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CALCULATION OF PLUTONIUM CRITICAL EXPERIMENTS USING ENDF/B-VI DATA

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INTRODUCTION

In the United States, plutonium data are needed to predict cycle lengths for reactors, burnup credit for transportation of discharged fuel, and certification of the nuclear safety margins of fuel repositories. Foreign countries have additional needs including nuclear safety in reprocessing, recycle fuel fabrication, and computation of reactor physics parameters for mixed oxide fuel cycles. Neutron spectra found in these applications span the energy range from thermal to a few MeV. Since most facilities rely on computational methods to demonstrate that operations are being conducted within approved limits, the accuracy of calculated k-effectives must be identified over a broad energy range. In 1995, a new data library was released[1] based on data from the ENDF/B-VI data files[2]. A study was initiated to determine if use of the new library resulted in significant changes in calculated k-effectives over those based on ENDF/B-V data.[3-4]

DATA LIBRARIES

ENDF/B-VI data were collapsed to a 199 group structure through the use of the NJOY[5] program. "ENDF/B-VI contains numerous significant changes relative to earlier versions. Improved experimental data and model predictions are included and several format changes were made to provide for better representation of the underlying physics and the extension to higher energies. ... Significant changes include: new resonance region evaluation for ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu (first updates since ENDF/B-III), simultaneous evaluation of several neutron standard cross sections, ... Reich-Moore formalism for resonance region(s) and representation (of) separate isotopic evaluations for structural elements[1]."

ENDF/B-V data were collapsed to a 238 group structure through the use of the AMPX[6] collection of programs. Limited validation studies of Pu-bearing experiments - 4 with fast spectra, 8 with thermal - are reported in Ref. 1. Calculated k-effectives ranged from 0.995 to 1.015.

Both libraries require further processing before the cross-section data can be used by neutron transport computer programs. The resonance energy region data for various nuclides are included in the libraries in a parametric fashion, enabling the user to generate "psuedo-problem-dependent" libraries based on the physical properties of the system. The term "psuedo" is applied because only the resonance energy region data can be adjusted, not the entire energy spectrum. The method applied to the unresolved resonance range is the same for both libraries but different methods are used for the resolved resonance range. Even if there were no differences in the data contained in the two collections, potentially significant differences in calculated k-effectives could result from the differing resonance treatments.

Resonance processing for the 199 group library is accomplished by the Bondarenko (f-factor) method. This method is essentially interpolation of a tabulation of resonance cross section values. The tabulation is as a function of temperature and the "background" cross section of the system[6]. "All materials (are) processed at temperatures of 300, 600, 1000, and 2100 degrees K and most materials (are) processed with 6 to 8 values of the background cross section."[1]

The resolved resonance range, which varies according to nuclide, for the 238 group library is formulated so that a Nordheim Integral Treatment[7] must be performed by executing the NITAWL module of AMPX. One nuclide, ^{241}Pu, has Bondarenko parameters for both the unresolved and resolved resonance energy ranges.

COMPUTATIONAL METHODS

All calculations were performed with modules from the SCALE [8] system. Monte Carlo calculations were performed for all benchmarks with

the KENO-V.a program as executed under the CSAS25 module. For spherical benchmarks, one-dimensional discrete ordinates calculations were performed with the XSDRNPM program as executed under the CSAS1X module.

SELECTED BENCHMARK EXPERIMENTS

The suite of benchmarks evaluated are those described in Appendix E of Ref. 9. Detailed descriptions of the experiments are contained in Refs. 10-12. Reference 10 notes the following criteria for experiment selection.

> The first criterion for the plutonium systems is to cover a wide range of H/Pu ratios. A wide range of H/Pu ratios implies a wide variation in the neutron spectrum which will emphasize the energy dependence of the neutron cross sections. The second criterion is to cover a range of ^{240}Pu contents... A third criterion is to minimize leakage, because the leakage calculation introduces another uncertainty into the analysis... Unreflected systems were chosen wherever possible because the reflector perturbs the neutron flux spectrum and adds an uncertainty to the calculation... (W)here more than one experiment is available for a given H/Pu ratio and ^{240}Pu content, the most recent experiment was selected for benchmark purposes because we deemed the more recent experiments to be the most reliable data.

Brief descriptions of the experiments are provided in Tables I and II. The Koponen citation number refers to a compilation of critical experiments prepared under the direction of B. L. Koponen, Lawrence Livermore Laboratory.[13] Reference 10 is assigned the I.D. of 2110, Ref. 11 is 2109 and Ref. 12 is 1727.

