# Fission yields in the symmetric region: Yields of the stable isotopes of tin and cadmium in the thermal neutron fission of <sup>235</sup>U and <sup>239</sup>Pu

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# (Received 12 May 1976)

The relative isotopic abundances of five isotopes of cadmium (111, 112, 113, 114, and 116) and eight isotopes of tin (117, 118, 119, 120, 121, 122, 124, and 126) have been measured in the thermal neutron fission of  $^{235}$ U and  $^{239}$ Pu. Samples of  $^{235}$ U and  $^{239}$ Pu were irradiated in a reactor; the elements were chemically separated and the isotopic abundances were mass spectrometrically determined. By suitably normalizing these relative yields, it has been possible to obtain the cumulative chain yields at 12 masses in the symmetric region of the mass distribution. These results indicate that there is a fine structure in the 113–115 mass region, especially in the fission of  $^{235}$ U. However, the mass distribution appears to be smooth between masses 116 and 126. The depression in the mass-yield curve around mass 115 may be related to the break around this mass number in the prompt neutron distribution as a function of mass.

 $\begin{bmatrix} \text{NUCLEAR REACTIONS, FISSION} & 235 \text{U}(n, f) \text{ and } 239 \text{Pu}(n, f) \text{ measured abundances} \\ & \text{of tin and cadmium isotopes.} \end{bmatrix}$ 

## INTRODUCTION

Fine structure in the mass distribution in fission has been a subject of considerable interest ever since its discovery.<sup>1</sup> Practically all the fine structure observed has been in the higher yield regions of mass numbers 133-145 and  $90-100.^{2-8}$  These studies have led to the conclusion that fine structure in mass distributions arises mainly as a result of the variations in the distribution of prompt neutrons as a function of fragment mass,<sup>9</sup> though there is some structure in the prompt mass distribution as well.

An important region of interest in the study of fine structure is the valley region in fission. The discontinuities in the neutron yield distribution appear in this region and this may indeed lead to some fine structure. In addition there have been suggestions of a third peak in the symmetric region in the fission of some heavy elements.<sup>10,11</sup> Calculations of Newson<sup>12</sup> and Thind and Tomlinson<sup>13</sup> also indicate dips around mass 127.

Recent complications of fission yield data<sup>14,15</sup> show that there are very few reliable measurements in the valley region and these are inadequate to define any fine structure in this region. A few radiochemical measurements have been carried out on certain individual fission products, the nuclides measured being some of the isotopes of palladium, silver, cadmium, and tin. Not only are the data limited to a few nuclides, but also the radiochemical yields given are unlikely to be very accurate because of the known difficulties arising out of the existence of isomers, nonavailability of accurate decay schemes, and the low fission yields in this region.

Since the isotopes of tin and cadmium cover most of the valley region in the mass distribution, a mass spectrometric measurement of the relative abundances of the isotopes of these elements produced in fission appears to be attractive from the point of view of yield measurement. However, these elements, especially tin, are difficult to measure on the mass spectrometer because of their poor sensitivity in the mass spectrometric measurements. The amounts of these elements available from permissible levels of irradiation are only in the nanogram range. Further, at these levels of sample handling, the likelihood of natural contamination is very high. Thus the mass spectrometric measurement of the relative fission yields of tin and cadmium isotopes calls for very "clean" separation techniques to extract nanogram quantities of these elements from highly radioactive fission product mixtures and very sensitive mass spectrometric techniques to accomplish the measurement. Only de Laeter and Thode<sup>16</sup> have succeeded in measuring fission product tin by mass spectrometry and their data are confined to the fission of <sup>233</sup>U. More recently they have measured the relative vields of the stable isotopes of cadmium in the fission of <sup>233</sup>U and 235U.17

In the present work we have measured the relative yields of the isotopes of both tin and cadmium in the thermal neutron fission of  $^{235}$ U and  $^{239}$ Pu. This covers the entire region from mass 111 to mass 126.

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## EXPERIMENTAL

# **Reagen ts**

All chemical reagents used in these experiments were specially purified to remove even traces of natural contamination. Distilled water was deionized twice by passing it through mixed bed ion-exchange columns. "Aristar" nitric acid and hydrochloric acid were further purified by ion exchange and stored in polyethylene bottles for use. The glass, polyethylene, polycarbonate, and Teflon ware used were cleaned by repeatedly rinsing them with purified nitric acid and water. The anion-exchange resin used throughout the experiment was Biorad AG  $1 \times 8$ .

