IMPORTANCE OF DELAYED NEUTRONS IN NUCLEAR RESEARCH

by

S. Das

Theoretical Physics Division

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Abstract: The report presents a comprehensive review of the uses of \( \beta^-n \) delayed neutrons in nuclear research with special emphasis on energy spectra and reactor applications. The review covers the following aspects: (1) delayed neutron in reactor analysis, (2) absolute delayed neutron yield, (3) delayed neutron decay constants, (4) delayed neutron energy spectra and their importance in high accuracy criticality calculations and precise evaluation of reactor kinetics characteristics, particularly fast breeders, (5) a chronological account of the developments in the measurement of delayed neutron energy spectra, both aggregate (composite) and from individual fission product isotopes during the last fifty years or so, (6) major spectrometry techniques for measuring delayed neutron energy spectra, (7) spectral analysis, (8) calculations of delayed neutron energy spectra from precursor data, and (9) sensitivity analysis of fast reactor dynamic behaviour to delayed neutron energy spectra. Finally, the report recommends a number of areas for future research work on delayed neutrons.

Keywords/Descriptors: DELAYED NEUTRONS; REVIEWS; BETA DELAYED NEUTRONS; REACTOR PHYSICS; REACTOR KINETICS; FISSION YIELD; NEUTRON EMISSION; ENERGY SPECTRA; NEUTRON SPECTRA; DECAY; NEUTRON SPECTROSCOPY; DELAYED NEUTRON PRECURSORS; SENSITIVITY ANALYSIS; FBR TYPE REACTORS; EXPERIMENTAL DATA
ABSTRACT

This report is a comprehensive review of the uses of $\beta^-$-delayed fission neutron information in nuclear research with special emphasis on energy spectra and the reactor applications. The report starts with a very brief introduction followed by a discussion of the applications of delayed neutron properties in nuclear structure and astrophysical studies. §3 gives the delayed neutron requirements for reactor physics purposes and points out the difference between the physical and the mathematical representations of the delayed neutron data. In §4 and §5 respectively, there are discussions of the total delayed neutron yield and the decay constants. §6 highlights the importance of an exact knowledge of delayed neutron energy spectra in high accuracy criticality calculation and in precise evaluations of reactor kinetics characteristics, particularly the fast breeders. The chapter also gives a skeletal description of the principal methods that are commonly used for determining the delayed energy spectra. §7 gives a chronologic account of the developments in the measurement of delayed neutron energy spectra, both aggregate (composite) and from individual fission product isotopes during the last fifty years or so, and compares the spectra, wherever possible, with the ENDF/B evaluations. A comparison is made of the energy spectra of a few well-known precursors measured at different laboratories. There are discussions on the major spectrometry techniques that are employed in the measurements of delayed neutron energy spectra as well as on the methods of performing spectral analysis. This covers the response function, the efficiency and the sensitivity of the spectrometers, their merits and demerits and their applicability. Calculations of delayed neutron energy spectra from precursor data, and decomposition of composite spectra into six-group delayed energy spectra using the summation and/or fitting procedures are described in §8. §9 reviews the work of several authors on the sensitivity of the kinetic response of fast reactors to delayed neutron energy spectra. Both direct and adjoint methods are discussed. §10 gives a summary. The report concludes in §11 with a number of recommendations for future investigations.
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§1. INTRODUCTION

The fundamental role of $\beta^-,n$ delayed fission neutrons in the operation and time-dependent behaviour of nuclear reactors has been known since the early days of research in the field of fission chain reactors and is now a matter of practical experience in hundreds of nuclear installations around the world. These delayed neutrons, thirty per cent of which are emitted in less than 1 s following fission, constitute less than one per cent of the total neutron emission. Though fission is the most common way of producing delayed neutrons, production of neutron rich nuclides unstable to neutron emission is not uniquely related to the fission process. There are also other processes in which delayed neutrons are produced such as: delayed neutron emission subsequent to beta decay of the neutron-rich Na29-Na31 isotopes, the delayed neutron emission from the isotopes B15, C18, N19, N20, Al-34, Al-35, P39. In Na29($\beta^-$)Mg29 decay scheme, five excited states of Mg29 are populated after $\beta$-delayed one-neutron emission. For Na30, 1n,2n delayed emissions have been reported [1]. The beta-delayed four neutron decay mode of B17 was reported for the first time by Dufour et al [2].

Delayed neutrons are so called not because the neutrons are delayed; the delay is because there are intermediate reactions before the neutrons are emitted. For example,

$$\begin{align*}
^{235}U & \xrightarrow{\text{fission}} ^{87}\text{Br} & \xrightarrow{\beta^-} ^{87}\text{Kr} & \xrightarrow{\text{d.n.}} ^{86}\text{Kr} \\
& \text{(Precursor \ Nuclide)} & \text{(Daughter)} & \text{(Grand \ Daughter)}
\end{align*}$$

The mechanism of delayed emission of neutrons in fission is well understood in principle [3,4]. The $\beta$-decay of a nuclide $(Z,N)$ with high decay energy, $Q_\beta$ can populate excited states lying above the neutron binding energy, $B$ of the daughter nuclide $(Z+1,N-1)$. These states, from which neutron decay into the nucleus $(Z+1,N-2)$ becomes energetically possible, may then deexcite through the emission of a neutron. The neutrons are promptly emitted but the overall time scale is governed by the half-life of the preceding $\beta$-decay of the nuclide $(Z,N)$, usually called the delayed neutron precursor (see figure A). The process of delayed neutron emission is most likely to occur in nuclides having a few neutrons in excess of a closed neutron shell because of the unusually low neutron binding energy in such nuclides.

§2. SIGNIFICANCE OF DELAYED NEUTRON EMISSION

Since the discovery of the delayed neutron by Roberts et al in 1939 [5], the majority of the interest in delayed neutron has evolved through the various aspects of nuclear technology like the design and operation of fission reactors, the measurement and interpretation of reactivity effects, the dynamics and safety analysis of nuclear reactors, detection of fuel element failures in fuel subassemblies of power reactors, fissile material assay of fuel elements, neutron flux monitoring by fissionable materials, determination of uranium content in urine from nuclear fuel
fabrication plant workers by delayed neutron counting [6], etc. While determining the worth of reactivity in a nuclear reactor from an analysis of the time behaviour of the delayed neutrons, the beta-decay properties (group yields and decay constants) of delayed neutrons which affect the length of reactor period are required [7]. The amount of decay heat after reactor shut-down is governed by the decay properties of beta-delayed neutrons [8]. Chemical properties of individual delayed neutron precursors are important in reactors whose precursors are transported out of the core region resulting in a corresponding loss of reactor control [9]. Examples of such systems are: reactors operating at very high temperatures, reactors with continuous fuel reprocessing and reactors with circulating fuel. However, interest in delayed neutron information goes beyond nuclear power as for example in nuclear structure and astrophysics applications.

2.1 Nuclear Physics Research

Studies of delayed neutron emission give important information on nuclear structure such as the matrix elements in beta-decay, level densities at medium excitation energies, the competition between neutron and gamma emission and through the decay energies involved on the shape of the nuclear energy surface. Delayed neutrons serve as a tool for studying the fission process yielding information on the fission fragment mass and charge distributions of various nuclei for different excitation energies. Knowing $P_\beta$ (delayed neutron emission probability) values and the yields of precursors, one can obtain an idea of the competition among the different types of fission. Improvements in theoretical nuclear models for estimating beta-decay properties using beta strength functions, $S_\beta$, etc. Klapdor [10-12] has shown that the structure in $S_\beta = \int \phi_j(E) B_j(E)/D \ MeV^{-1}$
sec\(^{-1}\) (with \(\varphi\) being the level density in the daughter nucleus, \(B\) the reduced transition probability, and \(D = 6250\ \text{sec}\) are decisive for the \(\beta\)-decay half-lives, the spectra of \(\beta\)-delayed particles and \(\beta\)-delayed fission rates and are crucial for obtaining reliable results. It is important to point out that the shape of the beta strength function, \(S\), is of great importance for various processes and problems [12]. These range, in nuclear physics, from the problem of determining fission barriers from beta-delayed fission, interpretation of reactor neutrino oscillation experiments, the production of transuranium elements by thermonuclear explosions to such technical problems as the control of Pu-fuelled fast breeder reactors and optimizing the emergency cooling system of reactors. In astrophysics, the shape of \(S\) governs the electron capture in degenerate cores of stars, the \(\beta\) neutrino cooling, the speed of gravitational collapse in the initial phase, the processes \((r\text{- or } n\text{-process})\) responsible for the production of heavy elements in the universe and the unsolved problem of identifying the astrophysical site of such processes.

Determination of \(\beta\)-decay properties by high resolution delayed neutron spectroscopy provides new insight into nuclear structure of far-unstable nuclei [13,14]. This is because for such nuclei, \(\beta\)-decay energies (\(Q\)) increase from less than one MeV to over 10 MeV (corresponding half-lives decrease down to milliseconds) and neutron binding energies (\(B\)) decrease with distance from \(\beta\)-stability as a result of which delayed neutron emission becomes the more important process. The overall shape of a delayed neutron spectrum is given by

\[
I_n(E_n) = \langle \frac{i}{i\gamma} \rangle \cdot \frac{\Gamma_n}{\Gamma_n + \Gamma_\gamma} E_n
\]

where, the first factor defines the \(\beta\)-decay intensity to neutron unbound levels and contains information about nuclear structure. The second factor gives the probability \(\Gamma\) of delayed neutron emission from these states according to their decay widths. For far-unstable nuclei having high level density \(\varphi(E)\), average \(\beta\)-decay properties are defined in terms of the \(\beta\)-strength function, \(S\), as:

\[
\langle I_\beta \rangle_{E_n} = \langle f.S_\beta(E) \rangle
\]

where, the average is taken over many narrowly spaced states instead of the individual levels and the Fermi function \(f (\sim 10^7)\) includes the kinematic and the phase-space effects. In delayed neutron emission, the influence of \(\gamma\) on the spectral shape of delayed neutrons is small since low-\(\gamma\) delayed neutrons are mostly emitted and the coulomb barrier plays an unimportant role. Consequently, the factor \(\Gamma\) in the equation approaches unity even at neutron energies of 50 to 100 keV. This makes it possible to extract from the spectra reliable information on high energy \(\beta\)-decay properties. For a detailed discussion of the average energies and beta strength function, readers may refer to [15,16]. Delayed neutron spectroscopy including \(n\gamma\)-coincidence measurements allows to extend our knowledge of excitation spectra of far-unstable nuclei.
beyond 10 MeV such as the verification of discrete nuclear structures beyond gross statistical properties, identification of nuclear shape changes via corresponding variations in the structure of $S_n$, determination of 'quenching' of the Gamow-Teller strength in exotic nuclei [16,17], determination of $Q_{\beta n}$, $B_n$ from the known $\beta$-decay energies and the measured $(Q_{\beta n} - B_n)$ value. Delayed neutron spectroscopy offers a tool for studying individual levels and for deriving reliable $Q(E)$ for far-unstable nuclei [18]. Knowledge of individual levels is important for interpreting the peak structure in delayed neutron spectra. This was tested in the case of the compound nucleus Kr87 [19] whose neutron-unbound levels are accessible to both $\beta$-decay and neutron capture. Besides Kr87, the only other nucleus that is a daughter of a delayed neutron precursor and that is also accessible with neutrons on a stable target is Xe137. However, the neutron emitting levels in Xe137 have relatively high spins $(5/2^+, 7/2^+, 9/2^+)$ and therefore, the overlap between $\beta$ decay and resonance neutron spectroscopy in Xe137 is not expected to be as clean or dramatic as in Kr87. In the delayed neutron spectrum of Br87, in the first 250 keV, fourteen resonances out of the seventeen observed p-wave capture resonances have been identified. With improved resolution of the techniques of neutron spectra measurements, it is possible to determine the natural line widths for individual states and to obtain better evaluation of level density. This way a preliminary value of $(300\pm40)$ keV was deduced for the natural width of the 14-keV resonance in Rb-95 by D.D. Clark, R.D. McElroy, T.-R. Yeh [15,p.449-454].

2.2 Astrophysical Applications

The rapid-neutron-capture process (r-process) of nucleosynthesis in astrophysics involves the progressive build-up of heavier isotopes via neutron captures proceeding on neutron-rich isotopes far off the valley of beta stability, interspersed by beta decays towards the stable regions [20]. The process is defined by the condition $r_\beta < r_n$, where $r_\beta$ is the life time for neutron capture and $r_n$ is the life time for beta unstable nuclei. Its abundance features reflect nuclear properties. Also, the process forms the important long-lived nuclear chronometers Th-232, U-238 and U-235 which are utilized for dating the galaxy. Although, the astrophysical site of the r-process nucleosynthesis is not yet identified, its association with type II supernovae is strongly suggested. Knowledge of r-process nucleosynthesis enables to put limits on the duration of galactic nucleosynthesis, on the age of the Galaxy and the Universe.

The character of the astrophysical r-process is dictated by nuclear properties in the neutron-rich region. One of these physics properties which is of fundamental importance in r-process calculations is the rate of beta-delayed neutron emission ($P_2n$ and $P_2\beta$). The other two are: neutron capture cross-sections and the beta-decay lives. The determinations of these properties are important to verifying empirical or physical models.

For the vast majority of nuclei in the r-process path, experimental information being non-available, it has become necessary to develope reliable theoretical methods for determining various inputs to the calculations. In determining neutron cross-
sections, substantial success has been achieved with the adoption of Hauser-Feshback or equivalent expression for the energy-averaged cross-section [21].

Thielemann et al [22] have calculated beta-delayed neutron emission employing the beta-strength distribution from Klapdor et al [23]. These calculated P values were used as input data in dynamical astrophysical r-process calculations for all nuclei with 75 < Z < 100 from the line of beta stability to the neutron drip line for three different mass formulae. Their results were consistent with the situation depicted by Howard and Möller [24]. Beta-delayed neutron emission also enters into the r-process network calculation through the system of differential equations for the time rate of change of the individual nuclear abundances:

\[
\frac{dY}{dt}(Z,A) = \sum_{Z',A'} \lambda_{Z',A'} Y_{Z',A'} + \sum_{Z',A'} N_A \langle \sigma v \rangle_{Z',A'} Y_{Z',A'} \frac{\tau_n}{\tau_n}
\]

Hence, the first term on the right-hand side includes beta decays and photodisintegrations; the second term includes all neutron-induced reactions.

With the assumption that the r-process abundances are produced under conditions of \((n,\gamma) \simeq (\gamma,n)\) equilibrium, Kratz et al [25] showed that beta-delayed neutron emission has an impact on the final shaping of the isotopic abundances after the r-process freeze-out and during the beta decays back to stability. They also showed that the isotopic ratios of In-131, In-133 and Ga-81, Ga-83 are related to the abundances of the stable isotopes Xe-131, Xe-132 and Br-81 and Kr-83 and that the beta-delayed neutron emission of In-133 and Ga-83 play an important role in these ratios.

Beta-delayed neutron emission is, usually, thought to smear and smooth out abundance fluctuations in the r-process path when they occur in a statistical way [26]. However, in the region 75 < A < 85, the predicted beta-delayed neutron emission seems to enhance the odd-even staggering, in agreement with the observed solar r-process abundances [27]. This behaviour could be due to strong \(\beta\)-delayed neutron branching from a few odd-mass isotopes located in or close to the r-process path.

In the explanation of solar system element abundances and of isotopic anomalies in meteorites [28], there are unresolved problems arising partly from our lack of knowledge of the relevant nuclear physics data for far-unstable nuclei like the \((n,\gamma)\) capture cross-sections \(\sigma_n\). These cross-sections are required while testing the validity of the classical \((n,\gamma)-(\gamma,n)\) equilibrium assumption. A direct measurement of \(\sigma_n\) for the short-lived nuclides being very difficult, theoretical 'average continuum cross-sections' from Hauser-Feshback (HF) calculations are commonly used in astrophysics [29]. But, HF calculations suffer from two drawbacks: their reliability depend to a great extent on a number of nuclear physics input parameters [30]; secondly, for nuclei with low \(\sigma(E)\), these calculations are replaced by Breit-Wigner (BW) resonance cross-sections. It has been demonstrated that a substantial part of these
input parameters can be derived from the 'inverse reaction' to n-capture, i.e., delayed neutron emission [19,30]. The actual inverse relationship demonstrated for Kr-87 is important for astrophysical applications in that high-resolution delayed neutron spectra of isotopes such as Br-87 permit us to determine the relevant parameters of special l-wave resonances as well as an experimental cross-section for the particular wave type(s). From this, partial cross-section and a reliable total $\sigma$-rate can be derived [30]. Using this method, level parameters of a number of unstable isotopes, for which high resolution delayed neutron spectra exist [31], have been determined. Results indicate that the $\sigma$-values obtained with correct input parameters differ considerably from the earlier HF-rates [29].

Sandler et al [32] have given an explanation of the Ca-Ti isotope anomalies found in high temperature meteoritic inclusions, in particular in the EK-1-4-1 inclusion from the Allende meteorite. In order to confirm this, the systems Ca47-Ca50(n,\gamma) were studied via their 'inverse reactions' K48-K51 (\beta,n) [33]. These systems being nuclei with low $\rho(E)$, their individual resonances dominate the total $\sigma$-rate. Combining the delayed neutron spectrum of K49 with shell model calculations, it has been shown that the low resonant contribution (of 2-3%) to the total Ca48 (n,\gamma) rate is due to the specific particle-hole structure of Ca49 which excludes both s-wave capture and delayed neutron emission up to about 1 MeV. This suggests that for the doubly-magic target Ca48, direct radiative capture is the dominant reaction mechanism. Another example is the possible non-statistical behaviour of the reaction Ca49+n [32].

In ref.[32], Sandler et al have also given an explanation of the Ti49/Ti50 abundance ratio. Although, existence of a low-energy s-wave resonance in Ca-50 could be verified from the delayed neutron spectrum of K-50, Sandler's suggestion could not be confirmed as measurements of partial decay widths of Ca-50 revealed a too low upper limit for the BW-rate. Further investigations have indicated that inclusion of the decay mode of delayed neutron emission of neutron-rich S to K isotopes into n-capture process models rather than finding $\sigma$ rates of particular isotopes may explain the solar Ca48/Ca46 abundance ratio and also the observed meteoritic Ca-Ti isotopic anomalies of EK-4-1 [33].

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§3. DELAYED NEUTRON IN REACTOR ANALYSIS

A satisfactory evaluation of the macroscopic effects of the delayed neutrons following fission in a nuclear reactor requires, among other data, an accurate knowledge of the delayed neutron data. These data consist of (distinct) quantities which are obtained either theoretically or experimentally and then evaluated and finally incorporated into nuclear data files such as the ENDF/B, the CEA data bank, JNDC (Japanese Nuclear Data Committee), etc. The quantities are: identification of the precursors of the delayed neutrons and the probability of their emission, the total delayed neutron yields, the delayed neutron energy spectra and "aggregate" descriptions, the half-lives, the fission yields and the average neutron energies. Of these, the ones that are of importance to the kinetics and safety calculations of nuclear reactors are:

(a) The absolute yield \(\bar{\nu}_d\) of delayed neutrons following fission induced by neutrons with energy up to about 10 MeV

(b) The energy distribution of the delayed neutrons \(x_d\)

(c) The division of the yield and the spectra into groups \((\beta_i, x_{di})\)

with more or less characteristic decay constants \(\lambda_i\).

In many reactor physics calculations, however, it is the total delayed neutron fraction \(\beta\) that is used. Beta is defined as the ratio of absolute delayed neutron yield to the average number of neutrons emitted per fission, \(\bar{\nu}_p\). But the quantity that determines the margin of control and therefore, the safety of a nuclear reactor is the reactor parameter, \(\beta_{\text{eff}}\) and not the nuclear \(\beta\). \(\beta_{\text{eff}}\) is the total effective fraction of delayed neutrons. The following relations hold good [1].

\[
\beta = \frac{\bar{\nu}_d}{\bar{\nu}_p} = \sum_i \beta_i \\
\beta_{\text{eff}} = \bar{\gamma} \beta = \sum_i \beta_{\text{eff},i} = \sum_i \gamma_i \beta_i \\
\beta_{\text{eff}}(t) = \left[ \sum_{k=1}^{I} \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E, t) \\
\times \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E', t) \\
\times \sum_{k=1}^{I} \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E, t) \\
\times \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E', t) \\
\times \sum_{k=1}^{I} \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E, t) \\
\times \int \int \int \nu_k(E') x_{di}(E) \beta_i \beta_k \Gamma_f(\vec{r}, E', t) \right] \, \, \, (\vec{r}, E', t)
\]

\((k, I) is for fissile isotopes, i, m is for delayed neutron precursor groups and \(\Phi^*\) is the adjoint flux).

Here, \(\bar{\gamma}\) and \(\gamma_i\) denote the neutron effectiveness factors and represent the effectiveness of a delayed neutron with respect to a prompt neutron in causing fission. It depends on the position and energy of the delayed neutron when it is born. Calculation of neutron effectiveness factor depends on the prompt and the delayed neutron spectra and on the details of the composition and construction of the reactor. It needs to be emphasized that the group structure of delayed neutrons can either be physical or mathematical, and that, it is only historical that in most
applications of delayed neutrons, the nuclear technology industry has been content to work with the much simpler temporal group representation of delayed neutron emission obtained from the measured aggregate data. These groups have no true physical basis and the generally accepted six-group delayed neutron data of Keepin et al [2,3] is a mathematical one. They originated as six-term, twelve-parameter optimum least-squares fit to experimentally measured count rates following fission pulse and saturation irradiation experiments in critical assemblies. Obviously, there are disadvantages in attempting to work with the large number of equations, one for each precursor, when studying reactor kinetics. It is only relatively recently that Perry et al [4] and Brady and England [5] have reported the results of kinetics calculations using individual precursor data.

The difference between the mathematical and the physical representation of delayed neutron data is that while in the former, different fissile isotopes have different $\lambda_i$'s and different $x_d$'s, in the latter, the $\lambda_i$'s and $x_d$'s for the physical precursor groups are independent of the fissile material in which the fission occurred. Since, each mathematical group represents delayed neutrons contributed from different fission product nuclei, the delayed neutron spectra of the mathematical groups will be time-dependent.

REFERENCES


§4. ABSOLUTE DELAYED NEUTRON YIELD

The absolute delayed neutron yield \((\bar{Y}_d)\) of a fissile isotope is the number of delayed neutrons emitted per fission. Since the first detection of delayed neutron emission, yields for a variety of fissionable isotopes have been measured, calculated and evaluated in the neutron energy range of 0 to about 10 MeV (the energy region of interest in reactor physics and design) using various forms of nuclear fission such as neutron fission, charged-particle fission, photofission and spontaneous fission. Many measurements have also been made near 14 to 15 MeV. For detailed reviews and discussions on the subject, readers may refer to [1-12].

