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APPROXIMATIONS TO SUMMATION CALCULATIONS OF DELAYED ENERGY AND SPECTRA FROM FISSION PRODUCTS

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R. J. LaBauve, T. R. England, M. G. Stamatelatos, and D. C. George

ABSTRACT

The purpose of this report is to provide users with simple analytical least-squares approximations to fission-product decay-energy and spectral results from summation calculations for fission burst or extended periods of fission. These approximate representations will enable users to circumvent the computational complexities associated with the detailed summation calculations.

I. INTRODUCTION

A current Los Alamos Scientific Laboratory (LASL) research effort involves calculations of fission-product decay energies (beta and gamma) and spectra from fast and/or thermal fission of a number of actinides -- ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu. The intent of these calculations is to provide a reliable source of information on delayed-energy release for a wide range of neutron irradiations (10^{-4}-10^{13} s) and for post-irradiation cooling times ranging from a fraction of 1 s to many years.[1-6] Although there are many areas of application for such information, emphasis has recently been placed on short cooling times. The great interest in calculations for short cooling times is related to some nuclear reactor safety aspects such as the loss-of-coolant accident (LOCA) condition. There is, however, also interest in long cooling times. The complexity involved in fission-product decay-energy calculations can be inferred from the following summary of the computational procedure used at LASL.

The ENDF/B-IV fission-product files[7] used as the initial data base contain neutron cross sections, decay constants, decay energies, and other decay data for 825 important fission products. They also contain fission yields for the same nuclides produced by 14 MeV, fast and thermal fission of a number of actinides including ^{232}Th, ^{235}U, ^{238}U, ^{239}Pu, etc. Any computer code that can accurately (within the limitations of the input data) calculate decay energies for wide ranges of irradiation and cooling times must be capable of efficiently handling enormous amounts of data. Several codes are now available.

One code now being used at LASL to calculate fission-product activities; beta, gamma, and total decay energies; fission-product gaseous inventories; etc., is CINDER-10.[8] The neutron cross sections for use in CINDER-10 have been generated by spectrum collapsing multigroup data generated with the MINX code.[9]

Two other codes, FPDCYS and FPSPEC, are used in conjunction with CINDER-10 when fission-product beta or gamma spectra are being calculated. The first code constructs multigroup (in arbitrary number of energy groups) beta and gamma spectra from individual fission-product nuclides for which spectral data exist in ENDF/B-IV files. These spectra, together with the corresponding activities and aggregate beta and gamma energies from CINDER-10, are further used by the FPSPEC code to calculate cumulative fission-product beta and gamma spectra for any desired irradiation and cooling times. The schematic of the entire code system is shown in Fig. 1.

The results of the above procedure have been successfully compared with experimental fisson-product gamma and beta spectra from ^{235}U thermal fission performed at LASL and the University of Illinois, respectively.[4,10] For the gamma-spectral comparisons with experiments at LASL (Figs. 2-5), gamma spectra were calculated in 150-group equal-grid energy structure from 0 to 7.5 MeV. CINDER-10 results have also been compared successfully with results from fission-product decay-energy experiments (^{235}U thermal fission) at LASL,[4] Oak Ridge National Laboratory (ORNL),[4] and Intelcom Rad Tech (IRT).

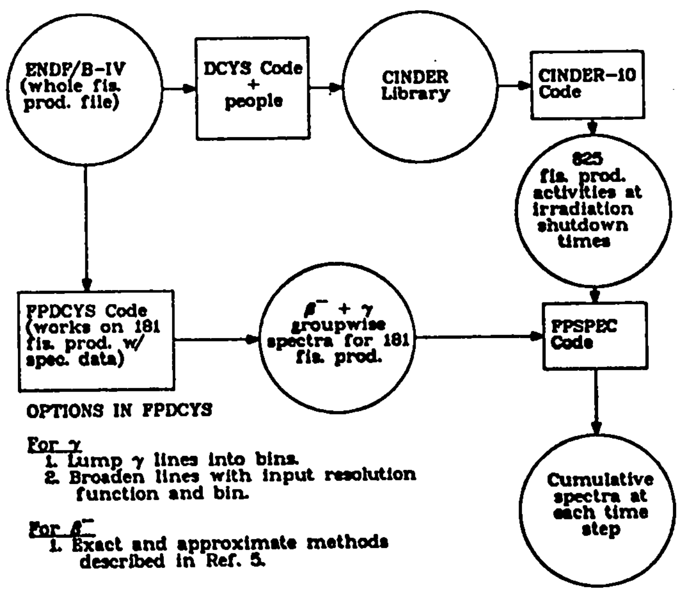

Fig. 1.
LASL code system for producing β^- and γ fission-product spectra.

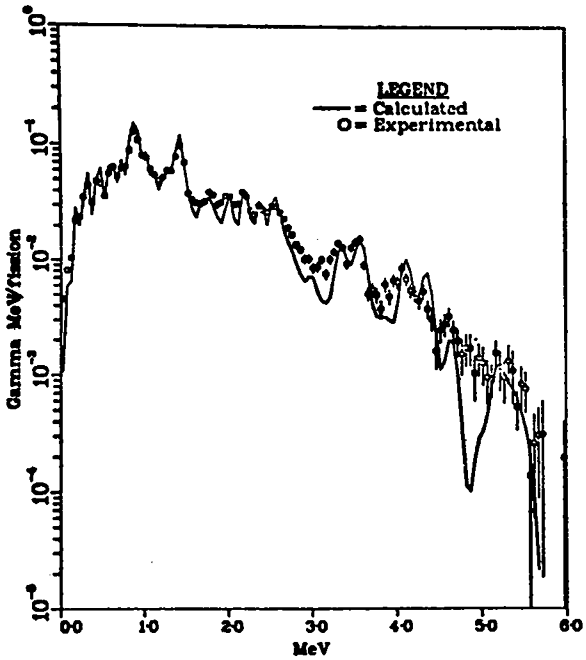

Fig. 2.
Gamma spectrum 5.56 h irradiation, 70 s cooling.

