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THEORETICAL ESTIMATES OF DECAY INFORMATION FOR "NON-EXPERIMENTAL" NUCLIDES

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ABSTRACT

Methods of estimating nuclear decay data for short-lived neutron rich nuclides are reviewed. The emphasis is on average decay energies. The connection with other data such as half-lives, delayed neutrons, and anti-neutrino spectra is noted within the context of the beta strength function. Integral data tests are made by comparisons of calculated and measured decay heat for ^{235}U.

INTRODUCTION

In decay heat summation calculations, about one-third of the computed decay heat immediately following a reactor scram arises from short-lived theoretical nuclides [1]. The term "theoretical" is used to indicate that the experimental decay data for these nuclides is too meagre to estimate their average decay energies (and sometimes half-lives). Although the fractional contribution of these nuclides decreases rapidly with increasing shutdown time, they still contribute nearly 10% at 100s cooling time, a time important to reactor Loss-of-Coolant-Accidents (LOCA). Moreover, since any estimates of their decay energies have relatively large uncertainties, they contribute disproportionately to the uncertainty in decay heat summation calculations.

This paper reviews ways that decay data can be theoretically estimated for very short-lived nuclides. The emphasis is on decay energies; however some consideration is given to half-lives, delayed neutron calculations, and even antineutrino studies. Much of the discussion is in the context of beta strength functions although other simple models are noted. Practical applications of these methods are discussed and carried through to data testing.

CONNECTION WITH BETA STRENGTH FUNCTIONS

The total number of beta decays to an electron energy range ΔE_β and a range of final state energies ΔE_f in the daughter nucleus is

$$\Delta N = T^{(-)} \rho_N(E_f) \, \Delta E_\beta \, \Delta E_f \ , \tag{1}$$

where $T^{(-)}$ is the beta-decay transition operator and $\rho_N(E_f)$ gives the density of states in the final nucleus (a δ-function for discrete transitions). Another convenient quantity is the relative beta feed to levels near E_f:

$$b(E_f) = \frac{1}{\lambda} \int_0^{Q_\beta - E_f} dE_\beta \left\{ T^{(-)} \right\} \rho_N \ , \tag{2}$$

where the normalizing integral is the decay constant

$$\lambda = \frac{\ln(2)}{T_{1/2}} = \int_0^{Q_\beta} dE_f \int_0^{Q_\beta - E_f} dE_\beta \left\{ T^{(-)} \right\} \rho_N \ . \tag{3}$$

The integrals in Eq. (2) can be evaluated in the customary way to give

$$b(E_f) = \left[\frac{\rho_N(E_f) \ |M|^2_{av}}{d} \right] f^{(-)}(Z, Q_\beta - E_f) \ T_{1/2} \tag{4}$$

where $f^{(-)}$ is the statistical rate function and $d = 6270$ s. Eq.(4) provides a natural definition for the beta strength function which is identified with the term in brackets and contains the nuclear structure information in the reduced matrix element $|M|^2_{av}$:

$$S_\beta(E_f) = \frac{1}{d} \rho_N(E_f) \ |M|^2_{av} \ . \tag{5}$$

Average beta and gamma energies are given by

$$\langle E_\beta \rangle = \int E_\beta \ dN \ , \tag{6}$$

and

$$\langle E_\gamma \rangle = \int E_\gamma \ dN \ . \tag{7}$$

One other item of interest is the relative beta feed to levels above the pairing energy

$$\alpha' = \int_{P}^{Q_\beta} b(E_f)\, dE_f \quad . \tag{8}$$

Next, following England [2], we use his approximation for allowed spectra,

$$\frac{dP}{dE_\beta} \; F(-Z,E_\beta) \propto \frac{W_\beta^2}{P^2} \quad , \tag{9}$$

where W_β is the total relativistic energy and the other symbols have their usual meaning, to further evaluate Eqs.(6-8). The results are