#### Sample preparation and irradiation

The uranium samples  $(93\%^{235}\text{U})$  were purified by making a solution in HCl, adsorbing on a small anion-exchange column in 6 *M* HCl, washing with 6 *M* HCl, and then eluting out with a minimum quantity of 1 *M* HCl. The purified solution was evaporated into quartz irradiation vials, flamed to convert uranium to the oxide form, and sealed in vacuum. Each sample vial contained 10 mg of <sup>235</sup>U. These sample vials were irradiated in the McMaster University reactor for a total of 3584 MWh during the period April–September 1972.

Plutonium was also purified by anion exchange. Pu(IV) was adsorbed on a small anion-exchange column in 7 M HCl. After washing with 7 M HCl, the plutonium was eluted with 7 M HCl containing 0.1 M HI. The hydriodic acid used was freshly purified by anion exchange. The plutonium was taken through one more cycle of purification and the product was evaporated into quartz vials, flamed, and sealed in vacuum. Each of the vials containing approximately 8 mg of plutonium was irradiated for about 3500 MWh in the McMaster University swimming pool reactor during the period June-November 1972.

#### Extraction of fission product tin and cadmium

The fission product tin and cadmium were separated from the irradiated samples after a cooling period of 4-6 months, using an anion-exchange separation method which was standardized earlier in inactive experiments. The irradiated uranium oxide was dissolved in HCl containing a trace of HF. The solution was cleared with two drops of HNO<sub>3</sub> to ensure the oxidation of all uranium to U(VI). This solution was evaporated to dryness, taken up in 6 *M* HCl and loaded on 1 ml of anionexchange resin in a 9.5 mm diam column which was preconditioned with 6 *M* HCl. After washing with 6 ml of 6 *M* HCl, the uranium was eluted out with about 10 ml of 1 M HCl. The tin fraction was eluted out with 10–15 ml of the eluant 1 MHNO<sub>3</sub> + 0.1 M HCl. Some of the cadmium also came with this fraction. The remaining cadmium was eluted with very dilute HNO<sub>3</sub>. The tin fraction was further purified by repeating the above procedure once again.

The plutonium oxide was dissolved in 7 M HCl. A few grains of NaNO<sub>2</sub> were added to adjust the oxidation state of plutonium to Pu(IV). The plutonium was adsorbed in about 1 ml of anion-exchange resin. After washing with 7 M HCl the plutonium was eluted with 7 M HCl +0.1M HI. The column was further washed with 3 M HCl and the tin fraction was eluted out using 1 M HNO<sub>3</sub> containing 0.1 M HCl. The cadmium fraction was eluted out with very dilute HNO<sub>3</sub>. The tin fraction was further purified by repeating the above procedure.

In order to minimize the contamination from naturally occurring isotopes, the purification and extraction procedures were carried out in a dry box set up in a fume hood. A positive pressure was maintained in the dry box by feeding in air through absolute filters and the high rate of exhaust in the fume hood ensured that the radioactive air coming out from the dry box did not diffuse to the operating side. In addition, adequate shielding was provided for hot samples. All these precautions made the chemical processing rather slow.

## Mass spectrometry

The isotopic analyses were carried out with a 38 cm radius tandem mass spectrometer described by McMullen, Fritze, and Tomlinson.<sup>18</sup> The mass spectrum was obtained by varying the accelerating voltage. A voltage stepping system with 10 positions made possible the automatic measurement of all tin isotopes and an electronic readout system was used to print out the intensities at each peak position in the digital form. A number of scans could thus be automatically recorded in quick succession.

As the sensitivity in the measurement of tin was a critical parameter, two different approaches were made in order to improve the sensitivity. An attempt was made to adapt the silica gel method used by Cameron, Smith, and Walker<sup>19</sup> for the measurement of nanogram quantities of lead samples. About 1 mg of moist silica gel was placed on the filament of a single filament ion source and the tin sample in about 1  $\mu$ 1 of 0.75 *M* H<sub>3</sub>PO<sub>4</sub> was evaporated onto this bead. Ions were produced by heating the filament. This method proved promising in the beginning, but the presence of what appear to be huge molecular ion beams at