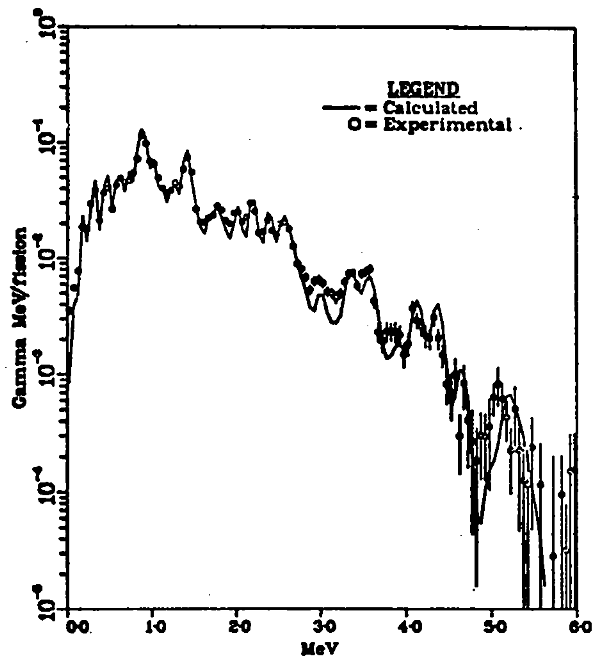

Fig. 3.
Gamma spectrum, 5.56 h irradiation, 199 s cooling.

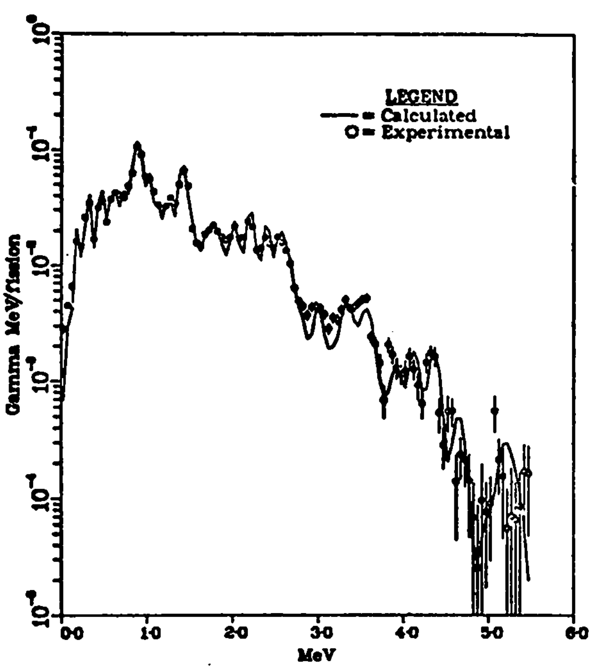

Fig. 4.
Gamma spectrum, 5.56 h irradiation, 388 s cooling.

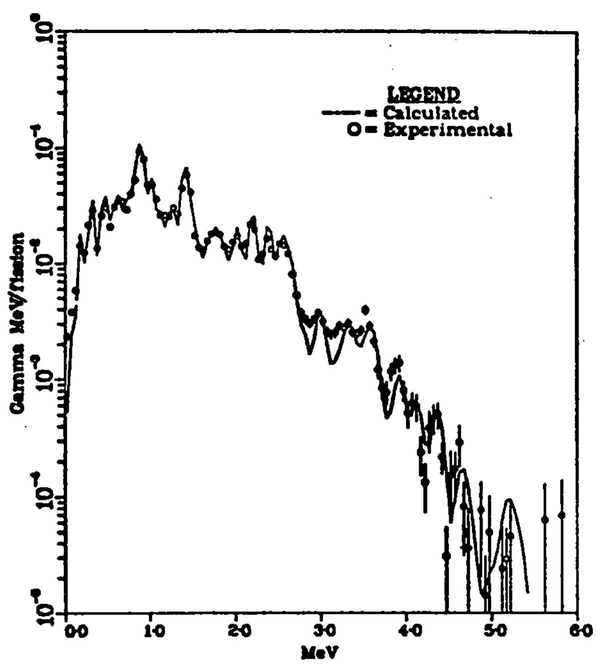

Fig. 5.
Gamma spectrum, 5.56 h irradiation, 660 s cooling.

The good agreement between summation calculations and experiments has demonstrated the reliability of the CINDER-10 code methodology and the adequacy of ENDF/B-IV data for many applications.

In view of the complexities associated with the calculations, there is a need to provide the users of the summation results with accurate, simple, and compact representations of these results that they can easily incorporate in their own specialized calculations. For this purpose, simple, analytical least-squares fits for summation results are provided so that

(1) spectra for additional intermediate cooling times can be rapidly interpolated and, more importantly,

(2) a histogram or analytical representation of a reactor power history can be folded with the fitted burst function and integrated to give decay spectra at any specific cooling times for a given irradiation history.

The purpose of this report is to demonstrate that a "broad-group" energy representation of the spectral data can be approximately fit with a sum of exponential functions that, when folded with a representation of a power history, give rise to new analytic functions that can be easily integrated. Also, the technique described can be applied to reducing experimental results to a burst function basis. This enables one to intercompare the results of different experiments and calculations.

II. FITTING THE CALCULATED DATA

The method used in fitting the calculated data is as follows. We assume the burst function $fc(t)$, in units of MeV/fission-s, for a particular energy group in the spectrum to be a linear combination of functions

$$fc(t) = \sum_{k=1}^{L} \alpha_k g_k(t) \quad , \tag{1}$$

where $g_k(t)$ can be any function but, to date, we are using $g_k(t) = e^{-\lambda_k t}$. The λ_k's are chosen by some consistent method but in this report they are not fitted, that is, the method we are describing is a single-parameter fit, a fit of the α_k's given adequately chosen λ_k's.[11] We have recently made two-parameter fits (λ_k and α_k) of functions of this type for the total fission-product decay power following ^{235}U and ^{239}Pu fission bursts.[1] This has given us insight into choosing the λ_k's for this single parameter fit. The single parameter fit described in this report demonstrates the feasibility of using burst functions. A single parameter fit is accurate for most uses; a two parameter fit requires fewer exponentials for a given accuracy and will be used in subsequent work.

