# Relative Yields of Stable Tellurium Isotopes in Neutron-induced Fission

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*Abstract*

The relative isotopic abundances of four isotopes of tellurium (125, 126, 128 and 130) produced in the thermal neutron fission of $^{233}U$ and $^{235}U$ have been measured for the first time by solid source mass spectrometry. Samples of $^{233}U$ and $^{235}U$ were irradiated in a reactor and chemically separated by ion exchange techniques to permit nanogram-sized samples of fission product tellurium to be analysed mass spectrometrically. The results for $^{235}U$ are in good agreement with published radiometric values, whereas our results for $^{233}U$ are the first experimental measurements in this mass range. The cumulative fission yields determined by mass spectrometry for ruthenium, palladium, cadmium, tin and tellurium show a smooth mass distribution in the symmetric region for both $^{233}U$ and $^{235}U$, except for a significant depression in the yield curve in the range 111–14.

## 1. Introduction

The precise measurements of yields of stable end products of fission product mass chains have increased our knowledge and understanding of the fission process, and together with other nuclear data have enabled improvements in reactor design and operation to be accomplished. Reviews of cumulative yields from neutron-induced fission have been published by Walker (1973), Crouch (1977) and Rider (1979). These authors provide a set of recommended fission product yields for a variety of fissile materials.

Fine structure in the mass distribution in fission has been of interest since its discovery by Thode and Graham (1947) using mass spectrometrically determined fission yields. Since then, fine structure has been observed in a number of other regions of the mass yield curve. The fine structure in mass distribution as a function of mass arises partly as a result of variations in prompt neutron emission with fragment mass, partly because of delayed neutron emission and partly because of even/odd effects in the prompt yields (Nifenecker *et al.* 1979). An important region of interest in the study of fine structure is that of symmetric fission—in the valley of the mass yield curve—where the yields are low. Discontinuities in the neutron yield distribution appear in this region, and there have also been suggestions of a third peak in the symmetric region in the fission of some heavy elements (Jensen and Fairhall 1960; Iyer *et al.* 1963). Thind and Tomlinson (1969) calculated cumulative yield curves in the region of symmetric fission by assuming a general neutron emission with mass curve in conjunction with prompt yield curves assumed smooth for $^{235}U$, and this indicated two depressed regions of cumulative yields over masses 125 and 110.

However, until recently there have been little accurate yield data available in the symmetric fission region. Radiochemically determined fission yields in this region are, in general, inaccurate because of difficulties arising from the existence of isomers, the unavailability of accurate decay schemes and the low fission yields. Mass yields in the valley region are in fact several hundreds of times smaller than in the peak regions for thermal neutron-induced fission. This gap in our knowledge is now being filled.

De Laeter and Thode (1969) were the first to report a mass spectrometric study of the cumulative fission yields of tin produced by thermal and epicadmium induced fission of $^{233}$U, and a similar study for cadmium was reported by de Laeter and Thode (1975) for thermal and epicadmium fission of $^{233}$U and $^{235}$U and the epicadmium fission of $^{238}$U. These studies enabled the mass range from 111 to 126 to be examined. The results indicated that the mass yield distribution for thermal induced fission of $^{233}$U was relatively smooth in the valley of symmetry except for a possible depression around mass 113. Nor was there any evidence to support the existence of a symmetric peak.

Mathews (1977) also carried out a mass spectrometric determination of the cumulative fission yields for cadmium and tin for thermal neutron fission of $^{235}$U and $^{239}$Pu. These results also revealed a smooth mass distribution in the 116–26 mass region and confirmed the existence of fine structure in the 113–15 mass region. Mathews (1977) postulated that the depression in the mass yield curve around 113–15 may be related to the break around these mass numbers in the prompt neutron distribution as a function of mass.

