Nuclear Data Sheets, Volume 112, Issue 12, December 2011, Pages 3135-3152
http://dx.doi.org/10.1016/j.nds.2011.11.009
Los Alamos unclassified report LA-UR 11-03445

Fission Product Yields for 14 MeV Neutrons on ^{235}U, ^{238}U and ^{239}Pu

M. Mac Innes*, M.B. Chadwick, and T. Kawano[1]

[1]*Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

(Received 24 June 2011, revised received 22 September 2011; accepted 14 October 2011)

We report cumulative fission product yields (FPY) measured at Los Alamos for 14 MeV neutrons on ^{235}U, ^{238}U and ^{239}Pu. The results are from historical measurements made in the 1950s-1970s, not previously available in the peer reviewed literature, although an early version of the data was reported in the Ford and Norris review. The results are compared with other measurements and with the ENDF/B-VI England and Rider evaluation. Compared to the Laurec (CEA) data and to ENDF/B-VI evaluation, good agreement is seen for ^{235}U and ^{238}U, but our FPYs are generally higher for ^{239}Pu. The reason for the higher plutonium FPYs compared to earlier Los Alamos assessments reported by Ford and Norris is that we update the measured values to use modern nuclear data, and in particular the 14 MeV ^{239}Pu fission cross section is now known to be 15-20% lower than the value assumed in the 1950s, and therefore our assessed number of fissions in the plutonium sample is correspondingly lower. Our results are in excellent agreement with absolute FPY measurements by Nethaway (1971), although Nethaway later renormalized his data down by 9% having hypothesized that he had a normalization error. The new ENDF/B-VII.1 14 MeV FPY evaluation is in good agreement with our data.

Contents

*Electronic address: macinnes@lanl.gov

I. INTRODUCTION

In a recent special issue on nuclear reaction data [1] we described the LANL technique for calibrating the radiochemical laboratory system for fission product analysis. We described there the LANL fission product yields measured in fission spectrum neutron energies in fast critical assemblies, for average neutron energies in the 0 – 2 MeV range, and the implications for a new ENDF/B-VII.1 fission product yield evaluation [2]. Both the previous work at fission spectrum energies, and the results reported here at 14 MeV, have influenced a new ENDF/B-VII.1 FPY evaluation for plutonium [3].

This paper expands our results for fission product calibration experiments to include results of high energy (14 MeV) neutron irradiations of ^{235}U, ^{238}U, and ^{239}Pu. The experiments described in this work were conducted during the 1950s, 1960s, and 1970s at the Los Alamos Cockcroft-Walton irradiation facility. The results reported herein were recorded in staff member laboratory notebooks and internal memos and reports [4], but they have not previously been published in peer-reviewed journals. Although these data are quite old, they still are important and they represent one of the relatively few

measurement sets today that provide an absolute scale for 14 MeV FPYs. Therefore we feel it beneficial to make them available to the broader nuclear science community, even though the passage of time might have made a few aspects of the experiments difficult to describe in detail.

Unlike the the Los Alamos thermal and fission spectrum neutron irradiations described in Ref. [1], fission chambers were not used to determine the fission reference in the experiments. Instead associated alpha-particle detection from the d-t reaction was used to determine the 14.1 MeV fluence, in conjunction with $Al(n,\alpha)$ monitor foils, along with the 14.1 MeV fission cross section of the target and the number of target atoms, to determine the number of fissions occurring in the target foil. In this reassessment of the fission product measurements, we have used the modern evaluated ENDF/B-VII.0 fission cross sections to determine the number of fissions in the target foils to arrive at fission product yields (FPY). As we describe in our previous paper, ^{99}Mo plays a pivotal role as the standard reference nuclide in our measurement laboratory. This paper, therefore focuses on the analysis of the ^{99}Mo fission product produced in the irradiations. Results for the high-energy irradiations are compared with ^{99}Mo results for thermal neutron irradiation of ^{235}U. Results for other fission products are determined through ratio measurements to ^{99}Mo using the standard R-value approach developed by Rod Spence at this laboratory in 1949 [1, 5]. Throughout this paper we have taken the liberty of reproducing sections of our previous paper that describe important concepts and techniques used at LANL for completeness and to assist the reader.

Los Alamos has traditionally used radiochemical methods to separate and measure fission product β decays in order to determine the number of fissions, or burnup, that have occurred in a sample. This approach did not require the determination, or use of, fission product yields. Rather an approach was developed that utilized "K-factors"; constants relating counter response for a fission product to the number of fissions associated with a sample. K-factors had to be determined initially through calibration experiments, such as those described herein. In this paper we describe how to use ratios of K-factors measured in high energy and thermal neutron spectrum for any fission product, known as Q-values, to convert better known thermal FPY to 14 MeV FPY. Definitions of K-factors, Q-values, etc are given in Sec. II. Section III describes the LANL Cockcroft-Walton irradiation facility, experimental setup, target sample composition, irradiation conditions, and counting methods for radiochemically-separated fission products. Results of the experiments are given in Sec. IV along with a determination of modern FPYs based on modern ENDF thermal FPYs. In the appendix, we summarize results of 14 MeV irradiation LANL R-value measurements and provide a comparison with other laboratories participating in interlaboratory calibrations.

II. DEFINITION AND HISTORICAL APPROACH TO LABORATORY CALIBRATION

The historical approach used to determine the number of fissions in a sample at Los Alamos relied on a calibration method called the K-factor method. This ingenious calibration method permitted accurate determination of the number of fissions in a sample by measuring the fission product β decay count rate and converting it to the number of fissions through multiplication by the laboratory measured K-factor. The K-factors were simply proportionality constants determined separately in a calibration experiment where the number of fissions and the β decay count rate were each measured and then ratiod. This paper describes how K-factors for high-energy neutron irradiations were determined. Once determined, the K-factor could be applied to fission product count rates in samples in which the total number of fissions produced in a high energy irradiation was not directly measured. K-factors were radiation detector dependent making them useful only within our Los Alamos measurement program.

The K-factor for our reference nuclide ^{99}Mo is defined as

$$K_{99} = \frac{F}{A_{99}}, \tag{1}$$

where A_{99} is the fission product count rate and F is the total number of fissions in the calibration experiment. Unlike the calibration experiments described in our previous paper, these experiments used neutron fluence measurements, total actinide target atoms and the fission cross section to determine the total number of fissions in a macroscopic sample. Once the macroscopic foil was dissolved, the sample activity of any fission product could be measured by β decay resolution or in more recent experiments by gamma counting techniques. The relationship between the K-factor method for determining fissions in a sample and the more modern gamma ray measurement technique is as follows:

$$F = A_j K_j = \frac{N_j}{Y_j}, \tag{2}$$

where N_j is the number of atoms j determined by gamma spectroscopy and Y_j is the fission product cumulative yield. Equation (2) indicates the inverse relationship of K-factors and fission product yields (FPY). To translate from one fission product to another, LANL Radiochemistry constructed ratios that allowed translation from one fission product nuclide, fissioning material, and energy of the irradiation to another. These ratios were defined as Q-values and R-values. The Q-values were ratios of single radionuclide K-factors or Y_j's in different fuel and neutron energy conditions relative to the reference fuel and neutron energy spectrum:

$$Q_j = \frac{K_{*,j}}{K_{14,j}} = \frac{Y_{14,j}}{Y_{*,j}}. \tag{3}$$

In Eq. (3), the $*$ refers to the reference fuel and spectrum (thermal neutron irradiation of ^{235}U). The subscript $14, j$ denotes any non-reference fissioning isotope produced in the high energy neutron spectrum. Q-values are radiation detector independent and are therefore useful to any laboratory. As Eq. (3) indicates, Q-values can be expressed in terms of ratios of FPY as wells as ratios of K-factors. A rearrangement of Eq. (3) indicates that FPY for the high energy spectrum can be related to the better known thermal FPY through Q-values:

$$Y_{14,j} = Q_j Y_{*,j}. \quad (4)$$

R-values are double ratios of fission product count rates, see Eq. (5). The numerator is the activity (or atoms) of any fission product in any fissionable material irradiated in a high-energy neutron spectrum relative to the activity (or atoms) of the reference fission product (^{99}Mo) for the same material at the same neutron energy. The denominator is the activity ratio of the same two fission products in the reference material (^{235}U) in the reference neutron energy spectrum (thermal). The R-values are radiation detector independent and laboratory independent.

$$R_j = \frac{A_{14,j}/A_{14,99}}{A_{*,j}/A_{*,99}} = \frac{Q_{14,j}}{Q_{14,99}}. \quad (5)$$

III. EXPERIMENTAL APPROACH

The experiments described in this paper used the Los Alamos Cockcroft-Walton accelerator facility. These irradiations used the T(d,n)^{4}He reaction produced by 350-keV deuterons on a Zirconium tritide target. The target actinide foils were accurately positioned at 90 degrees to the incident deuteron beam producing a 14.1-MeV irradiation. Aluminum monitor foils, the same diameter as the actinide target foils were positioned on each side of the actinide target foil in a sandwich arrangement. In addition, an aluminum foil packet was placed at 90 degrees to serve as a primary monitor. The ^{24}Na produced in the aluminum foils from the ^{27}Al(n,α)^{24}Na reaction was counted on a β-proportional counter and the data treated by least-squares analysis. The activity on the primary monitor foil was used in conjunction with the flux number obtained from an α counter monitor to determine the fluence at the primary monitor foil position. The α counter measures the α particles from the T(d,n)^{4}He reaction and was calibrated in terms of an accurately known fraction of the number of such events in the tritium target. The uncertainty (4%, assessed at the time) in number of D-T events is mainly in the measurement of the area of the diaphragm which determines the fraction of total α particles accepted by the counter (see Fig. 1) – though later we suggest that this uncertainty might be lower. The flux at any sample position is then calculated from the ratio of activity of the aluminum foil sandwich surrounding the actinide target foil to the activity in the primary monitor foil. This technique reduced the error caused by inaccurate measurement of distance from target to sample or beam wandering to less than 3% and eliminated error due to Zr-T target shadowing. The integrated neutron fluence was usually of order 1×10^{12} neutrons/cm^2 at our sample position in about a four hour irradiation. The preceding description was lifted from a paper by R.J. Prestwood *et al.*, who along with J. Gilmore and G. Knobeloch are responsible for the experimental results reported in this paper. The original experimenters used the same technique during the 1960s to determine the 14.1-Mev cross section of numerous non-fissile materials [7]. Results of the experimentally determined ^{27}Al(n,alpha)^{24}Na cross section from the monitor foil packet for several irradiations are shown in Sec. IV. In Fig 1, the position indicated for the "samples" was used by R. Prestwood in the non-fissile material n,2n cross section experiments. In all of the experiments described in this paper, the primary Al monitor foils (shown at 10 cm in Fig 1.) and the target packet(aligned between the primary monitor Al foils and the Zr-T target) were placed in the same line-of-site.