An attempt was also made to improve the sensitivity of the modified electron impact ion source used by de Laeter and Thode.<sup>16</sup> In this ion source a small ionization chamber was fitted on a triple filament button so that it completely surrounded a sample filament made of rhenium which was located in the center. A thoriated tungsten filament isolated electrically from the ionization chamber and sample filament, was used as the electron emitter. The electrons were accelerated towards the ionization chamber under a potential of approximately 30 V and, after passing through a collimator slit, they ionized sample atoms evaporated off the sample filament. Two sample magnets, rigidly mounted paraxially with the electron beam, enabled the electrons to spiral across the sample filament with a resultant increase in ionization efficiency. On heating the sample filament using a highly stable low voltage power supply, atoms of the sample were evaporated at a carefully controlled rate. They were then ionized by the electron beam and accelerated from the source through a potential of 10 kV.

Since phosphoric acid was found to enhance the sensitivity in the silica gel method, it was also used in conjunction with the modified electron impact ion source. The sample (about 25 ng of tin) was loaded onto a small tantalum sinter on the sample filament along with about a  $\mu$ l of 0.75 M  $H_3PO_4$ . This method greatly increases the sensitivity of the analysis. Steady beams of 100 mV or more could be obtained from about 25 ng samples at a sample filament current of 0.7 to 0.8 A. However, there was a molecular ion peak persistently appearing at mass 124. In some cases this could be minimized by selective heating, but could never be totally eliminated. Therefore, while analyzing fission product tin, some samples were loaded with phosphoric acid and others without it. This was to ensure that <sup>124</sup>Sn was also measured.

The mass spectrometer was operated at a resolution of about 1000, thus making it possible to resolve clearly metal peaks from hydrocarbons. The hydrocarbon peaks could be minimized by reducing the electron accelerating voltage, by frequently baking the instrument, and by exercising care in the loading of new samples. However, at some mass numbers, especially mass 110, the hydrocarbon peak had to be specially watched to ensure that it did not contribute to the metal peak.

As the scanning of the mass spectrum by varying the accelerating voltage introduced mass discrimination in the measured isotopic abundances, it was necessary to correct for this. By repeated analysis of natural tin samples, a correction factor was established and this factor (1.5% per mass unit) was used for correcting the measured abundances.

The cadmium ratios were similarly measured at a current of 0.4 to 0.5 A through the sample filament. The mass discrimination factor for the tin isotopes was assumed for cadmium isotopes also.

## RESULTS

#### Cadmium

The relative yields of cadmium isotopes from the fission of <sup>235</sup>U and <sup>239</sup>Pu are given in Tables I(a) and I(b), where results from different samples are reported. Only in the case of one sample [U(III)] was there some tin admixture at mass 116 and hence this value is not reported in Table I(a). The values are quoted after correcting for natural cadmium contamination using the abundance at mass 110 which amounted to only 5-20% of the abundance at mass 111, except in sample Pu(I)which showed about 30% contamination. In Table I the relative abundances at masses 113 and 114 are not separately given because the individual abundances do not represent fission yields on account of the large absorption cross section of <sup>113</sup>Cd. Hence only the sum of the abundances is given.

The estimated error, which is in general larger than the standard deviation for the set of measurements, takes into account the error introduced by correction for natural contamination.

#### Tin

The data on the relative abundances of tin isotopes from the fission of  $^{235}$ U and  $^{239}$ Pu are given in Tables II(a) and II(b). Tin was measured in sample No. U(III) after loading the sample in phos-

TABLE I.	Relative	abundances	of	cadmium	isotopes.
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Sample No.	111	112	113 + 114	116		
(a) In the fission of $^{235}\mathrm{U}$						
U(11)	1.000	0.900	1.465	0.865		
U(III)	1.000	0.965	1.560	•••		
Π(IΛ)	1.000	0.882	1.467	0.847		
Mean	1.000	0.92	1.49	0.86		
Estimated error		0.05	0.05	0.02		
(b) In the fission of <sup>239</sup> Pu						
Pu (l)	1.000	0.466	0.477	0.187		
Pu (II)	1.000	0.472	0.476	0.159		
Mean	1.000	0.47	0.48	0.17		
Estimated erro	$\pm 0.01$	$\pm 0.01$	$\pm 0.01$			