Shima *et al.* (1978) have measured the relative cumulative yields of nine stable and long-lived isotopes of palladium and ruthenium produced in the thermal neutron fission of $^{233}$U and the thermal and epicadmium neutron fission of $^{235}$U and $^{239}$Pu. This was the first time that the palladium yields had been measured mass spectrometrically, thus enabling the low mass side of the symmetric region to be accurately defined. Together with the previously determined cadmium and tin isotopes, the results confirm a significant depression in the mass yield curve at masses 111 and 114 for $^{233}$U and $^{235}$U thermal fission respectively. The only mass range in the symmetric region of fission which has not been determined by mass spectrometric techniques is at the high mass side of the valley region where the fission chains are terminated by the isotopes of tellurium. Tellurium isotope abundances have been difficult to measure because of the high ionization potential of tellurium, particularly with the small samples available from fission product yields. The present study reports the relative fission yields of the tellurium isotopes $^{125}$Te, $^{126}$Te, $^{128}$Te and $^{130}$Te produced by the thermal neutron-induced fission of $^{233}$U and $^{235}$U.

## 2. Experimental Procedures

Prior to irradiation, the uranium samples in the form of $U_3O_8$ were purified in the following manner. The samples were first taken into solution in 6 M HCl with a minimum of nitric acid. After evaporation to dryness they were redissolved in 6 M HCl and placed on a small anion exchange column (Dowex AG1-X8). The uranium was eluted with 2 M HCl and the eluant collected directly into small, long-necked, quartz vials. The samples were slowly evaporated to dryness by a heat lamp, and the vials were then sealed under vacuum. This procedure effectively removes any

possible tellurium contamination from the samples, since tellurium is strongly held on anion exchange resins in 2 M HCl.

The samples were irradiated in the evacuated quartz vials in the AAEC Research Establishment's reactor HIFAR for 28 days at a thermal neutron flux in excess of $10^{13}$ n cm$^{-2}$ s$^{-1}$. The fast neutron flux in the irradiation positions was less than $4 \times 10^{11}$ n cm$^{-2}$ s$^{-1}$. Thus the fast neutron fission rate was less than 0·01% of the total fission rate and could be completely ignored. The irradiation data for the various uranium samples are given in Table 1. A long cooling period occurred before the fission product tellurium was extracted. This allowed the activity of the irradiated samples to decrease, but more importantly this delay was necessary to enable a significant proportion of the mass 125 fission chain to reach $^{125}$Te, since one of the precursors, $^{125}$Sb, has a half-life of 2·7 yr (Lederer and Shirley 1978).

**Table 1. Irradiation data**
The irradiation time for all samples was 28 days

| Sample | Wt (mg) | Neut. flux[A] (cm$^{-2}$ s$^{-1}$) | Cooling time[B] (days) | Sample | Wt (mg) | Neut. flux[A] (cm$^{-2}$ s$^{-1}$) | Cooling time[B] (days) |
|---|---|---|---|---|---|---|---|
| | | $^{235}$U thermal | | | | $^{233}$U thermal | |
| 58 (1) | 9·9 | $1·6 \times 10^{13}$ | 2010 | 61 (6) | 9·5 | $2·46 \times 10^{13}$ | 2010 |
| 59 (2) | 3·1 | $2·28 \times 10^{13}$ | 855 | 62 (7) | 7·5 | $2·66 \times 10^{13}$ | 858 |
| 60 (5) | 5·7 | $1·8 \times 10^{13}$ | 872 | 63 (10) | 10·7 | $2·51 \times 10^{13}$ | 872 |

[A] Average thermal neutron flux.
[B] The period from the end of irradiation to the beginning of chemical separation.

