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TECHNIQUE FOR THE IDENTIFICATION OF DOMINANT DELAYED NEUTRON PRECURSORS

by

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ABASTRACT

The product of cumulative yield and probability of neutron emission is used to assess the relative importance of known delayed neutron precursors. Thirteen precursors are consistently dominant. Nonlinear fits to experimental delayed neutron decay data distinguish the decay constants of the three longest-lived dominant precursors: Br-87, I-137, and Br-88. Sensitivity calculations based on a six-to-seven group transformation lead to a proposed seven-group formulation in which the group decay constants are those of dominant precursors: Br-87, I-137, Br-88, Rb-93, I-139, Br-91, and Rb-96. An alternative six-group formulation is obtained by using the mean of the I-137 and Br-88 decay constants for group 2. Reactivity worth and transient analyses confirm that the positive reactivity scale is preserved the transformation. A known bias in the negative reactivity scale is eliminated by forcing the half-life of the longest-lived group to be the 55.9 s half-life of Br-87. The proposed use of dominant precursor decay constants offers significant simplifications in data analysis and the analysis of fast, epithermal, and thermal reactors with multiple fissioning nuclides.

1. INTRODUCTION

A technique for the identification of delayed neutron precursors has been developed based on the product of cumulative yield and probability of neutron emission. The motivation behind this work is to fix the decay constants of delayed neutrons to those of the dominant delayed neutron precursors. The desirability of identifying a single set of decay constants that would apply to all fissionable isotopes and be independent of the neutron energy spectrum has been addressed by several authors.[1,2,3] The main advantages of a fixed-decay constant representation are simplifying the analysis of epithermal and fast reactors with multiple fissioning isotopes, and improving the fit to experimental data while preserving the inferred positive reactivity scale associated with the original six-group representation.

It is well known that 271 delayed neutron precursors exist, but only a select number of those precursors contribute significantly to the decay of delayed neutrons.[4] Using data compiled by England and Rider, which lists fission yield and probability of neutron emission values for the 271 known delayed neutron precursors in 32 fissioning systems, thirteen precursors were identified that are consistently dominant for all fissioning systems.

The group decay constants that result from nonlinear least squares fits are empirical. They do not, by intent, correspond to actual decay constants of specific delayed neutron precursors. It is well know, however, that Br-87 is the longest lived precursor and the only precursor that contributes significantly to group 1. Occasionally an empirically determined decay constant for group 1 exactly matches the 0.0124 s^{-1} decay constant of Br-87. When the match is not exact, the difference is generally comparable to the estimated experimental error. The similarities in the empirical group decay constants listed in Table I for each of the five other groups suggest that there may be other dominant precursors that are common to a variety of fissioning systems.

2. IDENTIFICATION OF DELAYED NEUTRON PRECURSORS

The precursor yields that are listed by England and Rider are cumulative; that is, they indicate the total time-integrated decays of each precursor per fission accounting for both prompt precursor production and delayed production.[5] The product of a precursor's cumulative yield and its P_n value is the cumulative delayed neutron yield associated with the precursor. This yield-P_n product can be used to assess the relative importance of different precursors. Figure 1 shows the dominant delayed neutron precursors for the fast fission of U-235. The yield-P_n

product is plotted against the half-life of the 271 known delayed neutron precursors. From Figure 1, Br-87 is clearly the only precursor that contributes significantly to group 1. On the other hand, I-137 and Br-88 are both significant contributors to group 2. For all 32 fissioning systems included in the England and Rider database, Br-87, I-137, and Br-88 are the three longest-lived dominant precursors.

