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TITLE: DELAYED NEUTRON SPECTRA BY DECAY GROUP FOR FISSIONING SYSTEMS FROM ^{227}Th THROUGH ^{255}Fm

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DELAYED NEUTRON SPECTRA BY DECAY GROUP FOR FISSIONING SYSTEMS FROM [227]Th THROUGH [255]Fm

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ABSTRACT

The quality and quantity of delayed neutron precursor data has greatly improved over the past approximately 15 years. Supplementation of the data with model calculations and the use of models to extend the number of precursors to 271 is now practical. These data, along with improved fission-product parameters, permit direct calculations of aggregate behavior for many fissioning nuclides. The results can even by approximated using a few (usually six) temporal groups. This paper summarizes an extensive four-year effort to provide a complete set of evaluated data and emphasizes its use to generate the temporal approximations; precursor data and group values are intended for inclusion in ENDF/B-VI.

INTRODUCTION

Most applications of delayed neutrons use an approximate temporal group representation of measured aggregate data.[1] Such data have been limited to the few fissioning nuclides that have aggregate measurements, and even these have inadequate or no spectral measurements.

Improvements in the experimental techniques of isotope separation and neutron spectroscopy have made the study of delayed neutron emission from individual precursor nuclides more practical and productive over the past fifteen or so years. The quantity and quality of the delayed neutron emission probabilities and particularly the neutron emission spectra for the individual nuclides have been greatly improved. Such data are still inadequate for use in calculations of aggregate behavior from individual precursors. We have evaluated the measured data, supplemented it with model spectra for completeness, and added model values for probable unmeasured precursors. The resulting extensive data base will be incorporated into ENDF/B-VI.

This paper can only briefly describe the data base. We will concentrate on its use to produce aggregate delayed neutron yields, halflives, and spectra in the classical six-group representation for 43 fissioning systems from [227]Th to [255]Fm. Group data are also required in ENDF/B-VI. Comments and observations made concerning the use of more than six time groups are included. The application of the data in both its explicit and reduced (six temporal group representation) forms in the point reactor kinetics equations are also discussed. Results from beta-effective calculations in a simple Godiva-type system are presented, but this paper will concentrate on the precursor data base and emphasize the temporal group representations.

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PRECURSOR DATA BASE

Based on energetics, approximately 271 fission products should be delayed neutron precursors. Only a brief description of the types and sources of data for precursors, including fission-product yields, can be given here, along with a summary of the relative importance of the experimental data vs that provided by various model calculations.

Fission-product yields are based on a preliminary evaluation for ENDF/B-VI.[2] This comprises data at one or more neutron incident energies [denoted as thermal (T), fast (F), and high (H) and for spontaneous fission (S)] for 34 fissioning nuclides. Forty-three cases are included in this paper for 28 fissioning nuclides. Most of the yields are based on models.

Emission probabilities (Pn values) for 85 precursors have ben measured[3] and evaluated.[4] The evaluation also provides a fit to the parameters in the systematic Herrmann-Kratz equation[5] used to predict the unmeasured Pn's.

Spectra for 34 precursors have been measured.[6-8] Thirty of these were found to be inadequate in the measured energy range and had to be supplemented with nuclear models.[9,10] The same models were used to estimate the spectra for the remaining 237 precursors. Two models were used. The BETA code[10] was used to extend 30 measured spectra. Otherwise we used a modified evaporation model, described in Ref. 11, because it has the virtue of producing the shape of typical spectra without the need to know, as does the BETA code, energy levels, spins, and parities of precursors and daughters; such data are unknown for most short-leved nuclides. Figure 1 shows a comparison of measured and model results.
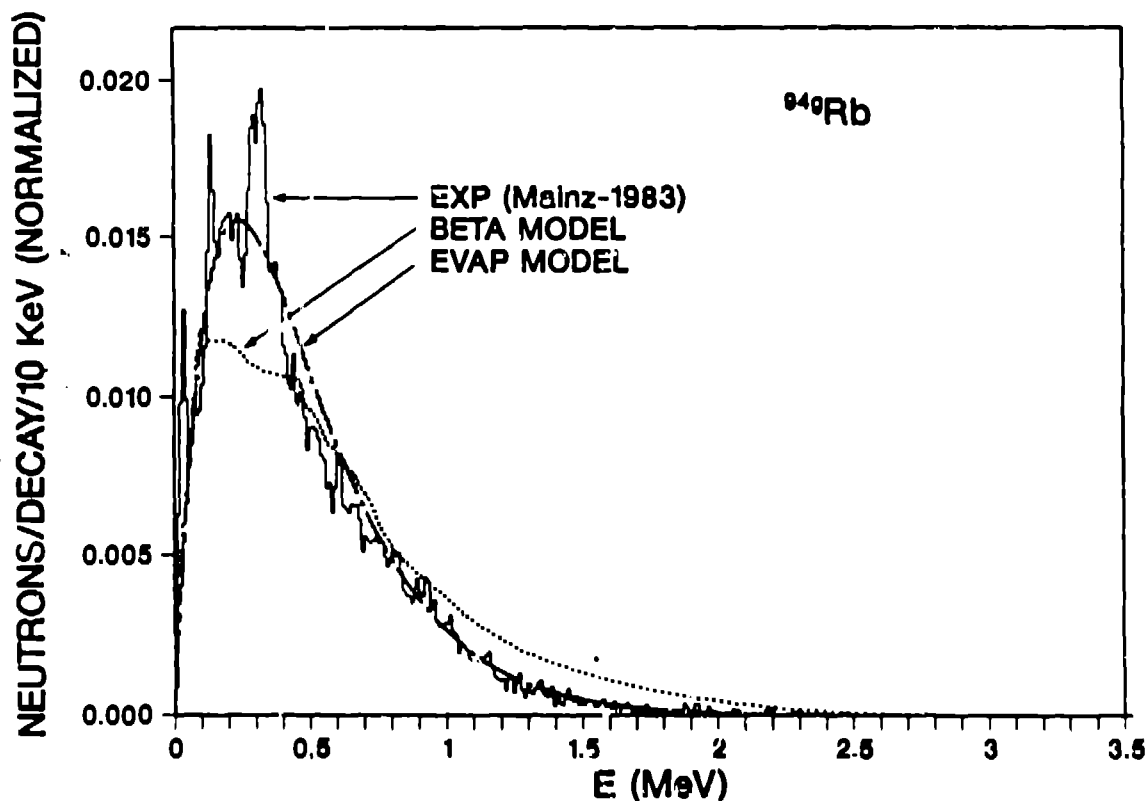

