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**THE CORRELATION OF ^{239}Pu THERMAL AND FAST-
REACTOR FISSION YIELDS WITH NEUTRON ENERGY**

October 1981

Idaho Falls, Idaho 83401

EXXON NUCLEAR IDAHO COMPANY, Inc.

Prepared For The
DEPARTMENT OF ENERGY

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THE CORRELATION OF ^{239}Pu THERMAL-[#]AND
FAST-REACTOR FISSION YIELDS WITH NEUTRON ENERGY

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ABSTRACT

The relative isotopic abundances and the fission yields for over 40 stable and long-lived fission products from ^{239}Pu fast fission were evaluated to determine if the data could be correlated with neutron energy. Only mass spectrometric data were used in this study. For some nuclides changes of only a few percent in the relative isotopic abundance or the fission yields over the energy range of thermal to 1 MeV are easily discernable and significant; for others the data are too sparse and scattered to obtain a good correlation. The neutron energy index used in this study is the $^{150}\text{Nd}/^{143}\text{Nd}$ isotopic ratio. The results of this correlation study are compared to the US Evaluated Nuclear Data File (ENDF) fast fission yield compilation. Several discrepancies are noted and suggestions for future work are presented.

SUMMARY

To better define fast reactor fission yields and to better establish the uncertainties associated with fast reactor yields, a study was conducted to investigate the correlation of ^{239}Pu fast fission yield data with neutron energy. Through the use of precise mass spectrometric isotopic abundance and isotope dilution measurements of many stable and long-lived fission product nuclides, both the change in the relative isotopic abundance, and the fast reactor fission yields for some nuclides are shown to correlate well with neutron energy. For others, the data are too sparse and scattered to obtain definitive correlations. The data used in this study were obtained from samples irradiated in several different fast reactors and were analyzed in different laboratories.

The energy index used for the correlation study is the isotope ratio $^{150}\text{Nd}/^{143}\text{Nd}$. This ratio can be measured with high precision, approximately 0.25%, and for ^{239}Pu fission changes approximately 20% over the energy range of thermal to approximately 1 MeV. The correlation of the isotopic abundances and the fission yields of the major fissions product nuclides is linear relative to the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio for a wide energy range.

As part of this study, the current ENDF yields, especially the fast yields, were compared to the results obtained from this work. Several discrepancies between the ENDF values and those obtained from this work are identified.

The major observations from this work are: 1) that fast fission yield values for some of the major fission products for ^{239}Pu fission can now be assigned for any neutron spectrum, 2) in many cases the data are too sparse and scattered to obtain a definitive correlation, 3) there are several discrepancies between the correlations developed in this study and the current ENDF data, and 4) there is a serious need for additional ^{239}Pu fast yield measurements, especially in the 800-1000 keV range.

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I. INTRODUCTION

In late 1980, this laboratory published a report¹ discussing the correlation of ^{235}U fission yields with neutron energy. Of particular interest was the energy range from thermal to about 1 MeV, the range associated with fast breeder reactors. By using only mass spectrometrically determined data which could be associated with a known neutron energy index it was shown that:

1) the differences in relative isotopic data from different experiments were well correlated with neutron energy, 2) many of the differences in fast yield values reported in the literature could be resolved when the values were correlated with neutron energy, and 3) that serious consideration should be given to the energy dependency factor to improve and make more meaningful existing and future fast reactor fission yield compilations.

This paper extends the ^{235}U yield-energy correlation study to the fissioning of ^{239}Pu . Many users of fission yield data, especially those involved in burnup measurements and fast breeder fuel development studies, have requested a value of $\pm 1\%$ as an uncertainty requirement for fast reactor fission yields^{2,3}. At the present time, essentially all compilations of fast reactor fission yields carry the assignment "fast yields," with no indication of the neutron energy associated with the quoted value. In some cases, the stated uncertainty associated with a particular yield is smaller than the change in the absolute yield for the neutron energy range which exists in a fast reactor. Thus, either the uncertainty value must be increased, which is counter-productive, or future fast yield compilations must, if possible, incorporate the energy dependency factor.

The purposes of this paper are: 1) to re-evaluate some of the more recent ^{239}Pu fast reactor yield data that can be associated with some neutron energy index, 2) to attempt to correlate these data with neutron energy to show the small changes in the yields that occur in fast reactors, 3) to compare these results with current compiled data, 4) to establish a more meaningful degree of confidence in the quoted values, and 5) to identify those areas where additional measurements are required to better define the yield-energy relationship.

II. THE DATA BASE

A. Thermal Fission Yields

Currently over a thousand individual references relative to thermal fission yields are available. The review, evaluation, and compilation of these data is a monumental task. Several individuals and organizations have undertaken this chore with varying degrees of vigor and continuity. Of these the most well documented and referenced works are those of Rider⁴ (this is essentially the U. S. Evaluated Nuclear Data File, ENDF/B-V(e)), E.A.C. Crouch⁵ (UK Fission Yield Data File), W. H. Walker⁶, and Lammer and Eder⁷ (now part of IAEA data file). In general, the recommended values presented by these evaluators are derived based on various statistical pooling and weighting techniques, and on the personal opinion and preferences of the individual evaluators.

In this paper, thermal yield data generated in our laboratory⁸ are used as the base values from which to determine the changes in yields with neutron energy. These data were selected because the measurement techniques (primarily isotope dilution mass spectrometry), the determination of the number of fissions based on the summation technique, and the general laboratory efforts were essentially identical to those used to measure the bulk of the fast reactor yield data given in this report. Hence, any serious systematic errors in standards, calibration, or technique should not affect the final results.

B. Fast Reactor Fission Yields

Compilations of fast reactor fission yields are not nearly as well developed as those for thermal yields. The two main factors responsible for this situation are: 1) the number of measurements reported for fast yields is small compared to thermal yield measurements (the number of ²³⁹Pu measurements is less than those for ²³⁵U), and 2) much of the reported fast yield data cannot be associated with a known neutron spectrum. As a result of this second item, evaluators and compilers of fast yield data, though they recognized that certain yields can change significantly with neutron energy, have had little choice other than to use pooling techniques similar to those used for thermal data. Thus, it is understandable

that fast yield compilations can show significant discontinuities for adjacent yields because the data were probably obtained from experiments conducted in significantly different neutron spectra.