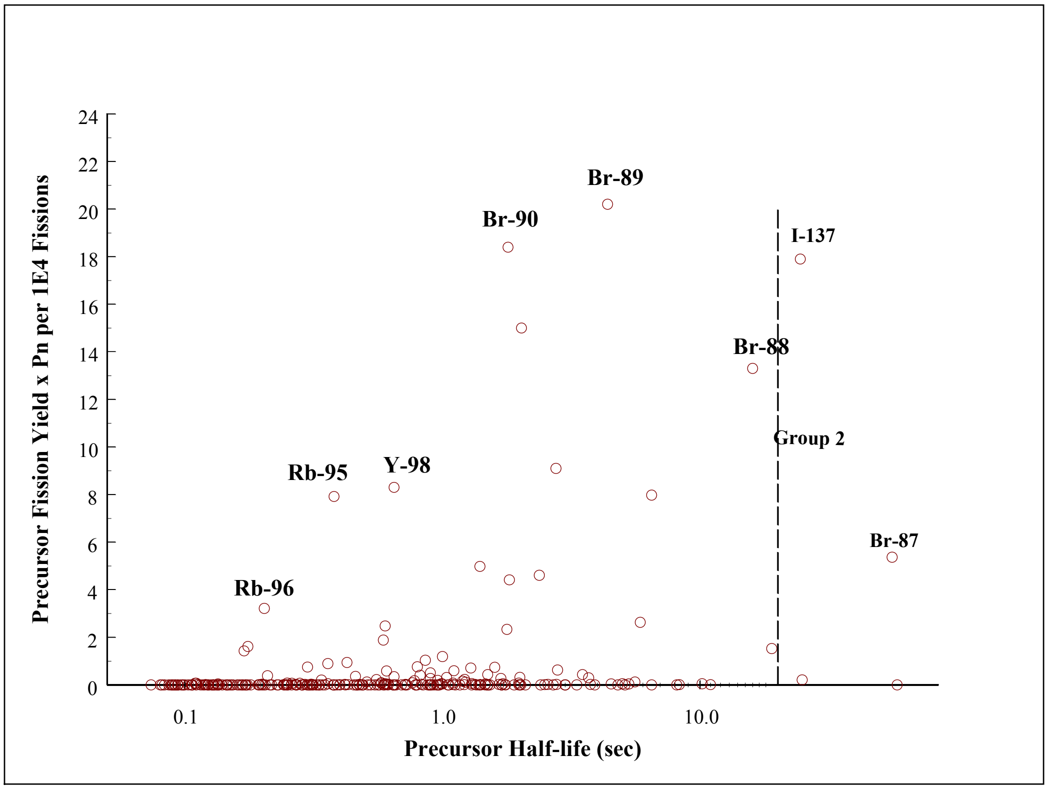

Figure 1. Dominant Delayed Neutron Precursor for U-235F

To substantiate the dominant contributions of Br-87, I-137, and Br-88, experimental decay data for U-235 and Np-237 was reanalyzed.[6] Since the half-life of Br-87 is significantly longer than the half-life of I-137, and in turn the half-life of I-137 is much longer than the half-life of Br-88, it is implicit that a careful analysis of the delayed decay data would allow the observation of these three longest-lived precursors. The method is simply to use the infinite irradiation data from Reference 6 and resolve the half-lives for the three longest-lived precursors individually. This is accomplished by fixing all other parameters and only iterating in one single parameter. For the first case, the iteration is performed on the half-life of Br-87. The linear least-square method yields a half-life value of 54.54 ± 0.92 seconds, which matches within one standard deviation the half-life of Br-87. In order to find the half-life of I-137, a least-square iteration is performed only on λ_2 while keeping all other parameters constant. The fitting routine returns a value of 24.84 ± 0.31 seconds, which is within one standard deviation to the half-life of I-137. Applying the same technique to solve for the half-life of λ_3 yields a slope equivalent of 16.59 ± 0.55 seconds, which corresponds to the half-life of Br-88. This demonstrates that the measured decay delayed neutron data provides experimental confirmation of the presence of Br-87, I-137, and Br-88 as the three longest-lived dominant precursors. Parish and Charlton[7] have also identified Br-87, I-137 and Br-88 as the dominant precursors in groups 1, 2a and 2b from delayed neutron measurements.