The reported delayed neutron yield measurements for fission caused by 14-15 MeV neutrons appear to fall into two groups: one of high values, measured prior to 1966, and one of low values, measured later. The latter values are in reasonable agreement with predictions and the former values are consistent within themselves when compared between isotopes at 14-15 MeV. Waldo et al [7] obtained an expression for the yield of beta-delayed-neutrons by a least-squares-fit of the available data (excluding \(^{237}\text{Np}\) photofission, \(^{234}\text{U}\) photofission and \(^{252}\text{Cf}\) spontaneous fission). The expression, which has an accuracy of \(\pm 9\%\), is:

\[
Y_{DN} \text{ (per 100 fissions)} = \exp(16.698 - 1.144Z_C + 0.377A_C)
\]

where, \(A_C\) and \(Z_C\) are the composite mass and charge of the fissioning material. Correlations such as this are useful in estimating delayed neutron yields for unmeasured nuclides. For example, the contribution of \(^{238}\text{Pu}\) or \(^{236}\text{U}\) fission in reactors with these minor contributions can be estimated using such a correlation.

Tuttle [4,5] prepared an extensive review of the measurements of \(\bar{Y}_d\) and also an evaluation. The uncertainties listed for the calculations were based on fission product yield uncertainties in ENDF/B-V and the \(P\) uncertainties. A 100% uncertainty was assigned to the estimated \(P\) values. Except for \(^{235}\text{U}\) thermal fission, the calculated uncertainties were probably small. The \(\bar{Y}_d\) calculations were sensitive to the yield distribution along each mass chain. This yield distribution was based on models using parameters derived largely from \(^{235}\text{U}\) independent yields. Errors in the most probable charge or nuclear pairing effects could readily alter the \(\bar{Y}_d\) values, even if mass chain yields were exact. Most (-90%) of the delayed neutrons are emitted from odd-Z precursors, and a relatively small error in the pairing could result in a large error in \(\bar{Y}_d\). More recently, Tuttle [11] combined the experimental information on delayed neutron yields from nuclear fission induced by broad-spectrum and narrow spectrum neutrons, photofission and spontaneous fission with the calculated values in a unified manner to provide a consistent set of energy-dependent evaluated yields for 44 fission reactions from \(^{227}\text{Th}(n,f)\) to \(^{256}\text{Fm}(sf)\). The data were integrated, extended and interpolated using the observed systematic dependence of yield on the parameters \(Z\), \(A\), and \(E\) [13] of the fissioning nucleus. The evaluation provided estimates for yields from zero...
excitation energy (corresponding to spontaneous fission) to fission induced by 15-MeV neutrons. The evaluation process consisted in first combining the prepared absolute and relative measurements to produce measured (and calculated) yield values at specified energies. These values were then used to generate a continuous pointwise curve. This curve and an adjusted curve from the systematics calculation were used to produce the evaluated yields. These evaluated yields were then used to convert the relative measurements to absolute values and the process continued.

Uncertainty estimates were provided over the energy range. They were determined by a combination of the uncertainties expected for the averaged values used in the first curve generation and the maximum of the expected and the observed uncertainties derived from the systematics calculation.

Comparison of the evaluated yield with the recommended yield from the 1974 evaluation of Cox [3], used in ENDF-B, showed good agreement below 7 MeV, but above this energy Tuttle's evaluation was significantly higher. The evaluation showed considerably more structure in the energy variation than was possible before. The evaluation also provided information on the delayed neutron yield for poorly known fission reactions, such as U-237(n,f) (see figure B below). For this reaction, the evaluated yield was based on the single input value calculated by T.R. England and B.F. Rider [10, pp.33-64] at an estimated effective neutron energy of 1 MeV.

![Fig. B: Evaluated yield values for neutron induced fission of U-237.](image)
The relation for $\beta$ (see §3) shows that error in $\beta$ comes from errors in $\bar{\nu}_d$ and $\bar{\nu}_p$. Error in beta-effective comes from error in the calculation of $\gamma$ due to the incomplete knowledge of delayed neutron spectra. According to Hammer [14], the target accuracy on beta-effective deduced from power reactor and critical experiment requirements is $\pm 3\%$ (1o). To achieve this, one has to determine the absolute yields with a $\pm 1.5\%$ error margin for the major fissile isotopes U235, U238 and Pu239. For a fast breeder reactor, one has to know the plutonium higher isotopes (Pu240, Pu241) data with an uncertainty of $\pm 7\%$ and for Pu242, Am241 the accuracy required is $\pm 10\%$. The present dispersion between the various evaluated results is higher than the maximum uncertainty required for the evaluated results. Also, major contributions are currently being made by the calculated values. There is, therefore, a need for additional experimental measurements of the broadest possible scope and comprehensiveness to complement the existing data base.

Although the total delayed neutron yield has been measured for many isotopes, the recent experimental work on precursors allows summation calculations such as the following [15] to determine $\bar{\nu}_d$.

$$\bar{\nu}_d = \int \int x_d(E, t) \, dE \, dt = \int P_n \int Y_f^n(t) \, dt$$

where, $x_d(E, t)$ is the delayed neutron spectrum, $P_n$ is the probability of delayed neutron emission by a precursor $n$ and $Y_f^n(t)$ is the effective time-dependent precursor yield from the fission process. The primary purpose of $\bar{\nu}_d$ calculations is to test the fission product yield.

Table 1 lists the results of recent $\bar{\nu}_d$ calculations and comparative results from evaluations and selected measurements. The T, F, and H following the fissionable nuclide refer, respectively, to the thermal, fast, and high energy (>-14 MeV) fission. Because of the large number of $\bar{\nu}_d$ measurements, particularly for some of the fissionable nuclides, only a selection of these are listed here.

Calculation marked ENDF/B-VI used preliminary ENDF/B-VI fission yields. Calculation marked ENDF/B-V used evaluated fission yields of the precursors and their $P$ values from ENDF/B-V. Generally, the calculated $\bar{\nu}_d$ is very good. $\bar{\nu}_d^n$ values for U238 and Th232 suggest that either an improvement in ENDF/B fission yields is needed or the evaluated experimental $\bar{\nu}_d$ values are in significant error. Table 2 compares relative abundances of the six time groups for three fuels. The values in Table 2 are expressed as a per cent of $\bar{\nu}_d$. Results of Tables 1 and 2 show that using the measured delayed neutron data for precursors, the model calculations and the evaluated fission product yields, one can do reasonably accurate summation calculations of the total $\bar{\nu}_d$ and its relative abundance in each of the conventional six time-groups for a variety of fissioning nuclides and neutron fission energies. Table 3 lists the total $\bar{\nu}_d$ and its dependence on the neutron fission energy as incorporated into ENDF/B-V [19]. Figure 1 gives a typical representation of it. Here, "linear" refers to a linear interpolation in $\bar{\nu}_d$ versus energy for the specified range, and "constant" refers to a constant value of $\bar{\nu}_d$ over the noted range.
Table 1: Comparison of total delayed neutron yield per 100 fissions

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th (F)</td>
<td>5.70 ± 1.08</td>
<td>4.76 ± 0.34</td>
<td>5.27</td>
<td>5.31 ± 0.23</td>
<td>4.96 ± 0.30</td>
</tr>
<tr>
<td>$^{232}$Th (H)</td>
<td>4.15 ± 1.07</td>
<td>3.03 ± 0.29</td>
<td>3.00</td>
<td>2.85 ± 0.13</td>
<td>3.1 ± 0.3</td>
</tr>
<tr>
<td>$^{233}$U (T)</td>
<td>0.968 ± 0.247</td>
<td>0.846 ± 0.066</td>
<td>0.740</td>
<td>0.667 ± 0.029</td>
<td>0.66 ± 0.04</td>
</tr>
<tr>
<td>$^{233}$U (F)</td>
<td>0.907 ± 0.157</td>
<td>0.916 ± 0.089</td>
<td>0.740</td>
<td>0.731 ± 0.036</td>
<td>0.78 ± 0.08</td>
</tr>
<tr>
<td>$^{233}$U (H)</td>
<td>0.704 ± 0.138</td>
<td>0.708 ± 0.095</td>
<td>0.420</td>
<td>0.422 ± 0.025</td>
<td>0.43 ± 0.04</td>
</tr>
<tr>
<td>$^{235}$U (T)</td>
<td>1.78 ± 0.15</td>
<td>1.77 ± 0.081</td>
<td>1.67</td>
<td>1.621 ± 0.05</td>
<td>1.58 ± 0.07</td>
</tr>
<tr>
<td>$^{235}$U (F)</td>
<td>2.07 ± 0.293</td>
<td>1.98 ± 0.18</td>
<td>1.67</td>
<td>1.673 ± 0.036</td>
<td>1.71 ± 0.17</td>
</tr>
<tr>
<td>$^{235}$U (H)</td>
<td>1.09 ± 0.193</td>
<td>0.978 ± 0.097</td>
<td>0.900</td>
<td>0.927 ± 0.029</td>
<td>0.95 ± 0.06</td>
</tr>
<tr>
<td>$^{236}$U (F)</td>
<td>2.32 ± 0.344</td>
<td>2.26 ± 0.19</td>
<td>-</td>
<td>2.21 ± 0.24</td>
<td>-</td>
</tr>
<tr>
<td>$^{238}$U (F)</td>
<td>4.03 ± 0.435</td>
<td>3.51 ± 0.27</td>
<td>4.40</td>
<td>4.39 ± 0.1</td>
<td>4.12 ± 0.25</td>
</tr>
<tr>
<td>$^{238}$U (H)</td>
<td>2.71 ± 0.385</td>
<td>2.69 ± 0.21</td>
<td>2.60</td>
<td>2.73 ± 0.08</td>
<td>2.83 ± 0.13</td>
</tr>
<tr>
<td>$^{237}$Np (F)</td>
<td>1.14 ± 0.158</td>
<td>1.28 ± 0.13</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>$^{239}$Pu (T)</td>
<td>0.763 ± 0.05</td>
<td>0.769 ± 0.058</td>
<td>0.645</td>
<td>0.628 ± 0.038</td>
<td>0.61 ± 0.05</td>
</tr>
<tr>
<td>$^{239}$Pu (F)</td>
<td>0.679 ± 0.093</td>
<td>0.724 ± 0.090</td>
<td>0.645</td>
<td>0.630 ± 0.016</td>
<td>0.65 ± 0.06</td>
</tr>
<tr>
<td>$^{239}$Pu (H)</td>
<td>0.379 ± 0.076</td>
<td>0.387 ± 0.062</td>
<td>0.430</td>
<td>0.417 ± 0.016</td>
<td>0.43 ± 0.04</td>
</tr>
<tr>
<td>$^{240}$Pu (F)</td>
<td>0.806 ± 0.109</td>
<td>0.923 ± 0.11</td>
<td>0.900</td>
<td>0.95 ± 0.08</td>
<td>0.88 ± 0.09</td>
</tr>
<tr>
<td>$^{241}$Pu (T)</td>
<td>1.39 ± 0.125</td>
<td>1.58 ± 0.13</td>
<td>1.62</td>
<td>1.52 ± 0.11</td>
<td>1.57 ± 0.15</td>
</tr>
<tr>
<td>$^{241}$Pu (F)</td>
<td>1.39 ± 0.165</td>
<td>1.49 ± 0.16</td>
<td>1.62</td>
<td>1.52 ± 0.11</td>
<td>-</td>
</tr>
<tr>
<td>$^{242}$Pu (F)</td>
<td>1.40 ± 0.169</td>
<td>1.41 ± 0.14</td>
<td>-</td>
<td>2.21 ± 0.26</td>
<td>1.6 ± 0.5</td>
</tr>
<tr>
<td>$^{252}$Cf (s)</td>
<td>0.608 ± 0.069</td>
<td>0.690 ± 0.092</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
Table 2: Comparison of relative group abundances (%) for U235 (T), Pu239 (F) and U238 (F)

<table>
<thead>
<tr>
<th>Fissile Nuclide</th>
<th>Precursor Group</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>235U (T)</td>
<td>Calculated (B-VI)</td>
<td>3.0</td>
<td>21.1</td>
<td>17.8</td>
<td>38.2</td>
<td>14.2</td>
<td>5.7</td>
</tr>
<tr>
<td></td>
<td>Calculated (B-V)</td>
<td>2.9</td>
<td>22.0</td>
<td>17.9</td>
<td>38.4</td>
<td>13.3</td>
<td>5.6</td>
</tr>
<tr>
<td></td>
<td>ENDF/B-V(Keepin)</td>
<td>3.8</td>
<td>21.2</td>
<td>18.8</td>
<td>40.7</td>
<td>12.8</td>
<td>2.6</td>
</tr>
<tr>
<td>239Pu (F)</td>
<td>Calculated (B-VI)</td>
<td>2.9</td>
<td>26.0</td>
<td>20.0</td>
<td>36.6</td>
<td>12.1</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>Calculated (B-V)</td>
<td>2.6</td>
<td>25.6</td>
<td>18.7</td>
<td>35.2</td>
<td>15.7</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>ENDF/B-V(Keepin)</td>
<td>3.8</td>
<td>28.0</td>
<td>21.6</td>
<td>32.8</td>
<td>10.3</td>
<td>3.5</td>
</tr>
<tr>
<td>238U (F)</td>
<td>Calculated (B-VI)</td>
<td>1.0</td>
<td>11.8</td>
<td>14.2</td>
<td>48.3</td>
<td>18.2</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td>Calculated (B-V)</td>
<td>1.1</td>
<td>15.5</td>
<td>15.2</td>
<td>43.3</td>
<td>19.0</td>
<td>6.0</td>
</tr>
<tr>
<td></td>
<td>ENDF/B-V(Keepin)</td>
<td>1.3</td>
<td>13.7</td>
<td>16.2</td>
<td>38.8</td>
<td>22.5</td>
<td>7.5</td>
</tr>
</tbody>
</table>

Table 3: Summary of ENDF/B-V Evaluation for Delayed Neutrons

<table>
<thead>
<tr>
<th>Fissionable Nuclide</th>
<th>( \bar{\nu} ) per 100 Fissions</th>
<th>Energy Range (MeV)</th>
<th>Spectra</th>
</tr>
</thead>
<tbody>
<tr>
<td>232Th</td>
<td>5.27</td>
<td>Constant 0 to 4</td>
<td>Same as 235U</td>
</tr>
<tr>
<td></td>
<td>5.27 to 3.00</td>
<td>Linear 4 to 7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.00</td>
<td>Constant 7 to 20</td>
<td></td>
</tr>
<tr>
<td>233U</td>
<td>0.740</td>
<td>Constant 0 to 4.5</td>
<td>Same as 235U</td>
</tr>
<tr>
<td></td>
<td>0.740 to 0.470</td>
<td>Linear 4.5 to 6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.470</td>
<td>Constant 6 to 14</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.470 to 0.420</td>
<td>Linear 14 to 15</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.420</td>
<td>Constant 15 to 20</td>
<td></td>
</tr>
<tr>
<td>235U</td>
<td>1.67</td>
<td>Constant 0 to 4</td>
<td>Group 4 spectra</td>
</tr>
<tr>
<td></td>
<td>1.67 to 0.900</td>
<td>Linear 4 to 7</td>
<td>used for groups 5 and 6</td>
</tr>
<tr>
<td></td>
<td>0.900</td>
<td>Constant 7 to 20</td>
<td></td>
</tr>
<tr>
<td>238U</td>
<td>4.40</td>
<td>Constant 0 to 4</td>
<td>Group 5 spectra</td>
</tr>
<tr>
<td></td>
<td>4.40 to 2.60</td>
<td>Linear 4 to 9</td>
<td>used for group 6</td>
</tr>
<tr>
<td></td>
<td>2.60</td>
<td>Constant 9 to 20</td>
<td></td>
</tr>
<tr>
<td>239Pu</td>
<td>0.645</td>
<td>Constant 0 to 4</td>
<td>Group 4 spectra</td>
</tr>
<tr>
<td></td>
<td>0.645 to 0.430</td>
<td>Linear 4 to 7</td>
<td>used for groups 5 and 6</td>
</tr>
<tr>
<td></td>
<td>0.430</td>
<td>Constant 7 to 20</td>
<td></td>
</tr>
<tr>
<td>240Pu</td>
<td>0.900</td>
<td>Constant 0 to 4</td>
<td>Same as 239Pu</td>
</tr>
<tr>
<td></td>
<td>0.900 to 0.615</td>
<td>Linear 4 to 7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.615</td>
<td>Constant 7 to 20</td>
<td></td>
</tr>
<tr>
<td>241Pu</td>
<td>1.62</td>
<td>Constant 0 to 4</td>
<td>Same as 239Pu</td>
</tr>
<tr>
<td></td>
<td>0.900 to 0.840</td>
<td>Linear 4 to 7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.840</td>
<td>Constant 7 to 20</td>
<td></td>
</tr>
</tbody>
</table>
Fig. 1: A typical representation of $\bar{J}_d$ dependence on neutron fission energy as incorporated into ENDF/ B-V.
range. The ENDF/B-V aggregate spectra shown in the table are based on Fieg's measurements [20] for time groups 1 through 4 or 5. Note that the U235 spectrum is used for Th232 and U233, and the Pu239 spectrum is used for Pu240 and Pu241. These spectra are normalised and are assumed to be independent of the incident neutron fission energy. The original review of spectra extended from about 79 keV to about 1.2 MeV. The lower cut-off was later changed using a straight line extension to E = 0 from the value at 79 keV.

REFERENCES


19) Fission-Product Decay Library Of the Evaluated Nuclear Data File (ENDF/B-IV and-V): Available from and maintained by the National Nuclear Data Centre (N.N.D.C.), Brookhaven National Laboratory, Upton, New York, USA.

§5. DELAYED NEUTRON DECAY CONSTANTS

The group decay constants $\lambda_i$ are usually deduced from the individual data $\lambda_{ik}$ per isotope. There are two ways of doing this. The first is to combine the $\lambda_{ik}$ as:

$$\lambda_i = \left( \sum_k \lambda_{ik} \beta_{ieff} \right) / \left( \sum_k \beta_{ieff} \right)$$

and the second is by combining $1/\lambda_{ik}$ as:

$$\left(1/\lambda_i \right) = \sum_k \left( \beta_{ieff}/\lambda_{ik} \right) / \left( \sum_k \beta_{ieff} \right)$$

where, $\beta_{ieff}$ is the effective fraction of delayed neutrons emitted by the fissile isotope $k$ in the delay group $i$. But due to the very close values of $\lambda_{ik}$ for the various fissile isotopes, the above two procedures for calculating $\lambda_i$ and $\bar{\lambda}$ (the mean decay constant) give results which are not very different from each other, the differences being less than two per cent. The group decay constants $\lambda_i$ do not depend very much either upon the fissile isotope $k$ (see Table 4) or upon the energy of the neutron causing fission (fast or thermal). Table 5 gives mean values of $\lambda_i$ for all the main isotopes (U and Pu), and compares the dispersion associated to these mean values with the uncertainties applied to the individual $\lambda_{ik}$ values. One notes that the differences between $\lambda_{ik}$ within one group and for various isotopes are not really significant due to the corresponding error margins. Consequently, the present accuracies given on the $\lambda_i$ seem sufficient for kinetics calculations.

As regards the yield repartition $\alpha_{ik}$ for each isotope, Keepin has determined the values with uncertainties which depend little on the isotope and more on the precursor group $i$. The mean values of these uncertainties group-wise are: 15%, 2.5%, 20%, 7%, 12% and 20%. The present uncertainties on $\alpha_{ik}$ seem acceptable for reactor applications. In particular, Beta-effective is insensitive to modifications in $\alpha_{ik}$.

The six groups delayed neutron scheme and the corresponding decay constants evaluated by Keepin are adopted in Delayed Neutron Constant evaluations. Though, Keepin's and Tuttle's delayed neutron data (group yield and decay constants) are widely used in reactor analysis, in recent times, similar data have been generated by others too. These new data have, however, not been in great use so far.

REFERENCE

Table 4: Decay constant per delayed neutron group and fissile element
($\lambda_i^{\text{sec}^{-1}}$)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Precursor Group, i</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
</tr>
<tr>
<td>$^{235}\text{U}$</td>
<td>0.0127</td>
</tr>
<tr>
<td>$^{238}\text{U}$</td>
<td>0.0132</td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>0.0129</td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>0.0129</td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>0.0128</td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>0.0129</td>
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</tbody>
</table>

Table 5: Mean values of group decay constant

<table>
<thead>
<tr>
<th>Delayed neutron group, $i$</th>
<th>Mean value for all U and Pu, $\lambda_i$</th>
<th>Dispersion on mean, $\lambda_i$ (%)</th>
<th>Error on individual, $\lambda_i$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.0129</td>
<td>±1.3</td>
<td>±3</td>
</tr>
<tr>
<td>2</td>
<td>0.0309</td>
<td>±3.3</td>
<td>±3</td>
</tr>
<tr>
<td>3</td>
<td>0.130</td>
<td>±6.7</td>
<td>±8</td>
</tr>
<tr>
<td>4</td>
<td>0.337</td>
<td>±4.9</td>
<td>±6</td>
</tr>
<tr>
<td>5</td>
<td>1.405</td>
<td>±8.1</td>
<td>±10</td>
</tr>
<tr>
<td>6</td>
<td>3.710</td>
<td>±8.9</td>
<td>±15</td>
</tr>
</tbody>
</table>
§6. DELAYED NEUTRON ENERGY SPECTRA

Of the three sets of numbers that are required in the dynamics and safety calculations of reactors, the least adequately known is the energy spectrum of delayed neutrons for each delayed group \( x_{di} (E) \) as well as for the composite spectra as a function of time, \( x_d(t) \) after fission. This inadequacy has been due as much to the complexity of measurements - arising from the very short life time of some of the precursors and the difficulty in separating them - as due to a lack of demand for exact delayed neutron data in thermal reactor calculations.

6.1 Importance of Delayed Neutron Energy Spectra in Fast Reactor Dynamic Calculations

Although the importance of delayed neutrons in controlling the rate of a fission chain reaction was noted as early as in 1940 [1] and the importance of the energy spectra of delayed neutrons in predicting the kinetic response of fast breeder reactors (and the space reactor systems) under fault conditions has been long recognised [2-5], the available information on the number and energy distribution of delayed fission neutrons is inadequate for the design of the coming generation of high power fast breeder reactors characterized by large quantities of fertile material (U-238 or Th-232) in the breeder/blanket reflector. The importance of an accurate knowledge of the delayed neutron spectra to the kinetic behaviour, control and safety analysis of fast reactors, particularly the breeders, is due to the following considerations:

(i) In \(^{235}\text{U}\) thermal systems, the total effective delayed fraction is often about 0.0065, because in such systems, fissions occur mainly in U-235. But in fast breeder reactors, which may contain large quantities of U-238 and Th-232 isotopes in the core and/or the breeder blanket, the neutron spectrum being fast, an appreciable fraction (upto 25%) of total fissions may occur in these nuclides which have high threshold energies for fission and whose delayed neutron yields are relatively high (\( \beta \) of U-238 = 0.0148; \( \beta \) of Th-232 = 0.0203). Depending on the energy with which delayed neutrons are born, they may or may not be able to produce fission in these isotopes. This can significantly alter the delayed effective fraction (group as well as total) and in certain cases can dominate the dynamic behaviour of the reactor. The high burn-up breeder reactors (Pu-U cycle) produce not only Pu-239 but also the higher isotopes Pu-240, Pu-241, Pu-242 in significant quantities. The delayed neutrons from these higher N/Z ratio species will also affect the kinetics and safety of such reactors.

