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THERMAL DATA TESTING OF ENDF/B-III AND PROGNOSIS FOR ENDF/B-IV

by

F. J. McCrosson

Savannah River Laboratory
E. I. du Pont de Nemours & Co.
Aiken, South Carolina 29801

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INTRODUCTION AND SUMMARY

One of the activities of the ENDF/B Cross Section Evaluation Working Group (CSEWG) is testing differential ENDF/B cross sections in integral benchmark experiments. In this paper, I will be presenting the ENDF/B-III data testing results as obtained by the CSEWG Data Testing Subcommittee. These results have provided a) a basis for evaluating the merit of ENDF/B-III in the analysis of thermal systems, and b) a reference to assist evaluators in the development of improved thermal neutron data for ENDF/B-IV.

The ENDF/B-III calculations for thermal systems were performed by personnel from six organizations (Slide 1): Floyd Wheeler (ANC); Jud Hardy (BAPL); D. S. Craig and M. Hughes (CRNL); Don Mathews (GGA); Lester Petrie (ORNL); and R. L. Reed and F. J. McCrosson (SRL).

Fifteen thermal benchmark experiments were analyzed (Slide 2):

- o Five unreflected spheres of uranyl nitrate (93 wt % ^{235}U) solution were analyzed to test the H_2O and ^{235}U cross sections. The ENDF/B-III calculations for the spheres yielded values of k_{eff} about 0.4% below experiment. This

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underprediction of criticality has been attributed to low thermal values for the product ν times σ_f for ^{235}U. In ENDF/B-IV, the product $\nu\sigma_f$ has been increased at thermal energies, and better agreement with experiment is expected.

- Two H_2O-moderated lattices of slightly enriched uranium rods and three D_2O-moderated lattices of natural uranium rods were analyzed to test the ^{238}U thermal and resonance region capture cross sections in addition to the ^{235}U and moderator cross sections. The criticality of the uranium lattices is underpredicted approximately 1.5% using ENDF/B-III. This underprediction is due primarily to a 10% overprediction of epithermal neutron captures by ^{238}U. ENDF/B-IV is expected to improve this situation somewhat, but calculations will continue to significantly overpredict epithermal ^{238}U captures.
- Five unreflected spheres of plutonium nitrate solutions were analyzed to test the ^{239}Pu cross sections. The calculations indicated that the ENDF/B-III cross sections tend to overpredict the criticality of the plutonium nitrate solutions; the degree of overprediction increases as the thermal neutron spectrum hardens. Typically, k_{eff} is about 1-2% high. No significant change is expected using Version IV.

Now the ENDF/B-III thermal data testing results will be discussed in more detail. A prognosis for ENDF/B-IV will follow the ENDF/B-III discussion.

DISCUSSION

Uranyl Nitrate Spheres (Slide 3)

CSEWG benchmark experiments ORNL-1, 2, 3, 4, and 10 refer to well-documented experiments performed in the early 1960's by R. Gwin and D. W. Magnuson[1] in which critical compositions were determined for aqueous solutions of ^{235}U in spherical geometry. The first four benchmarks in Slide 3 have the same critical radius, but have $H/^{235}U$ ratios which vary as a function of boric acid content. ORNL-1 and 10 contain no boron. These ORNL experiments were reanalyzed in 1968 by Alan Staub, D. R. Harris, and Mark Goldsmith of BAPL to include small corrections for the presence of the aluminum container, departures from sphericity, and room return.[2] The corrected measurements are useful for testing H_2O fast scattering data, the ^{235}U fission spectrum, thermal capture and fission of ^{235}U, and thermal absorption of hydrogen. Incidently, prior to these benchmark calculations, the $S(\alpha,\beta)$ thermal scattering law data for ENDF/B-III moderators had been tested and found satisfactory for predicting measured integral parameters associated with the diffusion length and pulsed neutron experiments.[3]

There is no ready explanation why the GGA results in Slide 3 are 0.2 to 0.3% higher than the BAPL and SRL results. The GGA and SRL calculations used S_n methods,* whereas the BAPL calculations were P_3 epithermally and double P_1 thermally with Marshak boundary conditions. Taken collectively, the BAPL, GGA, and SRL results indicate ENDF/B-III underpredicts k_{eff} by about 0.4%. This underprediction has been attributed in part to an underestimation of $\nu\sigma_f$ for ^{235}U at thermal energies.