The seven-group representation used in this paper differs from the traditional six-group representation by splitting traditional group 2 between Br-88 and I-137, as indicated in Figure 1. The dominant precursors for 12 of the most important fissioning systems are indicated in Table I. For all 32 fissioning systems included in the database, the 13 precursors listed in Table I are dominant. Br-87 is the dominant precursor in group 1. Iodine-137 and Br-88 are dominant in traditional group 2. For the proposed seven-group formulation, the dominant precursor in group 2 is I-137, and the dominant precursor in proposed group 3 is Br-88.

The relative importance ranks within groups 3, 4, and 5 (proposed groups 4, 5, and 6) change from one fissioning system to another, as indicated by the numbers in parentheses in Table I. With various permutations in rank, I-138, Rb-93, and Br-89 consistently dominate group 3 (proposed group 4). Rubidium-94, I-139, and Br-90 consistently dominate group 4 (proposed group 5). Yttrium-98, Br-91, and Rb-95 consistently dominate group 5 (proposed group 6). The dominant precursor in group 6 (proposed group 7) is Rb-96.

3. METHODOLOGY TO IDENTIFY DOMINANT PRECURSORS

Table I suggests a seven-group representation in which the decay constants of a dominant precursor could be used. The dominant precursors in groups 1, 2, 3 and 7 are Br-87, I-137, Br-88 and Rb-96, respectively. No single precursor consistently dominates proposed groups 4, 5, or 6 for all fissioning systems. A sensitivity analysis was, therefore, devised to identify a suitable dominant precursor for each of these groups. The first alternative investigated uses the half-lives of Br-89, Br-90, and Rb-95, which are the dominant precursors with the shortest half-lives (see Table I) in proposed groups 4, 5, and 6. Alternative 2 uses the corresponding dominant precursors Rb-93, I-139 and Br-91 with intermediate half-lives. Alternative 3 uses the corresponding longest-lived dominant precursors I-138, Rb-94, and Y-98.

Each of the three seven-precursor alternatives was tested by transforming six-group yields and decay constants for 12 fissioning systems, and then comparing the delayed neutron component of the inhour equation for the resulting seven-precursor models with those for the original six-group models.

The delayed neutron contribution to the inhour equation is

$$\rho_{\text{delayed}} = \frac{\beta}{1 + \frac{l_p}{T}} \sum_{i=1}^{6} \frac{a_i}{1 + \lambda_i T} \qquad (1)$$

where T is the reactor period, β is the delayed neutron fraction, l_p is the prompt neutron lifetime, and the abundances (a_i for $1 \le i \le 6$) sum to one. The integral characteristics of delayed neutrons must be preserved in transforming from six to seven groups. We use a prime symbol to distinguish constants of the new seven-group representation from those of the original six-group representation. Keeping β and l_p constant while equating delayed reactivities for the six- and seven-group representations requires

$$\sum_{i=1}^{7} \frac{a'_i}{1 + \lambda'_i T} = \sum_{i=1}^{6} \frac{a_i}{1 + \lambda_i T} \qquad (2)$$

Equating the delayed reactivities of both representations as T goes to zero requires that the new abundances sum to one; that is,

$$\sum_{i=1}^{7} a_i' = \sum_{i=1}^{6} a_i \equiv 1 \tag{3}$$

Equating six- and seven-group delayed reactivity terms as T goes to infinity requires that the mean decay time also be preserved; that is,

$$\sum_{i=1}^{7} \frac{a_i'}{\lambda_i'} = \sum_{i=1}^{6} \frac{a_i}{\lambda_i}; \tag{4}$$

The two preceding equations are linear in the seven unknown abundances. Five additional linear equations can be obtained by noting that

$$0 \le \sum_{i=1}^{6} \frac{a_i}{1+\lambda_i T} \le 1 \tag{5}$$

and requiring Equation (2) to hold at the five values of T, which make the right-hand side equal to 1/6, 2/6, 3/6, 4/6, and 5/6. In effect, the transformation breaks the original positive delayed reactivity scale into six increments of equal width and forces the new seven-group representation to match the original reactivity at the seven interval boundaries.

Table I. Dominant Precursors by Fission-yield-P_n Product.