Fig. 1. Delayed neutron spectra for nuclide ⁹⁴ˢRb.

The simple count of nuclides having measured data is misleading. The 85 having measured Pn values account for 80% or more of the total emission rate and the 34 having measured spectra account for 67% or more of the total. These contributions at reactor shutdown depend on the fissioning nuclide; e.g., for ^{235}U thermal fission, the respective contributions are 96% and 84%.

The largest effort in the evaluation was directed at model estimates of unmeasured spectra and the expansion of the incomplete measured spectra. This effort and the models are summarized in Refs. 9 and 11; it will be described in complete detail when Ref. 12 is published.

REDUCTION OF DATA INTO DECAY AND SPECTRA GROUPS

The use of a few temporal groups to represent the behavior of a large, unknown number of precursors started with aggregate experiments. It is still a convenient approximation for use in applications and can be duplicated from aggregate calculations of the individual precursors.

The fission product depletion code, CINDER-10, was used to calculate the activities of all precursor nuclides for various cooling times (to 300 seconds) following a prompt irradiation in each of the fissioning systems. These nuclide activities were folded in with the evaluated emission probabilities to produce aggregate delayed neutron emission values. The delayed neutron activity curves, as illustrated in Fig. 2, can be approximated mathematically as a sum of N exponenetials representing N-time-groups:

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

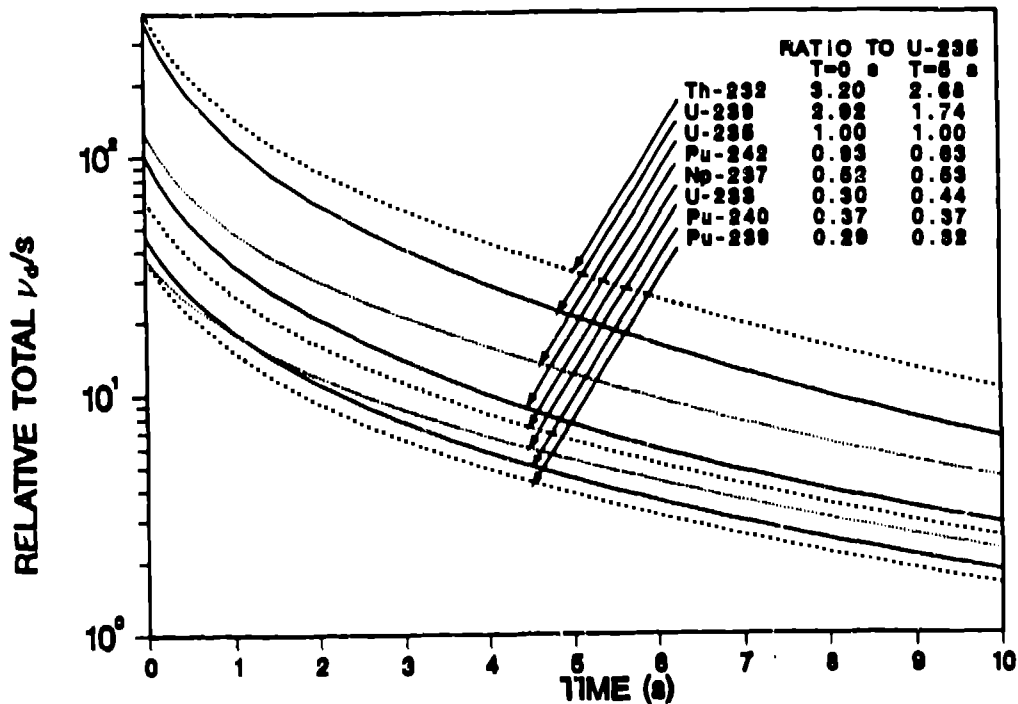|	RATIO TO U-235	
	T=0 s	T=6 s
Th-232	3.20	2.68
U-238	2.02	1.74
U-235	1.00	1.00
Pu-242	0.93	0.63
Np-237	0.62	0.63
U-233	0.30	0.44
Pu-240	0.37	0.37
Pu-239	0.29	0.32

Fig. 2. Total v_d/s for eight fuels vs time (pulse).

The parameters A_i and λ_i in Eq. (1) are determined using a nonlinear, least-squares fitting code. The data represents a pulse irradiation, implying that A_i is the product of the group decay constant, λ_i, and the group yield per fission, a_i.

Initial calculations for $^{235}U(F)$, $^{238}U(F)$, and $^{239}Pu(F)$ were performed using three, six, nine, and twelve groups. Increasing the number of groups from six to nine resulted in a significant improvement in the fit; however, the results from point kinetics calculations using both the six- and nine-group fits for prompt changes in reactivity did not reveal any significant differences. [12,13]

Based on these results and the general acceptance of a six-group representation, the fits for the remaining 40 fissioning systems were performed only for six-groups. Table I presents the normalized group abundances and decay constants for all 43 fissioning sytems.