In this study, to correlate ^{239}Pu yields with neutron energy, the criteria for entry into the data base were: 1) mass spectrometrically determined values, and 2) a measurement of the relative isotopic abundance of fission product neodymium. Although these criteria eliminated the bulk of the reported fast yield measurements, especially radiochemical measurements, the amount of data retained was sufficient to conduct the evaluation study and to identify specific problem areas. The criterion for the neodymium data is discussed in Section II-C.

The ^{239}Pu data used in this study include measurements from samples irradiated in the Experimental Breeder Reactor-II (EBR-II), the Dounreay Fast Reactor, the French fast reactor (RAPSODIE), the USSR reactor (BOR-60), and EBR-I. The primary data sources, details relative to various experiments, neutron spectrum, and specific comments on the experiments and data are given in Appendix I.

For all of the major fission products, mass spectrometrically measured relative isotopic data representative of at least three significantly different neutron energies, in addition to the thermal data, are available. In some cases however, absolute yield data for ^{239}Pu fission are only available for two different fast neutron environments. In general, the bulk of the data used in this study were those produced in this laboratory.

C. Neutron Energy Index

There are several conventions for expressing a neutron energy index. Some of the more common are mean neutron energy, median neutron energy, the mean neutron energy for fission of a certain fissioning nuclide, and the "spectral index", defined as the fission ratio of ^{238}U to ^{235}U ($^{238}\text{U}(n,f)/^{235}\text{U}(n,f)$).

In the previous study pertaining to the correlation of ^{235}U fission yields with neutron energy¹, a model was developed using the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio for ^{235}U fission as the energy index. This index was selected for use in that

study because previous studies^{2,3,9} showed a strong correlation of the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio with the "spectral index". This same ratio for ^{239}Pu fission is used in the present study to evaluate the correlation of ^{239}Pu yields with neutron energy.

The $^{150}\text{Nd}/^{143}\text{Nd}$ ratio is especially sensitive to neutron energy: as the energy of the incident fissioning neutron increases, the shape of the mass yield curve changes; the isotopic abundance of ^{143}Nd decreases, that of ^{150}Nd increases. The change in this ratio is approximately 20% between thermal and a mean neutron energy of 1 MeV. This change is quite significant considering the isotopic ratio of $^{150}\text{Nd}/^{143}\text{Nd}$ can be measured with an uncertainty of $\pm 0.25\%$ relative. Thus, small changes in this ratio, which are accurately measured, are significant with respect to the energy of the fissioning neutron. A unique feature of using the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio as the energy index is that the changes in both the isotopic abundances and the fission yields are linear with energy.

An additional benefit derived from using the $^{150}\text{Nd}/^{143}\text{Nd}$ isotopic ratio as the energy index is that it permitted the use of mass spectrometrically measured yield data when the neutron energy of the irradiation was unknown, provided the experimenter had measured the isotopic composition of the fission product neodymium. Thus, by using this technique it was possible to include several measurements in this study that otherwise, because of a lack of spectral information, could not be included.

A comparison of the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio for ^{235}U fission to that for ^{239}Pu fission is given in Figure 1. Also given in the lower portion of the plot is the corresponding mean neutron energy. The comparison is based on actual measured values of the $^{150}\text{Nd}/^{143}\text{Nd}$ ratio determined for samples irradiated in several different fast reactors.

The energy range covered in this study is from well moderated thermal reactors ($^{150}\text{Nd}/^{143}\text{Nd}$ ratio for ^{239}Pu fission = 0.2177) to the metal core of the now decommissioned fast reactor EBR-I ($^{150}\text{Nd}/^{143}\text{Nd}$ ratio for ^{239}Pu fission = 0.2465).

Figure 1. Comparison of the $^{150}\text{Nd}/^{143}\text{Nd}$ Isotopic Ratio with Neutron Energy for ^{235}U and ^{239}Pu Fast Fission

III. DATA TREATMENT AND CORRELATION

Most attempts to correlate fission yields with neutron energy have been restricted to those nuclides that show the most significant changes with energy (i.e., valley and extreme wing nuclides), and where the energy range is on the order of several MeV. Prior to the recent ^{235}U correlation report¹, essentially no correlation studies had been attempted for the energy region encompassed by thermal and fast reactors. The ^{235}U study showed that although the changes in the yields in this limited energy range are small (1-10% relative), such changes must be considered if the users' requirement of yields accurate to ± 1.0 -1.5% relative is to be achieved.

In both the ^{235}U study and in this effort to correlate ^{239}Pu yields with energy, only mass spectrometric data have been considered. This decision was predicated on the facts that: 1) the data could be associated with an energy index even if specific measurements of the neutron spectrum were not made, 2) mass spectrometric measurements can be made with smaller uncertainties than other measurements, and 3) small changes in the isotopic composition of a given fission product are usually significant and reliably measured. Much of the mass spectrometrically measured thermal and fast yield data in the literature originated from this laboratory. Generally these data took precedence in the decision making process used in the correlation study because: 1) all were obtained in a similar manner in the same laboratory, and 2) a more direct knowledge of the experimental details was available.

Two correlations of ^{239}Pu data were considered in this study. The first only involved the use of relative isotopic data; the second considered the reported literature values for the yields. Because the amount of useful ^{239}Pu data is significantly less than that for ^{235}U , some of the proposed correlation trends are open to question. This lack of data points to the need for more experimental data. Details of the evaluation studies follow:

A. The Correlation of ^{239}Pu Fission Product Relative Isotopic Abundance Data with Neutron Energy

The correlation of the relative isotopic composition of the fission products for ^{239}Pu fission with neutron energy was considered first because: 1) normally,

relative isotopic values are more accurately measured than are absolute fission yields, and 2) small but significant changes in the relative isotopic composition of a fission product element are easily detected, and, as such, may indicate a change in the yields or the structure of the mass yield curve. Relative fission product isotopic data are more accurate than absolute fission yield values. In practice the conversion of relative isotopic values to absolute yields requires a knowledge of the absolute number of atoms of the fission product nuclide and of the total number of fissions; hence, all absolute yield data must be considered potentially biased because of systematic errors associated with the measured absolute number of atoms of a specific fission product and/or the number of fissions.