Delayed Neutron Group													
Original	1	2		3			4			5			6
Proposed	1	2	3	4			5			6			7
Dominant Precursor													
Precursor	Br-87	I-137	Br-88	I-138	Rb-93	Br-89	Rb-94	I-139	Br-90	Y-98	Br-91	Rb-95	Rb-96
Half-life (s)	55.9	24.5	16.4	6.5	5.85	4.4	2.71	2.3	1.9	0.59	0.54	0.377	0.199
Fissioning System	Rank Within Group by Fission-Yield-P_n Product												
Th-232F	(1)	(1)	(1)	(2)	(3)	(1)	(3)	(1)	(2)	(3)	(1)	(2)	(1)
U-233F	(1)	(1)	(1)	(2)	(3)	(1)	(2)	(1)	(3)	(1)	(3)	(2)	(1)
U-233T	(1)	(1)	(1)	(2)	(1)	(3)	(1)	(2)	(3)	(1)	(3)	(2)	(1)
U-235F	(1)	(1)	(1)	(1)	(3)	(2)	(3)	(2)	(1)	(1)	(3)	(2)	(1)
U-235T	(1)	(1)	(1)	(2)	(3)	(1)	(3)	(1)	(2)	(2)	(3)	(1)	(1)
U-238F	(1)	(1)	(1)	(2)	(3)	(1)	(2)	(3)	(1)	(3)	(2)	(1)	(1)
Np-237F	(1)	(1)	(1)	(2)	(3)	(1)	(1)	(2)	(3)	(1)	(2)	(3)	(1)
Pu-239F	(1)	(1)	(1)	(1)	(3)	(2)	(1)	(2)	(3)	(1)	(3)	(2)	(1)
Pu-239T	(1)	(1)	(1)	(1)	(3)	(2)	(1)	(3)	(2)	(1)	(3)	(2)	(1)
Pu-240F	(1)	(1)	(1)	(2)	(3)	(1)	(1)	(3)	(2)	(1)	(3)	(2)	(1)
Pu-241F	(1)	(1)	(1)	(1)	(3)	(2)	(1)	(3)	(2)	(1)	(3)	(2)	(1)
Pu-242F	(1)	(1)	(1)	(1)	(3)	(2)	(1)	(2)	(3)	(1)	(3)	(2)	(1)

After performing the sensitivity analysis, alternative 2 resulted with the smallest maximum relative difference. For this reason, it is proposed that decay constants of the precursors associated with alternative 2 be used in lieu of empirically determined group decay constants. These precursors are Br-87, I-137, Br-88, Rb-93, I-139, Br-91, and Rb-96.

Table II presents the abundances obtained for the proposed dominant precursors by applying the transformation method to the conventional six-groups models selected for 12 important fissioning systems. As indicated in the last column of Table V, the maximum relative deviation of the seven-surrogate-precursor formulation from the corresponding empirical six-group formulation is very small, ranging from 1.8×10^{-5} (for Pu-242F) to 4.3×10^{-4} (for Th-232F and U-238F) for the 12 fissioning systems analyzed.

Table II. Abundances of Seven Dominant Precursors

Proposed group, surrogate precursor, and half-life								
Group	1	2	3	4	5	6	7	Maximum Relative Deviation[a]
Precursor	Br-87	I-137	Br-88	Rb-93	I-139	Br-91	Rb-96	
Half-life (s)	55.9	24.5	16.4	5.85	2.3	0.54	0.199	
Fissioning System	Abundances obtained from transformation of empirical Six-group formulations							
Th-232F	0.0346	0.0561	0.1159	0.0930	0.4830	0.1849	0.0325	4.3E-4
U-233F	0.0788	0.1666	0.1153	0.1985	0.3522	0.0633	0.0253	7.3E-5
U-233T	0.0787	0.1723	0.1355	0.1884	0.3435	0.0605	0.0211	6.5E-5
U-235F	0.0339	0.1458	0.0847	0.1665	0.4069	0.1278	0.0344	2.3E-5
U-235T	0.0321	0.1616	0.0752	0.1815	0.3969	0.1257	0.0270	8.2E-5
U-238F	0.0168	0.0239	0.1488	0.0254	0.4650	0.2222	0.0979	4.3E-4
Np-237F	0.0350	0.1983	0.0741	0.1428	0.3822	0.1490	0.0186	2.3E-4
Pu-239F	0.0312	0.2215	0.0670	0.1643	0.3703	0.1183	0.0274	8.9E-5
Pu-239T	0.0301	0.2522	0.0578	0.1728	0.3514	0.1153	0.0204	1.6E-4
Pu-240F	0.0224	0.2056	0.0777	0.1350	0.3914	0.1327	0.0352	2.5E-4
Pu-241F	0.0090	0.1780	0.0658	0.1094	0.4001	0.2004	0.0353	2.8E-4
Pu-242F	0.0023	0.1336	0.0722	0.1145	0.4361	0.2273	0.0140	1.8E-5