$$\frac{<E_\beta>}{m_e c^2} = \frac{1}{2\eta} \int_o^{Q_\beta} S_\beta(E_f) \left[X^4(X^2 + 4X + 5) \right] dE_f \ , \tag{10}$$

$$\frac{<E_\gamma>}{m_e c^2} = \frac{1}{\eta} \int_o^{Q_\beta} S_\beta(E_f) \left[X^4(X^2 + 5X + 10) \right] dE_f \ , \tag{11}$$

and

$$\alpha' = \frac{1}{\eta} \int_P^{Q_\beta} S_\beta(E_f) \left[X^3(X^2 + 5X + 10) \right] dE_f \ , \tag{12}$$

where

$$\eta = \int_o^{Q_\beta} S_\beta(E_f) \left[X^3(X^2 + 5X + 10) \right] dE_f \quad , \tag{13}$$

and

$$X = \frac{Q_\beta - E_f}{m_e c^2} \quad . \tag{14}$$

Given the uncertainties in $S_\beta(E_f)$, the allowed approximation is of little concern here.

One striking feature of these integrals is the large powers of X in the integrand. As a consequence, transitions to low lying states (small E_f) are strongly emphasized. These transitions are governed by beta decay selection rules so that the prediction of average decay energies is complicated, and the usefulness of strength functions is curtailed. The need for a proper treatment of these low-lying transitions was emphasized by Yoshida [3]. Nevertheless, in some applications such as reactor decay heat calculations where numerous short-lived nuclides can contribute together, the calculation of average decay energies with a slowly varying strength function still makes sense.

Delayed neutron yields and half-life systematics are also directly obtainable from these equations but are not pursued in detail here. Delayed neutron yields are given by the beta feed above the neutron separation energy with a correction for gamma competition [4], and half-lives are obtainable from Eq.(3).

Another closely related area of recent interest is the calculation of anti-neutrino spectra from fission reactors. These spectra are crucial to the interpretation of recent weak-interaction experiments designed to shed light on the fundamental properties of the neutrino [5]. Once the beta-strength function is known, the beta feed for fission-product nuclides with unknown branching can be determined. The desired reactor anti-neutrino spectrum is then obtained by folding in a spectrum for discrete transitions and summing over all fission products [5].

BETA STRENGTH FUNCTIONS

Both theoretical and experimental approaches have been used to obtain beta strength functions (see Hansen [6] for a comprehensive review). A major theoretical effort was initiated by Takahashi and Yamada [7] in their gross theory of beta decay. In their work, a smoothly varying strength function is obtained on the basis of assumed collective Fermi and Gamow-Teller excitations. Yoshida [3] has implemented their approach to obtain estimates of decay energies for a number of short-lived fission products important to decay heat.

On the experimental side, a large number of strength functions have been measured for short-lived neutron-rich nuclides at the OSIRIS facility by Aleklett, Nyman, and Rudstam [8]. They summarize their results by noting that the reduced matrix element $|M|^2_{av}$ in Eq.(4) is roughly constant with respect to the excitation energy E_f for energies above the pairing energy. This behavior is equivalent to a strength function proportional to the nuclear level density. Davis et al [5] have exploited this trend to calculate anti-neutrino spectra from fission reactors. As with the average decay energies, the transitions to low lying states must be treated separately. In the approach by Davis et al, systematics were developed for α' the relative beta feed above the pairing energy. The residual branching $1-\alpha'$ was then distributed among three hypothetical states at 0, P/3, and 2P/3 (P = pairing energy).

In the work based on the gross theory of beta decay, Yoshida [3] has used a slightly different method to account for transitions to low-lying states. Based on the gross theory, all the branching below a hypothetical state in the daughter nucleus at an energy Q_{oo} was collapsed to this state. Systematic values for Q_{oo} were then obtained by adjusting Q_{oo} to obtain the experimentally measured average decay energies for 19 nuclides. Values of Q_{oo} of 0 to 2.5 MeV were obtained with an adopted value of ~ 1 MeV. It was also found that adjustments of Q_{oo} to half-life data improved the prediction of average decay energies.

