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FAST REACTOR FISSION YIELDS FOR ^{241}Pu AND RELATIVE FISSION PRODUCT ISOTOPIC DATA FOR ^{239}Pu IRRADIATED IN ROW-4 OF EBR-II

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ABSTRACT

Absolute ^{241}Pu fast reactor fission yields are given for the stable and long-lived isotopes of rubidium, zirconium, molybdenum, xenon, cesium, barium, cerium, neodymium, and samarium, for a sample irradiated in Row-4 of the Experimental Breeder Reactor-II. The principal measurement technique for the fission product isotopes was isotope dilution mass spectrometry. The number of fissions was established by summing the number of fission product atoms in the heavy mass peak. Relative fission product isotopic data are given for a sample of ^{239}Pu also irradiated in Row-4 of the Experimental Breeder Reactor-II.

SUMMARY

Absolute ^{241}Pu fast reactor fission yields are reported for the stable and long-lived isotopes of rubidium, zirconium, molybdenum, xenon, cesium, barium, cerium, neodymium, and samarium. The results are from one sample of ^{241}Pu irradiated to 8 atom percent fission in Row-4 of the Experimental Breeder Reactor-II. The principal measurement technique for the fission product isotopes was isotope dilution mass spectrometry. The number of fissions was established by summing the number of fission product atoms in the heavy mass peak. In general, the uncertainty associated with the measured values is ~1%, relative. Previously, we reported yield data for samples irradiated to low burnup, 1 atom percent fission, in a softer neutron spectrum in Row-8 of EBR-II.

Also given are relative fission product isotopic data for a sample of ^{239}Pu irradiated in Row-4 of the Experimental Breeder Reactor-II.

For both ^{241}Pu and ^{239}Pu, detailed listing of the measured isotopic composition of the individual fission product elements is given and compared to that for samples of ^{241}Pu and ^{239}Pu irradiated in Row-8 of the Experimental Breeder Reactor-II. Small changes in the fission product isotopic composition are evident, especially for those elements on the wings of the mass yield curve where a broadening of the mass yield curve occurs as the energy of the fissioning neutron increases.

These new data will be used to augment the previous data in our continuing study of fission yields versus neutron energy.

CONTRIBUTING PERSONNEL

The wide scope of this project required contributions from a number of scientists and technicians with specialized disciplines. Individuals providing special skills for the successful completion of the project are listed below.

Chemistry

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Statistics

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Typing

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I. INTRODUCTION

For several years personnel at the Idaho Chemical Processing Plant have been involved in a program to accurately measure absolute fast reactor fission yields for many heavy element nuclides and to develop and recommend reliable methods for the accurate determination of burnup in a variety of fast reactor fuels. To obtain fission yield data characteristic of the neutron spectrum of a large mixed-oxide fueled LMFBR, samples of many heavy element nuclides were prepared and irradiated in Row-8 of the Experimental Breeder Reactor-II (EBR-II). Detailed reports [1,2,3,4] were issued giving fast reactor fission yield values for ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu. For all samples, the basic measurement technique was isotope dilution mass spectrometry, and the number of fissions was determined by summing the number of fission product atoms in the heavy mass peak.

In a companion program, experimenters at the Argonne National Laboratory (ANL) near Chicago prepared samples of several heavy element nuclides for irradiation in Row-2 of EBR-II. The purpose of that program was to provide samples for the measurement of fast reactor fission yields and to study the transmutation of heavy element nuclides in a fast reactor spectrum. The basic difference between the two irradiation programs involves the neutron spectrum and the degree of burnup: the ICPP samples were irradiated to $\sim$1 atom percent fission in Row-8 of EBR-II; the ANL samples were irradiated to 8-15 atom percent fission near the core center of EBR-II.

Following discharge of the ANL samples from EBR-II, funding limitations were imposed on the ANL program; consequently, only a few of the samples were dissolved and analyzed. The laboratory at ICPP collaborated in some of the analytical measurements by performing a significant fraction of the isotope dilution mass spectrometric measurements, but no formal reports were issued by either laboratory. Eventually, all funding for the ANL program was terminated, and a large fraction of the samples remained undissolved and unanalyzed.

In 1978, permission was granted to the ICPP Laboratory by USDOE to proceed with the dissolution and analysis of a selected number of the ANL samples to augment the yield data obtained from the ICPP samples irradiated in Row-8 of EBR-II. This report gives fast reactor yield data for one sample of ^{241}Pu, and relative fission product isotopic data for one sample of ^{239}Pu irradiated by ANL near the core center of EBR-II. Fission yield data for samples of ^{233}U and ^{235}U irradiated in Row-4 have previously been reported.[5]

For the present, we are not including a comparison of the current fission yield data relative to the ICPP Row-8 data. The comparison of fast reactor fission yield data and the correlation of the yields with neutron energy is the subject of a separate report to be issued in the fall of 1980. We have included, however, a comparison of the relative isotopic data for the Row-4 and Row-8 irradiation.