TABLE I

Delayed Neutron Six-Group Parameters

Fission Nuclide		1	2	Group 3	4	5	6
Th227T	alpha	0.1027	0.2182	0.1304	0.3555	0.1647	0.0284
	lambda	0.0128	0.0354	0.1098	0.2677	0.5022	2.0956
Th229T	alpha	0.0867	0.1907	0.1297	0.3887	0.1729	0.0312
	lambda	0.0128	0.0350	0.1123	0.2760	0.4950	2.0456
Th232F	alpha	0.0364	0.1259	0.1501	0.4406	0.1663	0.0808
	lambda	0.0131	0.0350	0.1272	0.3287	0.9100	2.8203
Th232H	alpha	0.0326	0.0997	0.1431	0.5062	0.1336	0.0848
	lambda	0.0130	0.0350	0.1307	0.3274	0.9638	3.1667
Pa231F	alpha	0.0826	0.2230	0.1608	0.3885	0.1050	0.0401
	lambda	0.0129	0.0347	0.1150	0.2856	0.6706	2.3111
U232T	alpha	0.1360	0.2745	0.1509	0.3052	0.1007	0.0326
	lambda	0.0128	0.0350	0.1073	0.2557	0.6626	2.0254
U233T	alpha	0.0674	0.1927	0.1383	0.2798	0.1128	0.2091
	lambda	0.0129	0.0333	0.1163	0.2933	0.7943	2.3751
U233F	alpha	0.0859	0.2292	0.1781	0.3516	0.1142	0.0409
	lambda	0.0129	0.0347	0.1193	0.2862	0.7877	2.4417
U233H	alpha	0.0900	0.2007	0.1912	0.3684	0.1090	0.0405
	lambda	0.0128	0.0378	0.1271	0.2981	0.8543	2.5314
U234F	alpha	0.0550	0.1964	0.1803	0.3877	0.1324	0.0482
	lambda	0.0131	0.0337	0.1210	0.2952	0.8136	2.5721
U234H	alpha	0.0808	0.1880	0.1791	0.3888	0.1212	0.0420
	lambda	0.0128	0.0364	0.1256	0.2981	0.8475	2.5696
U235T	alpha	0.0380	0.1918	0.1538	0.3431	0.1744	0.0890
	lambda	0.0133	0.0325	0.1219	0.3169	0.9886	2.9544
U235F	alpha	0.0350	0.1807	0.1725	0.3868	0.1586	0.0664
	lambda	0.0133	0.0327	0.1208	0.3028	0.8495	2.8530
U235H	alpha	0.0458	0.1688	0.1769	0.4079	0.1411	0.0595
	lambda	0.0131	0.0356	0.1246	0.2962	0.8260	2.6575
U236F	alpha	0.0302	0.1722	0.1619	0.3841	0.1775	0.0741
	lambda	0.0134	0.0322	0.1202	0.3113	0.8794	2.8405
U236H	alpha	0.0438	0.1540	0.1719	0.4018	0.1578	0.0707
	lambda	0.0131	0.0333	0.1252	0.3030	0.8802	2.8167
U237F	alpha	0.0178	0.1477	0.1445	0.3864	0.2095	0.0941
	lambda	0.0138	0.0316	0.1211	0.3162	0.9073	3.0368

TABLE I (Cont.)