Fortunately, in the isotope dilution mass spectrometric measurement of a given fission product element an error in the number of added spike isotope atoms translates equally to each isotope of the particular element being measured. Likewise, an error in the number of fissions used to establish the absolute fission yields is common to each isotope of the element being measured. Thus, while it is quite possible that the absolute yield values for the various isotopes of a given element may be in error, the relative isotopic composition of that element should be quite reliable.

For those reference sources that did not provide measured relative isotopic data, the absolute yield values were converted to relative isotopic data by the following technique: absolute yield values for the isotopes of a given fission product element were summed and each individual absolute yield was divided by the sum. This gave the relative isotopic composition of the fission product element and reduced the uncertainties to only those associated with the mass spectrometric measurement. Errors distinct to a single isotope (i.e., spectral impurity), though not eliminated, are rather easily identified, if more than a few percent.

An example of this method and the results for ^{143}Nd are given in Figure 2. In this case, relative isotopic data were reported in only about one-half of the references which gave yield data for the neodymium isotopes. The range in the reported yields for ^{143}Nd was several percent. After converting the absolute yield data to relative isotopic data, essentially all of the relative isotopic values for ^{143}Nd fell within $\pm 0.50\%$ relative of the best estimate of the change in the relative isotopic abundance of ^{143}Nd with the neutron energy index.

Figure 2. Change in Relative Isotopic Abundance of ^{143}Nd with Neutron Energy for ^{239}Pu Fission

This exercise clearly shows: 1) that while there were significant differences in the reported absolute yield values, most of the relative isotopic mass spectrometric measurements are quite reliable, and 2) that there is a gradual, but significant, change in the isotopic composition of ^{143}Nd with neutron energy.

The data for all of the major fission product elements were evaluated in a similar manner. Unfortunately, the available data for many of the fission products were not as plentiful as they were for neodymium. Hence, some of the correlation trends presented in this paper are open to question. It is in these cases where additional measurements are needed to clarify the trends. In all cases, the energy index, E_n , is the isotopic ratio $^{150}\text{Nd}/^{143}\text{Nd}$. The data are presented in Appendix II in graphical form in Figures A-II-1 through A-II-11, ordered by increasing mass number. In these figures, data from this laboratory, the Idaho Chemical Processing Plant, are identified by $\bullet$; data from other laboratories by $\square$; and, for reference, recent ENDF values by $\triangle$. For the ENDF data, it was assumed that all of the ENDF values were for a common neutron spectrum, denoted by the ENDF $^{150}\text{Nd}/^{143}\text{Nd}$ ratio for ^{239}Pu fission of 0.2290.

None of the data were subjected to any rigorous statistical treatment because in many cases reliable uncertainty values were not available. The indicated best line fits were drawn at the discretion and judgement of the author. The boundary limits were arbitrarily selected as $\pm 1\%$, relative. The primary purpose of presenting the data in this form is to show that in many cases changes of 1% in the relative isotopic abundance values are significant, and to identify those cases where the data are questionable.

B. The Correlation of ^{239}Pu Absolute Fission Yields with Neutron Energy

The second correlation study involved the direct use of literature yield values. For reasons previously stated (Section III-A), it was known that the yield data, and subsequent correlations, would be more scattered than the relative isotopic data; but the objective was to determine if the data were adequate and sufficiently reliable to describe the change in the yields with neutron energy. The same energy index, $^{150}\text{Nd}/^{143}\text{Nd}$ isotope ratio, was used for this evaluation. In all cases, the reported literature values were used without adjustments.

Like the relative isotopic abundance correlation study, the trend or best-fit lines were constructed solely at the discretion of the author. Because of better acquaintance with the data generated in this laboratory, and the fact that all of these data (thermal and fast yields) were obtained using the same techniques and equipment, these values were weighted more heavily in the decision making process. Recent ENDF values⁴ are included for the purpose of evaluation and comparison. In those cases where this evaluation differs significantly from the ENDF data, the difference is indicated by the use of a dashed-line.

The results of this study are presented in graphical form in Appendix-III in Figures A-III-1 through A-III-12. The data identification key for these plots is: ●, for data from this laboratory; □, for data from other laboratories; and ▲, for ENDF values.

One of the major problems encountered in this evaluation and correlation of ^{239}Pu fission yields with neutron energy was the lack of mass spectrometric yield data in the region of about 800 keV, mean neutron energy. This is especially noted for those fission products in the light mass peak for which little or no data are available for the energy range between row-8 of EBR-II (approximately 400 keV) and the core of EBR-I (approximately 1300 keV). In addition, there are questions relative to some of the EBR-I data. In many instances where the data are sparse and often scattered, the indicated trend line correlating the yields with neturon energy are based on the author's judgement and guidance from the ^{235}U data. The reader may disagree with the author's interpretation, but this is the penalty which must be paid when the data are less than desirable and only an educated estimate can be made.

Without question, there is a major need for additional fast fission yield measurements in the mean neutron energy range of 800 to 1000 keV if meaningful yield-energy relationships are to be established for ^{239}Pu fission products. Additional measurement in the 200-400 keV range will do little to clarify the proposed yield-energy relationship. This conclusion is quite evident when the yield-energy plots given in Appendix III are examined. If such an experiment were to be conducted in the US, it is recommended that the irradiation be done in row-2 of EBR-II.

IV. DISCUSSION AND EVALUATION

The results of the correlation of relative isotopic abundance data with neutron energy and the correlation of fission yields with neutron energy are discussed below for each of the major fission product elements. Scattered and seemingly erroneous data points and disagreements with current ENDF values are identified, and, if possible, an explanation is given. The fact that relative isotopic data are given for $E_n = 0.23$ and that no corresponding yield values are given is the result of not being able to quantify the number of fissions associated with this sample.¹⁰ For a better understanding of the evaluation process the reader should refer to the graphical data given in Appendix II and III when reading the following sections. In these discussions, E_n is used to identify the energy index, $^{150}\text{Nd}/^{143}\text{Nd}$ isotopic ratio.