II. IRRADIATION PACKAGE AND HISTORY

The plutonium isotopes used in this experiment were chemically purified by precipitation from solution using ammonium hydroxide followed by ignition for 2h at 900°C. Approximately 37 mg of $^{241}PuO_2$ and 120mg ^{239}Pu were placed in individual high purity nickel capsules, a precision fitted lid inserted, and the capsules sealed by welding. The cover gas was air. The individual nickel capsules were leak-checked by subjecting the sealed container to 500 psi helium gas pressure for 3h and then immersing the individual containers in water at 80°C. Leaks were indicated by the presence of bubbles on the container surface and such capsules were discarded.

The nickel capsules were 2.86cm long X 0.91cm OD; they had a wall thickness of 0.06cm and a weight of ~7.5g. The sample containing nickel capsules were placed in two different sample baskets; these baskets were loaded into a stainless steel inner-liner, and the liner was subsequently loaded into an outer stainless steel tube. Sodium bonding was used between the nickel capsules and inner-liner and also between the inner

and outer-liner. One of the sample baskets was placed at the reactor mid-plane and the other in the lower axial blanket of EBR-II. The sub-assembly was a standard EBR-II B-7 unit.

The nickel capsule identification numbers for those samples located at the reactor mid-plane, weight of the target material, and the isotopic composition is given below:

Sample No.	Isotope	PuO_2 wt. grams	Atom % ^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
14	^{241}Pu	0.0372	1.03	3.66	94.04	1.27
94	^{239}Pu	0.1199	99.38	0.61	0.009	--

Initially, the irradiation was to be conducted for 4Y in Row-2 of EBR-II to give atom percent fission values of 10-20%. After 2Y of irradiation in Row-2, the irradiation pin was discharged from the reactor and reloaded into another sub-assembly for continued irradiation; but, in a Row-4 position. During the Row-2 segment of the irradiation, the sample pin was located in the center of a 7-pin sub-assembly in which the surrounding pins all contained material for structural damage studies; this softened the neutron spectrum compared to a Row-2 fuel assembly. In the Row-4 irradiation, the sample pin was located in a corner position closest to core center. Although no spectrum monitors were included in the irradiation pin, we assumed that because of the structural samples in the Row-2 position, the average neutron spectrum during the irradiation approximated that of a Row-4 position. Hence, we refer to the overall irradiation as a Row-4 irradiation.