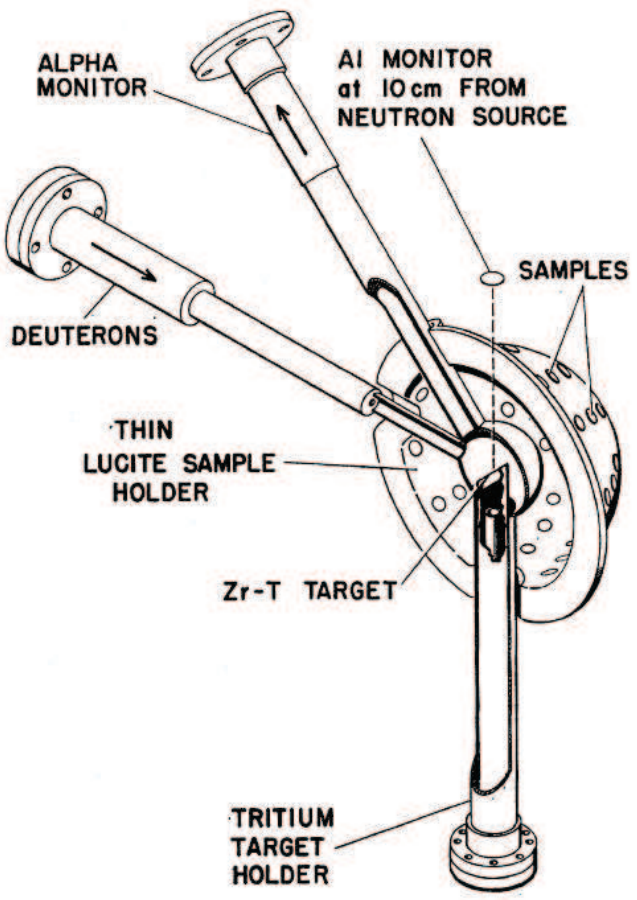

FIG. 1: Cockroft-Walton target and sample assembly.

A. December 1956 Irradiation

Four actinide target foils were irradiated for 12 hours in this experiment; one 93% enriched ^{235}U foil (HEU), one highly depleted ^{238}U foil (D-38), and two extremely pure ^{239}Pu foils ($^{240/239}$Pu ratio was 0.006) though one was not analyzed. All foils, both target and aluminum monitor foils were 1/2 inch in diameter. The HEU foil weighed 0.12 grams and was 93.27% ^{235}U, 1.05% ^{234}U, and 5.68% ^{238}U by weight. The D-38 foil weighed 0.268 grams with a $^{238/235}$U ratio of 3766:1. The isotopic composition of the uranium foils was determined by mass spectrometry. The uranium foils were each individually wrapped in 1.2 mil (0.0012 inches) aluminum foils to contain fission fragments. The plutonium foil mass was determined by alpha measurements on our standardized two-pi counters. The fraction of each plutonium isotope was determined by systematics for plutonium production available in the 1950s consistent with the measured fraction of ^{238}Pu activity in the plutonium of 0.0026. Based on the measured α activity, the plutonium foil weighed 0.1015 grams. The plutonium foils were coated with 5 mil copper. Aluminum monitor foils — 0.004 inch thick Dural disks weighing 32.2 mg each — were interspersed between each actinide foil. The total packet (see Table I) was wrapped in 15 mil cadmium and placed about 3 centimeters from the Zr-T neutron source. A pair of aluminum monitor foils was placed in the same plane 9 centimeters from the source. As described above, the two standard monitor foils were used in connection with the α counter monitor to determine the integrated fluence. Each of the aluminum sandwich foils were then related to the standard monitor foils by the simple ratio of ^{24}Na specific activity. The neutron fluence in the target actinide foil was determined from the average of the two aluminum foil monitors on either side of the target.

Following irradiation the foil package was separated and the individual foils and aluminum wrapper or copper plating was dissolved for subsequent radioanalytic measurements. The aluminum monitor foils were dissolved and purified samples of sodium were prepared for β counting of ^{24}Na and the data treated by least-squares analysis resolving the series of gross beta counts into a count rate extrapolated back in time to the end of irradiation. The fissile foils were dissolved and molybdenum was chemically separated and measured by β counting of the ^{99}Mo and the data treated by the β decay resolution techniques using least squares analysis.

The irradiated targets were dissolved in strong mineral acids. The solution was diluted to 25 mls with 4M HNO_3 and stored in a volumetric flask. Replicate aliquots were processed for radiometric determination of ^{99}Mo. The separation of molybdenum from fission products and actinides was facilitated by the unique chemical behavior of the hexavalent molybdate dianion (MoO_4^{2-}) on strong base anion exchange resin. A natural Mo carrier was added to an aliquot of the dissolved solution, which was equilibrated with ^{99}Mo as the MoO_4^{2-} anion upon heating the sample with dilute Br_2 in the presence of sulfuric acid. Molybdate was adsorbed onto an anion-exchange resin from 6M HCl solution, then washed successively with 0.1 M HCl-0.05M HF and 3M NH_4OH to remove most interfering ions. The Mo sample was then eluted from the column with 6M ammonium acetate. Molybdenum (IV) was selectively precipitated with alpha-benzoinoxime in the presence of oxalic acid, which served to optimize selectivity for Mo by preventing co-precipitation of potential interfering ions that may remain after the ion-exchange procedure. The isolated molybdenum alpha-bezoinoxime complex was ignited to MoO_3 in which form it was isolated onto a 1-inch filter paper disk, fixed with rubber cement, and mounted onto an aluminum planchet under a Mylar window for radiometric β decay resolution of the 2.75 day half life ^{99}Mo. The 6-hour ^{99m}Tc was allowed to reach equilibrium before data was taken for least-squares analysis.

TABLE I: Description of the foil packet used in the 1956 14 MeV irradiation. The neutron source came up from the bottom of this listing. There were actually two plutonium samples, but for simplicity we represent them as one in the table.

Material	Mass (mg)	Purpose
Al	32.2	Flux monitor
Al	32.2	Flux monitor
Al	32.2	Flux monitor
Cu	–	Can and catcher
Pu	101.5	Fissile material
Cu	–	Can and catcher
Al	32.2	Flux monitor
Al	–	Catcher foil
HEU	120.	Fissile material
Al	–	Catcher foil
Al	32.2	Flux monitor
Al	–	Catcher foil
D-38	268.1	Fissile material
Al	–	Catcher foil
Al	32.2	Flux monitor

B. May 1962 Irradiation

Two actinide target foils were irradiated for 9 hours in this experiment; along with several non-fissile foils as part of the experimental (n,2n) cross section measurement program. The two actinide foils were a 93% enriched ^{235}U foil (HEU) and a highly depleted ^{238}U foil (D-38). As in the 1956 experiment, the mass of the foils was determined by weighing. The HEU foil weighed 0.2949 grams and was 93.38% ^{235}U, 1.05% ^{234}U, and 5.57% ^{238}U by weight. The D-38 foil weighed 0.07715 grams with a $^{238/235}$U ratio of 1250:1. The isotopic composition of the uranium foils was determined by mass spectrometry. The uranium foils were each individually wrapped in 2 mil aluminum foils to contain fission fragments. Aluminum

TABLE II: Description of the foil packet used in the 1971 14 MeV irradiation. The neutron source came up from the bottom of this listing. There were actually two plutonium samples, but for simplicity we represent them as one in the table.

Material	Mass (mg)	Purpose
Al	23.35	Flux monitor
Al	23.38	Flux monitor
Al	23.40	Flux monitor
Al	–	Catcher foil
Oy	84.00	Fissile material
Al	–	Catcher foil
Al	23.58	Flux monitor
Al	–	Catcher foil
D-38	82.35	Fissile material
Al	–	Catcher foil
Al	23.56	Flux monitor
Al	–	Catcher foil
Oy	79.87	Fissile material
Al	–	Catcher foil
Al	23.53	Flux monitor
Ni	–	Can & catcher
Pu	92.83	Fissile material
Ni	–	Can & catcher
Al	23.10	Flux monitor
Al	–	Catcher foil
Oy	82.12	Fissile material
Al	–	Catcher foil
Al	23.42	Flux monitor

monitor foils were interspersed between each actinide foil and the non-fissile foils. A pair of aluminum monitor foils was placed in the same 90 degree plane 10.1 centimeters from the source. As described above, the two standard monitor foils were used in connection with the α counter monitor to determine the integrated fluence. Each of the aluminum sandwich foils was then related to the standard monitor foils by the simple ratio of ^{24}Na specific activity. The fluence of the target actinide foil was determined from the average of the two aluminum foil monitors on either side of the target.

Following irradiation the foil package was separated and the individual foils and aluminum wrapper were dissolved for subsequent radioanalytic measurements. The aluminum monitor foils were dissolved and purified samples of sodium were prepared for β counting of ^{24}Na and the data treated by least-squares analysis. The fissile foils were dissolved and molybdenum was chemically separated and measured by β decay resolution techniques.

C. November 1971 Irradiation

Seven actinide target foils were irradiated for 9 hours in this experiment; three 93% enriched ^{235}U foil (HEU), two highly depleted ^{238}U foil (D-38), and two plutonium foils with a ^{240}Pu content of 5.6 wt%. All foils, both target and aluminum monitor foils were 3/8 inch in diameter. The three HEU foils weighed between 80 and 85 milligrams and were 93.21% by weight ^{235}U, 1.02% ^{234}U, 0.67% ^{236}U, and 5.09% ^{238}U. The mass of ^{235}U in the HEU foils was determined by comparison fission counting using a dual fission chamber with a known mass of ^{235}U in one chamber and a known quantity of sample from the experiment in the other chamber. The two D-38 foils weighed a combined 82.35 milligrams (determined pre-irradiation by weighing) with a $^{238/235}$U ratio of 1250:1. The two foils were combined after the irradiation into one sample. The isotopic composition of the uranium foils was determined by mass spectrometry. The uranium foils were each individually wrapped in 2 mil aluminum foils to contain fission fragments. The plutonium foil mass (the two foils were combined after irradiation into one sample) was determined by α measurements on our standardized two-pi counters. The fraction of each plutonium isotope was determined by mass spectrometry except for ^{238}Pu that was determined by α spectroscopy. Based on the measured α activity and isotopic composition the plutonium combined sample weighed 92.8 milligrams. The plutonium foils were canned in 5 mils of nickel. The aluminum monitor foils were each weighed individually and interspersed with the target actinide foils as shown in Table II). A pair of aluminum monitor foils was placed in the 90 degree plane 10 centimeters from the source. As described above, the two standard monitor foils were used in connection with the alpha counter monitor to determine the integrated fluence. Each of the aluminum sandwich foils was then related to the standard monitor foils by the simple ratio of ^{24}Na specific activity. The neutron fluence in the target actinide foil was determined from the average of the two aluminum foil monitors on either side of the target.

Following irradiation the foil package was separated and the individual foils and aluminum wrapper or nickel can was dissolved for subsequent radioanalytic measurements. The aluminum monitor foils were dissolved and purified samples of sodium were prepared for β counting of ^{24}Na. The fissile foils were dissolved and molybdenum and neodymium were chemically separated and measured by β decay resolution techniques. The molybdenum separation procedure was described previously. Group separation of the latanides was accomplished using a series of precipitation/redissolution steps in which LaF_3 and $La(OH)_3$ were sequentially separated from the bulk of the fission product mixture. After initial purification, the individual lanthanides were separated from one another using an HPLC process that employs a cation-exchange stationary phase and alpha-hydroxyisobutyric acid mobile phase. The eluted neodymium fraction was precipitated as the oxalate, and fired at 900 degrees C to the blue oxide, Nd_2O_3. The radioanalytic β decay of the 10.98-day ^{147}Nd was measured through a 18.7 mg/cm^2 Al absorber to eliminate the ^{147}Pm daughter from the measured count rate. The resulting count rate data was resolved by least-squares analysis.

