Chapter 11

DELAYED NEUTRON SUMMATION CALCULATIONS

Introduction

The production of the JEF-2.2 fission product yield evaluated files was previously described in Part I, Chapter 3. The confidence which can be held in the validity of this, or any other evaluated nuclear data file, depends upon the tests that are applied to the data. These tests can be of two types. The first type are tests of internal consistency which are based upon the intrinsic physics or the empirical data on which the evaluation is based. The second type, external tests, are where the data are used to model a phenomenon based upon a real situation and the results of these calculations are compared with experimental measurements. One type of calculation was chosen to test the evaluated fission yield data that only requires the decay data to be known. This is the calculation of total delayed neutron emission. In this work very low values of neutron flux were assumed in the modelling so that cross-section effects could be ignored.

It should be noted that these delayed neutron calculations are a test of the fission product yields and are not produced as a recommendation for delayed neutron data.

Delayed neutron calculations

If we consider delayed neutron emission from fission products, the governing phenomenon is the decay of a fission product that leaves a daughter nucleus with sufficient excitation energy to throw off a neutron. For nuclides where this occurs the fraction of decays that produce a neutron is called the P_n value; these nuclides are short-lived and on the neutron rich side of the line of stability.

The total number of delayed neutrons per fission, $\overline{\nu}_d$, and the time dependence of the delayed neutron emission rate are important parameters for reactor design and safety studies, as they determine the kinetic response and behaviour of reactors. There exist three ways of determining $\overline{\nu}_d$:

- Experimentally from integral measurements, e.g. Keepin [1].
- From summation calculations, e.g. Liaw *et al.* [2] using cumulative fission yields and P_n branching ratios.
- By a more empirical method, proposed by Pai *et al.* [3] and modified by Tuttle [4], based upon systematics of the delayed neutron production with mass and charge of the fissioning compound nucleus.

The time dependence of delayed neutron emission can be determined by experiment or by summation calculations using the branching ratios, half-lives and inventories of the fission products following an irradiation, e.g. the work of Brady and England [5]. The proposed use of reprocessed fuel containing significant quantities of higher actinides has led to requests for the values of $\bar{\nu}_d$ for these nuclides so that their effects on the kinetic response of reactors can be estimated for safety studies. As experiments with these materials are often difficult due to the lack of reasonably sized samples and thus reported experiments are rare in the literature, the summation method may be the most reliable way for these $\bar{\nu}_d$'s to be estimated if it can be shown to be more accurate than the empirical extrapolation method of Pai [3] and Tuttle [4]. However the uncertainties in the yields and branching ratios of the delayed neutron emitters must be reviewed in order to decide whether the summation method is significantly accurate for practical use.

The delayed neutron emitters exist on the extremely neutron rich side of the independent fission yield distribution, where few fission product yield measurements have been made except for the more common actinides such as ^{235}U and ^{239}Pu. Thus the models used to predict the charge distribution of the fission yields will have a significant effect on $\bar{\nu}_d$. The different chain yield distributions for the fission of the higher actinides also mean that some precursors, relatively unimportant for ^{235}U fission, become much more significant. Especially important is the movement of the light mass peak towards higher mass as the mass of the fissioning nuclide increases. However, measurements of the P_n values have been based mainly upon ^{235}U fission so theoretical estimates of the P_n branching ratios become much more important when considering the higher actinides.

The neutron emission is a result of β^- decay producing a daughter which has sufficient energy to throw off a neutron. The probability of a nuclide emitting a neutron as a result of a β^- decay is referred to as the P_n. The fission products present determine the delayed neutron emission rate, n_{emit}, from the activity of these precursors:

$$n_{emit}(t) = \sum P_{ni} \lambda_i N_i(t)$$

where P_{ni} is the P_n for nuclide i, λ_i is the decay constant of i, and $N_i(t)$ is the number of i present at time t after the irradiation. N_i is determined by the initial fuel composition and the irradiation this receives. Therefore to generate the delayed neutron emission rate the irradiation must be specified and a calculation made of the inventory at each time t. However, the total delayed neutron emission per fission, $\bar{\nu}_d$, can be calculated by integrating over all time for a single fission. Thus:

$$\bar{\nu}_d = \sum_i P_{ni} R_i = \sum_i P_{ni} c_i$$

The total decays of nuclide i per fission, R_i, is equal to the cumulative fission product yield of i. Thus, for a pure sample of an actinide, if the cumulative yields, c_i are known the $\bar{\nu}_d$ can be calculated. Alternatively, if we consider a very long irradiation where all the fission products have reached equilibrium then the activity of each is the cumulative yield, thus producing the same formula. This equivalence is due to the definition of the cumulative yield.