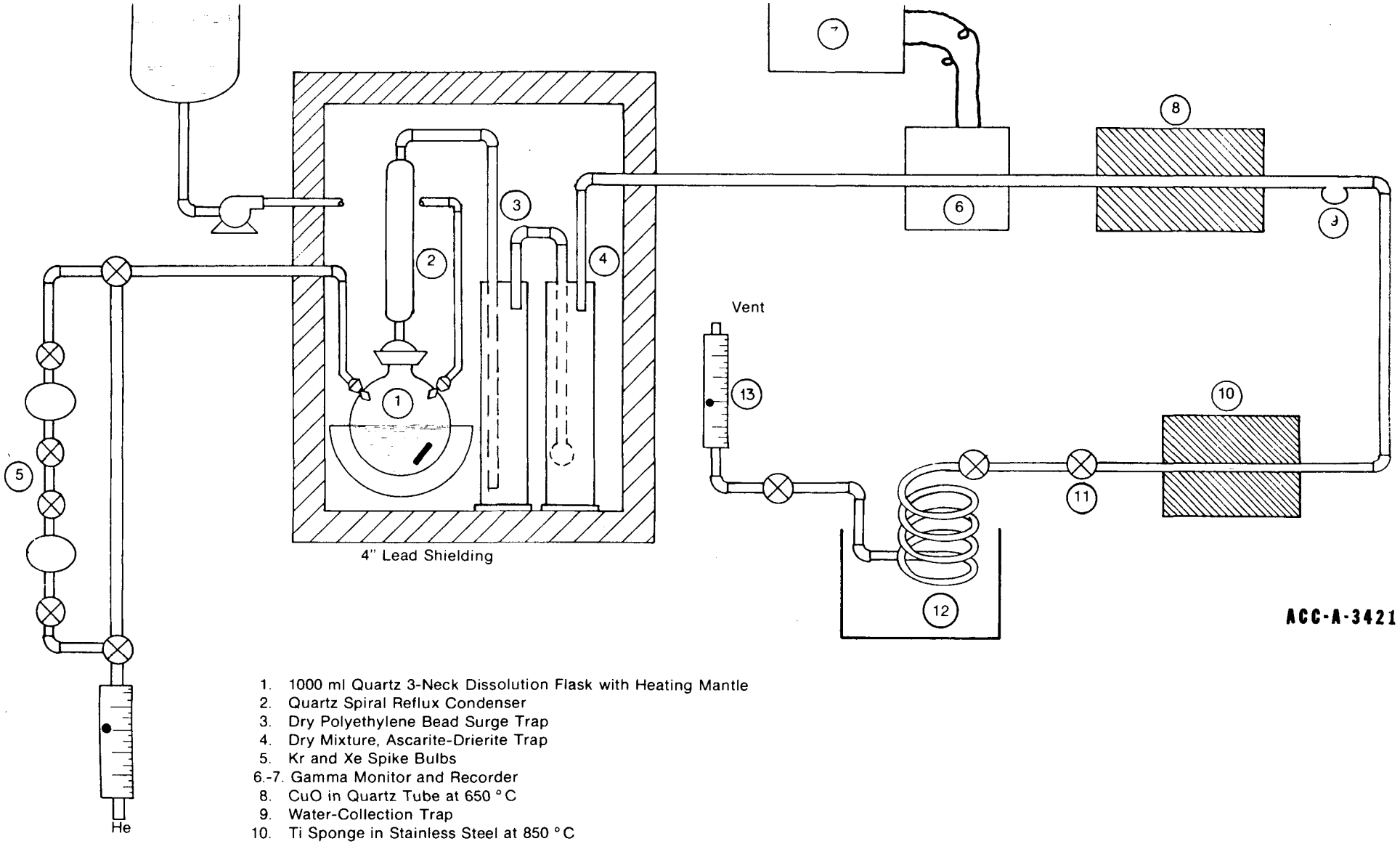
Specifically, the samples were irradiated in Row-2 from 10 December 1966 to 2 January 1969, during which time the reactor was operating 374 days and down 381 days. The estimated flux was 2.3×10^{15} n/cm^2/sec. The samples were then transferred to the Row-4 position and irradiated from 26 April 1969 to 9 September 1970, during which time the reactor operated 270 days and was down 345 days. The average estimated flux was 2.2×10^{15} n/cm^2/sec and the nominal power level $\sim$50 Mw.

III. SAMPLE DISSOLUTION AND ANALYSIS

The ^{241}Pu sample was dissolved first by using a mixture of 8M HNO_3-0.1M HF, then by adding small increments of 8M HBr in a quartz flask equipped with a quartz reflux condenser. A schematic diagram of the dissolution and gas collection apparatus is shown in Figure 1. Initially, the sample to be dissolved was placed in the quartz dissolver containing 50 mL of quartz-distilled water, and the entire system was purged with He. After purging, a slight positive pressure was placed on the system and all joints were monitored with a He leak detector to establish system integrity. The dissolving acid was added in two 100 mL increments and the temperature of the dissolver was increased. When the clad had been violated, as evidenced by the detection of ^{85}Kr activity, measured quantities of $^{78-80}Kr$ and ^{129}Xe isotopic spikes were introduced into the dissolution flask from the spike-addition manifold by switching the helium flow. The inert gases were swept from the dissolver flask and through the collection train (Figure 1) with helium at a flow of $\sim$100 cc/min during the entire dissolution.

Traps of polyethylene beads and Ascarite-Drierite removed spray, water vapor and some acidic gases. Hydrogen and organic vapors were removed on CuO at 650°C, and NO_x on titanium sponge at 850°C. The Kr-Xe fission gases and respective spikes were trapped on 5-A Molecular Sieve traps in liquid nitrogen. Two collection traps were used. Gas collection was continued for at least one hour after the dissolution was considered complete, as evidenced by cessation of the evolution of ^{85}Kr from the dissolver flask and by visual examination of the dissolver flask. The dissolution time was $\sim$24 hrs.

After dissolution, the two collection traps were removed and counted for ^{85}Kr. No ^{85}Kr activity was detected on the backup trap. The gases were removed from the primary collection trap and, the Kr and Xe separated using gas chromatography. The individual fission gases and respective spikes were collected upon exiting from the chromatograph and analyzed by mass spectrometry.