(ii) In a fast breeder reactor, the fissile and the fertile substances are usually not homogeneously distributed. In such a situation, the dynamic behaviour of different regions in the fast breeder will be characterized by different time-scales: "slow" in regions rich in U-238, Th-232 and Pu-240; "fast" in regions rich in U-235 (\( \beta = 0.0065 \)), Pu-239 (\( \beta = 0.002 \)) and Pu-241 (\( \beta = 0.0049 \)). Therefore, while investigating the possible damage that a local disturbance might cause in a fast breeder, it might be critical under certain circumstances to know the exact effective delayed
neutron fraction in each region of the reactor.

(iii) Delayed neutrons are more effective in causing fission than prompt neutrons because they are born with lower average energies and they suffer smaller leakage. One, therefore, expects markedly different effectiveness for delayed neutrons than for prompt neutrons. In fast reactors, most neutron interactions occur in fast (keV) regions, and therefore, an accurate knowledge of the delayed neutron spectrum will be of greater importance than in thermal reactors where almost all neutrons are slowed down before they produce fission [6].

Table 6 shows the results of neutron effectiveness calculations for four delayed groups and their composite value for six fast metal assemblies using the delayed neutron energy spectra of Batchelor and Bonner [7]. It is seen that for the bare assemblies, the individual $\gamma_i$'s differ from $\bar{\gamma}$ by less than 2% excepting the delay group 1 whose very low energy spectrum results in larger values. The three reflected fast assemblies, on the other hand, exhibit large differences among the individual $\gamma_i$ values. Thus in reflected fast systems, notably fast breeder reactors, the common assumption of equal effectiveness for all the six delayed groups ($\gamma_1$-$\gamma_6$) can lead to a wrong prediction of the peak power in fast reactor transient calculations. The need for and the importance of more accurate and complete delayed neutron spectral data for all the main fissile species is quite apparent for precise evaluation of neutron kinetics and control characteristics.

It is noted that both the $\gamma_i$ and $\bar{\gamma}$ exceed unity for the $^{235}\text{U}$ and U-233 bare systems. This is because in these thermal fissioning species, the lower energy of delayed neutrons means higher fission probability (greater $\Gamma_i$) and higher non-leakage probability [$\exp(-B_i\gamma_i)$] while slowing down. An anomalous situation occurs in the Pu-239 bare assembly whose $\gamma_1$ and $\bar{\gamma}$ are less than unity due to the dip in the Pu-239 fission cross-section in the vicinity of 0.5 MeV. One therefore expects $\gamma_1$ to exhibit the smallest effectiveness value for Jezabel since the second delay group spectrum peaks in the vicinity of 0.5 MeV.

In reflected fast systems, a considerable fraction of fissions occur in the U-238 of the reflector which has a large relative neutron yield. But the importance of the neutrons (prompt as well as delayed) born in the reflector being much lower than the importance of the neutrons born in the core, the major contribution to beta-effective comes from the core material in the three reflected systems. Consequently, despite the large relative neutron yields from U-238 in the reflector, the overall delay fractions are not radically different from the corresponding values in the bare assembly.

Figure 2 illustrates a practical application of neutron effectiveness calculations in reactor kinetics. It shows the period-reactivity relations calculated using the above values of $\gamma_1$ and $\bar{\gamma}$ for two reflected metal critical assemblies Popsy and 23 Flattop. Curves for the pure isotopes U-233, Pu-239, U-238 [7] and for Purnima 2, a U-233 fuelled BeO reflected aqueous homogeneous thermal
Table 6: Comparison of delayed neutron effectiveness values for six metal assemblies

<table>
<thead>
<tr>
<th>Assembly</th>
<th>$\gamma_1$</th>
<th>$\gamma_2$</th>
<th>$\gamma_3$</th>
<th>$\gamma_4$</th>
<th>$\bar{\gamma}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare $^{235}$U (Godiva)</td>
<td>1.096</td>
<td>1.028</td>
<td>1.050</td>
<td>1.033</td>
<td>1.034</td>
</tr>
<tr>
<td>Bare $^{239}$Pu (Jezebel)</td>
<td>0.963</td>
<td>0.942</td>
<td>0.947</td>
<td>0.944</td>
<td>0.945</td>
</tr>
<tr>
<td>Bare $^{233}$U (Skidoo)</td>
<td>1.123</td>
<td>1.055</td>
<td>1.078</td>
<td>1.059</td>
<td>1.06</td>
</tr>
<tr>
<td>$^{238}$U-reflected $^{235}$U (Topsy)</td>
<td>1.089</td>
<td>0.936</td>
<td>0.902</td>
<td>0.872</td>
<td>0.863 ($\bar{\gamma}_{cove} = 1.08$)</td>
</tr>
<tr>
<td>$^{238}$U-reflected $^{239}$Pu (Popsy)</td>
<td>0.739</td>
<td>0.668</td>
<td>0.585</td>
<td>0.498</td>
<td>0.530 ($\bar{\gamma}_{cove} = 1.35$)</td>
</tr>
<tr>
<td>$^{238}$U-reflected $^{233}$U (23 Flattop)</td>
<td>1.119</td>
<td>0.803</td>
<td>0.737</td>
<td>0.603</td>
<td>0.650 ($\bar{\gamma}_{cove} = 1.33$)</td>
</tr>
</tbody>
</table>
(iv) It is often assumed in reactor applications that the beta-effective is a constant during a particular transient calculation. But the relation for beta-effective (see §3) shows that the beta-effective is dominated by the ratio of the delayed neutron spectra \((\chi_d)\) to the prompt neutron spectra \((\chi^p)\) and the variables \(\phi\) (flux), adjoint flux \((\phi^a)\), \(\Sigma_f\) (macroscopic fission cross-section) are time-dependent. Therefore, beta-effective is time-dependent and the reactivity of the system in dollar units changes with time both locally and globally over the reactor depending upon the numbers used to describe the delayed neutron effects. To know the effects on power, power shape and power density, one must know the delayed neutron spectra for the different fissile materials in the reactor accurately. Consider, for example, a transient in a liquid-metal fast breeder reactor in which a segment of the core or subassembly is voided of sodium. In this segment, the neutron spectra will become harder. If significant amounts of U-238 are present in this region of the reactor, the relative number of fissions in U-238 will increase. Since a fission event in U-238 produces nearly six times more number of delayed neutrons than a fission event in Pu-239 and since the delayed neutrons emitted by the two nuclei have different energy spectra, the net effect might be a strong time-dependent beta-effective depending upon the exact interaction of the delayed neutron spectra with the other components of the reactor. The dynamics of the system will, therefore, depend on the delayed
neutron spectra used. The actual magnitude and importance of this effect would depend greatly on the composition and configuration of the reactor considered and can be ascertained only by detailed investigations.

(v) Measured values of reactor physics parameters obtained from integral experiments on critical assemblies represent a composite of many individual input parameters. Therefore, further refinement of delayed neutron numbers and the spectral data will contribute towards more definite cross-section evaluations from critical experiments. This will increase the accuracy and reliability of fast power reactor calculations.

(vi) The calculated and the measured values of reactivity are related to each other by beta-effective. Since beta-effective depends - among other quantities - on the prompt and the delayed neutron spectra, accurate determination of reactivity value requires an accurate knowledge of delayed spectral data. Efforts to improve the calibration of the reactivity scale between calculations and measurements by adjusting the total delayed neutron yields have been reported by D'Angelo [8].

(vii) In most analysis of critical assemblies and in the design of power reactors, it is commonly assumed that the delayed and the prompt neutrons are emitted with the same energy distribution. This is justified on the ground that the influence of this approximation is small in comparison with other effects such as the nuclear data uncertainties or the approximations made in modelling real reactor configuration. But, Kiefhaber [9] has recently reported that this assumption can lead to systematic deviations in k-effective of between -0.2 and +0.05%, the effect being more important for low-enriched k experiments and for highly enriched, high-leakage cores than for typical cores of LMFBR and their critical assemblies. It is, therefore, necessary that in cross-section adjustment procedures which usually cover a wide range of critical assemblies with fairly different nuclear characteristics, in precise prediction of nuclear parameters for an operating power reactor, or in high accuracy criticality calculation, the influence of exact delayed neutron spectra is taken into account.

(viii) Prompt neutron decay constant, $\alpha_{DC} = (\text{beta-effective/mean prompt neutron life time})$ can be subject to calculational error due to the uncertainties in the delayed neutron spectral data. Errors can also arise in the determination of prompt neutron life time using the measured values of $\alpha_{DC}$ and the calculated values of beta-effective ($L_p = \beta_{eff}/\alpha_{DC}$).

(ix) One of the design requirements of a nuclear reactor is that it should be operationally stable. This is normally achieved by constructing the reactor with a high operational safety margin. But higher the safety margin, higher is the cost. Hence to reduce costs, one needs to reduce the uncertainties in accident analysis. This means, in general, of improving and enlarging the existing basic experimental nuclear data, including the data on delayed neutrons.
(x) It has been pointed out several times that delayed neutrons do not always have a stabilizing influence on nuclear reactor dynamics [10,11]. Complete neglect of delayed neutrons would, in certain cases, result in a conditionally stable reactor, whereas including delayed neutron effects in the same reactor would make it unstable. Therefore, changes in beta-effective resulting from the uncertainties in the knowledge of delayed neutron spectral data may shift the reactor either into the unstable region or to a more stable region, depending on the error inherent in the neutron effectiveness calculations. Hence, to say whether a certain reactor configuration is stable or not, a precise evaluation of beta-effective is required and can be obtained only if delayed spectra are known accurately.

6.2 Determination of Delayed Neutron Spectra

In reactor applications, two types of delayed neutron energy spectra are, in principle, required: the equilibrium spectra for static calculations and the time-dependent spectra for dynamic calculations. These spectral data are obtained by commonly employing two independent approaches. The first is from microscopic weighted summation of measured or theoretically calculated separated precursor results (energy spectra, half-lives, cumulative fission yield and delayed neutron emission probability) and the second is the direct measurement of composite delayed neutron spectra containing contributions from all precursors decaying within specific time intervals after fission (§7). The direct measurement of aggregate delayed neutron spectra and their decomposition into six delay groups has the advantage that it avoids the uncertainties associated with the combining of many individual precursor spectra to produce the composite spectra. However, these measurements have not been very sensitive to the two shortest-lived Keepin groups, namely, groups 5 and 6. The summation method of generating delayed neutron energy spectra from precursors (§8) has the principal advantage that a single set of precursor data (delayed neutron emission probabilities and energy spectra) can be used to predict delayed neutron production for whatever group structure and fissioning system required, provided fission product yields are available [12]. Also, the method is mathematically more tractable than the decomposition method.

The basic relations used in the summation calculations are:

\[
Y_{in} = \sum_{k} \gamma_{k} Y_{i} P_{ni}
\]

\[
A_{i} = \sum_{n=1}^{\infty} Y_{in}
\]

\[
\lambda_{i} = \sum_{n=1}^{\infty} \gamma_{in} / \sum_{n=1}^{\infty} Y_{in}
\]

\[
A_{i} \chi_{di}(E) dE = \sum_{n=1}^{\infty} Y_{in} \chi_{dn}(E) dE
\]
\[ x_d(E,t) = \sum_{k=1}^{n} \sum_{i=1}^{m} \lambda_i^k X_d^i(E) \exp(-\lambda_i^k t) dE \]

where,

\( \phi \) = neutron flux,
\( N \) = number of fissile nuclei,
\( \sigma_{fk} \) = fission cross-section,
\( Y_i^k \) = cumulative fission yield in the \( i \)th precursor group for the \( k \)th fissile nuclide,
\( f_k \) = abundance factor of the \( k \)th fissile nuclide of which the fuel has \( M \) components,
\( \lambda_i^k \) = decay constant of the delayed neutron family (group) \( i \) for fissile material \( k \),
\( Y_n^i \) = yield of the \( n \)th precursor contributing to delayed neutron group \( i \),
\( \Lambda_n^i \) = disintegration constant of the \( n \)th precursor in the \( i \)th precursor group,
\( P_n^i \) = delayed neutron emission probability of the \( n \)th precursor in the \( i \)th group,
\( \Lambda_i^k \) = group abundance of the delayed neutron group \( i \) for the various fissile materials \( k \).

Alternatively, one can directly sum over the precursors and obtain the delayed neutron spectrum as:

\[ x_d(E,t) dE = \sum_{n} (\phi N \sigma^f Y_n P_n \Lambda_n) \exp(-\Lambda_n t) x_d^n(E) dE \]

The above basic procedures have been used to evaluate the six-group spectra and the gross energy spectrum under equilibrium condition at any time for a number of fissioning systems [See §8]. In practice, data by both the approaches (measurement and summation calculations) are needed to check for problems in the various measuring techniques and for comparing the measured composite spectra with the equivalent summation result. The current state of development is that the calculation of reactor physics parameters from \( P_n \) values, fission yields and spectra from separated nuclides has advanced to meet the
accuracy of data derived from experiments on aggregate ones.

Before closing this section, we want to point out that a number of calculations of delayed neutron energy spectra have been carried out using statistical model considerations [13-15]. Also, "analytic fits" to the experimental data have been made [16-18] using a constant $\beta^-$-strength function ($S_{\beta}$), proportional to either the level density ($q$) or given by the $\beta^-$ gross theory of $\beta$-decay [19,20]. Some of these results [14,15] showed a dramatic similarity of the overall spectra to the experimental spectra. It was, however, found that the model strength functions overestimated the neutron intensity above 0.3 MeV and underestimated at lower energies, and an analytical representation of the experimental $S_{\beta}$ gave a substantial improvement in the fit to the experiment. Mann et al [21] have used a statistical model with only one free global parameter, $a = \frac{N}{N+Z}$ ($a$ = level density parameter, $N$ and $Z$ are the number of neutrons and protons respectively in the daughter nucleus) to predict the beta decay properties ($\tau$, $E_x$, $E_y$, $E_z$, $P$) accurately and better than gross theory. The model, which is useful when many levels are involved, considered only levels in the daughter nuclide based on neutron single particle states. Figure C below compares the measured delayed neutron spectrum [16] with the model results for Rb-93. It is seen that while the fine structure observed in delayed neutron experiments is not predicted, the overall shape is well described, as is the probability of delayed neutron emission ($P_{\text{exp}} = 0.0137\pm0.0008$, $P_{\text{model}} = 0.0131$).

![Figure C: Delayed neutron spectrum of 93Rb.](image)

REFERENCES


§7. MEASUREMENT AND ANALYSIS OF DELAYED NEUTRON SPECTRA

Development in the measurements of delayed neutron energy spectra can broadly be divided into three stages. In the first phase, measurements of delayed neutron spectra were made with unseparated fission products. In the second phase, more precise data on delayed neutron spectra were obtained. These measurements showed not only greater structure but they also extended the energy range below 100 keV with an appreciable fraction of delayed neutrons in this low energy region. The third phase roughly pertains to the period following the 1979 Vienna workshop [1] and the 1983 Brookhaven meeting [2], during which the trend in the development of delayed neutron data has been to study delayed neutron emission from individual precursor nuclides. This approach, made possible through improvements in the experimental techniques of isotope separation and neutron spectroscopy, has become very productive during the last several years. The current state of development is that the calculation of reactor physics parameters from $P_r$ values, fission yields and spectra from separated nuclides has advanced to meet the accuracy of data derived from experiments on aggregate ones.

7.1 Phase 1

The first measurements of delayed neutron spectra were made by Burgy et al [3] followed by Hughes et al [4], Bonner et al [5], Batchelor and Hyder [6,7] and were limited to measurements on U-235. In each of these experiments, a sample was irradiated in a reactor for a predetermined time ($T_0$) and then transferred (via rabbit) to the measuring system and allowed to decay for a certain time ($T_d$) before recording started. In the measurements of Burgy et al, $T_d$ values varied from 0.5 to 50 sec and $T_0$ from 0.8 to 3.3 sec. Proton recoils were measured in a hydrogen-filled cloud chamber and spectra were observed. Energy resolution in the experiment was of the order of 100 keV, caused partly by poor statistics. The average energies lay in the region from 0.2-0.8 MeV. No simple relationship between energy and half-life was found. Figure 3 is the result of data graphically analyzed for the different groups and then combined to the steady-state distribution of the delayed neutron spectra. Hughes et al by varying $T_d$ from 1 s to 5 min and $T_0$ up to 5 min examined the attenuation of neutron intensity with distance in paraffin and compared the attenuation slopes to those derived from standard monoenergetic ($\gamma$,n) sources. Only mean neutron energies of some of the delayed neutron groups were derived. The resolution was of the order of 100 keV. Spectra were observed by Bonner et al by measuring proton recoils for $T_d = T_0$ = 1 sec. In Batchelor and Hyder's measurements, $T_d$ values ranged from 1.4 to 100 sec and $T_0$ values from 2 to 100 sec. Pulse heights were measured in a $^{3}$He-filled proportional counter and spectra were observed with a resolution of about 90 keV. In experiments where $T_d$ was varied, the data enabled experimenters to extract the spectra of the various half-life groups. Results of Batchelor and Hyder had the highest quality and are shown in figure 4. All the above measurements suffered from the following defects:

(a) Resolution being poor, it was not clear whether the spectra were really continuous with some superimposed fine structure (as obs-
Fig. 3: Equilibrium DN Spectrum from thermal-neutron fission of $^{235}$U obtained by Burgy et al.

Fig. 4: DN spectra from thermal neutron fission of $^{235}$U, measured by Batchelor and McK. Hyder and Bonner et al.

Resolution (full-width at half-height). Approximate Statistical Errors are indicated by Vertical Bars.
erved) or had a line structure.

(b) The magnitude of systematic effects from uncorrected background was uncertain. For example, whether the low- and the high-energy portions of the observed spectra were primarily due to the scattered prompt neutrons was not certain. The He^3 spectrometer in Batchelor and Hyder's experiments was particularly susceptible to scattered low-energy neutrons. Part of the apparent high-energy spectra might have been due to prompt neutrons arising from the delayed neutron caused fissions in the sample itself.

(c) The accuracy of separating the spectra into various groups was uncertain.

Later experiments by N.G. Chrysochoiades et al [8, pp. 213-227] and Chulik et al [9, 10] did not clear up the uncertainties either. These experiments, however, introduced the technique of neutron time-of-flight, triggering each flight-time measurement with the beta-decay detection from the precursor. The method involved coincidence measurement and had a poor signal-to-noise ratio caused by the intense accidental coincidence rate. The results were seen to differ markedly from the earlier four measurements in that a line structure was observed which was different from the previously observed continuous spectra. This was due to the improved resolution, which in Chulick's experiment varied from 6% at 100 keV to 20% at 500 keV. The observed distribution was also much different. For example, the concentration of neutrons at high energies (> 800 keV) did not agree at all. Figure 5 reproduces a spectrum from Chrysochoiades, figures 6 and 7 from Chulick. The spectrum in figure 5 was thought to be a mixture of spectra from the "traditional" groups 2 and 3. Figure 6 represents delayed neutrons from Cf252, which does not have the same groups as U235. This spectrum should correspond most closely to a mixture of the "traditional" groups 4 and 5. The structural spectrum in figure 6 includes eleven peaks from 95 keV to 430 keV. Figure 7 shows a comparison with the spectrum of Batchelor and Hyder which most closely corresponds to the results here.

Figure 8 shows the low energy combined spectrum of the mixture Br-87 and Br-88 obtained by Chrysochoiades et al [11] using a fast TOF beta-neutron coincidence technique which had excellent resolution at low energies (5% at 50 keV). The bromine isotopes from the fission products of U-235 were separated using a fast radiochemical technique. The structure observed in the spectra below 100 keV could possibly be due to the neutrons being emitted at discrete energies.

7.2 Phase 2

Since the early 1970s, the number of delayed neutron spectra measurements from fast- and thermal-neutron fission of different fissile isotopes has been steadily increasing. In 1972, Feig [12] measured spectra from the thermal fission of U-235 and from 14-MeV fission of U-235, U-238, Pu-239 using a proton recoil proportional counter. The spectra were measured for different time intervals and then resolved into four energy groups. The results for U-235 thermal fission superimposed on Batchelor and McK. Hyder spectra are shown
in figure 9 for four delayed groups and in figure 10 for the equilibrium spectrum. The agreement between the Feig measurements and the previous data is reasonable. The main difference is in the large amount of structure present in the Feig measurements as a result of his much better resolution, while the results of Batchelor and McK.Hyder are relatively smooth curves. Sloan and Woodruff [13,14] measured the delayed neutron spectra from thermal fission of U235, U233 and Pu239 using methane-and hydrogen-filled proportional counters; the first counter was for the 150 keV to 1.5 MeV interval and the second for 20 to 200 keV interval. Counting cycles of 4-, 12- and 25-s were used to enhance the different time groups. Their near-equilibrium spectrum (4-s cycle) is shown in figure 11.

Later, Eccleston and Woodruff [15,16] used the same technique to measure the near-equilibrium spectra from fast fission of Th-232, U-233, U-235, U-238 and Pu-239 using the Be9 (p,n) B9 reaction with 10 MeV protons as the neutron source. The Eccleston and Woodruff spectra for U-235 are reproduced and compared to several other measurements in figure 12. The results of Sloan and Woodruff, Eccleston and Woodruff are in good agreement with other measurements above about 200 keV. But, below 125 keV, these measurements show a significant increase in the amount of delayed neutrons emitted with several distinct peaks. However, the spectra at lower energies were not well supported by other measurements. Shalev and Cuttler [17] found a strong rise in the intensity of the delayed neutron spectra below 200 keV with a strong component at 120 keV, but were unable to obtain meaningful information below 200 keV because of the rapidly increasing detector efficiency at low energies and the deleterious effect of the high γ-ray counting rate. Using the Shalev He spectrometer, Evans and East [18] measured equilibrium delayed neutron spectra from fast fission of U235. In the experiment, 2.15 MeV protons were accelerated on a lithium target producing neutrons of energies between 30 and 420 keV. Their spectrum had a complex line structure as observed by Shalev [17] and by Sloan and Woodruff [13,14]. Neutron energy peaks corresponded to peaks previously observed. The overall shape of the spectrum, however, corresponded more closely to that measured by Batchelor and HYDER than that of Sloan and Woodruff. Evans and Kricks [19,20] followed a procedure similar to that of Evans and East to measure the spectra of equilibrium delayed neutrons from fast fission of U-235, U-238 and Pu-239. Energy resolution of the spectra varied from 16 keV for thermal neutrons to 54.3 keV for 1.5-MeV neutrons. The data, shown in figure 13, were similar to the spectra previously observed. The average energies were about 500 keV, somewhat in disagreement with the earlier results. The spectrum intensity below about 300 keV was found to be very sensitive to the wall-effect subtraction carried out by them, and hence, part of the average-efficiency difference could be experimental error.