The uncertainty in the calculated $\bar{\nu}_d$ can be estimated from above, by partial differentiation and assuming c and P_n are independent, as:

$$\sigma_{\bar{\nu}_d}^2 = \sum_i P_{ni}^2 \sigma_{c_i}^2 + \sum_i \sigma_{P_{ni}}^2 c_i^2$$

Summation calculations of $\bar{\nu}_d$

From the equations above values of $\bar{\nu}_d$ with uncertainties are easily calculated from the JEF-2.2 fission product yield and decay data files. This decay data was used to generate the cumulative yields from the independent yields. The values given in Table 11.1 are quoted per 100 fissions.

Table 11.1. Calculation of $\bar{\nu}_d$ using JEF2.2 decay data and fission yields

Nuclide	Neutron energy	Calculated	Measured*	Calculated/ Measured
Thorium-232	Fast	6.04559 ±4.55E-01	5.47±0.12 [4a]	1.105±0.08
Thorium-232	14 MeV	2.93874 ±2.52E-01	2.85±0.13 [4]	1.031±0.10
Uranium-233	Thermal	0.87778 ±8.45E-02	0.664±0.018 [4a]	1.322±0.10
Uranium-233	Fast	0.95255 ±1.15E-01	0.729±0.019 [4a]	1.307±0.12
Uranium-233	14 MeV	0.34425 ±6.88E-02	0.422±0.025 [4]	0.816±0.21
Uranium-234	Fast	1.19717 ±1.94E-01	1.06±0.12 [4a]	1.124±0.20
Uranium-235	Thermal	1.70768 ±1.17E-01	1.654±0.042 [4a]	1.032±0.20
Uranium-235	Fast	1.90981 ±2.01E-01	1.714±0.022 [4a]	1.166±0.11
Uranium-235	14 MeV	0.78986 ±8.16E-02	0.927±0.029 [4]	0.852±0.11
Uranium-236	Fast	2.32978 ±2.05E-01	2.31±0.26 [4a]	1.009±0.14
Uranium-238	Fast	4.26631 ±2.02E-01	4.510±0.061 [4a]	0.946±0.09
Uranium-238	14 MeV	2.39520 ±2.06E-01	2.73±0.08 [4]	0.877±0.16
Neptunium-237	Thermal	1.23220 ±1.55E-01	1.07±0.10 [8]	1.152±0.07
Neptunium-237	Fast	1.23409 ±8.88E-02	1.22±0.03 [7]	1.011±0.16
Plutonium-238	Thermal	1.47197 ±1.76E-01	0.456±0.051 [4a]	3.228±0.20
Plutonium-238	Fast	0.46987 ±7.49E-02	0.456±0.051 [4a]	1.030±0.19
Plutonium-239	Thermal	0.61740 ±5.61E-02	0.624±0.024 [4a]	0.989±0.10
Plutonium-239	Fast	0.69008 ±7.93E-02	0.664±0.013 [4a]	1.039±0.19
Plutonium-240	Fast	0.93974 ±1.12E-01	0.96±0.11 [4a]	0.979±0.17
Plutonium-241	Thermal	1.33637 ±1.35E-01	1.56±0.16 [4a]	0.857±0.14
Plutonium-241	Fast	1.45238 ±9.63E-02	1.63±0.16 [4a]	0.891±0.12
Plutonium-242	Fast	1.92750 ±1.39E-01	2.28±0.25 [4a]	0.845±0.13
Americium-241	Thermal	0.40910 ±6.62E-02	0.44±0.05 [8]	0.930±0.20
Americium-241	Fast	0.41147 ±7.70E-02	0.394±0.024 [7]	1.044±0.20
Americium-242m	Thermal	0.64864 ±8.38E-02	0.69±0.05 [8]	0.940±0.15
Curium-245	Thermal	0.50695 ±8.86E-02	0.59±0.04 [8]	0.859±0.19
Californium-252	Spontaneous	0.74153 ±1.64E-01	0.86±0.10 [6]	0.862±0.25

The evaluated values are based upon experiment and taken from the following sources: the evaluations of Tuttle [4], Tuttle [4a] and Manero [6], and where these evaluations do not contain data the experimental values reported by Benedetti [7] and Waldo [8] were used.