1. 1000 ml Quartz 3-Neck Dissolution Flask with Heating Mantle
2. Quartz Spiral Reflux Condenser
3. Dry Polyethylene Bead Surge Trap
4. Dry Mixture, Ascarite-Drierite Trap
5. Kr and Xe Spike Bulbs

6.-7. Gamma Monitor and Recorder

8. CuO in Quartz Tube at 650 °C
9. Water-Collection Trap
10. Ti Sponge in Stainless Steel at 850 °C
11. By-Pass Valve
12. Molecular Sieve 5A at - 190 °C (2 Traps)
13. Outlet Flowmeter and Vent

Fig. 1 Dissolution and Gas Collection Apparatus.

Following dissolution, the sample solution was allowed to stand several hours, then filtered and weighed. The filter was counted to determine completeness of dissolution. Weighed aliquots of the sample solution were removed and mixed with weighed quantities of the respective spike for each fission product element measured. The principal measurement technique was isotope dilution mass spectrometry. Usually, four spiked and four unspiked aliquots were analyzed for each element. Highly purified reagents were used in the chemical separations. A brief description of the chemical separation and mass spectrometric procedures used was previously reported.[2] The results for the ^{241}Pu sample are given in Section V of this report.

A similar dissolution scheme was used for the ^{239}Pu sample, but incomplete dissolution was obtained. Dissolution was continued using 8M HBr, but a small amount of residue still remained. Because several separate fractions of the dissolved sample resulted from this action, and because complete gas collection could not be assured; it was decided to only report relative fission product isotopic data for this sample.

IV. DETERMINATION OF NUMBER OF FISSIONS

The absolute number of fissions for the ^{241}Pu sample was determined using the summation technique, which is based on the fact that the sum of all of the fission product atoms in one of the peaks in the mass yield curve is equal to the total number of fissions. The preferred peak for measurement is the heavy mass peak, because the elements in this peak are more easily measured, and an accurate measurement of a larger fraction of this peak is possible. Accordingly, in this report, all yields are based on the sum of the atoms in the heavy mass peak.

Normally, it is not possible to measure all of the isotopes in the heavy peak, and some values have to be obtained by interpolation and estimation. For the mass region 125 to 130, the values were estimated assuming the shape of the mass yield curve to be similar to that for ^{241}Pu thermal fission, but with a more shallow valley.

For the region of 124 to one-half the mass of the compound nucleus less neutron emission, assumptions similar to that for the region 130 to 125 were used. The values for mass 139 and 141 are based on a linear interpolation between the measured number of atoms for mass 138 and 140, and 140 and 142, respectively. The atoms of mass 153 were estimated by plotting a smooth function through the measured values from mass 143 to 154. Extending this curve from mass 155 to 160 was the basis for the estimated number of atoms in this region of the mass yield curve. Liberal error estimates were assigned to all of the unmeasured values.

For the ^{241}Pu sample, 81.96% of the atoms in the heavy mass peak was measured. The actual measured, interpolated, and estimated number of atoms, and associated uncertainties are given in Table I. The relatively small error for the yields is the result of only analyzing one sample.

V. ^{241}Pu FAST REACTOR FISSION YIELDS AND ISOTOPIC COMPOSITION OF THE FISSION PRODUCT ELEMENTS

One sample of irradiated ^{241}Pu was dissolved and analyzed. The measured and estimated number of atoms for each individual fission product nuclide and the total number of fissions based on the sum of the number of the fission product atoms in the heavy mass peak is given in Table I. Also given is the fission yield for each nuclide obtained by dividing the measured or estimated number of atoms of that nuclide by the sum of the atoms in the heavy mass peak. The uncertainty estimates are based on propagating all known random and systematic errors.

The methods used to establish the estimated number of atoms for the unmeasured mass numbers in the heavy mass peak were detailed in Section IV.

In the light mass peak, no data are given for the isotopes of krypton and strontium because the amount of the natural element present was sufficiently large to preclude calculating a reliable natural correction factor. The value for mass 85 is based on the measured amount of ^{85}Rb corrected for the branching fraction (0.217) of ^{85m}Kr and decay of ^{85}Kr (10.76 yr). No

fission yield values are given for the ruthenium isotopes because a significant fraction of ruthenium plated-out on the wall of the dissolution flask and no reliable estimate of the fraction of the total ruthenium in solution could be made. The isotopic data given in Table II should, however, be valid as, no isotopic fractionation in the plate-out process is expected. The value for mass 99 was obtained by using linear interpolation between the adjacent measured masses (i.e., atoms (98+100)/2).

For the heavy mass peak, the following corrections were applied to the raw data: when ^{134}Ba and/or ^{134}Cs were detected in measurable quantities it was assumed to have resulted from neutron capture on ^{133}Cs rather than from independent fission and was added to the measured atoms of ^{133}Cs; in all cases, this correction was less than 1%.