RESULTS OF CALCULATIONS

K-effectives and spectral indices from the 238 group ENDF/B-V calculations and from the ENDF/B-VI calculations are shown in Table III. Results for Expt. 2110-29 could not be provided as that experiment contained gadolinium, an element not available in the 199 group library. Data from both sets of calculations are presented graphically in Fig. 1.

Several phenomena are visible in Fig. 1. Critical experiment spectra representative of the entire range of neutron energies appear to be represented. The spread in calculated k-effectives is large, considerably larger than the uncertainties in individual calculated k-effectives which are documented in Table III. The energy range of 10 to 1000 eV appears to have the largest disagreement between calculation and experiment - about 2.5%.

The comparison between ENDF/B-VI and V values is highlighted in Fig. 2. The differences between the 199 group k-effective and the 238 group k-effective for each benchmark are plotted as a function of spectral index for the benchmark.

Table I. Plutonium benchmarks

Koponen citation IDs	ID # source reference	Description
2110-1	1	Infinite system of ^{239}Pu and water
2110-2	2	Solution with high Pu content
2110-3	3	Solution with low Pu content
2110-4	4	Solution with low Pu content
2110-5,6	5	Polystyrene compacts with low ^{240}Pu
2110-34	6	Metal with low ^{240}Pu, Jezebel expts.
2110-7	7	Solution with intermediate Pu content
2110-8 to 2110-12	9	Polystyrene compact with high ^{240}Pu
2110-14	10	Solution with low Pu content
2110-15 to 2110-22	11	Polystyrene compact with very little H
2110-23 to 2110-28	12	Polystyrene compact with high ^{240}Pu
2110-13	13	Metal with high ^{240}Pu
2110-29	14	Solution with very high ^{240}Pu
2110-30, 31	15a	Solution with stainless steel shell
2110-32, 33	15b	Solution with extra stainless steel shell
2109-20	20	Solution with 25.4 cm concrete ref.
2109-21	21	Solution with 10.16 cm concrete ref.
2109-22	22	Solution with 10.16 cm concrete ref.
2109-23	23	Solution with Cd shell and 10.16 cm concrete ref.
2109-24	24	Solution with Cd shell and 32 cm water ref.
2109-25	25	Metal with natural U ref.
1727-09	IIIA1-13	Metal with natural U ref.
1727-10	VA-1	Metal with no ref.

Table II. Characteristics of critical experiments

Koponen citation ID no.[a]	H/Pu atom ratio	% ^{240}Pu ratio	Pu density (g/cm^3)	Geometry	Reflector
2110-1	3695	0	0.007	Infinite	None
2110-2	125	5	0.172	Sphere	None
2110-3	980	1	0.026	Sphere	30 cm water
2110-4	758	5	0.034	Sphere	6.6 mm steel
2110-5	15	2	1.12	Parallelpiped	None
2110-6	15	2	1.12	Parallelpiped	Plexiglas
2110-34	0	5	15.6	Sphere	None
2110-7	422	5	0.058	Slab	None
2110-8	50	18	0.37	Parallelpiped	None
2110-9,12	50	18	0.37	Parallelpiped	Plexiglas
2110-14	210	8	0.116	Cylinder	20 cm water
2110-15,16	0	18	5.8	Parallelpiped	None
2110-17,22	0	18	5.8	Parallelpiped	Plexiglas
2110-23	5	11	2.3	Parallelpiped	None
2110-24,28	5	11	2.3	Parallelpiped	Plexiglas
2110-13	0	20	15.7	Sphere	None
2110-29	623	43	0.041	Cylinder	20 cm water
2110-30,31	1067	4.6	0.024	Sphere	20 cm water
2110-32,33	1031	4.6	0.025	Sphere	2 mm steel + 20 cm water
2109-20	684	4.6	0.036	sphere	25.4 cm concrete
2109-21	684	4.6	0.0355	sphere	10.16 cm concrete
2109-22	496	4.6	0.0452	sphere	10.16 cm concrete
2109-23	454	4.6	0.0509	sphere	0.762 mm Cd + 10.16 cm concrete
2109-24	540	4.6	0.0469	sphere	0.762 mm Cd + 32 cm water
2109-25	0	4.8	15.375	sphere	19.6 cm natural U
1727-09	0	5.5	19.74	cylinder	water
1727-10	0	4.9	15.62	sphere	none

[a] Comma denotes inclusive, i.e., 17,22 means experiments 17 through 22.