U238F	alpha	0.0139	0.1128	0.1310	0.3851	0.2540	0.1031
	lambda	0.0136	0.0313	0.1233	0.3237	0.9060	3.0487
U238H	alpha	0.0195	0.1184	0.1490	0.3978	0.2081	0.1072
	lambda	0.0135	0.0320	0.1214	0.3142	0.9109	3.0196
Np237F	alpha	0.0400	0.2162	0.1558	0.3633	0.1659	0.0589
	lambda	0.0133	0.0316	0.1168	0.3006	0.8667	2.7600
Np237H	alpha	0.0326	0.1571	0.1589	0.3929	0.1789	0.0796
	lambda	0.0133	0.0322	0.1211	0.2933	0.8841	2.7922
Np238F	alpha	0.0216	0.1845	0.1519	0.3760	0.1861	0.0798
	lambda	0.0136	0.0308	0.1189	0.3077	0.8988	2.9676
Pu238F	alpha	0.0377	0.2390	0.1577	0.3562	0.1590	0.0504
	lambda	0.0133	0.0312	0.1162	0.2888	0.8561	2.7138
Pu239T	alpha	0.0306	0.2623	0.1828	0.3283	0.1482	0.0479
	lambda	0.0133	0.0301	0.1135	0.2953	0.8537	2.6224
Pu239F	alpha	0.0363	0.2364	0.1789	0.3267	0.1702	0.0515
	lambda	0.0133	0.0309	0.1131	0.2925	0.8575	2.7297
Pu239H	alpha	0.0678	0.1847	0.1553	0.3685	0.1750	0.0487
	lambda	0.0129	0.0353	0.1215	0.2885	0.8486	2.5587
Pu240F	alpha	0.0320	0.2529	0.1508	0.3301	0.1795	0.0547
	lambda	0.0133	0.0305	0.1152	0.2974	0.8477	2.8796
Pu240H	alpha	0.0534	0.1812	0.1533	0.3715	0.1849	0.0558
	lambda	0.0130	0.0329	0.1191	0.2918	0.8462	2.7080
Pu241T	alpha	0.0167	0.2404	0.1474	0.3430	0.1898	0.0627
	lambda	0.0137	0.0299	0.1136	0.3078	0.8569	3.0800
Pu241F	alpha	0.0180	0.2243	0.1426	0.3493	0.1976	0.0682
	lambda	0.0136	0.0300	0.1167	0.3069	0.8701	3.0028
Pu242F	alpha	0.0196	0.2314	0.1256	0.3262	0.2255	0.0716
	lambda	0.0136	0.0302	0.1154	0.3042	0.8272	3.1372
Am241T	alpha	0.0305	0.2760	0.1531	0.3122	0.1825	0.0457
	lambda	0.0133	0.0300	0.1145	0.2949	0.8818	2.6879
Am241F	alpha	0.0355	0.2540	0.1563	0.3364	0.1724	0.0454
	lambda	0.0133	0.0308	0.1130	0.2868	0.8654	2.6430
Am241H	alpha	0.0740	0.1757	0.1754	0.3589	0.1783	0.0377
	lambda	0.0129	0.0346	0.1267	0.3051	0.9536	3.3205
Am242mT	alpha	0.0247	0.2659	0.1512	0.3337	0.1756	0.0489
	lambda	0.0135	0.0301	0.1152	0.2994	0.8646	2.8107
Am243F	alpha	0.0234	0.2945	0.1537	0.3148	0.1656	0.0480
	lambda	0.0135	0.0298	0.1138	0.2986	0.8820	2.8111
Cm242F	alpha	0.0763	0.2847	0.1419	0.2833	0.1763	0.0375
	lambda	0.0130	0.0312	0.1129	0.2783	0.8710	2.1969
Cm245T	alpha	0.0222	0.1788	0.1672	0.3706	0.2054	0.0559
	lambda	0.0134	0.0307	0.1130	0.3001	0.8340	2.7686
Cf249T	alpha	0.0246	0.3919	0.1349	0.2598	0.1614	0.0273
	lambda	0.0135	0.0294	0.1053	0.2930	0.8475	2.4698
Cf251T	alpha	0.0055	0.3587	0.1736	0.2693	0.1688	0.0242
	lambda	0.0157	0.0288	0.1077	0.3246	0.8837	2.6314
Cf252S	alpha	0.0124	0.3052	0.1813	0.2992	0.1729	0.0290
	lambda	0.0136	0.0291	0.1068	0.3024	0.8173	2.6159
Es254T	alpha	0.0073	0.3148	0.1547	0.2788	0.2010	0.0435
	lambda	0.0194	0.0289	0.1048	0.3185	0.8332	2.7238
Fm255T	alpha	0.0060	0.4856	0.1766	0.1940	0.1160	0.0218
	lambda	0.0149	0.0287	0.1027	0.3130	0.8072	2.5768

In this table T, F, and H, refer to fission neutron incident
energies of thermal, fast, and high energy and S refers to
spontaneous fission.

Having determined the six-group parameters for each fissioning nuclide, the next logical step was to calculate a consistent set of six-group spectra.

Equation (1) is the mathematical representation of delayed neutron activity following a fission pulse. The physical representation using the individual fission-product precursor data is (ignoring coupling)

$$n_d(t) = \sum_{j=1}^{271} \lambda_j \, Pn_j \, YI_j \, e^{-\lambda_j t} \, , \tag{2}$$

where YI_j is the direct fission yield of nuclide j and Pn_j is its emission probability.

Both Eqs. (1) and (2) describe the delayed neutron activity per fission following a pulse and should be equivalent. In the present evaluation, it is required that

$$A_i e^{-\lambda_i t} = \sum_k f_{k,i} \, \lambda_k \, Pn_k \, YI_k \, e^{-\lambda_k t} \, , \tag{3}$$

where the subscript i represents mathematical group i, the summation is over all precursors contributing to group i, and $f_{k,i}$ is the fraction of delayed neutrons produced by precursor k that contribute to group i. It is assumed that the fission-product precursor may contribute to either or both of the adjacent mathematical groups determined by the decay constants. [Previous analyses of individual precursor data, with respect to its reduction to the six-group representation, simply assigned each precursor to a particular group based on either halflife or λ bounds.[6,14]] The sum of $f_{k,i}$ for any k must be unity. These fractions were determined by requiring the least-squares error

$$\int_0^\infty \left\{ \lambda_k e^{-\lambda_k t} - \left[f_{k,i} \, \lambda_i \, e^{-\lambda_i t} + (1 - f_{k,i}) \, \lambda_{i+1} \, e^{-\lambda_{i+1} t} \right] \right\}^2 dt \tag{4}$$

to be a minimum.[13,15] A modification of Newton's method was used to return the minimum of Eq. (4). The equilibrium group spectra were then found as

$$\Phi_i(E) = \sum_k f_{k,i} \, YC_k \, Pn_k \, \phi_k(E) \, , \tag{5}$$

where $\phi_k(E)$ is the normalized delayed neutron spectrum of precursor k and YC_k is the cumulative yield from fission for precursor k. The normalized six-group spectra for ^{235}U fast and thermal fission are given in Figs. 3-8 over a 1-MeV energy range in comparison with the six-group spectra taken from ENDF/B-V for ^{235}U. Differences between our thermal and fast spectra are much less than those with the ENDF/B-V evaluation. All spectra are normalized such that the integral over 1 MeV is unity in these comparisons.

Using the method outlined above, the group-one spectrum shown in Fig. 3, has three contributing precursor nuclides. The precursor ^{87}Br contributes 100% of its delayed neutrons to group one, as is expected; however, two additional precursors, ^{137}I and ^{141}Cs contribute about 20% of their delayed neutrons to group one. This result allows the group one spectrum to change for different fissioning systems, as suggested by ENDF/B-V data.

