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A study of calculated and measured time dependent delayed neutron yields

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A STUDY OF CALCULATED AND MEASURED TIME

DEPENDENT DELAYED NEUTRON YIELDS

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SUMMARY

Time dependent delayed neutron emission is of interest in reactor design, reactor dynamics, and nuclear physics studies. The delayed neutrons from neutron induced fission of ^{232}U, ^{237}Np, ^{238}Pu, ^{241}Am, ^{242m}Am, ^{245}Cm, and ^{249}Cf have been studied for the first time. The delayed neutron emission from ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu, and ^{242}Pu were measured as well.

The data were used to develop an empirical expression for the total delayed neutron yield. The expression gives accurate results for a large variety of nuclides from ^{232}Th to ^{252}Cf.

The data measuring the decay of delayed neutrons with time were used to derive another empirical expression predicting the delayed neutron emission with time. It is found that nuclides with similar mass to charge ratios have similar decay patterns. Thus the relative decay pattern of one nuclide can be established by any measured nuclide with a similar mass to charge ratio.

A simple fission product yield model was developed and applied to delayed neutron precursors. It accurately predicts observed yield and decay characteristics.

In conclusion, it is possible to not only estimate the total delayed neutron yield for a given nuclide but the time dependent nature of the delayed neutrons as well. Reactors utilizing recycled fuel or burning actinides are likely to have inventories of fissioning nuclides

which have not been studied until now. The delayed neutrons from these nuclides can now be incorporated so that their influence on the stability and control of reactors can be delineated.

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INTRODUCTION

Delayed neutron studies are of interest for three reasons:

(1) accurate delayed neutron values are essential to the design of any reactor system,

(2) the reactivity scale, through which reactors are controlled, is dependent on the effective delayed neutron fraction, and

(3) individual delayed neutron emitters are of great interest by themselves from the nuclear physics point of view. The origin and the energy distribution of delayed neutron emissions can be utilized to explore properties of neutron rich nuclei. Delayed neutron emission is dependent upon cumulative fission yields. It is possible to study the fission process and assess the validity of models which predict the yield of fission products by using a particular fission yield model to calculate delayed neutron yields and compare these with observed values.

Presently, time dependent delayed neutron yields are available only for the following nuclides: ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{252}Cf. These studies, with the exception of ^{252}Cf, were measured in low neutron flux facilities and required samples containing gram quantities. Use of neutron pulses as well

as continuous irradiations and short delay times between irradiation and counting enabled experimenters to measure accurately both short-lived and long-lived group parameters.

In this work, a high flux facility, the Livermore Pool Type Reactor (LPTR), with a thermal and fast flux of 3.5 X 10^{13} and 1.4 X 10^{13} n/cm^2-sec respectively, was used. The high flux which is available allowed use of samples containing milligram quantities. This is particularly significant since many of the samples used needed to be highly pure; consequently they were available only in milligram or microgram quantities. The LPTR was used to study delayed neutron emission from several nuclides which had not been studied. It was also used to study the delayed neutron data of several previously measured nuclides.

Chapter I provides a background of previous work in the fields of delayed neutron emission, individual precursor studies, and fission yield measurements. Chapter II outlines the equations which govern delayed neutron emission and also discusses fission product yield models. Chapter III outlines the best previous experimental method and the facilities used in this work. This chapter also includes the analytical methods used and the data obtained. Chapter IV uses the experimental data to develop an empirical delayed neutron yield model. Chapter V developes the delayed neutron prediction model used in this work and compares its results with observed values. Chapter VI contains the conclusions drawn. The computer programs developed for this work are included in Appendix A.

CHAPTER I

BACKGROUND

A. Delayed Neutron Emission

Delayed neutron emission is a complicated process. There is no barrier to prevent this emission other than the binding energy of the neutron. If neutron emission is energetically possible it proceeds immediately. If an atom (delayed neutron precursor) undergoes a beta decay which leaves the daughter in a highly excited state, immediate neutron emission can follow provided the excitation energy of the daughter exceeds the neutron binding energy. Thus the appearance of the neutron is controlled by the beta decay of the parent (precursor). Not all decays, however, will lead to neutron emission because beta decay can leave the daughter product in a large variety of excited states. The fraction of decays which do result in neutron emission is known as the delayed neutron probability for that emission and it is denoted by P_n.

In fission, one can calculate the yield of delayed neutrons from one particular precursor (^{87}Br for example) by multiplying the cumulative fission yield by the value of P_n for that precursor. By multiplying the fraction of fissions producing the nuclide in

question by the probability of that nuclide producing a delayed neutron and summing over all precursors, one obtains the observed delayed neutron yield from the fissioning material. To calculate the delayed neutron yield from some other material one needs only multiply the fission yields from that material by the appropriate P_n values. Delayed neutron yields dependend only upon the cummulative fission yields of the precursors in each fissioning nuclide.

In practice this is difficult to do. Over 100 delayed neutron precursors have been identified and it has only been recently that accurate P_n values became available. Furthermore accurate cumulative fission yields are quite rare except for thermal fission of ^{235}U. Thus actual calculation of delayed neutron yields is quite complicated.

For practical analysis of delayed neutrons it was found that the sum of six exponentials can be used to fit the decay characteristics with time of delayed neutrons. The underlying reason is that each individual precursor will decay exponentially and the decay of several precursors of similar half-lives can be closely approximated by one exponential with an "average" half-life. Thus depending upon the uncertainty in the experimental data some small number of artificial groups with effective half-lives will approximate the actual decay. The "best fit" may be found by a statistical approach. By increasing the number of groups a closer

fit to actual data can be obtained but at the cost of more variables (degrees of freedom). The standard deviation, SD, is defined as:

$$SD=\sqrt{\frac{1}{N-k}\sum_{i=1}^{N}\frac{[Y(i)_{obs}-Y(i)_{calc}]^2}{Y(i)_{obs}}}$$

where $Y(i)_{obs}$ is the ith observed data point and $Y(i)_{calc}$ is the corresponding calculated value using a specific number of groups, can be used as a criterion for determining the "best" number of groups. The number of groups which gives the lowest standard deviation is by definition the "best". There are N data points and k independent variables in the model. For a six-group model, there are six yields and six half-lives or twelve independent variables.

If the measured data are extremely accurate, one expects that the number of groups needed to give the "best" fit would be large, approaching the number of precursors. Practically however, the "best" fit to a particular data set is in the range of three to six groups. The uncertainty of the data and the acutal yield of the various precursors in a specific isotope combine, perhaps not uniquely, to produce the number of delayed groups for the "best" fit. ^{235}U fission may require six groups while ^{252}Cf may require only three groups. It appears that in ^{252}Cf, the lighter mass fission fragments are heavier than corresponding fragments in ^{235}U and this shift reduces the number of precursors.

Finally if a delay between irradiation and counting exists short-lived precursors may not be seen and so will not require another group. There is no magic number of groups (except one for each actual precursor) which best fits any observed fissioning material.

B. Time-Dependent Unseparated Delayed Neutron Yield Studies

Soon after the discovery of fission, Roberts et al[1] reported the existence of neutrons which made their appearance several seconds after the fission events. Roberts[2] concluded that these neutrons were not photoneutrons caused by gamma radiation from the fission products. A uranium blanket was placed around the fissioning sample and no additional neutrons were observed. In addition the decay periods of the gamma radiation groups did not correspond to the observed delayed neutron half-life; thereby proving the delayed neutrons were not related to the gamma radiation resulting from fission. Bohr and Wheeler[3] explained the existence of delayed neutrons as neutron emission from highly excited beta decay daughters of the origional fission products. Zeldovich and Kariton[4] first suggested the importance of delayed neutrons on the stability of possible critical systems in 1940.

During and after World War II, delayed neutron studies were confined to ^{235}U and ^{239}Pu because of their importance in reactor control

and weapons design. In 1947 Wilson[5] placed ^{235}U and ^{239}Pu samples near a BF_3 counter inside a paraffin moderating block and irradiated the samples with cyclotron neutrons. Irradiation and counting intervals were alternated every 5 minutes. During the counting intervals delayed neutron counts with time were observed. Snell[6] did similar work with natural uranium and was able to estimate the effect of ^{238}U fission by using fast and thermal neutrons. Similar experiments using ^{235}U and ^{239}Pu in graphite thermal columns were also done by De Hoffman and Feld[7], Redman and Saxon[8], and Hughes et al[9]. De Hoffman and Feld reported a delay of 2 seconds (transit time) between the end of irradiation and the beginning of counting. Hughes et al. controlled sample irradiation by a beam shutter and were the first to report very short-lived delayed neutrons.

With the growth of the nuclear industry more interest was shown, beginning in 1950, in other fissioning systems. Sun et al[10] irradiated natural uranium and thorium with neutrons from 15 MeV deuterons hitting carbon, B_4C, and LiF targets. Brunson[11] irradiated ^{232}Th, ^{233}U, ^{235}U, ^{238}U, and ^{239}Pu in the fast neutron core and the thermal neutron reflector of EBR-1. From the beginning the time-dependent decay pattern was characterized by a simple few group model as described earlier. Only the 4 longest delayed neutron groups were seen by Brunson. Rose and Smith[12] used the same nuclides in the fission spectrum of ZEPHYR and observed the 5 longest delayed neutron groups.

Keepin et al.[13] conducted a series of experiments at Los Alamos in 1956 which finally supplied detailed information for ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu. These experiments are described in detail later.

Cox et al.[14] measured delayed neutron emission from ^{252}Cf spontaneous fission by allowing fission fragments to be embedded in a rapidly moving tape and counting the neutron activity of the tape. The results showed that three groups, and not the six groups observed in ^{235}U fission, would characterize the delayed neutrons from this isotope adequately.

Moscati and Goldemberg[15] irradiated ^{232}Th and ^{238}U with gamma radiation pulses from a betatron and were able to study delayed neutrons from photofission. They suggested an empirical correlation between the delayed neutron yield and the quantity (A_c-3Z_c), where A_c and Z_c were the composite mass and charge of the fissioning nuclide. Nikotin and Petrzhak[16] did similar work for ^{232}Th, ^{235}U, ^{238}U and ^{239}Pu. For ^{238}U photofission the delayed neutron yield was found to be independent of the photon energy from 10 to 15 MeV. Caldwell et al.[17] reached the same conclusion for photofission between 6.8 and 9.4 Mev. Caldwell and Dowdy[18] studied photofission of ^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, and ^{239}Pu and again found no energy dependence in the delayed neutron yields.

Several experimenters used the (D,D) and the (D,T) reactions to provide neutron sources for fission studies of various materials.

Krick and Evans[19] studied fission in ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, and ^{242}Pu induced by neutrons from the $^{7}Li(p,n)$ and (D,D) reactions. These reactions provided neutrons of variable energies. The data indicated that the delayed neutron yield does not depend upon the neutron energy causing the fission between 0 and 5 MeV. Above 5 MeV the neutron yield appears to decrease. The results for 14 MeV neutrons are ambiguous; some report higher yields and some lower yields than from low energy neutrons.

A good summary of the work done with delayed neutron yields was given by Tuttle.[20]

C. ^{238}U Delayed Neutron Yield Controversy

Among the values reported by Keepin[13] for absolute delayed neutron yields was the absolute yield of ^{238}U fast fission. His other data agreed with prior and subsequent work but the absolute yield of ^{238}U differed significantly.

Work by Evans[19,21] indicated a higher yield even after corrections for miscalibration of the ^{99}Mo yield. The 17% disagreement was for some time unresolved and even today many investigators feel uncertain as to the true yield value. Subsequent studies seemed to confirm the higher value but it is important to try to discover the cause of Keepin's lower number.

Keepin relied on ^{99}Mo counting to determine the number of fissions in his samples. It appears that the same value of ^{99}Mo yield[22] was used for ^{235}U and ^{238}U fast fission. Keepin's ^{235}U

value is correct and agrees very well with all other experimenters.[20] Meek and Rider (1977) give a value of ^{99}Mo yield for ^{238}U which is 1.08±.03 times larger than that for ^{235}U. Thus Keepin's value would appear to be too low by this amount. If his value is corrected by this amount it agrees excellently with all other published values for ^{238}U fission. Thus there appears to be agreement on the delayed neutron yield in ^{238}U fast fission. Table 1 shows the published values and a mean value weighted by the quoted uncertainties of the individual reported values. The values are taken from Tuttle's paper.[20] The yield from fast neutron fission of ^{238}U is 4.44±.23 neutrons per 100 fissions.

D. Individual Precursor Studies

Due to the difficulty of extracting individual precursors with half-lives of the order of a few seconds, very little has been reported about individual delayed neutron precursors. Snell et al.[24] reported preparing fission products which were subjected to several chemical precipitations. They found delayed neutrons associated with the halogen precipitates. There was a delay between irradiation and separation of from 30 to 60 seconds. The half-lives of the two components (56 seconds and 23 seconds) corresponded with the two longest observed components of unseparated delayed neutron precursors. In a different experiment they found bromine and iodine could be separated from fission products in carbon tetrachloride and then by careful oxidation, separation of bromine and iodine was achieved.

Table 1 Reported ^{238}U Delayed Neutron Yields.[20]

INVESTIGATOR	NEUTRON ENERGY	YIELD (neut./100 fissions)
Keepin (1957)	Fission	4.12±.25 (excluded)
Tomlinson (1972)	Fission	4.40±.21
Manero & Konshin (1972)	Fission	4.37±.12
Cox (1974)	Fission	4.60±.25
Brunson (1955)	2.7 MeV	4.76±.74
Maksyutenko (1959)	2.4 MeV	4.37±.35
Maksyutenko (1959)	3.3 MeV	4.15±.38
Masters et al (1959)	3.1 MeV	4.84±.36
Cox & Whiting (1970)	0.9-2.4 MeV	4.46±.29
Clifford (1972)	1.8 MeV	4.72±.25
Cox (1974)	2.0 MeV	4.39±.26
Cox (1974)	3.0 MeV	4.35±.26
Keepin (adjusted)	Fission	4.45±.30
This Work (1980)	Fission	4.65±.35
MEAN WEIGHTED BY UNCERTAINTIES		4.44±.23

The 54 second activity was found in the bromine fraction and a 23.8 second activity found in the iodine fraction. The delay between irradiation and counting was about 30 seconds and the shorter lived bromine neutron activity was assigned to iodine impurities rather than to a different bromine isotope. ^{87}Br and ^{137}I were tentatively identified as the precursors because the half-lives of these nuclides had been roughly measured previously.

Sugarman[25] demonstrated that the 4.51 second delayed neutron component was due to a bromine isotope and the 1.52 second activity was due to an iodine isotope. This was done by studying the range of fission products in bakelite (light fission products have a longer range) and measuring the decay on a rapidly moving tape. Sugarman[26] also accurately studied the half-lives of ^{87}Br, ^{88}Br, ^{137}I, ^{138}I, and ^{139}I by milking known descendants from silver halide precipitates. In a milking process a purified sample is allowed to decay and later analyzed for decay products. The half-life with which these daughters appear gives the half-life of their parents. There was a 7 second delay after irradiation in this work.

It was long believed that only six delayed neutron precursors existed.[27] This belief was natural in view of the similarity of the half-lives of the 6 group parameters measured by Keepin and others for a variety of fissioning nuclides. Pappas[28,29] was the first to indicate that the group half-lives were due to combinations of multiple precursors.

Because they are easily separated from a target, gaseous and volatile fission products have been studied extensively. Stehney and Sugarman[30] used a gas sweeping technique in which bromine carrier was used to carry bromine fission products to a solution where they were precipitated with silver. By this technique they established the fission yield of ^{87}Br. Perlow and Stehney[31] improved the technique by decreasing the time between irradiation and counting, and thereby established the 15.5 second activity of ^{88}Br. Further modifications by Perlow and Stehney[32] using a gas burst and fast shutter established half-lives and relative yields for ^{87}Br, ^{88}Br, ^{89}Br, ^{90}Br, ^{137}I, ^{138}I, and ^{139}I. The same authors[33] used the same technique to study rare gases and established the 1.5 second krypton and 6 second rubidium as neutron emitters but the yield of neutrons from these fission products is extremely low. The krypton fission product contribution is less than 0.5% of the total. Neutron emission from xenon fission products was too low to be observed.

Tomlinson[34,35] formed volatile hydrides of antimony, arsenic, tin, and germanium which were carried by helium to a heated tube where the hydrides decomposed with a delay of 6 seconds after irradiation. The precursors thus established were ^{135}Sb, ^{85}As, and ^{86}As or ^{87}As. Del Marmol and Neve de Mevergnies[36] used a similar technique to establish that ^{85}As, with a half-life of 2.15 seconds, contributed 4% of all delayed neutrons.

Hermann et al.[37] extracted halogen precursors, as discussed above, from fission products and studied the remainder. They established that 2%, 8%, and 20% of the 55 second, 22 second, and 6 second delayed neutron groups were not from halogen precursors.

On-line mass spectrometers are well suited to delayed neutron precursor studies. Such instruments are quite capable of analyzing nuclides with half-lives of a fraction of a second and make possible the determination of the mass and charge. Amarel et al[38] identified ^{93}Rb, ^{94}Rb, ^{95}Rb, ^{142}Cs, and ^{143}Cs as delayed neutron precursors with the Orsay mass spectrometer. Ths fission products were produced in a heated graphite block from which the volatile fission products escaped. The stream of fission products was passed through an electrostatic deflector for charge separation and then curved in a magnetic field for mass separation. In a later paper the same authors[39] identified ^{97}Rb and ^{144}Cs as precursors and provided delayed neutron probabilities (P_n) for several rubidium and cesium nuclides.

Talbert et al[40] used the TRISTAN isotope separator to analyze short lived gaseous fission products. The P_n values for several krypton, rubidium, xenon, and cesium nuclides were determined. Roeckl et al[41] provided similar information for rubidium and cesium nuclides adding P_n values for ^{98}Rb, ^{145}Cs, and ^{146}Cs.

Asghar et al[42] used the Lohengrin mass separator to determine P_n values for ^{90}Br, ^{91}Br, ^{93}Rb, ^{94}Rb, ^{95}Rb, ^{137}I, ^{138}I, and ^{139}I;

and new P_n values for ^{91}Se, ^{94}Kr, ^{99}Sr, ^{99}Y, ^{134}Sn, and ^{138}Te. In this system fission products were allowed to recoil through a mass separator and then deposited on a moving tape. The tape was then neutron and beta counted to determine the ratio of neutron decays to beta decays, (P_n).

The SOLAR[43] mass spectrometer at Battelle-Pacific Northwest Laboratories was used to study rubidium and cesium precursors and to determine P_n values. Here a graphite oven was used in a neutron beam to produce ionized fission products which were charge separated and then mass separated to provide pure samples.

Kratz et al[44] used the volatilization of arsenic hydride to separate arsenic fission products from uranium solutions after fission. This work reported P_n values for ^{84}As, ^{85}As, ^{86}As, and cumulative fission yield for ^{87}As.

In 1973 Tomlinson[45] produced an excellent summary of all reported delayed neutron precursor data which served as a reference for most analyses after that time. Rudstam has, since that time, produced several summaries of delayed neutron precursor characteristics, the most recent of which was presented in March 1979.[46] This work represents the best summary of data currently available. Over 100 delayed neutron precursors have been identified and characterized.

With the advent of good delayed neutron precursor information, several authors have tried to compare observed delayed neutron yields with yields calculated using P_n values and fission yield data. Keepin's attempt[21] in 1965 showed poor agreement over most of the delayed neutron

groups. Schussler and Herrmann[47] in 1972 were able to get reasonable agreement for ^{235}U fission over all 6 groups.

Izak-Biran and Amiel[48] in 1975 were also able to get good agreement for ^{235}U fission. Rider and Meek[49] attempted a calculation of delayed neutron yields for a large variety of fissioning systems using known P_n values. Good agreement was found in some cases (thermal fission of ^{235}U and ^{239}Pu) and poor agreement in others (fast fission of ^{238}U and ^{232}Th). In view of the low quoted uncertainties of recent P_n values (10-20%) and the good agreement of calculated delayed neutron yields for cases where fission yields are well known it would appear that the errors involved in such a calculation come from errors in fission yields instead of errors in delayed neutron probabilities (P_n).

E. Fission Yield Measurements

The fission yield process was identified in 1938 by Hahn and Strassmann[50,51] when they demonstrated that the "activation" products of neutron irradiated uranium included elements of about half the atomic number of uranium (barium and lanthanum). Within a year over 100 papers had been written on the subject from around the world.[52] Bohr[53] suggested that thermal fission in uranium was caused by ^{235}U and with Wheeler[54] developed a liquid drop model which was used extensively to analyze fission.

Fermi[55] was responsible for the first quantitative analysis of fission yields. The technique used was to add a measured amount of a

particular element in a solution of uranium and fission products. After thorough mixing chemical separations were performed until a chemically pure sample of the element in question was obtained. This sample contained some fraction of the initial carrier and a minute amount of fission products of that element. The fractional recovery of the fission products was assumed to be the same as the fractional recovery of the carrier. The sample was then carefully counted to determine its absolute activity and thereby established the cumulative yield. Half-life differences made possible separations by isotope. This procedure was documented in Volume 9 of the Plutonium Project Record.[56] Fission of ^{235}U, ^{233}U, ^{239}Pu, and ^{238}U was analyzed in this way. Initially radiochemical techniques of this type provided all the available chain yield data. Such techniques are, however, limited to the longer-lived fission products and to the chain yield data.