Thermal benchmarks, those with energies less than 0.1 eV, appear to calculate slightly lower (0.003) with ENDF/B-VI than with V. Above 0.1 eV, only three differences are more than two standard deviations from zero. Two of these, 2110-23 and 2110-24, are experiments conducted with cadmium placed between the fissile material and the reflector. The third calculation, for 2110-05, is for a bare, plastic moderated oxide critical experiment. Considering the calculated k-effectives for a similar experiment, 2110-06, there is an indication that the 238 group calculation is unusually low.

Since its first reported use for nuclear criticality code validation[14], the parameter "average energy group of a neutron causing fission" (AEG) has been used as a spectral index to establish acceptable correlations of the AEG with computational biases and ranges of validation applicability. The AEG is not constrained to the presence of particular nuclides in a system as is H/fissile atom ratio. It allows for validation of codes/data with critical experiments having moderators different from the system being evaluated and it is easy to compute.

Because of energy-group boundary differences, calculations from both the 199 and 238 group libraries revealed that AEG is not an accurate comparative spectral index for the energy range of 100 to 50000 eV. Figure 3 shows a comparison of the spectral index based on energy corresponding to the average energy group for fissions (EAEG) to the index based on energy of the average lethargy (EAL) causing fission. The EAL is defined in Eq. (1):

$$EAL = \sum_i \frac{\left(ln E_i^U + ln E_i^L\right)}{2} F_i \Big/ \sum_i F_i \qquad (1)$$

where E_i^U and E_i^L are the upper and lower energy boundaries of group i and F_i is the number of fissions in group i. The EAL was defined by W. C. Jordan, Oak Ridge National Laboratory and is expected to be a part of the next version of the KENO program. The two indices agree for fast and thermal systems but vary greatly for epithermal systems.

Figure 4 provides a comparison of the EAEG index from the 199 group library to the EAEG index from the 238 group library. The ratio, which ideally should be very close to 1.0, varies by a factor of 1000 at high epithermal energies. In contrast, the ratios of the lethargy based indices, EALs, differ

Table III. Calculated Physics Parameters for Selected Pu-bearing Critical Experiments[a]