IV. RESULTS AND DISCUSSION

In this section we provide results of the K-factors (fissions/β-cpm), and Q-values (ratios of K-factors) for the three 14-MeV irradiations. As we explained earlier, K-factors are of vital interest for internal LANL assessments of the number of fissions that have occurred in actinides subject to a neutron fluence; however they apply only to the LANL β detectors used for the calibration experiments, and therefore have limited utility to other laboratories. The high-energy Q-value (the ratio of K-factors between thermal and high-energy neutron spectrum irradiations) is of direct importance to fission product yield studies, as it represents the ratio of fission product yield of high-energy neutron spectrum to thermal neutron spectrum irradiation of ^{235}U. The high-energy Q-value is especially useful because in general the thermal ^{235}U FPY are more accurately known than the 14 MeV FPY and the Q-value allows the determination of the 14 MeV FPY from thermal data as we show later in section IV.

A. K-factors

1. December 1956 high-energy result

Applying Eq. (1), the K-factor for ^{99}Mo is the total number of fissions in the target foil divided by the measured β-decay count rate for the total target foil. In Table III, we summarize the results of the K-factors measured in this experiment along with the thermal ^{235}U K-factor on the reference β counter at the time of the experiment. To illustrate the ^{99}Mo K-factor determination, we elaborate the calculations with the ^{239}Pu high-energy neutron irradiation experiment. The cross sections used for fissions of the uranium and plutonium isotopes are listed in Table IV.

$$\begin{aligned} &\text{Total target fissions} \\ &= \text{integrated target fluence} \\ &\times \sum \text{isotopic target atom} \times \text{isotopic cross section} \\ &= 2.116 \times 10^9 \quad \text{fissions}, \end{aligned} \tag{6}$$

which is derived from:

$$\begin{aligned} F &= 35150 \text{ cpm} \times 10^{24} \text{ Na/Al foil} \\ &\times \frac{4.306 \times 10^{12} \text{ fissions/cm}^2@9cm}{4405 \text{ cpm} \times 10^{24} \text{ Na/Al foil}} \\ &= 3.437 \times 10^{12}(\text{ fissions/cm}^2 \text{ in Pu target}) \\ &\times \{9.84 \times 10^{13} \cdot 2.07 + 1.02 \times 10^{19} \cdot 2.41 \\ &\quad +6.10 \times 10^{16} \cdot 1.62 + 8.13 \times 10^{14} \cdot 2.18 \\ &\quad +2.34 \times 10^{13} \cdot 1.98\} \times 10^{-24} \text{ cm}^2 \\ &= 2.116 \times 10^9 \quad \text{fissions}. \end{aligned} \tag{7}$$

TABLE III: December 1956 experiment ^{235}U (HEU), ^{238}U (D-38) and ^{239}Pu 14-MeV K-factors measured at LANL. The derived Q_{99} values use a thermal ^{235}U K-factor of 2.333×10^5 (the old counter 6 measured value [1]).

	K-factor (fission/cpm)	Unc. (%)	Q-value (no dim.)	Unc. (%)
^{235}U	2.701×10^5	2.17	0.8636	2.69
^{238}U	2.406×10^5	2.20	0.9696	2.72
^{239}Pu	2.487×10^5	2.64	0.9380	3.09

TABLE IV: Assumed 14.1 MeV fission cross sections, taken from ENDF/B-VII.0 [6].

	Cross Section (b)		Cross Section (b)
^{234}U	2.070	^{238}Pu	2.670
^{235}U	2.090	^{239}Pu	2.410
^{236}U	1.620	^{240}Pu	2.210
^{238}U	1.150	^{241}Pu	2.180
		^{242}Pu	1.980

Count rates were reported in cpm per ml A solution, where the A solution was the primary solution derived from the dissolution of the target foil, diluted to a volume of 25 mL. The total target cpm of ^{99}Mo was the cpm/ml times the total volume i.e. 340.3 cpm/ml $\times$ 25 ml = 8507.5 cpm. From these values, the ^{99}Mo K-factor for plutonium is 2.116×10^9 fissions/8507.5 cpm = 2.487×10^5 fission/cpm as shown in Table III. The ^{239}Pu content of the target foil accounted for 99.5% of the total ^{99}Mo count rate. Other K factors were determined in the same way.

The K-factor for the D-38 target was determined from a ratio of the total fissions in the ^{238}U portion of the foil to the ^{238}U portion of the ^{99}Mo count rate, even though the ^{235}U content of the D-38 foil is an insignificant fraction of the total atoms, the total fissions and the total ^{99}Mo count rate.

The K-factor for the HEU foil took into account the contribution of the ^{238}U isotopic fraction but treated the ^{234}U and any ^{236}U (the isotopic fraction was below detection level) as ^{235}U. The ^{238}U content of the HEU target foil accounted for about 4.5% the total ^{99}Mo count rate.

2. May 1962 K-factor result

In Table V we summarize the results of the K-factors for the May 1962 experiment. There were one ^{235}U target foil and one D-38 foil. The fissions in each foil were determined by the same procedure as we described above. The fluence was determined from the counts in the α counter monitor and the aluminum foil monitors at the 10.1-centimeter position. The aluminum foil sandwich monitors were then normalized to the 10.1-centimeter fluence determination through the relative ^{24}Na specific activity.

TABLE V: May 1962 experiment for ^{235}U (HEU) and ^{238}U (D-38) 14-MeV K-factors measured at LANL. The derived Q_{99} values use a thermal ^{235}U K-factor of 2.333×10^5 (the old counter 6 measured value).

Year	K-factor (fission/cpm)	Unc. (%)	Q-value (no dim.)	Unc. (%)
^{235}U	2.724×10^5	2.17	0.8565	2.69
^{238}U	2.430×10^5	2.20	0.9600	2.72

The K-factor for the D-38 target was determined from a ratio of the total fissions in the ^{238}U portion of the foil and the ^{238}U portion of the ^{99}Mo count rate even though the ^{235}U content of the D-38 foil is an insignificant fraction of the total atoms, fissions and ^{99}Mo count rate.

The K-factor for the HEU foil took into account the contribution of the ^{238}U isotopic fraction but treated the ^{234}U and ^{236}U, as ^{235}U. The ^{238}U content of the HEU target foil accounted for about 3.5% the total ^{99}Mo count rate.

3. November 1971 high-energy result

In Table VI we summarize the results of the K-factors for the November 1971 experiment. There were three ^{235}U target foils, two D-38 foils and two plutonium foils. The two D-38 foils were combined into one sample. Likewise, two plutonium foils were combined into one sample. The fissions in each foil were determined by the same procedure as we described above. The fluence was determined from the counts in the α counter monitor and the aluminum foil monitors at the 10-centimeter position. The aluminum foil sandwich monitors were then normalized to the 10-centimeter fluence determination through the relative ^{24}Na specific activity.

Due to the higher ^{240}Pu content, the fissions in ^{239}Pu in the 1971 experiment accounted for only 95% of the total ^{99}Mo count rate. However, the K-factor was treated as if all the plutonium was ^{239}Pu. That is, the total fissions in the plutonium foil computed from the sum of the product of the plutonium atoms times the isotopic cross section was ratiod to the total ^{99}Mo count rate.

The K-factor for the D-38 target was determined from a ratio of the total fissions in the ^{238}U portion of the foil and the ^{238}U portion of the ^{99}Mo count rate even though the ^{235}U content of the D-38 foil is an insignificant fraction of the total atoms, fissions and ^{99}Mo count rate. The K-factor for the HEU foil took into account the contribution of the ^{238}U isotopic fraction but treated the ^{234}U and ^{236}U, as ^{235}U. The ^{238}U content of the HEU target foil accounted for about 3.1% the total ^{99}Mo count rate.

TABLE VI: November 1971 ^{235}U (HEU), ^{238}U (D-38) and ^{239}Pu 14-MeV K-factors measured at LANL. The derived Q_{99} values use a thermal ^{235}U K-factor of 2.445×10^5 (in January 1970 the detector in counter 6 was replaced resulting in a new measured K-factor value [1]).

	K-factor (fission/cpm)	Unc. (%)	Q-value (no dim.)	Unc. (%)
^{235}U	2.879×10^5	2.93	0.8491	3.34
^{235}U	2.788×10^5	2.93	0.8769	3.34
^{235}U	2.843×10^5	2.93	0.8599	3.34
^{238}U	2.609×10^5	2.20	0.9371	2.72
^{239}Pu	2.648×10^5	2.66	0.9235	3.10

TABLE VII: Inferred ^{27}Al(n,α) cross sections from series of experiments, where the 14.1-MeV neutron fluence was determined independently. The average result, 121.6 mb ± 1.74%, agrees well with the ENDF/B-VII.0 evaluation at 14.1 MeV, 121.3 mb, see text. This provides confirmatory evidence on the accuracy of the 14.1 MeV neutron fluence that was determined.

Date	^{27}Al(n,α) (mb)
10-Mar-62	120.5
11-Apr-62	119.2
6-May-62	126.0
10-May-62	117.9
4-Jun-62	121.9
20-Sep-62	118.2
22-Jan-64	122.2
1-Sep-65	122.0
11-May-66	123.2
29-Apr-68	121.2
30-Sep-68	121.3
16-Dec-68	119.9
19-May-70	122.5
26-Mar-72	123.9
3-Nov-72	122.5
5-Sep-78	123.0

B. Uncertainty Assessments

In our previous paper we described the uncertainty in our K-factor determinations that made use of a calibrated NIST fission chamber to assess the number of

TABLE VIII: 14 MeV Q-values for molybdenum-99, LANL's reference fission product, based on an average of the various LANL replicate measurements. The Q-value is dimensionless and is a ratio of K-factors, but is also a ratio of 14 MeV and thermal ^{235}U FPYs and can therefore be compared with ENDF values, and with Nethaway (1971).