4. GOODNESS OF FIT FOR PROPOSED PARAMETERS

In recent analyses of delayed neutron decay data resulting from fast fission in U-235 and Np-237,[6] group abundances and decay constants were determined using the nonlinear least squares method of Levenberg and Marquardt.[8] The same method can be applied to perform the linear least squares fit that is required to determine abundances when the decay constants are set to those of the proposed dominant precursors. Table III summarizes the goodness of fit values obtained for both linear and nonlinear least squares fits to U-235F and Np-237F delayed neutron decay data. All computations were performed using the same computer, the same computer code, the same computational precision, and the same convergence criterion. With either six or seven groups, the goodness of fit obtained using linear least squares with the proposed dominant-precursor decay constants is slightly better (less) than that obtained using the same algorithm to determine both group yields and decay constants by nonlinear least squares. Although the six-group nonlinear fit is better than the seven-group nonlinear fit, the seven-group linear fit is marginally better than the corresponding six-group linear fit.

As indicated in Table III, a nonlinear fit using eight groups was attempted. The initial guesses for the group yields and decay constants were values suggested for fission in U-235 by Spriggs.[3] The eight-group fit did not converge, although fits based on nine and twelve groups did converge. Generally, the more groups used in a nonlinear least squares fit, the more ill-conditioned the equations one must solve, but there are many published sets of delayed neutron parameters that have been fit to eight or more groups. By loosening the convergence criterion, or increasing the computational precision, models with eight or more groups could no doubt be made to converge. However, the analyses presented in this paper are all based on six and seven-group models.

Table III. Goodness of Fit to Experimental Delayed-neutron Decay Data.

Number of Groups	Fissioning System	Type of Least Squares Performed	Reduced Chi-square
6	U-235F	Nonlinear	1.032
6	U-235F	Linear	1.006
6	Np-237F	Nonlinear	1.057
6	Np-237F	Linear	1.011
7	U-235F	Nonlinear	1.135
7	U-235F	Linear	1.003
7	Np-237F	Nonlinear	1.141
7	Np-237F	Linear	1.008
8	U-235F & Np-237F	Nonlinear	Did Not Converge

5. CONCLUSION

This study identified thirteen delayed neutron precursors that are consistently dominant out of 271 precursors for 32 fissioning systems. A rationale for using the decay constants of 7 of these dominant precursors in lieu of empirically determined decay constants has been presented. When the transformation used to develop this rationale is applied to widely used six-group reactivity models for important fissioning systems, the original positive reactivity scale is maintained. The goodness of fit to experimental delayed neutron decay data improved by using linear least squares with the suggested fixed decay constants instead of nonlinear least squares in which both decay constants and abundances are estimated.

It is not claimed that fixed-decay-constant representations are "better" than the original six-group models. Indeed the essential part of the reactivity scale is well preserved in transforming widely-used six-group models to corresponding fixed-decay constant models. The main advantages of a fixed-decay constant representation are simplifications in the analysis of epithermal and fast reactors with multiple fissioning isotopes, and improves the fit to experimental data while preserving the inferred positive reactivity scale associated with the original six-group representation.

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