All of the isotopes of neodymium were corrected for burnin and burnout by neutron capture using the following fast reactor capture cross section values:

^{141}Pr σc = 0.080b
^{143}Nd σc = 0.280b
^{144}Nd σc = 0.075b
^{145}Nd σc = 0.300b
^{146}Nd σc = 0.070n
^{148}Nd σc = 0.120b
^{150}Nd σc = 0.110b

The largest correction was to decrease the measured atoms of ^{146}Nd by 1.3%. All other corrections ranged from 0.5-1.0%.

Corrections to the samarium data were as follows: (1) the measured amounts of ^{147}Sm, ^{149}Sm, and ^{151}Sm were corrected for the calculated amounts of ^{146}Nd, ^{148}Nd, and ^{150}Nd burnout. (2) The measured amount of ^{148}Sm was added to ^{147}Sm and the measured atoms of ^{150}Sm were added to the measured atoms of ^{149}Sm; in the latter case, this amounted to increasing the atoms of ^{149}Sm, 3%.

The isotopic composition for each of the measured fission product elements for ^{241}Pu fast reactor fission is given in Table II. In all cases, except for xenon, the values are based on four individual unspiked analyses. The listed uncertainty for each isotopic abundance is the standard deviation of the mean for the multiple measurements and is only associated with the random error of the measurements. The results from this experiment are compared to those obtained from the Row-8 irradiation[2] in Table IV.

We attach particular importance to the unspiked isotopic data because it is more precise and accurate than fission yield data. It is especially important when comparing current results to other reported data, because for some elements the change in relative isotopic abundances can be directly correlated with neutron energy. In addition, we believe that evaluated or compiled fission yield data files must present relative yield values which closely agree with the unspiked isotopic composition of a given fission product element.

VI. FISSION PRODUCT ISOTOPIC DATA FOR ^{239}Pu FAST REACTOR FISSION

Fission yield data are not reported for the ^{239}Pu sample irradiated in Row-4 of EBR-II, because of unfortunate problems encountered in the dissolution.

Becasue there was no apparent evidence that isotopic fractionation of the fission products occurred during the dissolution, the portion of the dissolved sample containing the bulk of the fission products was analyzed to establish the relative isotopic composition of the fission products for the Row-4 irradiation. The results are given in Table III and compared to the relative isotopic data[2] for samples irradiated in Row-8 of EBR-II in Table V. The observations of this comparison are discussed in Section VII.

TABLE I

MEASURED ATOMS AND FAST FISSION YIELDS FOR ^{241}Pu

CAPSULE 14

EBR-II, Row-4

HEAVY MASS PEAK

Mass No.	Atoms X 10^{17}	Fission Yield	Standard Deviation
120-124	0.500±0.125(a)	0.585	0.146
125	0.155±0.039(a)	0.181	0.045
126	0.275±0.069(a)	0.321	0.080
127	0.470±0.118(a)	0.549	0.137
128	0.780±0.156(a)	0.912	0.181
129	1.210±0.182(a)	1.414	0.210
130	1.870±0.281(a)	2.186	0.322
131-Xe	2.732±0.013	3.194	0.033
132-Xe	3.941±0.014	4.607	0.046
133-Cs	5.604±0.025	6.551	0.066
134-Xe	6.421±0.013	7.506	0.071
135-Cs	6.152±0.019	7.192	0.070
136-Xe	5.859±0.024	6.850	0.069
137-Cs+Ba	5.445±0.076	6.365	0.099
138-Ba	5.129±0.318	5.996	0.348
139	4.827±0.338(a)	5.643	0.376
140-Ce	4.524±0.075	5.289	0.093
141	4.236±0.212(a)	4.952	0.240
142-Ce	3.948±0.065	4.616	0.081
143-Nd	3.989±0.083	4.663	0.090
144-Ce+Nd	3.564±0.074	4.167	0.080
145-Nd	2.852±0.059	3.335	0.064
146-Nd	2.359±0.049	2.758	0.053
147-Sm	1.895±0.016	2.215	0.027
148-Nd	1.675±0.035	1.958	0.038
149-Sm	1.236±0.006	1.445	0.015
150-Nd	1.057±0.023	1.235	0.025
151-Sm	0.775±0.009	0.906	0.014
152-Sm	0.628±0.004	0.735	0.008
153	0.476±0.047(a)	0.556	0.055
154-Sm	0.323±0.003	0.378	0.005
155-160	0.633±0.159(a)	0.740	0.184
SUM	85.540±0.802		
Measured	70.108±0.536 (81.96%)		
Estimated	15.432±0.598 (18.04%)		

(a) Estimated Atoms

TABLE I (Cont'd)