ID	238 group ENDF/B-V				199 group ENDF/B-VI			
	k-eff	1 std dev for k-eff	EAEG[b]	EAL[c]	k-eff	1 std dev for k-eff	EAEG[b]	EAL[c]
1727-09	0.9974	0.0018	5.38E+4	8.57E+4	0.9926	0.0016	3.63E+5	8.48E+4
1727-10	0.9940	0.0008	1.02E+6	1.14E+6	0.9941	0.0008	1.19E+6	1.10E+6
2109-20	1.0289	0.0012	1.28E-1	6.65E-2	1.0214	0.0012	1.00E-1	6.58E-2
2109-21	1.0234	0.0012	1.22E-1	6.47E-2	1.0221	0.0012	9.71E-2	6.39E-2
2109-22	1.0260	0.0011	1.69E-1	7.85E-2	1.0223	0.0011	1.21E-1	7.75E-2
2109-23	1.0223	0.0012	1.93E-1	8.64E-2	1.0175	0.0013	1.32E-1	8.50E-2
2109-24	1.0160	0.0015	1.60E-1	7.55E-2	1.0142	0.0015	1.16E-1	7.43E-2
2109-25	1.0059	0.0008	9.99E+5	1.09E+6	1.0051	0.0008	1.20E+6	1.09E+6
2110-01	0.9989	0.0007	6.17E-2	3.83E-2	0.9965	0.0007	4.63E-2	3.80E-2
2110-02	1.0048	0.0016	6.89E-1	3.14E-1	1.0029	0.0015	4.29E-1	3.05E-1
2110-03	1.0088	0.0017	9.03E-2	5.33E-2	1.0036	0.0018	7.29E-2	5.23E-2
2110-04	1.0102	0.0017	1.13E-1	6.24E-2	1.0079	0.0016	9.17E-2	6.11E-2
2110-05	1.0260	0.0009	9.94E+0	3.30E+1	1.0332	0.0009	1.30E+2	3.18E+1
2110.06	1.0295	0.0019	2.32E+0	5.58E+0	1.0320	0.0017	8.63E+0	5.58E+0
2110.07	1.0143	0.0009	2.02E-1	8.89E-2	1.0120	0.0009	1.37E-1	8.78E-2
2110-08	1.0047	0.0008	1.15E+0	1.59E+0	1.0052	0.0008	1.14E+0	1.54E+0
2110-09	1.0114	0.0016	1.04E+0	7.35E-1	1.0088	0.0015	8.46E-1	7.26E-1
2110-10	1.0067	0.0016	1.04E+0	7.22E-1	1.0096	0.0015	8.27E-1	7.02E-1
2110-11	1.0084	0.0016	1.04E+0	7.12E-1	1.0075	0.0015	8.21E-1	6.94E-1
2110-12	1.0059	0.0017	1.04E+0	7.02E-1	1.0072	0.0014	8.13E-1	6.85E-1
2110-13	0.9938	0.0009	1.13E+6	1.27E+6	0.9936	0.0009	1.36E+6	1.26E+6
2110-14	1.0061	0.0026	3.39E-1	1.44E-1	1.0029	0.0024	2.25E-1	1.41E-1
2110-15	0.9991	0.0011	5.43E+5	2.73E+5	1.0029	0.0009	5.00E+5	2.63E+5
2110-16	1.0240	0.0008	9.39E+5	1.01E+6	1.0228	0.0008	1.10E+6	1.01E+6
2110-17	1.0191	0.0024	7.11E+1	1.78E+3	1.0170	0.0028	3.99E+4	1.86E+3
2110-18	1.0368	0.0019	1.14E+2	3.93E+3	1.0343	0.0019	8.41E+4	4.02E+3
2110-19	1.0008	0.0019	1.43E+2	4.72E+3	1.0040	0.0016	9.59E+4	4.83E+3
2110-20	1.0248	0.0014	7.92E+1	2.46E+3	1.0247	0.0013	6.18E+4	2.48E+3
2110-21	1.0312	0.0014	6.94E+1	1.96E+3	1.0303	0.0013	5.01E+4	1.97E+3
2110-23	1.0094	0.0008	1.35E+2	1.59E+3	1.0159	0.0008	2.37E+4	1.52E+3
2110-24	1.0232	0.0014	2.12E+1	9.23E+1	1.0293	0.0011	1.38E+3	9.08E+1
2110-25	1.0234	0.0012	1.97E+1	8.38E+1	1.0250	0.0012	1.24E+3	8.47E+1
2110-26	1.0226	0.0012	1.67E+1	6.69E+1	1.0270	0.0012	8.50E+2	6.74E+1
2110-27	1.0249	0.0012	1.48E+1	5.72E+1	1.0246	0.0011	6.45E+2	5.70E+1
2110-30	1.0114	0.0015	8.65E-2	5.07E-2	1.0082	0.0015	6.86E-2	5.03E-2
2110-32	1.0086	0.0016	8.90E-2	5.21E-2	1.0105	0.0015	7.14E-2	5.16E-2
2110-34	0.9937	0.0008	1.10E+6	1.23E+6	0.9937	0.0009	1.32E+6	1.23E+6

[a]E-x = 10^{-x}

[b]EAEG=Energy corresponding to average energy group for fissions.

[c]EAL=Energy of the average lethargy causing fission.