Shalev and Cuttler [17,21,22] measured the delayed neutron spectra for Th-232, U-233, U-235, U-238 and Pu-239 using a new type of He-3 spectrometer and the IRR reactor as the neutron source. After irradiation, samples were transferred to the He3 counter by a pneumatic rabbit. By appropriate variations of the irradiation and the counting time and by recording the data as a function of time.
Fig. 9: Delayed neutron spectra from Group 1 through 4 from thermal neutron fission of $^{235}U$.

Fig. 10: Equilibrium delayed neutron spectra from thermal neutron fission of $^{235}U$, measured by Feig.

Fig. 11: Near-equilibrium delayed neutron spectrum from thermal neutron fission of $^{235}U$ (4-S cycle).

Fig. 12: Comparison of measured delayed neutron spectra from fast neutron fission of $^{235}U$ near equilibrium.
Fig. 13: Equilibrium spectra of delayed neutrons from fast fission of $^{235}\text{U}$, $^{238}\text{U}$ & $^{239}\text{Pu}$. 
delayed neutron groups 2 and 4 were accentuated. In figure 14, the delayed neutron spectra from fast fission of U-235 are shown. The upper and lower parts in the figure represent counting during a long cycle for different periods of time. It is seen that the different delayed neutron groups are emphasized. By neglecting the contribution of the third group and by appropriate pealing, Shalev and Cuttler were able to obtain delayed neutron spectra for groups 1 and 2 in the energy interval of 100 to 1200 keV. These data were in good agreement with the measurements of Batchelor and McK. Hyder, and with Feig's measurements, but data below 100 keV could not be produced as reported by Sloan and Woodruff and by Eccleston and Woodruff.

Recently, measurements of composite delayed neutron energy spectra from the thermal-and fast-neutron fission of U-235 have been reported by Tanczyn, Sharfuddin et al [23,pp.707-711;24,25] for eight delay-time intervals between 0.17 and 85.5 s. The experimental technique combined a helium-jet and tape transfer system with a beta-neutron time-of-flight spectrometer. The neutron energy range was from 10 keV to 2.0 MeV and was spanned with Li6-glass, plastic and liquid scintillators. Selection of this energy range enabled the experimenters to make measurements in the energy region of interest, namely, $E_\gamma < 200$ keV and $E_\gamma > 800$ keV. In the measurement, each delay-time was given equal sensitivity weighting. With this delay time, the measured spectra contained more than 50% of the combined contributions from groups 5 and 6 which were the least well known. Although this time range covered 90% of the total delayed neutrons emitted, the measurements actually encompassed only 78% due to the small gaps between the successive time intervals. Of the total 12% of delayed neutrons thus lost, 7% were lost in the interval less than 0.17 s and 3% in the interval greater than 85 s. Small gaps between the measured intervals accounted for the remainder of the missing yield. It should be noted that sensitivity to very short delay times is important since about 30% of all the delayed neutrons are emitted within 1 s following fission. The experiment combined not only high detection efficiency with good energy resolution but also minimized multiple neutron interactions and random background events. The energy resolution ($\Delta E/E$) was less than or equal to 10% for the Pilot U and the Bicron measurements, and 15% for the Li6-glass measurements. For Pilot U and BC 501, this resolution was maintained over the entire neutron energy range by correcting for the TOF spectra.

Typical TOF delayed neutron spectra are shown in figure 15. The top one was obtained using the Pilot U plastic, the middle one using the BC 501 liquid and the bottom one using the NE-912 Li-glass scintillators. Each TOF spectrum is seen to consist of a broad delayed neutron peak from beta-neutron coincidences, a sharp gamma-ray peak from beta-gamma coincidences, and a nearly isotropic random background arising primarily from chance coincidences between the betas and the gamma rays. The shoulder on the gamma-ray peak in the Pilot U and BC 501 measurements was due to the Compton back-scattering mainly from the structural materials in the counting room. The TOF spectrum was analyzed in three steps using the computer code DENTS [26] which is of general utility for the analysis of continuous neutron TOF spectra.
Fig. 14: DN spectra from fast neutron fission of $^{235}$U for two counting times.

Fig. 15: The TOF spectra measured in two delay-time intervals following the thermal neutron fission of $^{235}$U. Neutron energies (in kilo-electron-volts) are indicated with arrows.
Composite delayed neutron energy spectra for the eight delay time intervals are shown in figures 16 and 17. The total counts in each spectrum were normalized to $10^7$. The energy bin size of 10 keV roughly approximated the energy resolution. Systematic uncertainty in the background was much less than 3%. The uncertainty in the 10 keV yield was estimated to be 100%. For purposes of comparison, composite spectra were constructed from the six-group spectra of the ENDF/B-V compilation [27] as well as from Rudstam's compilation [28] for U-235 based on the Studsvik measurements of delayed neutron spectra for individual precursors. These comparison spectra, also normalized to $10^7$, are shown as dotted curves in the figures.

The spectra show a smooth evolution from short to long delay times with the ones at the two extremes displaying the most dissimilar characteristics. Comparison of the measured spectra with those generated from ENDF/B-V and Rudstam show the approximate overall agreement and the regions of differences. The ENDF composite delayed neutron spectra change little with delay time, but this is not so with the measured spectra. The reason for this is partly due to the groups 4, 5, and 6 spectra being identical in the compilation. Further, the low energy peak and the high energy cut-off in the ENDF spectra appear to be quite artificial. In contrast to ENDF, the composite delayed neutron spectra derived from the individual precursors data of Rudstam evolve in a manner quite similar to the present work. In particular, the agreement is close for the three intervals between 2.1 and 29 sec. The authors have stated that for delay times less than 2 s, the low-energy structure in Rudstam's spectra might be slightly exaggerated due to the incomplete nature of the individual precursor data bases, particularly for groups 5 and 6.

A delayed neutron equilibrium spectrum from thermal fission of U-235 was calculated from the composite spectra measurements by two methods: the direct integration method and the matrix inversion method. In the direct integration method, the full range of delay times in the composite delayed neutron spectra was spanned and the delayed neutron fractions as a function of delay time were used. In the matrix inversion method, only the six-group representation of delayed neutron spectra was used. Spectra at every delay time interval was unnecessary since a mathematical solution exists for any set of six independent delayed neutron spectra measurements. The two methods yielded nearly indistinguishable equilibrium spectra for a given set of $(\beta_i, \lambda_i)$. Figure 18 shows the spectra generated using the direct integration method and two sets of $(\beta_i, \lambda_i)$. The solid curve at the top was calculated using the $(\beta_i, \lambda_i)$ set from ENDF/B-V and the solid curve at the bottom employed the set from Rudstam. In the figure, the dotted curve at the top was generated from the ENDF/B-V group spectra and the one at the bottom from Rudstam's group spectra. It is seen that the agreement in the overall shape is satisfactory. The two main discrepancies with ENDF are its low-energy peak and its high-energy cut-off. The Rudstam spectrum has a somewhat fewer high energy neutrons but differs mainly in the persistence of the low energy peaks. The effect of these differences was found to decrease the average energy of the equilibrium spectrum by about 50 keV compared to the measured values.
Fig. 16: Composite neutron energy spectra measured by Tanczyn et al. (solid curves) following thermal neutron induced fission of $^{235}$U, compared with the spectra generated from ENDF/B-V (dotted curves) for the same time intervals.
Fig. 17: Composite DN energy spectra measured by Tanczyn et al (solid curves) compared with the spectra generated from the six-group spectra of Rudstam (dotted curves) for the same time intervals.
Fig. 18: Equilibrium spectra generated by Tanczyn et al. using the direct integration method (solid curves) compared with those generated from the six group spectra and $(\beta_i, \lambda_i)$ parameter sets from ENDF/B-V (TOP) and Rudstam (BOTTOM).
A search was made to find any changes in the composite delayed neutron spectra caused by a change in the energy of the neutrons inducing fission in U-235 [29]. The search was performed over the eight successive delay time intervals with the mean energy of fast neutrons at 1.8 MeV. The eight fast minus thermal delayed neutron difference spectra indicated no measured changes of delayed neutron spectra between thermal-and fast-induced U-235 fission. This result was consistent with the calculated difference spectra from England's compilation. It was, however, noted that the delayed neutron spectral differences could be substantially greater for a much harder neutron spectrum such as in the deuterium-tritium fusion process, where second-and third-chance fission modifies the yield of delayed neutron precursors.

Figure 19 shows the measured composite delayed neutron energy spectra following thermal neutron induced fission of Pu239 for four delay time intervals together with the corresponding ones from U-235 fission by Couchell et al [30,pp.215-241]. The spectra are seen to be very similar for short delay times, but show an increasing difference for longer delay times. The average neutron energies of the Pu239 delayed neutron spectra were approximately 10 keV lower than those of U235 for the two shortest delay intervals, and 40 to 50 keV lower for the two longest delay intervals. This trend differs somewhat from the results of composite measurements reported by Kratz and Gabelmann [23,pp.661-673] who found that the shapes and average energies of U-235 and Pu-239 delayed neutron spectra were quite similar for delay times ranging from about 1 to 50 seconds.

Atwater, Goulding et al [30,pp.255-274] have measured delayed neutron spectra from individual short pulse (about 50 μsec) fission of small U-235 samples using a small NE 213 proton recoil neutron spectrometer and the Godiva fast-burst reactor as the neutron source (fast neutron flux ~ E+13 n/cm²). Data were acquired in sixty-four 0.5 s time bins and over an energy range of 1-7 MeV where very little is known about the delayed neutron spectra. The unfolding technique used to obtain the energy spectrum from the pulse-height distribution was the "least-squares" method of Cook [31]. The unfolded data started at 1 MeV since the threshold for the neutron detector was 700 keV. Figure 20 shows a typical delayed neutron energy spectrum for time bin of 5-10 sec together with the calculation of England et al [30,p.270]. The early time data suggest a distribution that falls less steeply with energy than the calculations indicate.

7.3 Phase 3

During the last one decade or so, following the 1979 Vienna workshop and the 1983 Brookhaven meeting, the trend in the development of delayed neutron data has been to study delayed neutron emission from individual precursor nuclides. This approach has become possible through improvements in the experimental techniques of isotope separation and the neutron spectroscopy. In their reviews, S. Amiel et al [8,pp.115-145] presented a list of thirty-seven isotopes, Del-Marmol [32] of twenty-five isotopes and Tomlinson [33] of forty-five together with their half-lives and emission probabilities (Pᵣ). These isotopes were the actual physical...
Fig. 19: A comparison of composite DN energy spectra from thermal neutron induced fission of $^{239}$Pu (solid curve) and $^{235}$U (dotted curve)
Fig. 20: Neutron spectrum (5-10 seconds after Godiva burst)
precursors of the delayed neutrons.

Measurements of delayed neutron spectra for separated precursors have been made \([11,30,34-48]\) at a number of laboratories using a variety of techniques: on-line mass spectrometry, He-3 spectrometry, proton-recoil technique, the time-of-flight method and the neutron scintillator. The basic requirement in measuring the spectra of an individual precursor is to have some fast method of separation of the particular fission product after the fissile sample is irradiated. These measurements indicate considerable variation in the quality (statistical accuracy and resolution) of the spectra depending upon the technique and the laboratory doing the work. They also indicate that while the overall shapes and peak structures can be reproduced by different laboratories using the same or different techniques, the relative intensities of spectra at energies below 200 keV are sensitive to the data processing techniques and the experimental environment. Data analysis procedures are yet to be standardized.

Reeder and Warner have developed a ring ratio technique \([49]\) to estimate the average energies of delayed neutrons from Br-87-Br89, Rb92-Rb98, I-137 and Cs141-Cs147 precursors from the ratio of counts in rings of counter tubes embedded in different thicknesses of polythene moderator \([42,pp.239-263;50]\). The technique has the advantage that it requires relatively little measuring time and has no cut-offs at high or low energies.

In figures 21 and 22, we present the neutron energy spectra of the four nuclides (As-85, Br-87) and (Sb-135, I-137) measured after rapid chemical separation of the individual precursors \([38,51,52]\). These nuclides represent characteristic pairs in the light and heavy mass regions in fission, the member of each pair differing only by a pair of protons. The corresponding neutron emitters have \(N=51\) and \(N=83\). Consequently, the nuclides lead to nuclei with major closed shells after neutron emission. For the nuclides Br-87 and I-137, the neutron decay proceeds entirely to the ground state of the final nucleus where as for As-85 and Sb-135, the large energy windows \((Q_{d,n})\) result in neutron emission to several excited states. In the cases of Br-87 and I-137, the maximum neutron energies of about 1.3 MeV and 1.7 MeV respectively are in agreement with the energy range \((Q_{d,n})\) available for neutron emission. A dominant feature of the spectra from As-85 and Sb-135 is the absence of appreciable neutron intensity at high energies. For As-85, the intensity above 1.6 MeV is less than 3% of the total and for Sb-135 the intensity above 2.2 MeV is less than 4% of the total. This is despite the fact that ranges of 4.9 MeV (As) and 4.2 MeV (Sb) are possible. Through \(\gamma\)-ray studies, it has been demonstrated that the effect is due to strong neutron emission to excited final states \([11,51,53]\).

The neutron energy spectra shown in figures 23 and 24 are from isotopes-separated precursors Rb92-Rb98 \([54]\). They have the following main features:

(i) Only little high-energy neutron intensity even if energy ranges upto 5.8 MeV (Rb98) are available for neutron emission.

(ii) A systematic variation, within the isotope sequence, of the peak structure and the overall spectrum shape with the mass number.
Figure 23: Delayed neutron spectra from $^{95}$As.
Fig. 22: Delayed-neutron spectra from $^{133}\text{Sb}$ and $^{137}\text{I}$ decay.
Fig. 23: Delayed-neutron spectra from the decay of $^{92}\text{Rb}$, $^{94}\text{Rb}$, $^{96}\text{Rb}$ and $^{98}\text{Rb}$ precursor nuclides.
Fig. 24: Delayed-neutron spectra from the decay of $^{93}\text{Rb}$, $^{95}\text{Rb}$, $^{97}\text{Rb}$ nuclides. Bottom figure to the right is the pulse height distribution of $^{95}\text{Rb}$ delayed neutrons.
(iii) Neutron spectra of odd mass Rb precursors decaying into even-even Sr nuclei exhibit prominent line structure and account for the majority of the total neutron intensity. A sudden drop in neutron intensity above about 850 keV is observed.

(iv) Even-mass Rb precursors decaying into odd-mass Sr final nuclei show a larger continuous neutron distribution superimposed by only a few strong peaks which contain less than 20% of the total neutron intensity. The main neutron intensity is concentrated at energies below 400 keV.

Experiments have shown that the above variation in the shape of the spectrum and the absence of appreciable high-energy neutron intensity are due to the strong neutron emission to excited states in the Sr final nuclei. Figure 25 shows the decay of Rb93-Rb95 to levels in Sr92-Sr94. It is clearly seen that in the case of the even-even final nuclei Sr-92 and Sr-94, neutron emission can lead to only a few widely-spaced excited states, whereas in the case of the final nucleus Sr-93, a total of about 30 narrowly spaced levels at excitation energies up to 3 MeV are available for neutron decay. Thus many neutron branches with comparable intensities superimposed lead to the complex neutron energy spectrum of the even-mass precursor Rb-94, with only a few strong well-resolved neutron lines. This "odd-even effect" in neutron spectrum shapes is also observed for the bromine, iodine and cesium isotope sequences.

Using the detection and analysis philosophy described earlier in (7.2), Atwater et al carried out spectral measurements of separated isotopes. Figure 26 shows the results of preliminary analysis for Rb-97 with the experimental data normalized to the theoretical calculation. A steeper fall-off with neutron energy than theory is seen.

7.4 Comparison of Spectra [2]

Table 7 shows three data sets for each of the Br, Rb and Cs

<table>
<thead>
<tr>
<th>Source</th>
<th>Laboratory</th>
<th>Spectra</th>
<th>Detector</th>
</tr>
</thead>
<tbody>
<tr>
<td>K.L.Kratz</td>
<td>Mainz</td>
<td>$^{87}$Br, $^{93}$Rb, $^{94}$Rb, $^{95}$Rb, $^{143}$Cs</td>
<td>$^3$He</td>
</tr>
<tr>
<td>P.Ray</td>
<td>Penn. State</td>
<td>$^{87}$Br</td>
<td>Proton Recoil</td>
</tr>
<tr>
<td>G.Rudstam</td>
<td>Studsvik</td>
<td>$^{87}$Br, $^{93}$Rb, $^{94}$Rb, $^{95}$Rb, $^{143}$Cs</td>
<td>$^3$He</td>
</tr>
<tr>
<td>P.L.Reeder</td>
<td>Pacific North West Laboratory</td>
<td>$^{93}$Rb, $^{94}$Rb, $^{95}$Rb, $^{143}$Cs</td>
<td>$^3$He</td>
</tr>
</tbody>
</table>
Fig. 25: Neutron emission to excited states in $^{92-94}$Sr
Fig. 26: $^{97}$Rb pulse height spectrum.
precursors for comparison [43]. In all the three data sets, much of the peak structure and overall intensity trends were reproduced except for the Studsvik data where below 200 keV, the intensities were generally higher. The intensities of the individual peaks depend to some extent on the detector resolution and also on the laboratory where the measurement is made. In figure 27 for Rb95, the Mainz spectrum has a very intense peak at 13.7 keV, the area under the peak corresponding to 10% of the area between the limits of 100 to 1100 keV. The same peak is observed in the Reeder's spectrum but the intensity is only 4% of the intensity between 100 and 1000 keV. This illustrates the sensitivity of the low energy region to details of data processing. For Br87 (figure 28), there is reasonable agreement as to the peak positions and relative intensities for the two spectra measured with the He detectors. However, the spectrum measured with a proton recoil detector has much greater intensities at low energies although the peak positions are reproduced well. The average energy for the spectrum measured with a proton recoil detector is about 140 keV whereas the He spectrometer data give average energies of about 220 keV. The average energy for Br87 measured by the ring-ratio technique gave a value of about 150 keV which tends to support the proton recoil result.

7.5 Principal Detection Methods [2,30,42]

The principal detection methods employed in the measurements of the energy spectra of β-delayed neutrons from fission products in the energy range of 0.01 MeV to about 4 MeV are: proton-recoil spectrometry, Time-of-flight measurements and the 3He(n,p) spectrometry. We shall now discuss each of these techniques in brief.

7.5.1 Proton-recoil Spectrometry

Very clean measurements of fast neutron energy spectra can be performed with recoil proton telescopes. In these spectrometers, the energy and direction of the proton from a hydrogen-rich material are measured with scintillators or semi-conductor counters. Using pulse shape discrimination in a CSI (Tl) proton detector, it is possible to measure neutron spectra down to very low energies. However, the technique has a very low efficiency (about 10⁻⁵) and because of this, it is not commonly used in β-delayed neutron spectrometry.

The conventional proton-recoil system uses methane or hydrogen-filled proportional counters at several atmospheres pressure and γ-ray discrimination via pulse-shape analysis [55]. Using this system, time-dependent neutron spectra from the thermal-neutron fission of U-235 were measured by Sloan and Woodruff [13,14]. Later, Eccleston and Woodruff [15,16] used it to measure the two-parameter data of near-equilibrium delayed neutron spectra produced by fast-neutron induced fission of different fissile isotopes. The main problem in analyzing proton-recoil data consists in the transformation of the registered two-parameter data to one-parameter proton-recoil distributions which are then differentiated using the PSNS code [see Report ANL-7394, Jan. 1968, 130p] to give the neutron spectra. Accuracy of neutron spectra determination requires that corrections are made for: the energy loss per ion pair, the neutron scattering on heavy
Fig. 27: Comparison of $^{95}$Rb delayed-neutron spectrum measured at different laboratories.
Fig. 28: Comparison of delayed-neutron spectrum for $^{87}$Br measured at different laboratories.
nuclei (carbon, nitrogen), the non-ideal electric-field effects and for wall-and-end effects. It has been found that the (near-) equilibrium delayed neutron spectra obtained by proton-recoil spectrometry show considerable differences in the energy distributions, especially in the low-energy portion of delayed neutrons, when compared to He-spectrometry neutron energy spectra.

Ray and Kenney [39] used a methane filled proton-recoil proportional counter of the above type to measure the delayed neutron spectrum from Br-87 beyond 100 keV. Major peaks were observed at 131 and 192 keV and much smaller peaks at 261, 337, 420 and 520 keV. The observed spectrum was significantly softer compared to the previously reported data. Greenwood and Caffrey [2,48] reported on measurements of the energy spectra of delayed neutrons for the isotope-separated fission-product precursors Rb93-Rb97 and Cs143-Cs145 over the energy region from about 10 to 1300 keV using gas-filled proton-recoil proportional counters at the TRISTAN ISOL facility. These measurements showed systematic deviations in intensities between the INEL data and the other reported data sets in the neutron energy region above about 800 keV. Subsequently [30, pp.199-214], they carried out a series of measurements using the same experimental set up to obtain additional delayed neutron spectral information at these higher energies and below about 20 keV using high pressure CH gas-filled proportional counters and a liquid scintillation counter. Tables 8 and 9 summarize the delayed neutron spectral intensity distributions obtained by combining the present data with their earlier data [2,48]. Quantitative intercomparison of the present data with the previous data measured using He3 ionization chambers is given in Table 10 for the widely studied Rb95 isotope. It is seen that while the most recent INEL data and the Rudstam data [2] appear to be reasonably consistent in the energy region above 770 keV, the present INEL neutron intensities fall off much faster than those reported by Kratz [2].

7.5.2 Fast Neutron Time-of-flight Spectrometers

In principle, the best method for precise measurements of fast neutron energy spectra is the TOF technique. Through the developments of accelerators capable of delivering beam pulses of good quality, of large organic scintillation detectors, and of fast electronics, the energy resolution in TOF neutron spectroscopy has the same quality as that obtained for charged particles. The method is basically very simple and consists in measuring the flight time of a neutron ejected from a target over a distance and then calculating the neutron energy from it. The start pulse is taken from a pick-up coil in front of the target or from an associated particle emitted in the reaction. The stop pulse is taken from the neutron detector. In the case of delayed neutrons, the associated particles emitted are the β-rays.

The energy resolution ΔE is determined by the time uncertainty Δt, since the flight distance (l) can be measured with high precision. The expected energy resolution is given by [56]:

ΔE = Δt \cdot l
Table 8: Delayed neutron spectral intensities (relative) for the Rb precursor isotopes.