Sample No.	126	124	122	121	120	119	118	117
			(a) In <sup>23</sup>	<sup>15</sup> U fission	1			
$\mathbf{U}(\mathbf{III})$	1.000		0.31	0.053	0.275	0.256	0.35	0.274
			$\pm 0.02$	$\pm 0.017$	$\pm 0.011$	±0.028	$\pm 0.05$	$\pm 0.011$
U(IV)	1.000	0.518	0.283	0.069	0.245	0.235	0.175	0.263
		$\pm 0.045$	$\pm 0.03$	• • •	•••	$\pm 0.05$	• • •	$\pm 0.040$
Mean	1.000	0.52	0.30	0.06	0.26	0.25	0.26	0.27
Estimated error		$\pm 0.05$	$\pm 0.02$	$\pm 0.02$	±0.05	$\pm 0.03$	$\pm 0.09$	$\pm 0.02$
			(b) In <sup>23</sup>	<sup>9</sup> Pu fissic	on			
Pu(I)	1.000	0.44	0.23	0.04	0.12	0.16	0.15	0.15
		±0.005	$\pm 0.002$		±0.008	$\pm 0.002$	$\pm 0.002$	$\pm 0.002$
Pu (II)	1.00	0.395	0.23	0.03	0.152	0.155	0.144	0.154
		±0.01	±0.005		±0.003	$\pm 0.005$	$\pm 0.005$	$\pm 0.002$
Mean	1.00	0.42	0.23	0.035	0.14	0.16	0.15	0.152
Estimated error		±0.02	$\pm 0.005$	±0.005	±0.02	$\pm 0.01$	$\pm 0.01$	±0.005

TABLE II. Relative abundances of tin isotopes.

phoric acid medium. Therefore in this case mass 124 is not reported as there was interference from a molecular ion peak at that mass number. The U(IV) sample was, on the other hand, loaded in nitric acid and therefore mass 124 could also be measured. Both Pu(I) and Pu(II) were loaded in the nitrate form only. A number of different sets of spectra were taken at different filament temperatures and beam currents. Each set was separately corrected for natural contamination, as this factor appeared to vary from set to set, probably as a result of some tin contamination originating from the instrument itself. The abundance at mass 116 was used to correct for natural contamination and, in order to ensure that there was no cadmium contribution at mass 116, mass 114 was also monitored during the measurement. In fact tin was always measured after the complete evaporation of cadmium, the latter being measured at a filament current of 0.45 to 0.5 A and the former at 0.7 to 0.8 A. The contamination level (at mass 117) was about 60% in the uranium samples and about 50% in the plutonium samples. The average values of the different sets of corrected abundances are given in Table II against each sample number together with the standard deviation. The mean value for two different samples is then given as well as an estimated error which takes into account the dispersion in the values both between the samples and among different sets.

### DISCUSSION

#### Effect of charge distribution

In the case of the heavier isotopes of both tin and cadmium, it is necessary to examine whether the measured yield represents the cumulative chain yield. For the mass 126 chain the most probable charge  $(Z_p)$  is 49.01 for <sup>235</sup>U fission and 49.70 for <sup>239</sup>Pu fission.<sup>20,21</sup> The measured yield at <sup>126</sup>Sn would therefore be about 96% of the chain yield in <sup>235</sup>U fission and 93% of the chain yield in <sup>239</sup>Pu fission. The corresponding figures for <sup>124</sup>Sn are 99.5% and 98.5%. Since these calculated values themselves are only estimates, no effort has been made to correct the measured relative yields accordingly. On the other hand, even for the heaviest cadmium isotope only a negligibly small fraction of the chain yield is lost to elements having higher atomic numbers.

#### Relative and absolute yields

Only by normalizing the relative yields of the tin isotopes with those of cadmium isotopes one can get the relative yields from masses 111 to 126. One way of doing this is to place the yields at masses 116 and 117 on a smooth curve. This approach seems justified in view of the nature of the curve in this region.

In order to obtain absolute yields from these relative values, it is necessary to normalize the yield at one mass number against a well established fission yield. In the region of masses 110 to 126, the only radiochemical yeilds that have a reasonable agreement in literature are those for the isotopes of silver. The yield of <sup>111</sup>Ag recommended by Walker,  $^{15}$  viz. 0.018% for  $^{235}$ U fission and 0.28% for <sup>239</sup>Pu fission, were chosen for the normalization. The fission yields thus obtained after normalization are quoted in Table III along with the literature values. Most of the literature values are quoted as updated by Meek and Rider.<sup>14</sup> The yield vs mass curve for the entire region from mass 111 to mass 126 is given in Fig. 1. The radiochemical yield at mass 109 for <sup>235</sup>U is also

TABLE III. Yields of tin and cadmium isotopes in the thermal neutron fission of  $^{235}$ U and  $^{239}$ Pu.