Uranium Lattices (Slide 4)

Benchmark experiments TRX-1 and 2 correspond to two of the lattices described by J. Hardy, Jr.[4] These lattices contain slightly enriched (1.3%) uranium rods with diameters of 0.4915 cm. Benchmarks MIT-1, 2, and 3 are well-documented D_2O lattice experiments performed in the early 1960's at MIT under the Heavy Water Lattice Project.[5] The MIT experiments were performed in a subcritical exponential facility and involved D_2O-moderated lattices of natural uranium rods with diameters of 2.565 cm. In addition to material bucklings, the TRX and MIT series of experiments determined several important activation parameters:

*DTF-IV was used in the GGA calculations and ANISN was used in the SRL calculations.

ρ^{28} = The ratio of epithermal-to-thermal ^{238}U captures

δ^{25} = The ratio of epithermal-to-thermal ^{235}U fissions

δ^{28} = The ratio of ^{238}U fissions to ^{235}U fissions

These benchmark experiments directly test the thermal and epithermal cross sections for ^{238}U capture and ^{235}U fissions and the ^{238}U fast fission cross section. They are sensitive to ^{238}U inelastic scattering, the ^{235}U fission spectrum, and the moderator cross sections. By way of comparison, these lattices have a much softer neutron spectrum than a typical pressurized water reactor. Thus, for a PWR, the ratio of epithermal-to-thermal ^{235}U fissions is three times greater than for the TRX-1 lattice, which has the hardest spectrum of the benchmark cases in Slide 4.

The calculated values of k_{eff} reported by the laboratories differ by as much as 1%. Part of this large variation may be attributed to the multiplicity of calculational methods used. Some laboratories used Monte Carlo, others the S_n approximation, and others integral transport theory. All the laboratories used detailed resonance treatments to account for resonance self-shielding, but again, there were variations in the methods and approximations. Differences can arise in the initial step of processing the ENDF/B point-wise data to multigroup form, but some of the laboratories were able to compare multigroup edits to eliminate this possibility.

The calculated values of ρ^{28} (Slide 5) correspond to a thermal cutoff energy 0.625 eV. These calculated values are about 10% higher than experiment. This suggests epithermal ^{238}U neutron capture is being overpredicted by about 10%. This overprediction of epithermal ^{238}U capture largely accounts for the 1.5 to 2.0% underprediction of k_{eff} for the benchmark lattices. One of the objectives of ENDF/B-IV was to reduce this discrepancy through careful reevaluation of the ^{238}U resonance cross sections. The effect of this new evaluation will be discussed later when we present some preliminary ENDF/B-IV data testing results.

The calculated and measured values of δ^{25} in this slide (Slide 6) correspond to a thermal cutoff energy of 0.625 eV. The agreement between calculation and experiment is good for the TRX lattices, but poor for the MIT lattices. It can be observed that the CRNL and SRL results are significantly higher than the GGA and ORNL results for both the TRX and MIT series of experiments. This trend is believed to arise from differences in the approximations used in the resonance treatments. The CRNL and SRL calculations treated ^{235}U resonances individually and neglected the effects of overlapping of ^{238}U and neighboring ^{235}U resonances. The GGA and ORNL calculations, on the other hand, properly accounted for all the overlap effects.

The calculations for the H_2O-moderated TRX lattices suggest no significant shortcomings in the ENDF/B-III epithermal ^{235}U fission cross sections. Therefore, the large differences between the calculated and measured values of δ^{25} for the D_2O-moderated MIT lattices are not likely to stem from deficiencies in the ^{235}U cross sections themselves. Nor is it likely the problem lies in the measurements or in the definition of the thermal cutoff energy. The D_2O lattices are far more sensitive to the resonance overlap effects than the H_2O lattices, so perhaps the differences between calculated and measured values of δ^{25} lie in an associated effect, viz., the representation of interference resonance scattering.

There is good agreement between the calculated and measured values of the ratio of ^{238}U fissions to ^{235}U fissions (Slide 7). These calculations for δ^{28} test not only the ^{238}U fast fission cross section, but also the ^{238}U inelastic cross section and the ^{235}U and ^{238}U fission neutron spectra.

Plutonium Nitrate Spheres (Slide 8)

This last slide describing ENDF/B-III data testing results summarizes calculated eigenvalues for five unreflected spheres of plutonium nitrate solution. The first two benchmarks in the series were performed by R. C. Lloyd, et al., at Battelle-Northwest in 1966.[6] PNL-3, 4, and 5 denote experiments by F. E. Kruesi, et al., performed at Hanford in 1952.[7] These experiments, which have hydrogen-to-^{239}Pu atom ratios ranging from 124 to 1204, are useful for testing H_2O scattering data, cross sections for thermal neutron capture and fission by ^{239}Pu, and the ^{239}Pu fission spectrum. Although their inventories are not defined as precisely as more recent experiments, the simplicity of these bare, homogeneous spheres makes them particularly attractive for calculational benchmarks.