Fig. 1. Calculated k-effectives for plutonium fueled critical experiments

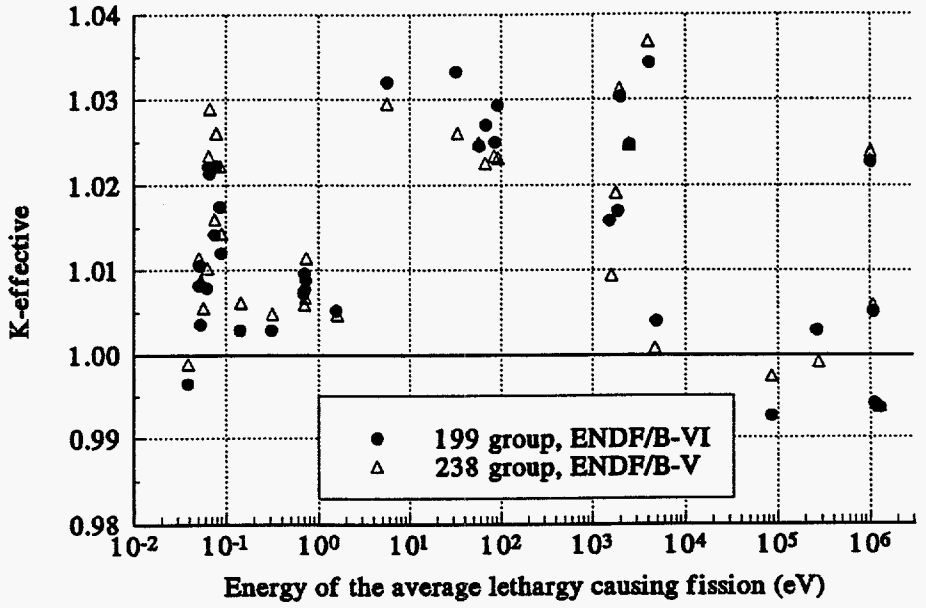


Fig. 2. Changes in k-effectives due to updated cross-section data

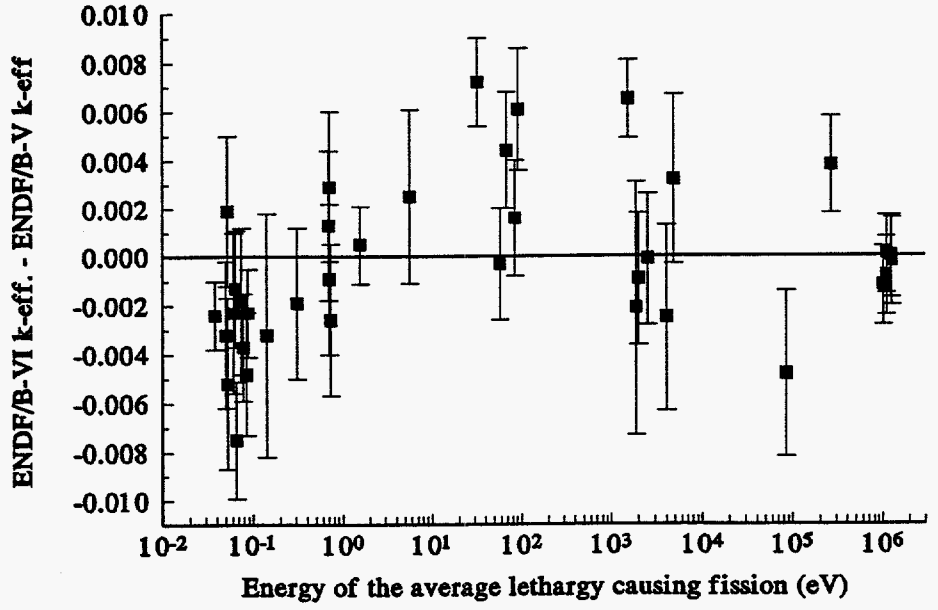


Fig. 3. Comparison of spectral indices for 199 group, ENDF/B-VI library

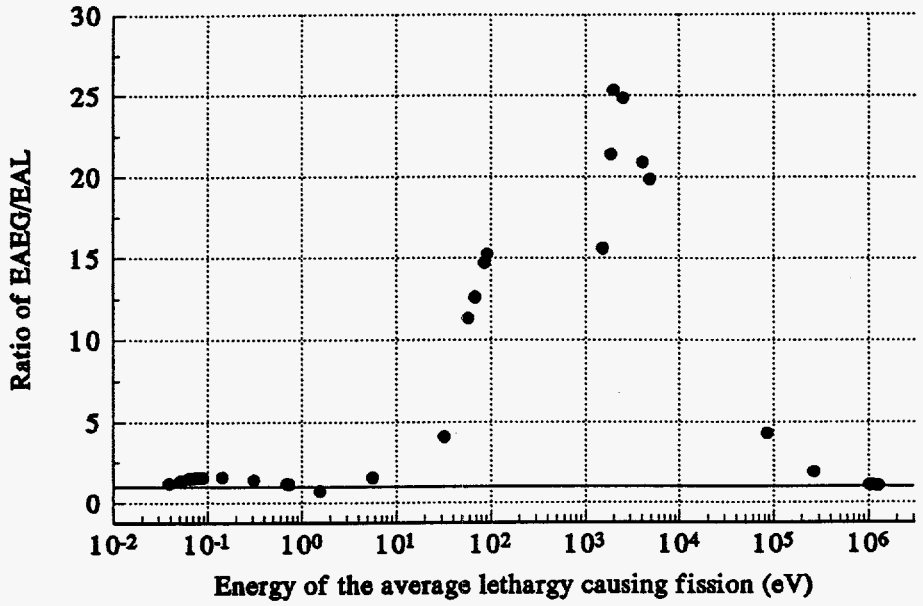
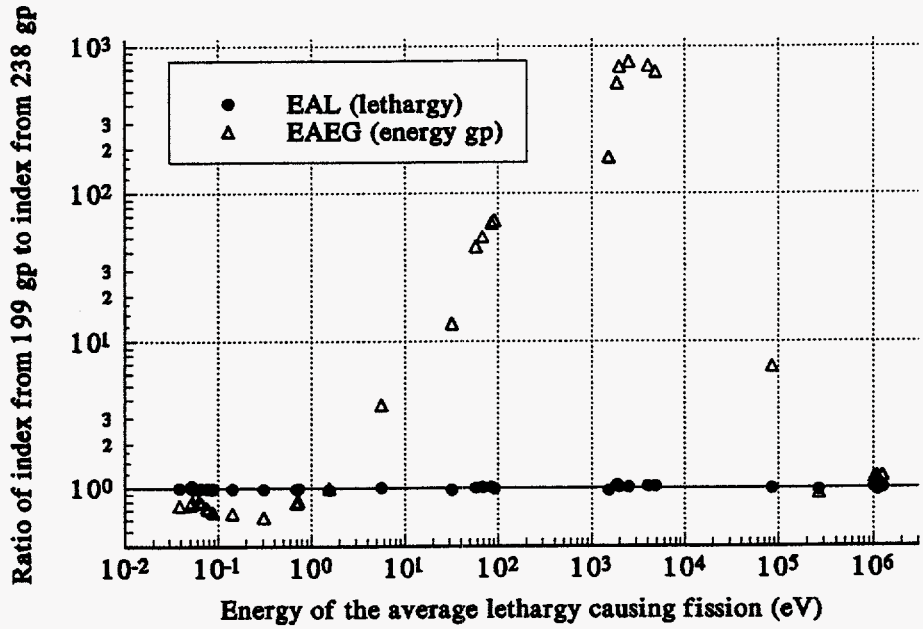