As can be seen from Table 11.1 there is a tendency to over-predict $\overline{\nu}_d$ for masses below 238 and under-predict those above. The evaluated uncertainties are given as one standard deviation. For the main systems a recent study [9] based upon the currently available experimental data considered the previous evaluated uncertainties to be low, and suggested larger values which should be associated with the results. The uncertainties of the other experimental values measured relative to these are thus also brought into question.

It is interesting to note that the system with the poorest fit to the Z_p model (thermal neutron fission of ^{233}U) also has the worst C/E values.

It must be remembered that these calculations are very sensitive to short lived nuclides far from stability and the P_n values used. Thus study of the sensitivity of these calculations to the Z_p parameters and different P_n data sets will give more information on the properties of the calculations.

Sensitivity of $\overline{\nu}_d$ to Z_p parameters

The sensitivity of $\overline{\nu}_d$ to the Z_p parameters was studied by considering the fractional change in $\overline{\nu}_d$ following a small change in each Z_p parameter used to generate a set of unadjusted yields. These yield sets were not adjusted to fit physical constraints, as this would alter the independent yields used in the calculation. This study was made with the UKFY2 fission yields and its corresponding decay data file (Preliminary JEF-2, 1991). Each of the eight parameters x was varied in turn by + and -1%, and the sensitivity $S(x)$ to x found from:

$$S(x) = 100\frac{\overline{\nu}_d(x+1\%) - \overline{\nu}_d(x-1\%)}{2\overline{\nu}_d(x)}$$

The results of this calculation are shown in Table 11.2. This shows the 1% sensitivities to the Z_p parameters for the thermal and fast neutron fission of ^{235}U.

Table 11.2. Sensitivity of $\overline{\nu}_d$ to input Z_p model parameters

System	$\Delta Z(A' = 50)$	$\frac{d\Delta Z}{dA'}$	$\overline{\sigma}_z$	$\overline{\sigma}_{50}$	$\overline{F}_z$	$\overline{F}_n$	$\Delta A'_z$	ΔZ_{max}
^{235}U T	-0.44	-0.046	1.2	0.0010	-0.99	0.10	-0.0012	-0.0010
^{235}U F	-0.47	-0.10	1.0	0.00045	-0.95	0.037	-0.000021	-0.000023

Variations of + and -10% were also made, but the calculated sensitivities were not found to change significantly. This suggests the sensitivity to the parameters are not rapidly changing.

These results shows that $\overline{\sigma}_z$, $\overline{F}_z$ and $\Delta Z(A' = 50)$ are the most important Z_p parameters for the calculation of $\overline{\nu}_d$. The two parameters $\overline{\sigma}_z$ and $\Delta Z(A' = 50)$ largely determine the shape and positions of the Gaussian fractional independent fission yield distributions, and hence the yields of the neutron-rich precursors. The dependence on $\overline{F}_z$ reflects the preponderance of odd-Z delayed neutron precursors.

A detailed understanding of how these three Z_p parameters change between different systems would thus improve the results of summation calculations.

Sensitivity of $\bar{\nu}_d$ to different P_n sets

To study the effect of different P_n datasets upon $\bar{\nu}_d$, calculations of the equations above were carried out using the UKFY2 cumulative yields with different P_n datasets. It should be noted that if the different P_n values had been used in the production of the UKFY2 the file would alter the predicted cumulative yields. Thus the $\bar{\nu}_d$ would be altered. However, previous work [9,10] had showed that for most mass chains these differences in chain yields would be small. This effect was, therefore, ignored for the purpose of this study.

The results for the thermal neutron fission of ^{235}U and ^{239}Pu, and the fast neutron fission of ^{235}U and ^{238}U are shown in Table 11.3. The number of delayed neutron emitters in each file are shown in the table with a flag to show whether the set includes experimental (E), model prediction (M) or both (EM). Also the results of the two later calculations with the JEF2.2 decay data are shown for comparison.