THEORETICAL ESTIMATES OF DECAY DATA

In spite of the rough approximations inherent in the beta-strength function approach, Eqs.(10 & 11) are quite constraining and in general predict that $\langle E_\beta \rangle$ and $\langle E_\gamma \rangle$ are roughly proportional to Q_β. In fact, the simple prescription $\langle E_\beta \rangle = \langle E_\gamma \rangle = \frac{1}{3} Q_\beta$ used by Tobias [9] and by Blachot and Fiche [10] works quite well.

A more elaborate parameterization was used for ENDF/B-IV [11]. About 150 nuclides with experimental decay energies were used to obtain the following equations:

$$\langle E_\beta \rangle / Q_\beta = 0.474 + 0.0177P + 0.00406(N-Z) - 0.00252A, \quad (15)$$

$$\langle E_\gamma \rangle / Q_\beta = 0.0399 - 0.0110\,P + 0.0100(N-Z) + 0.000191A. \quad (16)$$

For example, for ^{87}Br these formulas give $\langle E_\beta \rangle / Q_\beta = 0.344$ and $\langle E_\gamma \rangle / Q_\beta = 0.214$ with $Q_\beta = 6.6$ MeV, values quite close to the $1/3\ Q_\beta$ prescription. It is interesting to note the significant (N-Z) term for $\langle E_\gamma \rangle$. As one moves toward neutron rich nuclei, the trend is for a greater proportion of gamma energy. A similar trend was noted by Spinrad's group at Oregon State University.

For ENDF/B-V, a beta-strength function approach was used similar to the work of Davis et al [5] in estimating antineutrino spectra. A constant reduced nuclear matrix element was assumed, or equivalently a strength function proportional to the nuclear level density ρ_N. There are large uncertainties in the strength functions and, accordingly, a simple constant temperature formula for ρ_N was chosen. Because of the importance of beta transitions to low-lying states, one additional parameter Θ was introduced which describes the relative normalization of an assumed constant nuclear density below the pairing energy:

$$\rho_N(E_f) = \begin{cases} \Theta & E_f < P \\ (1-\Theta)\, e^{E_f/T} & E_f \geq P \end{cases} \quad . \qquad (17)$$

The nuclear temperature was parameterized by a single variable C_T:

$$T = C_T \, T_{GC} \quad , \qquad (18)$$

where T_{GC} is the nuclear temperature determined by Gilbert and Cameron [12]. The normalization Θ was allowed to have a very simple A-dependence by introducing two parameters Θ_1 and Θ_2 for the light and heavy fission product mass peaks respectively. The three parameters Θ_1, Θ_2, and C_T were then determined by fitting to the decay energies for 276 nuclides evaluated by Reich for ENDF/B-V [11]. Additionally, measured α' values for 67 nuclides reported by Aleklett et al, were included in the fit. The results obtained are $\Theta_1 = \Theta_2 = 0.5$ and $C_T = 3$. The least-squares residuals were not sensitive to very small changes in these parameters and rounded values were selected for convenience. This parameterization was then used to generate average beta and gamma energies for all ENDF/B-V fission products where experimental values were unavailable. For a few nuclides, decay energies were estimated by Reich and Bunting by the direct use of measured strength functions [13].