A. Krypton

The relative isotopic abundances of ^{83}Kr , ^{84}Kr , and ^{86}Kr correlate well with neutron energy over the full energy range from thermal to EBR-I (approximately 1300 keV). All values fall within $\pm 1\%$ of the best line estimate. The largest change in relative isotopic abundance is for ^{83}Kr , approximately 6%; this compares to an 11% change in the relative isotopic abundance of ^{83}Kr for ^{235}U fission for the same energy range. The current ENDF values fall within the boundary limits and indicate changes similar to the individual experimental values. All experimental data points originated from this laboratory.

The correlation of the fission yields of ^{83}Kr , ^{84}Kr , and ^{86}Kr with neutron energy also is quite good, with all of the data, except one set of values, falling within $\pm 1\%$ of the best estimate. In all cases, the one set lying outside that range ($E_n = 0.221$) lies within $\pm 2\%$ of the best estimate. This particular sample was located in a soft spectrum in the axial blanket of row-8 of EBR-II. For all three krypton isotopes, the indicated change in the yields is in the same direction as indicated by the ENDF data but it is of a greater magnitude.

As will become evident in reviewing these data, considerable weight was given to the EBR-I values ($E_n = 0.2465$) in constructing the correlation line when the author believed these data to be reliable. The need for new data in the region $E_n = 0.23 - 0.24$ is quite evident.

B. Rubidium

In general, the relative isotopic abundance data correlates well with neutron energy. The magnitude of the evaluated changes in the abundance values is somewhat less than that indicated by the ENDF values, but of no apparent serious consequence.

The measured yields for ^{85}Rb and ^{87}Rb also correlate well with neutron energy; however, a major discrepancy is indicated in the change in the yield of ^{87}Rb with neutron energy based on this evaluation compared to that indicated by the ENDF data. For the same energy range, the measured values given in this report indicate that the yield for ^{87}Rb increases from 1.00% to approximately 1.07% (7% relative increase); the ENDF values indicate no change. Review of the ENDF fast yield data is suggested.

All of the measured values originated solely from this laboratory.

C. Strontium

Both isotopes of strontium, ^{88}Sr and ^{90}Sr , show excellent correlation of their relative isotopic abundance values with neutron energy. All of the measured data as well as the ENDF values lie within about $\pm 0.5\%$ of the best line estimate. No significant change with neutron energy is indicated.

The measured yield values also show good correlation with neutron energy, but indicate a change with energy opposite to that indicated by the ENDF values. For both ^{88}Sr and ^{90}Sr , the ENDF thermal values are several percent larger than our values,⁸ and the fast values a few percent less. It is very difficult to reconcile these differences and a review of the ENDF data, especially the thermal values, is indicated. The major questions to be addressed are, whether the absolute yields for ^{88}Sr and ^{90}Sr are valid, and do they increase or decrease with increasing neutron energy⁴?

D. Zirconium

The bulk of the relative isotopic values for fission product zirconium show a good correlation with neutron energy (Figure A-II-4). Where scatter does occur the values are generally within $\pm 2\%$ of the best line estimate.

Reasonable agreement with the ENDF Values, except for ^{96}Zr , is indicated.

The major problem identified in the evaluation of zirconium data is with the absolute yield values and the comparison with the ENDF values. For all of the zirconium isotopes, the yield-energy trend lines are in direct opposition to that based on the ENDF values (Figure A-III-4). This evaluation concludes that the yields for all of the zirconium isotopes increase with neutron energy; the ENDF values indicate that the yields decrease with energy, in some cases (^{94}Zr) quite dramatically.

Historically, the mass spectrometric measurement of fission product zirconium has presented problems; the major one being significant amounts (5-50%) of natural zirconium contamination which had to be subtracted from the total measured zirconium to obtain the residual fission product component. The major source of the natural zirconium is thought to arise from the use of quartzware for the sample dissolution and sample storage. Normally, in the manufacturing process, quartzware is fired and annealed in zirconia-lined furnaces. This natural zirconium contamination could contribute to the scatter in the yield values shown in Figure A-III-4. This is one of those cases where additional measurements, specifically designed to minimize the natural contamination factor, might aid in establishing more definitive trend lines.

In general, the ENDF values for thermal fission are approximately 2-4% greater than the value measured by this laboratory⁸ and used in this study. This difference, coupled with the fact that the ENDF fast yield values are generally lower than our measured values combined to give the indication that the yields of the zirconium isotopes decrease with energy.

One additional factor to consider in attempting to reconcile this problem is the fact that the ENDF relative isotopic values calculated from the ENDF yield values are in agreement with the measured values (Figure A-II-4). This could indicate a relatively consistent bias in all of the ENDF yield values. Review of both thermal and fast ENDF values is indicated.

E. Molybdenum

The majority of the relative isotopic abundance values for the molybdenum isotopes shows good correlation with neutron energy. Little change with energy is indicated.

There are no thermal isotopic data or thermal yield data from our laboratory used in this evaluation. In our thermal yield measurement study⁸, it was determined that some of the fission product molybdenum had combined with the fission product ruthenium and palladium to form a finely divided intermetallic material that did not dissolve. Thus, the ENDF values were used as the thermal data points.

From the available molybdenum yield values (Figure A-III-5), it is next to impossible to establish a definitive yield-energy relationship for the molybdenum isotopes. The trend lines given in Figure A-III-5 show the molybdenum yields decreasing with neutron energy. This decision was based on two items: 1) the ENDF values indicated a decrease with energy, and 2) to conserve mass in the light mass peak, the molybdenum yields must decrease considering that all of the light mass isotopes up to this point tend to increase with energy.

The author recognizes that the above arguments may be challenged based on the scatter of the data, and is open to other suggestions.

