 10^{-8}$	

Figs. 3–5 we present a detailed comparison of how the yields of several products vary with the mass number of the uranium target isotope. The yields have all been measured with the same counting efficiencies. The only systematic differences are due to errors in the fission cross sections or to the assumption of a pure 14.8-MeV neutron source. Figure 3 shows the yield variation for three products on the low-mass wing; Fig. 4, the yield variation for four high-yield products; and Fig. 5, the yield variation for four products on the high-mass wing. As might be expected, all of the plots exhibit a smooth variation with uranium mass number. It is difficult to say on the basis of these measurements alone whether or not there is an odd-even effect on the yields. The products on the low-mass wing have a much larger variation than the others do.

#### SUMMARY

We have measured the yields of 25 products from fission of  $U^{233}$  with 14.8-MeV neutrons. The yields of products on the wings of the mass-yield curve have been fitted by a Gaussian function. By

means of this function, the yields of a number of unmeasured low-yield products have been calculated and tabulated. The Gaussian curve is somewhat similar to those found previously for  $U^{235}$  and  $U^{238}$  fission.

We find that the peak-to-valley ratio is unusually small ( $\sim 3.1$ ). The yields of products in the valley are higher than those found for 14.8-MeV neutron-induced fission of other nuclides. For example, the peak-to-valley ratio is  $\sim 6.0$  for 14-MeV fission of  $U^{235}$ . We are also in rather serious disagreement with earlier measurements by others of yields of valley products. Our valley yields are about 70% higher. We feel that this is probably due to the effect of low-energy fission in the earlier work. A small fraction of thermal neutrons in the 14-MeV neutron spectrum can easily cause the observed discrepancy in yield.

A comparison of the yields of a number of products from 14.8-MeV fission of three isotopes of uranium shows a general smooth variation of yield with uranium mass number. The products on the low-mass wing of the fission-yield curve show the largest variation.

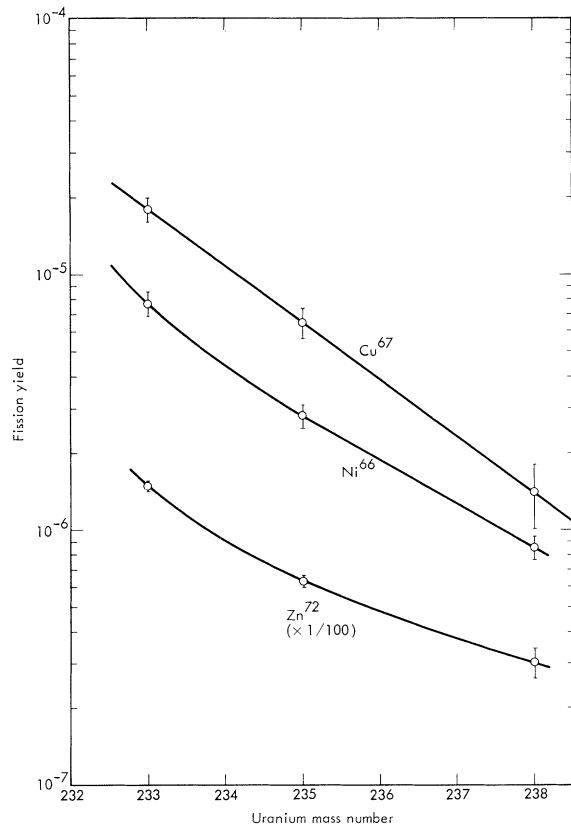


FIG. 3. Variation of fission yield with uranium mass number. Low-yield products of light fragment.