Let fx(t) be values of the burst function calculated using the code system of Fig. 1 at cooling time t and let us choose specific values t_i over which the α_k's are to be fit. Denote fx_i, $i = 1, 2, \ldots, N$ and fc_i, $i = 1, 2, \ldots, N$ where $N \geqslant L$. For generality, assume a weighting function w_i and minimize

$$X = \sum_{i=1}^{N} (fx_i - fc_i)^2 \, w_i \quad , \tag{2}$$

$$\frac{\partial x}{\partial \alpha_\ell} = -2 \sum_{i=1}^{N} (fx_i - fc_i) g_{\ell i} \, w_i = 0 \quad , \tag{3}$$

$$\ell = 1, 2, \ldots, L \quad .$$

Incorporating w_i into fx_i and fc_i we have

$$\sum_{i=1}^{N} fx_i g_{\ell i} = \sum_{k=1}^{L} \alpha_k \sum_{i=1}^{N} g_{\ell i} g_{ki} \quad , \tag{4}$$

$$\ell = 1,2,\ldots, L \quad .$$

The g's and fx's are known, and we therefore have L linear equations for the L unknown α's.

A small code, ERDALEW, was written to fit calculated fission-product gamma-decay power following a ^{235}U thermal fission burst. This code was preceded by a routine, FOSTBIN, that rebins the fine group data into a coarser group structure with arbitrarily chosen energy boundaries. An 8-group energy structure shown in Table I was chosen for this test case and 2 points per time decade from 0.1 to 10^9 s were chosen for the α_k fit. The λ's were calculated between pairs of points, that is,

$$\lambda_k = \log\,(fx_i/fx_{i+1})/(t_{i+1} - t_i) \quad . \tag{5}$$

Results of this test case are shown in Figs. 6-13. It can be seen from these figures that all fits are generally good except for group 7 in the vicinity of 10^5 s where growth reverses the slope of the decay curve.

TABLE I

EIGHT-GROUP ENERGY STRUCTURE USED IN SAMPLE

Group No.	Lower Energy Boundary (MeV)	Upper Energy Boundary (MeV)
1	0.00	0.25
2	0.25	0.50
3	0.50	0.75
4	0.75	1.00
5	1.00	1.50
6	1.50	2.50
7	2.50	4.00
8	4.00	7.50

III. APPLICATION OF BURST FUNCTION FIT TO CALCULATION OF DECAY POWER AFTER FINITE IRRADIATION

The fitted burst function can be folded with a reactor power history so that decay spectra from irradiated fuel can be calculated as a function of cooling time. Consider a reactor operated at variable power $P(t')$, $0 < t' < T$, for a time interval T followed by a shutdown period t_s. Let

$$fc = \sum_{k=1}^{L} \alpha_k e^{-\lambda_k t} \quad , \tag{6}$$

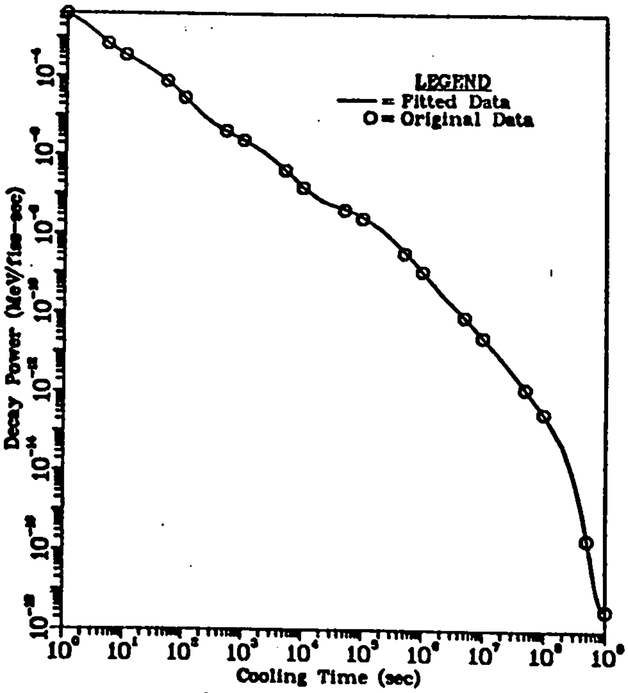

Fig. 6.
Gamma decay power following ^{235}U thermal fission burst for group 1 energy range 0.0 to 0.25 MeV.

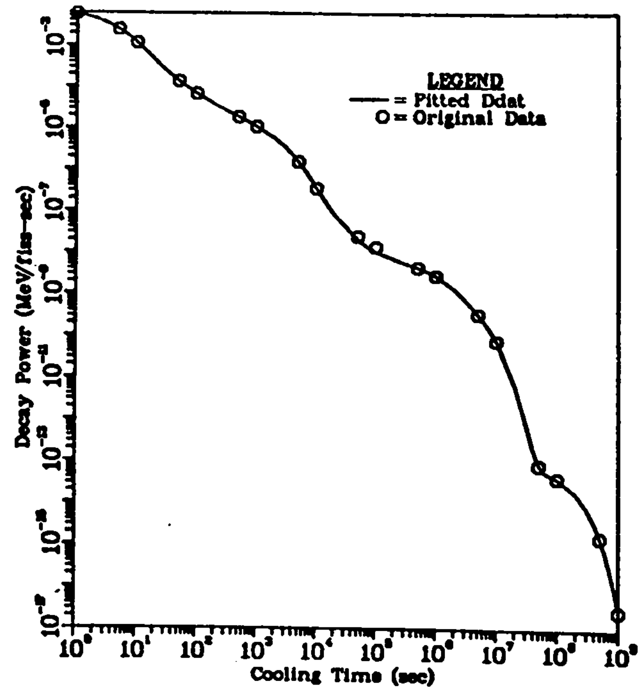

Fig. 7.
Gamma decay power following ^{235}U thermal fission burst for group 2 energy range 0.25 to 0.5 MeV.