Table 11.3. $\bar{\nu}_d$ calculated using different P_n datasets

Fission yield file	Decay data file	Number of P_n	^{235}U (thermal)	^{235}U (fast)	^{238}U (fast)	^{239}Pu (thermal)
UKFY2(1990)	JEF-2 (1991)	94 EM	1.6354	1.8492	3.9039	0.5884
UKFY2(1990)	Lund(1986)	83 E	1.4455	1.5963	3.5420	0.5050
UKFY2(1990)	Mann(1986)	88 E	1.5665	1.7629	3.6896	0.5970
UKFY2(1990)	Brady (1988)	271 EM	1.6995	1.9092	4.0218	0.6131
UKFY2(1990)	Klapdor(1989)	209 M	1.2572	1.4044	3.2950	0.4697
UKFY2(1990)	JEF-2 (May 1991) + Klapdor (1989)	251 EM	1.6447	1.8541	4.0491	0.5895
JEF-2.2(1993)	JEF-2.2 (1993)	165EM	1.7071	1.9092	4.2611	0.6171

E – Experimental data, M – Modelled data, EM – Combination of experimental and modelled data

This work shows that the majority of the delayed neutron emission comes from precursors whose P_n values have been measured. For the thermal fission of ^{235}U only around 6% of the total comes from modelled P_n values. Interestingly, using all modelled P_n values decreases the value. This may indicated that the modelled P_n values are unrealistically small.

The Keepin six-group model

As described above the neutron emission rate following a neutron irradiation can be calculated from an inventory calculation using the equations above. However, in practice, reactor kinetics codes consider a small set of “lumped fission products” with a set of a representative decay constants and yields. This approach was pioneered by Keepin [1], who found that a set of six “lumped fission products” gave a good approximation to measurements. The delayed neutron activity following a single fission pulse of one "average" fission was thus approximated by Keepin [1] by a six-group representation:

$$n_{emit}(t) = \overline{\nu}_d \sum_{k=1}^{6} \alpha_k \lambda_k e^{-\lambda_k t}$$

and similarly for a long constant irradiation, producing one fission per second, as:

$$n_{emit}(t) = \overline{\nu}_d \sum_{k=1}^{6} \alpha_k e^{-\lambda_k t}$$

where t is the time after the irradiation, α_k are the normalised group strengths and the λ_k are the decay constants for the six delayed neutron emitting groups. For these conditions to be applicable the pulse must be too short for any precursor to decay significantly during the irradiation. Similarly the long irradiation condition only applies if all precursors have reached equilibrium before the end of the irradiation.

It is an interesting result, which also applies to decay heat calculations, that at zero time after the long irradiation the neutron emission is equal to the integral of neutron emission following a single "average" fission pulse over all time after the irradiation.

JEF/DOC-830 describes calculations of neutron emission made using UKFY2. The decay data used for this work was based upon a preliminary version of JEF-2 (1991), with the P_n values extended with the work of Lund [11] and Klapdor [12]. The half-lives were also extended using the Japanese Chart of the Nuclides [13]. To generate the Keepin six-group constants using the UKFY2 data it was first necessary to use the above equations and the inventory code FISPIN to generate the n_{emit}. Both a single fission pulse (10^6 fission/s for 10^{-6} s) and a "long" irradiation (1 fission/s for 10^{13} s) were modelled. The cooling time steps after the irradiation ranged from zero to 500 seconds. Two hundred and four (204) time steps were chosen to accurately reproduce the rapidly changing curves. Keepin's six-group model was fitted to the pulse and infinite irradiation data simultaneously (i.e. 408 data points) using the Levenberg-Marquardt method [14] as applied by Press, *et al.* [15]. The $\overline{\nu}_d$ values used were taken from the zero time long irradiation results. The full results of these calculations are presented in JEF/DOC-830.

As well as fitting the twelve α_k and λ_k parameters, an attempt was made to fit the six α_k values with a constant set of λ_k to allow simplification in reactor calculations where more than one of the nuclides are present. In the first case the set of average λ_k values reported by Keepin [1] (Table 4-9, page 91) were used. In a second case the results were obtained using the set of λ_k values calculated for the thermal neutron fission of ^{235}U. The effects of these approximations were then studied. The twelve parameter fits give the best results. These seldom vary by more than 1% from the calculation. However the two approximations (using the fixed λ_k sets) show considerably higher variation from the FISPIN calculations. These results are described in detail in JEF/DOC-830.