Precursor and group spectra are in energy bins of 10 keV for up to 300 groups. In some applications[11] we have used the same bin structure to > 8.5 Mev, but these calculations did not use temporal groups.
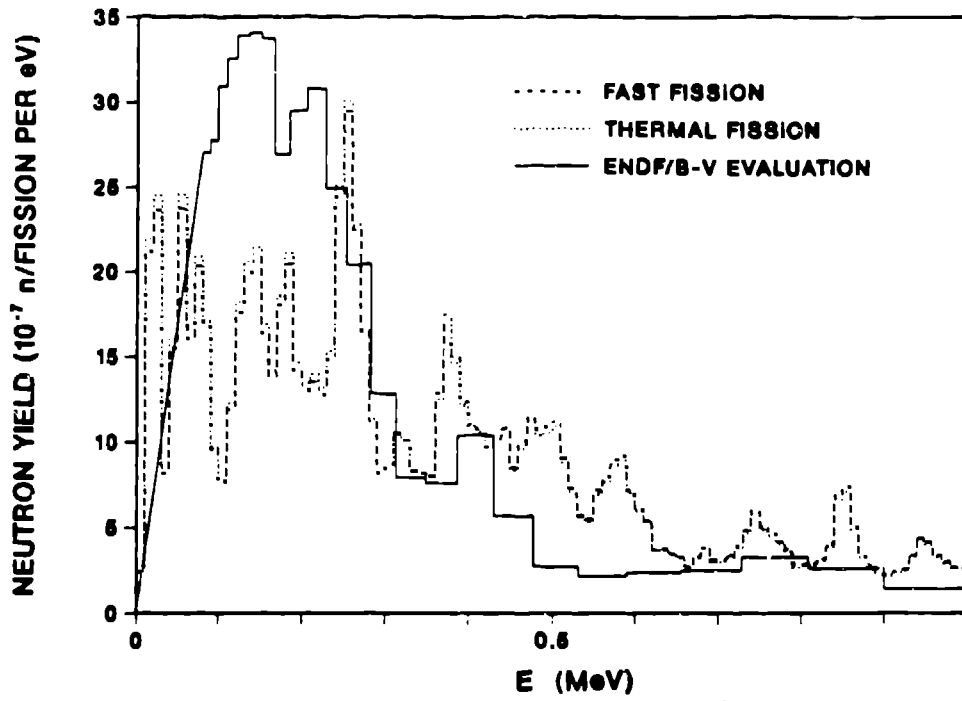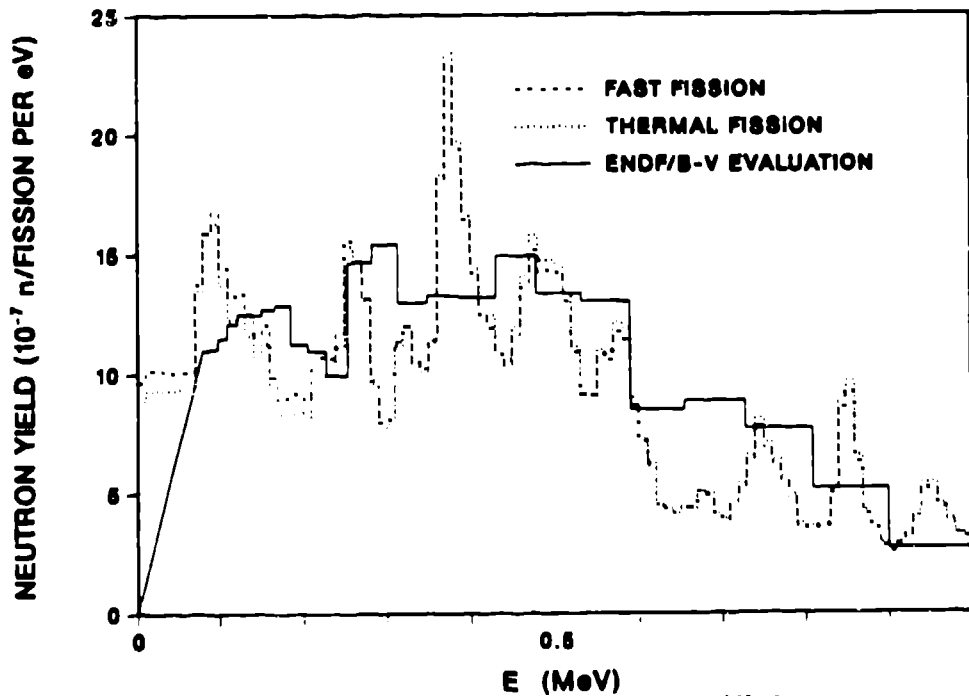