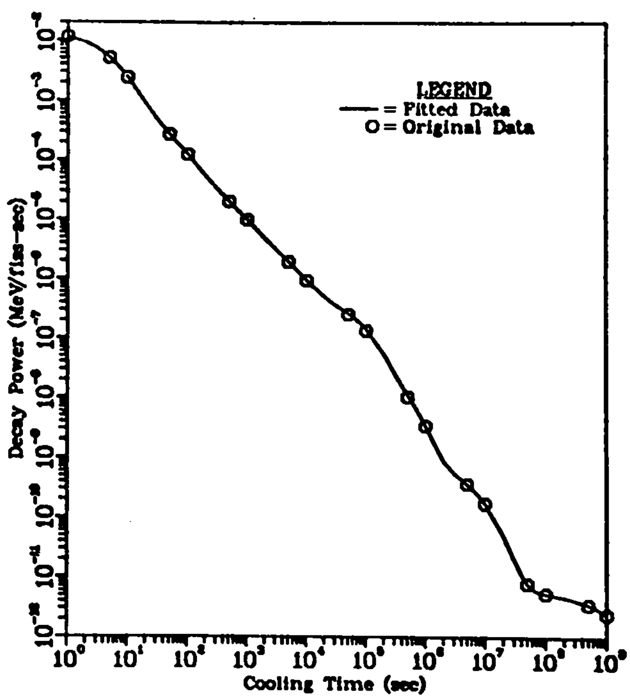

Fig. 8.
Gamma decay power following ^{235}U thermal fission burst for group 3 energy range 0.5 to 0.75 MeV.

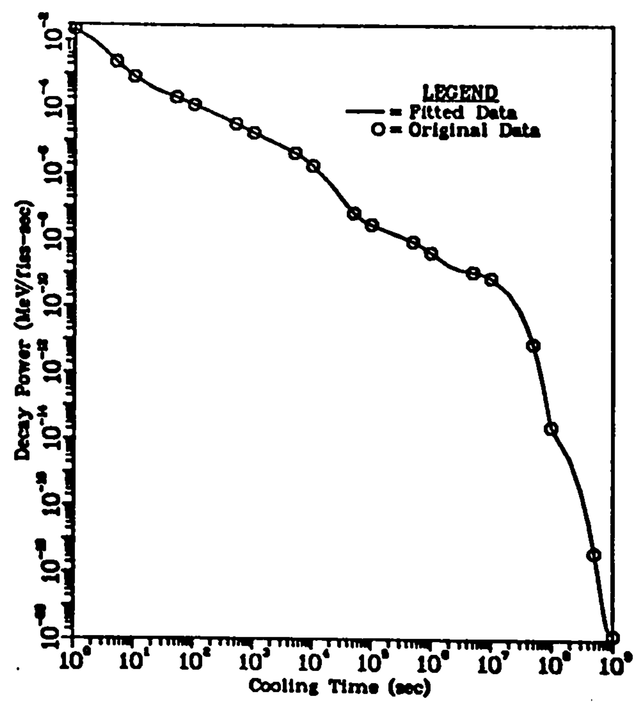

Fig. 9.
Gamma decay power following ^{235}U thermal fission burst for group 4 energy range 0.75 to 1.0 MeV.

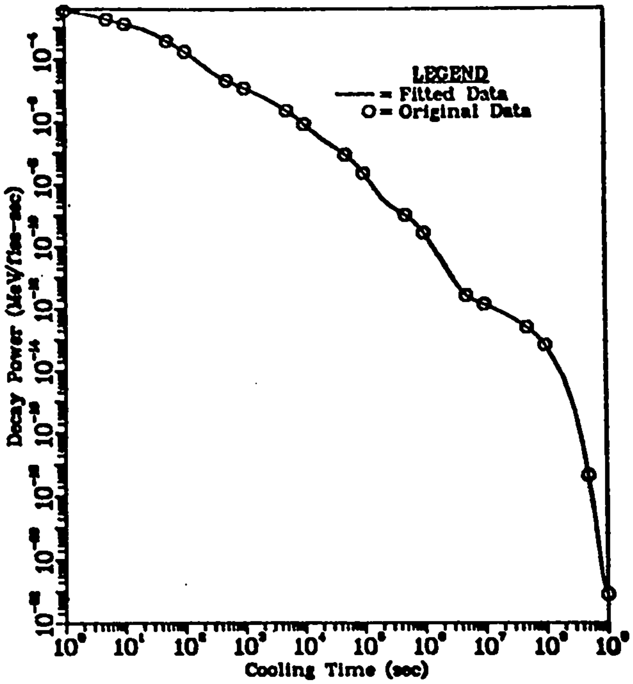

Fig. 10.
Gamma decay power following ^{235}U thermal fission burst for group 5 energy range 1.0 to 1.5 MeV.

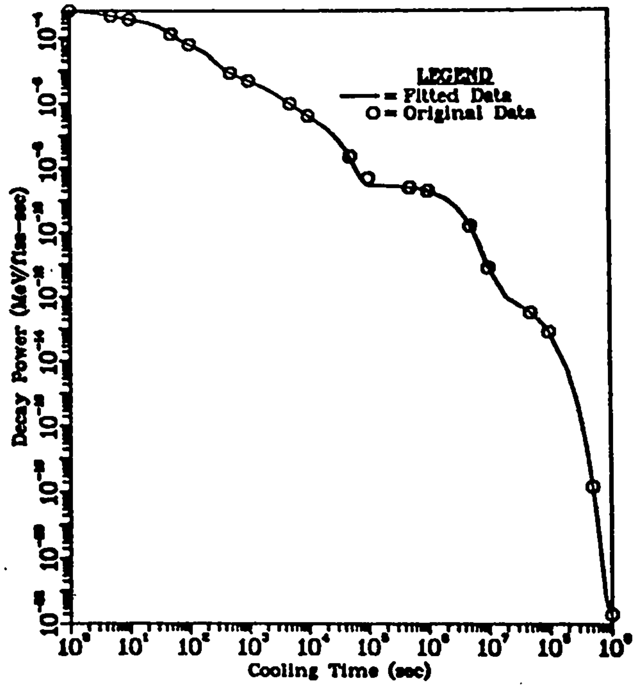

Fig. 11.
Gamma decay power following ^{235}U thermal fission burst for group 6 energy range 1.5 to 2.5 MeV.