MEASURED ATOMS AND FAST FISSION YIELDS FOR ^{241}Pu

CAPSULE 14

EBR-II, Row-4

LIGHT MASS PEAK

Mass No.	Atoms X 10^{17}	Fission Yield	Standard Deviation
85-Rb	0.384±0.001	0.448	0.004
87-Rb	0.688±0.002	0.805	0.008
90-Sr+Zr	1.331±0.015	1.556	0.022
91-Zr	1.653±0.023	1.932	0.032
92-Zr	2.053±0.029	2.400	0.040
93-Zr	2.558±0.034	2.990	0.048
94-Zr	2.831±0.039	3.309	0.055
95-Mo	3.243±0.025	3.791	0.046
96-Zr	3.679±0.049	4.301	0.070
97-Mo	3.851±0.021	4.502	0.049
98-Mo	4.118±0.022	4.815	0.052
99	4.619±0.231(a)	5.399	0.275
100-Mo	5.119±0.032	5.984	0.067

(a) Estimated Atoms

TABLE II

RELATIVE ISOTOPIC COMPOSITION OF THE FISSION PRODUCT ELEMENTS FOR ^{241}Pu IRRADIATED IN ROW-4 OF EBR-II

Rubidium	
85	0.3578±0.0005
87	0.6422±0.0004
Zirconium	
91	0.1294±0.0004
92	0.1607±0.0008
93	0.2003±0.0007
94	0.2215±0.0013
96	0.2880±0.0006
Molybdenum	
95	0.1983±0.0008
97	0.2359±0.0002
98	0.2523±0.0003
100	0.3136±0.0005
Ruthenium	
101	0.3168±0.0007
102	0.3354±0.0004
104	0.3466±0.0005
Xenon	
131	0.1441±0.0003
132	0.2078±0.0002
134	0.3386±0.0004
136	0.3088±0.0005
Cesium	
133	0.3270±0.0004
135	0.3590±0.0001
137	0.3140±0.0001
Cerium	
140	0.5340±0.0010
142	0.4660±0.0010
Neodymium*	
143	0.2574±0.0002
144	0.2300±0.0002
145	0.1841±0.0002
146	0.1522±0.0001
148	0.1081±0.0001
150	0.0682±0.0002
Samarium*	
147	0.3901±0.0010
149	0.2544±0.0005
151	0.1595±0.0016
152	0.1294±0.0004
154	0.0665±0.0002

*Corrected for neutron capture reactions.

TABLE III

RELATIVE ISOTOPIC COMPOSITION OF THE FISSION PRODUCT ELEMENTS FOR ^{239}Pu IRRADIATED IN ROW-4 OF EBR-II

Krypton	
83	0.1970±0.0004
84	0.3106±0.0004
86	0.4923±0.0007
Rubidium	
85	0.3659±0.0004
87	0.6341±0.0004
Strontium	
88	0.3928±0.0006
90	0.6072±0.0011
Zironium	
91	0.1399±0.0004
92	0.1642±0.0008
93	0.2108±0.0006
94	0.2250±0.0005
96	0.2603±0.0008
Molybdenum	
95	0.2034±0.0001
97	0.2412±0.0001
98	0.2551±0.0001
100	0.2952±0.0004
Ruthenium	
101	0.3374±0.0002
102	0.3359±0.0002
104	0.3266±0.0004
Xenon	
131	0.1646±0.0003
132	0.2259±0.0004
134	0.3118±0.0004
136	0.2976±0.0004
Cesium	
133	0.3301±0.0002
135	0.3580±0.0001
137	0.3118±0.0002
Cerium	
140	0.5365±0.0006
142	0.4635±0.0006
Neodymium*	
143	0.2687±0.0003
144	0.2284±0.0001
145	0.1865±0.0001
146	0.1515±0.0001
148	0.1030±0.0001
150	0.0618±0.0001
Samarium*	
147	0.4085±0.0004
149	0.2536±0.0005
151	0.1547±0.0001
152	0.1267±0.0001
154	0.0566±0.0001

*Corrected for neutron capture reactions.

These isotopic values will be of value in evaluating the change in the fission product isotopic composition as a function of neutron energy, and also to provide an aid in developing a function which may describe the change in fission yields as a function of neutron energy.

VII. COMPARISON OF FISSION PRODUCT RELATIVE ISOTOPIC ABUNDANCES

One of the initial objectives of the ANL high burnup irradiation was to obtain data relative to the transmutation of heavy element and fission product nuclides under prolonged exposure to a fast neutron flux. To assess the possibility of the existance of any large fast reactor neutron capture cross sections in any of the fission product decay chains, the relative isotopic abundances of the major fission product elements for ^{233}U and ^{235}fast fission previously reported,[5] were examined. A comparison of the relative fission product isotopic abundances for 1^{a}/oF and 15^{a}/oF for ^{233}U fast fission and 1^{a}/oF and 10^{a}/oF for ^{235}U fast fission, show no evidence for any large unknown fast reactor fission product capture cross sections.