Mass	<sup>235</sup> U fissi	on yields (%)	<sup>239</sup> Pu fission yields (%)			
No.	Present work	Radiochemical	Present work	Radiochemical		
111	0.018 <sup>a</sup>	0.018 <sup>b, c</sup>	0.28 <sup>a</sup>	0.28 <sup>c</sup>		
112	0.0165	0.008-0.012, d 0.013 e	0.132	$0.10-0.12, d 0.11^{\circ}$		
113	0.015	0.013 <sup>e</sup>	0.076	0.065, <sup>f</sup> 0.082, <sup>g</sup> 0.076		
114	0.012	• • •	0.057			
115	•••	0.0104, <sup>c</sup> 0.0105 <sup>e</sup>	•••	0.038, <sup>c</sup> 0.037 <sup>e</sup>		
116	0.015	• • •	0.048	•••		
117	0.015 <sup>h</sup>	0.010 <sup>i</sup>	0.048	•••		
118	0.015 h	• • •	0.048	• • •		
119	0.014	• • •	0.051	• • •		
120	0.015	•••	0.045	•••		
$121^{j}$	0.0034	0.0008-0.012, 0.006 <sup>e</sup>	0.011	0.0003 e		
122	0.017	• • •	0.073			
124	0.030	•••	0.134			
125	•••	$0.029^{\rm c}$	• • •	$0.100^{\circ}$		
126	0.057	•••	0.318			

<sup>a</sup> Normalized at this mass number.

<sup>b</sup> Croall (Ref. 27).

<sup>c</sup> Values recommended by Walker (Ref. 15).

<sup>d</sup> All unreferenced values are taken from the compilation of Meek and Rider (Ref. 14).

<sup>e</sup> Values recommended by Meek and Rider (Ref. 14).

<sup>f</sup> Croall and Willis (Ref. 26).

<sup>g</sup> Jain (Ref. 22).

<sup>h</sup> Cuninghame, Kitt, and Rae (Ref. 25).

<sup>i</sup> Metcalf (Ref. 28).

<sup>j</sup> Yield of 76 yr  ${}^{121}$ Sn<sup>m</sup>.

plotted in Fig. 1(a) to show how the curve is rising towards lower mass numbers.

Only the sum of the yields at masses 113 and 114 were measured and resolving this into the individual components is difficult in the absence of accurate information on the neutron flux and self-shielding correction. In fact the identification of any fine structure in this region depends on the apportioning of the sum of the yields to masses 113 and 114. In the case of  $^{235}$ U fission it is seen that the sum of the yields at 113 and 114 is less than twice the yield at 116. Therefore, whatever way the division is made, at least one of the yields would be less than that at mass 116. A rough calculation of the absorption of neutrons by fission product <sup>113</sup>Cd shows that the abundance of <sup>114</sup>Cd in fission is less than that of <sup>113</sup>Cd. About 50% of the <sup>113</sup>Cd produced in fission is estimated to be converted to <sup>114</sup>Cd. Since the measured abundance of <sup>113</sup>Cd is about 0.5 relative to <sup>114</sup>Cd. the production rate of the latter in fission is less than that of the former. In view of the fact that this calculation only shows the trend, the yield at mass 113 was obtained by extrapolating the smooth curve from mass 112 to 113. Once the yield at 113 is estimated, that at 114 is automatically established. The values given in Fig. 1(a)and in Table III are obtained this way. The radiochemical yield of  $^{115}$ Cd (Table III) also seems to support this trend and makes the depression in the mass-yield curve at this point pronounced (see Fig. 1).