F. Ruthenium

The correlations of the relative isotopic abundance data for ^{101}Ru , ^{102}Ru , and ^{104}Ru are given in Figure A-II-6. Essentially all of the values lie within $\pm 1\%$ of the best estimate trend line. The isotopic abundances show little or no change with neutron energy and no discrepancy with the ENDF values. For ^{235}U fission, the relative isotopic abundance of ^{104}Ru increased 19% over the same energy range.

Although yield data are given for the ruthenium isotopes as a function of neutron energy (Figure A-III-6), no correlation of the data with energy is given.

In the author's opinion, the data are too sparse and scattered to establish a definitive trend. No thermal yield data from the Idaho laboratory are included because in nearly all of our experiments ruthenium plated on the wall of the dissolver vessels or precipitated from solution as an intermetallic material with molybdenum and palladium and, therefore, complete recovery of the ruthenium was not obtained. In addition to the lack of thermal data, the ENDF values are quite discrepant with the measured values.

If reliable ruthenium yield data are desirable, it appears that a special experiment must be conducted to obtain the values. Serious consideration should be given to the irradiation of a Pu-Al alloy rather than the oxide. Additionally, well researched dissolution procedures must be used to obtain complete dissolution and stability of the ruthenium in the dissolved product.

G. Xenon

The changes in the isotopic composition of the xenon isotopes as a function of neutron energy are shown in Figure A-II-7. The indicated relative changes in the isotopic abundances range from an increase of approximately 6% for ^{131}Xe to a decrease of approximately 2% for ^{136}Xe for a neutron energy range of thermal to 1300 keV. These changes are considerably less than those for ^{235}U fission where the relative isotopic abundance of ^{131}Xe increases approximately 20% and that for ^{134}Xe decreases approximately 9%, over the same energy range. Also given in Figure A-II-7 are the relative isotopic values for the various xenon isotopes for 14.7 MeV fission¹⁸. In all cases the trends established in the low energy range are continuous to the high energy range. The significance of the 14.7 MeV data is discussed in detail in Reference 1. The bulk of the xenon isotopic measurements are from this laboratory; data for one other experiment¹¹, are also given. These other data tend toward higher isotopic abundances for the low mass xenon isotopes and a lower abundance for mass 136. In general, the experimental trend lines are in agreement with the ENDF values, except for ^{136}Xe where the ENDF data indicate a slight increase in the abundance of ^{136}Xe compared to a decrease described by the measured data.

The yield data given in Figure A-III-7, unfortunately, are much more scattered than the relative isotopic data. Of particular concern are the results from the "TACO" Experiment and those from the analysis of the EBR-I fuel¹⁷. The trend lines shown in Figure A-III-7 were constructed primarily based on thermal⁸ and EBR-II¹⁰ data, with the ENDF and 14.7 MeV data used as a guide. Results in the 800-1000 keV range would be most helpful in clarifying this problem and in establishing more confidence in the construction of the trend lines.

H. Cesium

The correlation of the relative isotopic values for the cesium isotopes with neutron energy is shown in Figure A-II-8. The results from this laboratory (ICPP) fall within approximately 0.5% of the best line fit, while the results from the TACO experiment¹¹ show some scatter. This may be the result of natural contamination because natural cesium is monoisotopic (^{133}Cs) and a common contaminant. In general, the relative isotopic abundance of the cesium isotopes changes little with neutron energy. All of the ENDF values fall within $\pm 1\%$ of the best line estimate.

The correlation of the cesium yield data with energy is shown in Figure A-III-8. Again, if data in 800-1000 keV range were available more confidence could be placed on the indicated trends. A classic example of the need for additional data is shown in Figure A-III-8 for ^{135}Cs . The scatter of the EBR-II Row-8 data and that of the TACO experiment¹¹ relative to the thermal value is such that almost any trend line could be constructed, either increasing or decreasing with energy. For this case the value at $E_n = 0.2465$ is the governing point. Clearly, a value at $E_n = 0.235$ would be most helpful in this case.

For both ^{135}Cs and ^{137}Cs , the ENDF values indicate a much greater relative change in yields with energy than does the author's evaluation. This point should be resolved especially for ^{137}Cs , which is one of the more widely used monitors for the measurement of fission density and burnup. Of particular concern are the ENDF fast yield data.

I. Barium

No relative isotopic data are given for barium because only one stable barium fission product isotope, ^{138}Ba , is mass spectrometrically measured. Any measured ^{137}Ba is considered to be a decay product of ^{137}Cs and is summed with the measured atoms of ^{137}Cs to give the total number of mass 137 atoms. The measured yields of ^{138}Ba as a function of neutron energy are given in Figure A-III-9. The value at $E_n = 0.2465$ is probably in error as a result of natural contamination problems. The ENDF values are consistent with the measured values. Little energy effect is indicated.

J. Lanthanum

Lanthanum, like barium, has only one stable fission product isotope, ^{139}La ; hence, no relative isotopic data are given for lanthanum. Unlike barium, however, it is quite difficult to obtain an accurate and reliable mass spectrometric analysis of fission product ^{139}La . This stems primarily from the fact that natural lanthanum is greater than 99.99% ^{139}La , and thus it is difficult to accurately measure the very small ^{138}La peak to obtain an accurate estimate of the correction for natural lanthanum contamination. The wide scatter in the results given in Figure A-III-9 is believed due to the inability to make accurate corrections for natural lanthanum. This scatter precluded constructing any reliable trend line to estimate the correlation of ^{139}La yields with neutron energy.

The thermal value, ϕ , shown in Figure A-III-9 is not a measured value but rather is the value derived from a linear interpolation between the measured yields for ^{138}Ba and ^{140}Ce . No judgement is presented relative to the ENDF values.

K. Cerium

The correlation of the relative isotopic abundance of ^{140}Ce and ^{142}Ce with neutron energy is shown in Figure A-III-10. All of the measured values, except those for EBR-I fall within $\pm 1\%$ of the best line estimate. The measurement of the EBR-I samples, which was done several years ago, may be subject to error because of natural contamination. The effect of neutron energy on the change in the relative abundance of the cerium isotopes is more pronounced for ^{239}Pu fission than for ^{235}U fission. For ^{239}Pu fission, the relative abundance of ^{140}Ce increases about 10% for the energy range of thermal to 1300 keV, compared to 2% for ^{235}U fission.