Mass spectrometers have come to play a serious role in fission yield measurements. Such machines make it possible to study much shorter-lived nuclides. A typical approach was the isotope dilution method. In this case fission yields of several isotopes were measured against one standard of known abundance and long half life. For example isotopes of ruthenium were measured relative to ^{106}Ru. This technique was used to provide fission yields for isotopes of strontium, zirconium, molybdenum, cerium, barium, cesium, and neodymium.[57]

Katcoff[58] produced a summary of fission yields as of 1960 (largely radiochemical) and Farrar et al[59] produced a separate fission

yield set, in 1962, from a series of mass spectrometer experiments. While chain yield work is continuing (expecially for transuranic nuclides and for chains of low yield) the emphasis has shifted to independent yield measurements. A discussion of the terms independent yield, cumulative yield, and chain yield is given in the next chapter.

Independent fission yields are, in general, very hard to measure. Primary fission products tend to be very neutron rich and so highly unstable. Because of their very short half-lives it is usually quite difficult to study them. It is hard to separate independent yields from cumulative yields.

In a few cases shielded nuclei exist. These nuclides can not be formed by beta decay because their would-be parents are stable. ^{130}I is such a nuclide. Thus any ^{130}I found in fission products must have come from direct formation in fission.

By rapid separations it is possible in some cases to determine independent yields. If a chemical separation is performed before significant beta decay of the parent occurs the yield observed will be the independent yield of the daughter. The total yield after decay of the parent is the cumulative yield. ^{140}La is an example of this technique since its parent, ^{140}Ba, has a 12.8 day half-life. Unfortunately such cases are limited and tend to be far away from the majority of primary fission products.

Wahl[60] developed a technique for determining the relative cumulative yield of noble gas fission products. Noble gases escape immediately from barium stearate powder. By comparing the amount of

daughters in the stearate powder to those elsewhere in the irradiation container he determined the ratio of fission products which decay through either krypton or xenon and thus excaped the powder. This technique has been used for a large number of fissioning materials and often provides the only cumulative yield information available for these materials.

In summary then, chain yields are known for a large number of chains for almost all fissioning materials. This is because several hours after irradiation the major activity in a sample of fission products are the long-lived members at the bottom of the beta decay chains. Gamma counting can easily determine the abundance of these members. Independent fission yields (and cumulative yields of the nuclides near the beginning of the beta decay chains) are much harder to measure because of the short half-lives involved. Such measurements have been performed using mass spectrometers but only for ^{233}U and ^{235}U thermal fission. The only method available for other fissioning materials is studying the limited number of shielded or noble gas fission products. Thus for almost all fissioning materials very little independent and cumulative yield information is available upon which to base predictions of fission yields.

Better yield information will be slow in coming. Most fissionable materials are quite rare, extremely toxic, and highly radioactive. Such samples must therefore be small and preferably non-destructively analyzed. It is unlikely that the large facilities with sufficiently high fluxes and sophisticated analyzers will be anxious to contaminate

these analyzers with such materials in the near future.

CHAPTER II

THEORY

When an atom fissions a large variety of fission product pairs may be formed. Conservation of charge and mass apply in the following way:

$$(Z_c,A_c) \xrightarrow{\text{fission}} (Z_1,A_1) + (Z_2,A_2) + \nu n,$$

$$Z_c = Z_1 + Z_2 \quad \text{and} \quad A_c = A_1 + A_2 + \nu$$

where Z_c and A_c are the charge and mass of the nuclide when it fissions (ie. neutron induced fission of ^{235}U would give Z_c=92 and A_c=236). Z_1 and A_1 are the charge and mass of one of the fission fragments and Z_2 and A_2 are the charge and mass of the other fragment. ν is the number of prompt neutrons emitted.

In general, fission products are very neutron-rich and quickly undergo beta decay. Beta decay increases the charge of the fission product while leaving its mass unchanged. In general several beta decays occur until the nuclide in question is no longer unstable to beta decay. Thus after one decay or k decays we have:

$$(Z_1,A_1) \xrightarrow{\text{decay}} (Z_1+1,A_1) + \beta^- \quad \text{and}$$

$$(Z_1,A_1) \xrightarrow{\beta \text{ decay}} \cdots \xrightarrow{\beta \text{ decay}} (Z_1+k,A_1) + k\beta^-.$$

For any particular type of fission (such as thermal neutron fission of ^{235}U) there is a certain probability that a given fission product may be formed directly from that fission event. The independent fission yield for a fission product (Z_1,A_1) is the probability that a given fission event will produce directly a fission product of charge Z_1 and mass A_1.

Notice, however, that the fission product (Z_1-1,A_1) will also produce the nuclide of interest, (Z_1,A_1) by undergoing beta decay. The cumulative fission yield for the fission product (Z_1,A_1) is the probability that a fission event will result in the formation of (Z_1,A_1) either directly or via beta decay. Thus the cumulative fission yield for (Z_1,A_1) is just the sum of the independent yields for (z,A_1) where $z \leq Z_1$. The chain yield for mass A_1 is the sum of all independent yields for (z,A_1) for all z. The chain yield is thus the probability of a given fission event producing a fission product of mass A_1. The cumulative yield, $C_y(Z_1,A_1)$ and the chain yield $C_c(A_1)$ are given by the equations:

$$C_y(Z_1,A_1) = \sum_{z=0}^{Z_1} I_y(z,A_1) \quad \text{and} \quad C_c(A_1) = \sum_{z=0}^{\infty} I_y(z,A_1) \quad \text{where}$$

$I_y(Z_1,A_1)$ is the independent yield for the nuclide (Z_1,A_1).

The relative independent yield for the nuclide (Z_1,A_1) is simply the ratio of the independent yield of a nuclide to the chain yield for that mass, A_1. Thus

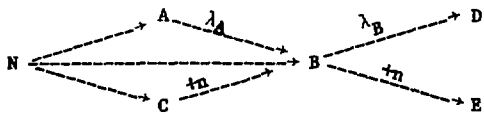
$$RI_y(Z_1,A_1) = \frac{I_y(Z_1,A_1)}{C_c(A_1)}$$

where $RI_y(Z_1,A_1)$ is the relative independent yield. Similarly the relative cumulative yield is the cumulative yield normalized to unity or:

$$RC_y(Z_1,A_1) = \frac{C_y(Z_1,A_1)}{C_c(A_1)} \quad .$$

In general as a fission product undergoes beta decay it becomes more and more stable (that is the half-life of each daughter increases). Because of this it is often possible to neglect the half-lives of the early members of a decay chain since they rapidly decay into the more stable daughters. By measuring the abundance of the longest-lived daughter of a decay chain the chain yield of that chain is found. For this work cumulative yield information on delayed neutron precursors is required. Unfortunately usually only chain yields are available. Fission yield models are used to calculate cumulative yields from observed chain yields.

Imagine a reactor environment in which the a fissioning source, N, is producing atoms of materials: A with a charge and mass of (Z_1-1,A_1), B with a charge and mass of (Z_1,A_1), and C with a charge and mass of (Z_1,A_1-1). In addition material A decays into material B and material C can undergo neutron capture and become B. Material B decays into D whose charge and mass are (Z_1+1,A_1), and can also undergo neutron capture to become E with a charge and mass of (Z_1,A_1+1). This situation is shown as



The differential equation governing this situation is:

$$\frac{dB(t)}{dt} = -\lambda_B B(t) + \lambda_A A(t) + N\sigma_f\phi I_y(B) + C(t)\sigma_C\phi - B(t)\sigma_B\phi$$

where λ_B and λ_A are the decay constants for nuclides A and B. The fission cross-section is σ_f for nuclide N, ϕ is the neutron flux, and $I_y(B)$ is the independent yield of B. The neutron absorption cross-sections for nuclides B and C are σ_B and σ_C.

For the nuclides of interest in this work neutron capture is insignificant. This can be seen since in this work the nuclides of interest have: $\lambda_B \simeq 0.1$ sec^{-1}, $\phi \simeq 10^{13}$ n/cm^2-sec, and $\sigma_B \simeq 10$ b (10^{-23} cm^2). Thus $\lambda_B >> \sigma_B\phi$. The same is true for material C so neutron capture in the fission products will be neglected. There are situations with fission products such as ^{135}Xe and ^{149}Sm where capture can not be neglected but they are of no interest here.

If the parents of A are very short-lived they quickly reach equilibrium so that

$$\frac{dA(t)}{dt} = -\lambda_A A(t) + N\sigma_f\phi C_y(A)$$

where $C_y(A)$ is the cumulative fission yield of A (remember all of A's parents quickly decay into A). Therefore

$$A(t) = \frac{N\sigma_f\phi C_y(A)}{\lambda_A}\left[1-e^{-\lambda_A t}\right].$$

For the nuclide B the equation is

$$\frac{dB(t)}{dt} = -\lambda_B B(t) + N\sigma_f\phi I_y(B) + N\sigma_f\phi C_y(A) - N\sigma_f\phi C_y(A)e^{-\lambda_A t}$$

and noting that $C_y(B) = I_y(B) + C_y(A)$ gives the equation:

$$B(t) = N\sigma_f\phi C_y(B)\,\frac{[1-e^{-\lambda_B t}]}{\lambda_B}\,\left[1-\frac{C_y(A)}{C_y(B)}\,\frac{\lambda_B}{\lambda_A-\lambda_B}\,e^{-\lambda_A t}\right] .$$

For the very neutron-rich fission products of interest here we find

a) $C_y(B) >> C_y(A)$ and

b) $\lambda_A \simeq 10\lambda_B$ (note $^{87}Se \rightarrow ^{87}Br$, $^{88}Se \rightarrow ^{88}Br$, and $^{137}Te \rightarrow ^{137}I$, etc.)

Therefore

$$B(t) \simeq N\sigma_f\phi C_y(B)\,\frac{[1-e^{-\lambda_B t}]}{\lambda_B} .$$

After an irradiation time, t_o, and a period of decay, t',

$$B(t') = N\sigma_f\phi C_y\,\frac{[1-e^{-\lambda_B t_o}]}{\lambda_B}\,e^{-\lambda_B t'} .$$

The rate of decay of $B(t')$ is given by $\lambda_B B(t')$ and, since $P_n(B)$ is the probability of delayed neutron decay, the activity of delayed neutrons from the decay of B is given by

$$DN(t') = N\sigma_f\phi C_y(B)[1-e^{-\lambda_B t_o}]\,e^{-\lambda_B t'}P_n(B)$$

The delayed neutron activity (in neutrons per second) for all delayed neutron precursors is given by the summation:

$$DN(t') = \sum_B N\sigma_f\phi C_y(B)[1-e^{-\lambda_B t_o}]\,e^{-\lambda_B t'}P_n(B)$$

where the variable L covers all delayed neutron precursors.

For a continuous irradiation ($\lambda_B t_o$) $\rightarrow \infty$ where the sample is counted immediately after irradiation ($\lambda_B t'$) $\rightarrow 0$ we have

$$DN(t'=0) = \sum_B N\sigma_f\phi C_y(B)P_n(B) \ .$$

Counting a sample immediately after a burst irradiation implies:

$$[1-e^{-\lambda_B t_o}] \rightarrow (\lambda_B t_o) \quad \text{and} \quad (\lambda_B t') \rightarrow 0 \quad \text{so that}$$

$$DN(t'=0) = \sum_B \lambda_B t_o N\sigma_f\phi C_y(B)P_n(B) \ .$$

Notice in this case the extra factor of λ_B. Thus short-lived delayed neutron emitters (where λ_B is large) are emphasized in a burst irradiation while a continuous irradiation emphasizes the longer-lived precursors. This is reasonable because the short-lived precursors will approach equilibrium much faster than will the long-lived ones.

Often the limitation on data taking in an experiment is the dead-time of the counter. Thus the sample activity must be limited. A given delayed neutron activity can be obtained by continuous irradiation at low power or a short irradiation at high power. In the former case the long-lived precursors are emphasized and in the later case the short-lived precursors are emphasized.

There are a large number of delayed neutron precursors, but it has been found that six artificial groups may be used to accurately reproduce the observed delayed neutron decay. In this case

$$DN(t') = \sum_{i=1}^{6} A_i N\sigma_f\phi[1-e^{-\lambda_i t_o}] \ e^{-\lambda_i t'}$$

where λ_i is the decay constant for group i. The absolute group yield, A_i, is the probability that a fission event will produce

a delayed neutron in group i. The units of $DN(t')$ are therefore neutrons per second.

Group I corresponds exactly to ^{87}Br so that $A_I = P_n(^{87}Br) \cdot C_y(^{87}Br)$. In general one would expect the summation of A_I over all groups to give the total delayed neutron yield. This is in fact the case. Notice again that a burst irradiation will emphasize the shorter-lived groups and a continuous irradiation will emphasize the longer-lived groups.

Prediction of independent and cumulative fission yields is very difficult due to the lack of data in all cases except ^{235}U and ^{233}U thermal fission. Attempts have been made however. Coryell et al[62] noted that if independent yields for a given mass chain were plotted as a function of the charge of the fission product, Z_I, the curve could be fitted by a Gaussian function. The peak of the curve defines the most probable charge, Z_p. This value is in general not an integer and is essentially the average charge of the fission products of a given chain. Therefore

$$Z_p(A_I) = \sum_{z=0}^{\infty} z\ RI_y(z,A_I)$$

where z is the charge of each fission product and all are of mass A_I. RI_y is the relative independent fission yield for that particular nuclide.

A nuclide which is neutron-rich will undergo β^- decay. A proton-rich nuclide will undergo β^+ decay. Therefore, in general

there exists one charge, Z_a, which is most stable for a given mass chain, A_1. Occassionally it is observed that there are two stable (non-radioactive) members to a chain (eg ^{128}Te and ^{128}I). In this case ^{128}Te would decay to ^{128}Xe except that it must pass through ^{128}I first and this is energetically unfavorable because of the even-odd effect which is discussed below. The systematics of beta decay are described in detail by Friedlander et al.[62]

In any case it is possible to assign a charge, Z_a, to a given chain which is the most stable charge in that chain. Coryell suggested that the quantity (Z_p-Z_a) was the same for both the light and heavy fission fragments. This is the equal charge displacement model and in essence it suggests that a fissioning atom will divide in such a way that both fragments are equally unstable to beta decay and both will undergo an equal number of beta decays before attaining stability. This is, of course, a statistical model that must be averaged over a large number of fissions since it is not possible to speak of a non-integer number of decays in any particular fission event. What little independent fission yield data which were available tended to confirm this hypothesis.

Wahl[60] compiled all available yield data (including to a large extent his own noble gas yield data) and was able to empirically determine Z_p values. Wahl assumed relative independent fission yields,

$$RI_y(Z,A) = c \int_{Z-.5}^{Z+.5} \exp[-\frac{(z-Z_p)^2}{2\sigma_z^2}] \, dz$$

where c is a normalization constant so that the sum of all relative independent yields in a given chain is unity. Wahl found σ_z=0.56

provided the best fit of the data. Thus for a given fission yield this equation could be solved for Z_p. By this method he calculated Z_p value for most mass chains in ^{235}U thermal fission. These values generally confirmed the equal charge displacement model but were useful in themselves, because they were experimentally determined. Other authors have confirmed the accuracy of this approach.[63,64]

Recently, mass spectrometer work has contributed greatly to the independent yield data available. Clerc et al[65] have published relative independent fission yields for several mass chains from 90 to 104. In their experiment fission products escaped from the irradiation foil and were mass separated by a magnetic field. The separated fission products then were absorbed in a stack of carbon foils and the energy deposited was related to the charge on the fission products. Siegert et al[66] produced similar data from the same facility at Lohengrin by studying the energy loss in a thin silicon-barrier detector. These data provided charge distributions from mass 79 to 100.

With the large data base for ^{235}U thermal fission it was possible to establish a complete set of Z_p values for this fissioning isotope. These values have been reproduced in several reports but a common set is listed in Rider and Meek.[49]

Nethaway[67] proposed an empirical method of calculating Z_p for the fissioning nuclides based on the limited fission yield data available for other nuclides and excitation energies. Rider and Meek

published very complete tables of fission yields (both experimental and calculated) based on a Gaussian distribution and Nethaway's model. The model consists basically of calculating the change in Z_p due to changes in charge, mass, and excitation energy of the fissioning nuclide away from the reference values for thermal fission of ^{235}U. Thus

$$\Delta Z_p = a(Z_c-92) + b(A_c-236) + c(E^*-6.52)$$

where Z_c, A_c, and E^* are the composite charge, mass, and excitation energy of the fissioning material and a, b, and c are constants determined from experimental data. From conservation of charge and mass one would expect Z_p to change as Z_c and A_c change from material to material. The excitation energy is important in an indirect way. Prompt neutron emission increases as the excitation energy increases. Conservation of mass therefore forces the fission fragments each to have less mass. This causes a shift in Z_p in the same way that a decrease in A_c causes a shift in Z_p. On the average 7 MeV of excitation energy increases prompt neutron emission by about one. One could then say, in a sense, an increase in excitation energy of 7 MeV decreases A_c by one. Thus one would expect 14 MeV fission to have somewhat different fission yield distributions than thermal fission would have.

It is interesting to note that while prompt neutron emission is strongly dependent upon excitation energy delayed neutron emission is not. The more excitation energy in the fission event the more

highly excited are the fission products and therefore the more likely that the binding energy of the neutron will be exceeded and prompt neutron emission will occur. However the excitation energy of the fission fragment is rapidly lost either by neutron emission or gamma emission. The fission fragment is therefore in its ground state long before it undergoes beta decay (which is a much slower process). Delayed neutron emission depends only on the energetics of beta decay from the ground state of the precursor. Any excitation the precursor possessed was lost long before beta decay occurs.

However prompt neutron emission decreases the excess of neutrons which exist in fission fragments. Statistically speaking, an increase in excitation energy increases prompt neutron emission which decreases the yield of neutron-rich fission products (such as delayed neutron precursors). This decrease in the yield of precursors is reflected in a decrease in the yield of delayed neutrons. As an example, if a sample of ^{240}Pu is given an extra 7 MeV of excitation energy in fission its prompt neutron emission will increase by one and it will in many respects be the same as low energy fission of ^{239}Pu. Since ^{239}Pu is observed to have fewer delayed neutrons than ^{240}Pu one would expect high energy fission of ^{240}Pu to decrease the delayed neutron yield. This simple analysis would predict a decrease of about 5% in the delayed neutron yield per MeV of additional excitation energy.

It is approximately true to say that fission products are

formed with the same mass-to-charge ratio as the fissioning material. Thus

$$\frac{A_1}{Z_{1p}} \simeq \frac{A_2}{Z_{2p}} \simeq \frac{A_c - \nu}{Z_c} \qquad \text{where}$$

$A_c - \nu$ is the composite mass of the fissioning material minus the average number of prompt neutrons emitted. Z_{1p} and Z_{2p} are the most probable charges of the two fission products.

Delayed neutron emission is always associated with fission products that have a large mass-to-charge ratio. This is because if the ratio is large the neutron binding energy is low and the beta decay energy is high which is needed to have delayed neutron emission. The higher this ratio is the more likely delayed neutron emission (P_n) becomes. It is for this reason that for a given element, Z_1, the P_n values increase as the mass is increased.

Cumulative yields, C_y, for a given mass, A_1, tend to decrease as the charge is decreased (since each beta decay increases the charge). Therefore for a given charge, Z_1, cumulative fission yields decrease as the mass, A_1, is increased. Delayed neutron emission is dependent upon the product of P_n and C_y. Since, for a given element, P_n increases with mass but C_y decreases the delayed neutron contribution will first increase and then decrease as the mass is increased.

If the mass of the fissioning material is increased while keeping the charge constant (eg. $^{235}U \rightarrow {}^{238}U$) the mass-to-charge ratio of the fission products will increase. Therefore the cumulative yields of precursors will increase and an increase in delayed neutrons is seen.

In general a fissioning material produces one fission product with a mass of about 90 and another with a mass of about 140. Delayed neutron emitters also appear to be concentrated in these mass regions. For this reason the light and heavy mass peaks both produce significant delayed neutron emission.

It has been found, however, that as one increases the mass of the fissioning material the heavy mass peak remains at about 140 while the light mass peak increases correspondingly. Therefore in heavy nuclides such as ^{245}Cm and ^{252}Cf the light fission yield peak is shifted away from the light delayed neutron precursors. For this reason delayed neutron emission from the light mass peak decreases dramatically as the mass of the fissioning material is increased.

A topic of recent interest is the "even-odd" effect. In a fissioning material with an even number of protons it has been seen that fission products with even charge are more abundant than those of odd charge. This is due to the extra energy required to break a proton-proton bond to provide two odd fission products. The effect is expected to be most obvious in materials with low excitation energy in fission. The effect should be insignificant as the excitation energy becomes large compared to the 1.7 MeV proton-proton bond. [68] The same effect is not expected to be significant with neutron pairing due to the emission of prompt neutrons from the fission products.