	Q_{99}-value (this work)	ENDF /B-VII.1	ENDF /B-VII.0	Nethaway 1971
^{235}U	0.861 ± 3.2%	0.841	0.841	–
^{238}U	0.956± 3.3%	0.933	0.933	0.948
^{239}Pu	0.931± 3.3%	0.927	0.777	0.928

fissions in the target macrofoil. In this work the uncertainty in the number of fissions depends on the accuracy of the Cockcroft-Walton α particle monitor, the number of atoms in the target foil, and the fission cross section at 14.1 MeV. In Section III we cited the accuracy of the α-particle monitor as 4% based upon statements made by R. Prestwood who made and published extensive measurements of cross sections using this experimental irradiation facility [7]. These cross section measurements required knowledge of the fluence, number of atoms in the target foil, and an absolute calibration for counting the radioactive product. At Los Alamos, the absolute disintegration rate of a radioactive isotope was determined by either the gamma sum coincidence method [9] or 4-π β-γ coincidence method [10] depending on the decay scheme. The 4-π β-γ coincidence method was developed by J. Balagna in the early 1950s. The γ sum coincidence method was developed at our laboratory by D. Barr and J. Gilmore in the early 1960s independent of the work reported in Ref. [9]. For each 14.1-MeV irradiation used to determine cross sections at the Cockcroft-Walton facility we would also determine the ^{27}Al(n,alpha)^{24}Na cross section. In Table VII we list results for the aluminum cross section checks determined between the early 1960s and late 1970s. The average value of 121.6 mb is to be compared with the current 14.1 MeV ENDF-VII value of 121.2 mb; a value only 0.2% higher. The standard error (statistical) in the measured cross section during this period was 1.74%. We believe this is an accurate reflection of the reproducibility in the fluence determination and is also a reasonable estimate of the total uncertainty, because we assess the systematic uncertainty to be small since the (n,alpha) cross section agrees very well (0.2%) with the ENDF value. To this value we added in quadrature the assessed uncertainty in the 14.1-MeV ENDF-VI fission cross section of the major isotope (^{235}U, ^{238}U, or ^{239}Pu) [17] and the uncertainty in the number of target atoms. The mass of the D-38 foils was determined by weighing after cleaning with weak acid and the isotopic composition by mass spectroscopy. The same was true of the HEU foils used in the 1956 and 1962 experiments. We assessed the uncertainty in the isotopic composition of the major uranium atoms to be 0.3%. The mass of the ^{235}U in the HEU foils in the 1971 experiment was determined by comparison fission counting using aliquots from the dissolved foils. The isotopic composition of the foils was determined by mass spectroscopy. We assessed a 2% uncertainty in the ^{235}U mass of the HEU. The plutonium mass was determined by alpha spectroscopy. The isotopic composition of the plutonium was determined by mass spectroscopy. We assessed a 1.5% overall uncertainty to the major isotopic plutonium atom content.

The uncertainty in the ^{99}Mo count rate (the denominator in the K-factor) was described in our previous paper where we explained that the reproducibility of the overall measurements of ^{99}Mo was a better estimate of uncertainty than the precision of measurements on replicate samples on any one experiment. This uncertainty was assessed for β counting to be approximately 1.1%. Thus we ascribe an overall uncertainty to our 14 MeV K-factor determinations of 2.2% for the D-38 experiments. The 1971 HEU experimental uncertainties were higher at 2.9%. Finally, the uncertainty in the plutonium K-factors was 2.6%.

The uncertainty in the Q-values was determined by adding the 14.1-MeV K-factor uncertainty in quadrature with the thermal K-factor (1.6% from Ref. [1]). This resulted in uncertainties of between 2.7-3.3% as we show in Tables III, V, and VI.

Our fission product yield uncertainties come from combining in quadrature the aforementioned Q-value uncertainties, the R-value uncertainties, and the thermal FPY uncertainties, see Tables IX, X, and XI. The method we use involving ratios to thermal FP data enables us to obtain smaller absolute FPY uncertainties than are sometimes quoted in other methods. (The thermal FPY uncertainties, taken from ENDF, are quite small owing to them being based on a weighted average of many measurements from around the world, including accurate mass spectrometry measurements).

Other corrections were considered and not included because they were insignificant under the specific experimental conditions including other neutron sources from thermal and epithermal room return and in the plutonium sample, spontaneous fission of ^{240}Pu. In 1950, W. Nyer [8] conducted relative cross section measurements at the Cockcroft-Walton facility using a dual fission chamber. He determined that the high thermal cross section of ^{235}U required the chamber to be wrapped in cadmium to cut out thermal neutrons. In the 1956 experiment, we found documentation that the foil packet was cadmium wrapped. Although this documentation was not found in later experiments, it is highly likely that all experiments were wrapped in cadmium because they were conducted by the same staff members who were well aware of the thermal neutron room return issue. In his article, Nyer also determined the correction for epithermal neutrons. Based on information in his article, we estimate the correction for epithermal neutrons to be a few tenths percent. Although we think, therefore, that it is likely that corrections due to room return were been minimized because LANL experimentalists were certainly aware of this concern, the passage of time since these experiments were done makes it hard for us to definitively state that no such contamination occurred.

In January 1960, J. Gilmore, R. Prestwood, G. Knobleoch, and D. Barr determined the equilibrium activity level of ^{99}Mo due to spontaneous fission of ^{240}Pu. A value of 0.52 cpm of ^{99}Mo/milligram of ^{240}Pu was obtained based on our standard counting geometry. Using this value, the contribution due to ^{240}Pu spontaneous fission is about 0.04% of the observed 99Mo activity in the 1971 experiment at the time of separation from plutonium. The spontaneous fission contribution in the 1956 experiment was a factor of 5 less.

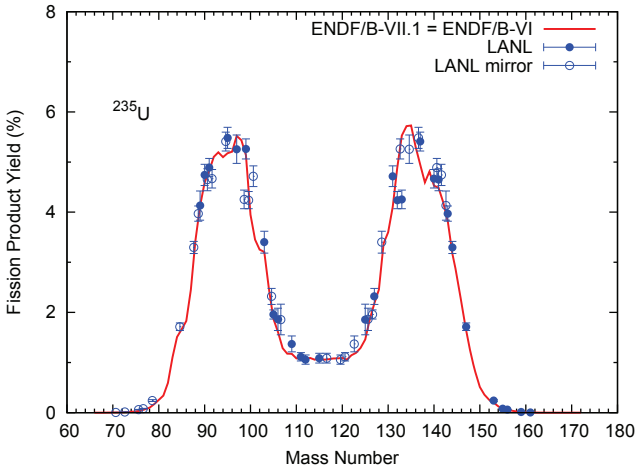

FIG. 2: ^{235}U cumulative FPYs at 14 MeV.

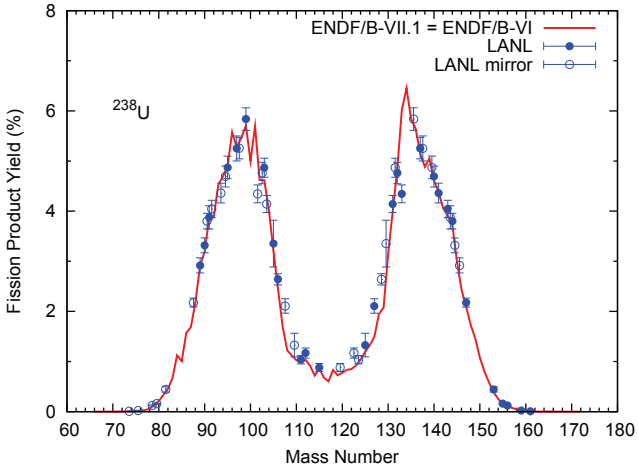

FIG. 3: ^{238}U cumulative FPYs at 14 MeV.

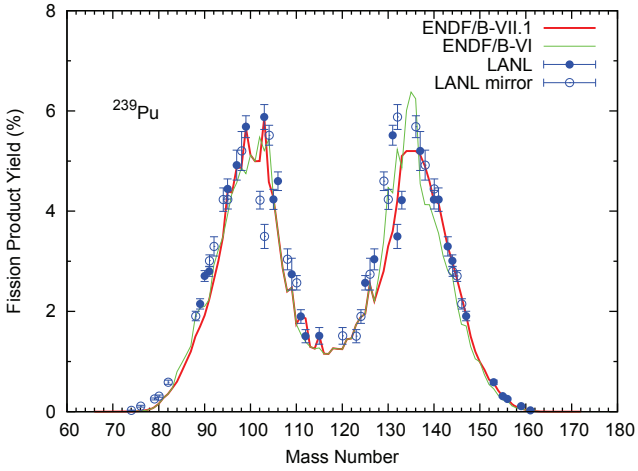

FIG. 4: ^{239}Pu cumulative FPYs at 14 MeV.

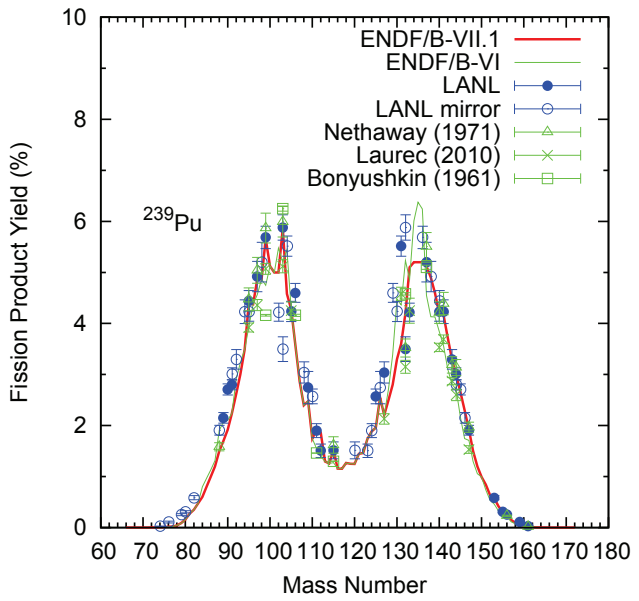

FIG. 5: ^{239}Pu cumulative FPYs at 14 MeV, with comparison to other measurements.

C. Q-values and Fission Product Yields

As we described in Sec. II, K-factors were used by Los Alamos to determine the number of fissions in a sample, but they are individually of little utility except in our measurement facility. The ratio of the 14 MeV K-factor to the thermal ^{235}U K-factor (the Q-values, see Table VIII) are however, applicable to the nuclear scientific community because they are equivalent to the ratio of fission product yields. This relationship was defined in Sec. II:

$$Q = \frac{K_{\rm ref}}{K_i} = \frac{Y_i}{Y_{\rm ref}}, \tag{8}$$

leading to the 14 MeV FPY in terms of the thermal FPY:

$$Y_i = Q_i Y_{\rm ref}. \tag{9}$$

Although thermal FPY, $Y_{j,235}$ have been measured at Los Alamos (see appendix A table 12) with fission products atoms determined from high resolution γ-ray spectroscopy and fissions being determined by β-decay resolution through the use of our K-factors, for this discussion the thermal FPY will be adopted from ENDF/B-VI. We do this because these values will likely be used in future ENDF FPY upgrades and the uncertainty in ENDF values are smaller than our measurements benefiting from the high precision mass spectrometry measurement. Moreover, γ-ray determined FPYs are laboratory dependent quantities — depending on γ-ray detector calibrations and the decay scheme used in converting the count rate deduced from the spectroscopic measurement to atoms.

TABLE IX: Measured 14-MeV neutron induced ^{235}U fission product yields from LANL. The Q_{99} value was taken as 0.861± 3.23%, based on LANL K-factor experiments. We also show for comparison two Livermore experiments by Nethaway from 1983 (documented in 1993 [12]) — but these are relative, and so we use parentheses around them since Nethaway obtained these magintitudes by normalization to Rider's yields at the time.