The indicated absolute change in the fission yields of the cerium isotopes, especially ^{140}Ce , is quite significant: greater than 10% relative for the energy range covered in EBR-II (Figure A-III-10). Additional measurements for mass 140 at higher energies ($E_n = 0.23\text{-}0.24$) should be made to confirm if the change is as large as now perceived and to resolve the discrepancy with the ENDF values. Of particular concern are the ENDF thermal values. This is especially important because ^{140}Ba - ^{140}La yield data are commonly used in estimating the number of fissions and in fission rate measurements.

If additional mass spectrometric measurements are made for the cerium isotopes, special care must be taken to accurately correct for natural contamination. The problems in the accurate measurement of the absolute number of fission product cerium atoms is in many ways similar to that associated with ^{139}La . The most abundant isotope in natural cerium is ^{140}Ce , 88.5%. Thus, it is crucial to obtain an accurate correction for natural cerium. The measurement of ^{136}Ce and/or ^{138}Ce can be used as natural monitors provided special techniques are used to accurately measure the small abundance of these isotopes. To obtain more reliable values for ^{136}Ce , we use a two stage mass spectrometer and peak stepping at two different intensity levels.¹⁰ First the oxide beam of mass 140, 142, and 144 is measured. Then the beam intensity is increased about 20 fold and the 136 to 144 ratio is measured. Because the atom abundance of mass 144 is less than the abundance of mass 142 but greater than mass 136, the measurement of a large 140/136 ratio is not required. This provides a means to improve the accuracy of the measurement.

L. Neodymium

The largest number of mass spectrometric fission product measurements reported in the literature for ^{239}Pu fission is for neodymium. This primarily results from the use of the neodymium isotopes, especially ^{148}Nd , as burnup monitors, and, hence, requires the need to accurately know the fission yields of the neodymium isotopes.

The correlation of the relative isotopic data with neutron energy for the neodymium isotopes is given in Figure A-II-10. The values for the Nd isotopes 143, 145, 146, 148, and 150 are given relative to the sum of these isotopes. The value for ^{144}Nd is relative to all of the neodymium isotopes. The data are presented in this manner because not all experimentors reported data for mass 144. In all cases, except for two 144 values, the relative isotopic data fall within $\pm 1\%$ of the best line estimate. The exceptions may be the result of incorrect decay corrections or may be based on radiochemical measurements for ^{144}Ce . The neodymium relative isotopic data for ^{239}Pu show changes similar to that for ^{235}U . The relative abundance of the light mass neodymium isotopes decreases with neutron energy, while the abundance of the heavy mass neodymium isotopes increase with energy (Figure A-II-10).

The values for the absolute fission yields for the neodymium isotopes, given in Figure A-III-11, show considerably more scatter than the relative isotopic data. The consistently high yields reported by Robin¹⁴ ($E_n = 0.2296$) may be the result of an inaccurate estimate of the number of fissions or an error in the number of isotope dilution spike atoms added to the sample. If these values are disregarded, the remaining yield values fall within $\pm 2\%$ of the best line estimate, describing the change in yields with neutron energy.

M. Samarium

The isotope abundance values for the fission product samarium isotopes are given in Figure A-II-11. The values for the samarium isotopes 147, 149, 151, and 152 are given relative to the sum of these isotopes because one set of literature data¹¹ did not report values for ^{154}Sm . The relative isotopic abundance values for ^{154}Sm are relative to the sum of all of the fission product samarium isotopes. Our measured relative isotopic values for ^{151}Sm and ^{152}Sm for thermal fission⁸ appear discrepant compared to the fast fission values (Figure A-II-11). This discrepancy in the thermal values may be the result of the large neutron capture correction required for ^{151}Sm . From the data given in Figure A-II-11 it appears that the actual correction for ^{151}Sm capture to ^{152}Sm may be larger than the applied calculated correction. For ^{147}Sm and ^{149}Sm , the correction for thermal neutron capture was based on adding the measured amount of capture product to the target nuclide to obtain the total amount of original target. The measured amount of ^{148}Sm was added to ^{147}Sm , and the ^{150}Sm was added to ^{149}Sm .

The reported yield values for the samarium isotopes show more scatter (Figures A-III-12) than the relative abundance data. For ^{147}Sm , ^{149}Sm , and ^{151}Sm , all of the data, except the EBR-I values¹⁷, fall within about $\pm 2\%$ of the best line estimate. The values for ^{152}Sm are more scattered. The high thermal yield value for ^{152}Sm may be the result of incomplete correction of ^{151}Sm capture to ^{152}Sm . As expected, the higher mass samarium isotopes show significant increases in their yields with increasing neutron energy.

V. CONCLUSIONS AND RECOMMENDATIONS

A study was conducted to evaluate and to determine if existing ^{239}Pu fission yield data could be correlated with neutron energy over the energy range encompassed by thermal and fast reactors. A prior study using mass spectrometrically measured ^{235}U fission yield data¹ showed that both the changes in the relative isotopic abundance values and the fission yields of the major fission product nuclides were well correlated with neutron energy. The same degree of success in establishing correlation trends was not obtained for ^{239}Pu because the amount of measured ^{239}Pu data is less than that for ^{235}U and the data are more scattered. Thus, not only was it more difficult to evaluate the data, but in some cases, it was impossible to establish definitive correlation trends for many of the ^{239}Pu fission products.

The neutron energy index used in this study, like the ^{235}U study¹, is the isotope ratio $^{150}\text{Nd}/^{143}\text{Nd}$. A unique feature of this index is that the resulting correlations are linear for the energy range studied.

Figure 3 compares the changes in the yields with neutron energy derived from this study with those listed in ENDF. The figure gives the percent change in the fission yields as a function of mass number where the percent change in the yields is defined as:

$$\left(\frac{\text{fast fission yield}}{\text{thermal fission yield}} - 1 \right) \times 100$$

The values for the fast fission yields were taken from the data given in the plots in Appendix-III at the point $E_n = 0.2300$ (approximately 600 keV). In general, the data follow the expected trends for the change in the structure of the mass yield curve for increasing neutron energy, although there are a few questionable points.