<table>
<thead>
<tr>
<th>Neutron energy range (keV)</th>
<th>Relative Neutron Intensity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{93}$Rb</td>
</tr>
<tr>
<td>E &lt; 11.6</td>
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</tr>
<tr>
<td>11.6 - 19.0</td>
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</tr>
<tr>
<td>19.0 - 31.1</td>
<td>1.4</td>
</tr>
<tr>
<td>31.1 - 45.0</td>
<td>1.1</td>
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<tr>
<td>45.0 - 65.2</td>
<td>1.9</td>
</tr>
<tr>
<td>65.2 - 83.5</td>
<td>2.1</td>
</tr>
<tr>
<td>83.5 - 106.8</td>
<td>4.4</td>
</tr>
<tr>
<td>106.8 - 136.7</td>
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<td>136.7 - 175.6</td>
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<td>175.6 - 224.1</td>
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<td>13.4</td>
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<tr>
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<td>13.3</td>
</tr>
<tr>
<td>470.0 - 601.7</td>
<td>12.1</td>
</tr>
<tr>
<td>601.7 - 770.2</td>
<td>9.7</td>
</tr>
<tr>
<td>770.2 - 985.9</td>
<td>3.5</td>
</tr>
<tr>
<td>985.9 - 1262.0</td>
<td>1.0</td>
</tr>
<tr>
<td>1262.0 - 1615.5</td>
<td>0.3</td>
</tr>
<tr>
<td>1615.5 - E</td>
<td>0.02</td>
</tr>
<tr>
<td>2068.0 - 2647.2</td>
<td>0.04</td>
</tr>
<tr>
<td>2647.2 - 3150</td>
<td></td>
</tr>
</tbody>
</table>

$E_c$ (cut-off energy) = $13.8 (^{93}\text{Rb}), 8.0 (^{94}\text{Rb}), 7.1 (^{95}\text{Rb}), 8.0 (^{96}\text{Rb}), 7.1 (^{97}\text{Rb})$

$E_u$ (keV) = $1971.3 (\text{Rb93,Rb96}); 2017.0 (\text{Rb97}); 2068.0 (\text{Rb94,Rb95})$

$AE = 2E (\Delta t/t) = 0.028 \ (\Delta t/\lambda) E^{1.5}$

However, the real resolution in the experiments includes the energy spread of the beam and the thickness of the target. The TOF technique is the only spectrometer type where the energy resolution is not an inherent factor of the system. It can essentially be defined by the experimenter. Crawford et al [57] obtained the TOF energy spectrum of delayed neutrons from Rb95 using plastic and lithium-glass scintillators in the energy range of 10 to 1200 keV. The spectra showed good overall agreement with the spectra obtained from the $^3\text{He} (n,p)$ spectrometry. Analysis of the low-energy part of the Rb95 spectrum from the lithium glass detector showed, in agreement with the results of the Mainz-group who used a $^3\text{He}$ ionization chamber, that there was a prominent group of delayed neutrons at 13.7±0.2 keV. The width of this group was about 0.9 keV. The estimated energy resolution was 0.2 keV at the main peak and 0.5 keV at the 25.5 keV peak. There were also two significant peaks at 11.2 keV and 25.5 keV with a width of about 2 keV. In addition, there were numerous other peaks over the whole energy range. Later, Crawford and Kellie [30,pp.299-308] carried out measurements using
Table 9: Delayed neutron spectral intensities (relative) for the Cs precursor isotopes.

<table>
<thead>
<tr>
<th>Neutron energy range (keV)</th>
<th>(^{143})Cs</th>
<th>(^{144})Cs</th>
<th>(^{145})Cs</th>
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</tr>
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<td>2.9</td>
</tr>
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<td>4.9</td>
<td>3.2</td>
</tr>
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<td>4.6</td>
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<tr>
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<td>11.0</td>
<td>8.9</td>
<td>8.6</td>
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<td>7.3</td>
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<td>3.1</td>
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<td>1.7</td>
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</tr>
<tr>
<td>1615.5 - 2017.0</td>
<td>0.001</td>
<td>0.1</td>
<td>0.3</td>
</tr>
</tbody>
</table>

\(E_c\) (cut-off energy) = 10.2, 10.2 and 7.8 keV \(^{143-145}\)Cs respectively.

Table 10: \(^{85}\)Rb - Comparison of \(^3\)He and proton recoil results

<table>
<thead>
<tr>
<th>Neutron energy range (keV)</th>
<th>(^3)He Spectrometer</th>
<th>Proton-Recoil Detector</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Rudstam</td>
<td>Reeder</td>
</tr>
<tr>
<td>0 - 39.8</td>
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<td>2.4</td>
</tr>
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<tr>
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<td>0.3</td>
</tr>
<tr>
<td>2647.2 - 3386.6</td>
<td>0.3</td>
<td>0.3</td>
</tr>
</tbody>
</table>
the fission product mass separator OSTIS at the ILL, Grenoble. Beta-particles from the decay of fission products were detected by a small plastic scintillator. Two neutron detectors, the NE 213 liquid scintillator and the NE 912 lithium glass scintillator were used. For Rb-94 and Rb-95, flight paths were 2.5 m and covered the energy range from about 160 keV to 10 MeV. With the glass scintillator, short flight paths of 30 to 45 cm were used in order to cover the energy region below 100 keV. In figures 29 to 31, some of their results are presented. The errors shown in the figures are purely statistical based on the spectra themselves. No account was taken of the possible uncertainties in the efficiency corrections. The results for Rb-94 taken with the NE 213 liquid scintillator are shown in figure 29 and compared to those of the Mainz group who used 3-He spectrometers. No normalization was included, so the intensity scales are arbitrary. The agreement is generally good. The results in figure 30 show little evidence of significant yields above 2 MeV. Figure 31 shows the low-energy part of the delayed neutron spectrum for Rb-95 taken with the Li glass detector. The well-known low energy peak at about 14 keV is seen.

7.5.3 3He-Spectrometers

In the spectroscopy of neutrons in the energy range of 0 to 3 MeV, a He3 gas proportional counter provides the best combination of energy resolution and detection efficiency [58]. But 3He-spectrometry is not always a straightforward technique. With the He3 (n,p) detection method, the energy spectra of delayed neutrons from fission have been studied both from fissionable material and from separated fission products [59]. Thirty-three individual precursors have been measured which include precursors of primary interest in nuclear technology. However, for nearly fourteen of these spectra, the low energy part up to about 100 keV is lacking and there is a general discrepancy in the energy/intensity distributions between the OSIRIS and the Mainz neutron spectra. These discrepancies call for detailed studies of the He3-spectrometer characteristics with particular emphasis on detector resolution, efficiency and spectrum distortion effects.

The latest versions of the 3He spectrometers are cylindrical gridded ionization chambers with guard tubes to reduce fringing fields [17,60]. Neutrons are detected through the reaction

\[ ^3\text{He} + n \rightarrow p + ^3\text{He} + 763.8 \text{ keV} \]

which has a smooth and well measured cross-section in the energy range of the delayed neutrons. Competing reactions with comparative cross-sections are He3 (n,n') He3 and H1 (n,n') H1. The spectrometers are filled with a gas mixture consisting mainly of 3-He, Ar and Methane. They are surrounded by thermal neutron shields which reduce the thermal neutron count rate by a factor of about 50 to 20. The energy calibration of the He3 spectrometers and their response function to mono-energetic neutrons are usually obtained using neutrons from the 7Li (p,n) Be reaction produced with Van de Graaff accelerators. Scattered neutrons from the detector, the
Rb-94
Crawford and Kellie

Kratz et al

Figure 29

Rb-95 (2.5m flight path)
Background chosen to give zero average counts from 7 to 30 MeV

Rb-95 (Li glass spectrum)

Figure 30

Figure 31

NEUTRON ENERGY (MeV)

RELATIVE INTENSITY CORRECTED FOR EFFICIENCY

NEUTRON ENERGY (MeV)
surrounding material and the wall effects give a continuous pulse
distribution between the fast and the thermal neutron peak. Of major
influence are the recoil effects in the elastic scattering of
neutrons with protons and the He-3 nuclei. Because the Q-value of
the He3 (n,p) H3 reaction is 763.8 keV, this recoil effect shows up
beyond the thermal neutron peak only for neutron energies greater
than about 1.0 MeV.

The response function is determined with the aim of unfolding
experimental neutron pulse-height spectra [42,61-63]. The technique
used in some of these works utilizes a combination of pulse height
and rise time discrimination against scattering events in the
detector. This results in a significant decrease in efficiency since
for high energies a major fraction of the response appears below the
full energy peak. Beimer et al [64] have measured and fitted the
whole detector response in the energy range of 130-3030 keV. The
fitting was done with regard to the fast neutron peak, the wall
effects and recoil distributions of elastically scattered protons
and helium nuclei. This makes the total intensity of the
experimental spectrum useful for evaluating complex neutron spectra.
This is specially important for high energies where the major part
of the response is found in the low energy tail. The sixteen-
parameter response function was:

\[
R(E,E_n) = \left[ A_{17} (E-A_{18}) + A_{19}\right] \left[ 1 + \exp\left(\frac{(E-A_{18})}{A_{15}}\right) \right] \\
+ \left[A_{11} (A_{13}-E)+A_{12}\right] \left[ 1 + \exp\left(\frac{(E-A_{13})}{A_{14}}\right) \right] \\
+A_6 \exp\left[\frac{-((E_n+764)-E)}{A_7}\right] \text{ERFC} \\
+A_4 \exp\left[\frac{-((E_n+764)-E)}{A_5}\right] \text{ERFC} \\
+\delta_1 A_1 \exp \left[-(1/2)\left(\frac{(E_n+764)-E}{A_3}\right)^2\right] \\
+\delta_2 A_8 \exp \left[-(1/2)\left(\frac{(E_n+764)-E}{A_9}\right)^2\right]
\]

with \( \delta_1 = 1 \) and \( \delta_2 = 0 \) for \( E < (E_n+764) \)
\( \delta_1 = 0 \) and \( \delta_2 = 1 \) for \( E > (E_n+764) \)

and the energies are in keV. The peak energy \( (E_n+764 \) keV)
corresponds to the fit parameter \( A_n \). The function \( R \) is valid for
neutron energies \( E_n \) in the range 100 to 3100 keV and for
absolute energies \( E \) from 150 keV. A least-squares fit using the
algorithm of Marquardt [65] which combines a gradient search with an
analytical solution developed from linearization of the model
function is used. The fitting was done by minimizing the chi square.
To predict the shape of the detector response for any neutron
energy, the parameters were determined by linear interpolation
between the tabulated parameter values given in ref.65. Conclusions
drawn from the study are:

(i) The \(^3\)He spectrometers can be used with confidence for neutron
energies up to at least 3000 keV. Care must, however, be taken
with the interpretation at low energies due to the unexpected structure.

(ii) The peak structure of the beta-delayed neutron spectrum was caused by the \(^{3}He\) spectrometer itself;

(iii) Scattering from the detector wall is a possible source for the distorted detector response;

(iv) The appearance and energy of the "ghost peaks" is due to back-scattering in the detector wall.

Ohm and Kratz [30, pp. 175-198] have developed a computational technique for response function analysis that removes the unwanted side effects in the neutron spectra without disturbing the efficiency function. The method takes into account all the relevant physical processes in detail and a relatively simple description of the detector response is derived instead of many-parameter polynomial fitting routines [64, 66, 67]. Though the analysis method was demonstrated for the well-structured \(\beta dn\)-spectrum of the precursor \(1.26\ s\ K49\), the response correction is applicable to \(\beta dn\) spectra measured under different experimental conditions and geometries provided minor adjustments to the magnitude of the wall effect are applied.

The wall-effect magnitude was calculated as a simple function of the total energy as:

\[
P_{w,\text{tot}}^{\text{exp}}(E_t) = P_{w,\text{tot}}^{\text{exp}}(P_1.E_t)
\]

where, \(P_{w,\text{tot}}^{\text{exp}}(E_t) = 1 - \frac{a_1}{(a_1 + a_2 E_t + a_3 E_t^2)}\) and \(a_1, a_2, a_3\) are the parameters of the parabola that were fitted to the data points. \(E_t\) is the total energy carried by the products of the reaction \(He3(n,p)H3\). \(P_1\) is a parameter that is determined for each standard spectrum.

The proton recoil plateau was described by:

\[
N_H(E_r) = [4p(CH_4). \sigma_{n,n}^H.(1-P_{w,\text{tot}}^{H}(E_r)) \Delta E] [N_n,p(E_n).P_2]/[p(^3He)].
\]

Here \(p\) denotes the partial gas pressure, \(\sigma_{n,n}^H\), the scattering cross-section of neutrons by protons, \(E_r\) the \(^n\text{p}\text{ recoil energy and } \Delta E\) the channel width. \(P_2\) is a parameter and \(N_r\) is the number of proton recoil events.

The \(^3\text{He}\) recoil distribution was described by:

\[
N_{He}(E_r) = [16\pi(1-P_{w,\text{tot}}^{He}(E_r)). \Delta E.N_n,p(E_n).P_4 . \frac{d\sigma/d\Omega(E_n,Cos \theta)}{[3E_n. \sigma_{n,p}(1-P_{w,\text{tot}})]}
\]
The $^3\text{He}(n,n')^3\text{He}$ differential cross-section was represented by Legendre polynomials:

$$
\frac{d\sigma}{d\Omega}(E_n, \cos \theta) = \sum_{l=0}^{3} a_l(E_n) \cdot P_l(\cos \theta)
$$

The computer program developed for the stripping procedure corrects the experimental neutron pulse-height distributions for the complete detector response. The program starts with the last (highest-energy) channel, labelled $N$, assuming it to contain only $^3\text{He}(n,p)^3\text{H}$ full-energy events. The corresponding wall-effect and scattering distributions are calculated and subtracted from all the channels lower in pulse height. Then the program handles channels $N-1,N-2$, etc successively in the same way, until the thermal neutron peak is reached.

The major difficulties with the $^3\text{He}$ spectrometers having impact on neutron spectroscopy, especially the energy resolution are: wall effects, recoil effects, spread in pulse rise, $\gamma$-ray sensitivity (high $\gamma$-ray input rates causing spectrum distortion because of neutron-$\gamma$ pile-up) and acoustic effects (give spurious pulses in neutron spectrum). Under normal experimental conditions, an energy resolution of better than 12 keV for thermal neutrons and about 20 keV for 1 MeV neutron was achieved for the Cutler spectrometer. For Shalev type, a resolution of 14.6 keV for thermal neutrons and of about 45 keV for 1 MeV neutrons have been reported [68].

As delayed neutron emission is always accompanied by intense $\beta$- and $\gamma$-ray emission, the sensitivity of the spectrometer to pile-up from high energy $\beta$-particles and $\gamma$-rays may be the limiting factor in delayed neutron spectroscopy. The total counting rate can be reduced by inserting lead between the source and the detector. Lead, in addition to having a high absorption cross-section for the $\beta$- and $\gamma$-rays, has a high threshold energy for inelastic neutron scattering. The influence of lead absorbers on the energy resolution and the peak shape of the monoenergetic neutrons has been tested with neutrons from the $\text{Li}_7(p,n)\text{Be}_7$ reaction. It was found that a 2 mm thick lead absorber increased the tenth-width of the neutron peaks by approximately 10% where as the half-width was not affected. In practice, for low input-rates, 2-3 mm thick layers of lead was sufficient. The influence of these absorbers on the peak shape, its energy and the spectrometer detection efficiency was found to be negligible. In a recent paper, Albert E. Evans [69, pp.1343-1346] has measured the spectrum of neutrons from a 24-keV iron-filtered neutron beam from the Omega West Reactor [70] at 1 MW and at full power of 8 MW. It was found that at 8 MW, the $n-\gamma$ pile up on the high-energy side of the 24-keV peak nearly obscured the 72- and 128-keV neutron peaks which were clearly visible in the low-power spectrum. Also noted was the prominence of the 24-keV coincidence peak in the high-power spectrum. According to Evans, if the spectrum to be measured contains a significant component of neutrons greater than 1.5 MeV, the neutrons should be incident at an angle of 15° to 30° from the detector axis. If the neutron flux is isotropic and has a high-energy component, a severe problem in unfolding the spectrum will occur.
Efficiency measurements [60,64] indicate a decrease at low energy which follows the 1/v variation of the ³He (n,p) ²H cross-section, then a rapid decrease above 1 MeV where wall effects due to charge collection problems become severe and a departure from the smooth monotonic decrease of the efficiency with energy in the vicinity of \( E_\text{th} = 130 \text{ keV} \) and \( E_\text{th} = 340 \text{ keV} \). Evans, Franz have suggested that differences in the spectrometer construction could be important in determining the performance characteristics of ³He spectrometers. Using a Monte Carlo approach, Sailor and Prussin [71] have shown that the decrease in the efficiency with increasing energy is a function of the source to detector distance. This necessitates efficiency correction for 'high' geometry measurements.

There are two major features which put constraints on the applicability of the three types of spectrometers discussed. The first is the energy dependence of the system resolution which is shown in figure 32 for the three spectrometer types. It is seen that the TOF and the proton-recoil spectroscopy are the superior methods in the low-energy range up to about 150 keV and 300 keV respectively.

![Figure 32: Comparison of detector resolution (FWHM) for a ³He spectrometer, a proton-recoil proportional counter (2.5 atm H₂/CH) and typical TOF system (flight path 50 cm, time resolution 3 ns).](image)

In contrast, ³He ionization chambers provide an adequate energy resolution of about 12 to 25 keV over the entire delayed neutron energy range of interest up to 3 MeV, where the performance of the other two detector systems worsens drastically. The increased FWHM around 300 keV is due to multiple scattering effects [30,p.153]. The second feature relates to the potential spectrum distortions whose origin are different for the three detection systems. However, in all the three cases, background effects may obscure the low-energy part of the delayed neutron spectra and thus may raise doubts as to the exact intensity distributions below about 150 keV.

The disadvantages of the delayed neutron detection systems can be minimized by careful experimentation and by taking due caution in the spectrum analysis. For example, by minimizing spectrum
distortion effects, it has been possible to resolve delayed neutrons with energy greater than 10 keV from the thermal neutron peak in \(^{3}\)He spectra.

7.6 Spectral Analysis

The conversion of a measured pulse spectrum \(C_i\) obtained with an energy-sensitive detector to a true particle energy spectrum \(\phi_j\) is called spectral analysis. Expressed mathematically:

\[
C_i = \sum_j R_{ij} \phi_j
\]

where, \(i\) denotes the energy channel \((E_i)\) and the matrix \(R_{ij}\) the response function of the detector. The components of \(R_{ij}\) give the probability that a particle of energy \(E_j\) will give a pulse of energy \(E_i\). The total probability that the particle of energy \(E_j\) will give rise to a pulse is the efficiency of detection \(\epsilon_j\).

\[
\epsilon_j = \sum_i (C_i/\phi_j) = \sum_i R_{ij}
\]

Several methods are available for doing spectral analysis. The simplest of them is to use the matrix inversion and obtain

\[
\phi_j = (\sum_i C_i D_i)/D
\]

where \(D\) is the determinant of the system and \(D_i\) is the appropriate cofactor. A drawback of this method is that unphysical negative spectral values may occur. The difficulty can be overcome by means of a modified unfolding technique [72].

Another way is to use an iterative technique originally developed by Greenberger and Shalev [73]. The method uses a complete set of premeasured detector response functions for monoenergetic neutrons to fit the entire spectrum in a least squares approach and to subtract the continuous pulse distribution below the respective fast neutron peaks. The program starts from a zeroth approximation of the energy spectrum, applies the response function to the approximate energy spectrum, deduces the expected pulse spectrum and compares it to the measured one. The \(\phi_j\) values are then adjusted in successive approximation until a set of values is obtained which yields a pulse spectrum in agreement with the measured one. In simple cases, a deconvolution method can be used provided the response function has a cut off, i.e., \(R_{ij} = 0\) for \(i > 1\). But the iterative method ignores the experimental conditions to which the response function is very sensitive. Besides, the technique is not very suitable for an error analysis. The deconvolution method cannot be used directly since the response function has no sharp cut-off in practice due to the presence of high-energy tail. In the
above methods the error at low energies are large because of contributions from all the higher energies. Evans and Krick [20], Weaver et al [42,pp.207-238], Rudstam [67] all of them used the above mentioned subtraction processes and applied fixed detector response functions. Franz et al [38,60] used response functions adapted to the experimental conditions during the spectrum measurements by varying the peak-to-plateau ratio. In addition to the continuum subtraction, corrections for thermal neutrons and γ-ray pile-up are incorporated before the residual 'net' pulse height distribution is converted to a neutron spectrum by dividing it by the efficiency function of the detector.

A method that circumvents the above problems is the modified deconvolution method [62] which is a combination of the iterative and the deconvolution methods. In this method, one starts with the highest channel 'n' containing information and adjusts its content to obtain

\[ C'_n = C_n - \sum_{j=1}^{n-1} R_{nj} \phi_j \]

and then the corresponding neutron intensity \( \phi' \) which is closer to the true value than \( \phi_n \). One more iteration gives a better and the final value (\( \phi'' \)) of the number of neutrons of energy \( E \). After determining the highest energy point of the neutron spectrum by the above-mentioned procedure, all pulse values are adjusted as:

\[ C_j \rightarrow C'_j = C_j - R_{jn} \phi'_n \]
\[ \sigma^2(C_j) \rightarrow \sigma^2(C'_j) + R_{jn}^2 \sigma^2(\phi'_n) \]

The above steps are repeated for the next channel and so on until all the channels have been converted to a set of neutron intensities \( \phi''_j \) with statistical variances \( \sigma^2(\phi''_j) \).

In the method for spectrum analysis reported by Walker et al [44,pp.265-267;61], the initial starting flux distribution was obtained by dividing the measured pulse-height distribution by the detection efficiency and the response function was computed using the equation

\[ Y(E) = P(1) \left[ \exp\left(-0.5\left(\frac{E-E_n}{P(2)}\right)^2\right) \right.\]
\[ + P(3) \exp\left(-0.5\left(\frac{E-(E_n-P(4))}{P(5)}\right)^2\right) \]
\[ + P(6) \omega(E) \]

where \( Y(E) \) is the magnitude of the pulse height distribution at an energy \( E \), \( \omega(E) \) is the wall effect prediction, \( P(1) \) is a normalising factor and \( P(2) \) to \( P(6) \) are terms determined from a non-linear least squares fit to the data. The computed result (product of flux
distribution and response function) was compared with the measured pulse height distribution by means of a chi-squared test. The iteration is continued till the change in chi-squares between two successive iterations is less than some pre-determined value when the flux distribution is determined. An analysis of errors is made by calculating the full covariance error matrix for the unfolded spectra. The basis of the operation is to vary each parameter within the unfolding operation by a known amount, and then repeat the unfolding. Thus each channel of the pulse-height distribution is in turn changed by 1 standard deviation and the unfolding repeated in each case. Similarly, each parameter which is used to define the response function, both in shape and efficiency, is varied, and a revised unfolded distribution produced. In this manner, a difference matrix $D$ between each of the unfolded cases and the unperturbed result is generated, and the error on the unfolded spectrum $V_y$ is calculated from the relationship:

$$V_y = D^T V_x D$$

where $V_x$ is the covariance matrix of the input parameters. It is important to note that the method of determining the covariance matrix is independent of the method employed to unfold the data. Using this procedure, the uncertainties in the individual unfolded spectra and in the differences between them were evaluated for a set of four measurements of delayed neutron spectra from fast fission in U-235 [74]. The results shown in figure 33 indicate a systematic trend of increasing intensity at delayed neutron energies less than 200 keV as a function of increasing primary neutron energy. This trend is statistically significant and presumably arises from variations in the fission yields of delayed neutron precursors. The letters P, Q, T and E refer to the four measurements conducted with mean primary neutron energies of 0.94, 1.76, 1.44 and 0.49 MeV respectively. Similar results for other fission products were reported by W.J. Maeck [44, pp.91].
Shikoh Itoh [69, pp. 1367-1370] has developed an unfolding procedure based on the maximum likelihood method which incorporates the Poisson statistics of neutron detection. The solution spectrum is given by the matrix equation

\[ \hat{c} = R^+ [I - A U_0 (U_0^+ U_0)^{-1} U_0^+] c, \quad R^+ = VS^{-1} U_1^T \]

where, \( c \) = counts in the ith channel, \( \lambda \) = expectation of \( c \), \( \Lambda = \text{diag} (\lambda_1, \ldots, \lambda_n) \), \( \hat{\phi} \) = number of neutrons belonging to the jth energy group, \( R_{ij} \) = the probability that a jth group neutron produces a count in the ith channel, \( R = USV^T \) is the response matrix and \( \hat{\lambda} = R\hat{\phi} \). Results of his numerical calculations showed that the procedure converged after three or four iterations and the differences between these solutions and the least-squares method were little. The appearance of negative values in \( \hat{\phi} \) were found to be more dependent on the statistical accuracies of \( c \)'s than on the methods used. Itoh has introduced a factor called "error magnifying factor" (F) whose asymptotic value F indicates the intrinsic limitation of spectrometers that need unfolding procedures. The factor F can be used to compare the efficiency of various unfolding algorithms and also to plan experiments.