Amiel et al[69] summarized the experimental evidence for such an effect and indicated several fissioning materials where the effect appeared to occur. If one calculated the expected yield of a fission product (using the methods outlined above) one noticed that the evenly charged fission products were more abundant by a factor of (1+a) and the odd nuclides less abundant by a factor of (1-a), where "a" was the even-odd effect. For instance in ^{235}U thermal fission the effect appeared to be about 22±7%[69].

The even-odd effect has significant effect upon the delayed neutron yield which can be calculated for any fissioning material. Since almost all delayed neutron precursors have an odd charge the larger the even-odd effect the smaller the calculated delayed neutron yield would be.

Izak-Biran and Amiel[71] found the calculated delayed neutron yield for fast fission of ^{235}U was too large if no even-odd effect was assumed and too low if a 10% effect was assumed. This roughly agrees with the observed even-odd effect for fast ^{235}U fission of 10±10%.[69] For fast fission of ^{233}U the even-odd effect needed to give agreement in delayed neutron yields was somewhat more than 10%. Alexander and Krick[72] also noted the result of various even-odd effects and found reasonable agreement in ^{235}U fission by assuming a 25% effect for thermal fission, a 10% effect for 2 MeV neutron induced fission, and no even-odd effect for 3.3 MeV neutron induced fission.

Because of the lack of experimental independent and cumulative

yields for almost all fissioning materials fission models must be relied upon. The accuracy of such models, especially in view of the even-odd effect, is extremely suspect however.

CHAPTER III

EXPERIMENTAL METHODS

Delayed neutrons are studied by inducing fission through either a short neutron burst or continuous irradiation over a specified time span. Pulsed studies emphasize the short-lived components and continuous irradiations emphasize the long-lived components. Because of the presence of prompt fission neutrons the delayed neutron precursors must be separated from continuing sources of fission. This may be accomplished in several ways.

One clever method used by Cox et al.[14] was to capture fission product recoils from ^{252}Cf spontaneous fission on a rapidly moving tape and to count the delayed neutrons far from the californium source.

Another method is to count the sample in place but to remove the source of neutrons causing fission. This is easily done with cyclotron pulses of neutrons or gamma rays. Hughes used a rotating shutter to stop a thermal neutron beam which was used to cause fission. Such approaches cause negligible delays between the end of irradiation and the start of counting and are capable of detecting extremely short-lived precursors. The main problems are low intensity, which causes poor counting statistics,

and questions about the excitation energy of the fissioning system. Since delayed neutrons are of practical value in fission reactors data collected from photofission or high energy particle fission are not of direct use.

The most often used approach has been to rapidly transfer the fissioned sample away from the irradiation point to a low background counting station. Keepin[13] used a system which transferred the sample a quarter of a mile in 50 msec. Unless such a fast shuttle is used the shortest-lived delayed neutrons will be lost. Because Keepin's system possessed the best of all combinations, his data remain the standard against which all other work is compared*. The only two materials which were not studied by Keepin and have been reported elsewhere are ^{252}Cf and ^{241}Pu reported by Cox.[14,73]

A. Keepin's Work

R. Keepin[13] conducted his work at the Los Alamos Godiva facility. This pulse reactor was capable of producing neutrons in fast burst or continuous operation. In either case the sample underwent the same number of fissions (about 3×10^{12} fissions per irradiation). For each nuclide studied a sample was prepared and repeatedly exposed to either a burst irradiation or a continuous

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*This is only true for relative time dependent yields. A large number of total delayed neutron yields have been published. In particular Keepin's ^{238}U total yield value appears to be in error.

irradiation for a total of 40 of each, except for ^{235}U fast fission in which 80 bursts and continuous irradiations were used.[21] The two types of irradiations enabled Keepin to establish small uncertainties on both his long-lived and short-lived groups. Each sample consisted of a few grams of material.

The neutron spectrum of the Godiva reactor was a slightly degraded fission spectrum. The samples studied were ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu. A large polyethylene block was used to moderate the Godiva neutrons for thermal fission of ^{233}U, ^{235}U, and ^{239}Pu. Because the block was located away from the center of Godiva the thermal neutron studies had lower fluxes and hence higher uncertainties in the group parameters derived.

Each sample was delivered to a 4π counting system after irradiation in about 50 msec. Thus essentially no correction for transit time was needed. There was no evidence of groups with half-lives shorter than 0.2 seconds or longer than 56 seconds. The counter used was a $^{10}BF_3$ proportional counter modified to be energy insensitive within 5% from 23 keV to 1.5 MeV. The counter's dead-time was measured to be about 1 μsec.

B. Experimental Set-Up Used In This Work

Measuring delayed neutron yields from transuranic nuclides which are only available in small quantities made it necessary to utilize a high flux neutron facility so that good statistical accuracy

could be obtained. By comparison, Keepin's work required gram quantities of material because of the low neutron flux available to him. Typical samples used in this work ranged from about a microgram for nuclides with large thermal fission cross-sections to several milligrams for materials with low thermal fission cross-sections. The Livermore Pool Type Reactor (LPTR) was well suited for this work. A pneumatic system whose irradiation head was in a thermal flux of 3.5×10^{13} neutrons/cm^2-sec was a convenient auxiliary system of the reactor facility. The flux of neutrons above 1 MeV was approximately 1.4×10^{13} neutrons/cm^2-sec. Such fluxes were often sufficient to cause saturation of the delayed neutron counter on sample arrival. In these cases the sample was irradiated at high power and counted and then later irradiated at lower power and recounted to provide the data missed by the previous run because of counter saturation. Each irradiation was for 90 seconds (the irradiation limit of the sample container).

After neutron counting for 800 seconds the sample was pneumatically transferred to a GeLi detector and the gamma radiation from the unseparated fission products was analyzed. In each case a standard of 93% enriched ^{235}U was also irradiated and counted to provide calibration of the absolute delayed neutron yield of the sample. Fission yields were used to calculate the ratio of total fissions in the sample relative to the standard for each of the gamma emitters studied.

A fission fragment distribution of any sample with a significant thermal fission cross-section was considered to have been due to thermal fission. This is because fast fission cross-sections are normally of the order of a barn and thermal cross-sections are large and can be well over 1000 barns. The high energy neutron spectrum of the reactor was a degraded fission spectrum. The exact average fission neutron energy for fast fission samples varied with the fission threshold of each sample but studies have indicated that delayed neutron yields have very little dependence on the neutron energy. A fission spectrum was assumed.

At first the sample transit time to the neutron detector was about three seconds which was totally unsuitable for this work. By optimizing the flight path and increasing the gas pressure this time was reduced to about 1 second. To provide even shorter transit times a second neutron detector was constructed and placed in the pneumatic line at the reactor top. The transit time was thus reduced to less than 0.4 seconds. The same samples that had been counted in the main detector were later irradiated again for 90 seconds and counted in this counter.

Regretably the LPTR had no pulsing capability. In order to emphasize the shorter-lived delayed neutrons the samples were irradiated for as short a time as possible (about 4 seconds) and then counted by the neutron counter on the reactor top. Several data sets were thus collected with each sample.

The pneumatic system was a highly automated and computer controlled system which utilized several photosensors and a sound sensor to provide timing for the experiments. Figure 1 is a diagram of the main neutron detector. This detector was located eight feet below ground level and about 150 feet of flight-path from the reactor core. The shielding of this counter was sufficient to essentially eliminate any background. The counter itself consisted of 20 ^{3}He gas proportional counting tubes placed concentrically around the sample and embedded in polyethylene. Between the sample and the polyethylene was a 1.5 inch thick lead gamma shield. The gamma sensitivity of the counter was tested and found to be completely negligible. The 20 detector tubes were connected so that the dead-time of one counter did not affect the counts in the others. This gave a high efficiency (≈30%) counter with a short dead-time (≈3.1 μsec). It was capable of tolerating count rates up to 100,000 counts per second provided dead-time corrections were made. This was determined using a ^{235}U standard irradiated at low power and high power and comparing the observed count rates with time.

The second neutron counter consisted of a single BF_3 tube embedded in polyethylene and shielded from the sample by 0.5 inches of lead. The entire assembly was surrounded by two inches of borated polyethylene. Figure 2 is a drawing of this unit. A small background from reactor neutrons was measured when the unit was placed on the reactor room floor, but on the reactor top the background was

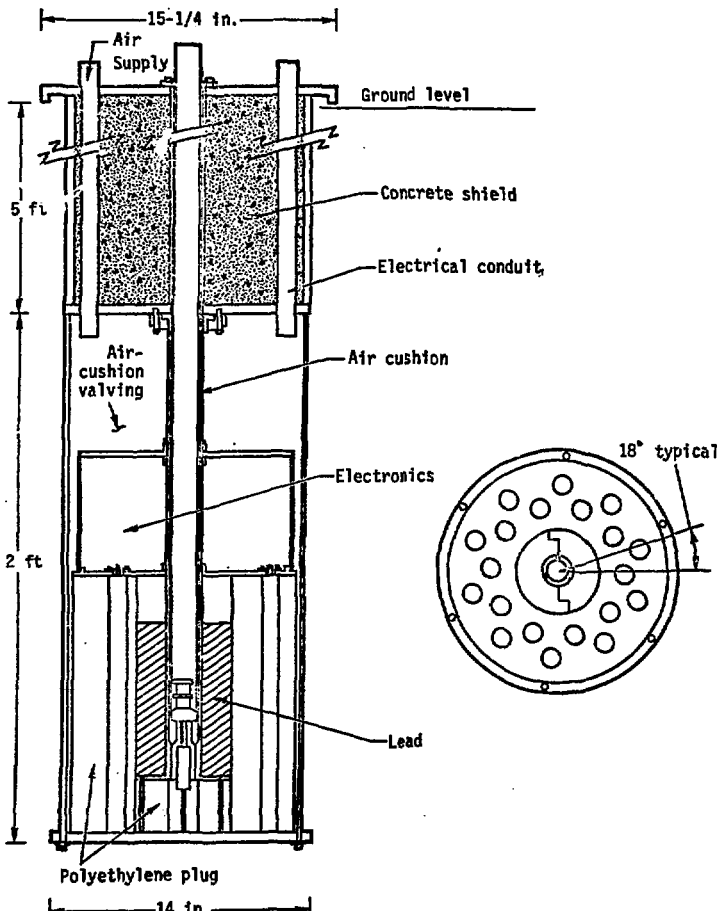

Figure 1. Diagram of the delayed-neutron detector.
(Taken From UCID 16911-76-3, Lawrence Livermore Lab.)

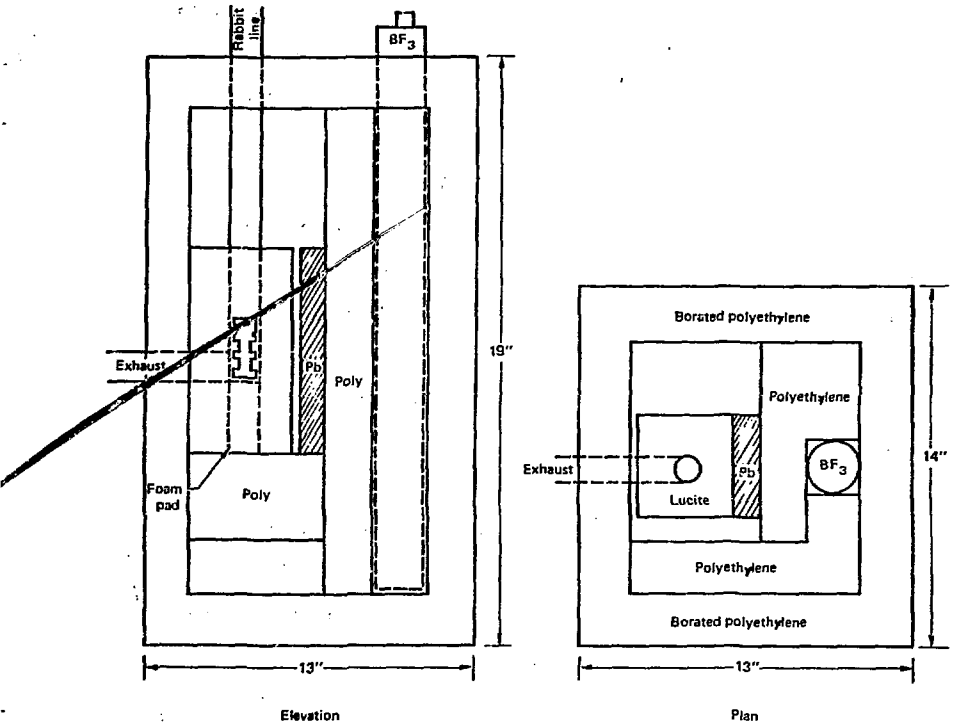

Figure 2. Diagram of the Neutron Detector Located on the Reactor Top.

entirely negiligible. Plots of delayed neutrons from previously measured nuclides showed essentially no variation from those found here. This indicates negligible energy dependence in either counter.

Count signals were recorded on a 1024 channel Tracor Northern TN-1705 multichannel analyzer. A channel size was selected from 0.04 to 0.8 seconds per channel depending upon the total counting time desired.

Gamma counting was done with one of the 4 mated coaxial GeLi detectors associated with the pneumatic system. Each counter was set in an identical movable holder to provide variable but reproducible counter geometry. Counts were begun about 30 minutes after irradiation and extended over 8 hours. Another count was taken 24 hours later to emphasize the longer-lived fission products. A similar count was taken on a ^{235}U standard so that the ratio of fissions in the sample to those of the ^{235}U standard was found. It was normally possible to use over a dozen different fission products to establish this ratio. The one exception was ^{237}Np. In this case the decay of ^{238}Np formed from neutron capture precluded gamma counting below 1 MeV. Fortunately several fission products have gamma energies above this energy and these were used to find the fission ratio. The 60 keV gamma peak associated with ^{241}Am caused counter dead-time problems. This was removed by shielding the gamma counters with 0.06 inches of cadmium which stopped the low energy gammas but allowed the higher energy gammas through.

Chain yields used in this work were generally taken from Rider and Meek[49]. For some nuclides for which the yields were not given by Rider and Meek the information was taken from Crouch[50]. For chain yields from ^{238}Pu, calculated yields listed by Sidebottom[75] were used. Chain yields for ^{232}U were taken from work by Monohan et al[76]. The fission products normally used to calculate the total fissions were: ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{103}Ru, ^{131}I, ^{132}I, ^{135}I, ^{139}Ba, ^{140}La, and ^{143}Ce. For each fission product the counts in the photopeak associated with that fission product were subtracted from the background counts on either side of the peak. The ratio of counts in the sample nuclide to the ^{235}U standard was divided by the fission yield ratio for that fission product. This was the fission ratio. The values obtained from all the fission products were averaged and were normally quite consistent.

Energy sensitivity of the delayed neutron counters was of serious concern since a change in counter efficiency with neutron energy would give a distorted count rate. This is because delayed neutrons are similar to gamma rays in that those with the highest energies also tend to have the shortest half-lives. Thus if a counter had a higher counting efficiency for low energy neutrons one would observe an enhancement of the 55 second and 22 second groups relative to the shortest-lived (most energetic) groups. Plots of data taken with the two delayed neutron detectors used in this work were compared with plots of Keepin's data (taken with

an energy insensitive detector). No variation was observed for any nuclide. Since there is a very large change in relative group yields from ^{232}Th and ^{238}U to ^{233}U and ^{239}Pu this was considered a sufficient check on the energy sensitivity of the counters.

C. Sample Impurities

Since fission cross-sections at thermal energy are normally much larger than those at high energy small impurities of fissile nuclides could bias the results of fast fission studies. As an example the presence of 0.1% ^{235}U in a ^{238}U sample would dominate the results of the delayed neutron study and even lesser amounts would bias the observed yield downward. This is because the total yield of ^{235}U delayed neutrons is about one-third the yield of ^{238}U.

Samples with huge thermal cross-sections such as ^{249}Cf, ^{245}Cm, and ^{242m}Am required little concern about impurity contamination. In other cases pure samples were obtained by chemical means. ^{232}Th, ^{237}Np, ^{241}Am, ^{239}Pu, and ^{233}U were such nuclides. Isotope separation was required in the other cases. ^{235}U (93% enriched) was readily available. Ultra pure ^{238}U (99.999% ^{238}U) was cadmium covered and was thus suitable for study. Ultra pure ^{242}Pu (0.032% fissile impurities) was studied with and without cadmium cover and no difference was observed so it was considered acceptable. Isotopically separated standards of ^{241}Pu, ^{232}U, and ^{238}Pu were also obtained.

Some nuclide samples were rejected. A ^{240}Pu sample with 1%

^{239}Pu impurity was rejected. A ^{244}Pu sample contained enough ^{241}Pu to dominate the results. A very pure sample of ^{243}Am still contained enough ^{241}Am to bias the results so it too was rejected. Finally a ^{251}Cf sample contained enough ^{252}Cf to cause an unacceptable spontaneous fission neutron background. This information is summarized in Table 2.

It is important to note that the results themselves provide a check on the sample purity. As is discussed later in this work, it is possible to predict with good accuracy the total delayed neutron yield a nuclide will have. This yield varies dramatically from nuclide to nuclide and so a sample, which gives a different absolute yield than expected, will immediately be suspect.

Such an example was the ^{242}Pu sample. The absolute yield observed was considerably lower than expected (probably due to the even-odd effect). This could have been due to impurities (^{239}Pu and ^{241}Pu) with lower absolute yields and large thermal cross-sections. However, covering the sample with cadmium would have decreased this effect and the change would have been noticed. Cadmium covering a sample of pure ^{242}Pu would have no effect since thermal neutrons do not cause fission in ^{242}Pu. In a nuclide with a large thermal fission cross-section (such as ^{239}Pu or ^{241}Pu) the ratio of fissions in a bare sample to a cadmium-covered sample is about 10. Since no change was observed in the delayed neutron yield of the ^{242}Pu sample the low absolute yield was assumed to be real.

Table 2. Difficult Samples

NUCLIDE OF INTEREST	IMPURITY	SOLUTION
^{238}U	^{235}U	Use of high purity ^{238}U covered with cadmium. (99.9991% ^{238}U)
^{240}Pu	^{239}Pu	Sample not used.
^{242}Pu	^{241}Pu	Use of high purity ^{242}Pu, checked with cadmium-covered sample. (99.90% ^{242}Pu)
^{244}Pu	^{241}Pu	Data dominated by ^{241}Pu fission. Data not used.
^{243}Am	^{241}Am	Data dominated by ^{241}Am fission. Data not used.
^{244}Cm	^{245}Cm	Sample not used.
^{251}Cf	^{252}Cf	Data dominated by spontaneous fission neutron background. Data not used.

D. Analysis of Data

Keepin established that delayed neutron data may be treated as a linear superposition of six exponentials. In this work, for a given sample, the optimal number of groups varied. The data were plotted and approximate yields and half-lives were found by "peeling off" the longest-lived components. These values were then used as initial guesses for a least squares fitting routine. This program produced one "best fit" for all the data from the various irradiation times and power levels for the number of groups used. The number of groups providing the smallest standard deviation was the best fit.

Least Squares Fitting

It can be assumed that the delayed neutron data, Y(t), obey the expression:

$$Y(t) = \sum_{i=1}^{N} pA_i(1-e^{-\lambda_i t_o})e^{-\lambda_i t}$$

where N is the number of groups assumed, A_i and λ_i are the yield and decay constants for group i, t_o is the length of irradiation, and p is a constant of proportionality depending upon the reactor power level, counting channel size, and detector efficiency.

With the initially guessed values of A_i' and λ_i' a function Z(t) was defined such that it was the difference between the observed data and the best guessed calculated data:

$$Z(t) = Y(t) - \sum_{i=1}^{N} pA_i'(1-e^{-\lambda_i t_o})e^{-\lambda_i t} \quad .$$

The ideal values, A_i and λ_i may be expressed in terms of A_i' and λ_i' by

$$A_i = A_i' + \Delta A_i' \qquad \text{and} \qquad \lambda_i = \lambda_i' + \Delta\lambda_i'.$$

If E^2 is the sum of the squared differences between the experimental points and the calculated points then it is also the sum of the squared differences between Z(t) and the contributions of $\Delta A_i'$ and $\Delta\lambda_i'$ so that

$$E^2 = \sum_{t=0}^{\infty} W(t)\ [Z(t) - \sum_{i=1}^{N} p(1-e^{-\lambda_i' t_o})e^{-\lambda_i' t}(\Delta A_i' - A_i'\Delta\lambda_i' t) + \sum_{i=1}^{N} pA_i' e^{-\lambda_i'(t+t_o)}\ \Delta\lambda_i' t_o]^2$$

where W(t) is the statistical weighting function which turns out to be the inverse of Y(t). If E^2 is minimized the best fit is obtained. This is done by differentiating with respect to $\Delta A_L'$ or $\Delta\lambda_L'$ so that

$$\frac{dE^2}{d\Delta A_L'} = 0 = -\sum_{t=0}^{\infty} W(t)\ [Z(t) - \sum_{i=1}^{N} p(1-e^{-\lambda_i' t_o})e^{-\lambda_i' t}(\Delta A_i' - A_i'\Delta\lambda_i' t) + \sum_{i=1}^{N} pA_i' e^{-\lambda_i'(t+t_o)}\Delta\lambda_i' t_o][p(1-e^{-\lambda_L' t_o})e^{-\lambda_L' t}]$$

and

$$\frac{dE^2}{d\Delta\lambda_L'} = 0 = -\sum_{t=0}^{\infty} W(t)\ [Z(t) - \sum_{i=1}^{N} p(1-e^{-\lambda_i' t_o})e^{-\lambda_i' t}(\Delta A_i' - A_i\ \Delta\lambda_i' t) + \sum_{i=1}^{N} pA_i\ e^{-\lambda_i'(t+t_o)}\Delta\lambda_i' t_o\ ]\ [\ p(1-e^{-\lambda_L' t_o})e^{-\lambda_L' t}(-A_L' t) \quad pA_L\ e^{-\lambda_L'(t+t_o)} t_o A_L'].$$

These equations were solved for $\Delta A_i'$ and $\Delta\lambda_i'$ by converting to matrix

notation and inverting the matrix of the coefficients of $\Delta A_i'$ and $\Delta \lambda_i'$.