Fig. 4. Comparison of indices (EAL and EAEG) for 199 and 238 libraries



from 1.0 by less than 5% over the entire energy range.

The EAL has all of the advantages previously mentioned for the AEG and also appears to give a truer representation of the energy dependence of fissions, i.e., the influence of the group structure of the library on the computed parameter is reduced. This characteristic is important when determining applicability of critical experiment data to a system being evaluated.

STATISTICAL ANALYSES

The mean k-effective for all of the ENDF/B-VI calculated k-effectives is 1.013 $\pm$ 0.012. Tests for correlations between ENDF/B-VI-calculated k-effectives and the parameters listed in Table II were conducted. No evidence of linear correlation of calculated k-effective to EAL, H/Pu ratio, ^{240}Pu content of the plutonium, or plutonium density in the critical experiment was found. Oxide, parallelpiped geometry experiments calculated significantly higher than the spherical experiments (1.019 vs. 1.009). Oxide experiments calculated higher than nitrate experiments (1.019 vs. 1.011) and both were higher than metal experiments (0.995).

For criticality safety evaluations it is imperative to identify a function which defines an acceptable degree of subcriticality. One methodology in use at Oak Ridge National Laboratory is defined in[15]. Based on these studies, one would conclude that separate limits should be developed for nitrate, oxide, and metal systems. If this practice is followed, the methodology in[14] leads to the identification of subcritical limits on k-effectives of 0.954, 0.970, and 0.974 for metal, oxide, and nitrate systems, respectively. These limits correspond to a 95% confidence interval on 99.9% of the population with the population treated as estimates of a single parameter.

SOURCES OF ERROR

"At low energies, many nuclides have resonances that are wide when compared with the scattering ranges for the mixtures in a particular configuration. If the Bondarenko factors used in the calculation are based on the narrow resonance approximation (the case with the 199 group library), this can produce cross-section values that are too high. Systems with nuclides whose resonances overlap also cause inaccuracies."[6]

CONCLUSIONS

There is little improvement in calculated single-unit plutonium systems k-effectives due to the use of the 199 group ENDF/B-VI library when compared to the 238 group ENDF/B-V library. Plutonium systems calculate approximately 1.3% high for k-effective although there are variations according to the physical form of the Pu in the experiment. The parameter, "energy of the average lethargy causing fission" appears to be a useful spectral index for comparing critical experiments to systems being evaluated and appears to be more robust than the "average energy group for fissions" parameter. For the code/library configuration considered, a calculated k-effective + 2 standard deviations being less than 0.95 should provide an adequate margin of subcriticality for single unit, Pu-bearing systems.

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