Figures 11.1-11.6 are an example of the results obtained from the calculations. They show the delayed neutron emission rates for the thermal neutron fission of ^{235}U and ^{239}Pu, and the fast neutron fission of ^{238}U. Both the pulse and long irradiation results are shown. To show this work in context the figures plot the results of the FISPIN calculation, the six-group parameter calculations and the six-group parameters published by other workers relative to the FISPIN calculation.

Figure 11.1. The delayed neutron emission rate following a pulse and long irradiation for the thermal neutron fission of ^{235}U

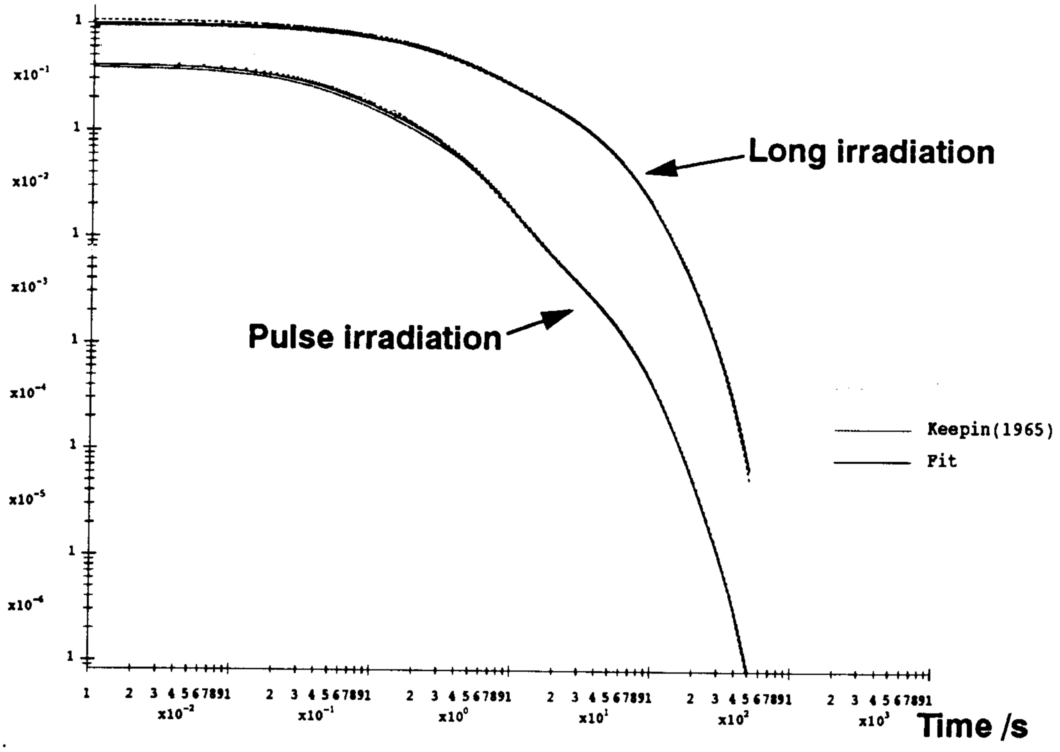


Figure 11.2. Percentage difference between the long irradiation FISPIN calculations and six-group parameters for the thermal neutron fission of ^{235}U