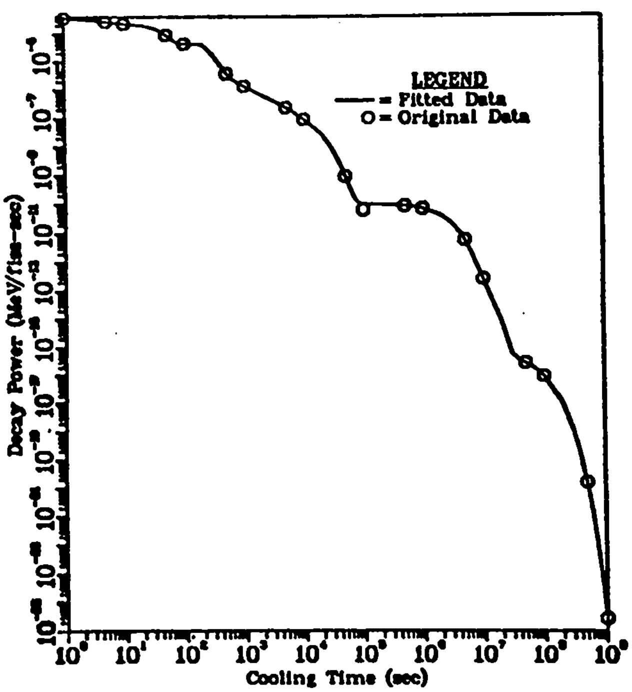

Fig. 12.
Gamma decay power following ^{235}U thermal fission burst for group 7 energy range 2.5 to 4.0 MeV.

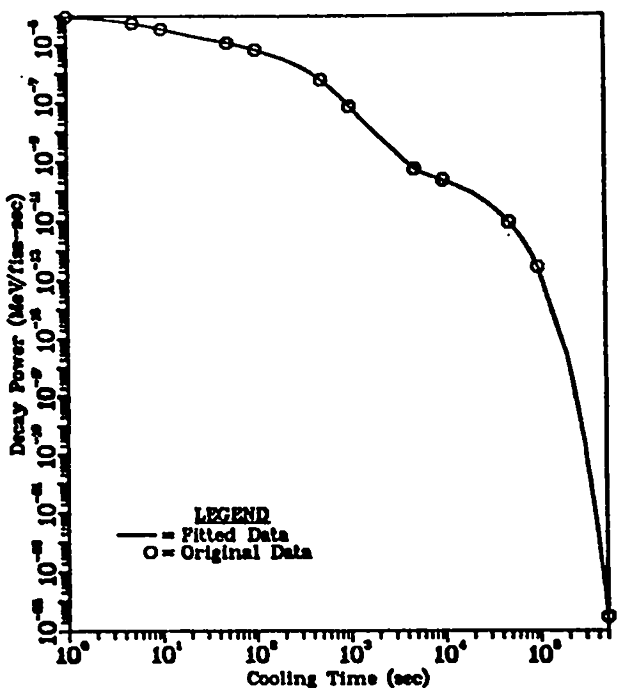

Fig. 13.
Gamma decay power following ^{235}U thermal fission burst for group 8 energy range 4.0 to 7.5 MeV.

t = time since fission burst,

$P(t')$ = power in watts at time t',

$K = 0.32042 \times 10^{10}$ W-s/fis,

T = total time at power,

t_s = shutdown time of interest, measured from T, and

$F(T+t_s)$ = decay-energy release at time $(T + t_s)$ for some energy bin (MeV/s).

Then,

$$F(T+t_s) = \int_0^T \frac{P(t')}{K} fc(T+t_s-t')dt' \tag{7}$$

or

$$F(T+t_s) = \int_0^T \frac{P(t')}{K} \sum_{k=1}^{L} e^{-\lambda_k (T+t_s-t')} dt' \quad . \tag{8}$$

Assume, for example, that the power history can be approximated by J histograms with a power of P_j at irradiation time T_j. Then,

$$F(T+t_s) = \sum_{j=1}^{J} \frac{P_j}{K} \sum_{k=1}^{L} \alpha_k \int_{T_{j-1}}^{T_j} e^{-\lambda_k (T+t_s-t')} dT \tag{9}$$

or

$$F(T+t_s) = \sum_{j=1}^{L} \frac{P_j}{K} \sum_{k=1}^{L} \frac{\alpha_k}{\lambda_k} \left[e^{-\lambda_k (T+t_s-T_j)} - e^{-\lambda_k (T+t_s-T_{j-1})} \right] \quad . \tag{10}$$

A small program, CALDEGS, was written to implement the burst functions to calculate decay power following extended fuel irradiation. The results were checked by comparing them with CINDER-10 integrated spectrum calculations[8] (without neutron absorption) for gamma-power, beta-decay power, and gamma- plus beta-decay power after 20 000 h constant irradiation.

First, a fit to the ^{235}U thermal fission burst decay curves was made with the ERDALEW code. Results are shown in Figs. 14-16. The sharp change in slope at 5×10^{10} s necessitated a 2-segment fit that caused some error in the region 10^{10} s where the segments joined.

Next, the CALDEGS code was used to calculate the decay curves after 20 000 h constant power irradiation time. Results are shown in Figs. 17-19 in units of MeV/fission. Again, the greatest deviation is in the vicinity of 10^{10} s where the 2 fitted burst segments were joined. Such calculations, using the fitted exponentials, are roughly equivalent to calculating a single 3-nuclide chain (or less, because nuclide cross sections and decay energies are not involved). The required storage is negligible and such fits can be used in spatial calculations.