If $H(L,I) = \sum_{t=0}^{\infty} W(t)(1-e^{-\lambda_L' t_o})e^{-\lambda_L' t}\, p(1-e^{-\lambda_I' t_o})e^{-\lambda_I' t}$ and

$D(L) = \sum_{t=0}^{\infty} W(t)(1-e^{-\lambda_L' t_o})e^{-\lambda_L' t}\, Z(t)$ then in matrix form:

$$\begin{pmatrix} \vdots \\ D(L) \\ \vdots \end{pmatrix} = \begin{pmatrix} \vdots \\ \cdots\ H(L,I)\ \cdots \\ \vdots \end{pmatrix} X \begin{pmatrix} \vdots \\ \Delta A_I' \\ \vdots \end{pmatrix} .$$

This assumes the two sets of equations have been separated and only one difference is considered in each case (ie $\Delta \lambda_I' = 0$ in this case and $\Delta A_I' = 0$ in the next case). The other set of equations in matrix form is

$$\begin{pmatrix} \vdots \\ F(L) \\ \vdots \end{pmatrix} = \begin{pmatrix} \vdots \\ \cdots\ G(L,I)\ \cdots \\ \vdots \end{pmatrix} X \begin{pmatrix} \vdots \\ \Delta \lambda_I' \\ \vdots \end{pmatrix} \quad \text{where}$$

$F(L) = \sum_{t=0}^{\infty} W(t)\, Z(t)\, (t - te^{-\lambda_L' t_o} - t_o e^{-\lambda_L' t_o})e^{-\lambda_L' t}$ and

$G(L,I) = \sum_{t=0}^{\infty} W(t) p(t - te^{-\lambda_L' t_o} - t_o e^{-\lambda_L' t_o})e^{-\lambda_L' t}(-A_I)(1-e^{-\lambda_I' t_o})e^{-\lambda_I' t}$.

By inverting the H(L,I) and G(L,I) matricies equations for $\Delta A_I'$ and $\Delta \lambda_I'$ were found.

$$\begin{pmatrix} \vdots \\ \Delta A_I' \\ \vdots \end{pmatrix} \quad \begin{pmatrix} \vdots \\ \cdots\ H(L,I)\ \cdots \\ \vdots \end{pmatrix}^{-1} \quad X \quad \begin{pmatrix} \vdots \\ D(L) \\ \vdots \end{pmatrix} \quad \text{and}$$

$$\begin{pmatrix} \vdots \\ \Delta\lambda_I' \\ \vdots \end{pmatrix} = \begin{pmatrix} & \vdots & \\ \cdots & G(L,I) & \cdots \\ & \vdots & \end{pmatrix}^{-1} \times \begin{pmatrix} \vdots \\ F(L) \\ \vdots \end{pmatrix} .$$

New estimates of A_I and λ_I were then made using the corrections found above. Finally the uncertainty in A_I and λ_I was found by taking the square-root of the Ith diagonal element of the appropriate inverse matrix. The inverse matrices are known as error matrices because of this property.[13,80]

The program KEEP computed the necessary summations over time for the data and the program MATINV calculated the inverse matrices and supplied the values of $\Delta A_I'$ and $\Delta\lambda_I'$. These corrections were then added to the old parameters and a new set of initial guesses was formed for KEEP. By this process any desired level of convergence was obtainable. In reality the process was continued until each iteration caused changes which were much smaller than the errors associated with the parameters.

A standard deviation was calculated with each fit. One expected a standard deviation of 1.0 simply due to variations in the data. Any deviation in excess of 1.0 would have indicated a poor fit to the data. Thus the standard deviation was used as a measure of the goodness of the fit. At some point increasing the number of groups did not statistically improve the standard deviation and thus the best number of groups was established. In every case this deviation was near unity, indicating a good fit.

As a check on the agreement between the results reported here and those reported previously, a program was written which calculated the mean square difference (MDS) and the mean square difference ratio (MSDR). from the following equations

$$MSD = \sqrt{\int_{0.4}^{800} [Y(t)_{other} - Y(t)_{waldo}]^2 \, dt} \quad \text{and}$$

$$MSDR = \frac{MSD}{\int_{0.4}^{800} Y(t)_{other} \, dt} \quad .$$

In this case $Y(t)_{other}$ and $Y(t)_{waldo}$ are the yields with time using the previously reported data and the data reported in this work. Table 3 shows a summary of the ratios obtained. It is evident that the agreement was excellent. In all cases the mean square difference ratio was less than 1%.

E. Transit Time Correction

Unfortunately, due to the relatively long transit time even with the second neutron detector (<0.4 second delay), a fraction of the shortest delayed neutrons were missed. By analyzing Keepin's delayed neutron yields it was clear that the fraction of the delayed neutron count observed at t=0.4 seconds to that at t=0.0 seconds was a function of the quantity $\frac{Ac}{Zc}$, where A_c and Z_c were the composite mass and charge of the fissioning material. Simply put, neutron rich nuclides gave more delayed neutrons with short half-lives. This was easily understood since the larger the ratio $\frac{Ac}{Zc}$ was the

Table 3. The Mean Square Difference Ratio (MSDR*) Between This Work and Other Reported Delayed Neutron Yields as a Function of Time.

NUCLIDE	MSDR*
^{232}Th	0.0020
^{233}U	0.0033
^{235}U	0.0022
^{238}U	0.0019
^{239}Pu	0.0074
^{241}Pu	0.0046

*The Mean Square Difference Ratio is a method of comparing two sets of delayed neutron yields as a function of time. It is a measure of the variation with time between the two data sets divided by the value of one of the sets over time. Since the data in this work was taken over the period from 0.4 seconds after irradiation to 800 seconds this is the interval chosen for comparison.

$$(MSD)^2 = \int_{0.4}^{800} [Y_{other}(t)-Y_{waldo}(t)]^2 \, dt \qquad \text{and}$$

$$MSDR = \frac{\sqrt{(MSD)^2}}{\int_{0.4}^{800} Y_{other}(t) \, dt}$$

where $Y_{other}(t)$ and $Y_{waldo}(t)$ are each calculated using the respective group parameters reported by other experimenters (Keepin and Cox) and reported here.

more neutron-rich the fission products were and therefore the shorter-lived. Specifically the fraction of delayed neutrons missing at t=0.4 seconds was fit to the equation:

$$F = 1 - \frac{Y(t=0)}{Y(t=0.4)} = 1.9207 \frac{Ac}{Zc} - 4.788 \quad \pm(2\%).$$

This correction was then applied to the observed delayed neutron yield at t=0.4 seconds to give an absolute yield at t=0 seconds. The correction was of the order of 10% and so the error associated with this correction was small (0.2%). Typically this did not change the result at all. In all cases except ^{232}Th and ^{238}U the effect was minimal. For nuclides where a short-lived group was observed (that is $\lambda > 0.7$ sec^{-1}) no correction was applied. For nuclides where a short-lived group was not observed it was assumed that a short-lived group with low yield was missed due to the long transit time. In these cases the correction was applied. The difference between the observed yield (extrapolated to t=0) and the calculated absolute yield using this correction was assigned to a short-lived group. In all cases this group was very small. Keepin suggested a value of 0.514±.013 seconds for the average half-life of his Group V neutrons (λ=1.35 sec^{-1}) and so this value was arbitrarily assigned to this group.

The value of adding this group is questionable. It does not contribute significantly to any numerical results. Its only purpose is to estimate the very short-lived components for comparison to calculated yields presented later.

^{232}Th and ^{238}U were treated somewhat differently. Because these two nuclides were extremely neutron rich, the correction needed was significant. However both of these nuclides were studied by Keepin so that accurate information was available about the decay from t=0 to t=0.4 seconds. In these two cases the shortest-lived group yield and decay constant were modified to accurately reflect the decay observed from t=0 to t=0.4 seconds.

F. Results

The following tables summarize the data collected in this work. Group decay constants are listed in the first column, group absolute yields in the second, and group relative yields (normalized to unity) in the third.

The total measured yield is compared with other reported or predicted yields in the "boxed" summary. Predictions were made using a correlation suggested by Tuttle[20]. Also included is a description of the purity of the sample and the standard deviation of the data from group parameters.

Finally, where available, the group parameters reported by other experimenters are listed. The values listed for ^{242}Pu were predictions based on calculated yields done by Bohn[77] and not measured values.

All measurements were relative to the ^{235}U delayed neutron yield. The value suggested by Tuttle for ^{235}U thermal fission is 1.654±.033 neutrons per 100 fissions. This value was assumed for this work and agrees well with the value suggested by Rider and

Meek[49] of 1.67±.07 neutrons per 100 fissions.

Table 4. ^{232}Th Decay Constants, Absolute Group Yields, Relative Yields, and Total Delayed Neutron Yield are Compared with Keepin's Values.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01251±.00002	A_1=0.1809±.0069	A_1/A_t=0.0360±.0014
λ_2=0.03241±.00012	A_2=0.704±.027	A_2/A_t=0.1402±.0054
λ_3=0.1327±.0025	A_3=1.33±.059	A_3/A_t=0.265±.012
λ_4=0.437±.020	A_4=2.02±.12	A_4/A_t=0.402±.024
λ_5=1.79±.64 *	A_5=0.79±.29 *	A_5/A_t=0.157±.058
	$A_t=\Sigma A_i$=5.02±.26	

* Group 5 was modified to fit Keepin's relative yields until t=0.4 seconds.

Measurement Summary

The corrected absolute yield is 5.02±.26 neutrons per 100 fissions. This compares with Tuttle's value of 5.47±.12 and Rider and Meek's value of 5.27±.40 neutrons per 100 fissions.

The standard deviation of the data is 1.037.

The sample was about 0.5 gram of ^{232}Th foil (>99.5% ^{232}Th.

Keepin's Values

Decay Constant (sec^{-1})	Absolute Yield (Neutron per 100 fissions	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0124±.0002	A_1=0.169±.012	A_1/A_t=0.034±.002
λ_2=0.0334±.0011	A_2=0.744±.037	A_2/A_t=0.150±.005
λ_3=0.121±.005	A_3=0.769±.108	A_3/A_t=0.155±.021
λ_4=0.321±.011	A_4=2.212±.110	A_4/A_t=0.446±.015
λ_5=1.21±.090	A_5=0.853±.073	A_5/A_t=0.172±.013
λ_6=3.29±.297	A_6=0.213±.031	A_6/A_t=0.043±.006
	$A_t=\Sigma A_i$=4.69±.20	

Table 5. ^{232}U Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01276±.00004	A_1=0.0524±.0040	A_1/A_t=0.120±.009
λ_2=0.03502±.00029	A_2=0.131±.010	A_2/A_t=0.300±.023
λ_3=0.1439±.0059	A_3=0.134±.014	A_3/A_t=0.334±.032
λ_4=0.396±.045	A_4=0.113±.012	A_4/A_t=0.256±.027
λ_5=1.35 *	A_5=0.007±.039 *	A_5/A_t=0.016±.089 *
	$A_t=\Sigma A_i$=0.437±.033	

* λ_5=1.35 is assumed and A_5 is calculated from the corrected total yield.

Measurement Summary

The corrected absolute delayed neutron yield is 0.437 ±.033 neutrons per 100 fissions. The predicted yield using Tuttle's correlation is 0.493±.054 neutrons per 100 fissions.

The standard deviation of the data is 1.004.

The sample was several micrograms of ^{232}U whose assay was 99.99% ^{232}U.

Table 6. ^{233}U Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Keepin's Values.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01239±.00004	A_1=0.0551±.0037	A_1/A_t=0.0751±.0051
λ_2=0.0259±.0019	A_2=0.070±.027	A_2/A_t=0.095±.037
λ_3=0.0398±.0024	A_3=0.160±.024	A_3/A_t=0.218±.033
λ_4=0.161±.010	A_4=0.175±.024	A_4/A_t=0.238±.033
λ_5=0.287±.028	A_5=0.188±.030	A_5/A_t=0.256±.041
λ_6=1.32±.40	A_6=0.084±.013	A_6/A_t=0.115±.018
	$A_t=\Sigma A_i$=0.733±.047	

Measurement Summary

The measured absolute yield is 0.733±.047 neutrons per 100 fissions. This compares with Tuttle's value of 0.698 .013 and Rider and Meek's of 0.74±.04 neutrons per 100 fissions.

The standard deviation of the data is 1.022.

The sample was several micrograms with the following assay: 4 ppm ^{232}U, 95.1% ^{233}U, 0.5% ^{234}U, 0.8% ^{235}U, 0.1% ^{236}U, and 3.5% ^{238}U.

Keepin's Values

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0126±.0003	A_1=0.057±.003	A_1/A_t=0.086±.003
λ_2=0.0337±.0006	A_2=0.197±.009	A_2/A_t=0.299±.004
λ_3=0.139±.006	A_3=0.166±.027	A_3/A_t=0.252±.040
λ_4=0.325±.030	A_4=0.184±.016	A_4/A_t=0.278±.020
λ_5=1.13±.40	A_5=0.034±.016	A_5/A_t=0.051±.024
λ_6=2.50±.42	A_6=0.022±.009	A_6/A_t=0.034±.014
	$A_t=\Sigma A_i$=0.66±.03	

Table 7. ^{235}U Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Keepin's Values.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01255±.00003	A_1=0.0566±.0011	A_1/A_t=0.0342±.0007
λ_2=0.0309±.0001	A_2=0.356±.007	A_2/A_t=0.2175±.0043
λ_3=0.114±.0001	A_3=0.346±.011	A_3/A_t=0.2089±.0065
λ_4=0.328±.007	A_4=0.672±.018	A_4/A_t=0.406±.011
λ_5=2.06±.31	A_5=0.303±.045	A_5/A_t=0.183±.027
	$A_t=\Sigma A_i$=1.654±.033	

Measurement Summary

The absolute yield for ^{235}U is assumed to be 1.654±.033 as listed by Tuttle. Rider and Meek list a value of 1.67±.07 neutrons per 100 fissions.

The standard deviation of the data is 1.062.

The sample was several micrograms of 93.7% enriched ^{235}U comprising 0.2% in platinum wire.

Keepin's Values

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0127±.003	A_1=0.052±.005	A_1/A_t=0.038±.004
λ_2=0.0317±.0012	A_2=0.346±.018	A_2/A_t=0.213±.007
λ_3=0.115±.004	A_3=0.310±.036	A_3/A_t=0.188±.024
λ_4=0.311±.012	A_4=0.624±.026	A_4/A_t=0.407±.010
λ_5=1.40±.12	A_5=0.182±.015	A_5/A_t=0.128±.012
λ_6=3.87±.55	A_6=0.066±.008	A_6/A_t=0.026±.004
	$A_t=\Sigma A_i$=1.58±.05	

Table 8. ^{238}U Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Keepin's Values.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01254±.00003	A_1=0.0487±.0040	A_1/A_t=0.0105±.0013
λ_2=0.03032±.00010	A_2=0.557±.042	A_2/A_t=0.1198±.0091
λ_3=0.08691±.0031	A_3=0.358±.035	A_3/A_t=0.0770±.0073
λ_4=0.2453±.0035	A_4=1.656±.140	A_4/A_t=0.356±.029
λ_5=0.705±.051	A_5=1.212±.124	A_5/At=0.261±.027
λ_6=2.5±1.1 *	A_6=0.82±.50 *	A_6/A_t=0.18±.11 *
	$A_t=\Sigma A_i$=4.65±.35	

* Group 6 was modified to fit Keepin's data to 0.4 seconds after irradiation.

Measurement Summary

Corrected absolute yield is 4.65±.35 neutrons per 100 fissions which compares with 4.60±.25 listed by Rider and Meek and 4.51±.61 by Tuttle. The analysis of all reported yields gave a yield of 4.44±.23 neutrons/100 fission.

The standard devaition of the data is 1.025.

The sample was 0.1 gram of ultra pure ^{238}U with the assay: 1 ppm ^{233}U, 1 ppm ^{234}U, 6 ppm ^{235}U, 1 ppm ^{236}U and the rest ^{238}U.

Keepin's Values (Absolute Yield Normalized to 4.44 neutrons per 100 fissions)

Decay Constants (sec^{-1})	Absolute Yield (% Neutrons per fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0132±.0003	A_1=0.0577±.004	A_1/A_t=0.013±.001
λ_2=0.0321±.006	A_2=0.608±.009	A_2/A_t=0.137±.002
λ_3=0.139±.005	A_3=0.719±.089	A_3/A_t=0.162±.020
λ_4=0.358±.014	A_4=1.72±.05	A_4/A_t=0.388±.012
λ_5=1.41±.07	A_5=1.00±.06	A_5/A_t=0.225±.013
λ_6=4.02±.21	A_6=0.33±.02	A_6/A_t=0.075±.005
	$A_t=\Sigma A_i$=4.44±.23	

Table 9. ^{237}Np Decay Constants, Absolute Group Yield, Relative yield, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group yield/ Total Yield)
λ_1=0.01258±.00004	A_1=0.0368±.0034	A_1/A_t=0.0345±.032
λ_2=0.0306±.00034	A_2=0.244±.024	A_2/A_t=0.229±.023
λ_3=0.0653±.016	A_3=0.070±.033	A_3/A_t=0.066±.031
λ_4=0.139±.019	A_4=0.153±.065	A_4/A_t=0.143±.061
λ_5=0.328±.030	A_5=0.424±.053	A_5/A_t=0.397±.050
λ_6=1.62±.69	A_6=0.132±.031	A_6/A_t=0.124±.029
	$A_t=\Sigma A_i$=1.068±.098	

Measurement Summary

The measured absolute yield is 1.060±.098 neutrons per 100 fissions. This compares to a predicted yield of 1.02±.12 using Tuttle's correlation.

The standard deviation of the data is 1.033.

The sample consisted of several milligrams of ^{237}Np with the following assay: 0.7% thorium, 0.1% uranium, 0.01% plutonium, with the rest neptunium.

Table 10. ^{238}Pu Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01262±.00013	A_1=0.0197±.0031	A_1/A_t=0.0426±.0067
λ_2=0.03026±.00035	A_2=0.142±.022	A_2/A_t=0.314±.047
λ_3=0.0851±.012	A_3=0.0528±.031	A_3/A_t=0.114±.067
λ_4=0.197±.023	A_4=0.0815±.013	A_4/A_t=0.176±.028
λ_5=0.356±.051	A_5=0.151±.024	A_5/A_t=0.327±.052
λ_6=1.35 *	A_6=0.015±.087 *	A_6/A_t=0.033±.19 *
	$A_t=\Sigma A_i$=0.461±.073	

* λ_6=1.35 is assumed and A_6 is calculated from the corrected total yield.

Measurement Summary

The corrected absolute yield is 0.461±.073 neutrons per 100 fissions which compares with the predicted value of 0.455 ±.051 neutrons per 100 fissions using Tuttle's correlation.

The standard deviation of the data is 1.033.

The sample was several micrograms of isotopically pure ^{238}Pu. The assay was as follows: <0.1% ^{238}U, 0.1% ^{239}Pu, and the rest ^{238}Pu.

Table 11. ^{239}Pu Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Keepin's Values.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01246±.00001	A_1=0.01895±.0009	A_1/A_t=0.0269±.0013
λ_2=0.02941±.0008	A_2=0.1825±.0089	A_2/A_t=0.260±.013
λ_3=0.0714±.0036	A_3=0.0780±.0087	A_3/A_t=0.111±.013
λ_4=0.212±.018	A_4=0.158±.031	A_4/A_t=0.225±.044
λ_5=0.324±.048	A_5=0.147±.031	A_5/A_t=0.209±.046
λ_6=1.28±.25	A_6=0.119±.015	A_6/A_t=0.170±.021
	$A_t=\Sigma A_i$=0.703±.049	

Measurement Summary

The measured absolute yield is 0.703±.049 neutrons per 100 fissions which compares with 0.645±.05 listed by Rider and Meek and 0.655±.012 neutrons per 100 fissions listed by Tuttle.