Each irradiated sample was dissolved, whilst still in its quartz vial, by repeated treatment with hot, concentrated HCl/HNO$_3$/HF acids. The final solution was evaporated to dryness, dissolved in 0·3 ml of 12 M HCl, and transferred to a 1 ml anion exchange column (Dowex AG1-X8, 100–200 mesh). A further three column volumes of 12 M HCl were added to elute the bulk of the cationic species, although some elements which would normally be cationic in solution are retained as anions as a result of complex ion formation at this chloride ion concentration. Tellurium was then eluted with ten column volumes of 0·5 M HCl. The eluate was then evaporated to dryness with a few drops of concentrated nitric acid, and then redissolved in 0·5 M HCl. This solution was transferred to a 0·75 ml cation exchange column (Dowex AG50-X8, 200–400 mesh), primarily to separate tellurium from uranium. Uranium is quantitatively retained on a cation column in 0·5 M HCl, whilst tellurium is eluted. This procedure also provided another stage of separation of tellurium from other species and enabled the final eluate to be sufficiently pure for mass spectrometric analysis.

The HCl and HNO$_3$ acids used in the separation procedure were prepared by sub-boiling distillation of the analytical grade acids in a quartz still, followed by sub-boiling distillation in a Teflon still. The water used for dilution and cleaning was deionized, then distilled in Pyrex followed by sub-boiling distillation in a quartz still. The ion exchange resins were supported on quartz wool in quartz tubes and were subjected to a rigorous cleaning procedure before use. All other laboratory ware used was made of Teflon, and careful clean-room techniques were used throughout.

Isotopic analyses were performed on a 30 cm radius, 90° magnetic sector, solid source mass spectrometer equipped with an electron multiplier. The samples were loaded on previously outgassed, zone-refined rhenium filaments with a phosphoric acid/silica gel mixture as the activator. Details of the mass spectrometric procedures are described by Smith *et al.* (1977). The fission-product samples produced ion beams of approximately $10^{-14}$ A for several hours. Terrestrial tellurium contamination, resulting either from the chemical processing or ion-source memory, could be detected by monitoring mass 124. However, no evidence of tellurium contamination was observed throughout the experiment. No other isobaric interferences occur at masses 125, 126, 128 and 130.

## 3. Results and Discussion

The measurement of the isotopic composition of tellurium using thermal ionization techniques has only recently been achieved (Smith *et al.* 1978). This technique allows the isotopic composition of nanogram amounts of tellurium to be measured, thus enabling fission yields for tellurium to be determined by mass spectrometric techniques for the first time.

**Table 2.   Observed isotopic abundances of fission product tellurium**

The mean values are in *italics*; those for $^{125}$Te have not been calculated since the value for each sample depends on the cooling time of each sample

| Sample | $^{125}$Te | $^{126}$Te | $^{128}$Te | $^{130}$Te |
|---|---|---|---|---|
| | | $^{235}$U thermal | | |
| 58 (1) | $0\cdot0129\pm0\cdot0001$ | $0\cdot00086\pm0\cdot00016$ | $0\cdot1816\pm0\cdot0011$ | 1 |
| 59 (2) | $0\cdot00824\pm0\cdot00017$ | $0\cdot00059\pm0\cdot00045$ | $0\cdot1818\pm0\cdot0012$ | 1 |
| 60 (5) | $0\cdot00852\pm0\cdot00011$ | — | $0\cdot1828\pm0\cdot0009$ | 1 |
| | | *$0\cdot00080\pm0\cdot00015$* | *$0\cdot1821\pm0\cdot0009$* | *1* |
| | | $^{233}$U thermal | | |
| 61 (6) | $0\cdot0311\pm0\cdot0006$ | $0\cdot00390\pm0\cdot0003$ | $0\cdot446\pm0\cdot003$ | 1 |
| 62 (7) | $0\cdot0300\pm0\cdot0008$ | $0\cdot00412\pm0\cdot0005$ | $0\cdot446\pm0\cdot006$ | 1 |
| 63 (10) | $0\cdot0311\pm0\cdot0005$ | $0\cdot00394\pm0\cdot0002$ | $0\cdot444\pm0\cdot002$ | 1 |
| | | *$0\cdot00396\pm0\cdot0002$* | *$0\cdot445\pm0\cdot002$* | *1* |