Many more short-lived fission products have experimental half-lives than have experimental decay energies, and the estimation of theoretical half-lives is of less practical concern. These estimates can be obtained the same as for decay energies; however simple parameterizations suffice. In general these descriptions are close to the well known rule [14]

$$T_{1/2} \propto Q_\beta^{-5} \quad . \qquad (19)$$

DATA TESTING

This section gives a very short review of the effect of these theoretical estimates in decay heat summation calculations. Figure 1 shows a breakdown of the relative contributions of three categories of fission-product nuclides to the decay heat following a fission pulse of ^{235}U (thermal):

(1) 276 experimental nuclides evaluated by Reich [11],
(2) 38 nuclides evaluated by Reich [13] on the basis of measured strength functions, and
(3) 430 nuclides with theoretical decay energies,

all from ENDF/B-V [11]. For decay times beyond 100 s, the contribution of the theoretical nuclides rapidly becomes negligible. For a more realistic finite reactor operating history, their contribution is further diminished.

Decay heat calculations based on three different evaluations of decay data are compared in Figure 2. Graphs are shown separately for the total decay heat and the beta and gamma components. Each graph displays the fractional deviation of the calculation from a recent comprehensive decay heat evaluation for ^{235}U [15]. ENDF/B-IV yields [11] were used in all cases. The dotted curve labeled ENDF-IV represents the use of the complete ENDF/B-IV fission product data file. The ENDF-V (prelim) dashed curve uses theoretical decay energies based on the same parameterization Eqs. (15 & 16) used for ENDF/B-IV but with updated Q_β values. The substantial changes seen are mostly due to revised decay data (half-lives and decay energies) for the experimental nuclides. Also, many nuclides that were classed as theoretical in version IV have been changed to experimental in version V of ENDF/B. Finally, the solid curves, labeled ENDF-V (revised), represent the use of decay energies based on the strength function parameterization described above. The small differences seen between the use of the different decay energy parameterizations reflect their relative consistency and the dominant effect of the experimental nuclides.

The lack of improvement in the decay heat calculations based on the latest theoretical decay energies, especially for the gamma component, is disappointing. Unless the experimental decay heat evaluations are seriously in error, the calculated gamma component is about 20% low for decay times less than 100 s. Since this component is low even at 200 s where the experimental nuclides contribute over 90% of the total gamma energy, there is an indication of a systematic error in the experimental evaluations for the short-lived nuclides. Such an error would also affect the theoretical estimates since they are based in part on the experimental values. As a practical matter, one must continue to depend strongly on experimental decay heat assessments for the short cooling times. Improvement in the calculated values is still highly desirable, however, because of the flexibility such calculations afford and because once their general validity is established, they can be confidently applied in areas where direct measurements are unavailable.

The assessment of uncertainties in model estimates of decay energies is difficult, especially where one is extrapolating beyond the measured values. Some general comments can still be made however. The standard deviations obtained from the variance between the theoretical values and the experimental values were 0.37 MeV for $\langle E_\beta \rangle$ and 0.82 MeV for $\langle E_\gamma \rangle$. The larger value for $\langle E_\gamma \rangle$ reflects the larger dispersion of experimental values for $\langle E_\gamma \rangle$. Beta Q-values are typically known for experimental nuclides. However many [16] of the Q-values for the theoretical nuclides are estimated from semi-empirical mass formulas. This source of uncertainty contributes, very roughly, an additional 25% uncertainty [16]. Because this source of uncertainty is expected to be strongly correlated over the different nuclides, it is an important component of the total uncertainty in decay heat calculations at very short (<100 s) cooling times.

Not too long ago, decay heat summation calculations were thought to be very unreliable at very short times because of the lack of data for the short-lived nuclides. This situation has been dramatically improved. Nevertheless there is still room for substantial improvement, especially for the separate beta and gamma components.