The standard deviation of the data is 1048.

The sample consisted of about a milligram of ^{239}Pu with the following assay: 0.01% ^{238}Pu, 93.6% ^{239}Pu, 5.7% ^{240}Pu, 0.65% ^{241}Pu.

Keepin's Values

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0128±.0005	A_1=0.021±.006	A_1/A_t=0.035±.009
λ_2=0.0301±.0022	A_2=0.182±.023	A_2/A_t=0.298±.035
λ_3=0.124±.009	A_3=0.129±.030	A_3/A_t=0.211±.048
λ_4=0.325±.036	A_4=0.199±.022	A_4/A_t=0.326±.033
λ_5=1.12±.39	A_5=0.052±.018	A_5/A_t=0.086±.029
λ_6=2.69±.48	A_6=0.027±.010	A_6/A_t=0.044±.016
	$A_t=\Sigma A_i$=0.61±.03	

Table 12. ^{241}Pu Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Cox's Values.

Decay Constants (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01296±.00010	A_1=0.0195±.0012	A_1/A_t=0.0125±.008
λ_2=0.0296±.0002	A_2=0.324±.017	A_2/A_t=0.208±.011
λ_3=0.0663±.0079	A_3=0.0860±.018	A_3/A_t=0.055±.008
λ_4=0.196±.009	A_4=0.473±.036	A_4/A_t=0.304±.022
λ_5=0.694±.047	A_5=0.598±.035	A_5/A_t=0.384±.022
λ_6=1.35 *	A_6=0.058±.089 *	A_6/A_t=0.037±.056 *
	$A_t=\Sigma A_i$=1.56±.12	

* λ_6=1.35 is assumed A_6 is calculated from the corrected total yield.

Measurement Summary

The corrected absolute yield is 1.56±.12 neutrons per 100 fissions which compares with Cox's value of 1.57±.15. The standard deviation of the data is 1.040. The sample was isotopically separated ^{241}Pu with <0.1% ^{240}Pu and 0.1% ^{242}Pu.

Cox's Values[73]

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0128±.0002	A_1=0.0156±.0047	A_1/A_t=0.010±.003
λ_2=0.0299±.0006	A_2=0.357±.009	A_2/A_t=0.229±.006
λ_3=0.124±.013	A_3=0.279±.039	A_3/A_t=0.173±.025
λ_4=0.352±.018	A_4=0.608±.078	A_4/A_t=0.390±.050
λ_5=1.61±.15	A_5=0.284±.030	A_5/A_t=0.182±.019
λ_6=3.47±1.7	A_6=0.025±.008	A_6/A_t=0.016±.005
	$A_t=\Sigma A_i$=1.57±.15	

Table 13. ^{242}Pu Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield are Compared with Bohn's Calculations.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.0134±.00027	A_1=0.0221±.0027	A_1/A_t=0.0119±.0015
λ_2=0.0295±.0015	A_2=0.316±.104	A_2/A_t=0.170±.057
λ_3=0.0409±.014	A_3=0.0616±.097	A_3/A_t=0.033±.054
λ_4=0.127±.0056	A_4=0.322±.030	A_4/A_t=0.173±.017
λ_5=.397±.033	A_5=0.721±.071	A_5/A_t=0.388±.039
λ_6=2.22±.87	A_6=0.523±.169	A_6/A_t=0.281±.093
	$A_t=\Sigma A_i$=1.97±.23	

Measurement Summary

The Measured absolute yield is 1.97±.23 neutrons per 100 fissions which compares with Evans' value of 1.5±.5 neutrons per 100 fissions and a predicted value using Tuttle's correlation of 2.17±.25 neutrons per 100 fissions.

The standard deviation of the data is 1.097.

The sample was several milligrams of isotopically pure ^{242}Pu with the following assay: 0.012% ^{238}Pu, 0.009% ^{239}Pu, 0.011% ^{241}Pu, 99.90% ^{242}Pu and 0.008% ^{244}Pu.

Calculated Group Parameters Using Bohn's [77] Calculations

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield Total Yield)
λ_1=0.0128±.0003	A_1=0.0060±.0025	A_1/A_t=0.004±.001
λ_2=0.0314±.0013	A_2=0.293±.123	A_2/A_t=0.195±.32
λ_3=0.128±.009	A_3=0.242±.102	A_3/A_t=0.161±.048
λ_4=0.325±.020	A_4=0.202±.085	A_4/A_t=0.412±.153
λ_5=1.35±.09	A_5=0.327±.137	A_5/A_t=0.218±.087
λ_6=3.70±.44	A_6=0.015±.006	A_6/A_t=0.010±.003
	$A_t=\Sigma A_i$=1.5±.5	

Table 14. ^{241}Am Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yields (Neutrons per 100 fissions)	Relative Yields (Group Yield/ Total Yield)
λ_1=0.01271±.00003	A_1=0.0185±.0022	A_1/A_t=0.0369±.0044
λ_2=0.02985±.00004	A_2=0.146±.018	A_2/A_t=0.291±.036
λ_3=0.1519±.003	A_3=0.154±.019	A_3/A_t=0.307±.038
λ_4=0.446±.022	A_4=0.154±.020	A_4/A_t=0.307±.040
λ_5=2.63±2.11	A_5=0.036±.048	A_5/A_t=0.072±.097
	$A_t=\Sigma A_i$=0.509±.060	

Measurement Summary

The measured absolute yield is 0.509±.060 neutrons per 100 fissions which compares with a predicted value of 0.439 ±.048 neutrons per 100 fissions using Tuttle's correlation.

The standard deviation of the data is 1.117.

The sample was several milligrams of ^{241}Am prepared from decay of weapons grade plutonium. The assay was as follows: 1.8% ^{237}Np, <0.1% all other fissionable impurities, and the rest ^{241}Am.

Table 15. ^{242m}Am Decay Constants, Absolute Group Yields, Relative Yield, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01273±.00005	A_1=0.0176±.0012	A_1/A_t=0.0256±.0017
λ_2=0.03002±.00011	A_2=0.195±.013	A_2/A_t=0.284±.018
λ_3=0.0930±.0054	A_3=0.0822±.0092	A_3/A_t=0.120±.013
λ_4=0.2462±.0067	A_4=0.244±.026	A_4/A_t=0.355±.037
λ_5=0.656±.083	A_5=0.119±.013	A_5/A_t=0.173±.018
λ_6=1.35 *	A_6=0.030±.045 *	A_6/A_t=0.044±.065 *
	$A_t=\Sigma A_i$=0.688±.045	

* λ_6 =1.35 is assumed and A_6 is calculated from the corrected total yield.

Measurement Summary

The corrected absolute yield is 0.688±.045 neutrons per 100 fissions which compares with Tuttle's correlation prediction of 0.65±.07 neutrons per 100 fissions.

The standard deviation of the data is 1.070.

The sample consisted of 1 μgm ^{242m}Am. The assay was as follows: 0.79% ^{241}Am, 99.21% ^{242m}Am, <0.007% ^{243}Am, and no other elements.

Table 16. ^{245}Cm Decay Constants, Absolute Group Yield, Relative Yield, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.01335±.00009	A_1=0.01397±.0009	A_1/A_t=0.02360±.0017
λ_2=0.03031±.00014	A_2=0.1793±.012	A_2/A_t=0.303±.020
λ_3=0.104±.014	A_3=0.054±.017	A_3/A_t=0.0912±.028
λ_4=0.211±.011	A_4=0.174±.031	A_4/A_t=0.294±.050
λ_5=0.537±.073	A_5=0.136±.016	A_5/A_t=0.230±.022
λ_6=1.35 *	A_6=0.035±.056 *	A_6/A_t=0.059±.093 *
	$A_t=\Sigma A_i$=0.592±.039	

* λ_6=1.35 is assumed and A_6 is calculated from the corrected total yield.

Measurement Summary

The corrected absoulte yield is 0.592±.039 neutrons per 100 fissions which compares with the predicted value of 0.62 ±.07 neutrons per 100 fissions using Tuttle's correlation.

The standard deviation of the data is 1.036.

The sample contained 0.59 μ gm of ^{245}Cm with the following assay: 0.218% ^{244}Cm, 0.215% ^{246}Cm, 0.013% ^{247}Cm, 0.231% ^{248}Cm, and the rest ^{245}Cm. The only other elements observed were curium daughters.

Table 17. ^{249}Cf Decay Constants, Absolute Group Yields, Relative Yields, and Total Delayed Neutron Yield.

Decay Constant (sec^{-1})	Absolute Yield (Neutrons per 100 fissions)	Relative Yield (Group Yield/ Total Yield)
λ_1=0.012851±.000021	A_1=0.00765±.00056	A_1/A_t=0.0284±.0021
λ_2=0.03037±.000039	A_2=0.09435±.0069	A_2/A_t=0.3507±.026
λ_3=0.1678±.0037	A_3=0.102±.0086	A_3/A_t=0.379±.032
λ_4=0.541±.063	A_4=0.0628±.0069	A_4/A_t=0.233±.026
	$A_t=\Sigma A_i$=0.267±.019	

Measurement Summary

The measured total yield is 0.267±.019 neutrons per 100 fissions which compares with the predicted value of 0.27±.03 neutrons per 100 fissions using Tuttle's correlation.

The standard deviation of the data is 1.15.

The sample consisted of several micrograms of ^{249}Cf obtained from the decay of ^{249}Bk. It contained <0.1% fissile impurities.

CHAPTER 4

RESULTS AND DISCUSSION

Comparison of the present results with those of Keepin and Cox indicates extremely good agreement. For the long lived groups the uncertainties quoted in this work are much smaller. This is due to better statistics available from the high fluxes used. The count rate in this work was several hundred times that obtained by Keepin from his multiple irradiations. For the shorter lived groups this work had larger uncertainties due to the non-pulsing nature of the experiments and the significant transit times.

In all cases the longest-lived group (^{87}Br) agrees with the values quoted elsewhere. For group II the agreement is also excellent except that in the cases of ^{233}U and ^{242}Pu this work appears to have separated the ^{137}I and the ^{88}Br contributions into two groups. Normally it is not possible to distinguish between ^{137}I ($T_{\frac{1}{2}}$=24.5 sec) and ^{88}Br ($T_{\frac{1}{2}}$=16 sec) and both are lumped in Group II. In ^{233}U and ^{242}Pu the statistics associated with this group were sufficient to make this distinction.

In shorter-lived groups some difference begins to appear. One reason for this is that in some cases it was found that the best fit was obtained with five groups instead of six. Naturally in this case the group parameters would tend to merge together.

Secondly, because of the poor statistics for the shortest lived group in this work variations from other reported values are not surprising. In reality it is not important that the group parameters reported here exactly correspond with those reported elsewhere. What is important is that when all the groups are taken together they give the same decay pattern as observed in other work. This is the case.

A. ^{87}Br and ^{137}I Fission Yield Analysis Using Group I and II Yields

It is worth noting that the values quoted by Keepin varied by more than the reported uncertainty for the half-life of Group I. Since it is known that Group I represents only ^{87}Br this must mean that the much larger Group II yield must be affecting the fit. The same effect was observed in this work. Although the decay constants reported here are more accurate than previously reported values there is still a variation in values. The values do cluster around $\lambda_1 = 0.01255 \pm .00003$ sec^{-1} in many cases. In cases where the yield of Group II is extremely large deviation from this value was observed.

The half-life obtained for ^{87}Br is $55.23 \pm .13$ seconds which agrees reasonably well with the mean measured value[78] of $55.6 \pm .2$ seconds. The best P_n value for ^{87}Br is $2.38 \pm .08\%$.[46] Thus by dividing the observed ^{87}Br (Group I) yield by 2.38% the cumulative fission yield of ^{87}Br is obtained. The results of this calculation are listed in Table 18. Also are included are experimentally measured

Table 18. Derived Cumulative Fission Yield for ^{87}Br.

FISSIONING NUCLIDE	^{87}Br YIELD %	DERIVED ^{87}Br FISSION YIELD %	RECOMMENDED[49] YIELD %
^{232}Th	0.180±.007	7.56±.39	<7.15±.20
^{232}U	0.052±.004	2.18±.18	
^{233}U	0.055±.004	2.31±.19	2.20±.13
^{235}U	0.056±.001	2.35±.09	2.27±.14
^{238}U	0.048±.004	2.02±.18	1.36±.44, <1.44±.04
^{237}Np	0.036±.003	1.51±.14	1.73±.07
^{238}Pu	0.019±.003	0.80±.13	
^{239}Pu	0.0190±.0009	0.80±.05	0.73±.04
^{240}Pu	0.022±.003	0.92±.13	<1.01±.16
^{241}Pu	0.018±.001	0.76±.05	0.61±.05, <0.80±.06
^{242}Pu	0.019±.003	.080±.13	<0.86±.14
^{241}Am	0.018±.002	0.76±.09	
^{242m}Am	0.017±.001	0.71±.05	
^{245}Cm	0.0122±.0009	0.51±.04	
^{249}Cf	0.0072±.0006	0.30±.03	

cumulative yield values listed in Rider and Meek.[49] In some cases the cumulative fission yield of ^{87}Br was not known or uncertain so in these cases the cumulative fission yield of the beta decay daughter ^{87}Kr was included with a "<" because the yield of ^{87}Kr is definitely greater than that of ^{87}Br.

^{87}Br is obtained from the observed group I yield by correcting for the difference between λ_1 and the decay constant for ^{87}Br of 0.01247 sec $^{-1}$. The time at which Group I and Group II yields are the same is taken as the reference time. At this time the neutrons being observed are those of ^{87}Br and by extrapolating back using the decay constant for ^{87}Br instead of that for Group I we find a slightly different yield for ^{87}Br than we had for Group I. The equations used are:

$$Y_{Br}e^{-\lambda_{Br}t_o} = A_1e^{-\lambda_1 t_o} \text{ so that } Y_{Br} = A_1e^{-(\lambda_1-\lambda_{Br})t_o}.$$

It is noted that the agreement in cumulative yields is excellent except for ^{238}U. In view of the accuracy of this method and the general agreement obtained for other nuclides we conclude the cumulative yield value for ^{87}Br and ^{87}Kr reported by Rider and Meek is in error for ^{238}U fast fission.

A similar, but more complicated analysis can be made for the yield of ^{137}I from Group II data. In general in Group II the major contributor is ^{137}I with smaller contributions from ^{88}Br and

^{137}Te. The contribution of ^{88}Br decreases with increasing fissioning nuclide mass so that in most instances well over 80% of the contributions come from ^{137}I. The contribution of ^{88}Br and ^{136}Te are estimated using the fission yield model described later and this is subtracted from the observed Group II ($T_{\frac{1}{2}} \simeq 23$ seconds) yield. The fission yield of ^{137}I is then calculated by dividing by the P_n value of ^{137}I of 6.6±.6%.[46] The results of these calculations are shown in Table 19.

Rider and Meek list a few experimentally measured cumulative fission yields which agree well with the values obtained by this analysis. They also list recommended fission yields using calculations where measurements are not available. These values also agree with the values obtained by analyzing delayed neutrons. For several nuclides, however, no report of fission yields has been made and this analysis provides new information.

For other delayed neutron groups it is not practical to try to separate out individual precursors. It is, however, of interest to compare group yields with sums of individual precursors which is done later in this work.

B. Empirical Model for Total Delayed Neutron Yield

It was found some time ago that delayed neutron yields exhibit a relationship with the quantity (A_c-3Z_c) where A_c and Z_c are the composite mass and charge of the fissioning material. The

Table 19. Derived Cumulative Yield for ^{137}I.

FISSIONING NUCLIDE	GROUP II YIELD %	^{88}Br & ^{136}Te CONTRIBUTION	DERIVED ^{137}I YIELD %	OBSERVED[49] ^{137}I YIELD %	VALUE SUGGESTED BY Rider & Meek[49]
^{232}Th	0.704±.027	0.433	4.11±1.4	5.15±.82	5.3[illegible].59
^{232}U	0.131±.010	0.100	0.48±.34		
^{233}U	0.230±.036	.140	1.36±.71	1.67±.10	1.6[illegible].07
^{235}U	0.358±.007	.15[illegible]	3.03±.58	3.46±.21	3.22±.19
^{238}U	0.557±.042	.162	5.99±1.0		5.31±.85
^{237}Np	0.244±.024	.087	2.37±.51		2.90±.67
^{238}Pu	0.142±.022	.040	1.54±.39		
^{239}Pu	0.183±.009	.048	2.03±.28	2.57±.21	2.43±.14
^{240}Pu	0.238±.016	.059	2.71±.41		2.58±.59
^{241}Pu	0.324±.017	.065	3.92±.51	3.86±.23	4.13±.25
^{242}Pu	0.316±.104	.086	3.48±1.6		3.70±.85
^{241}Am	0.146±.018	.024	1.85±.34		
^{242m}Am	0.195±.013	.039	2.36±.33		
^{245}Cm	0.179±.012	.032	2.23±.30		
^{249}Cf	0.094±.007	.012	1.25±.17		
^{252}Cf	.0347±.0009	.020	3.04±.34		2.29±.73

reason for such a dependence has never been explained. At first glance one would expect to see a dependence on the mass to charge ratio of the parent nuclide ($\frac{A_c}{Z_c}$) which is normally about 2.57. This is because nuclides with the same mass to charge ratio should fission into products that also have the same mass to charge ratio. The mass to charge ratio of the fission products determines the amount of delayed neutron emission since the larger the ratio the more neutron rich the fission products and the more likely they are to decay by neutron emission.

An additional effect is also observed. Since the heavy fission peak is more or less constant an increase in A_c causes the light fission yield peak to shift. Delayed neutron precursors are concentrated in two groups near the light and heavy fission yield peaks (A≈90 and A≈140). Increasing the mass of the fissioning material, A_c, causes the light fission yield peak to shift away from the light delayed neutron precursors. The result is a decrease in the delayed neutron yield. To compensate for this loss one must increase the mass to charge ratio by more than 2.57. It is not surprising then that leaving the quantity (A_c-3Z_c) constant leaves the delayed neutron yield constant. Increasing the quantity increases the delayed neutron yield exponentially.

One can also least-squares fit the observed delayed

neutron data to an exponential of the form:

$$Y_{DN}(\text{per 100 fissions}) = \exp(a+bZ_c+cA_c).$$

If this is done one finds that the neutron induced fission data fits very well, whereas some of the photofission and spontaneous fission data fit roughly but with greater dispersion. It may well be that the quoted uncertainties on these data are too small. It may also be that, because these are somewhat different processes, the yield may be affected.

The least-squares fit of the available data (excluding ^{237}Np photofission, ^{234}U photofission, and ^{252}Cf spontaneous fission) fits the equation:

$$Y_{DN}(\text{per 100 fissions}) = \exp(16.698-1.144Z_c+0.377A_c) \quad (\pm 9\%).$$

Figure 3 is a plot of the measured delayed neutron yields that have been reported in this work and elsewhere against the function above.

Tuttle previously used many of the same points to find a fit of the form:

$$Y_{DN} = \exp[14.638+.1832(A_c-3Z_c)\frac{Ac}{Zc}] \quad (\pm 11.3\%).$$

This correlation was used to predict total delayed neutron yields for the nuclides studied in this work.

Such correlations are quite useful in estimating delayed neutron yields for unmeasured nuclides. For example the contri-

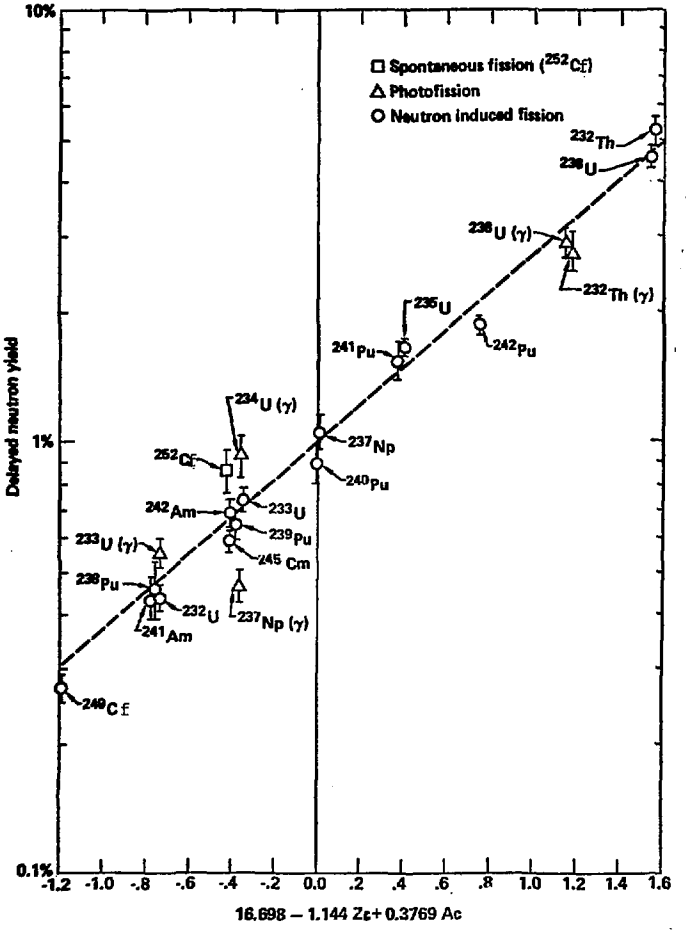

Figure 3. Plot of the Total Delayed Neutron Yield for Various Nuclides Versus the Quantity (16.698-1.144 Z_c+0.3769 A_c) where Z_c and A_c are the Composite Charge and Mass of the Fissioning Nuclide.

bution of ^{238}Pu fission or ^{236}U fission in reactors with these minor contributors can be estimated using such a correlation. If actinide burning reactors are ever designed any other delayed neutron yields will have to be estimated in this way.