	Thermal ENDF FPY ^{235}U (%)	Unc. (%-relative)	Evauated R-value (this work) (no dim.)	Unc. (%)	**14 MeV FPY LANL (%)**	14-MeV FPY *(relative)* Nethaway'83a (%)	14-MeV FPY *(relative)* Nethaway'83b (%)
Sr-89	4.733	1.4	1.014	6.08	**4.131 ± 7.03 %**		
Sr-90	5.782	1.4	0.953	2.75	**4.743 ± 4.47 %**		
Y-91	5.828	1.0	0.974	1.53	**4.890 ± 3.71 %**	(5.27)	(4.71)
Zr-95	6.503	2.0	0.979	0.53	**5.483 ± 3.84 %**	(5.29)	(5.27)
Zr-97	5.984	2.8	1.020	3.28	**5.257 ± 5.39 %**	(4.62)	(4.62)
Mo-99	6.109	2.0	1.000	—	**5.261 ± 3.80 %**	(5.02)	(5.03)
Ru-103	3.031	2.0	1.30	5.14	**3.401 ± 6.39 %**	(3.09)	(3.09)
Rh-105	0.964	2.8	2.36	1.85	**1.959 ± 4.66 %**	(1.80)	(1.79)
Ru-106	0.4016	2.0	5.38	11.32	**1.861 ± 11.94 %**	(1.92)	(1.66)
Pd-109	0.03122	11	51	1.40	**1.371 ± 11.55 %**		
Ag-111	0.01738	6.0	74.4	3.09	**1.114 ± 7.48 %**	(1.10)	(1.10)
Pd-112	0.01304	8.0	94.3	0.60	**1.059 ± 8.65 %**	(1.12)	(1.13)
Cd-115	0.01158	8.0	100.5	1.83	**1.002 ± 8.82 %**		
Cd-115m	0.001005	8.0	97.7	1.45	**0.085 ± 8.75 %**		
Sb-125	0.034	4.0	63.5	15.66	**1.859 ± 16.48 %**	(1.24)	(1.14)
Sb-127	0.157	6.0	17.2	1.24	**2.319 ± 6.93 %**	(1.46)	(1.44)
I-131	2.89	1.4	1.895	2.61	**4.716 ± 4.38 %**		
Te-132	4.295	2.0	1.147	1.33	**4.241 ± 4.03 %**	(4.02)	(4.00)
I-133	6.697	1.4	0.738	2.59	**4.254 ± 4.37 %**		
Cs-136	0.0055	20	45.4	7.22	**0.215 ± 21.51 %**	(0.237)	(0.220)
Cs-137	6.188	0.7	1.015	1.11	**5.409 ± 3.49 %**	(5.02)	(5.11)
Ba-140	6.215	1.4	0.872	1.78	**4.666 ± 3.94 %**	(4.49)	(4.47)
Ce-141	5.847	2.8	0.923	2.02	**4.647 ± 4.73 %**	(4.38)	(4.38)
Pr-143	5.956	1.0	0.774	1.82	**3.971 ± 3.84 %**		
Ce-144	5.5	1.0	0.696	1.32	**3.296 ± 3.63 %**	(3.19)	(3.19)
Nd-147	2.247	2.0	0.886	2.17	**1.714 ± 4.38 %**	(1.62)	(1.62)
Sm-153	0.1583	6.0	1.77	3.24	**0.242 ± 7.54 %**	(0.209)	(0.209)
Eu-155	0.0321	6.0	2.915	0.73	**0.080 ± 6.85 %**	(0.0789)	(0.07883)
Eu-156	0.01485	6.0	4.78	2.43	**0.061 ± 7.24 %**	(0.0535)	(0.0514)
Gd-159	0.001009	8.0	14.9	4.00	**0.012 ± 9.51 %**		
Tb-161	0.0000853	6.0	65.7	7.48	**0.005 ± 10.12 %**	(0.0044)	(0.0041)

We define FPY by combining Eqs. (5) and (9) where $Y_{\rm ref}$ is the ^{235}U thermal yield for the j-th fission product, R_j is the R-value for the j-th nuclide as defined in Eq. (5), and Q_{99} is the Q-value for ^{99}Mo in the 14 MeV irradiation (Tables III–VI, and Table VIII). In appendix A table 13, we show the R-values measured at Los Alamos, Livermore, and DoD laboratories. In Tables IX – XI, we show the ENDF thermal FPY, and together with R-values from appendix A and the average Q-values for ^{99}Mo, we construct our recommended high-energy FPY. Uncertainties are the simple quadrature treatment of the three values (thermal FPY, ^{99}Mo Q-values, and R-value) combined to determine the FPY.

The measured 14-MeV neutron induced ^{235,238}U and ^{239}Pu fission product yields from LANL are given in Tables IX–XI, and compared with some Livermore (Nethaway) results.

Our results are very close to those we previously documented in an internal LANL report [4]. For the important ^{147}Nd 14 MeV FPY from ^{239}Pu, though, we note a significant change where our new result (FPY=1.91%) is lower than the value we previously reported (FPY=2.03%). The reason for this change is that our new evaluation of the ^{147}Nd FPY is lower because it includes a broader suite of measured R-value data from Los Alamos, Livermore, and the US Department of Defense, as shown in Table XVI. Our new 14 MeV plutonium evaluated R-value 0.912 is a weighted average of five US measurements (Table XI; see footnote too) compared with 0.962 Ref [4] (which was an average of just 2 values, a LANL measurement of 0.942 and a value of 0.982 from a "practice run" for this same measurement which we have since rejected). It is interesting that our new evaluated 14 MeV neutron on ^{239}Pu neodymium R-value, 0.912± 3.2% is consistent both with the Nethaway 1971 value (0.909) and the CEA value from Laurec (0.884), see Table XVI.

TABLE X: Measured 14-MeV neutron induced ^{238}U fission product yields from LANL. The Q_{99} value was taken as 0.956± 3.30%, based on LANL K-factor experiments. We also show for comparison two Livermore experiments by Nethaway: the 1971 results [11] were absolute, and agree well with our LANL data; the 1983 results (documented in 1993 [12])) are relative, and so we use parentheses around them since Nethaway obtained these magintitudes by normalization to Rider's yields at the time.

	Thermal ENDF FPY ^{235}U (%)	Unc. (%-relative)	Evauated R-value (this work) (no dim.)	Unc. (%)	**14 MeV FPY LANL (%)**	14-MeV FPY Nethaway'71 (%)	14-MeV FPY *(relative)* Nethaway'83 (%)
Sr-89	4.733	1.4	0.645	3.70	**2.917 ± 5.16 %**		
Sr-90	5.782	1.4	0.601	2.71	**3.318 ± 4.50 %**		
Y-91	5.828	1.0	0.696	4.88	**3.876 ± 5.97 %**		
Zr-95	6.503	2.0	0.784	2.52	**4.872 ± 4.61 %**	4.90± 5%	(4.85)
Zr-97	5.984	2.8	0.918	1.96	**5.251 ± 4.75 %**	4.98± 5%	(4.79)
Mo-99	6.109	2.0	1.000	—	**5.837 ± 3.86 %**	5.79± 5%	(5.68)
Ru-103	3.031	2.0	1.68	0.50	**4.866 ± 3.89 %**	4.66± 5%	(4.64)
Rh-105	0.964	2.8	3.64	13.21	**3.353 ± 13.90 %**		(2.87)
Ru-106	0.4016	2.0	6.88	1.70	**2.640 ± 4.22 %**		(2.47)
Ag-111	0.01738	6.0	62.4	4.29	**1.036 ± 8.08 %**		(0.982)
Pd-112	0.01304	8.0	93.9	0.56	**1.170 ± 8.67 %**		(1.034)
Cd-115	0.01158	8.0	72.8	1.55	**0.806 ± 8.79 %**	0.784± 10%	
Cd-115m	0.001005	8.0	78.1	6.61	**0.075 ± 10.89 %**	0.565 (check) ± 5%	
Sb-125	0.034	4.0	40.9	16.94	**1.329 ± 17.72 %**		(1.06)
Sb-127	0.157	6.0	14.1	0.50	**2.108 ± 6.87 %**		(1.34)
I-131	2.89	1.4	1.5	2.00	**4.142 ± 4.11 %**		
Te-132	4.295	2.0	1.16	2.28	**4.761 ± 4.49 %**	4.55± 5%	(4.52)
I-133	6.697	1.4	0.679	2.00	**4.345 ± 4.11 %**		
Cs-136	0.0055	20	5.03	1.15	**0.026 ± 20.3 %**		(0.022)
Cs-137	6.188	0.7	0.889	2.31	**5.257 ± 4.09 %**	5.19± 5%	(4.93)
Ba-140	6.215	1.4	0.790	2.64	**4.692 ± 4.46 %**	4.54± 5%	(4.63)
Ce-141	5.847	2.8	0.781	1.19	**4.361 ± 4.49 %**	4.27± 5%	(4.25)
Pr-143	5.956	1.0	0.711	2.52	**4.044 ± 4.28 %**	3.84± 5%	
Ce-144	5.5	1.0	0.723	2.23	**3.802 ± 4.11 %**	3.72± 5%	(3.75)
Nd-147	2.247	2.0	1.01	1.40	**2.176 ± 4.11 %**	2.14± 5%	(2.10)
Sm-153	0.1583	6.0	2.94	9.52	**0.444 ± 11.73 %**		(0.361)
Eu-155	0.0321	6.0	5.18	0.70	**0.1589 ± 6.89 %**		(0.158)
Eu-156	0.01485	6.0	8.73	5.49	**0.1239 ± 8.78 %**	0.107± 5%	(0.105)
Gd-159	0.001009	8.0	25.1	7.75	**0.0242 ± 11.62 %**		
Tb-161	0.0000853	6.0	104	2.92	**0.0085 ± 7.45 %**	0.00824± 10%	(0.0069)

D. Comparisons with Other Measurements and with ENDF

Figs. 2–5 compare our 14 MeV FPY data with the original ENDF/B-VI England and Rider evaluation [13] which was carried over to ENDF/B-VII.0 [6]. The figures also show comparisons against 14 MeV ENDF/B-VII.1 [3] which has been updated only for plutonium. In the case of plutonium we also show comparisons with Laurec's recently-published 14 MeV data [14] in Fig. 5, and with older data from Nethaway (1971) [11] and Bonyushkin [15].

For the uranium isotopes, the agreement between our LANL results and ENDF/B-VII.1=ENDF/B-VI is very good. This is not surprising since some of the data used to create the B-VI evaluation came from Los Alamos (Ford and Norris) – from some of the the same measurements we describe here. Furthermore, for these cases the fundamental nuclear data used to determine the number of fissions – the 14.1 MeV fission cross sections – have not changed substantially over the last few decades.

This is not the case for plutonium, where more precise fission cross section measurements over the last few decades have led to the evaluated 14.1 MeV fission cross section decreasing by 15–20%. Because of this, we assess the number of fissions in the plutonium sample to be significantly lower than Los Alamos originally assessed, and consequently our updated 14 MeV plutonium FPYs are significantly higher. This issue was documented in recent years in a report by Schecker *et al.* [4] – one of a series of reports where LANL documented its modern fission basis. The K-factors and Q-values for our ^{99}Mo reference nuclide that we describe in this paper are essentially identical to those we reported in 2005 [4] - at 14 MeV this paper reports, for ^{235,8}U and ^{239}Pu, molybdenum Q-values of 0.861, 0.956, and 0.931, to be compared with our 2005 (Schecker) values of 0.870, 0.961, and 0.937 – the very small (less than 1%) differences arising from our reanalysis of the original measurements).