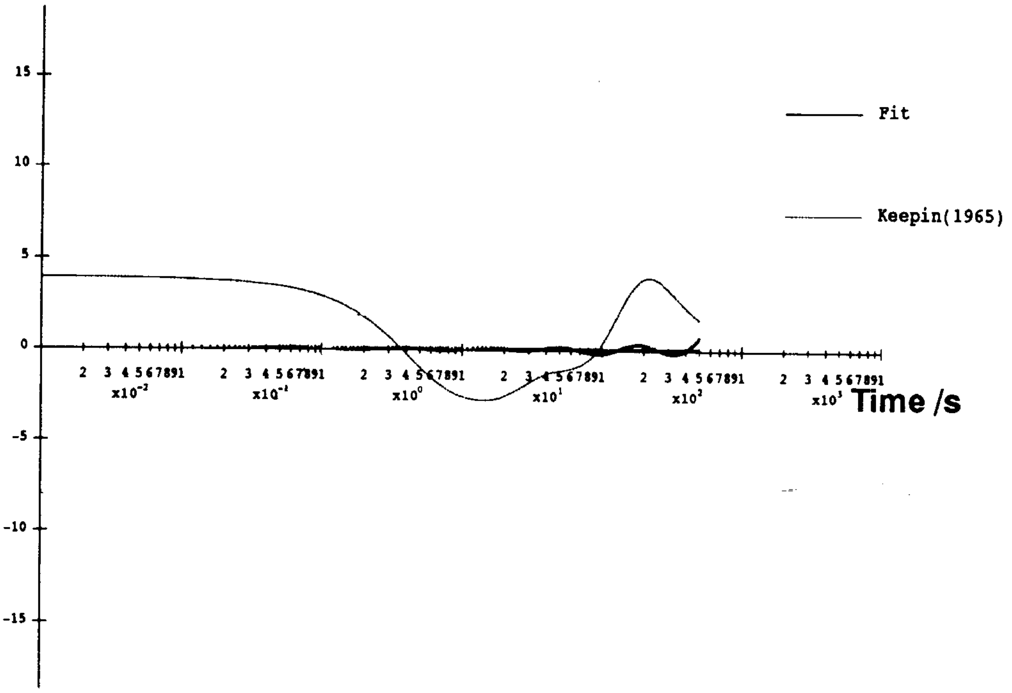


Figure 11.3.The delayed neutron emission rate following a pulse and long irradiation for the thermal neutron fission of ^{239}Pu

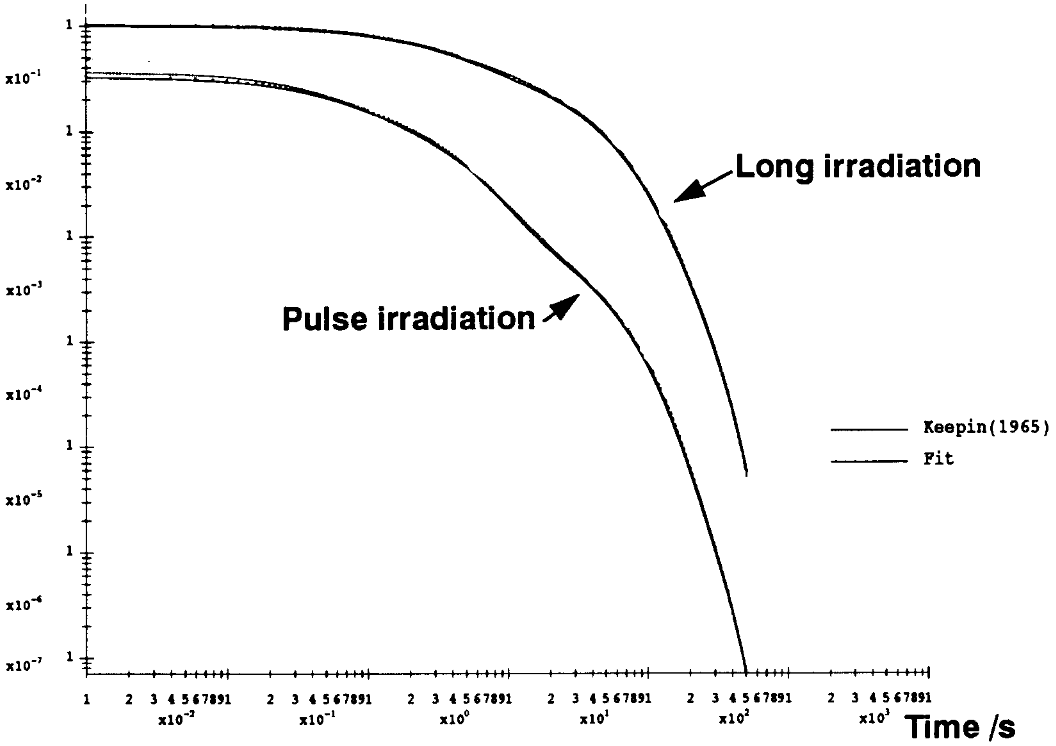


Figure 11.4. Percentage difference between the long irradiation FISPIN calculations and six-group parameters for the thermal neutron fission of ^{239}Pu