Also given in Figure 3 are the corresponding changes based on ENDF values for those cases where the indicated changes in the yields are more than a few percent different than the values derived from this evaluation.

Figure 3. Change in ²³⁹Pu Fission Yields as a Function of Mass Number

Where such discrepancies occur, the ENDF values are plotted with the symbol, Δ . Several discrepancies are identified, especially on the light mass peak. Of particular concern is the fact that many of the indicated changes in the yields established by this study are directly opposite to those indicated by the ENDF values. For example, this study concludes that the yield for ^{90}Sr increases with neutron energy, while the ENDF data show the yield decreasing with increasing energy. Many of the ENDF values, both thermal and fast, need to be re-evaluated and reviewed.

A major observation of this work is the lack of yield data in the 800 to 1000 keV energy range. At the present time little or no data are available between the energy range of approximately 400-1300 keV. This is especially the case for the nuclides in the light mass peak. Yield data in this energy range are highly desirable to more definitively establish trends in the change in yields with neutron energy. The measurement of yields for samples of ^{239}Pu irradiated in the row-2 position of EBR-II would help appreciably to resolve this problem.

Specific comments and identified problems are listed below.

1. Light Mass Peak

- a. Striking discrepancies are noted for the isotopes of Sr and Zr where the changes in the yields (direction) derived from this study appear to be opposite to those indicated by ENDF data.
- b. Additional Mo measurements are needed to clarify the existing yield data, which are quite scattered.
- c. The Ru yield data are highly questionable and additional measurements are needed. Because of the unpredictable nature of Ru chemistry, the experiment must be designed with the specific intention of obtaining Ru yields.

2. Heavy Mass Peak

- a. The yield data for the xenon isotopes are quite scattered.

Measurements in the 800-1000 keV range would be most desirable.

- b. Additional measurements of Cs yields are desirable. The ENDF value for ^{135}Cs fast fission should be examined.
- c. No ^{138}Ba yield data exist for energies greater than that of row-8 in EBR-II. Additional measurements at higher energies would be beneficial.
- d. The data for ^{139}La are quite scattered and it was impossible to construct a definitive trend line.
- e. There is a serious discrepancy in the cerium yield data used in this study and that contained in ENDF. Complete review of ^{140}Ce and ^{142}Ce data is recommended.
- f. The bulk of the Nd data lie within $\pm 2\%$ of the best fit trend lines. Measurements in the 800-1000 keV range would be helpful in the construction of more definitive trend lines.

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APPENDIX I

Brief Description of Experiments and Data Used in Correlation Study

(References identified in Appendix I can
be found in the general Reference Section)

APPENDIX-I

Brief Description of Experiments and Data

The symbol used in the data plots in Appendices II and III, and the corresponding $^{150}\text{Nd}/^{143}\text{Nd}$ isotopic ratio are given adjacent to each discussion.

Thermal Fission Yields

$^{150}\text{Nd}/^{143}\text{Nd}$ Ratio

- - 0.2177 W. J. Maeck, et.al.⁸ Engineering Test Reactor (ETR) - Idaho National Engineering Laboratory. Seven samples of 99.12% enriched $^{239}\text{PuO}_2$ sealed in high purity aluminum capsules were irradiated in the reflector region of the ETR. Three samples were irradiated to approximately 1°OF , and four to approximately 50°OF . The ^{239}Pu capture-to-fission ratio was approximately 0.48. All seven samples were analyzed mass spectrometrically for the stable and long-lived fission products of krypton, rubidium, strontium, zirconium, molybdenum, ruthenium, xenon, cesium, barium, cerium, neodymium, and samarium. The number of fissions was determined by the summation method. The values reported in this work were used as the basic thermal neutron comparison points in the correlation study.
- △ - 0.2185 B. F. Rider⁴ - evaluated data. Essentially the same as the US ENDF/B-V(e) data file.

Fast Reactor Fission Yields

$^{150}\text{Nd}/^{143}\text{Nd}$ Ratio

- - 0.2215 W. J. Maeck,¹⁰ EBR-II, Row-8, Idaho National Engineering
- - 0.2237 Laboratory.
- - 0.2239 Four samples of 99.10% $^{239}\text{PuO}_2$ sealed in high purity nickel
- - 0.2249 capsules were irradiated in four different axial positions

in Row-8 of EBR-II for 50 days at a power level of approximately 62 Mw. The samples were analyzed mass spectrometrically for the stable and long-lived fission products of krypton, rubidium, strontium, zirconium, molybdenum, ruthenium, xenon, cesium, barium, lanthanum, cerium, neodymium, and samarium. The number of fissions was determined using the summation technique. The sample corresponding to $E_n = 0.2215$ was irradiated in the axial blanket of Row-8; hence, the sample was exposed to a softer spectrum.

- - 0.2254

□ - 0.2281

□ - 0.2299

L. Koch¹¹ - TACO Experiment, RAPSODIE Reactor. Three separately encapsulated samples of enriched ^{239}Pu were inserted into standard rods of a RAPSODIE fuel bundle and irradiated to an approximate fluence of 10^{23} n/cm^2 , during 1970-1971. The samples were analyzed mass spectrometrically at the Institute for Transuranium Elements, Karlsruhe, for the stable and long-lived isotopes of xenon, cesium, neodymium, and samarium. The number of fissions was established using the summation technique. The results for the three samples are presented separately. The corresponding Nd energy index is given at the left.
- - 0.2286

V. Ya. Gabeskiriya, et.al.,¹² USSR BOR-60 Reactor. Two ^{239}Pu targets were irradiated and analyzed for fission product cesium, cerium, and neodymium. The measurement technique was isotope dilution mass spectrometry. The fission yields were calculated by normalization to the sum of the yields of ^{145}Nd plus ^{146}Nd (6.85%) as reported in Reference 17. No independent measurement of the number of fissions was made.
- - 0.2290

V. M. Sinclair, W. Davies,¹³ Dounreay Fast Reactor (DFR). A sample of 94% enriched $^{239}\text{PuO}_2$ was irradiated for a period of approximately 18 months, giving approximately $5^a/\text{oF}$. Mass spectrometric measurements were made for the isotopes of neodymium. Radiochemical data are given for ^{90}Sr , ^{137}Cs , and ^{144}Ce .