We are only aware of 2 other 14 MeV plutonium ex-

TABLE XI: Measured 14-MeV neutron induced ^{239}Pu fission product yields from LANL, constructed from thermal ^{235}U FPY from ENDF and our evaluated 14 MeV ^{239}Pu R-values, and a Q_{99} value of 0.931± 3.28% which was based on an average of two LANL K-factor experiments. We also show for comparison two Livermore experiments by Nethaway: the 1971 results [11] were absolute, and agree well with our LANL data; the 1983 results (documented in 1993 [12])) are relative, and so we use parentheses around them since Nethaway obtained these magintitudes by normalization to Rider's yields at the time – they are notably smaller than the 1971 results and the LANL data.

	Thermal ENDF FPY ^{235}U (%)	Unc. (%-relative)	Evauated R-value (this work) (no dim.)	Unc. (%)	**14 MeV FPY LANL (%)**	14-MeV FPY Nethaway'71 (%)	14-MeV FPY *(relative)* Nethaway'83 (%)
Sr-89	4.733	1.4	0.488	3.19	**2.150 ± 4.8**		
Y-90	5.782	1.4	0.503	1.76	**2.707 ± 4.0**		
Y-91	5.828	1.0	0.516	0.93	**2.799 ± 3.6**		
Zr-95	6.503	2.0	0.734	2.36	**4.443 ± 4.5**	4.32 ± 5%	(3.96)
Zr-97	5.984	2.8	0.877	4.10	**4.884 ± 6.0**	4.87 ± 5%	(4.06)
Mo-99	6.109	2.0	1.000	—	**5.686 ± 3.8**	5.67 ± 5%	(4.98)
Ru-103	3.031	2.0	2.08	1.76	**5.879 ± 4.2**	5.79 ± 5%	(4.98)
Rh-105	0.964	2.8	4.72	1.90	**4.235 ± 4.7**		(3.58)
Ru-106	0.4016	2.0	12.3	1.20	**4.598 ± 4.0**		(3.86)
Pd-109	0.03122	11	94.4	1.40	**2.743 ± 11.**		
Ag-111	0.01738	6.0	117.4	2.18	**1.899 ± 7.2**		(1.67)
Pd-112	0.01304	8.0	124.1	0.74	**1.506 ± 8.7**		(1.77)
Cd-115	0.01158	8.0	130.0	5.30	**1.401 ± 10.**	1.47 ± 10%	
Cd-115m	0.001005	8.0	120.2	11.06	**0.112 ± 14.**	0.094 ± 5%	
Sb-125	0.034	4.0	81.2	2.20	**2.570 ± 5.6**		(1.65)
Sb-127	0.157	6.0	20.8	0.50	**3.039 ± 6.9**		(1.73)
I-131	2.89	1.4	2.05	0.50	**5.514 ± 3.6**		
Te-132	4.295	2.0	0.875	5.74	**3.496 ± 6.9**	3.40 ± 5%	(2.89)
I-133	6.697	1.4	0.677	2.20	**4.220 ± 4.2**		
Cs-136	0.0055	20	164.4	3.11	**0.842 ± 20.**	0.814 ± 5%	(0..814)
Cs-137	6.188	0.7	0.903	6.66	**5.201 ± 7.5**	5.32 ± 5%	(4.24)
Ba-140	6.215	1.4	0.731	2.57	**4.229 ± 4.4**	4.07 ± 5%	(3.61)
Ce-141	5.847	2.8	0.777	3.43	**4.230 ± 5.5**	4.24 ± 5%	(3.52)
Pr-143	5.956	1.0	0.595	4.71	**3.297 ± 5.8**	3.08 ± 5%	(2.94)
Ce-144	5.5	1.0	0.588	1.92	**3.011 ± 3.9**	3.03 ± 5%	(2.57)
Nd-147	2.247	2.0	0.912	3.21	**1.908 ± 5.0**	1.90 ± 5%	(1.72)
Sm-153	0.1583	6.0	3.98	1.96	**0.586 ± 7.1**		(0.472)
Eu-155	0.0321	6.0	10.4	1.40	**0.3107 ± 7.0**		(0.278)
Eu-156	0.01485	6.0	18.4	3.20	**0.2543 ± 7.6**	0.225 ± 5%	(0.194)
Gd-159	0.001009	8.0	117.0	14.00	**0.1099 ± 16.**		
Tb-161	0.0000853	6.0	312.8	20.31	**0.0248 ± 21.**	0.020 ± 10%	(0.023)

periments where absolute FPYs were determined: that of Nethaway (1971) [11] at Livermore, which used methods similar to ours to determine the number of fissions, though germanium gamma-ray detection was used to measure the FP atoms; and that of Laurec, which used a fission chamber to determine the number of fissions. We show the numerical values from the Nethaway 1971 experiment in Tables IX–XI, and the Laurec values were given in the recent Nuclear Data Sheets publication [14]. Our results agree well with Nethaway's 1971 measurements, but not with Laurec's plutonium data (which are closer to the original ENDF/B-VI evaluation) which are generally lower than our FPYs. We do not know why the discrepancy with Laurec exists. The plutonium FPY results from Nethaway's 1971 experiment [11] appear to have been revised just a few (2-4%) percent lower for reasons we do not know, when reported a year later by Nethaway and Prindle [16] – and then, as we decribe in more detail below, Nethaway made a further adjustment down by 9% owing to concerns he had on the overall fission yield normalization that should add to 200%. As we have said, our results agree well with the original reported Nethaway 1971 data [11]. Also, as we describe in the Appendix, if we update Nethaway's 1971 results to use modern more accurate ENDF/B-VII.0 data for the 14.1 MeV plutonium fission cross section and the the ^{27}Al(n,α) monitor foil cross section, the determined yields would *increase* by about 4% for 14 MeV neutrons on plutonium.

Other FPY measurements have been reported for plutonium, but they were not absolute – in the sense that the absolute number of fissions was not determined, and instead was estimated by putting a smooth curve through the relative FP yields and requiring the integrated FPY distribution be 200%. But because only a small fraction

TABLE XII: Summary of LANL measured Q, R, and FPYs for fission product experiments, for neutrons incident on ^{235,238}U and ^{239}Pu. Superscript "fs" refers to fission spectrum values we reported last year in the paper by Selby *et al.* [1] with average neutron energies 1.3-1.5 MeV (except for ^{99}Mo which is at 0.6 MeV), whilst superscript "14" refers to 14.1 MeV measured values reported in this paper. The FPYs are constructed using ENDF/B-VII.1=ENDF/B-VI thermal FPYs, as opposed to LANL in-house measured values.

FP	R^{fs}_{235U}	R^{14}_{235U}	R^{fs}_{238U}	R^{14}_{238U}	R^{fs}_{239Pu}	R^{14}_{239Pu}
Zr-95	0.967±1.8%	0.979±0.5%	0.752±2.8%	0.784±2.5%	0.727±2.9%	0.734±2.4%
Mo-99	1	1	1	1	1	1
Cs-137	1.003±1.2%	1.015±1.1%	0.931±1.4%	0.889±2.3%	1.030±1.8%	0.903±6.7%
Ba-140	0.953±0.5%	0.872±1.8%	0.908±0.5%	0.790±2.6%	0.836±1.0%	0.731±2.6%
Ce-144	0.909±2.3%	0.696±1.3%	0.800±2.7%	0.723±2.2%	0.659±2.9%	0.588±1.9%
Nd-147	0.976±0.5%	0.886±2.2%	1.162±0.6%	1.010±1.4%	0.916±0.8%	0.912±3.2%
	Q^{fs}_{235U}	Q^{14}_{235U}	Q^{fs}_{238U}	Q^{14}_{238U}	Q^{fs}_{239Pu}	Q^{14}_{239Pu}
Zr-95	0.966±2.6%	0.843±3.2%	0.771±3.6%	0.750±4.1%	0.738±3.5%	0.683±4.1%
Mo-99	1.000±1.9%	0.861±3.2%	1.025±2.2%	0.956±3.3%	1.015±2.0%	0.931±3.3%
Cs-137	1.003±2.2%	0.874±3.4%	0.954±2.6%	0.850±4.0%	1.045±2.7%	0.841±7.4%
Ba-140	0.953±2.0%	0.751±3.6%	0.931±2.3%	0.755±4.2%	0.849±2.2%	0.681±4.2%
Ce-144	0.909±3.1%	0.599±3.5%	0.820±3.3%	0.691±4.0%	0.699±3.5%	0.547±3.8%
Nd-147	0.976±2.0%	0.763±3.8%	1.191±2.3%	0.966±3.6%	0.930±2.2%	0.849±4.6%
	FPY^{fs}_{235U}	FPY^{14}_{235U}	FPY^{fs}_{238U}	FPY^{14}_{238U}	FPY^{fs}_{239Pu}	FPY^{14}_{239Pu}
Zr-95	6.30±3.0%	5.48±3.8%	5.01±3.8%	4.87±4.6%	4.80±3.8%	4.44±4.5%
Mo-99	6.11±2.4%	5.26±3.8%	6.26±2.6%	5.84±3.9%	6.20±2.4%	5.69±3.8%
Cs-137	6.20±2.3%	5.41±3.5%	5.91±2.7%	5.26±4.1%	6.47±2.7%	5.20±7.5%
Ba-140	5.91±2.2%	4.67±3.9%	5.78±2.5%	4.69±4.5%	5.27±2.4%	4.23±4.4%
Ce-144	5.00±3.2%	3.30±3.6%	4.51±3.4%	3.80±4.1%	3.68±3.6%	3.01±3.9%
Nd-147	2.19±2.4%	1.71±4.4%	2.68±2.7%	2.18±4.1%	2.09±2.6%	1.91±5.0%

of all FPs were measured, this approach has large uncertainties, often 10% or more. This was the approach used by Bonyushkin. Another approach was used by Nethaway in his 1983/1984 *relative* experiment [12]: he obtained only relative FPYs but converted them to an absolute scale by re-normalizing to some of the FPYs evaluated at the time by Rider (which could, of course, be deficient). For this reason although we provide these relative values in Tables IX–XI we do not show the Nethaway 1983/84 data in Fig. 5 (the FPY values lie lower than our LANL plutonium data); but we do use these data in the R-value evaluations as shown in Tables XIV–XVI.

V. CONCLUSIONS

Our 14 MeV ^{235,8}U FPYs are consistent with other literature values, but our ^{239}Pu FPYs tend to be higher than some previous estimates. Our values for plutonium are supported by Nethaway's 1971 Livermore experiment, but not by Laurec's CEA experiment.

One final issue is of some concern. The recent 14 MeV ENDF/B-VII.1 plutonium evaluation was strongly influenced by our data reported here, but in order to preserve the integral of 200%, some FPYs for FPs adjacent to those measured by us were assumed to be significantly lower, resulting in some spikes and discontinuities in the FPY evaluation that are not well grounded physically. Ultimately this was because the evaluator (Chadwick) placed a higher premium on matching the LANL and Nethaway-1971 plutonium FPY data than on requiring a smooth FPY distribution. Indeed, in the early 1970s Nethaway noted this same issue with his absolute plutonium 14 MeV FPY data (which agree well with ours) and at that time took a different approach [16]: he concluded that he and Prindle must have made an unidentified 9% normalization error on the 14 MeV neutron fluence assessment, and then he renormalized his plutonium 14 MeV FPYs down by this amount [16], enabling him to put a smooth FPY doubled-hump distribution through these data and match 200% for the integral. The new ENDF/B-VII.1 approach by Chadwick instead reproduces the absolute scale of the plutonium-239 FPYs reported herein, because three US experiments agreed in this absolute scale (LANL 1956, LANL 1971, and Nethaway 1971); nevertheless we still conclude that there remain significant uncertainties in the absolute magnitude of the 14 MeV plutonium FPY.