IV. EQUIVALENT BURST FUNCTION FOR FINITE IRRADIATION RESULTS

Equation (1) permits use of general functions $g_k(t)$; therefore, the above techniques can also be applied to reduce experimental results to a burst function basis and thereby enable one to compare results for different irradiation times on a common basis. Current decay-heat and spectral measurements are made following a short irradiation time that can be assumed to have a constant fission rate. Let S = constant fission rate. Then, from the arguments above,

$$F_1(t) = S \sum_{k=1}^{L} \frac{\alpha_k}{\lambda_k} e^{-\lambda_k t}(1-e^{-\lambda_k T}) \quad \left(\frac{MeV}{s}\right) \quad . \tag{11}$$

If we measure the total energy release over interval Δt about $t = t_c$, the average is

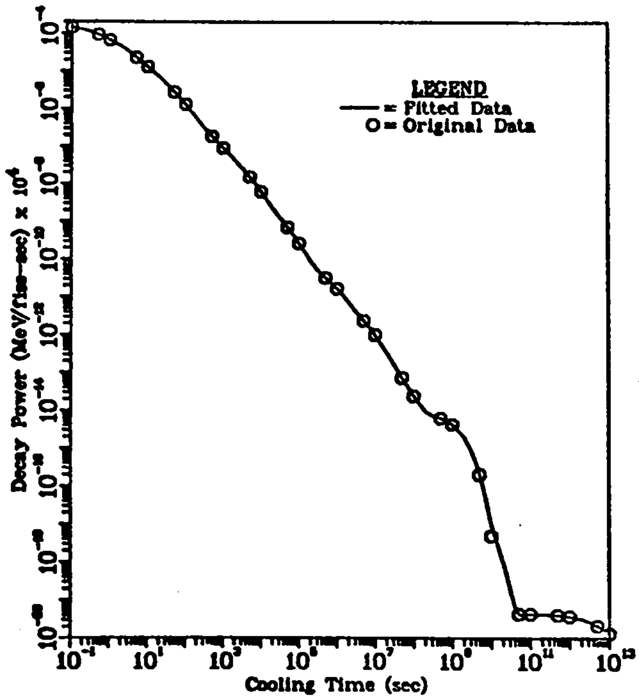

Fig. 14.
Total decay power from ^{235}U thermal burst (gammas and beta$^-$).

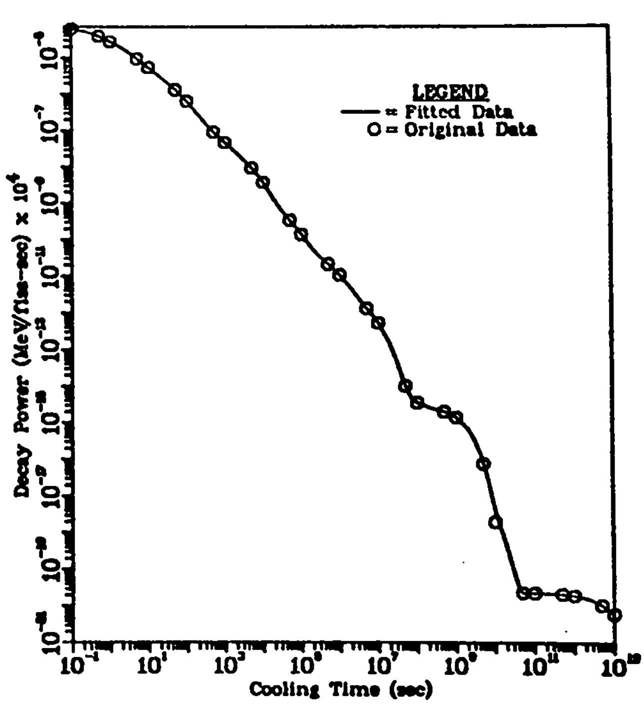

Fig. 15.
Total decay power from ^{235}U thermal burst (gammas only).

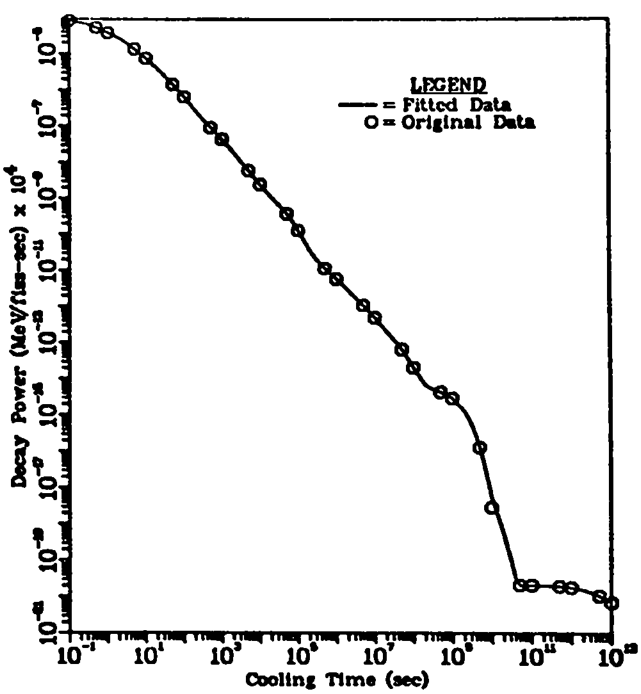

Fig. 16.
Total decay power from ^{235}U thermal burst (beta$^-$ only).

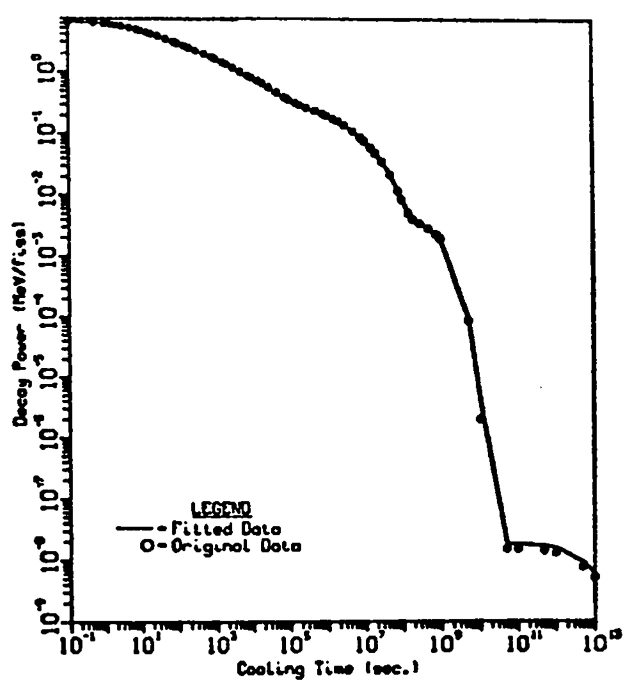

Fig. 17.
Total fission-product decay power from ^{235}U thermal fission after 20 000 h irradiation time (beta$^-$ only).