Fig. 3. Group 1 normalized v_d spectra ^{235}U.
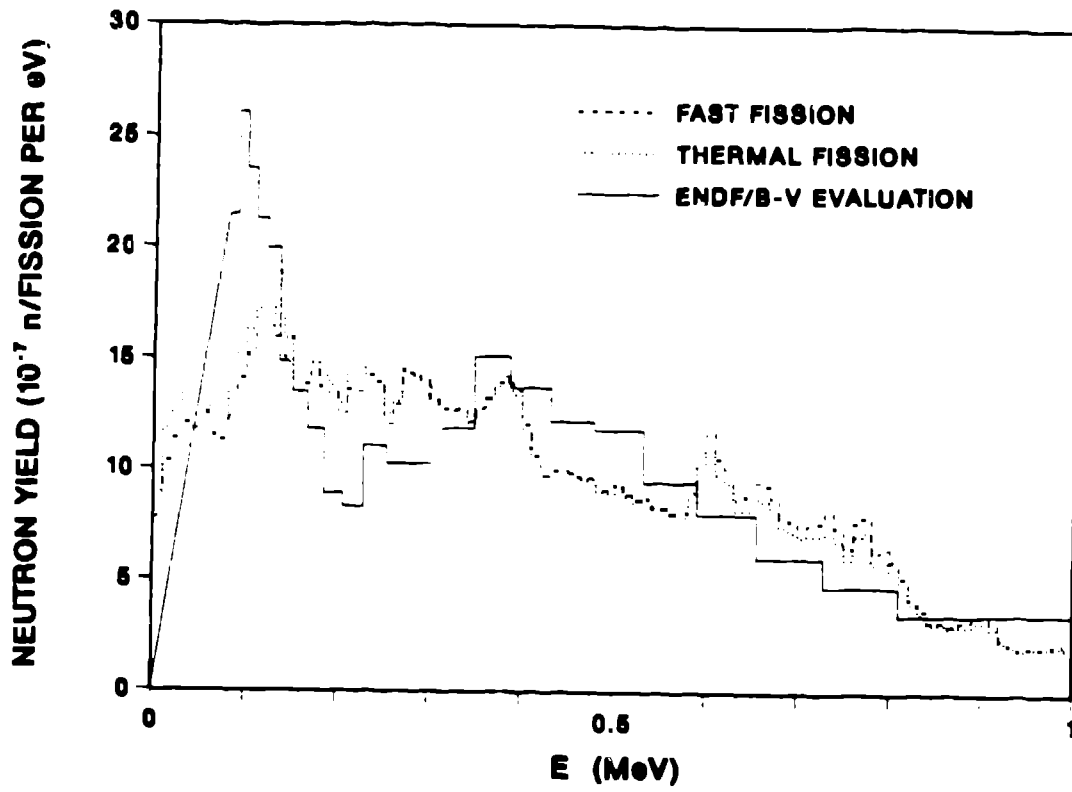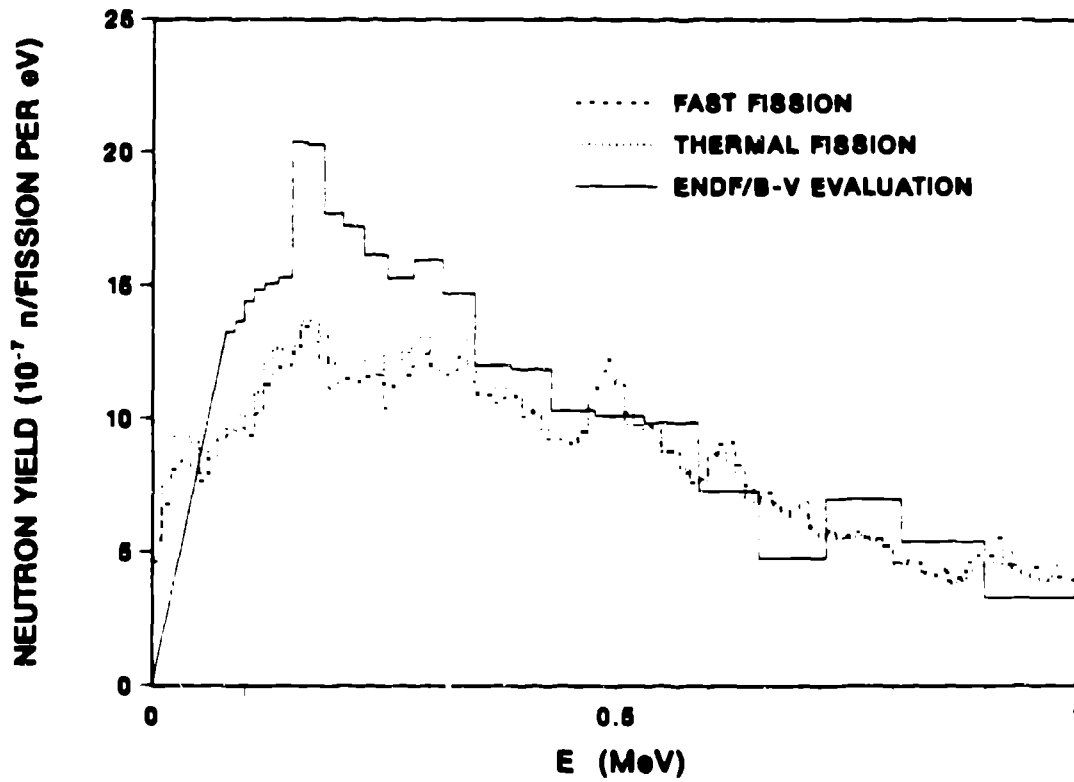

Fig. 4. Group 2 normalized v_d spectra ^{235}U.

Fig. 5. Group 3 normalized v_d spectra ^{235}U.