One important fact which must be kept in mind when comparing the data in Tables IV and V is that the data are for different neutron spectra; one for an irradiation in Row-4 with a mean neutron energy of ∿700keV, and the other in Row-8 with a mean neutron energy of ∿400keV. Also the samples irradiated in Row-4 were taken to high burnup levels, 8-15^{a}/oF, while the samples in Row-8 were only irradiated to ∿1^{a}/oF. In our estimation, some of the small differences in the relative isotopic abundances for those elements on the wings of each mass peak where the neutron energy effect is known to be most pronounced, are due primarily to the difference in the neutron spectra. This is especially evident for the heavy isotopes of neodymium and samarium on the wing of the heavy mass peak and for the light isotope of rubidium in ^{241}Pu fission on the wing of the light mass peak. The increase in the relative abundance of ^{131}Xe for the ^{241}Pu sample irradiated in Row-4 probably is the result of an

TABLE IV

COMPARISON OF FISSION PRODUCT RELATIVE ISOTOPIC ABUNDANCES FOR SAMPLES OF ^{241}Pu IRRADIATED IN ROW-4 AND ROW-8 OF EBR-II

	Row-4	Row-8
Krypton		
83	a	0.1752±0.0003
84	a	0.3078±0.0006
86	a	0.5169±0.0002
Rubidium		
85	0.3578±0.0005	0.3456±0.0001
87	0.6422±0.0004	0.6544±0.0001
Strontium		
88	a	0.3864±0.0006
90	a	0.6136±0.0006
Zirconium		
91	0.1294±0.0004	0.1264±0.0011
92	0.1607±0.0008	0.1559±0.0007
93	0.2003±0.0007	0.1981±0.0012
94	0.2215±0.0013	0.2274±0.0004
96	0.2880±0.0006	0.2922±0.0013
Molybdenum		
95	0.1983±0.0008	0.1979±0.0001
97	0.2359±0.0002	0.2349±0.0002
98	0.2523±0.0003	0.2506±0.0001
100	0.3136±0.0005	0.3166±0.0003
Ruthenium		
101	0.3168±0.0007	0.3139±0.0002
102	0.3354±0.0004	0.3324±0.0007
104	0.3466±0.0005	0.3537±0.0003
Xexnon		
131	0.1441±0.0003	0.1398±0.0006
132	0.2078±0.0002	0.2041±0.0005
134	0.3386±0.0004	0.3465±0.0006
136	0.3088±0.0005	0.3096±0.0005
Cesium		
133	0.3270±0.0004	0.3258±0.0003
135	0.3590±0.0001	0.3557±0.0002
137	0.3140±0.0001	0.3185±0.0001
Cerium		
140	0.5340±0.0010	0.5351±0.0012
142	0.4660±0.0010	0.4649±0.0012
Neodymium		
143	0.2574±0.0002	0.2568±0.0001
144	0.2300±0.0002	0.2346±0.0001
145	0.1841±0.0002	0.1828±0.0001
146	0.1522±0.0001	0.1528±0.0001
148	0.1081±0.0002	0.1066±0.0001
150	0.0682±0.0002	0.0664±0.0001
Samarium		
147	0.3901±0.0010	0.3942±0.0005
149	0.2544±0.0005	0.2560±0.0004
151	0.1595±0.0016	0.1607±0.0004
152	0.1294±0.0004	0.1240±0.0001
154	0.0665±0.0002	0.0650±0.0005

a) No data obtained.

TABLE V

COMPARISON OF FISSION PRODUCT RELATIVE ISOTOPIC ABUNDANCES FOR SAMPLES OF ^{239}Pu IRRADIATED IN ROW-4 AND ROW-8 OF EBR-II