A few more words can be said on this issue. Unfortunately the field of FPY measurements has many examples where data sets from different laboratories are inconsistent, and one must always be concerned that some systematical error could be present. For ^{239}Pu in particular we have noted the contradiction between these 14 MeV LANL experiments and the CEA Laurec experiments. As we discussed earlier (at the appropriate request of a referee) one might speculate that contamination from room return thermalized neutrons could be present in these LANL experiments. If this happened, because of the large thermal fission cross section of ^{235}U and ^{239}Pu there would have been more fissions present than we estimated, and our inferred FPY would be erro-

neously biased. Two arguments would count against this hypothesis: (1) the LANL experimentalists were aware of this potential problem and would have used Cd wrappings to mitigate against the effect; and (2) if the problem occurred for ^{239}Pu it would likely have occurred for ^{235}U too, yet our results and Laurec's appear to be fairly consistent for ^{235}U. Having said this, the possibility of such a systematic error being present in the data reported here is not zero, supporting our statement at the end of the last paragraph that " there remain significant uncertainties in the absolute magnitude of the 14 MeV plutonium FPY".

The 14 MeV FPY we present here are close to the 14 MeV FP data we assessed a few years ago in an internal Los Alamos report [4], except that for the important ^{147}Nd FPY our plutonium FPY assessment has decreased by 6%-relative, from FPY=2.03% to 1.91% owing to our updated R-value for neodymium based on a broader suite of measured R-value data. This new value agrees with the absolute FPY measured value of 1.90% by Nethaway (1971), but is still significantly higher than the Livermore *evaluated* value of 1.67% [22]. We hope that future experiments, for example those being presently initiated by Becker, Wilhelmy, Vieira *et al.*[20] at TUNL, and those beginning by Tovesson, White *et al.* at LANSCE, will one day help resolve some of these remaining puzzles and provide a definitive determination of the absolute magnitude of some of the key 14 MeV FPYs for plutonium. These same experiments, together with nuclear modeling insights such as the work by Lestone [23] also in this edition of Nuclear Data Sheets, will help better understand the FPY energy dependence up to 14 MeV.

This publication completes the documentation of Los Alamos' radiochemical fission product yields in the thermal and fast [1], and 14 MeV energy ranges. For convenience we summarize these data in Table XII.

Acknowledgments

We acknowledge useful discussions with Eric Bauge, John Becker, Jerry Wilhelmy, David Vieira, John Lestone, Carol Burns, Chuck Wilkerson, and Ken Moody who also provided us with some of Livermore's internal Chemistry Department technical notes. We also note Don Barr's important contributions to this work over the years.

[1] H. D. Selby, M. R. Mac Innes, D. W. Barr, A. L. Keksis, R. A Meade, C. J. Burns, M. B. Chadwick, and T. C. Wallstrom, "Fission Product Data Measured at Los Alamos for Fission Spectrum and Thermal Neutrons on ^{239}Pu, ^{235}U and ^{238}U", Nucl. Data Sheets **111**, 2891 (2010).

[2] M. B. Chadwick, T. Kawano, D. W. Barr, M. R. Mac Innes, A. C. Kahler, T. Graves, H. Selby, C. J. Burns, W. C. Inkret, A. L. Keksis, J. P. Lestone, A. J. Sierk, and P. Talou, "Fission Product Yields from Fission Spectrum n+^{239}Pu for ENDF/B-VII.1", Nucl. Data Sheets **111**, 2923 (2010).

[3] M. B. Chadwick *et. al.*, "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields, and Decay Data", Nuclear Data Sheets **112**, 2887 (2011).

[4] J. A. Schecker, M. R. Mac Innes, D. W. Barr, and W. C. T. Inkret, "A Re-evaluation of LANL's Historic Radiochemistry Constants," LA-UR-05-3760, Los Alamos National Laboratory (2005).

[5] R. W. Spence, "Current Radiochemical Research at Los Alamos", U.S. Atomic Energy Commission Report AECD-2625 (LADC-677) (1949).

[6] M. B. Chadwick, P. Obložinský, M. Herman *et al.*, "ENDF/B-VII.0: Next Generation Nuclear Data Library for Nuclear Science and Technology," Nuclear Data Sheets, **107**, p. 2931 (2006).

[7] R.J. Prestwood and B.P. Bayhurst, "(n,2n) Excitation Functions of Several Nuclei from 12.0 to 19.8 MeV", Phys. Rev. 121, no. 5., pp 1438–1441 (1961).

[8] W. Nyer, Warren, "Fission Cross Sections of Thorium 232, Uranium 233, 235 and 236 and Plutonium 239 Relative to Uranium 238 for 14 MeV Neutrons", Los Alamos National Laboratory Report LAMS-938 (1950)

[9] G.A. Brinkman, A.H.W. Aten, J.Th. Veenboer, "Absolute standardization with a NaI(Tl) crystal-I: Calibration by means of a single nuclide", The International Journal of Applied Radiation and Isotopes, Vol. 14, March 1963, pp. 153-157; G.A. Brinkman, A.H.W. Aten, J.Th. Veenboer, "Absolute standardization with a NaI(Tl) crystal-II determination of the total efficiency", The International Journal of Applied Radiation and Isotopes, Vol. 14, October 1963, pp. 433-437.

[10] R. Gunnink. L.J. Colby, Jr, and J.W. Cobble, "Absolute Beta Standardization Using 4 Pi Beta-Gamma Coincidence Techniques", Anal. Chem. 31 (5), p 796 (1959).

[11] D. L. Nethaway to M. Mac Innes, Private communication, "Fission Product Yields at 14 MeV", Lawrence Livermore National Laboratory, October 12, (1971).

[12] D. L. Nethaway, "Fission Product Yields from FLATTOP and RTNS-2 Irradiations in 1983/1984", Lawrence Livermore National Laboratory Memorandum, Feb 2, (1993).

[13] T. R. England and B. F. Rider, "Evaluation and Compilation of Fission Product Yields," ENDF-349, LA-UR-94-3106, Los Alamos National Laboratory (1994) [available at `http://t2.lanl.gov/publications/yields/apxA.txt`].

[14] J. Laurec, A. Adam and T. de Bruyne, E. Bauge, T. Granier, J. Aupais, O. Bersillon, G. Le Petit, N. Authier, and P. Casoli, Nucl. Data Sheets **111**, 2965 (2010).

[15] E.K. Bonyushkin, Atomnaya Energia 10, 10 (1961).

[16] D. L. Nethaway and A. L. Prindle, "Evaluation of fission yields for ^{239}Pu plus 14 MeV neutrons", Lawrence Livermore National Laboratory Chemistry Department Technical Note No. 72-48, September 21 (1972). See also Note No. 72-51 for ^{235,8}U, October 25 (1972).

[17] A. D. Carlson, V. G. Pronyaev, D. L. Smith, N. M. Larson, Z.-P. Chen, G. M. Hale, F. J. Hambsch, E. V. Gai, S.-Y. Oh, S. A. Badikov, T. Kawano, H. M. Hofmann, H. Vonach, and S. Tagesen, Nuclear Data Sheets **110**, 3215 (2009).

[18] G. P. Ford and A. E. Norris, "A Compilation of Yields from Neutron-Induced Fission of ^{232}Th, ^{235}U, ^{236}U, ^{237}Np, ^{238}U and ^{239}Pu Measured Radiochemically at Los Alamos," LA-6129, Los Alamos National Laboratory (1976).

[19] B.P. Bayhurst, J.S. Gilmore, R.J. Prestwood, J.B. Wilhelmy, N. Jarmie, B.H. Eellike, and R.A. Hardcopy, "Cross Sections for (n,xn) Reactions Between 7.5 MeV and 28 MeV", Phys. Rev. C 12, no. 2, pp 451–465 (1975).

[20] J.A. Becker, D.J. Vieira, R.S. Rundberg, J.B. Wilhelmy, R. Macri, C. Hagmann, M.A. Stoyer, A. Tonchev, "Modernizing the fission basis", Lawrence Livermore National Laboratory report LLNL-PROP-246546 (2010).

[21] D.L. Nethaway to M. Mac Innes, Private communication, "Fission Product Yields at 14 MeV", (February 1993).

[22] D.L. Nethaway, "PROPHET Fission Yields", Nuclear Chemistry Division Memorandum LJW-45-85-189, Lawrence Livermore National Laboratory (1985).

[23] J.P. Lestone, "Energy dependence of plutonium fission product yields", Nuclear Data Sheets, this issue (2011).

Appendix A

As we described in our previous paper [1], Los Alamos developed a comprehensive calibration program using reactor and fast critical irradiation facilities. We also produced fission product activities for US laboratory intercalibration efforts. The most common fission product calibration used thermal neutron irradiation of ^{235}U. Irradiated foils were dissolved to create a stock solution that was distributed to US laboratories involved in the interlaboratory calibration program. These yearly reference calibrations formed the basis of comparison between laboratories and provided each laboratory an internal check on fission product measurement capabilities. In Table XIII we show results of Los Alamos and Livermore thermal fission yields derived from interlaboratory calibrations run between mid-1970 to early-1990. The atoms were measured in the respective laboratories and ratiod to the fissions in the stock solution as reported by Los Alamos using their fission-chamber calibrated β-decay resolution technique for ^{99}Mo.

Occasionally, Los Alamos prepared intercalibration solutions of fission products using actinide fissile materials irradiating with neutrons of different energies. Results of these calibrations were compared through the R-value method because these values are laboratory independent quantities because detector-specific biases cancel in the ratios used.

In the 1963–64 time frame, Los Alamos prepared solutions from foil irradiations of HEU, D-38 and plutonium using the Cockcroft-Walton high-energy neutron spectrum facility. Fission product measurements were made by Los Alamos, Livermore, and various US DoD laboratories. The HEU used in this irradiation was 93% enriched in ^{235}U. The D-38 was depleted to a $^{238/235}$U ratio of 325:1. The plutonium had a ^{240}Pu content of 5.5%. Results of this series of calibrations are presented in Tables XIV, XV, XVI, listed as LANL, LLNL, and DoD. Independently, Livermore conducted fission product research using high-energy neutron irradiations in the early 1970's and again in the early 1980's. The neutron energy of the irradiations was typically between 14.3 and 14.8 MeV depending on target foil placement. The composition of the fissile materials used by Livermore was essentially the same as the earlier Los Alamos interlaboratory calibration experiments. We used Livermore results of the yearly interlaboratory thermal calibrations where available (see table IX) to construct R-values from the reported results of Nethaway [11, 21]. For those nuclides not reported in the interlaboratory calibrations, we used the Livermore evaluated thermal FPY [22]. According to Nethaway [11], the 1971 results for plutonium were the average of nine irradiations. Some of the irradiations also used aluminum and gold flux monitors to determine high-energy FPY for ^{99}Mo. Nethaway's reported 14 MeV plutonium FPY for ^{99}Mo (FPY=5.67%, Table XI), when we correct for modern nuclear data cross sections gives FPY=5.91% (a 4% correction for plutonium, comprising of a 7.4% increase owing to updating his fission cross section of 2.59 b to the modern value of 2.412b, and a 3% decrease owing to updating his aluminum (n,alpha) monitor cross section of 125 mb to the modern ENDF/B-VII.0 value of 121.2mb). The 1971 results for ^{238}U are the average of three irradiations according to Nethaway. Absent additional information, we chose to weight the constructed R-values from the 1971 Livermore experiments as one value in the averaging process. The constructed R-value results are shown in Tables XIV, XV, XVI.