The number of fissions was determined from the fission loss of ^{239}Pu . In general, all of the yield data for the neodymium isotopes appear reasonable, except the value for mass 144, which appears low relative to other measurements.

Δ - 0.2290

B. F. Rider,⁴ evaluated fast reactor yield data. This is essentially the US ENDF/B-V(e) data file. For this correlation study, it was assumed that all of the evaluated data were from a single source, corresponding to a Nd energy index of $E_n = 0.2290$.

□ - 0.2296

M. Robin, et.al.,¹⁴ RAPSODIE Reactor, CEA. The details of this experiment are sparse. Only neodymium data are presented.

● - 0.2300

Rel. Isotopic
data only

W. J. Maeck, A. L. Erikson, R. L. Tromp,¹⁵ EBR-II, Row-4. A sample of 99.38% enriched $^{239}\text{PuO}_2$ sealed in a nickel capsule was prepared by the Argonne National Laboratory (ANL) and irradiated during the period 1967-1970 in Row-4 of EBR-II. After being in storage for several years, the sample was transferred to our laboratory in 1979 for analysis. Because of problems encountered in the dissolution of the sample, absolute fission yield data were not obtained for this sample. However, there was no apparent evidence that isotopic fractionation of the fission products occurred during the dissolution. Therefore, that 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 EBR-II Row-4 irradiation.

□ - 0.2420

M. Robin, et.al.,¹⁶ RAPSODIE Reactor, CEA. Small samples of ^{239}Pu (approximately 98% enriched) were irradiated in a special pin in the fast reactor RAPSODIE. The amount of neodymium formed was measured and the neodymium yields for ^{239}Pu were calculated relative to those for the fast fission of ^{235}U .

● - 0.2465

F. L. Lisman, et.al.¹⁷, EBR-I, Idaho National Engineering Laboratory. A ^{239}Pu fuel rod (90% Pu - 10% Al, clad in Zircaloy - 2) was mechanically declad and fuel subsequently dissolved for analysis. Isotope dilution mass spectrometry was used to measure the stable and long-lived fission products of krypton, rubidium, strontium, zirconium, molybdenum, ruthenium, cadmium, xenon, cesium, barium, cerium, neodymium, and samarium. The number of fissions was established by summing the number of atoms in the heavy mass peak.

14.7 MeV

K. A. Pietrzak, et.al.,¹⁸ USSR. Samples of ^{239}Pu were irradiated with 13.7 MeV neutrons. The noble gases were thermally extracted from the target material and mass spectrometrically analyzed using a high sensitivity gas mass spectrometer.

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APPENDIX II

Change in Relative Isotopic Abundance
of the Fission Products as a Function
of Neutron Energy

Figure A-II-1. Relative Isotopic Abundance of Krypton

Figure A-II-2. Relative Isotopic Abundance of Rubidium

Figure A-II-3. Relative Isotopic Abundance of Strontium

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Figure A-II-4. Relative Isotopic Abundance of Zirconium

Figure A-II-4. Relative Isotopic Abundance of Zirconium (Continued)

Figure A-II-5. Relative Isotopic Abundance of Molybdenum

Figure A-II-5. Relative Isotopic Abundance of Molybdenum (Continued)

Figure A-II-6. Relative Isotopic Abundance of Ruthenium

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Figure A-II-7. Relative Isotopic Abundance of Xenon

Figure A-II-7. Relative Isotopic Abundance of Xenon (Continued)

Figure A-II-8. Relative Isotopic Abundance of Cesium

Figure A-II-9. Relative Isotopic Abundance of Cerium

Figure A-II-10. Relative Isotopic Abundance of Neodymium

Figure A-II-10. Relative Isotopic Abundance of Neodymium (Continued)

Figure A-II-11. Relative Isotopic Abundance of Samarium

Figure A-II-11. Relative Isotopic Abundance of Samarium (Continued)

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APPENDIX III

Changes in Fission Yields As a Function of Neutron Energy

Figure A-III-1. Change in Krypton Fission Yields with Neutron Energy

Figure A-III-1. Change in Krypton Fission Yields with Neutron Energy (Continued)

Figure A-III-2. Change in Rubidium Fission Yields with Neutron Energy

Figure A-III-3. Change in Strontium Fission Yields with Neutron Energy

Figure A-III-4. Change in Zirconium Fission Yields with Neutron Energy

Figure A-III-4. Change in Zirconium Fission Yields with Neutron Energy (Continued)

Figure A-III-4. Change in Zirconium Fission Yields with Neutron Energy (Continued)

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Figure A-III-5. Change in Molybdenum Fission Yields with Neutron Energy

Figure A-III-5. Change in Molybdenum Fission Yields with Neutron Energy (Continued)

Figure A-III-6. Change in Ruthenium Fission Yields with Neutron Energy

Figure A-III-6. Change in Ruthenium Fission Yields with Neutron Energy (Continued)

Figure A-III-7. Change in Xenon Fission Yields with Neutron Energy

Figure A-III-7. Change in Xenon Fission Yields with Neutron Energy (Continued)

Figure A-III-8. Change in Cesium Fission Yields with Neutron Energy

Figure A-III-8. Change in Cesium Fission Yields with Neutron Energy (Continued)

Figure A-III-9. Change in ^{138}Ba and ^{139}La Fission Yields with Neutron Energy

Figure A-III-10. Change in Cerium Fission Yields with Neutron Energy

Figure A-III-11. Change in Neodymium Fission Yields with Neutron Energy

Figure A-III-11. Change in Neodymium Fission Yields with Neutron Energy (Continued)

Figure A-III-11. Change in Neodymium Fission Yields with Neutron Energy (Continued)

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Figure A-III-12. Change in Samarium Fission Yields with Neutron Energy

Figure A-III-12. Change in Samarium Fission Yields with Neutron Energy (Continued)

Figure A-III-12. Change in Samarium Fission Yields with Neutron Energy (Continued)