In the method of Gadjokov and Jordanova [75], neutron spectra, \( F(E_n) \) are unfolded by differentiating the recoil-proton spectrum, \( P(E_p) \) using the equation [76]:

\[ F(E_n) = \frac{d}{dE_p} (P(E_p)/dE_p) \cdot (K(E_p, E_n))^{-1} \]

Here, \( E_p \) is the proton energy, \( E_n \) the neutron energy and \( K(E_p, E_n) \) is the spectrometer transfer function. The derivatives are computed on the basis of global orthonormal-polynomial fits supported by suitable functional transforms.

\[ (dP/dE_p) = (dP/da) (g.dL/dE_p)^2 + P(a).g.(d^2L/dE_p^2) \]

where, \( a \) is the pulse amplitude, \( g \) the spectrometer gain factor and \( L(E_p) \) is the light-output function. A three-factor representation is used for the response transfer function [77]:

\[ K(E_p, E_n) = e(E_n) \cdot S(S_p, E_n) \cdot B(E_p) \]

where, \( e(E_n) \) is the crystal efficiency, \( S(S_p, E_n) \) is the recoil-proton distribution when a neutron of energy \( E_n \) is scattered, and \( B(E_p) \) is the edge-effect function. Using the method outlined above, several spectra were unfolded. In particular, the peaks at energies 2.1, 3.2, 4.8, 5.6, 6.8, 7.85 and 9.7 MeV of the Pu-Be spectrum were resolved in agreement with other experimental and calculated data.
The errors introduced into the delayed neutron intensity by this unfolding procedure are estimated to be about:

\[10\% \quad (E_n < 100 \text{ keV})\]
\[7\% \quad (100 \text{ keV} < E_n < 1 \text{ MeV})\]
\[10-20\% \quad (E_n > 1 \text{ MeV})\]

These errors include all possible uncertainties in the response function, efficiency, correction for scattered and thermal neutrons, and \(\gamma\)-ray pile-up.

Though the main properties of the \(^3\)He ionization chambers are more or less well understood by now, discrepancies still exist between the near-equilibrium and the individual precursor neutron spectra accumulated by various authors using the same type of spectrometer, and there is disagreement among the data obtained with different detection methods.

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§8. THEORETICAL ASPECTS

The first evaluation of delayed neutron emission spectra was made by Saphier et al [1] using the delayed neutron spectra from twenty fission product isotopes measured at Studsvik laboratory [2-4]. The results were presented in a 54 energy-group structure for applications in nuclear technology and covered thermal neutron fission (U-233, U-235, Pu-239 and Pu-241), fast-neutron fission (Th-232, U-235, U-238, Pu-239) and high-energy (14.7 MeV) fission (U-235, U-238). The 54 multi-group energy structure was chosen because it was thought to be convenient and easily adaptable to other existing energy group structures like the ABN 26 group. However only the first 30 energy groups were presented, since there were no measured spectra below 50 keV. The upper limits (in keV) of the twenty-four lower energy groups were: 50, 45, 40, 35, 31, 28, 25, 22, 20, 18, 16, 14, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, and 1.

In calculating the spectra, they followed a least-squares fit method with the requirement that

$$\sum \lambda_i \beta_i \exp(-\lambda_i t) = \sum \lambda_n \sum \beta_n \exp(-\lambda_n t)$$

where the summation extends over all the isotopes in the ith precursor group. It was assumed in the evaluation that a fission product precursor 'n' contributed to two adjacent mathematical groups (i,i+1) with the condition that

$$\lambda_{i+1} < \lambda_n < \lambda_i$$

It was also required that the least-squares error

$$\int \left( \lambda_n \exp(-\lambda_n t) - \sum q_{n,i} \lambda_i \exp(-\lambda_i t) + q_{n,i+1} \lambda_{i+1} \exp(-\lambda_{i+1} t) \right)^2 dt$$

be a minimum. Here, $q_{n,i}$ and $q_{n,i+1}$ are the contributions of the nth fission product to the ith and the (i+1)'th delayed neutron group, and $q_{n,i} + q_{n,i+1} = 1$. By the least-squares fit process, the best values of $q_{n,i}$ for each fission product precursor for each fissile isotope were obtained and the delayed neutron spectrum for a particular delayed neutron group was calculated from the delayed neutron spectra of the fission product isotopes using the equation

$$x_i(E) = \left(1/B_i \right) \sum q_{n,i} \sum \beta_n \exp(-\lambda_n t)$$

with $n = 1, 2, ... , N$. \end{quote}
where, $B_i = \sum_{n} q_{n} P_i^n$, $Y$ is the delayed neutron contribution to the $i$th delayed group. In the evaluation, $P_i$ values were either those given by Tomlinson [5] or were obtained by a least squares fit. Another parameter that was required in the fitting process was $Y$, the fission product yield of the precursors. As stated by the authors, "large liberties were taken when $Y$ values had to be adjusted." Average equilibrium spectra for each fissile isotope were calculated by weighing each delayed neutron group spectra with appropriate $p_i$.

Figure 34 shows the measured near-equilibrium spectra superimposed on Saphier's evaluated equilibrium spectra. Although the two curves were normalized differently, there is reasonable agreement between the two above 150 keV. Below this energy, large discrepancies are seen.

The authors, while observing the large differences between the delayed neutron energy spectra from U-235 fast fission as measured by Eccleston and Woodruff [§7, ref.15] and the delayed neutron spectra from U235 thermal fission as given by Sloan and Woodruff [§7, ref.14], noted the absence of the large peaks at 33 keV and 60 keV in the measurement of Eccleston and Woodruff but reported by Sloan and Woodruff. They attributed this phenomenon to the possible differences in the relative fission product yields of some of the short-lived precursors and suggested that more precise data on fission product yields were required since the error in the fission product yield was as much as 64 per cent.

Figure 35 shows a comparison of the results of Saphier's evaluation with some older spectral data. A good agreement above 400 keV is seen; but below this energy, the evaluated spectrum gives a significant amount of structure and relatively more delayed neutrons below 200 keV.

The isotopic spectra required in the evaluation of the delayed neutron spectra by Saphier et al were measured by Shalev and Rudstam but for the short-lived isotopes which contribute to groups 5 and 6, mainly to group 6. The contributors to group 6 were Se89, Rb95, Rb96, Rb97 of which only the spectrum of Rb95 was then available. Rb95 was also a contributor to group 5. Therefore, in the sixth group, only about 30 per cent of the spectral emission was known and it belonged to Rb95. All the neutrons emitted in the precursor group 1 were assumed to be from Br87. In groups 1 and 2, hundred per cent of the spectra were known; in group 3 about 98 per cent, in group 4 about 94 per cent, and 85 per cent in group 5. Thus the data presented could be considered a complete set except for group 6.

Figure 36 shows a sample six-group spectra of delayed neutrons. The large amount of structure seen does neither represent the experimental resolution nor the great precision and detail of the data. They were of numerical origin, mainly the result of the spectral lines having been prepared directly by a computer program in which the input spectra were first segmented into sixteen hundred intervals of 1 keV width and then recombined appropriately.
Fig. 34: Equilibrium delayed-neutron spectrum from fast-neutron fission of $^{239}\text{Pu}$.

Fig. 35: Comparison of Saphier's evaluated delayed-neutron equilibrium spectra ($^{235}\text{U Thermal}$) with a few older measurements.
Fig. 36: Calculated six (precursor) group-delayed-neutron energy spectra from high energy (14.7 MeV) neutron fission of $^{238}$U.
A summation calculation similar to Saphier's was carried out by Rudstam [6] to provide updated group parameters (abundances and half-lives), group spectra and composite spectra for a variety of fissionable nuclides, namely, Th-232, U-233, U-235, U-236, U-238, Np-237, Pu-239, Pu-240, Pu-241, Pu-242 and Cf-252. In his evaluation, he used the spectral data, $\chi_{\beta}(E)$ from thirty-one precursors [3,4,7-18] to construct the group energy spectra, $\chi_i(E)dE$ which were assumed to be unchanged in shape during the cooling period. The experimental range was extended towards the extremes by a semi-empirical approach. The precursors were classified into six groups with half-life limits from group 1 to group 6 set respectively at: 55.6 s, 30 to 10 s, 10 to 4 s, 4 to 1.4 s, 1.4 to 0.4 s and less than 0.4 s. This grouping of precursors was somewhat arbitrary since there were precursors with half-lives which could be placed in either of two adjacent groups. The group spectra of a given fuel component $k$ were evaluated from the relation:

$$A_{ki} \chi_{ki}(E) dE = \sum_{n=1}^{\gamma} a_{in} \chi_{n}(E) dE$$

and then the resulting delayed neutron spectrum at any cooling time was calculated from the expression:

$$\chi_d(E,t) = \prod_{k=1}^{M} \sum_{x=1}^{6} f_x A_{ki} \exp(-\lambda_{ki}) \chi_{ki}(E) dE$$

where, the fuel was supposed to consist of $M$ fissile components with appropriate abundance factors $f_x$ determined from the fuel composition. The group abundances $A_{ki}$ and the decay constants $\lambda_{ki}$ were calculated from the corresponding precursor data using the relations

$$A_{ki} = \sum_{n=1}^{\gamma} a_{in}$$

$$\lambda_{ki} = \left( \sum_{n=1}^{\gamma} a_{in} \Lambda_{in} \right) / \left( \sum_{n=1}^{\gamma} a_{in} \right)$$

Due to the statistical cancellations of errors, error in the measured precursor spectra contributed very little to the error in the calculated delayed neutron group spectra except at the low and the high energy ends of the spectra. The main source of errors came from the incomplete nature of the precursor spectral data. There were a number of known precursors for which measured spectra were not available then and these corresponded, typically for U-235, to 0, 0.5, 2, 13, 54 and 17 per cent of the effect for groups 1 to 6 respectively. To account for this in the calculation, Rudstam gave the missing precursors the same spectrum as the partial group spectrum of the $\gamma$ known precursors of the group. This was given by the expression:
In the error analysis, the errors of the unknown spectra were assumed to be proportional to the spectral value with a proportionality constant, $f_e$. This assumption was also made for precursors with published spectra without error analysis and for parts of spectra obtained by the extrapolation procedure. Though the contribution of the missing precursors of importance to delayed neutrons were neglected, it was pointed out that for higher mass fissile materials, they can not be excluded.

Spectra calculated by Rudstam showed good agreement with integral measurements in the energy range of 100 to 1200 keV with a pronounced fine structure in group 1 and group 2 spectra. This fine structure, which was absent in the integral curves, was because the individual precursor spectra were measured with higher resolution. Spectra of the same group but from different nuclei were found to be very similar. For example, the differences between the group spectra of U-235 and Pu-239 were found to be minute and rarely outside the uncertainties attributed to the spectral points. Comparison of the spectra for groups 2, 3 and 4 from thermal-neutron induced fission of U-235 with those of Batchelor and Hyder, and of Feig showed that the agreement between the different determinations was very good. For group 3, the agreement was excellent, especially, with the spectrum of Batchelor and Hyder. For group 4, the agreement was good. Figures 37 and 38 show the normalized spectra for groups 2 and 3 respectively. The errors of the evaluated spectra are indicated at regular intervals. They were built up from both the statistical and the systematic errors.

The agreement of calculated total abundances with the recommended values was acceptable except for fast fissions of U-238 and Pu-242 where the calculated values were too low. Comparison of average neutron energies of the delayed spectra with their direct determinations showed that the agreement was satisfactory. The largest deviations occurred in group 2 because of the rapidly changing fission yield ratio between the two dominating members, Br-88 and I-137.

The authors have suggested that in applications, their group parameters and group spectra for U-235 be used while calculating neutron spectra. Only for group 2, the data for Pu-239 are to be used.

Reeder and Warner [19] used a procedure similar to Saphier et al and Rudstam to calculate the delayed neutron group and equilibrium spectra by summing the spectra of individual precursors by weighting them with the appropriate cumulative fission yields and $P_n$ values. But unlike Saphier et al and for simplicity, they did not divide the spectrum of a particular precursor into adjacent half-life groups, since the equilibrium spectra should be independent of whether such a division was performed or not. The calculated equilibrium spectra were compared to an approximate spectrum based on a Maxwellian distribution with just one parameter, the average energy. The
Fig. 37: Delayed neutron energy spectrum for Group 2, normalized to unity, of 235U. The uncertainty (±1σ) is indicated at 50 keV intervals.

Fig. 38: Delayed neutron energy spectrum for Group 3, normalized to unity, of 235U.
The formula used for the comparison was the one frequently used to describe prompt neutron spectra [20], namely:

\[ N(E) \propto E^{0.5} \exp(-E/T) \]

where,

- \( N(E) \) = number of neutrons per unit energy interval
- \( E \) = neutron energy
- \( T \) = "temperature" parameter

The average energy, \( \bar{E} \), is related to the "temperature" parameter by:

\[ \bar{E} = (3/2)T \]

In the comparison, two sets of experimental spectra for individual precursors measured at two different laboratories were used. The first set was from the Mainz laboratory [10] and included eighteen spectra. These spectra had the best resolution and accuracy and accounted for forty-five per cent of the total delayed neutron yield for U-235 (T) fission. When summed, this set gave an equilibrium spectrum that could be reasonably approximated by a Maxwellian spectrum with an average energy of 574 keV (see figure 39). But, the equilibrium average energy calculated from \( E \) values listed in Reeder's paper [19] was 440 keV based on eighty-eight per cent of the total yield. This indicated that the spectra excluded from the calculation must, on the whole, have lower energies and, therefore, the true equilibrium spectrum would be better approximated by a Maxwellian curve with \( \bar{E} = 440 \) keV. The peak structure seen in figure 39 is largely due to the I-137, which contributes nearly twenty-eight per cent of the calculated spectrum. This would correspond to about twelve per cent of the true equilibrium spectrum. Hence, the peaks would still be less prominent. The second set was from the Studsvik laboratory [7] and contained twenty-four spectra accounting for seventy-nine per cent of the total delayed neutron yield. The calculated equilibrium spectrum from these Studsvik data could not be fitted by a Maxwellian since the semi-empirical extrapolation resulted in a large peak at low energies. However, measured equilibrium spectra having a large number of neutrons at energies below 100 keV such as in figure 40 could not be fitted by a simple Maxwellian-shaped spectrum. Reeder and Warner also calculated an approximate delayed neutron spectrum for each precursor. They obtained a reasonable overall agreement with the I-137 spectrum from Mainz laboratory but with little peak structure (figure 41). In figure 42 is the comparison made with the Rb-94 spectrum from Studsvik laboratory. The semi-empirical extrapolation produced a large peak at the lowest energies characteristic of the Studsvik spectra and totally out of phase with the Maxwellian curve. But, there was a reasonable agreement between the measured Rb-94 spectrum from Mainz and the Maxwellian spectrum calculated using the corresponding \( \bar{E} \) value. It was found that except for Rb-95, the data from Mainz tended toward
Fig. 39: Calculated equilibrium spectrum of delayed neutron for $^{235}$U (T) using precursor spectra from Mainz.

Fig. 40: Equilibrium delayed neutron spectrum for fast-neutron-induced fission of $^{235}$U.

Fig. 41: Energy spectrum of delayed neutrons from $^{94}$Rb.

Fig. 42: Energy spectrum of delayed neutrons from $^{137}$I.
low intensity at low energies (< 50 keV). The authors also calculated the average energies of the six-group and the equilibrium spectra by weighting $\bar{E}$ values by the individual precursor fission yield and $P_n$ values and summing over the appropriate precursors. For this, they used the experimentally measured average energies of 34 delayed neutron precursors. They found a reasonable agreement between the calculated values and the limited experimental data available at that time.

England et al [21,22] have reported an evaluated library for 271 delayed neutron precursors and have calculated six-group parameters and spectra for 43 fissioning systems (Th-227 to Fm-255) that produce results consistent with the explicit precursor results. Although some disagreement with ENDF/B-V was observed, the major improvement was in the delayed neutron group spectra, in producing data for unmeasured systems and in expanding the incompletely measured spectra. ENDF/B-V contains six-group spectra for seven fissioning nuclides in a 28 energy-group structure that extends only to about 1.2 MeV, where as the present group spectra cover twenty-eight fissioning nuclides in a fine 10 keV energy bin structure and extend to 3.0 MeV, the maximum range of the experimental data for any precursor. For some applications, the energy range was extended to more than 8.5 MeV but without the temporal groups. The normalized spectra, and the group constants (to a lesser degree) were found to be nearly independent of the incident neutron energy. Results related to beta-effective were meant to check the self-consistency of the precursor and the group data. Delayed neutron spectra calculated for various delay intervals compared well with some recent measurements.

England et al found that of the available measured spectra for thirty-four precursors [23], thirty were inadequate in the measured energy range. These, they supplemented with two nuclear models: the BETA code [24] to extend the thirty measured spectra, and a modified evaporation model [25,26] for the remaining 237 precursors. Evaporation model had the advantage that it could produce the shape of typical spectra without knowing — unlike the BETA code — the energy levels, the spins, and the parities of precursors and their daughters, since these data are unknown for most short-lived nuclides. Figure 43 shows a comparison of the measured data with the model results. For the simple evaporation model, this comparison is typical, but is one of the better comparisons with predictions from the BETA code. It is clear from the figure that neither of the model predicts the detailed variation of the spectra, and the evaporation model does not predict the low-energy values observed in a few measured spectra.

The thirty-four precursors, having measured spectra, account for sixty-seven per cent or more of the total emission rate. These contributions, at reactor shut-down, depend on the fissioning nuclide. For U-235 thermal fission, the contributions are eighty-four per cent. In the calculations, England et al classified the relative importance of the individual precursors into three ranges: those contributing 10 per cent or more, those between 1 and 10 per cent, and those between 0.1 and 1 per cent. For some fuels, a single precursor nuclide contributes an excess of 10 per cent to the total
Fig. 43: Comparison of measured delayed-neutron energy spectra with Brady and England's model generated results for the nuclide $^{94}_{49}$Rb.
delayed neutron yield and in some cases, it can be as much as 20 to 30 per cent. Of the 271 precursors, nine have no measured data. They are: Ga-84, Ge-84, As-88, Se-90, Kr-96, Kr-98, Nb-105, Rb-91, Sb-137. Three of these (As-88, Rb-91, Nb-105) can be responsible for as much as 1 to 10% of the total delayed neutron yield in some fuels. Hence, the authors recommended the experimental determination of the $P_n$ values for these precursors which would reduce the uncertainty in aggregate calculations.

Unlike in the earlier analysis of the individual precursor data, which defined half-life bounds using the U-235 six-group evaluation, the method used by England et al to determine the group half-lives and abundances was independent of any fixed half-life bounds. Equity required that the energy spectra for each group should be determined in a consistent manner. For calculating the group parameters, the activities of all precursor nuclides following a pulse irradiation in each of the fissioning systems was first calculated for 39 cooling times up to 300 sec. These nuclide activities were then folded in with the evaluated emission probabilities to compute the aggregate delayed neutron emission values. By approximating the delayed neutron emission as a sum of $N$ exponentials ($N$ = number of time groups)

$$n_d(t) = \sum_{i=1}^{N} A_i \exp(-\lambda_i t)$$

and using a non-linear least-squares fitting routine, the parameters $A_i$ and $\lambda_i$ were determined. Here, the constant $\lambda_i$ represents an effective decay constant for the $i$th delayed group of precursors and the coefficient $A_i$ represents the initial emission of delayed neutrons. $A_i$ was found to be the product of the group decay constant ($\lambda_i$) and the group yield per fission ($a_i \nu_0$), where $a_i$ is the normalized group abundances. Calculations for neutron-induced fast fission of U-235, U-238 and Pu-239 were performed using three, six, nine, and twelve delayed groups. Increasing the number of groups from six to nine resulted in a significant improvement in the fit. However, point kinetics calculations using both the six-and the nine-group fits for step changes in reactivity did not reveal any comparable significant differences. The calculated delayed neutron activities using the new six-group parameters derived from precursor calculations for U-235 (F) showed good agreement with the experimental data [27]. The above six-group fits were performed for all the forty-three fissioning systems, and the normalized group abundances and decay constants were determined. The equilibrium group spectra were found as

$$x_{di}(E) = \sum_{n} f_{n,i} Y_{n} P_n x_{dn}(E)$$

with the assumption that a delayed neutron precursor contributed to either or both of the adjacent temporal groups as determined by the decay constants in the inequality
\[ \lambda_i < \lambda_n < \lambda_{i+1} \]

In the equation above, \( x_{dn}(E) \) is the normalized delayed neutron spectrum of precursor \( n \), \( y_n \) is the cumulative yield from fission of the precursor \( n \) and \( f_{n,i} \) denotes the fraction of delayed neutrons produced by precursor \( n \) that contribute to the temporal group \( i \) with the requirement that

\[ f_{n,i} + f_{n,i+1} = 1. \]

This condition ensured that the aggregate (total) spectrum calculated by summing the six-group spectra would be the same as that calculated by summing the contributions from the individual precursors. The fractions \( f_{n,i} \) were determined by requiring the least-squares error

\[
\int_0^\infty \left[ \lambda_n \exp(-\lambda t) - [f_{n,i} \lambda_i \exp(-\lambda_i t) \\
+ (1-f_{n,i}) \lambda_{i+1} \exp(-\lambda_{i+1} t)]\right]^2 dt
\]

to be a minimal \[1,28\]. Using this approach, England et al computed the normalized (to unity) six-group spectra for U-235 fast and thermal fission over a 1 MeV energy range and compared them with the six-group spectra of ENDF/B-V for U-235. Figure 44 shows a typical result for group 1. It is seen from the figure that the spectra calculated from the evaluated precursor data provide a much more detailed structure than the earlier ENDF/B-V spectra \[29\]. In the energy region from 1 to 79 keV, where the ENDF/B spectra have been simply extrapolated to zero, the current spectra reveal several low energy peaks of varying intensity. Since in this evaluation, group 1 has three contributing nuclides, namely: Br-87, I-137, Cs-141, this result allows the group 1 spectrum to vary for different fissioning systems depending upon the relative yields of these nuclides as suggested by ENDF/B-V data. In a U-235 fuelled system, the precursor Br-87 contributes all (100 per cent) of its delayed neutrons and the other two precursors contribute about 20% each.