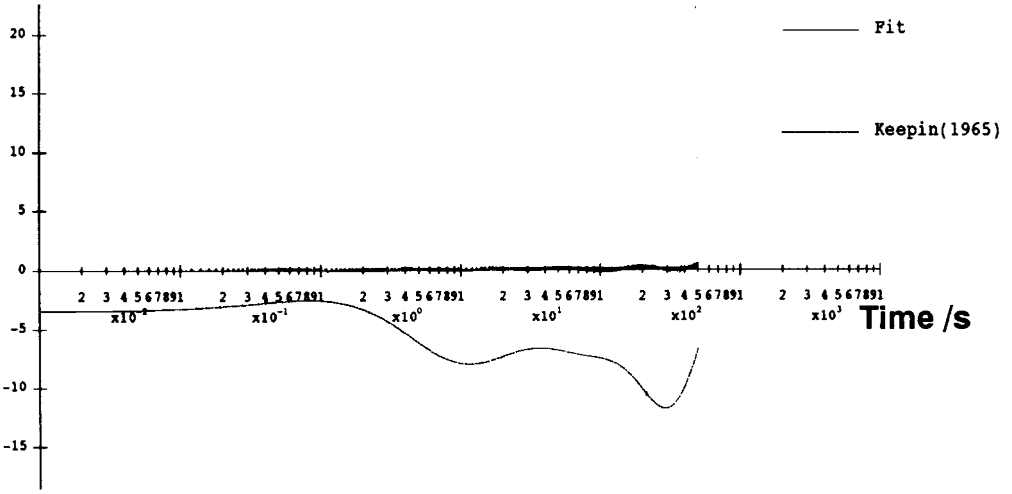


Figure 11.5. The delayed neutron emission rate following a pulse and long irradiation for the fast neutron fission of ^{238}U

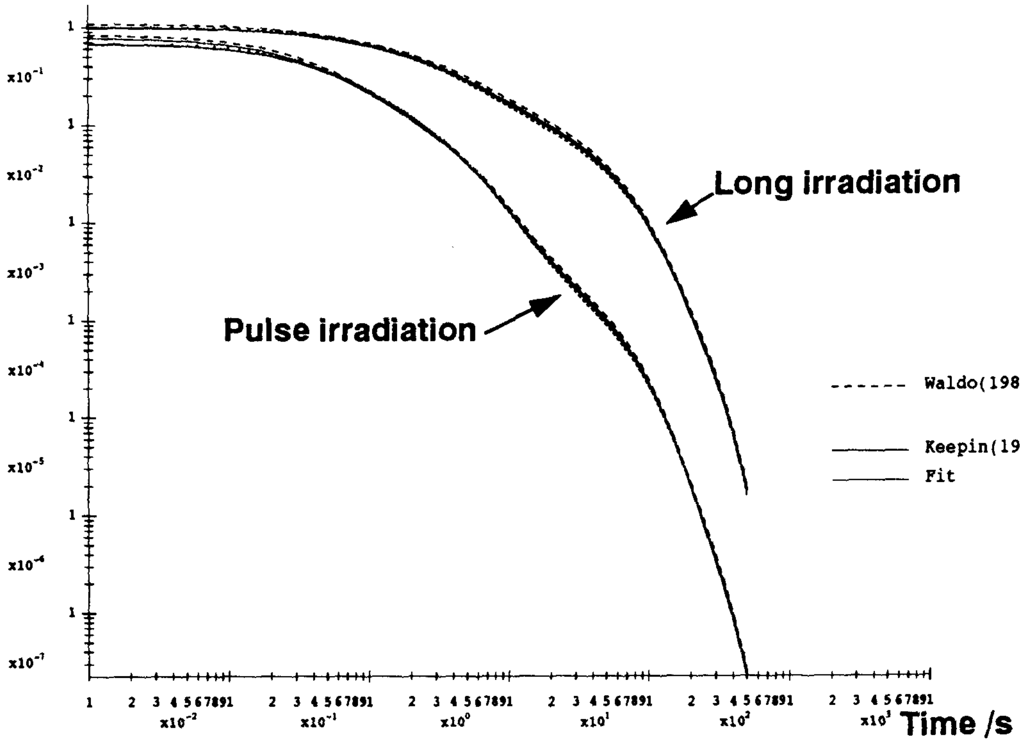
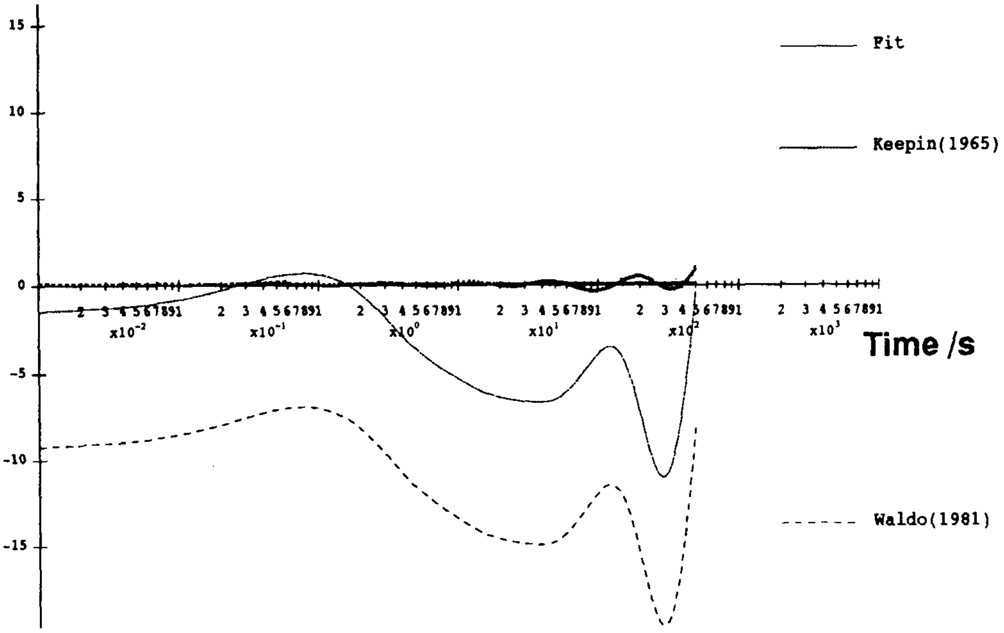