Fig. 6 Group 4 normalized v_d spectra ^{235}U.
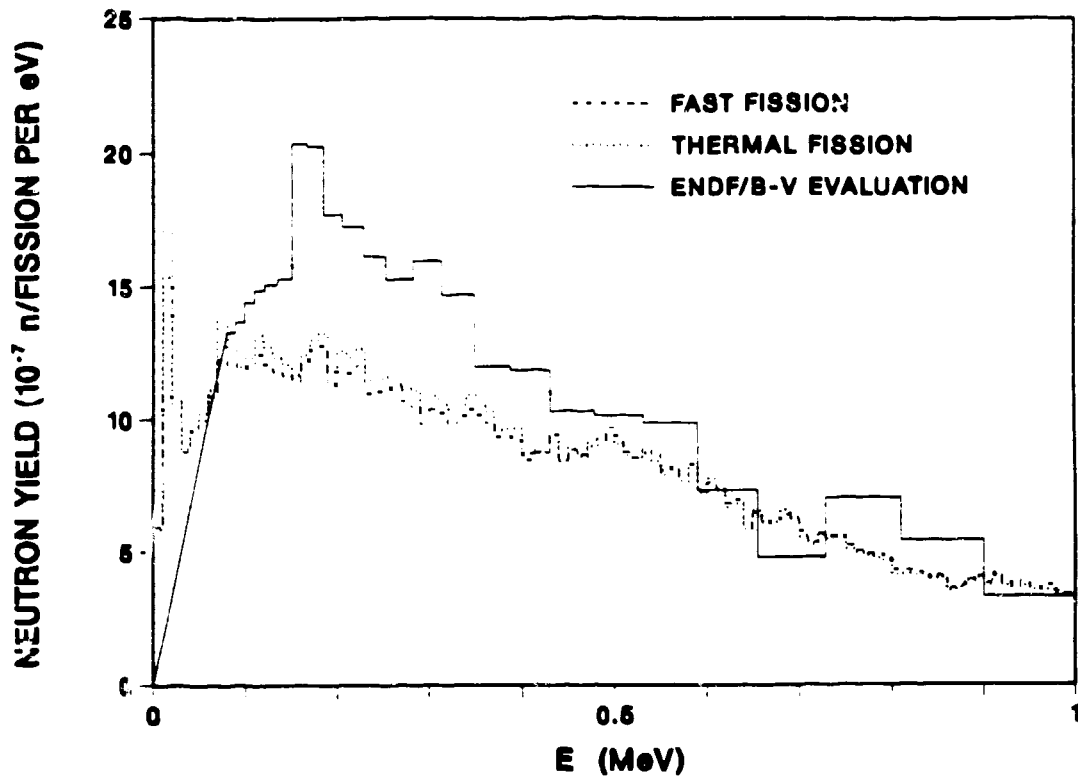
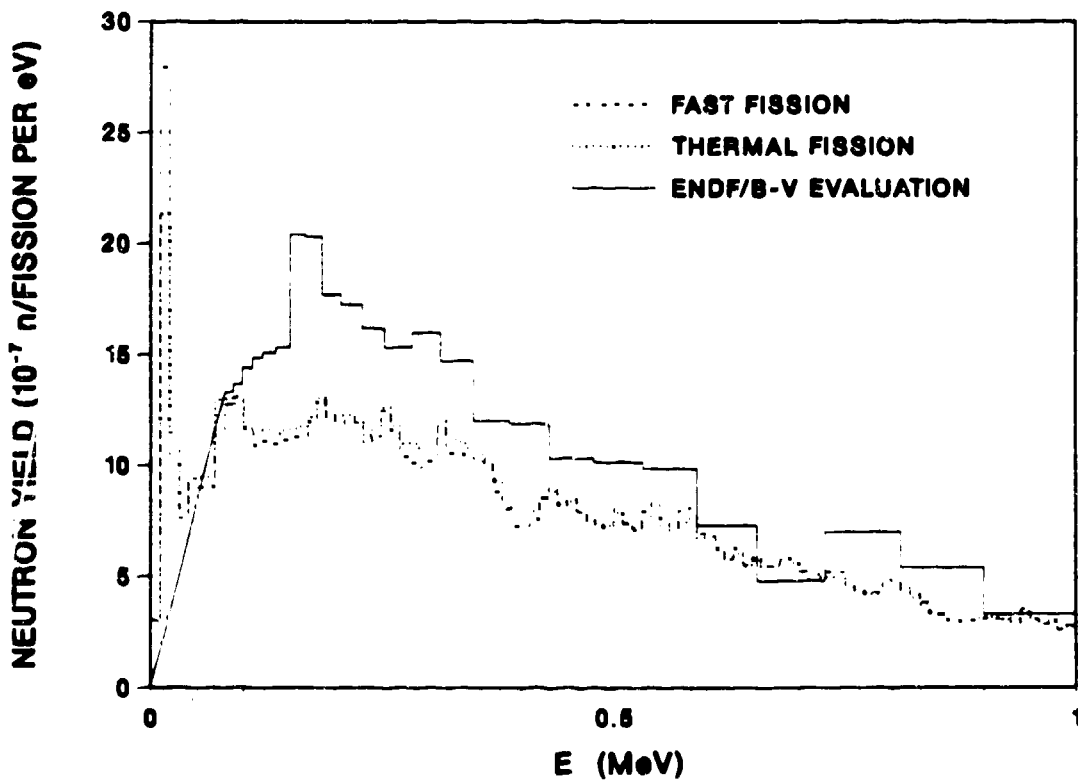Fig. 7. Group 5 normalized ν_d spectra ^{235}U.

Fig. 8. Group 6 normalized ν_d spectra ^{235}U.

APPLICATIONS OF THE GROUP DATA

The accuracy of the six-group parameters is difficult to quantify as it is influenced by not only the uncertainties included in the basic data that was used in calculating the delayed neutron activity curves (i.e., the direct fission yields, halflives, and emission probabilities) but also by the uncertainty introduced by the least-squares fit to that data. Likewise, the calculation of the uncertainties for the group spectra is not straightforward because of the method used to calculate the fractional contribution from each precursor to the various groups.