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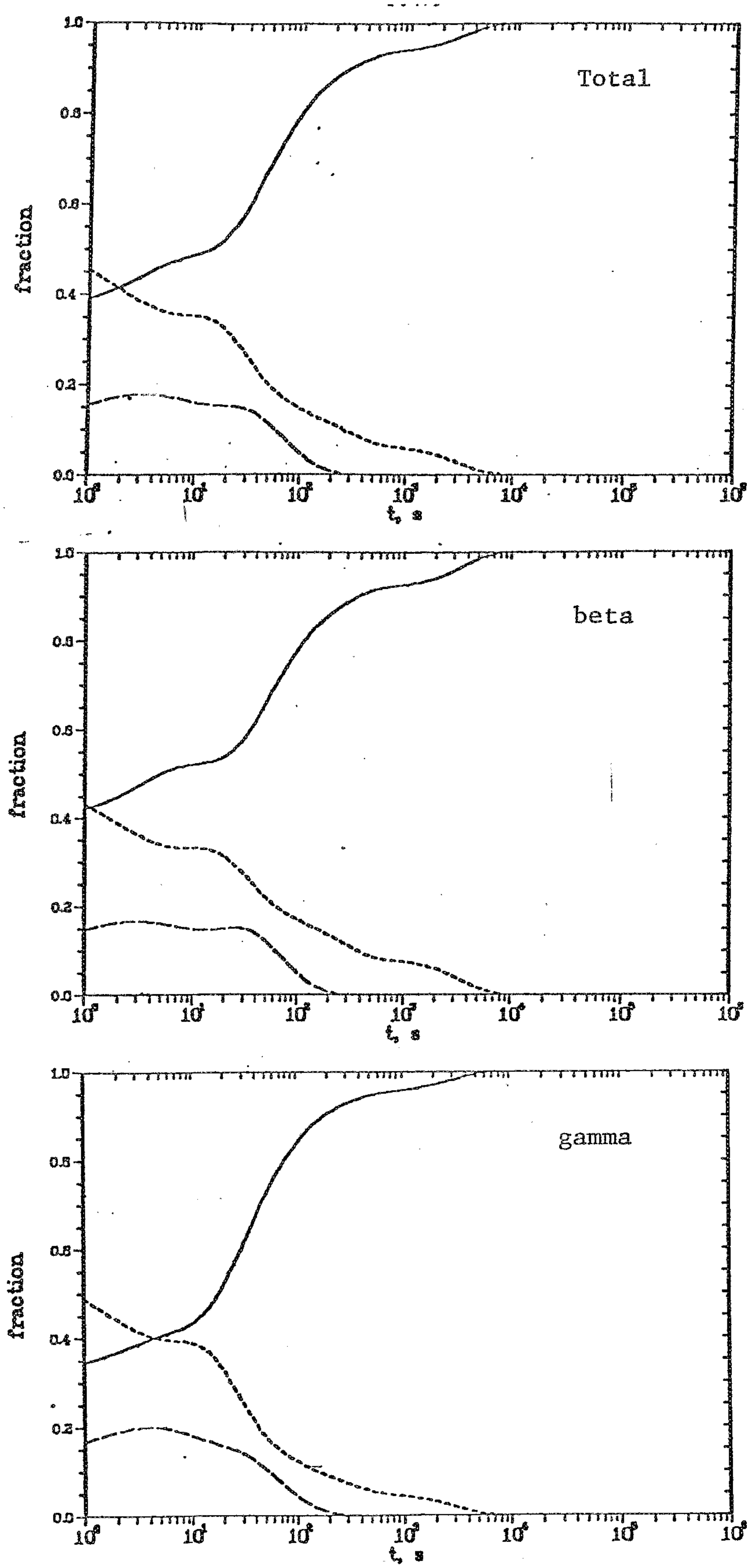

Figure 1. Fractional Contributions to Calculated Decay Heat for a Pulse Fission of ^{235}U (thermal). ——denotes experimental nuclides by Reich [11]; ---- denotes measured strength functions [13]; ...... denotes theoretical.