C. Relative Time Dependent Yields

As stated earlier, neutron-rich nuclides (such as ^{232}Th and ^{238}U) not only have more delayed neutrons than neutron poor nuclides (such as ^{233}U and ^{239}Pu), but more of these delayed neutrons have a short half-life as well. Because the average fission product from a neutron rich nuclide is farther from stability than the average fission product from a neutron poor nuclide its half-life is naturally less. Figure 4 is a plot of the relative delayed neutron yield (normalized to unity) with time for all available data. In this plot it is obvious that the delayed neutrons from ^{238}U die off much more quickly than do those from ^{232}U. One can calculate the uranium equivalent mass for all the nuclides studied. The uranium equivalent mass of a nuclide of mass A and charge Z is simply $92*\frac{A}{Z}$. If this quantity is calculated for each nuclide one notes an orderly progression from ^{232}U to ^{238}U including the non-uranium nuclides. Thus it is possible not only to estimate the total delayed neutron yield for a given nuclide but the time dependent nature of the delayed

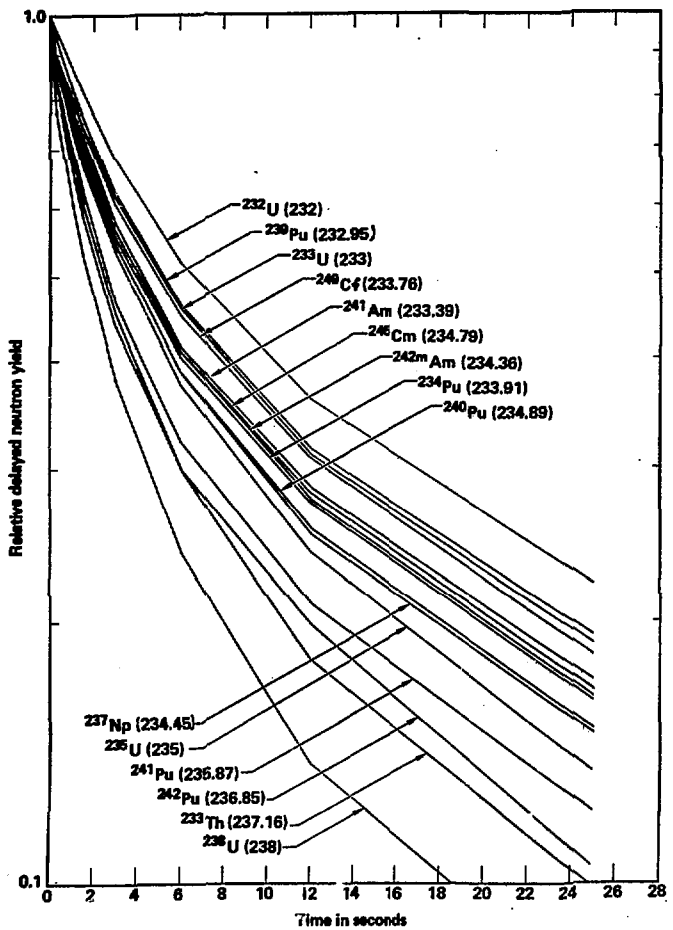

Figure 4. Plots of the Relative Delayed Neutron Yield For Various Nuclides With Time. The numbers in parentheses are Uranium equivalent; they are obtained by multiplying A_c/Z_c by 92.

neutrons as well. Reactors utilizing recycled fuel or burning actinides are likely to have inventories of fissioning nuclides which have not been studied so far. The delayed neutrons from these nuclides could have a perturbing effect on the stability and control of such reactors.

CHAPTER V

GENERALIZED FISSION YIELD MODEL

A. Model Development

As stated previously, it is generally accepted[67] that independent fission yields can be fit to a Gaussian distribution. Specifically if the relative independent yield is calculated (by dividing by the chain yield) and is plotted versus fission product charge for a given mass, the resulting Gaussian has a width parameter of σ_z=0.56. Recent studies indicate that σ_z = 0.53 may be better but at the moment the evidence is inconclusive. The center of this curve is located at Z_p. For a fission product of mass A and charge Z the relative independent yield is given by the expression:

$$RI_y = c(1+a) \int_{Z-.5}^{Z+.5} \exp[-\frac{(z-Z_p)^2}{2\sigma_z^2}] \, dz$$

where c is a normalizing constant (so that the total of all relative independent yields in a chain is unity) and "a" is the even-odd effect. If the variable X is defined by the equation:

$$X = (\frac{Z-Z_p}{\sigma_z})$$

then

$$RI_y = (1+a) \, F(x)$$

where F(x) is the integral function above and the tabulation used

in this work was listed in Bevington[80]. Independent yields may be calculated by multiplying the relative value by the chain yield and the cumulative yield may be obtained by summing from z=0 to z=Z, where Z is the charge of the nuclide of interest.

In the case of the even-odd effect, if the charge of the fission product is even,"a" is positive,and if it is odd,"a" is negative. For thermal fission of ^{233}U and ^{235}U, for instance, the even-odd effect is about 22%. There is no even-odd effect in fissioning nuclides which have an odd charge.

If one had an accurate formula for Z_p one could then calculate cumulative fission yields for any fissioning nuclide and any fission product of interest. Sufficient experimental data are available to calculate Z_p values for thermal fission of ^{233}U and ^{235}U.[70] A plot of the resulting values as a function of mass is shown in Figure 5. It was noted in calculating these values that evenly charged fission products resulted in Z_p values that were on the average 0.11 charge units less than the average and odd fission products gave a Z_p of 0.11 charge units larger than the average value, $\overline{Z}_p$ This is just the even-odd effect again. The $\overline{Z}_p$ values obtained were fitted to the equations:

$\overline{Z}_p = 0.4153A-1.19$ (A<116) and $\overline{Z}_p=0.4153A-3.43$ (A>116) for ^{235}U

and $\overline{Z}_p = 0.4.53A-.856$ (A<116) and $\overline{Z}_p=0.4153A-2.54$ (A>116) for ^{233}U.

A least squares fit of the values listed in Rider and Meek gives essentially the same result. It was hoped that deviation of $\overline{Z}_p$

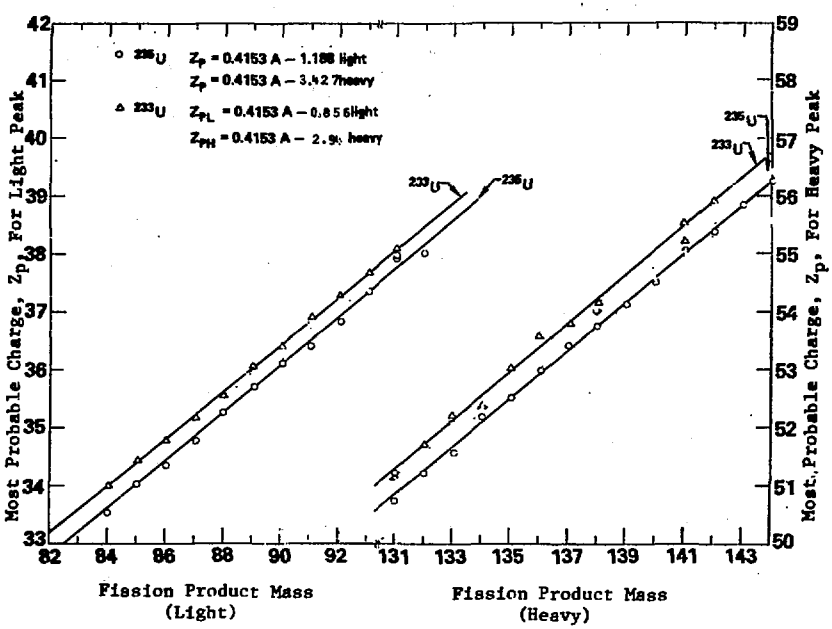

Figure 5. Plot of Z_p Values Versus Mass For ^{233}U and ^{235}U For Light and Heavy Fission Products.*

*For Fission Products of a given mass, A, the average charge (averaged over all observed charges) is known as the most probable charge, Z_p.

for other nuclides would only depend upon the composite mass to charge ratio so it was decided to use as a description of $\overline{Z}_p$ for any composite system of mass A_c and charge Z_c the following equations:

$$\overline{Z}_p = 0.4153A - 1.19 + 0.167*(236 - 92*\frac{Ac}{Zc}) \qquad (A<116)$$

and

$$\overline{Z}_p = 0.4153A - 3.43 + 0.243*(236 - 92*\frac{Ac}{Zc}) \qquad (A>116)$$

The values chosen fit the observed values for ^{233}U and ^{235}U. Thus for a fissioning material of mass A_c and charge Z_c the fission yield of a fission product of mass A and charge Z can be calculated. No attempt has been made to insert an even-odd effect which is the major difference between the approach used here and that used by Rider and Meek. Their formulation for Z_p was derived by using a correlation reported by Nethaway.

Now with this cumulative fission yield model and the most current P_n and half-life values for all the known delayed neutron precursors it is possible to calculate not only the total delayed neutron yield from fission but the time dependence of this yield. The P_n and half-life values used are included at the end of this work. They were taken from Rudstam's latest work[46] and from Rider and Meek.

The results of the calculations are summarized in Table 20. Not only is the calculated absolute yield compared to measured values but the relative yield (normalized to unity) at several points in time are compared to observed values after a continuous irradiation.

Table 20. Comparison of Measured Absolute Delayed Neutron Yields and the Decay of the Relative Yields to Calculated Values for Various Nuclides.

	Absolute Yield per 100 fissions	Relative Yield (Normalized to Unity) t=0.4	t=0.7	t=1.0	t=1.5	t=3.0	t=6.0	t=12.0	t=25.0
^{232}Th Obs	5.27±40	.84	.76	.69	.61	.45	.30	.18	.098
^{232}Th Calc	5.24	.84	.76	.69	.61	.45	.29	.17	.093
^{232}U Obs	0.44±.03	.95	.91	.80	.74	.61	.46	.31	.19
^{232}U Calc	0.45	.93	.88	.85	.79	.66	.51	.35	.22
^{233}U Obs	.74±.04	.90	.86	.80	.74	.61	.46	.31	.19
^{233}U Calc	.79	.92	.87	.83	.77	.63	.47	.32	.19
^{235}U Obs	1.67±.07	.87	.81	.75	.67	.53	.37	.24	.14
^{235}U Calc	1.67	.88	.82	.76	.69	.54	.37	.23	.13
^{238}U Obs	4.44±.23	.78	.68	.61	.53	.38	.24	.14	.073
^{238}U Calc	4.43	.79	.69	.62	.52	.36	.22	.13	.067
^{237}Np Obs	1.06±.10	.87	.81	.75	.68	.54	.39	.25	.15
^{237}Np Calc	1.04	.89	.82	.77	.70	.55	.39	.26	.15
^{238}Pu Obs	0.46±.07	.91	.87	.82	.76	.63	.46	.32	.19
^{238}Pu Calc	0.43	.90	.85	.81	.74	.60	.44	.30	.18

Table 20 Continued

	Absolute Yield per 100 fissions	Relative Yield (Normalized to Unity) t=0.4	t=0.7	t=1.0	t=1.5	t=3.0	t=6.0	t=12.0	t=25.0
Obs	.645±.050	.88	.83	.76	.69	.56	.41	.28	.15
^{239}Pu Calc	.68	.89	.83	.78	.72	.57	.41	.28	.16
Obs	0.90±.09	.87	.80	.74	.67	.53	.39	.26	.15
^{240}Pu Calc	1.05	.88	.81	.76	.69	.54	.38	.25	.15
Obs	1.57±.15	.84	.76	.71	.62	.46	.32	.21	.12
^{241}Pu Calc	1.57	.86	.79	.73	.65	.50	.35	.23	.13
Obs	1.97±.23	.83	.72	.65	.57	.43	.29	.19	.11
^{242}Pu Calc	2.46	.83	.75	.68	.60	.45	.30	.19	.11
Obs	0.51±.06	.90	.83	.78	.72	.58	.42	.29	.17
^{241}Am Calc	0.45	.90	.84	.79	.73	.59	.43	.30	.18
Obs	0.69±.05	.88	.82	.78	.71	.57	.41	.28	.17
^{242m}Am Calc	0.69	.89	.83	.78	.70	.56	.41	.27	.17
Obs	0.59±.04	.87	.82	.77	.71	.57	.42	.28	.17
^{245}Cm Calc	0.75	.89	.83	.77	.71	.56	.41	.28	.17

Table 20. Concluded

	Absolute Yield per 100 fissions	Relative Yield (Normalized to Unity) t=0.4	t=0.7	t=1.0	t=1.5	t=3.0	t=6.0	t=12.0	t=25.0
^{249}Cf Obs	0.27±.02	.92	.87	.80	.74	.60	.45	.31	.18
^{249}Cf Calc	0.36	.90	.85	.81	.75	.62	.47	.34	.21
$^{252}Cf(sf)$ Obs	.86±.10	.78	.67	.59	.49	.36	.25	.17	.11
$^{252}Cf(sf)$ Calc	0.86	.87	.79	.73	.66	.51	.36	.24	.14

As can be seen the agreement is generally excellent. It is perhaps too good in view of the expected even-odd effect which should be reflected in variations in the observed yields from calculated yields.

The even-odd effect is expected to be large in non-fissile nuclides such as ^{232}Th, ^{238}U, ^{240}Pu, and ^{242}Pu. Indeed the measured yields for the plutonium isotopes do appear lower than calculated. However for ^{232}Th and ^{238}U the measured yield does not seem to indicate a significant even-odd effect exists.

As noted earlier the odd Z nuclides ^{237}Np, ^{241}Am, and ^{242m}Am should not have an even-odd effect and fission yield measurements on ^{239}Pu and ^{241}Pu seem to show no effect either.[81] One notes the model does an excellent job predicting the total yield in all these cases. Indeed the only area where there is poor agreement is at very large masses (for ^{245}Cm and ^{252}Cf). It is interesting though that the fit is again good for ^{252}Cf spontaneous fission.

B. Comparison of Experimental and Calculated Total Yields

The only comprehensive attempt at calculating delayed neutron yields for a variety of nuclides has been the work of Rider and Meek[49]. The approach used here is essentially the same except in some cases more current P_n values are used and the Z_p model is different. Rider and Meek also included a postulated even-odd effect for each nuclide studied. It is useful to compare the experimentally determined values

with those calculated by Rider and Meek, This is done in Table 21. Also included is a calculation using Nethaway's correlation (used by Rider and Meek) but without the even-odd effect. Nethaway's Z_p correlation is as follows:

$$Z_p(Z_c,A_c,E^*)=Z_p(92,236,6.52)+a(Z_c-92)+b(A_c-236)+c(E^*-6.52)$$

where for the light mass fission fragments:

$$a=.414\pm.016,\ b=-.143\pm.007,\ \text{and}\ c=.0174$$

and for heavy fission fragments:

$$a=.547\pm.010,\ b=-.188\pm.004,\ \text{and}\ c=.051-.0023(A_H-130).$$

Here A_H is the mass of the heavy fission fragment, and Z_c, A_c, and E^* are the composite charge, mass, and excitation energy of the fissioning nuclide.

It is seen that the model used in this work gave by far the best agreement. Poor agreement was found only for ^{242}Pu, ^{245}Cm, and ^{249}Cf. In these cases the calculated yields were too high possibly because of a large even-odd effect in these nuclides. The Nethaway correlation gave very poor agreement which is not surprising because no even-odd correction was applied. The Rider and Meek results agreed reasonably well but gave poor agreement for ^{232}Th and ^{238}U where a large even-odd effect was assumed and may well not exist. Poor agreement was also obtained for ^{237}Np and ^{242}Pu. In the case

Table 21. Comparison of Experimental and Calculated Absolute Delayed Neutron Yields

NUCLIDE	OBSERVED YIELD %	CALC. YIELD THIS WORK %	RIDER & MEEK[49] YIELD %	NETHAWAY CORREL. %
^{232}Th	5.27±.40	5.24	4.66	5.98
^{232}U	0.44±.03	0.45		0.75
^{233}U	0.74±.04	0.79	0.83	1.11
^{235}U	1.67±.07	1.67	1.72	2.02
^{238}U	4.60±.25	4.43	3.31	4.06
^{237}Np	1.07±.10	1.04	1.22	1.29
^{238}Pu	0.46±.07	0.43		0.55
^{239}Pu	0.65±.05	0.68	0.74	0.72
^{240}Pu	0.90±.09	1.05	0.86	1.11
^{241}Pu	1.57±.15	1.57	1.51	1.43
^{242}Pu	1.97±.23	2.46	1.33	1.84
^{241}Am	0.51±.06	0.45		0.48
^{242m}Am	0.69±.05	0.69		0.62
^{245}Cm	0.59±.04	0.75		0.56
^{249}Cf	0.27±.02	0.36		0.20
^{252}Cf(sf)	0.86±.10	0.86	0.63	0.67
^{238}U(γ,f)	2.91±.09	3.25		
^{235}U(γ,f)	1.02±.04	1.16		

of ^{242}Pu, again a large even-odd effect was assumed and perhaps the real effect is smaller.

C. Comparison of Group Yields

It is useful to group precursors by half-life and compare their calculated yields to the observed group yields. This was done in the following tables (22-31). Note that the precursor groupings were not necessarily fixed due to the various group half-lifes obtained for different fissioning nuclides. For a given fissioning nuclide the average of each of the two adjacent group decay constants was used as the cut-off point for placement of individual precursors. In a general way these comparisons indicate the contribution of individual precursors to each delayed neutron group.