It must be pointed out here that there is no report in literature of a measurement of the yield of  ${}^{113}Ag$  in  ${}^{235}U$  fission. Meek and Rider<sup>14</sup> have quoted the radiochemical measurement of Cuninghame et al.<sup>25</sup> However, the latter authors determined only a "practical" ratio of <sup>90</sup>Mo/<sup>113</sup>Ag and the extrapolated yield at mass 113 was used in order to correct this ratio for counting efficiencies. The yields reported in literature for <sup>113</sup>Ag in the fission of <sup>239</sup>Pu have been measured by the comparison method which assumes the yield in <sup>235</sup>U fission.<sup>22</sup> Thus, here also the reported values are not reliable. Hence, the extrapolation method was used to estimate the contribution of mass 113 in the total yield of 113 and 114 in the <sup>239</sup>Pu case also. This gives a smooth curve [see Fig. 1(b)] unless one takes into account the radiochemical yield of <sup>115</sup>Cd [dotted line in Fig. 1(b)].

## Errors in measured yields

The errors in the yields reported in Table III arise from three different sources:

(1) the errors in the relative abundance measure-



FIG. 1. Fission yield vs mass curve for the symmetric region in the thermal neutron fission of (a)  $^{235}$ U, (b)  $^{239}$ Pu, and (c)  $^{233}$ U.

ments quoted in Tables I and II;

(2) the error involved in normalizing the relative abundances of tin and cadmium isotopes with respect to each other;

(3) the error in the yield of  $^{111}$ Ag used for normalization.

The first of these, which is the error in the experimental measurement, is given in Tables I and II. The second is difficult to estimate, though from the general trends in the yields in this region this error is likely to be rather small. The third component is also difficult to assess but does not affect the relative values which alone define the nature of the curve in Fig. 1. Therefore, no error assignment is made on the yields reported in Table III and the error bars shown in Fig. 1 are for the relative measurements only. It must be pointed out here that the possibility of mass 116 having a higher or lower yield than 117 cannot be ruled out. However, this will only enhance the fine structure in this region.

# Comparison with literature data

It is clear from Table III that there are very few radiochemical yields with which the present results can be compared. The only reported measurements at the mass numbers of our interest are on the nuclides <sup>111,112,113</sup>Ag, <sup>117</sup>Cd, <sup>121</sup>Sn, and <sup>123</sup>Sn. The yield at mass 111 has been used for normalization and that at 113 has already been discussed. The radiochemical yield of <sup>112</sup>Ag is somewhat less than our value, but this is not surprising since the radiochemical yield is obtained by resolving the activity of 3.2 h <sup>112</sup>Ag from that of 5.3 h <sup>113</sup>Ag. The decay scheme at mass 117 is too complicated to give very accurate yield values.

The yield listed for mass 121 is the independent yield of 76 yr  ${}^{121}$ Sn<sup>m</sup>, as  ${}^{121}$ In does not decay to the isomeric state of the tin isotope. A relatively large uncertainty has been quoted for this measurement (see Table II) because the measured intensities were very low. The values reported in literature for the yield of <sup>121</sup>Sn<sup>m</sup> range from 0.0008% to 0.012%.14 Meek and Rider have recommended an yield of 0.006%. Our measured value is about half that and even this is an upper limit. There is no reported measurement for the yield of  ${}^{121}Sn^m$ in <sup>239</sup>Pu fission. Meek and Rider have recommended 0.0003% as an estimate. Our value is much higher than this. It is remarkable that the measured independent yield is so high, suggesting thereby that most of the independent yield at tin in the mass 121 chain is in the form of the 76 yr isomer.

de Laeter and Thode<sup>16</sup> measured the relative yields of tin isotopes in the fission of <sup>233</sup>U only. However, they later reported<sup>17</sup> the measurement of the yields of cadmium isotopes in the fission of both <sup>233</sup>U and <sup>235</sup>U. Their relative yields in the latter case agree very well with the present measurements.

## Fine structure in the mass-yield curve

It is interesting to note that the mass distribution in the 116-126 mass region is quite smooth and does not appear to have any fine structure for either of the fissioning nuclides, viz.  $^{235}$ U and  $^{239}$ Pu. The radiochemical yields reported for mass 125 are somewhat lower than indicated by the smooth curves in Fig. 1. But this alone cannot be used to define the curve here because the error in the radiochemical yield could be high. Therefore the relative abundances of the tin isotopes were alone chosen to define the mass-yield curve. de Laeter and Thode<sup>16</sup> also came to the conclusion that the mass yield curve was smooth in this region for the neutron-induced fission of <sup>233</sup>U. Thind and Tomlinson<sup>13</sup> had predicted a sharp depression in the mass distribution at around mass 125. Calculations of Newson<sup>2</sup> also showed a similar trend. Only the vields of the odd masses are missing between 120 and 126 in the present work. The fact that a smooth