	Row-4	Row-8
Krypton		
83	0.1970±0.0004	0.1963±0.0001
84	0.3106±0.0004	0.3114±0.0006
86	0.4923±0.0007	0.4923±0.0006
Rubidium		
85	0.3659±0.0004	0.3656±0.0006
87	0.6341±0.0004	0.6344±0.0006
Strontium		
88	0.3928±0.0006	0.3947±0.0004
90	0.6072±0.0011	0.6053±0.0004
Zirconium		
91	0.1399±0.0004	0.1345±0.0003
92	0.1642±0.0008	0.1646±0.0009
93	0.2108±0.0006	0.2060±0.0003
94	0.2250±0.0005	0.2321±0.0005
96	0.2603±0.0008	0.2628±0.0002
Molybdenum		
95	0.2034±0.0001	0.2104±0.0005
97	0.2412±0.0001	0.2389±0.0003
98	0.2551±0.0001	0.2533±0.0003
100	0.2952±0.0004	0.2974±0.0007
Ruthenium		
101	0.3374±0.0002	0.3317±0.0004
102	0.3359±0.0002	0.3369±0.0003
104	0.3266±0.0004	0.3314±0.0008
Xexon		
131	0.1646±0.0003	0.1641±0.0001
132	0.2259±0.0004	0.2256±0.0004
134	0.3118±0.0004	0.3158±0.0003
136	0.2976±0.0004	0.2940±0.0003
Cesium		
133	0.3301±0.0002	0.3306±0.0005
135	0.3580±0.0001	0.3575±0.0001
137	0.3118±0.0002	0.3119±0.0005
Cerium		
140	0.5365±0.0006	0.5275±0.0003
142	0.4635±0.0006	0.4725±0.0004
Neodymium		
143	0.2687±0.0003	0.2713±0.0002
144	0.2284±0.0001	0.2304±0.0001
145	0.1865±0.0001	0.1852±0.0001
146	0.1515±0.0001	0.1512±0.0002
148	0.1030±0.0001	0.1018±0.0001
150	0.0618±0.0001	0.0601±0.0001
Samarium		
147	0.4085±0.0004	0.4132±0.0004
149	0.2536±0.0005	0.2533±0.0002
151	0.1547±0.0001	0.1587±0.0002
152	0.1267±0.0001	0.1209±0.0002
154	0.0566±0.0001	0.0539±0.0002

increase in the yields of the valley nuclides with increasing neutron energy.

In general, the change in the relative isotopic abundances of the fission product elements near the top of each mass peak is small, generally less than 1%, relative. One exception is the isotopic composition for cerium for the ^{239}Pu samples. At this time the reason for this difference is unknown; but, analytical problems are suspected. The measurement of a small natural component in fission product cerium is a difficult task.

For all practical purposes, the data in Tables IV and V do not indicate the existance of any significantly large fast reactor fission product capture cross sections in any of the major decay chains.

VIII. ERROR ANALYSIS

The standard deviation associated with each reported fission yield is given in Table I and includes allowances for known sources of systematic errors. The standard deviation was calculated by first approximating the functions used to calculate the fission yields, by the linear terms of the Taylor series expansion, and then applying the ususal method of calculating the standard deviation of a linear sum of random variables.

The procedure consisted of three distinct steps: (1) the propagated standard deviation of the measured number of atoms of each element was calculated. This computation included the systematic effect of uncertainty in the spike concentrations, the random mass spectrometer errors in the spike, natural, fission product, and mixed spike-fission product isotopic fractions, and the Xe spike volume uncertainties. One recognized uncertainty not included is the systematic error in the element concentration resulting from systematic errors in the mass spectrometer. (2) The standard deviation of the total number of atoms in the heavy peak was calculated using the above standard deviations for the measured atoms and estimated atoms. (3) The standard deviation of each yield from each capsule was calculated using the standard

deviations of the total number of fissions, the atoms of the specific element, and the fraction of the specific isotope. The standard deviation of the isotopic fractions includes the random mass spectrometer errors and the estimated systematic uncertainty in the mass spectrometer.

IX. COMMENTS TO EVALUATORS AND COMPILERS OF FISSION YIELD DATA

The fast reactor fission yield values for ^{241}Pu given in this report are for a specific location and neutron spectrum in EBR-II; the Row-4 position. In a number of instances the yields of certain fission products, especially those on the wings of the mass yield curve are significantly different from the yields obtained from samples irradiated in the Row-8 position of EBR-II.[2] The $^{150}Nd/^{143}Nd$ isotopic ratio for ^{241}Pu fission for the Row-4 irradiation is 0.2650 compared to 0.2586 for the Row-8 irradiation and 0.2300 and 0.2243 respectively for ^{239}Pu fission. This is similar to the trends established for ^{235}U fission and supports the broadening effect on the wings of the mass yield curve with increasing neutron energy. The estimated mean neutron energy for the Row-4 and Row-8 irradiation is $\sim$700 keV and 400 keV, respectively. These differences again emphasize the need for a procedure of correlating fast reactor fission yields with neutron energy.

From an evaluation viewpoint, it must be remembered that the uncertainties associated with the yields given in this report are quite low, many less than 1%, relative. This primarily results because only one capsule was analyzed and possible random variations between capsules is not included.

Based on our experience and studies, we suggest consideration be given to limiting the errors associated with compiled and evaluated yields to a lower value of 1%, relative.

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