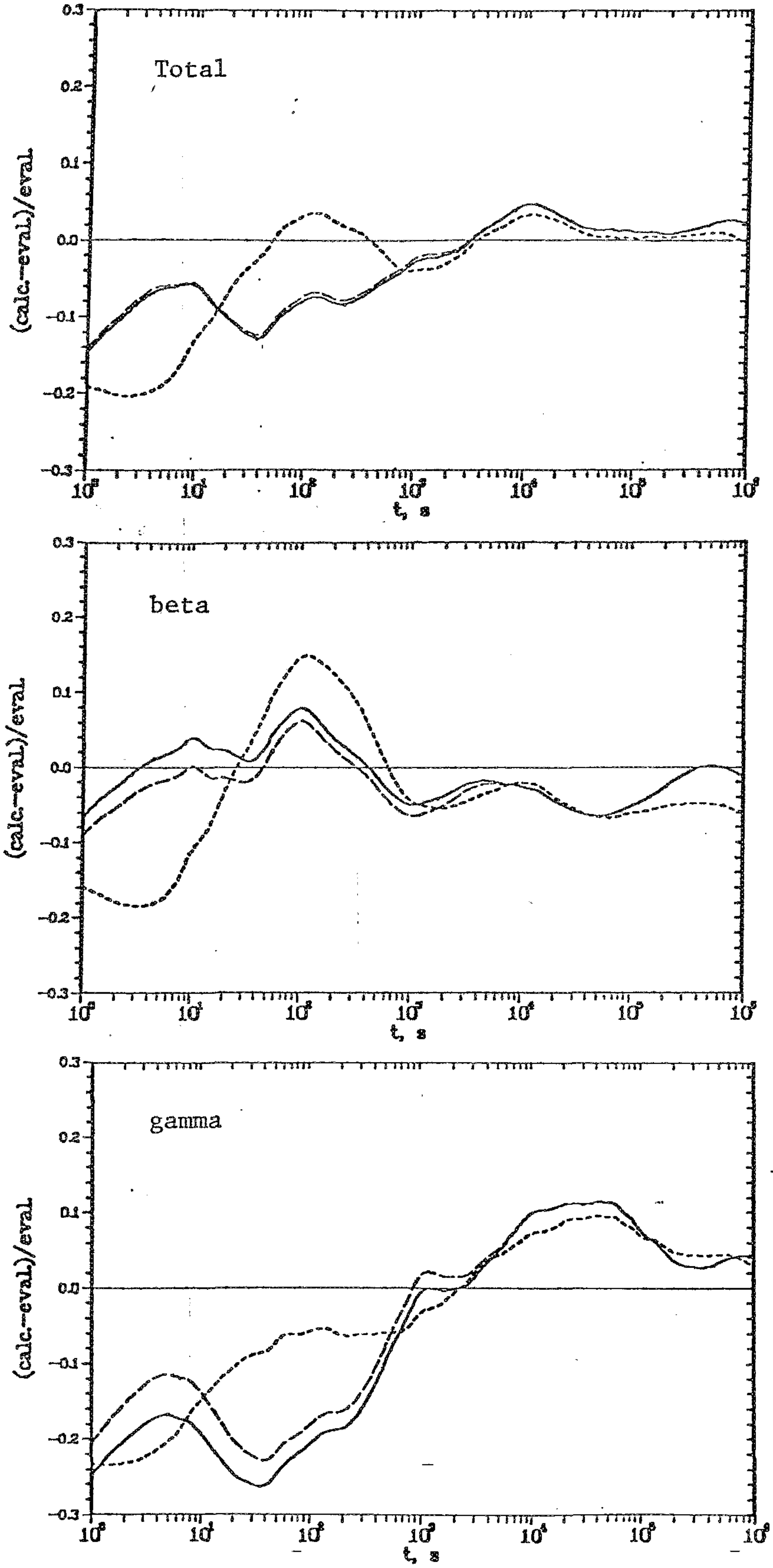

Figure 2. Comparison of Calculated and Evaluated Decay Heat for a Pulse Fission of ^{235}U (thermal). ——— denotes ENDF-V (revised) ---- denotes ENDF-V (prelim); ..... denotes ENDF-IV.