A reasonable check on the group abundances and decay constants would be to use them in a point kinetics calcuation. These calculations were performed for step changes in reactivitiy in a ^{235}U(F) system and the results are given in Fig 9. The point kinetics equations were modified[13,16] for the calculation using the explicit precursor data. A total of 386 nuclides were required in that calcuation to include the 271 percursors and their parents. Agreement of the fitted six-groups with the explicit calculations is very good. The results using the ENDF/B-V six-group parameters is also given for comparison; the number of delayed neutrons produced is constant for all cases.
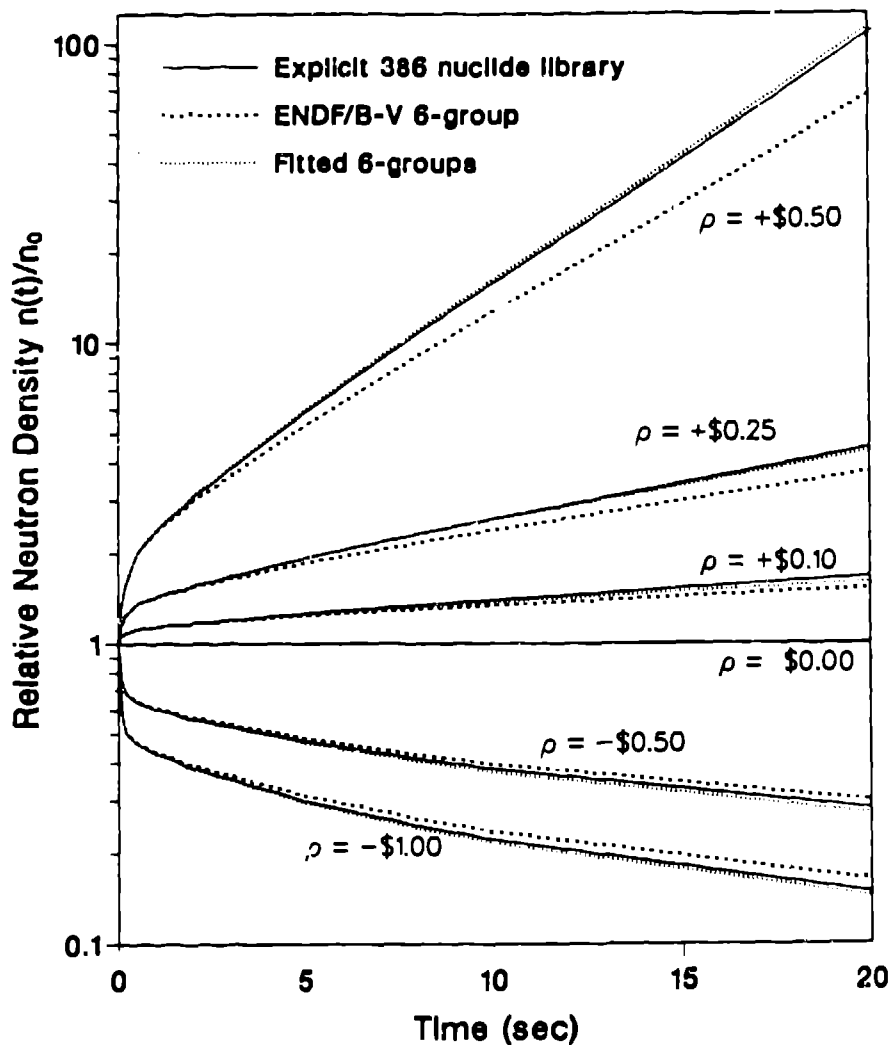

F: -. 9. Calculated neutron density following step reactivity (ρ) inputs for ^{235}U fast fission.

Rossi-alpha (β_{eff}/generation time) calculations were performed to insure that the group spectra produce results which are consistent with those using the aggregate spectra derived from the individual precursor data. The one-dimensional transport theory code, ONEDANT[18] was used to model a Godiva reactor (a bare sphere of enriched ^{235}U metal) and to calculate the fluxes and adjoint fluxes needed for the perturbation calculation. This was accomplished with a modified version of the PERT-V code[19] which uses first order perturbation theory based on the multigroup diffusion model and calculates the effective delayed neutron fraction, β_{eff}, and the mean generation time. The PERT-V code allows the user to input either a single delayed neutron spectrum or individual group spectra, the results for Rossi-alpha, α_{DC}, calculated using each of these options are given below:

Aggregate Spectrum	-1.1185×10^6 s^{-1}
Six-Group Spectra	-1.1123×10^6 s^{-1}.

The perturbation calculations were performed using the six-group decay constants and fractional abundances for $^{235}U(F)$ from Table I, and a total delayed neutron yield of 0.0167 neutrons per fission, which is the value recommended in ENDF/B-V.

The measured value of α_{DC} noted in the Cross Section Evaluation Working Group Benchmard Specifications (BNL 19302), Fast Reactor Benchmark No. 5 is $-(1.11 \pm 0.02) \times 10^6$ s^{-1}. The ratio of the six-group result to the experiment is 1.0020 and that using the aggregate spectrum is 1.0076. These results are in excellent agreement and provide an adequate support of the methods used to derive the six-group spectra.

SUMMARY

An evaluated library for 271 delayed neutron precursors has been completed. Six-group parameters and spectra have been calculated for 43 fissioning systems that are consistent with the explicit precursor results. Although some disagreement with the ENDF/B-V is observed, the major improvement has been in the delayed neutron group spectra and in data produced for unmeasured systems. ENDF/B-V contains six group spectra for only seven fissioning nuclides and these are in a very coarse bin structure that extends only to about 1.2 MeV, whereas the present group spectra cover 28 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). The normalized spectra, and the group constants to a lesser degree, appear to be nearly independent of the incident neutron energy and therefore the $\bar{\nu}_d$ data recommended for inclusion in ENDF/B-VI contain only one set of group constants and spectra (usually that of the fast system) for each of the 28 fissioning nuclides. The delayed neutron yields are a function of incident neutron energy, especially for high energy (14-MeV) fission and are given as they were in ENDF/B-V. The actual values for $\bar{\nu}_d$ recommended for ENDF/B-VI, but not the abundances or spectra, are those taken from the previous ENDF/B-V evaluation[17] or newly evaluated measurements. Nuclides with no reported measurement of $\bar{\nu}_d$ in the literature were assigned the calculated values. We have presented some results related to β_{eff} and point kinetics; these were primarily used to check the validity of the precursor and group data.

ACKNOWLEDGMENTS

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