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curve can be drawn through them suggests that if there is any depression in yields in this region it must be so sharp as to be defined by a single mass number. However, it seems unlikely that this fine structure would appear at one of the three odd masses 121, 123, and 125 in the fission of three different nuclides, viz. <sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu. It may be pointed out here that the recommended radiochemical yields at mass 123 are 0.0169% and 0.044%, respectively, in <sup>235</sup>U and <sup>239</sup>Pu fission.<sup>14</sup> These values are lower than the interpolated values obtained by drawing a smooth curve through the normalized yields in this work. Even though Erdal, Williams, and Wahl<sup>23</sup> quote very good precision in their radiochemical measurements, the general agreement on this yield among different laboratories is not good. Thus it is not clear whether the radiochemical yield at mass 123 is defining a structure or questioning the interpolation and normalization in this work as well as the errors in the radiochemical yield measurements.

On the other hand, there appears to be a fine structure in the mass region 113-115. This is clearly seen for the case of <sup>235</sup>U fission in Fig. 1 and Table III. The situation is less clear for  $^{-239}$ Pu fission unless one takes into account the radiochemical yield at mass 115 (dotted line in Fig. 1). In the case of <sup>239</sup>Pu fission mass 113 comes at the foot of a steeply falling curve and therefore it cannot be fixed very accurately by extrapolation. However, it is worth noting that in Fig. 1(b) the curve is not symmetric, the left hand side falling more sharply than the right hand side. In general one would expect that in the prompt mass distribution this curve would be symmetric. Further, if there is no change in the slope of the neutron distribution for the region, the mass distribution after neutron emission would also not show such an increase in the slope for the low mass side. Thus the large slope of Fig. 1(b) around masses 112-114 may itself be a depression in yields. In fact, if we fold the curve Fig. 1(b) around the middle, the yields at 113 and 114 are found to be less than those for the corresponding right hand side masses by about 20-25%. In Fig. 1 we have also plotted the relative yields of cadmium and tin isotopes in the thermal neutron fission of <sup>233</sup>U as measured by de Laeter and Thode,<sup>17</sup> but normalized at mass 111 using radiochemical yields. The curve shows a depression at mass 111. It may be pointed out here that

the normalization with radiochemical yields at mass 111 is not responsible for the dip in the curve at mass 111. The yield at mass 111 is less than that at 112 and yields must in general increase towards the lower masses.

Any fine structure in the mass 111–115 region is likely to be connected with the change in the slope of the prompt neutron distribution. The depression in yields in this region may thus be related to the discontinuity around mass 110–115 in the sawtooth distribution of prompt neutrons as a function of fragment mass. In fact the first ramp of the sawtooth distribution in the case of  $^{235}$ U may be rising all the way up to mass 115 before changing slope. This interpretation receives support from the calculations of Prakash et al.<sup>24</sup> and does not conflict with neutron yield measurements because the statistics of counting and the mass resolution in actual neutron measurements (e.g., Apalin et al.<sup>29</sup>) do not permit an accurate definition of the maximum of the sawtooth curve.

If the maximum in the neutron distribution appears at mass 115, there is bound to be a depression in the measured yield at this point as the loss in yield due to neutron emission from this mass is not compensated by the increase in yield as a result of neutron emission by higher masses. In the case of  $^{233}$ U [Fig. 1(c)] the dip in the mass-yield curve and hence the maximum in the neutron yield distribution appears at mass 111.

In Fig. 1 it is interesting to note that the flat portion of the valley is wider for  $^{235}$ U than for  $^{239}$ Pu by about 2–3 mass units and still wider for  $^{233}$ U. This is clearly related to the fact that the light wing peak occurs at a higher mass number in  $^{239}$ Pu fission than in  $^{235}$ U or  $^{233}$ U fission.

## ACKNOWLEDGMENTS

This work was carried out at the McMaster University, Hamilton, Ontario, Canada during 1972-73. The author wishes to thank the Atomic Energy of Canada Limited for granting him a post doctoral felloship and the Bhabha Atomic Research Centre for granting him leave of absence for this period. He is extremely grateful to Professor H. G. Thode and Professor R. H. Tomlinson who not only provided all the facilities but also valuable guidance. He also acknowledges the assistance rendered by Dr. W. B. Clarke and Mr. E. M. Beaver.

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