Table 22.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{232}Th.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.41.±.09	.1209±.0069	^{87}Br	55.6	.1598
II	21.29±.08	.704±.027	^{137}I	24.5	.3767
			^{136}Te	17.5	.0255
			^{88}Br	16.0	.4075
			Others		.0027
			Total		.8124
III	5.05±.09	1.33±.06	^{138}I	6.53	.2346
			^{93}Rb	5.85	.0805
			^{89}Br	4.38	.5689
			^{94}Rb	2.76	.3090
			Others		.2456
			Total		1.439

Table 22. continued Th^{232}

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	1.58±.07	2.02±.12	^{139}I	2.38	.2221
			^{85}As	2.03	.7071
			^{98}Y	2.0	.1009
			^{90}Br	1.92	.4691
			^{104}Nb	1.0	.0093
			^{144}Cs	1.002	.0673
			^{86}As	.9	.0973
			Others		.3587
			Total		2.023
V	.39±.14	.786±.29	^{140}I	.60	.2153
			^{145}Cs	.58	.0676
			^{91}Br	.542	.0771
			^{141}I	.47	.0788
			^{95}Rb	.38	.1141
			^{92}Br	.36	.0310

Table 22. concluded Th^{232}

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{96}Rb	.201	.0475
			Others		.1724
			Total		.803
All Groups		5.02±.26	All Precursors		5.238

Table 23.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{232}U.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	54.32±.17	.0524±.0040	^{87}Br	55.6	.0493
II	19.79±.16	.131±.010	^{137}I	24.5	.0503
			^{136}Te	17.5	.0007
			^{88}Br	16.0	.0947
			Others		.0010
			Total		.1467
III	4.82±.20	.134±.014	^{138}I	6.53	.0095
			^{93}Rb	5.85	.0209
			^{89}Br	4.38	.0705
			^{94}Rb	2.76	.0453
			Others		.0025
			Total		.1487

Table 23. continued ^{232}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{139}I	2.38	.0024
			^{85}As	2.03	.0240
			^{98}Y	2.0	.0286
IV	1.75±.20	.113±.012	^{90}Br	1.92	.0246
			^{104}Nb	1.0	.0001
			^{114}Cs	1.002	.0003
			^{86}As	.9	.0011
			Others		.0080
			Total		.0891
			^{140}I	.60	.0005
			^{145}Cs	.58	.0001
V	.514	.007±.039	^{91}Br	.542	.0017
			^{141}I	.47	.0001
			^{95}Rb	.38	.0070

Table 23. concluded ^{232}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{92}Br	.36	.0003
			^{96}Rb	.201	.0011
			Others		.0038
			Total		.0146
All Groups		0.437±.033	All Precursors		0.4484

Table 24.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{233}U.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.94±.18	.0551±.0037	^{87}Br	55.6	.0633
II	26.76±1.96	.070±.027	^{137}I	24.5	.0999
III	17.42±1.05	.160±.024	^{136}Te	17.5	.0023
			^{88}Br	16.0	.1378
			Others		.0016
			Total		.1417
IV	4.31±.27	.175±.024	^{138}I	6.53	.0207
			^{93}Rb	5.85	.0315
			^{89}Br	4.38	.1222
			Others		.0017
			Total		.1761

Table 24. continued ^{233}U.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 Fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
V	2.42±.24	.188±.030	^{94}Rb	2.76	.0815
			^{139}I	2.38	.0074
			^{85}As	2.03	.0525
			^{98}Y	2.0	.0483
			^{90}Br	1.92	.0518
			^{104}Nb	1.0	.0005
			^{144}Cs	1.002	.0012
			^{86}As	.9	.0027
			Others		.0234
			Total		.2693
VI	.53±.16	.084±.013	^{140}I	.60	.0021
			^{145}Cs	.58	.0004
			^{91}Br	.542	.0039
			^{141}I	.47	.0002
			^{95}Rb	.38	.0143

Table 24. concluded: ^{233}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{92}Br	.36	.0007
			^{96}Rb	.201	.0029
			Others		.0112
			Total		.0357
All Groups		.733±.047	All Precursors		.786

Table 25.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{235}U.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.23±.13	.0566±.0011	^{87}Br	55.6	.0514
II	22.43±.07	.358±.007	^{137}I	24.5	.2224
			^{176}Te	17.5	.0098
			^{88}Br	16.0	.1482
			Others		.0035
			Total		.3839
III	6.08±.13	.346±.011	^{138}I	6.53	.0939
			^{93}Rb	5.85	.0493
			^{89}Br	4.38	.2019
			Others		.0059
			Total		.3600

Table 25. continued ^{235}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	2.11±.05	.672±.018	^{94}Rb	2.76	.1830
			^{139}I	2.38	.0505
			^{85}As	2.03	.0930
			^{98}Y	2.0	.0983
			^{90}Br	1.92	.1425
			Others		.0904
			Total		.6577
V	.336±.050	.303±.045	^{104}Nb	1.0	.0025
			^{144}Cs	1.002	.0107
			^{86}As	.9	.0082
			^{140}I	.60	.0246
			^{145}Cs	.58	.0063
			^{91}Br	.542	.0166
			^{141}I	.47	.0049
			^{95}Rb	.38	.0512

Table 25. concluded ^{235}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{92}Br	.36	.0047
			^{96}Rb	.201	.0165
			Others		.0698
			Total		.2160
All Groups		1.654±.033	All Precursors		1.669

Table 26.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{238}U.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.27±.13	0.0487±.0040	^{87}Br	55.6	.0365
II	22.93±.086	.557±.042	^{137}I	24.5	.3637
			^{136}Te	17.5	.0405
			^{88}Br	16.0	.1211
			Others		.0083
			Total		.5336
III	7.98±.29	.358±.035	^{138}I	6.53	.2267
			^{93}Rb	5.85	.0585
			^{89}Br	4.38	.2528
			Others		.0399
			Total		.5779

Table 26. continued ^{238}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	2.82	1.656±.140	^{94}Rb	2.76	.3173
			^{139}I	2.38	.2619
			^{85}As	2.03	.1559
			^{98}Y	2.0	.1527
			^{90}Br	1.92	.2545
			Others		.5657
			Total		1.7080
V	.98±.07	1.212±.124	^{104}Nb	1.0	.0195
			^{144}Cs	1.002	.0561
			^{86}As	.9	.0273
			^{140}I	.60	.2821
			^{145}Cs	.58	.0843
			^{91}Br	.542	.0672
			^{141}I	.47	.1249

Table 26. concluded ^{238}U

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			Others		.4776
			Total		1.139
			^{95}Rb	.38	.1473
			^{92}Br	.36	.0360
VI	.28±.12	.82±.50	^{96}Rb	.201	.0959
			Others		.1558
			Total		.435
All Groups		4.65±.35	All Precursors		4.430

Table 27.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{237}Np.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.10±.18	.0368±.0034	^{87}Br	55.6	.0330
			^{137}I	24.5	.1840
			^{136}Te	17.5	.0073
II	22.65±.25	.244±.024	^{88}Br	16.0	.0800
			Others		.0042
			Total		.2755
III	10.61±2.6	.070±.033	^{138}I	6.53	.0622
			^{93}Rb	5.85	.0349
IV	4.99±.68	.153±.065	^{89}Br	4.38	.0892
			Others		.0034
			Total		.1275

Table 27. continued ^{237}Np.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
V	2.11±.19	.424±.053	^{94}Rb	2.76	.1177
			^{139}I	2.38	.0279
			^{85}As	2.03	.0521
			^{98}Y	2.0	.0904
			^{90}Br	1.92	.0588
			^{104}Nb	1.0	.0044
			^{144}Cs	1.002	.0050
			^{86}As	.9	.0038
			Others		.0953
			Total		.4564
VI	.428±.182	.132±.031	^{140}I	.60	.0115
			^{145}Cs	.58	.0029
			^{91}Br	.542	.0073
			^{141}I	.47	.0020

Table 27. concluded ^{237}Np.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{95}Rb	.38	.0328
			^{92}Br	.36	.0021
			^{96}Rb	.201	.0096
			Others		.0132
			Total		.0814
All Groups		1.06±.10	All Precursors		1.036

Table 28.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{238}Pu.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	54.92±.57	.01968±.0031	^{87}Br	55.6	.0201
II	22.19±.27	.1419±.022	^{137}I	24.5	.0942
			^{136}Te	17.5	.0021
			^{88}Br	16.0	.0383
			Others		.0024
			Total		.1370
III	8.15±1.15	.0528±.031	^{138}I	6.53	.0206
			^{93}Rb	5.85	.0176
			Others		.0005
			Total		.0387

Table 28. continued ^{238}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	3.52±.41	.0815±.013	^{89}Br	4.38	.0381
			^{94}Rb	2.76	.0538
			Others		.0032
			Total		.0951
V	1.95±.28	.151±.024	^{139}I	2.38	.0061
			^{85}As	2.03	.0158
			^{98}Y	2.0	.0506
			^{90}Br	1.92	.0172
			^{104}Nb	1.0	.0027
			^{144}Cs	1.002	.0009
			^{86}As	.91	.0009
			Others		.0203
			Total		.1145

Table 28. Concluded ^{238}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Nuet/100 fiss)
VI	.514	.015±.087	^{140}I	.60	.0016
			^{145}Cs	.58	.0003
			^{91}Br	.542	.0016
			^{141}I	.47	.0002
			^{95}Rb	.38	.0095
			^{92}Br	.36	.0003
			^{96}Rb	.201	.0025
			Others		.0103
			Total		.0263
All Groups		.461±.073	All Precursors		.432

Table 29.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{239}Pu.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	55.63±.05	.01895±.0009	^{87}Br	55.6	.0178
II	23.57±.64	.1825±.0089	^{137}I	24.5	.1548
			^{136}Te	17.5	.0047
			^{88}Br	16.0	.0437
			Others		.0041
			Total		.2073
III	9.71±.49	.0780±.0087	^{138}I	6.53	.0424
			^{93}Rb	5.85	.0229
			Others		.0014
			Total		.0667

Table 29. continued ^{239}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	3.27±.28	.158±.031	^{89}Br	4.38	.0488
			^{94}Rb	2.76	.0805
			Others		.0052
			Total		.1345
V	2.14±.32	.147±.031	^{139}I	2.38	.0169
			^{85}As	2.03	.0229
			^{98}Y	2.0	.0741
			^{90}Br	1.92	.0279
			^{104}Nb	1.0	.0048
			^{144}Cs	1.002	.0026
			^{86}As	.9	.0015
			Others		.0440
			Total		.1947

Table 29. concluded ^{239}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
VI	.54±.11	.119±.015	^{140}I	.60	.0059
			^{145}Cs	.58	.0012
			^{91}Br	.542	.0031
			^{141}I	.47	.0009
			^{95}Rb	.38	.0203
			^{92}Br	.36	.0008
			^{96}Rb	.201	.0028
			Others		.0266
			Total		.0616
All Groups		.703±.049	All Precursors		.683

Table 30.

Comparison of Measured Group Parameters Versus Calculated Precursors Contributions for ^{240}Pu.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	53.56±1.21	.022±.003	^{87}Br	55.6	.0197
II	22.14±.38	.238±.016	^{137}I	24.5	.2215
			^{136}Te	17.5	.0098
			^{88}Br	16.0	.0491
			Others		.0050
			Total		.2854
III	5.14±.42	.162±.044	^{138}I	6.53	.0846
			^{93}Rb	5.85	.0283
			^{89}Br	4.38	.0610
			Others		.0042
			Total		.1781

Table 30. continued ^{240}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	2.08±.19	.315±.027	^{94}Rb	2.76	.1116
			^{139}I	2.38	.0420
			^{85}As	2.03	.0382
			^{98}Y	2.0	.0903
			^{90}Br	1.92	.0425
			^{104}Nb	1.0	.0076
			^{144}Cs	1.002	.0071
			^{86}As	.9	.0029
			Others		.1062
			Total		.4484
V	0.511±.077	.119±.018	^{140}I	.60	.0174
			^{145}Cs	.58	.0045
			^{91}Br	.542	.0058
			^{141}I	.47	.0033

Table 30. concluded ^{240}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{95}Rb	.38	.0323
			^{92}Br	.36	.0020
			Others		.0369
			Total		.1002
			^{96}Rb	.201	.0118
VI	0.172±.033	.029±.006	Others		.0058
			Total		.0176
All Groups		.088±.06	All Precursors		1.051

Table 31.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{241}Pu.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	53.48±.41	.0195±.0012	^{87}Br	55.6	.0149
II	23.42±.16	.324±.017	^{137}I	24.5	.3083
			^{136}Te	17.5	.0167
			^{88}Br	16.0	.0485
			Others		.0062
			Total		.3797
III	10.5±1.3	.086±.018	^{138}I	6.53	.1430
			^{93}Rb	5.85	.0285
			Others		.0086
			Total		.1801
			^{89}Br	4.38	.0660
			^{94}Rb	2.76	.1362

Table 31. continued ^{241}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	3.54±.16	.473±.036	^{139}I	2.38	.0866
			^{85}As	2.03	.0430
			^{98}Y	2.0	.1056
			^{90}Br	1.92	.0575
			Others		.1509
			Total		.6458
V	1.00±.07	.598±.035	^{104}Nb	1.0	.0129
			^{144}Cs	1.002	.0166
			^{86}As	.9	.0046
			Others		.1050
			Total		.1391
			^{140}I	.60	.0578
			^{145}Cs	.58	.0134
			^{91}Br	.542	.0095

Table 31. concluded ^{241}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
VI	.514	.058±.089	^{141}I	.47	.0124
			^{95}Rb	.38	.0495
			^{92}Br	.36	.0040
			^{96}Rb	.201	.0180
			Others		.0440
			Total		.2086
All Groups		1.56±.07	All Precursors		1.568

Table 32.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{242}Pu.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	51.[illegible]04	.0221±.0027	^{87}Br	55.6	.0192
II	23.50±1.19	.316±.104	^{137}I	24.5	.3992
III	16.95±5.8	.062±.10	^{136}Te	17.5	.0280
			^{88}Br	16.0	.0580
			Others		.0069
			Total		.0929
IV	5.46±.24	.322±.030	^{138}I	6.53	.2075
			^{93}Rb	5.85	.0323
			^{89}Br	4.38	.0961
			^{94}Rb	2.76	.1810
			Others		.0606
			Total		.5775

Table 32. continued ^{242}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
V	1.75±.15	.721±.071	^{139}I	2.38	1.677
			^{85}As	2.03	.0615
			^{98}Y	2.0	.1239
			^{90}Br	1.92	.0931
			^{104}Nb	1.0	.0153
			^{144}Cs	1.002	.0310
			^{86}As	.9	.0079
			^{140}I	.60	.1203
			^{145}Cs	.58	.0355
			^{91}Br	.542	.0182
			Others		.4549
			Total		1.1293
VI	.31±.12	.523±.169	^{141}I	.47	.0409
			^{95}Rb	.38	.0751
			^{92}Br	.36	.0088

Table 32. concluded ^{242}Pu

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{96}Rb	.201	.0481
			Others		.0681
			Total		.2410
All Groups		1.97±.23	All Precursors		2.460

Table 33.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{241}Am.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	54.54±.13	.0185±.0022	^{87}Br	55.6	.0097
II	23.22±.03	.146±.018	^{137}I	24.5	.1257
			^{136}Te	17.5	.0031
			^{88}Br	16.0	.0211
			Others		.0032
			Total		.1531
III	4.56±.09	.154±.019	^{138}I	6.53	.0325
			^{93}Rb	5.83	.0133
			^{89}Br	4.38	.0229
			^{94}Rb	2.76	.0489
			^{139}I	2.38	.0108

Table 33. continued ^{241}Am

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			Others		.0062
			Total		.1346
IV	1.55±.076	.154±.020	^{85}As	2.03	.0104
			^{98}Y	2.0	.0622
			^{90}Br	1.92	.0125
			^{104}Nb	1.0	.0039
			^{144}Cs	1.002	.0015
			^{86}As	.9	.0006
			^{140}I	.60	.0032
			^{145}Cs	.58	.0006
			^{91}Br	.542	.0014
			^{141}I	.47	.0004
			Others		.0390
			Total		.1357

Table 33. concluded ^{241}Am

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
V	.263±.211	.036±.048	^{95}Rb	.38	.0132
			^{92}Br	.36	.0003
			^{96}Rb	.201	.0035
			Others		.0015
			Total		.0185
All Groups		0.509±.060	All Precursors		0.453

Table 34.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{242m}Am.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	54.45±.21	.0176±.0012	^{87}Br	55.6	.0137
			^{137}I	24.5	.1630
			^{136}Te	17.5	.0063
II	23.09±.09	.195±.013	^{88}Br	16.0	.0328
			Others		.0045
			Total		.2066
			^{138}I	6.53	.0532
			^{93}Rb	5.85	.0170
III	7.45±.43	.0822±.0092	^{89}Br	4.38	.0402
			Others		.0025
			Total		.1129

Table 34. continued ^{242m}Am

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	2.82±.081	.244±.026	^{94}Rb	2.76	.0635
			^{139}I	2.38	.0249
			^{85}As	2.03	.0217
			^{98}Y	2.0	.0690
			^{90}Br	1.92	.0236
			Others		.0638
			Total		.2665
V	1.06±.13	.119±.013	^{99}Y	1.40	.0112
			^{104}Nb	1.0	.0077
			^{100}Y	.756	.0145
			Others		.0022
			Total		.0356
			^{140}I	.60	.0098
			^{145}Cs	.58	.0023

Table 34. concluded ^{242m}Am

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
VI	.514	0.030±.045	^{91}Br	.542	.0030
			^{141}I	.47	.0016
			^{95}Rb	.38	.0176
			^{92}Br	.36	.0009
			^{96}Rb	.201	.0059
			Others		.0093
			Total		.0504
All Groups		0.688±.045	All Precursors		0.686

Table 35.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{245}Cm.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	51.92±.35	.0140±.0009	^{87}Br	55.6	.0094
			^{137}I	24.5	.2031
			^{136}Te	17.5	.0083
II	22.87±.10	.179±.012	^{88}Br	16.0	.0236
			Others		.0046
			Total		.2396
III	6.70±.91	.054±.017	^{138}I	6.53	.0734
			^{93}Rb	5.85	.0128
			Others		.0035
			Total		.0897
			^{89}Br	4.38	.0331
			^{94}Rb	2.76	.0533

Table 35. continued ^{245}Cm

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
IV	3.29±.17	.174±.031	^{139}I	2.38	.0366
			^{85}As	2.03	.0186
			^{98}Y	2.0	.0570
			^{90}Br	1.92	.0230
			Others		.0286
			Total		.2502
V	1.29±.18	.136±.016	^{143}Cs	1.78	.0137
			^{135}Sb	1.71	.0181
			^{99}Y	1.40	.0102
			^{100}Y	.756	.0150
			Others		.0389
			Total		.0959
			^{140}I	.60	.0161
			^{145}Cs	.58	.0033

Table 35. concluded ^{245}Cm

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
VI	.514	.035±.050	^{91}Br	.542	.0026
			^{141}I	.47	.0029
			^{95}Rb	.38	.0165
			^{92}Br	.36	.0008
			^{96}Rb	.201	.0060
			Others		.0133
			Total		.0615
All Groups		0.592±.039	All Precursors		0.746

Table 36.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{249}Cf.

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	53.94±.09	.00765±.00056	^{87}Br	55.6	.0043
II	22.82±.03	.0944±.0069	^{137}I	24.5	.1293
			^{136}Te	17.5	.0034
			^{88}Br	16.0	.0086
			Others		.0035
			Total		.1448
III	4.13±.09	.102±.009	^{138}I	6.53	.0356
			^{93}Rb	5.85	.0066
			^{89}Br	4.38	.0091
			^{94}Rb	2.76	.0218
			^{139}I	2.38	.0139
			^{85}As	2.03	.0065

Table 36. concluded ^{249}Cf

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			^{98}Y	2.0	.0303
			Others		.0184
			Total		.1422

Table 37.

Comparison of Measured Group Parameters Versus Calculated Precursor Contributions for ^{252}Cf (spontaneous fission)

Group	Half-Life (sec)	Observer Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
I	20.0±.5	.22±.01	^{87}Br	55.6	.0036
			^{137}I	24.5	.2035
			^{136}Te	17.5	.0086
			^{88}Br	16.0	.0109
			^{138}I	6.53	.1074
			^{93}Rb	5.85	.0072
			^{89}Br	4.38	.0181
			Others		.0152
			Total		.3745
II	2.0±.4	.29±.04	^{94}Rb	2.76	.0356
			^{139}I	2.38	.0725
			^{85}As	2.03	.0075
			^{98}Y	2.0	.0404

Table 37. continued ^{252}Cf

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
			Br	1.92	.0149
			Nb	1.0	.0087
			Cs	1.002	.0182
			As	.9	.0007
			Others		.1580
			Total		.3565
			^{140}I	.60	.0427
			^{145}Cs	.58	.0157
			^{91}Br	.542	.0025
			^{141}I	.47	.0105
III	0.5±.4	.35±.10	^{95}Rb	.38	.0135
			^{92}Br	.36	.0135
			^{96}Rb	.201	.0058
			Others		.0397
			Total		.1313

Table 37. concluded ^{252}Cf

Group	Half-Life (sec)	Observed Group Yield (Neut/100 fiss)	Precursor	Half-Life (sec)	Calculated Yield (Neut/100 fiss)
All Groups		0.86±.10	All Precursors		.862

D. The Even-Odd Effect

Rider and Meek's calculated delayed neutron yields were too low in all three of the cases where an even-odd effect of greater than 30% was assumed (^{232}Th, ^{238}U, and ^{242}Pu). It would appear that a lower even-odd effect should have been used. Indeed the magnitude of the difference was such that no even-odd effect seems to exist for ^{232}Th and ^{238}U. Using the $\bar{Z}_p$ model developed in this work the only nuclides whose experimentally determined yields were lower than the calculated yields were ^{240}Pu, ^{242}Pu, ^{245}Cm, and ^{249}Cf. In view of the good fits for nearby nuclides with low known even-odd effects (^{239}Pu, ^{241}Pu, ^{241}Am, and ^{242m}Am) it seems likely that these nuclides do have significant even-odd effects. Table 38 shows a summary of known information. In a few cases estimates of the even-odd effect have been made although ^{233}U and ^{235}U thermal fission are the only reliable ones. In column 3 is indicated the effect assumed by Rider and Meek with an inequality expressing the direction the even-odd effect should go to give a fit with observed delayed neutron data (ie <<32.7% means the assumed value of 32.7% even-odd effect was much too large according to the delayed neutron yield actually observed).

Column 4 shows the information about the calculational model used in this work. No even-odd effect was used so for cases where the delayed neutron yield was lower than calculated the even-odd

Table 38. Estimated Even-Odd Effect for Fissioning Nuclides

NUCLIDE	MEASURED EFFECT %	MODIFIED RIDER & MEEK VALUE %[49]	THIS WORK %	RECOMMENDED VALUE %
^{232}Th	38±13	<<32.7	≈0	≈0
^{232}U			≈0	≈0
^{233}U	22±7	>21.0	≈5	≈20
^{235}U	22±7	>22.8	≈0	≈20
^{238}U	20±11	<<32.9	≈0	≈0
^{237}Np	0.0	>0.0	≈0	0.0
^{238}Pu			≈0	≈0
^{239}Pu	11±9 ≈0	>17.1	≈5	≈10
^{240}Pu		<24.4	≈4	≈10
^{241}Pu	≈0	<20.6	≈0	≈0
^{242}Pu		<<36.4	≈36	≈30
^{241}Am	0.0		≈0	0.0
^{242m}Am	0.0		≈0	0.0
^{245}Cm			≈20	≈20
^{249}Cf			≈10	≈10
$^{252}Cf(sf)$		<<5.0	≈0	≈0
$^{238}U(\gamma,f)$			≈13	≈13
$^{235}U(\gamma,f)$			≈10	≈10

effect indicated is the effect which when used gave agreement between calculation and observed yields.