Both the GGA and SRL results in Slide 8 were obtained using the S_4 approximation and multigroup cross sections with P_1 scattering. The overprediction of criticality for the PNL spheres in Slide 8 worsens as the thermal neutron spectrum hardens, i.e., as the ratio of hydrogen atoms to ^{239}Pu atoms decreases. The origin of this overprediction has not been determined. Perhaps, without violating the precision of the data, judicious adjustments to the ^{239}Pu capture and fission cross sections in the vicinity of the 0.3-eV resonance could be made which would yield reasonable values of k_{eff}. The overprediction of criticality is expected to continue with ENDF/B-IV since the shape of the ^{239}Pu thermal cross sections is identical in Versions III and IV, and relatively small adjustments have been made to the 2200-meter/sec values.

ENDF/B-IV Cross Sections and Preliminary Data Testing Results

Now consider some of the changes which we might expect as the ENDF/B-IV ^{235}U and ^{238}U cross sections become available. The ^{235}U and ^{238}U cross sections for ENDF/B-IV have been completely reevaluated over the full energy range from 0 to 20 MeV. Slide 9 lists those new features in ENDF/B-IV which will affect calculations for thermal systems.

The 2200-meter/sec cross sections for ^{235}U have been revised according to the least squares adjustment described by J. R. Stehn;[8] the shapes of the thermal cross sections are the same as in Version III. These changes in the thermal ^{235}U cross sections tend to increase predictions of criticality. However, these increases are offset somewhat by a hardening of the ^{235}U fission spectrum, since the average energy of the neutrons emitted by thermal ^{235}U fission has been increased from 1.95 to 1.9835 MeV.

The ENDF/B-IV thermal capture cross sections for ^{238}U are uniformly about 1% lower than ENDF/B-III and have a 2200-meter/sec value of 2.70 barns compared to 2.72 barns in ENDF/B-III. A complete reanalysis of the resonance cross sections for ^{238}U was performed. The new ^{238}U cross sections yield effective ^{238}U capture resonance integrals for the uranium benchmark lattices which are about 0.3 barn lower than ENDF/B-III.

To get some indication of what might be expected with ENDF/B-IV, some of the uranium benchmark experiments were analyzed at Savannah River. The SRL calculational methods were the same as described previously, viz., the ORNL spheres were calculated using the S_n code ANISN with proper account being taken for resonance self-shielding effects; the TRX and MIT lattices were analyzed with integral transport theory and the Nordheim resonance treatment. The zero leakage integral transport results were leakage corrected by subsequent B_1 calculations for equivalent spatially homogeneous cells.

The second column in Slide 10 lists the average k_{eff} for ENDF/B-III as determined by the BAPL, GGA, and SRL calculations. The third column gives the difference in k_{eff} when the ENDF/B-III cross sections for ^{235}U and ^{238}U are replaced with ENDF/B-IV data. This difference does not reflect any change which might result from the new ENDF/B-IV evaluations of hydrogen and oxygen. The observed increase in k_{eff} is primarily the result of a 0.7% increase in the ENDF/B-IV ^{235}U thermal values for $\delta\sigma_f$. Fast leakage is increased somewhat using ENDF/B-IV because the effective nuclear temperature for the ^{235}U fission spectrum is 1.30 MeV in Version III and 1.323 MeV in Version IV. The last column in the slide gives the estimated value of k_{eff} for ENDF/B-IV. This value was obtained by applying the difference in Column 3 to the average value in Column 2. It is observed that the improvement in going from Version III to Version IV is small, but prediction of criticality with ENDF/B-IV is reasonably good.

The third column in Slide 11 indicates that ENDF/B-IV yields about a 0.7% increase in k_{eff} for the TRX lattices and a 1.0% increase for the MIT lattices. Nevertheless, these very preliminary ENDF/B-IV results still indicate significant underprediction of k_{eff}. This poor agreement, in contrast to the good agreement for the uranyl nitrate spheres, suggests the problem lies in the ^{238}U cross sections and/or in the calculational models.