We also list R-value results reported by Ford and Norris from Los Alamos measurements made in the 1950s. The Los Alamos results reported by Ford and Norris from the 1963 – 64 (the column labeled LANL) interlaboratory calibrations have been updated by Mac Innes using improved thermal ratios to construct the R-values. The uncertainties shown in these tables reflect the precision of the measurements in the case of DoD. The values reported by Ford and Norris, Livermore, and Los Alamos included the full statistical treatment of uncertainties in the R-values for replicate measurements. We have used selected simple average results from Tables XIV, XV, XVI, to construct the average (evaluated) R-values and 14 MeV fission product yields in section V. The ^{90}Sr results for ^{235}U and ^{238}U fission is the average of two results reported by Ford and Norris. The ^{136}Cs result for ^{238}U fission is the average of three results reported by Ford and Norris.

TABLE XIII: FPY for thermal neutrons on ^{235}U (in percent). Note that the values we list below for Livermore (LLNL) are not values reported directly by LLNL, but rather values we assess based on Livermore's quoted number of measured FP atoms in the Intercal Program. The uncertainties quoted for the LANL and LLNL data do not include systemmatic uncertainties.

	LANL		LLNL		ENDF	
	FPY	Unc.(%)	FPY	Unc.(%)	FPY	Unc.(%)
Y-91			6.341	1.86	5.828	1.00
Zr-95	6.426	1.47	6.597	1.13	6.503	2.00
Zr-97	5.709	2.16	5.743	0.89	5.984	2.80
Mo-99	6.096	2.25	6.188	1.32	6.109	2.00
Ru-103	3.091	2.64	3.007	2.40	3.031	2.00
Rh-105	0.97	4.18			0.964	2.80
I-131	2.92	4.72			2.890	1.40
Te-132	4.093	3.79	4.098	4.36	4.295	2.00
Cs-137	6.25	2.16	6.146	1.51	6.188	0.70
Ba-140	6.226	1.65	6.243	1.92	6.215	1.40
Ce-141	5.834	1.84	5.864	1.29	5.847	2.80
Ce-143	6.109	2.07	5.935	1.58	5.956	1.00
Ce-144	5.76	3.48	5.589	1.48	5.500	1.00
Nd-147	2.233	2.33	2.279	1.46	2.247	2.00

TABLE XIV: 14-MeV R-values and uncertainties for ^{235}U (HEU) from various measurements, relative to ^{99}Mo. The Ford and Norris value is also a Los Alamos measurement. The values from Laurec are shown for comparison.

	LANL		LLNL		DoD		Ford & Norris		Nethaway 1983		Nethaway 1983		Laurec 2010
	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value
Sr-89	1.013	0.60	0.939	1.38	1.012	0.54	1.09	2.40					
Sr-90													
Y-91			0.962	2.08	0.961	0.81	0.99	2.50	1.102	8.02	0.984	8.02	
Zr-95			0.975	2.96	0.975	1.04			0.986	3.14	0.98	3.20	1.001
Zr-97					1.048	1.28	1.05	1.40	0.992	5.55	0.99	4.11	1.037
Ru-103							1.38	3.00	1.27	3.14	1.27	3.44	1.237
Rh-105							2.41	3.00	2.34	3.56	2.33	3.15	
Ru-106							4.86	3.00	6.05	8.00	5.23	2.40	
Pd-109													
Ag-111	74.3	0.51	74.2	1.74	77	0.36	76.3	2.90	74.4	2.74	70.4	2.34	
Pd-112					93.9	0.56	94.7	1.30	123.4	6.76	116.3	6.62	
Cd-115	99.2	0.47	98.9	0.90	101.4	0.61	101.4	1.90	103.3	2.93	98.7	2.87	
Cd-115m	98.7	2.36			96.7	0.90							
Sb-125							74.6	3.00	60.5	4.52	55.4	4.32	
Sb-127							20.6	3.00	17.3	1.10	17	1.15	13.405
I-131							1.62	2.20	1.86	1.32	1.93	1.16	1.850
Te-132							1.13	3.80	1.16	1.75	1.15	1.93	1.191
I-133									0.724	3.55	0.751	3.62	0.986
Cs-136			44.6	2.22	40.6	3.62	46	2.70	49.7	1.21	46	1.26	47.3
Cs-137			0.85	18.0	1.148	7.44			0.987	3.69	1.003	3.26	
Ba-140	0.877	0.62	0.846	1.03	0.883	1.03	0.86	1.50	0.885	3.16	0.88	3.20	0.894
Ce-141			0.905	2.56	0.949	0.85			0.92	3.14	0.917	3.14	0.929
Pr-143	0.799	0.81			0.77	0.65	0.77	80.0	0.764	3.32	0.768	3.40	0.771
Ce-144	0.69	0.74	0.684	1.47	0.708	0.61	0.69	4.50	0.701	3.54	0.702	3.58	0.699
Nd-147[a]	0.869	1.87	0.823	1.93	0.861	0.95			0.876	3.14	0.879	3.14	0.871
Sm-153			1.84	1.24	1.8	0.53			1.73	3.55	1.72	3.41	
Eu-155									2.93	2.00	2.9	2.26	
Eu-156	4.82	1.02	4.79	3.81	4.75	0.52	4.6	1.70	4.96	1.05	4.77	1.05	
Gd-159					14.9	4.00							
Tb-161	66.2	2.15			72	0.74			64.6	1.75	60.1	1.75	

[a]For ^{147}Nd, additionally, LANL Nov. 1971 measured R=0.907, 0.908, 0.901.

TABLE XV: 14-MeV *R*-values and uncertainties for ^{238}U from various measurements relative to ^{99}Mo. The Ford and Norris value is also a Los Alamos measurement. The values from Laurec are shown for comparison.

	LANL		LLNL		DoD		Ford & Norris		Nethaway 1971		Nethaway 1983		Laurec 2010
	R-value	Unc,%	*R*-value	Unc,%	*R*-value	Unc,%	*R*-value	Unc,%	*R*-value	Unc,%	*R*-value	Unc,%	*R*-value
Sr-89	0.666	0.39	0.619	2.28	0.65	1.01	0.69	2.40					
Sr-90													
Y-91			0.72	1.88	0.672	1.10					0.867	13.0	
Zr-95			0.789	3.37	0.755	0.68			0.793	1.50	0.799	0.50	0.818
Zr-97					0.898	0.83	0.94	1.40	0.924	2.00	0.911	2.10	0.947
Ru-103											1.68	0.50	1.668
Rh-105							3.98	3.00			3.3	2.00	3.898
Ru-106											6.88	1.70	
Ag-111	61.8	0.40	62.1	4.32	63.6	1.15	65.8	2.90			58.5	3.30	
Pd-112					72.7	2.39					94.4	5.30	
Cd-115	73.5	0.43	72.3	1.05	71.2	2.25	74	1.90	72	1.50	73.8	2.00	
Cd-115m	81.7	1.44			74.4	1.12				1.50			
Sb-125							36	3.00			45.8	5.50	
Sb-127							14.1	3.00			14	0.50	9.178
I-131											1.5	2.00	1.523
Te-132							1.14	3.80	1.19	1.60	1.15	0.50	1.191
I-133											0.679	2.00	0.877
Cs-136					3.43	9.00	5.1	2.70			4.06	5.00	
Cs-137					0.782	8.30			0.903	1.50	0.856	2.90	
Ba-140	0.813	0.49	0.764	2.75	0.787	1.05	0.79	1.50	0.777	2.10	0.809	0.70	0.791
Ce-141			0.769	2.60	0.789	1.06			0.777	1.70	0.787	0.50	0.802
Pr-143	0.735	1.86			0.706	1.16			0.692	1.80	0.709	0.90	0.709
Ce-144	0.726	0.90	0.705	1.69	0.717	1.12	0.75	4.50	0.711	3.30	0.731	1.50	0.724
Nd-147[a]	1.03	1.84	0.969	1.22	1.02	0.88			1.002	2.10	1.001	0.50	0.981
Sm-153			3.15	1.12	3.04	1.41					2.62	1.20	
Eu-155											5.18	0.70	
Eu-156	8.9	1.24	9.55	3.71	8.71	0.75	8.1	1.70	8.57	1.40	8.57	0.50	
Gd-159					25.1	7.75							
Tb-161	101	1.80			107	0.97			104.8	6.00	89.6	1.50	

[a]For ^{147}Nd, additionally, LANL Nov. 1971 measured R=1.121.

TABLE XVI: 14-MeV R-values and uncertainties for ^{239}Pu from various measurements relative to ^{99}Mo. The Ford and Norris value is also a Los Alamos measurement. The values from Laurec are shown for comparison.

	LANL		DoD		Ford & Norris		Nethaway 1971		Nethaway 1983		Laurec 2010
	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value	Unc,%	R-value
Sr-89	0.499	0.75	0.477	0.57							
Y-90			0.503	1.76							
Y-91			0.516	0.93							
Zr-95			0.744	0.58			0.714	3.00	0.744	0.50	0.744
Zr-97			0.896	0.79	0.833	1.40	0.923	1.30	0.881	4.10	0.910
Ru-103							2.11	2.50	2.06	0.50	2.101
Rh-105									4.72	1.90	5.631
Ru-106									12.3	1.20	
Pd-109											
Ag-111	120.4	0.76	118	0.59	117.1	2.90			114.2	1.30	
Pd-112			124.7	0.75	123.4	1.30			184.5	2.50	
Cd-115	132.6	1.02	97.7	5.20	121.8	1.90	137.9	3.80	127.6	1.30	
Cd-115m			110.8	3.70			129.6	4.20			
Sb-125									81.2	2.20	
Sb-127									20.8	0.50	16.537
I-131									2.05	0.50	1.952
Te-132							0.91	3.90	0.839	0.50	0.908
I-133									0.677	2.20	0.774
Cs-136	163.1	1.64	162.3	1.55			160.4	1.10	171.9	0.50	175.1
Cs-137			1.9	11.0			0.945	5.32	0.843	2.10	
Ba-140	0.754	0.81	0.738	0.68			0.712	0.96	0.72	0.50	0.700
Ce-141			0.797	0.68			0.788	1.10	0.747	0.70	0.760
Pr-143			0.596	1.18			0.566	3.20	0.622	1.10	0.602
Ce-144	0.598	0.86	0.592	0.82			0.591	1.00	0.572	1.60	0.576
Nd-147[a]	0.942	1.92	0.877	0.62			0.909	3.00	0.942	0.50	0.884
Sm-153			4.03	0.69					3.92	2.80	
Eu-155									10.4	1.40	
Eu-156	19.2	1.36	17.8	1.13			18.4	0.93	18.2	0.70	
Gd-159			117	14.0							
Tb-161	261.1	2.38	389	0.89			259.7	2.00	341.3	1.00	

[a]For ^{147}Nd, additionally, LANL Nov. 1971 measured R=0.892.