The practice in ENDF/B-V of approximating the missing groups 5 and 6 spectra by group 4 data is apparent from figures 44 and 45. In ENDF/B-V, only the spectra for U-235, U-238 and Pu-239 are evaluated, and these are used to represent all the other fissioning nuclides. The small differences seen between the calculated U-235 thermal and fast spectra suggest that there is little dependence on the incident neutron energy. The ENDF/B-V spectra are also independent of the incident neutron energy. The authors have included the uncertainties in the individual precursor spectra in ENDF/B-VI but not the uncertainties in the group spectra.
Fig. 44: Comparison of normalized (to unity) delayed-neutron group 1 & group 4 spectra for $^{235}$U fast and thermal fission.
Fig 45: Group 5 and Group 6 normalized (to unity) delayed-neutron spectra for $^{235}$U.
In order to ensure that the group spectra produced results that were consistent with those obtained using the aggregate spectra derived from the individual precursor data, the authors carried out Rossi-alpha, $\alpha_{DC}$ (= beta-effective/neutron generation time) calculations on the Godiva reactor using the PERT-V code [30] which had provision to input either a single delayed neutron spectrum or individual group spectra. They found that the ratio of the six-group spectra result to the measured value [31] of $\alpha_{DC}$ was 1.0020, and that using the aggregate spectrum the ratio was 1.0076. The excellent agreement between the two results provided support for the methods used to derive the six-group spectra.

A further check on the consistency of the group spectra with the aggregate spectrum calculated from the evaluated precursor data was done by comparing the measured time-delay interval spectral data of the University of Lowell [32] with the calculated ones. Calculations of spectra for the eight delay time intervals were carried out using both the individual precursor data and the six-group data. The results from the two calculations as well as the measured spectra were found to be in very good agreement for all the eight delay intervals. There was also excellent agreement between the two calculations. The most notable differences between the calculated and the measured spectra were in the delay intervals (1.2 to 1.9 s), (2.1 to 3.9 s), (4.7 to 10.2 s), (12.5 to 29.0 s) and (35.8 to 85.5 s) with the last delay interval showing the greatest differences (see figure 46). The measured spectra for the above delay intervals showed a dominant low-energy peak, whereas as the calculations predicted similar low-energy peaks but with slightly lower intensities. In figure 46, the measurement depicts a dominant low-energy peak that is not observed with any significant intensity in the calculations. This discrepancy was attributed to the following:

(i) The dominant delayed precursors contributing to this delay interval are those that make up "group 1", namely: Br-87, I-137 and Cs-141 whose delayed neutron spectra were measured using He3 spectrometry which had poorer resolution at lower energies.

(ii) The Lowell group have stated that in the 35.8 to 85.5 s interval, while normalizing, the TOF spectrum had too severe a gamma-ray background because of which the spectrum of the neighbouring time interval was used as an estimate of the spectrum below 130 keV. And, it is precisely in the energy region below 130 keV that the largest differences are observed.

Villani et al [33] have developed a constraint least-squares method for decomposing composite spectra, measured at six or more delay time intervals following fission, into six-group delayed neutron energy spectra. The constraining condition was chosen to yield not only stable and non-negative solutions, but also to provide good fits to the measured spectra. The method was applied to obtain six-group spectra from the eight previously measured [32] composite delayed neutron spectra following thermal fission of U-235. The solutions were shown to be unique for a large range of constraint spectra and the dependence of the solutions on the choice of six-group parameters ($\beta_1, \lambda$) was also examined. The method, suggested by techniques developed for unfolding pulse-height spectra
Fig. 46: Comparison between calculated and measured (Lowell) time delay interval spectra (delay interval 35.8 to 85.5 s).
measured with proton recoil scintillators \([34,35]\), does not increase the mathematical complexity of the matrix inversion problem over that of the standard least-squares analysis. The method is applicable to spectral decomposition problems involving overlapping detector response functions and allows resolution of the spectrum into a finer time (energy) grid than is possible using standard spectral fitting procedures. The method can also be applied to unfold gamma-ray spectra measured with NaI (Tl) in the study of gamma-ray decay heat released by fission products following neutron-induced fission of reactor fuel materials.

In the fitting procedure, the quantity minimized was:

\[
\chi^2(E_\mu) = \sum_{j=1}^{M} \left[ \sum_{i=1}^{6} \frac{(\hat{y}_j(E_\mu) - f_j(E_\mu))/\sigma_j(E_\mu)}{(g_i(E_\mu) - x_i(E_\mu))/\delta_i(E_\mu)} \right]^2 + \lambda_D \sum_{i=1}^{6} \left[ (g_i(E_\mu) - x_i(E_\mu))/\delta_i(E_\mu) \right]^2
\]

where,

\[
\hat{y}_j(E_\mu) = \sum_{i=1}^{6} a_{ji} x_i(E_\mu)
\]

represent the modified spectra obtained by removing group 1 contribution from each of the measured spectra.

\[
x_i(E_\mu) = \text{the energy spectrum (normalized to same area of } 10^4 \text{) of the } i^{\text{th}} \text{ delayed neutron group}
\]

\[
a_{ji} = \frac{\beta_i}{\beta} \left[ \exp(-\lambda_i t_{j1}) - \exp(-\lambda_i t_{j2}) \right]
\]

\[
\lambda_D = \text{A positive damping co-efficient. It is an adjustable parameter.}
\]

\[
\delta_i^2(E_\mu) = \frac{\sum_{j=1}^{M} \sigma_j^2(E_\mu)}{M^2}
\]

is the statistical average of the measured uncertainties.

\(f_j, g_i(E_\mu)\) are a suitable set of positive "guess" spectra that act as a constraint to prevent the solutions \(x_i(E_\mu)\) from being highly oscillatory or negative. The energy spectrum is divided into energy bins \(\mu = 1, \ldots, M\) and each spectrum is over a finite time interval \((t_{j1}, t_{j2})\) within which all delay times have equal weighting. \(j = 1, \ldots, M\) are the measured composite spectra \(y_j(E_\mu)\). The group 1 spectrum, \(x_1(E_\mu)\) as well as the six-group parameters \((\beta_i, \lambda_i)\) were those of England and Brady [36].

The analysis was carried out using the SIXGP computer code with a 20-keV-wide smoothing function for each measured spectrum and a bin width of 10-keV. The guess spectra \(g_i(E_\mu)\) were all taken to be the average of the measured composite spectra.
Solutions for $\chi_i(E)$ were obtained for values of $\lambda_D$ ranging from 0 to 1.0. Figure 37 shows the delayed neutron energy spectra for group 2 to 6 obtained with $\lambda_D = 0.03, 0.1$ and 0.3. It is seen that each of the fine spectrum has the general shape expected of a delayed neutron spectrum and shows little oscillatory structure. Within each group, the solutions are nearly identical in shape and similar in structure. The $\lambda_D = 0.03$ solution is the best since it is the smallest among the damping co-efficients in the admissible range that produces physically acceptable solutions, i.e., solutions that do not show structure whose width is less than the energy resolution of the detection system. Comparison of the eight measured delayed neutron spectra [32] with the six-group parameterization using the $\lambda_D = 0.03$ spectra of the above figure plus the assumed group 1 spectrum showed that the fit was excellent. Figure 48 shows the comparison for the two time intervals of 0.17 to 0.37 s and 35.8 to 85.5 s.

The uniqueness of the six-group solutions was established by calculating $\chi_i(E)$ with the constraint spectra, $g_i(E)$ taken to be the measured delayed neutron spectra for which group $i$ had the largest contribution. The results were nearly identical to those shown in figure 47. The uniqueness of the detailed structure was tested by representing the second set of constraint spectra $g_i(E)$ with Maxwellian spectra, each having the same average energy as one of these spectra. The solutions $\chi_i(E)$ were virtually identical to those obtained previously. These results suggest that the solutions are unique for a class of constraint spectra having shapes similar to the measured delayed neutron spectra.

By generating synthetic composite spectra using Monte Carlo simulation studies, it was concluded that while the experimental uncertainties in the measured composite spectra are magnified in deducing the six-group spectra, they do not lead to unstable solutions. A more likely cause of the instability was considered to be coming from the six-group approximation of the time variation of the delayed neutron energy spectrum.

In order to test the dependence of the solutions $\chi_i(E)$ on the six-group parameters $(\beta_i, \lambda_i)$, six-group parameters from two other compilations (ENDF/B-V and Rudstam) were examined. It was found that in general there was a good agreement among the three sets. However, the ENDF/B-V parameters for group 6 were considerably different from those of the other two sets. Rudstam's group parameters [6] gave group spectra $\chi_i(E)$ identical to those obtained with England and Brady's parameters. Use of the ENDF/B-V group parameters produced solutions mostly similar to those obtained with the other two sets except for group 6 whose solution displayed significant differences especially above 1.5 MeV neutron energy, where a much higher delayed neutron yield resulted from the ENDF parameters.

As an example of the extent of adequacy of the six-group delayed neutron representation in reactor kinetics calculations, we present in figure 49 typical reactor rod-drop calibration curves calculated
Fig. 47: Comparison of $^{235}$U group solutions obtained by the constrained least-squares method for $\lambda_D = 0.03$, 0.1 and 0.3.

Fig. 48: Comparison of measured $^{235}$U composite delayed neutron spectra with fits deduced from the $\lambda_D = 0.03$ group solutions.
for U-235 thermal fission using the ENDF/B-V neutron precursor data [37]. The results indicate that a reactivity measurement evaluated at $3.00 with the explicit nuclide data would be evaluated at $3.23 with the ENDF/B-V six-group functions.

Fig. 49: Comparison of 105-precursor and ENDF/B-V six-group rod calibration curves ($^{235}$U thermal fission).

REFERENCES


§9. SENSITIVITY STUDIES

The analysis of variations in the response of a reactor with respect to variations in the input parameter is called the sensitivity analysis of a reactor. The resulting variation in the response is the uncertainty in the response. The response can be power, power density, reactivity, energy, temperature, etc. and the input parameter can denote a parameter or a set of parameters such as the cross-sections, delayed neutron constants, etc. In the present investigation, the parameters of interest are the spectra of delayed neutrons.

The aims of doing sensitivity analysis of fast reactor dynamic behaviour to delayed neutron energies are: (1) To provide guidance on the accuracy required in the measurements of delayed neutron spectral data; (2) To demonstrate the importance of the delayed neutron spectra in reactor static and dynamic calculations; (3) To reduce the uncertainty in accident analysis and optimize on the reactor core design since considerable uncertainty exists in the intensities of spectra in the low (<100 keV) and the high energy (>1.5 MeV) regions.

The sensitivity analysis of a reactor can be performed in two ways: first to calculate directly the reactor response to various input parameters, here delayed neutron spectra, and second to use adjoint methodology [1]. In the latter, there are two approaches: the 'operator' approach and the 'matrix' approach. Both the approaches have been used to solve linear and non-linear problems in reactor physics and engineering [2-21]. Cacuci et al [22] have developed a general sensitivity theory for treating problems characterized by systems of non-linear equations with non-linear responses. The theory utilizes the concept of Fréchet derivative in Hilbert space [23] to unify the differential, the variational and the matrix approaches to sensitivity theory in reactor physics. It is shown that they all lead to identical sensitivity expressions. A response functional of the form

\[ R[\vec{x}, \vec{a}] = \int_{\vec{\phi}} F[\vec{x}(\vec{\phi}), \vec{a}(\vec{\phi}), \vec{\phi}] \, d\vec{\phi} \]

is written as a function of the system state vector \( \vec{x} \) and the input parameter vector \( \vec{a} \). \( \vec{a} \) and changes in it, \( d\vec{a} \) are real numbers. \( \vec{\phi} \) is the phase-space position vector. The variation of the response functional is obtained by taking the Fréchet differential of the equation for \( R \) to obtain

\[ \delta R = \int_{\vec{\phi}} F_{\vec{x}} \delta \vec{x}(\vec{\phi}) \, d\vec{\phi} + \int_{\vec{\phi}} F_{\vec{a}} \delta \vec{a}(\vec{\phi}) \, d\vec{\phi} = \delta R_{\vec{x}} + \delta R_{\vec{a}} \]
where, \( F' = (F'_1, \ldots, F'_{N_{\text{r}}} \) and \( F'' = (F''_1, \ldots, F''_{N_{\text{r}}} \) are vectors. \( \delta R_\alpha \) is interpreted as the "direct effect" term accounting for those changes \( \delta \alpha \) that produce explicit changes in the response, and \( \delta R_\beta \) is the "indirect effect" term accounting for indirect changes in \( R \) due to those changes \( \delta \alpha \) that produce changes \( \delta X \). It is shown that

\[
\delta R = \sum_{i=1}^{N_{\text{r}}} \left( \delta R / \partial \alpha_i \right) \delta \alpha_i = \left( \delta R / \partial \alpha, \delta \alpha \right) = \delta R_\alpha + \left( R, S, \delta \alpha \right) + P [\delta \alpha, \delta \alpha].
\]

where, the inner product \( \left( \delta R, \delta \alpha \right) \) is obtained by taking the Frechet derivative of the equations governing the system, taking their adjoint and defining the solution vector of these equations to be \( \delta \alpha \). The \( S \) vector is the Frechet derivative of the governing equations with respect to \( \alpha \). \( P[\delta \alpha, \delta \alpha] \) is the bilinear concomitant [24] associated with the adjoint operation. It may be emphasized that the term adjoint referred to here is different from the term adjoint flux used in reactor physics. In adjoint methodology, the adjoint function \( () \) is adjoint to \( \delta X \) and not to \( X \), the state vector of the system. Therefore, \( () \) can not be interpreted as an importance function. It is rather a measure of the importance of the change in the response due to differential changes in the input data field.

Secondly, knowledge of the forward solution \( X \) is required to solve the adjoint system for \( \delta \alpha \). This is not the case with linear forward systems where the forward and the adjoint systems can be solved independently. More recently, Cacuci [25] has suggested a new direction for the analysis of non-linear models of nuclear systems by presenting a unified methodology for global analysis that synthesizes and extends the current scopes of sensitivity analysis and optimization. The methodology permits global computation of the essential features (maxima, minima, limit points, bifurcation points) of the system, provides sensitivities at any design point of interest and accommodates both equality and inequality constraints. The approach is to search for essential global features of the physical system and responses under consideration instead of investing huge efforts in the calculation of higher order sensitivities. These global features correspond to the global roots and singularities of the equation

\[
F(u) \equiv [N^*(u), N(u), S(u), K(u)] = 0.
\]

The potential applicability of this methodology was illustrated by solving test problems which involved multiple critical points and bifurcations [26,27] and comprising both equality and inequality constraints. The results showed that the methodology is reliable and yields accurate results. This unified formula recasts the problem of determining the special features (i.e., critical points) into a fixed point problem of the form
whose global zeroes and singular points within the phase-space \((u, \lambda)\) are related to the special features of the original problem. The fixed point problem \(G(u, \lambda) = 0\) is solved by using a global algorithm based on the so-called confirmation methods [28]. This algorithm also provides automatically the local sensitivities at any phase-space point \((u, \lambda)\) of interest. These mathematical methods have been developed to achieve computational economy in those problems of sensitivity analysis where determination of the effects of large parameter variations on physical systems is prohibitively expensive. The adjoint approach to making sensitivity study has the advantage that it is not necessary to repeatedly solve the governing kinetics equations. This makes it computationally less expensive and is of particular importance when large systems of equations are involved.

Both the direct and the adjoint methods have been used to make sensitivity analysis of reactor dynamic behaviour to uncertainties in the delayed neutron spectra. The earliest reported sensitivity study seems to be that of Yiftah and Saphier [29]. Using the code "FREDY" [30] which solved the equations of a point kinetic model, they showed how various \(\gamma_i\)'s (delayed neutron effectiveness) affect the reactor's response. A step of reactivity \(\delta k = 0.002\) for 0.4 sec and a reactivity ramp of \(\delta k = 0.01/\text{sec}\) for 0.3 sec were applied to a reactor which had the delayed groups of U-235. For each of this study, it was found that the peak power attained varied greatly with the delayed neutron effectiveness chosen. Generally, the smaller the delayed neutron effectiveness, the larger was the power excursion attained. In a subsequent sensitivity study of LMFBR transient behaviour, Saphier et al [31] chose four reactors: the 300 MWe SNR prototype fast breeder, the 250 MWe PHENIX prototype fast reactor, the 1000 MWe G.E. Commercial fast breeder and a very large slab reactor with data based on the INTERCOMPARISON system No.8 given by Okrent. The improved version of the SHOVAV [32] code was used to solve the one dimensional time-dependent diffusion equation by the source projection method [33]. A four energy group and six delayed neutron spectra were used in the calculation which excluded feedback effects. The four cores were perturbed exactly by the same amount in \(\Gamma_a\), namely \(\Delta \Gamma = -0.000348\). It was found that the total excursion power after 2 sec was 207\% when delayed spectra were equal to prompt spectra, and was \((238.5 \pm 1.5)\%\) depending upon whether Batchelor, Feig or Shalev spectra were used. In all the four cases, the power excursion was lower when a higher value for \(\beta\) for U-238 was used. In SNR and PHENIX, decrease in total power was about 3\%, 2 sec after the beginnings of perturbation; for GE the decrease was about 5\%, and for the Intercomparison-8 cores, the decrease was about 15\%. No simple correspondence between the differences among the kinetic parameters and the reactor transient response was found. It was concluded that detailed spatio-temporal calculations should be performed to evaluate the exact power excursion and that dynamic calculations performed in the various stages of fast reactor safety analysis should be performed with more caution with regard to the data used and should be performed for clean as well as for high BU cores. Later, Saphier et al [34] performed more detailed calculations using the multi-group two-dimensional dynamic code, FX2
[35,36]. In one series of calculations, a small portion of the Clinch River Breeder Reactor (CRBR) core was voided and the resulting transient was simulated for 5 s. The calculations were performed in 26 energy groups and six groups of delayed neutrons were used for each of the fissile isotopes. In each run, the following delayed neutron spectra were used: the evaluated spectra of Saphier et al [34], the spectra of Batchelor and McK Hyder from thermal fission of U-235 [§7,ref.6], the spectra of Feig [§7,ref.12], and the prompt fission spectra replacing delayed neutron spectra. It was found that the differences among beta-effective and among the relative powers 5 s after the beginning of the perturbation were negligible. However, by replacing these delayed spectra by prompt spectra, beta-effective increased by 12%, and the resulting power was lower by 8%. In the second series of calculations, the total core was voided by a ramp function of 0.10 s duration resulting in partial fuel melting. In this prompt critical power excursion, significant differences in the achieved power level occurred but the differences in the total beta-effective were negligible. Thus at t=0.1 s, while for Feig, Saphier, Batchelor and Hyder spectra, changes in beta-effective were negligible, the difference in the power level was within about 7%. But, difference in the achieved power relative to prompt spectra was about 4.E+5 per cent and in beta-effective about 15 per cent. Similar calculations were performed by using a single average spectra for all the isotopes, average spectra for each fissile isotope, and detailed group spectra. Negligible differences among the calculations were reported. This marginal sensitivity to differences in the delayed neutron spectra was attributed to the relatively small size and a hard spectrum of the CRBR core. Calculations were also carried out in a very large oxide core with a much softer spectrum and a different composition [37]. A narrow region of the core was perturbed by a large change in the absorption cross section (step perturbation) resulting in a significant flux shape and spectral change. The resulting transients, when different spectra were used, showed that the spread among the transients was larger than in the earlier two cases. An interesting result in this case was the transient in which the delayed-neutron spectra had a low-energy component with 46% of its neutrons emitted below 50 keV. For this the power at 0.4 s was about 2050 MW, where as for the prompt spectra it was about 1600 MW, and for the other spectra (Burgy et al [§7,ref.3], Bonner et al [§7,ref.5], Feig et al [§7,ref.12]), it was about 1450 MW. Based on these studies, the authors drew the following conclusions:

(i) Although there are significant differences among the various delayed neutron spectra when observed in fine-group structure, the influence of these differences is usually negligible when broad energy bands are used in multi-group calculations in the delayed-critical region. But, if there are large perturbations where spectral changes can be appreciable, or if significant amounts of high-threshold fissile materials are present in the core, the uncertainties in the resulting power level can not be neglected. Under such circumstances, it is desirable that delayed neutron spectra with fine group structure are used in dynamics calculations. Under such circumstances, they recomme-

nded that their spectra be used.
(ii) Significant errors in transient calculations will result in all cases if prompt instead of delayed neutron spectra are used.

(iii) More measurements of delayed neutron spectra of individual precursors are required below 150 keV, and possibly to as low as 10 keV, because these data can have a significant impact on transients in a large LMFBR.

(iv) Improved knowledge of precursor yields ($Y_n$) and delayed neutron emission probabilities ($P_n$) are needed, though they may not have a significant impact on the combined spectra. Delayed neutron spectra from fast fission of Pu-240, Pu-241 and Pu-242 need to be evaluated since LMFBR recycled plutonium fuel will contain significant amounts of these isotopes.