Column 5 shows estimates of the actual even-odd effects. In the odd Z fissioning cases the effect is zero. In some cases the effect has been well measured (^{233}U and ^{235}U). In the other cases a best estimate of the relative accuracies of the indicators was used. While being a rough measure of the effect it is none-the-less useful to have some measure of the even-odd effect to substantiate theoretical arguments on the subject. The most interesting nuclides to study in this regard appear to be ^{242}Pu and ^{245}Cm.

CHAPTER VI

CONCLUSIONS

It is concluded that total delayed neutron yield can be simply expressed in terms of an empirical fit and that this fit is accurate for a large variety of nuclides from ^{232}Th to ^{252}Cf. The fit does indeed hold in the previously unmeasured region between ^{242}Pu and ^{252}Cf.

Secondly the time dependent decay of delayed neutrons can also be expressed with an empirical fit. Nuclides with similar $\frac{Ac}{Zc}$ ratios have similar relative decay patterns. Thus the relative decay pattern for one nuclide may be estimated by another measured nuclide with a similar mass to charge ratio.

Most importantly it has been found that it is possible to accurately reproduce the observed yield and decay characteristics of delayed neutrons using a simple fission yield model and known precursor characteristics. Such an approach is more realistic than an empirical correlation because it relies on the actual mechanism of delayed neutron production.

Having established that such a complex tabulation of precursors is possible and the P_n values are accurate, delayed neutron yields then can be used to study fission yields for these precursors. Cumulative fission yields for ^{87}Br and ^{137}I have been derived by studying the group-wise decay of delayed neutron emission for a

large number of nuclides in a non-destructive fashion. Such a technique can be used to test fission yield models in general since if they do not predict the observed delayed neutron yields they are not accurate.

Finally delayed neutron studies indicate that the even-odd effect is not yet well understood. Nuclides such as ^{232}Th and ^{238}U, were supposed to have large even-odd effects, and yet seem instead to have very small effects. Estimates of the size of the even-odd effect have been made for a large variety of nuclides.

APPENDIX A

COMPUTER PROGRAMS

The programs used in this work were written in FORTRAN for use on an LSI-11 minicomputer. Because of the limitation in memory size great use was made of interactive data files. The programs were therefore slow running but this was of small consequence.

The program TX was used to calculate fission yields and multiply the yields of delayed neutron precursors by their P_n values and thereby predict delayed neutron yields. A data file (File 1) was prepared with the chain yields for masses from 79 to 150 for the fissioning nuclide in question. File 2 contained precursor data (precursor charge, mass, half-life, and P_n) ordered by half-life from ^{87}Br to ^{99}Rb. Table 39 shows the values used in this data file. The file was the same for all fissioning nuclides studied. Figure 6 is the program listing for program TX. File 3 contained values for integrals of a Gaussian function and was treated as a data look-up file when conversion was needed from $(Z-Z_p)$ to

$$\int_0^{Z+.5} \exp\left[-\frac{(z-Z_p)^2}{2\sigma_z^2}\right] dz \ .$$

Starting with the first precursor, ^{87}Br, the program calculated the appropriate Z_p for that mass (A=87) using the fission yield model

Table 30. Delayed Neutron Parameters (P_n and Half-Life Values) Used In This Work.[46,49]

Precursor Charge	Mass	P_n(%)	Half-Life (sec)	Precursor Charge	Mass	P_n(%)	Half-Life (sec)
35	[illegible]	2.38	55.6	39	98	3.4	2.0
55	[illegible]	0.036	24.9	43	109	1.7	2.0
53	1[illegible]	[illegible]	24.4	35	90	21.2	1.92
52	136	[illegible]	17.5	32	83	0.17	1.9
35	88	6.7	16.0	42	110	1.3	1.892
41	103	0.13	15.66?	36	92	0.033	1.85
51	134	0.108	10.4	41	105	2.9	1.8
56	147	5.2	10.0	56	150	0.24	1.798
53	138	5.3	6.53	55	143	1.68	1.78
37	93	1.39	5.85	54	141	0.044	1.73
33	84	0.090	5.6	55	142	0.091	1.71
34	87	0.190	5.6	51	135	15.6	1.71
37	92	0.012	4.5	31	80	0.8	1.66
35	89	13.5	4.38	34	88	0.6	1.52
40	104	0.11	3.783	47	122	1.4	1.5
39	97	0.06	3.7	50	133	0.02	1.47
57	149	0.81	2.864	52	138	6.3	1.4
31	79	0.094	2.86	39	99	1.2	1.4
52	137	2.50	2.8	36	93	1.96	1.29
37	94	10.4	2.76	54	142	0.42	1.24
30	79	1.1	2.74	31	81	11.9	1.23
49	129	3.5	2.5	32	84	10.	1.2
53	139	9.42	2.38	38	100	5.	1.046
56	147	5.2	2.23	50	134	17.	1.04
33	85	50.0	2.03	42	109	0.53	1.033
49	127	0.65	2.0	55	144	3.	1.002

Table 39. (continued)

Precursor Charge	Precursor Mass	P_n(%)	Half-Life (sec)	Precursor Charge	Precursor Mass	P_n(%)	Half-Life (sec)
41	104	0.71	1.0	52	139	6.3	0.424
54	144	0.73	1.0	34	89	5.	0.41
56	149	0.03	0.917	47	123	4.6	0.39
33	86	12.	0.9	37	95	8.8	0.384
49	128	0.057	0.84	35	92	22.	0.362
43	110	3.1	0.83	55	146	13.2	0.335
48	128	0.11	0.83	53	143	18.	0.328
51	136	23.	0.82	31	83	56.	0.31
38	98	0.36	0.8	54	143	1.2	0.3
39	100	5.5	0.756	50	135	8.6	0.291
33	87	44.	0.73	51	137	20.	0.284
57	150	0.94	.648	49	131	1.73	0.28
31	82	21.9	0.6	34	91	21.	0.27
53	140	23.	0.6	32	86	22.	0.259
38	99	3.4	0.6	32	85	20.	0.234
55	145	13.3	0.585	55	147	25.4	0.21
49	130	1.38	0.58	36	94	5.7	0.208
40	105	1.4	0.559	37	96	14.2	0.201
34	90	11.	0.555	35	93	41.	0.201
35	91	10.9	0.542	53	142	16.	0.196
41	106	5.5	0.535	37	97	28.	0.17
36	95	9.5	0.5	49	132	4.3	0.13
56	148	23.9	0.5	37	98	16.	0.119
53	141	39.	0.47	37	99	15.	0.076
38	97	0.27	0.43				

```
C THIS PROGRAM CALCS DN FRACTION
      DIMENSION COUNT(8), TIME(8)
     TOT=0.
       WRITE(7,993)
993      FORMAT(3X,'TYPE AC,THEN ZC F12.3')
         READ(5,994) AC,ZC
994       FORMAT(F12.3)
      WRITE(7,936)
936      FORMAT(3X,'TYPE EX. ENERGY')
       READ(5,994) EST
        WRITE(7,995)
995      FORMAT(3X,'TYPE E/O')
         READ(5,996) EPSI
996       FORMAT(F12.6)
997      FORMAT(3X,'TYPE NU')
     TIME(1)=0.4
      TIME(2)=0.7
     TIME(3)=1.0
     TIME(4)=1.5
     TIME(5)=3.
      TIME(6)=6.
      TIME(7)=12.
      TIME(8)=25.
     DO 445 I=1,8
     COUNT(I)=0.
445     CONTINUE
      D=.030
1     READ(2,901)Z,A,PN,HL
      IF(A.LT.1)GO TO 1
     IF(Z.GT.200) GO TO  100
    GO TO 3
901   FORMAT(4F7.3)
3     REWIND 1
2    READ(1,902)ZA,YC
980   FORMAT(3X,2(F12.6,3X))
      IZA=ZA
     IA=A
      IF(IZA.NE.IA) GO TO 2
902      FORMAT(2F12.6)
      IZ=Z/2
     IZ=IZ*2
      EPS=EPSI*-1.
   IF(Z-IZ.GT.0.1) EPS=EPSI
     K=.87
    IF(Z-IZ.GT.0.1) K=1.19
```

Figure 6. Printout of Program TX for Calculating Delayed Neutron Yields For Any Nuclide of Interest.

```
      XN=A-Z
       N=XN/2
      N=N*2
      EPS2=.193*EPS
       IF(XN-N.GT.0.1) EPS2=-.193*EPS
      EPS2=0
      PPB=(EST-6.52)*(.0509-.00233*(A-130))
      CFT=.547*(ZC-92)-3.171-.254*(AC-236)+PPB
       IF(A.LT.118)CFT=.474*(ZC-92)-.169*(AC-236)+.0174*
     C (EST-6.52)-.541
       ZP=0.4153*A-1.70+.167*(236-92*AC/ZC)+EPS-Z
      IF(A.GT.117) ZP=0.4153*A-3.954+.243*(236-92*AC/ZC)-Z+EPS
     DD=ZP/.56
      CT=1
      IF(DD.LT.0) CT=-1
      IF(DD.LT.0)DD=-1*DD
      REWIND 3
4     READ(3,908)X1
      READ(3,908)Y1
         IF(DD.GT.3.9) DD=3.9
      IF(X1.GT.DD) GO TO 5
      X2=X1
       Y2=Y1
      GO TO 4
5      Y=Y1-(Y1-Y2)/(X1-X2)*(X1-DD)
       Y=(1-Y)/2
      IF(CT.LT.0) Y=1-Y
       Y=Y*YC
      PY=Y*PN
      TOT=TOT+PN*Y
      XL=-0.6931/HL
    DO 446 ILQ=1,8
      COUNT(ILQ)=COUNT(ILQ)+PY*EXP(XL*TIME(ILQ))
446    CONTINUE
       WRITE(7,990)Z,A,Y,PY,HL,TOT
990      FORMAT(1X,6(F8.4,3X))
       GO TO 1
100       WRITE(7,906) TOT
906      FORMAT(3X,'PU239TH TOTAL D NEUTS = ',F12.6)
908       FORMAT(F12.8)
      DO 284 I=1,8
      COUNT(I)=COUNT(I)/TOT
       WRITE(7,937) TIME(I),COUNT(I)
937     FORMAT(6X,F5.2,3X,F12.8)
284     CONTINUE
       END
```

Figure 6 (continued)

$$(A<116) \qquad Z_p = 0.4153 - 1.19 + 0.167\left(236 - 92*\frac{Ac}{Zc}\right) \quad \text{or}$$

$$(A>116) \qquad Z_p = 0.4153 - 3.43 + 0.243\left(236 - 92*\frac{Ac}{Zc}\right)$$

where Z_c and A_c were the compound charge and mass of the fissioning nuclide.

Next $(Z-Z_p)$ was calculated and the corresponding integral of the Gaussian found in File 3. This quantity was the relative cumulative yield for the precursor in question (^{87}Br). This was then multiplied by the chain yield (A=87) and the P_n value (P_n=2.38%) to give the delayed neutron contribution from that precursor (^{87}Br) to the entire delayed neutron yield.

This process was repeated for all precursors and a summation of all individual contributions gave the total calculated delayed neutron yield for that fissioning nuclide.

In addition the program calculated the decay of each precursor for several specific times (eg. 0.4 sec, 1 sec, et cetera) and calculated the relative time dependent neutron yields for those times. The calculated values could then be compared to experimental values.

The program KEEP calculated the summations necessary to do the least squares fitting of the experimental data. In this case File 3 contained the experimental data as a function of time. File 1 was the input initial group yields and decay constants to be used. File 2 was the output file which contained the summations which were

```
      DIMENSION SUM(12,12),X(12),XL(6),B(6),XY(12),D(12)
     REWIND 1
      REWIND 2
    NORDER=8
     NN=0
    NARDER=NORDER
      NOR2=NORDER/2
     DO 99 KKY=1,NOR2
      READ(2,901) B(KKY)
      READ(2,901) XL(KKY)
99    CONTINUE
      DO 10 K=1,NOR2
     WRITE(7,903) K
903    FORMAT(3X,'B(',I1,')=')
      READ(2,901) A
904     FORMAT(3X,'XL(',I1,')=')
     READ(2,901) AQ
     XL(K)=XL(K)+AQ*0.9
     B(K)=B(K)+A*0.9
      IF(B(K).LT.100.) B(K)=100.
    IF(XL(K).GT.5.) XL(K)=5.
10       CONTINUE
23     TOT=0
     XX=0
     DO 3 I=1,NORDER
     XY(I)=0
     DO 3 J=1,NORDER
    SUM(I,J)=0
3     CONTINUE
     REWIND 3
35       CONTINUE
    READ(3,901) DT
      READ(3,901) TO
     READ(3,901) CH
     READ(3,901) TIR
901    FORMAT(E14.7)
       DO 53 KS=1,NOR2
       ET=-1.*TIR*XL(KS)
     D(KS)=CH*(1-EXP(ET))
   WRITE(7,901) D(KS)
53       CONTINUE
    T=TO-DT
```

Figure 7. Printout of Program KEEP for Calculating Best Fits for Delayed Neutron Data for a Given Number of Groups.

```
1       T=T+DT
         TOT=0
         READ(3,901)C
         IF(C.LT.-.0010) GO TO 400
          IF(C.LT.0.01) GO TO 35
         CXX=C
        CCC=C
          IF(DT.GT.0.05) CCC=C-196.13
          IF(DT.GT.0.7) CCC=C-785.72
         C=CCC
          DDX=C/CH
948           FORMAT(2F12.3)
         NN=NN+1
         DO 2 I=1,NOR2
        X(I)=D(I)*EXP(-1*XL(I)*T)
        TOT=TOT+X(I)*B(I)
       J=I+NOR2
         X(J)=-1*D(I)*B(I)*T*EXP(-1*XL(I)*T)
2      CONTINUE
       YY=C-TOT
        XX=XX+YY**2/CXX
        DO 4 I=1,NORDER
        XY(I)=XY(I)+YY/C*X(I)
        DO 4 J=1,NORDER
        SUM(I,J)=SUM(I,J)+X(I)*X(J)/C
4        CONTINUE
          GO TO 1
400        CONTINUE
        DO 456 KKY=1,NOR2
       WRITE(1,901) B(KKY)
        WRITE(1,901) XL(KKY)
456         CONTINUE
         DO 5 I=1,NARDER
         WRITE(1,942)(SUM(I,J),J=1,NORDER)
943          FORMAT(1X,10(E10.3))
942        FORMAT(2X,5(E14.7))
902         FORMAT(2X,8(F11.4))
5       CONTINUE
        WRITE(1,942)(XY(J),J=1,NORDER)
        XX=XX/NN
       WRITE(7,996) XX
 996       FORMAT(1X,E12.5)
     END
```

Figure 7. (continued)

used as input for the matrix inverting program. The summations calculated are those summations over time listed on page 49 in the least squares fitting section of Chapter III.

The program MATINV took File 2 input data and created the inverse of the matricies shown on page 49. The program for matrix inversion was taken from Bevington[80]. This inverse was then multiplied by the left hand side of the equation on page 49 to give the values for $\Delta A_I'$ and $\Delta \lambda_I'$. These values were added to the old values to create new estimates of A_I and λ_I and these values were put in File 1 for use as input to KEEP.

The diagonals of the inverse matrix represented the squares of the errors associated with A_I and λ_I so this matrix was printed out. When the changes in A_I and λ_I were very small compared to the errors convergence was considered complete.

```
      DIMENSION ARRAY(12,12),IK(12),JK(12),AA(12),BB(12)
      ERX=1*10.**7.
     REWIND 1
      REWIND 2
     NORDER=8
      NARDER=NORDER
      NOR2=NORDER/2
      DO 225 KKY=1,NORDER
      READ(1,977) AA(KKY)
977      FORMAT(E14.7)
       WRITE(2,977) AA(KKY)
976      FORMAT(E12.4)
225     CONTINUE
        DO 67 I=1,NORDER
942     FORMAT(2X,5(E14.7))
       READ(1,942)(ARRAY(I,J),J=1,NARDER)
903          FORMAT(2(2X,I3))
905       FORMAT(E12.5)
67       CONTINUE
68        CONTINUE
       DO 690 J=1,NARDER
      NADER=NORDER
907      FORMAT(1X,10(E8.2))
690     CONTINUE
10      DET=1.
11       DO 100 K=1,NORDER
       AMAX=0.
21      DO 30 I=K,NORDER
         DO 30 J=K, NORDER
23        ABAMA=AMAX
       IF(AMAX.LT.0) ABAMA=-1*AMAX
24     ABARR=ARRAY(I,J)
        IF(ARRAY(I,J).LT.0) ABARR=-1*ARRAY(I,J)
      IF(ABAMA.GT.ABARR) GO TO 30
24        AMAX=ARRAY(I,J)
       IK(K)=I
       JK(K)=J
30       CONTINUE
31      IF(AMAX.NE.0) GO TO 41
32     DET=0.
        GO TO 140
41       I=IK(K)
      IF(K.GT.I) GO TO 21
       IF(K.EQ.I) GO TO 51
43     DO 50 J=1,NORDER
      SAVE=ARRAY(K,J)
```

Figure 8. Printout of Program MATINV for Calculating Inverse Matricies

```
      ARRAY(K,J)=ARRAY(I,J)
50       ARRAY(I,J)=-1*SAVE
51        J=JK(K)
       IF(J.LT.K) GO TO 21
       IF(J.EQ.K) GO TO 61
53      DO 60 I=1,NORDER
       SAVE=ARRAY(I,K)
      ARRAY(I,K)=ARRAY(I,J)
60     ARRAY(I,J)=-1*SAVE
61     DO 70 I=1,NORDER
       IF(K.EQ.I) GO TO 70
63      ARRAY(I,K)=-1*ARRAY(I,K)/AMAX
70      CONTINUE
71      DO 80 I=1,NORDER
       DO 80 J=1,NORDER
       IF(K.EQ.I) GO TO 80
74        IF(J.EQ.K) GO TO 80
75    ARRAY(I,J)=ARRAY(I,J)+ARRAY(I,K)*ARRAY(K,J)
80      CONTINUE
81       DO 90 J=1,NORDER
       IF(J.EQ.K) GO TO 90
83      ARRAY(K,J)=ARRAY(K,J)/AMAX
90       CONTINUE
        ARRAY(K,K)=1./AMAX
100     DET=DET*AMAX
101     DO 130 L=1,NORDER
       K=NORDER-L+1
        J=IK(K)
       IF(J.LE.K) GO TO 111
105    DO 110 I=1,NORDER
      SAVE=ARRAY(I,K)
      ARRAY(I,K)=-1*ARRAY(I,J)
110      ARRAY(I,J)=SAVE
111     I=JK(K)
      IF(I.LE.K) GO TO 130
113     DO 120 J=1,NORDER
       SAVE=ARRAY(K,J)
       ARRAY(K,J)=-1*ARRAY(I,J)
120      ARRAY(I,J)=SAVE
130      CONTINUE
140       CONTINUE
       DO 666 I=1,NORDER
          DO 666 J=1,NORDER
```

Figure 8. (continued)

```
666       CONTINUE
        DO 69 J=1,NARDER
       WRITE(7,988) (ARRAY(I,J),I=1,NORDER)
988         FORMAT(1X,1P6(E11.4))
900        FORMAT(1X,6(E10.3))
69        CONTINUE
         WRITE(7,987) DET
987       FORMAT(3X,'DET=',E12.5)
          READ(1,942)(AA(J),J=1,NORDER)
902           FORMAT(2X,8(E11.4))
       DO 223 KKP=1,NORDER
       DO 223 KKQ=1,NORDER
       BB(KKP)=BB(KKP)+AA(KKQ)*ARRAY(KKQ,KKP)
223       CONTINUE
       DO 233 IKK=1,NOR2
       IKJ=IKK+NOR2
        WRITE(2,908) BB(IKK)
       WRITE(2,908) BB(IKJ)
908         FORMAT(E14.7)
233         CONTINUE
       END
```

Figure 8. (continued)

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VITA

Raymond Waldo was born on July 14, 1950 in Glendale, California. He received his Bachelor of Science Degree in Physics from the California Institute of Technology in June of 1972.

Upon graduation Mr. Waldo was employed at the Jet Propulsion Laboratory as a computer analyst. He was responsible for code development and statistical analysis of health statistics under Dr. Peter Gottlieb in this project for the County of Los Angeles.

From 1973 to 1975 Mr. Waldo was a Peace Corps Volunteer. He was employed as a Physics and Mathematics teacher in the college preparation program of Sandy Point High School, St. Kitts, West Indies.

In September of 1975 Mr. Waldo began graduate studies at the Georgia Institute of Technology under an ERDA (now DOE) Traineeship. He received a Master of Science Degree in Nuclear Engineering in September of 1976. During this period Mr. Waldo became trained and licensed as a reactor operator for the Georgia Tech Research Reactor under Dr. Monte Davis. He subsequently participated in the core design and analysis of a Gaseous Core Actinide Burning Reactor under Dr. Joseph Clement.

In January of 1977 Mr. Waldo became Shift Supervisor of the Livermore Pool Type Reactor at the Lawrence Livermore Laboratory. In this position he was responsible for sample approval, core analysis, fuel management, and shift scheduling. Mr. Waldo became Acting

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