Figure 11.6. Percentage difference between the long irradiation FISPIN calculations and six-group parameters for the fast neutron fission of ^{238}U



Other experimentalists who have published experimental determined six-group parameters include Keepin [1] and Waldo [8].

The differences from the "long" irradiation case FISPIN results are shown for each of the six-group parameter sets in Figure 11.2.

When comparing the results it should be borne in mind that experiments have difficulty in measuring the neutron emission at very long times after irradiation due to the fall off of the delayed neutron emission to below the experimental noise. Also, the short-lived groups cannot be measured directly as moderated neutrons from the irradiation will still be present. One common technique to measure the short-lived groups is to use a pulsed irradiation. The long-lived groups and the moderated neutrons then become a background that can be subtracted. However, at very short times, this background will swamp the neutron emission being measured. Thus the short and long measurements will not be as accurate as those at the middle of the range. Also, the accuracy of the six-group model will be less than that for $\overline{\nu}_d$.

The six-group half-lives vary from ~0.2 to 60 seconds. Thus if any neutron emission occurs outside of this time window it cannot be accurately represented by the model.

The majority of the differences in the figures can be attributed to the different values of $\overline{\nu}_d$ used in the calculations. This can be seen on the figures showing the differences, because at zero time after the "long" irradiation the neutron emission rate will equal this value. Thus the differences at zero time are directly related to the values used.

In the region up to 200 seconds the remaining differences are of the same order as the uncertainty on $\overline{\nu}_d$. For times greater than 200 seconds the neutron emission has dropped to such a level that the differences have no practical significance.

Conclusions

Above we have shown reasonable agreement between summation calculations and experimental measurements. This suggests that the JEF-2.2 fission product yields and decay data give a good approximation to physical reality. However, it must be stressed that the above delayed neutron calculations were carried out to test the JEF-2.2 yield and decay data. The calculated delayed neutron parameters (in JEF/DOC-830) are therefore not recommended for applications as no comprehensive analysis has been made of all the available delayed neutron measurements to validate this work.

Since the completion of these calculations, earlier this decade, there has been much work carried out as part of the WPEC Subgroup 6, which will soon be published. This includes a compilation of all the published delayed neutron data parameters. Also included is interesting new work based upon fitting the delayed neutron emission to a larger number of delayed neutron groups, but where a group is dominated by one precursor the time constant is assumed to be the decay constant of this nuclide. We direct the interested reader to the Subgroup 6 report and references therein.

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