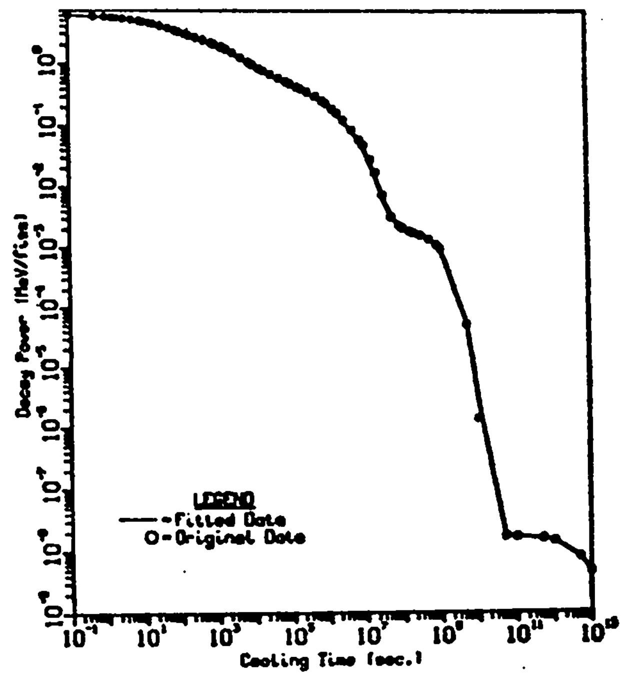

Fig. 18.
Total fission-product decay power from ^{235}U thermal fission after 20 000 h irradiation time (gammas only).

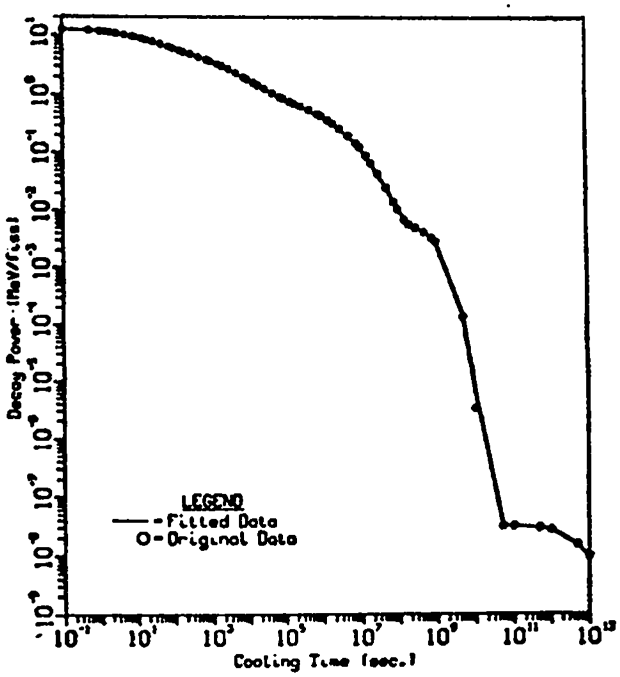

Fig. 19.
Total fission-product decay power from ^{235}U thermal fission after 20 000 h irradiation time (gammas and beta$^-$).

$$\langle MeV/fis \rangle = \int_{t-\frac{\Delta t}{2}}^{t+\frac{\Delta t}{2}} F(t')dt' \quad , \tag{12}$$

where

$$F(t) = \frac{F_1(t)}{S} = \sum_{k=1}^{L} \frac{\alpha_k}{\lambda_k} e^{-\lambda_k t}(1-e^{-\lambda_k T}) \quad . \tag{13}$$

Integrating,

$$\left\langle \text{MeV/fis} \right\rangle = \frac{1}{\Delta t} \sum_{k=1}^{L} \frac{\alpha_k}{\lambda_k^2} (1-e^{-\lambda_k t_c})(e^{\lambda_k \frac{\Delta t}{2}} - e^{-\lambda_k \frac{\Delta t}{2}})$$

$$= \frac{1}{\Delta t} \sum_{k=1}^{L} \frac{\alpha_k}{\lambda_k^2} (1-e^{-\lambda_k T})(1-e^{-\lambda_k \Delta t}) \left[e^{-\lambda_k (t_c - \frac{\Delta t}{2})} \right] \quad , \qquad (14)$$

which are the functions to be used in fitting the experimental data. We have thus obtained an equivalent burst representation of a finite irradiation experiment.

To demonstrate the applicability of this unfolding technique, let us consider the data in Table II, which contains a comparison of experimental results for a 20 000-s ^{235}U thermal irradiation experiment[12] with a CINDER calculation of the same experiment using ENDF/B-IV data. Equivalent burst functions were derived for each set of numbers shown in Table II using the method above. For this calculation, 13 values of λ_k were chosen from $\lambda_k = 0.1$ to $\lambda_k = 1.0 \times 10^{-5}$, amounting to 3 λ_k's per decade. Table III shows the α_k's derived for each case. Finally, each burst function was computed using the derived parameters and compared with a burst function calculated directly by the CINDER code and using ENDF/B-IV data. This comparison is shown in Fig. 20. Note that the parameters derived in unfolding the calculation for the corresponding experiment give a very good fit to the directly calculated burst. The maximum difference of about 5% at the initial 10-s cooling time step probably indicates that a better initial value of λ_k could be chosen. Also, it should be noted that differences between calculation and experiment are much greater when the experiment is unfolded as can be seen by comparing Fig. 20 with Fig. 21, which is an E/C plot for the experiment. The shapes of the curves in the two figures are similar, although the inflection points are somewhat displaced along the cooling time axis. These comparisons indicate that this technique of reducing experiments to equivalent burst functions offers an adequate method for intercomparing experiments and calculations.