Onega and Florian [38] performed a sensitivity analysis to study the response of reactor power and power density to uncertainties in the delayed neutron spectra using the adjoint operator formalism of Cacuci et al. The reactor that served as the basis for their study was the reference mixed oxide (MOX) design used in the International Nuclear Fuel Cycle Evaluation (INFCE) fast breeder reactor (FBR) studies [39]. In using Cacuci's adjoint formulation [22], Onega and Florian made a one-dimensional (radial) time-dependent diffusion calculation which involved two neutron energy groups [($10^{6}$ eV, 0.11 MeV) and (0.11 MeV and 10 MeV)], two precursor groups (delayed neutrons were produced only by the isotopes U-238 and Pu-239), and cylindrical geometry without taking temperature reactivity feedback and burn-up into account. The accidents studied were ejections of a central control-rod assembly with ejection times of 2, 10 and 30 s. The control-rod ejection was simulated by ramping (reducing linearly in time) the poison cross-section in the central control region from its critical value to zero. The slope of the ramp was determined by the rod-ejection time. The reactor response resulting from changes in the delayed neutron spectra was predicted from the sensitivity derivatives which were calculated by solving the adjoint equations. The predictions were also verified by recalculating the responses by directly solving the forward transient equations. The power-density sensitivity calculations were very similar to those for the power. In the analysis, the state vector ($\vec{X}$) was:

$$\vec{X} = [x^1, x^2, c^0_1, c^0_2, c^g_1, c^g_2]$$

and the parameter vector (data field), $\vec{a}$ was

$$\vec{a} = (x^1_1, x^1_8, x^2_1, x^2_8, x^1_1, x^1_9, x^2_1, x^2_8, x^1_2, x^1_9, x^2_2, x^2_9)$$

$$= (\alpha_1, \alpha_2, \alpha_3, \alpha_4, \alpha_5, \alpha_6, \alpha_7, \alpha_8)$$
The sensitivity of the response due to a change in the component $\alpha_i$ was

\[
\frac{dR}{d\alpha_i} \bigg|_{\alpha_0} = 2\pi H \int_0^L \int_0^\infty \left[ \psi^1(r,t;\alpha_0^i) S_1^i(r,t;\alpha_0^i) + \psi^2(r,t;\alpha_0^i) S_2^i(r,t;\alpha_0^i) \right] r \, dr \, dt \\
- 2\pi H \int_0^L \int_0^\infty \psi^j(r,0;\alpha_0^i) \phi^j(r,0;\alpha_0^i) \, r \, dr
\]

where, the $\psi^3(r,0;\alpha_0^i), \ldots, \psi^6(r,0;\alpha_0^i)$ are found by solving the steady-state Fréchet differentiated two-group equations. The $\psi^j(r,0;\alpha_0^i)$ are the derivatives of $\phi^1, \phi^2, \phi^3, \phi^4, \phi^5, \phi^6$ with respect to $\alpha_i$.

Using the linear power response functional

\[
P(t;\alpha_0^i) = \int_0^L \int_0^\infty w_f \left[ \Gamma_1^i \phi^1(r,t;\alpha_0^i) + \Gamma_2^i \phi^2(r,t;\alpha_0^i) \right] \delta(t-\bar{t}) \, 2\pi r H \, dr \, dt
\]

and the weight functions for the two-group fluxes

\[
f(r,t) = w_f \Gamma_1^i \delta(t-\bar{t}) \\
g(r,t) = w_f \Gamma_2^i \delta(t-\bar{t}),
\]

the sensitivity derivatives $dP/d\alpha_i$ were calculated. From these, the change $\Delta P$ in the power was calculated for a given change $\Delta \alpha$ in the parameter vector. Knowing the response $P(t,\alpha_0^i)$ evaluated using the unperturbed parameter vector $\alpha_0^i$ and the sensitivity derivatives, the responses $P(t,\tilde{\alpha})$ were calculated, where, $\tilde{\alpha} = \alpha_0^i + \Delta \alpha$ is the perturbed parameter vector. The power response was given by

\[
P(t;\tilde{\alpha}) = P(t,\alpha_0^i) + \Delta P
\]

Some of the changes in $\alpha$ used were: $\Delta \alpha = -\Delta \alpha$, $\Delta \alpha = 2\Delta \alpha$, and $\Delta \alpha = 3\Delta \alpha$. For a given response, the adjoint equations were solved in order to calculate the sensitivity derivatives. In the calculation, $\alpha_0^i$ was either 0.83 or 0.17 and $\Delta \alpha_i$ was either 0.02 or -0.02. Sensitivity derivatives, $dP/d\alpha_i$, are positive and have units of MWth. The perturbation, $\Delta \alpha$ in the parameter vector is interpreted as an uncertainty in the parameter vector $\alpha_0^i$. So the change in the power response, $\Delta P$, is also interpreted as the uncertainty in the reactor power level.
Their sensitivity computations showed that small uncertainties in delayed neutron spectra could lead to large uncertainties in reactor power responses. For example, an uncertainty of only 2.4% in the fast components of the delayed neutron spectra resulted in a 6.1% uncertainty in the predicted reactor power level after 25 s into the transient for a rod-ejection time of 2 s, whereas an uncertainty of 18.4% in the reactor power level after 25 s into the transient. The largest uncertainty observed in the predicted power level was 23.8% at 45 s into the 30-s rod-ejection time transient and resulted from a 7.2% uncertainty in the fast components of the delayed neutron spectral data. The behaviour of the uncertainties in the power-density responses was exactly the same as the behaviour of the power responses. They increased with time during a transient and were more for rapid transients than for slow transients. The power and power density were found to be most sensitive to uncertainties in the energy spectrum of delayed neutrons from fissions in U-238 which contribute to precursor group 2 and neutron energy group 1 \((\chi^1_{238})\). The responses were least sensitive to uncertainties in \(\chi^2\). Results indicated the need for improving the accuracy of delayed neutron spectral data: the component needing most attention was \(\chi^2\) and the spectral data for \(\chi^1\) needed to be improved. Since \(\chi^2\) was an integral quantity, i.e., the second precursor group in the model contained the 2, 0.5 and 0.2 s half-life precursor groups, and energy group 1 ranged from 10 MeV down to 0.11 MeV, the authors felt that detailed spectral measurements in these precursor groups for U-238 should be performed.

Das and Walker [40-42] carried out a sensitivity analysis of an oxide fuelled LMFBR design used some years ago at the Argonne National Laboratory, USA. One-dimensional space-time kinetic calculations in multi-group diffusion theory approximation were carried out using the computer program QX-1 [43]. A loss of sodium coolant was assumed to produce an increase in reactivity and the range of ramp rates was varied from 0.57 mk/sec to about 1153 mk/sec. Nine energy groups compressed from 26 in the original code, six precursors, and Doppler feedback were used in the calculations. The influence of delayed neutron energy spectra on fast reactor dynamics was studied by calculating reactor characteristics for seven assumed spectra, i.e., the initial reference spectrum and the six variants of it. Delayed neutron spectra were varied from very soft, with nearly half the delayed neutrons below 40 keV (designated \(\chi^{1,5}_{dai}\)), to excessively hard, close to a prompt neutron spectrum (designated \(\chi^{1,5}_{dai} = \text{Prompt}\)). Although a collapsed nine-energy-group structure was used, four groups \(\chi^1\), \(\chi^2\), \(\chi^3\) and \(\chi^4\) were made zero for the initial spectra thus leaving only five populated groups (designated \(\chi^{1,5}_{dai} = 5\)). Delayed neutron energy spectra were altered by altering the fractions in the various energy groups. For the kth fuel isotope

\[
x^{3,4}_{dai,k} = \frac{\int \chi^3_{ki}(E) dE}{\int \chi^4_{ki}(E) dE} / \int \chi^d_{ki}(E) dE
t_{\text{all energies}}
\]
where, $\chi_{di}(E)$ is the energy spectrum of delayed neutron group $i$ from the $k$'th fuel isotope and $E_{g-1}$ and $E_{g}$ are the upper and lower energies for the $g$th energy group respectively. All the calculations were carried out with the initial and the altered neutron spectra normalized to unity. Of the nine transients studied, four were in the prompt critical domain and the rest in the delayed critical domain. Sensitivity computations showed that for all the transients and for all the spectra, the spread in peak power was about 1.2-6% and generally increased with input rate for both the prompt and the delayed critical transients. Power response (relative power and peak power) was most sensitive to the relative distribution of neutrons in the less than 200 keV energy region and when the delayed neutron spectrum was given the same spectrum as the prompt neutrons. For example, when a large component (46%) of neutrons was put in the less than 40 keV region, peak power in the prompt excursion resulting from a ramp rate of about 0.14/s increased by 3% but decreased by about 6% when $\chi_{di}=$prompt. In the same transient, at t=25 ms, total fission power varied by as much as a factor of about 7 depending upon whether the reference spectrum or $\chi_{di}=$4(I) spectrum was used; this factor decreased to about 1.3 at 50 ms. The spread in the energy released within the transient was about 15%. Harder the neutron spectrum, greater was the fraction of the energy released in the first prompt burst but as the transient proceeded, the harder spectrum had the opposite effect. After the first burst, the integrated energy released showed larger differences, e.g., at t=50 ms, the softest spectrum, $\chi_{di}=$4(I) gave a release 13% greater than the initial reference spectrum, where as the hardest spectrum ($\chi_{di}=$prompt) gave 20% less. For postulated accidents involving delayed critical transients, the fuel took many seconds to reach its melting point but only milliseconds with prompt criticality; the spread in these times was only a few percent over all the seven assumed delayed neutron spectra. In the delayed critical region, the spread in the maximum $\Delta k$ reached was as much as 67% for a ramp rate of about 0.6E-3 per sec and decreased to about 0.5% for 57.5E-3 per sec, but in the prompt critical domain the spread increased with ramp rate from about 1.5% to 6%. The authors concluded that the main cause of any sensitivity of fast reactor kinetic behaviour to delayed neutron spectra was through changes in the effective delayed neutron fraction, $\beta_{eff}$. The rather lower sensitivity of the present reactor compared to earlier studies was, it was felt, due to the hard spectrum for the prompt neutrons in the core, and it was suggested that calculations were made with a softer spectrum characteristic of a large power breeder reactor with more diluents in the core. It was also suggested that calculations are done to obtain results with finer energy structures in order to find the optimum number of neutron energy groups for sensitivity studies.

Das [44,45] has performed a sensitivity analysis of the kinetic response of a 500 MWe carbide-fuelled fast breeder reactor [46] to large uncertainties in delayed neutron energies using the concepts of 'Delayed Spectrum Factor' and 'Beta Growth Factor'. The 'Delayed Spectrum Factor'($K$) is a delayed neutron spectrum-dependent parameter and is a measure of the deviation of the beta-effective from its reference value resulting from the uncertainty in the delayed neutron spectra. It is unity for the reference spectrum and
not equal to unity for other spectra. The 'Beta Growth Factor' \( Q_B \) was the time variation of beta-effective during a transient with the condition that

\[
\beta_{\text{eff}} = \prod_{i=n}^{m} \beta_{i\text{eff}} \quad (t > 0)
\]

for all values of \( K_B \), i.e., for all the delayed spectra. It was assumed that the neutron effectiveness was the same for all the delayed precursor groups and that the total change in beta-effective is equally distributed over the precursor groups. In the computation, \( K_B \) was varied from 0.8 up to a maximum of 1.4 in steps of 0.1 and \( Q_B \) was taken as 0.5 mk/sec for all the spectra and was continued throughout the excursion. Positive reactivity accidents were postulated with reactivity rate varying from 10 mk/sec to 800 mk/sec and a simple reactor model was calculated using a point kinetics code SENSTVTY [47], six precursor groups and Doppler feedback. Results of sensitivity computations showed that while the achieved power level was sensitive to the shape of the delayed neutron spectrum and varied by as much as a factor of about 10 at a particular time, the spread was conservatively almost always within about 27% for the peak power attained and about 20% for the accident energy released. The spread in the maximum reactivity reached was within 18%. The time spread in the melting of fuel resulting from the uncertainty in the delayed neutron spectrum was estimated to be in milliseconds, the spread decreasing with the increase of ramp rate. Compared to the reference spectrum, the melting of fuel was delayed for the softer spectra, but occurred earlier for the hard spectra. The peak power decreased with the hardening of the spectrum, and the spread in peak power decreased with input rate. Thus a 20% uncertainty in the beta-effective resulting from the uncertainties in the delayed spectral data gave rise to a 30% uncertainty in the peak power for ramp reactivity rate of 10 mk/sec, but for 200 mk/sec, the uncertainty was only about 6%. The spread in the pulse energy released during the excursions increased with input rate. The author has suggested that the above study should be backed up with multi-group, multi-dimensional spatio-temporal calculations using, preferably, measured delayed neutron spectral data and the effects of dimensionality, neutron energy group structure, reactor configurations and compositions on the sensitivity analysis should be examined.
REFERENCES


5) P. Roussopoulos: Comptes Rendus 236 (1953) 1858-1860.


47) S. Das: 'SENSTVTY - A Point Kinetics Fortran Programme to Study the Sensitivity of a Fission Reactor' (Unpublished).
§10. CONCLUDING REMARKS

In this review, we have given a summary of the diverse role $\beta^-$,n delayed neutrons play in various problems of nuclear science and technology. We have shown that quite apart from the practical value of data on fission-product delayed neutrons for reactor applications, the phenomenon of beta-delayed neutron emission affords unique empirical clues to neutron cross-sections of far unstable nuclides including cross-sections of nuclei in excited states. The key point is to exploit the inverse relationship between neutron emission and absorption by compound nuclear levels above the neutron binding energy and to explore the possibility of obtaining data of sufficient quality and quantity to allow construction of empirically based cross-section of very neutron-rich nuclides. Such cross-sections as well as individual line properties and level densities are of interest in astrophysics and in nuclear theory. In some cases, these cross-sections may be dominated by individual isolated resonances, and in others they may be viewed as averages over a number of closely spaced resonances. In either way, delayed neutron energy spectra are a unique source of data for constructing cross-sections of nuclides that are completely inaccessible to direct measurements. Another utility of delayed neutron information is in the neutron emission probability ($P_n$), whose accurate determination over as wide a range of short-lived nuclides as possible, is important to verifying empirical or physical models employed in astrophysical r-process. By studying neutron rich nuclei with atomic number around 100, which are known to be strongly deformed, it has been demonstrated that comparison of present shell model predictions with gross beta decay properties such as half-life, $P_n$ and the beta strength function ($S_{\beta}$) may provide a new and simple way to identify Nilson parameters and orbitals, and to determine nuclear deformation. Thus there is a strong and close link between applications and the testing of nuclear models which have largely been developed by studying nuclei near neutron-rich stability. Since the nuclei produced after fission are neutron rich compared to stable nuclei and some of these lie far off the valley of stability, the ability of these nuclear models to predict the behaviour of these exotic and highly unstable species would be an excellent testing ground for these models. For example, the ability to predict beta-decay parameters which are essential in predicting the radioactivity of reactor cores after shut-down is closely linked to nuclear models and the nuclear mass formulae.

Not only in the report we have brought out the present status of delayed neutron data (both experimental and theoretical), but also have created awareness that a continuing need exists for more precise and detailed delayed neutron data. To obtain these delayed neutron parameters, one requires accurate and complete information of the fission fragment yields, half-lives, delayed neutron emission probabilities and energy spectra. Emission probabilities and decay times should be known to within 1 to 5% for the primary precursors, especially for half-lives less than 20 sec. Energy spectra of precursors should be known to at least within 20%. Calculated yield values and decay times should agree with the measured values to within 5 to 10%. Establishment of a complete individual precursor data base is a desirable long term goal to generating delayed
neutron parameters.

The delayed neutron emission probability and the fission product yield form input to the calculations of total delayed neutron yield \( \bar{\nu}_d \) and the delayed neutron spectrum. Recent work has shown that uncertainty in summation calculations is approximately equally due to fission yields and \( P \) values. The situation for precursor \( P \) values and group abundances are quite satisfactory and except for a few precursors, there is good agreement between the \( P \) values measured at different laboratories. Over 80 precursors have been evaluated and new precursors such as Cu75 and Cu76 have been identified. \( P \) values of precursors with just over 60 neutrons are reported to be lower than expected, such as the Y100-Y102 precursors. This is thought to be due to deformation. \( P \) values can be estimated theoretically to about a factor of 2.5 using the equation of Kratz and Herrmann. They can also be calculated using models. The models have the advantage that they can give isomeric ratios which the equation can not. Gross theory \( P \) 's for Rb and Cs have been found to be in better overall agreement with experimental data than the TDA (Tamm-Dankoff Approximation) predictions. From measurement of delayed neutron-gamma angular correlations for Rb94 and Rb95, it has been shown that Rb95 has about 15% f-wave emission. As regards fission product yields, a large number of independent and cumulative yields have been measured for thermal neutron-induced fission in U235 and for thermal and fast fission in other fissile nuclides. The importance of independent fission yields in nuclear fission reactors is mainly in three respects: control of nuclear reactors, size of emergency cooling systems in power stations and microscopic criticality studies. The U235 yields agree satisfactorily with evaluated data but for Pu239 the agreement is not so good. The evaluated yields need adjustment. The U235 data show much better correlations with neutron energy than do the Pu239 data. There is a definite need for more Pu239 yield measurements especially in the region of 500-1000 keV. No extensive study of isomeric yields seem to have been made. These data are needed not only for evaluations but also to test existing theories. Future work should provide more data in this field. There is need to evaluate fission product yield data by more than one evaluators and target accuracies need to be redefined. In the context of evaluation needs, the more important fissioning nuclides are: Th232, U233, U235, U238, Np237, Pu239 and Pu241. On the whole, a generally accepted reference yield data set containing measured data as well as extensive models for unmeasured data should be available to the nuclear community.

Information on total delayed neutron yield for each fissioning isotope is important to analytical calculation of materials containing mixtures of nuclides such as light-water reactor fuels where the isotopes change with increasing exposure as uranium is consumed and plutonium is produced. Even information on minor isotopes like U236 and Pu241 is important, since analytical analysis of these materials is required in reactor safety and safeguards modelling for these materials throughout the fuel cycle. In view of the major contributions currently being made by the calculated values, there is need for additional experimental measurements of the broadest possible scope and comprehensiveness to complement the existing data base. Aggregate and time-group re-evaluations of \( \bar{\nu}_d \) particularly for groups 5 and 6 - are also necessary.
The data sets for energy spectra used so far in energy-dependent reactor calculations are not at the desired level of accuracy. Although, a number of new integral measurements of delayed neutron spectra have become available, so far no consistent and generally accepted set of delayed neutron spectra for the entire energy range of interest exists. Direct measurements of composite delayed neutron spectra have the advantage that they can be performed over a much shorter time frame than generating spectra from individual precursor data. The aggregate spectra not only furnish an independent method of obtaining six-group spectra but also serve as a check to summation calculations. Comparison of delayed neutron energy spectra measured at different laboratories using the same or different techniques shows, on the one hand, a remarkable degree of consistency with regard to the existence of peak structure and the overall spectral energy distribution. On the other hand, certain systematic differences are observed both at lower energies (< 150 keV) and at higher energies (> 700 keV). These inconsistencies should be resolved by further analysis and measurements. In theoretical calculations of delayed neutron spectra, the gross theory, the statistical model, the Tamm-Dankoff Approximation (TDA) model and the Random Phase Approximation (RPA) model can each be used by paying careful attention to input parameters. Test results of spectra calculation and multichannel neutron emission have given reasonably good results when compared to experiments. An interesting observation from the calculations of delayed neutron spectra is that the shape of the spectrum is the same for thermal, fast and 14 MeV neutron induced fission in spite of large changes in the fission yields and total delayed neutron yields.

As regards the average delayed neutron energies, there is discrepancy in the estimates made by different groups. They need to be resolved. It is important to note that average energies should not be calculated from incomplete spectra, especially from those that lack the high energy part.

On the experimental side, measuring delayed neutron spectra has developed from a low-resolution detection to a high-resolution spectroscopic method over the years with a wide range of techniques having been applied to the measurements.

The major conclusions that have emerged from the various studies conducted on the sensitivity of fast reactor kinetic behaviour to delayed neutron energy spectra are: (a) Dynamic calculations performed for fast reactor safety analysis should be performed with more caution with respect to the data used and should be performed for clean as well as for high burn-up cores; (b) Significant errors in transient calculations would result in all cases if prompt (very hard spectrum) instead of delayed neutron spectra are used; (c) The reactor kinetic response depends considerably on the fraction of delayed neutrons in the low energy region i.e. on the degree of softness of the delayed neutron spectrum; (d) For large perturbations or for reactors having significant amounts of high-threshold fissile material, it is desirable that delayed neutron spectra with fine group structure are used in dynamics calculations; (e) There is need to improve the accuracy of delayed spectral data.
§11. RECOMMENDATIONS

Future work on Delayed Neutrons can focus on the following areas:

(1) Use of delayed neutron information in nuclear physics and astrophysics.

(2) Improvement in the existing theoretical models in order to obtain better estimates of unmeasured spectra.

(3) Improvement in the experimental techniques of measuring delayed neutron spectra with particular reference to efficiency and resolution.

(4) More accurate information of the structure in the delayed neutron spectra and the true intensities of delayed neutrons at both the high (>1.5 MeV) and the low ends (<100 keV) of each spectrum.

(5) Generation of delayed neutron spectral data both by microscopic summation of separated precursor results and also by analysis of aggregate spectra from unseparated precursors to check for problems in one technique or the other. Comparison of the experimentally obtained aggregate spectra with the equivalent summation results. Measurements using several different basic approaches would help in identifying the serious systematic errors.

(6) Use of both the aggregate and the microscopic data to evaluate the best "few" group representation of delayed neutron spectra and inclusion of the up-dated delayed neutron data into data files (ENDF/B, JEF).

(7) Adequacy of the widely used six-group representation of delayed neutron precursor groups versus explicit precursor representation should be examined.

(8) Neutron spectral data for individual precursors should be sent to data banks so that they are easily accessible to all users.

(9) Measurements of energy spectra of delayed neutrons coming from short-lived nuclides.

(10) Measurement of delayed neutron spectra of important individual precursors like bromine, iodine by different techniques to provide a check for inconsistencies.

(11) Measurements of aggregate delayed neutron spectra for $^{238}\text{U}$, $^{233}\text{U}$ and Th232 to complement the existing data for U235, Pu239 all of which are important for fast reactors. Delayed neutron spectral data from fast fission of Pu240, Pu241 and Pu242 are required. In particular, the fraction of delayed neutron having energies above the thresholds of fertile materials like U238, Th232 and Pu240 are important.

(12) More P measurements on individual precursors to reduce the present discrepancies in the P values and to confirm results of
single measurements. \( P_n \) values should be measured for precursor nuclides As88, Y100-Y103 and Sb138 and re-measured for other As and Sb isotopes.

(13) More detailed evaluations of \( P_n \) values. For example, \( \beta \)-strength function with smooth shapes can be used in \( P_n \) calculations.

(14) Sensitivity studies should be made to better determine which precursors need to be measured and to what accuracy.

(15) Since corrections to \( \nu_A \) also depend on knowledge of spectra, a reassessment of corrections to \( \nu_A \) measurements should be made with the improved status of the delayed neutron spectra.

(16) Delayed neutron spectra, using (n-\( \gamma \)) coincidence techniques, should be used to obtain improved \( \beta \)-strength function models.

(17) Interlaboratory comparison of spectrometers using an Am/Li source should be made.

(18) High resolution spectra for energies between 1 keV to 100 keV should be measured for selected fission products to develop data for neutron cross-sections and for further understanding of the low energy results.

(19) The influence of delayed neutron spectrum on fast reactor criticality should be studied.

(20) Use of diffusion or transport theory methods to study the sensitivity of reactor dynamic behaviour to assumed or experimentally measured delayed neutron energy spectra for different reactor configurations, different core spectra, different neutron energy groups and different energy group structure.

(21) To explore the possibility of doing further work on sensitivity theory or the refining of existing generalized sensitivity theory for applications to problems in reactor kinetics, thermal-hydraulics or other areas of physics and technology.

(22) Development of computer code based on adjoint methodology.

(23) The discrepancy in the average energy of delayed neutron spectra should be sorted out.