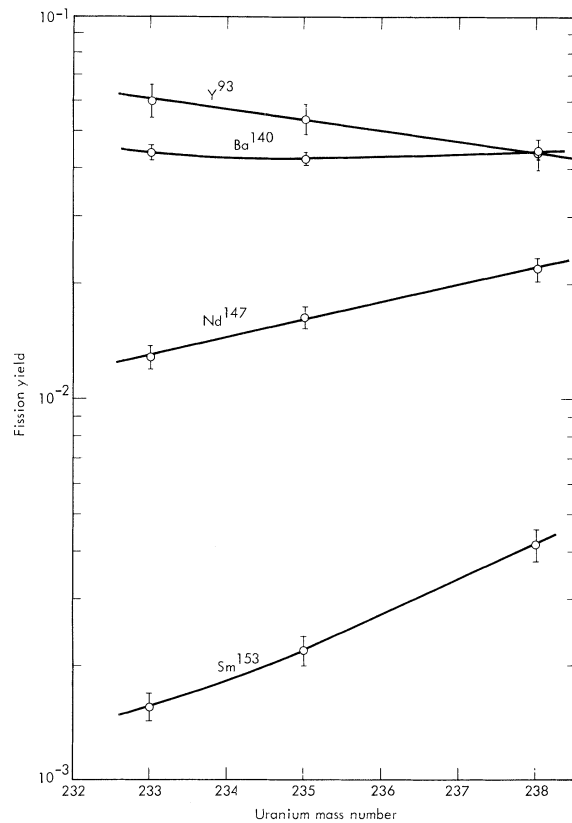


FIG. 4. Variation of fission yield with uranium mass number. High-yield products.

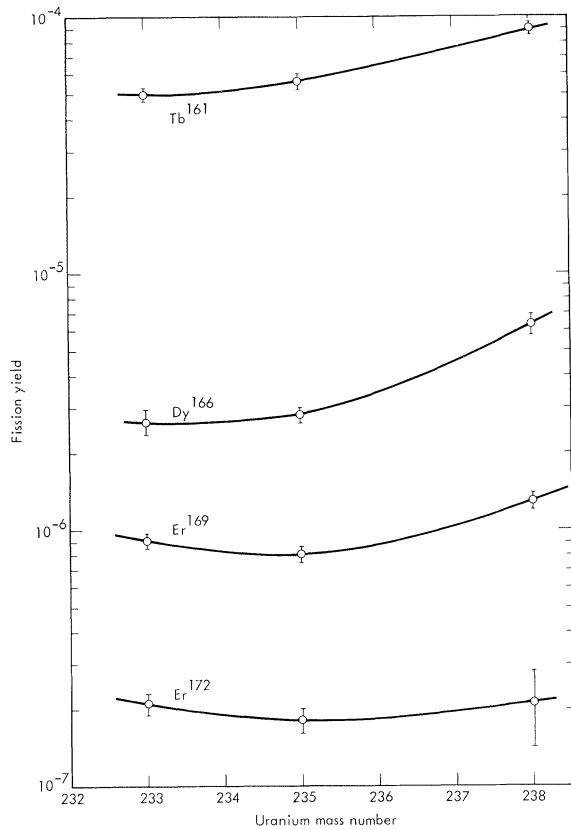


FIG. 5. Variation of fission yield with uranium mass number. Low-yield products of heavy fragment.

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<sup>1</sup>E. K. Bonyushkin *et al.*, *At. Energ. USSR* **10**, 13 (1961).

<sup>2</sup>K. D. Borden and P. K. Kuroda, *J. Inorg. Nucl. Chem.* **31**, 2623 (1969).

<sup>3</sup>D. R. Nethaway, B. Mendoza, and T. E. Voss, *Phys. Rev.* **182**, 1251 (1969).

<sup>4</sup>R. Booth, Lawrence Radiation Laboratory Report No. UCRL-70183, 1967 (unpublished).

<sup>5</sup>M. Lindner, Lawrence Radiation Laboratory Report No. UCRL-14258, 1965 (unpublished).

<sup>6</sup>National Academy of Sciences - National Research Council, Nuclear Science Series on the radiochemistry of the various elements.

<sup>7</sup>R. Rieder and H. Munzer, *Acta Phys. Australia* **23**, 42 (1966).

<sup>8</sup>D. G. Vallis, Atomic Weapons Research Establishment Report No. AWRE-O-76/66, 1966 (unpublished).

<sup>9</sup>The calibrated radioactive solutions were obtained from the International Atomic Energy Agency, Vienna, Austria.

<sup>10</sup>W. Hart, United Kingdom Atomic Energy Authority Report No. AHSB(S)R124, 1967 (unpublished).

<sup>11</sup>A new table of values of  $Z_p$  (the most probable charge) for each mass number in the range 72 to 161 is given by A. C. Wahl, R. A. Rouse, and J. C. Williams, in *Proceedings of the Second International Atomic Energy Symposium on Physics and Chemistry of Fission, Vienna, Austria, 1969* (International Atomic Energy Agency, Vienna, Austria, 1969), Paper No. SM-122/116.

<sup>12</sup>M. Hillman, Brookhaven National Laboratory Report No. BNL-846, 1964 (unpublished).

<sup>13</sup>K. Wolfsberg, Los Alamos Scientific Laboratory Report No. LA-3169, 1965 (unpublished).