The observed isotopic abundances of fission product tellurium, normalized to mass 130, are given in Table 2. The fission yields have not been corrected for mass discrimination, nor for the effect of long-lived precursor nuclides in the 125 and 126 isobaric fission chains. Thus, the measured values for $^{125}$Te vary from sample to sample, depending on the cooling period between irradiation and chemical analysis as specified in Table 1, and therefore mean values have not been listed for this isotope. The stable isotope $^{126}$Te was measured with some difficulty, since it only represents a very small fraction of the mass 126 fission chain. This is due to the fact that its radioactive precursor, $^{126}$Sn, has a half-life of $10^5$ yr (Lederer and Shirley 1978). Thus, over the period of this experiment an insignificant amount of $^{126}$Sn has decayed to $^{126}$Te. However, small, independent fission yields for $^{126}$Sb and $^{126}$Te are produced in the fission process and it is this tiny fraction of the total mass 126 chain which has been measured (and listed in Table 2). Rider (1979) suggested that $0\cdot3\%$ of the total cumulative fission yield of the mass 126 chain will be produced at $^{126}$Sb

and $^{126}$Te for $^{235}$U, whereas the corresponding value for $^{233}$U is $1\cdot9\%$. Crouch (1977) also listed calculations of the independent fission yields, and suggested that $0\cdot7\%$ and $3\cdot2\%$ of the cumulative fission yield for the mass 126 fission chain is contributed by $^{126}$Sb and $^{126}$Te for $^{235}$U and $^{233}$U respectively. Thus, the recommended values given by Rider (1979) and Crouch (1977) suggest that the independent yields of $^{126}$Sb and $^{126}$Te comprise from $6\cdot3$ to $4\cdot6$ times as much of the cumulative yield for $^{233}$U than for $^{235}$U. It is of interest to note that the measured values in Table 2 show that the relative amount for $^{233}$U of these independent yields is approximately five times the corresponding amount for $^{235}$U.

Table 3. Relative cumulative yields of tellurium isotopes corrected for radioactive decay and mass discrimination for thermal neutron fission of $^{235}$U and $^{233}$U

Errors include estimates of mass spectrometer discrimination and $^{125}$Te precursor data in addition to the analytical errors from Table 2

| Mass | This work | Walker (1973) | Crouch (1977) | Rider (1979) |
|------|-----------|---------------|---------------|--------------|
| | | $^{235}$U thermal | | |
| 125[A] | $0\cdot018\pm0\cdot002$ | $0\cdot017$ | $0\cdot018$ | $0\cdot017$ |
| 126 | | $0\cdot031$ | $0\cdot037$ | $0\cdot036$ |
| 128 | $0\cdot180\pm0\cdot001$ | $0\cdot200$ | $0\cdot182$ | $0\cdot221$ |
| 130 | 1 | 1 | 1 | 1 |
| | | $^{233}$U thermal | | |
| 125[A] | $0\cdot068\pm0\cdot004$ | $0\cdot046$ | $0\cdot049$ | $0\cdot047$ |
| 126 | | $0\cdot104$ | $0\cdot105$ | $0\cdot104$ |
| 128 | $0\cdot439\pm0\cdot003$ | $0\cdot400$ | $0\cdot384$ | $0\cdot403$ |
| 130 | 1 | 1 | 1 | 1 |

[A] $^{125}$Te yields have been corrected for the decay of the mass 125 fission chain.

The experimental observations given in Table 2 have been corrected for mass discrimination and radioactive decay and are shown in Table 3, together with the recommended values of Walker (1973), Crouch (1977) and Rider (1979). A mass discrimination of $0\cdot625\%$ per mass unit was used to correct the data in Table 2, based on a value of $0\cdot68\%$ per mass unit determined by the double spike technique for cadmium isotopes measured in the mass spectrometer used in this project (Rosman and de Laeter 1975). However, since this value for tellurium is an estimate, the errors have been reassessed to allow for the uncertainty in the mass discrimination correction factor. The yield data for $^{125}$Te were corrected for the radioactive transformations involved in the mass 125 fission chain, using Bateman's equations (see e.g. Lapp and Andrews 1972). The major precursor is $^{125}$Sb which has a half-life of $2\cdot7$ yr. The uncertainties in the nuclear decay parameters are reflected in the errors associated with the $^{125}$Te fission yields listed in Table 3, which are at the $2\sigma$ level and include an assessment of all the errors involved in the measurements. No estimate of the cumulative yields for the 126 fission chain has been made since the quality of the data and the uncertainty in the proportion of fractional independent yields to cumulative yields are not known with sufficient certainty. A far better method of measuring the cumulative yield for mass 126 is to measure $^{126}$Sn (de Laeter and Thode 1969; Mathews 1977).