TABLE II

MeV/FISSION COMPARISON FOR 20 000-s IRRADIATION EXPERIMENT FOR U-235 THERMAL FISSION

Cooling Time (s)	Experimental Decay Heat (MeV/Fis)	Calculated With CINDER Code and ENDF/B-IV Data	Decay Heat (Exp/Calc)
10	8.1500	7.7795	1.0476
15	7.4600	7.2389	1.0305
20	6.9950	6.8425	1.0223
30	6.3730	6.2761	1.0154
40	5.9500	5.8734	1.0130
50	5.6360	5.5618	1.0133
60	5.3790	5.3087	1.0132
70	5.1620	5.0968	1.0128
80	4.9780	4.9155	1.0127
90	4.8200	4.7579	1.0130
100	4.6810	4.6193	1.0134
150	4.1780	4.1115	1.0162
200	3.8450	3.7804	1.0171
300	3.4190	3.3552	1.0190
400	3.1350	3.0780	1.0185
500	2.9200	2.8727	1.0165
600	2.7460	2.7090	1.0137
700	2.5980	2.5723	1.0100
800	2.4740	2.4546	1.0079
900	2.3630	2.3510	1.0051
1000	2.2640	2.2583	1.0025
1500	1.8860	1.9013	0.9920
2000	1.6270	1.6499	0.9861
3000	1.2830	1.3113	0.9784
4000	1.0670	1.0915	0.9775
5000	0.9111	0.9362	0.9732
6000	0.7998	0.8197	0.9757
7000	0.7195	0.7287	0.9874
8000	0.6480	0.6553	0.9888
9000	0.5886	0.5948	0.9896
10000	0.5401	0.5440	0.9929
15000	0.3803	0.3778	1.0066
20000	0.2918	0.2874	1.0152
30000	0.1947	0.1923	1.0124
60000	0.0860	0.0880	0.9770
100000	0.0450	0.0455	0.9894

TABLE III

ALPHA FITS USING ACTUAL EXPERIMENTAL AND CALCULATED RESULTS

	Lambda	Alpha Fit Using Actual Experiment	Alpha Fit Using Calculated Experiment
1.	1.0×10^{-1}	3.055×10^{-1}	1.766×10^{-1}
2.	5.0×10^{-2}	-7.697×10^{-3}	2.225×10^{-2}
3.	2.0×10^{-2}	5.073×10^{-2}	3.917×10^{-2}
4.	1.0×10^{-2}	1.399×10^{-4}	5.319×10^{-3}
5.	5.0×10^{-3}	7.257×10^{-3}	6.397×10^{-3}
6.	2.0×10^{-3}	5.222×10^{-4}	5.587×10^{-4}
7.	1.0×10^{-3}	1.084×10^{-3}	9.026×10^{-4}
8.	5.0×10^{-4}	3.904×10^{-4}	3.935×10^{-4}
9.	2.0×10^{-4}	1.784×10^{-4}	1.865×10^{-4}
10.	1.0×10^{-4}	8.494×10^{-6}	2.245×10^{-5}
11.	5.0×10^{-5}	3.779×10^{-5}	2.671×10^{-5}
12.	2.0×10^{-5}	-3.667×10^{-7}	4.063×10^{-6}
13.	1.0×10^{-5}	6.375×10^{-6}	5.087×10^{-6}

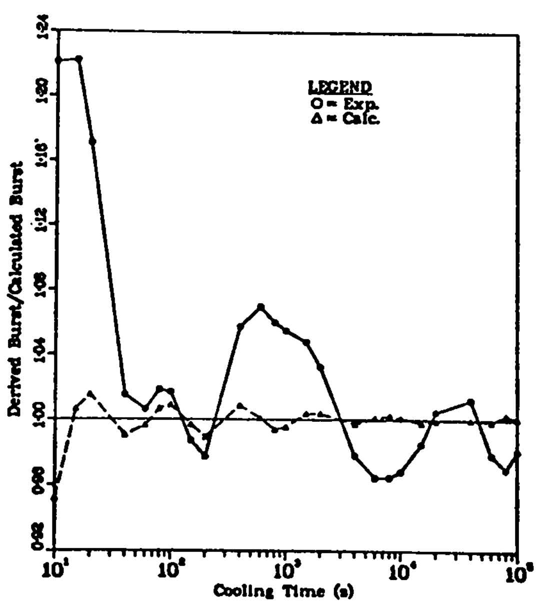

Fig. 20.
Bursts derived from 2×10^4 s irradiation experiment and from CINDER calculation of the same experiment compared with direct CINDER calculation using ENDF/B-IV data.

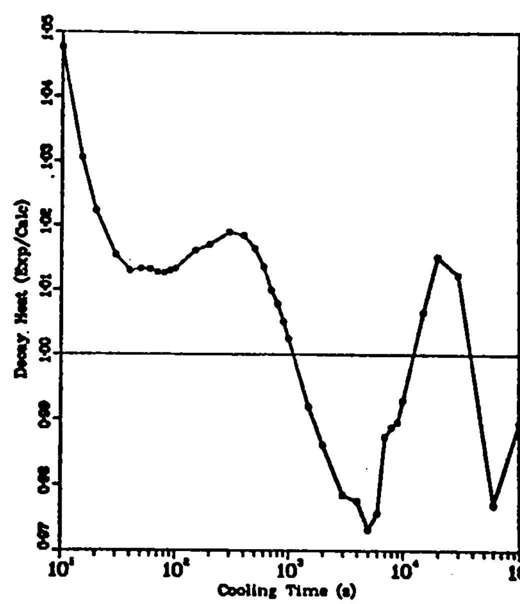

Fig. 21.
^{235}U thermal fission 2×10^4 s irradiation experiment compared with CINDER calculation using ENDF/B-IV data.

V. SUMMARY

We have demonstrated that a linear combination of functions (here exponential functions) can be used to obtain fits to results of summation calculations of fission-product gamma spectra and beta-decay spectra following a ^{235}U thermal fission burst. It was noted that these calculations, to which the fits were made, were obtained by a LASL code system. These codes have been successfully applied to the calculation of and subsequent comparison with recent total decay-heat and spectral measurements. The data-fitting technique was applied to irradiated fuel results, which compared favorably with detailed summation calculations. Typical comparisons of calculated and experimental spectra are given in this report. The total heating has also been compared with a recent LASL experiment. Between 20 and 100 000 s, the calculations are within ~2% of the experimental results. Finally, we have demonstrated that applying these techniques to unfolding experimental results to derive equivalent burst functions provides a sensitive method for intercomparing experiments and calculations.

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