The measured yields for $^{235}$U thermal neutron-induced fission are in good agreement with the recommended values of Crouch (1977), and in reasonable

agreement with those of Walker (1973) and Rider (1979). A number of radiochemical measurements of the fission yields for these mass chains have in fact been carried out, whereas for the thermal neutron-induced fission of $^{233}$U there is an almost complete absence of experimental data. It is therefore not surprising that there are significant variations between the measured mass spectrometric yields and the estimated values given by the other authors listed in Table 3. The present values therefore represent the first direct yield measurements for $^{233}$U in this mass range.

Shima *et al.* (1978) have combined the cumulative mass fission yield data for $^{235}$U (thermal fission), from their measurements on ruthenium and palladium (masses ranging from 101 to 110), with the cadmium and tin isotope yields reported by de Laeter and Thode (1975) and Mathews (1977). Although the absolute yields of masses ranging from 101 to 110 are well established by normalization to a recommended value of $5\cdot04\pm0\cdot05\%$ at mass 101, the absolute yield values in the 111–26 mass range are much more uncertain. Shima *et al.* (1978) have normalized the relative mass spectrometer yields of the cadmium and tin isotopes in two different ways. By normalizing to a recommended value of $0\cdot018\%$ (based on the radiometric yield measurement of $^{111}$Ag), a good match is obtained with the palladium yields, producing a yield curve in the valley symmetrical about a mass $\bar{A}$ of $116\cdot79$. However, if the cadmium and tin data are normalized to recommended yields for masses 116 and 117, based on radiometric yields at masses 115, 121, 123 and 125, there is a considerable mismatch between the palladium and cadmium data. If the tellurium yields are normalized to the radiometric measurements at mass 125, there is good agreement with the radiometric yield at mass 127 of $0\cdot124\pm0\cdot015\%$ (Erdal *et al.* 1969) and a smooth transition from the tin to tellurium fission yields.

The situation with respect to the thermal fission of $^{233}$U is even more uncertain. Shima *et al.* (1978) have normalized the ruthenium and palladium yields to an absolute value of $3\cdot22\pm0\cdot02\%$ at mass 101, and then normalized the relative cumulative yield data for tin (de Laeter and Thode 1969) and cadmium (de Laeter and Thode 1975) by three different procedures—to a yield of $0\cdot014\%$ at mass 116, a yield of $0\cdot023\%$ at mass 111 and a yield of $0\cdot21\%$ at mass 126. The three procedures for normalization have extremes which differ by a factor of three and only the last matches up with the ruthenium–palladium data. The tellurium fission yields, as measured in this project, can also be made to match with the cadmium–tin yields normalized to a yield of $0\cdot21\%$ at mass 126 by normalizing to an absolute yield of $0\cdot116\%$ at mass 125. The mass spectrometrically determined fission yields indicate a wider valley than do the radiometric values, especially at mass 130. However, this reflects the situation found by Shima *et al.* (1978) for the low mass side of the fission valley.

Now that mass spectrometrically determined cumulative fission yields exist for all the major elements in the symmetric region, it is imperative that absolute yields for at least the palladium, cadmium, tin and tellurium isotopes be determined by the isotope dilution technique, to enable the shape of the fission valley to be determined with great confidence, and to define more precisely the position and magnitude of fine structure in this region of the mass distribution curve.

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