Measured and calculated values of ρ^{28} are compared in Slide 12. The preliminary ENDF/B-IV ^{238}U cross sections reduce the ENDF/B-III results for ρ^{28} by less than 2%; a 10% reduction of epithermal ^{238}U capture would have yielded not only agreement with the ρ^{28} measurements, but also, good values for k_{eff}. Such a 10% reduction, however, can not be justified by current differential cross section measurements. Great care was taken in the ENDF/B-IV evaluation to reduce the possibility of spurious p-wave resonances and excess absorption through systematic experimental errors.

CONCLUSIONS AND PROGRAM (SLIDE 13)

In conclusion, the preliminary test results for ENDF/B-IV indicate reasonably good prediction of criticality for the ORNL spheres, but criticality for the uranium lattices will be significantly underpredicted. This underprediction arises primarily because the lattice calculations overestimate ^{238}U resonance capture. Further reduction of the epithermal ^{238}U capture cross sections to force agreement does not appear warranted by current differential cross section measurements; hence, we may be facing problems in the calculational methods themselves. To help resolve this question, plans are under way to conduct a seminar on resonance calculations in the fall. Hopefully, through a discussion of current calculational methods and possible extensions, a course of action can be established for improving the predictions for the uranium lattices.

The criticality of the plutonium nitrate spheres is overpredicted by 1 to 2% with ENDF/B-III, and no significant change is expected with ENDF/B-IV. This overprediction appears to be a function of spectrum hardness. To better understand this problem, the CSEWG Data Testing Subcommittee is preparing specifications for additional homogeneous experiments performed at the Battelle-Northwest Critical Mass Laboratory.[9,10] These more recent experiments involve plutonium-uranium mixtures, and they are somewhat more complex geometrically than the older PNL spheres, but their inventories are better defined. Several plutonium-fueled uniform lattice experiments are also being added to the CSEWG benchmarks. We encourage the measurement of plutonium activation parameters similar to ρ^{28} and δ^{25} for uranium lattices. Such plutonium activation parameters would be extremely useful in plutonium data testing.

Finally, we would like to have more organizations participate in the ENDF/B data testing program. Although the utilities were not active in the ENDF/B-III data testing effort, we are looking forward to their participation in Version IV data testing. This will provide additional balance to the ENDF/B effort and will surely result in a broader spectrum of thermal data testing benchmarks than we now have.

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Slide 1

PARTICIPATING ORGANIZATIONS

- AEROJET NUCLEAR CO. (ANC)
- BETTIS ATOMIC POWER LABORATORY (BAPL)
- CHALK RIVER NUCLEAR LABS. (CRNL)
- GULF GENERAL ATOMIC (GGA)
- OAK RIDGE NATIONAL LABORATORY (ORNL)
- SAVANNAH RIVER LABORATORY (SRL)

Slide 2

ENDF/B-III BENCHMARK RESULTS

- o UNREFLECTED SPHERES OF URANYL NITRATE SOLUTION:
  k_{eff} ∿0.4% low

- o H_2O- AND D_2O-MODERATED URANIUM LATTICES:
  k_{eff} ∿1.5% low

- o UNREFLECTED SPHERES OF PLUTONIUM NITRATE SOLUTION:
  k_{eff} 1 to 2% high

Slide 3

URANYL NITRATE SPHERES (ENDF/B-III)

			k_{eff}		
BENCHMARK	RADIUS, cm	$H/^{235}U$	BAPL	GGA	SRL
ORNL- 1	34.595	1378	0.9965	0.9999	0.9973
ORNL- 2	34.595	1177	0.9963	0.9995	
ORNL- 3	34.595	1033	0.9933	0.9963	
ORNL- 4	34.595	971	0.9947	0.9980	0.9958
ORNL-10	61.011	1835	0.9931	0.9956	0.9935

Slide 4

CRITICALITY OF URANIUM LATTICES (ENDF/B-III)

A. H_2O-Moderated Lattices

		k_{eff}					
BENCHMARK	MOD/FUEL	ANC	BAPL	CRNL	GGA	ORNL	SRL
TRX-1	2.35	0.9741	0.9872	0.9808	0.9791	0.985	0.9766
TRX-2	4.02	0.9823	0.9913	0.9876	0.9924	0.998	0.9859

B. D_2O-Moderated Lattices

		k_{eff}			
BENCHMARK	MOD/FUEL	CRNL	GGA	ORNL	SRL
MIT-1	20.74	0.9801	0.9888	0.984	0.9735
MIT-2	25.88	0.9804	0.9925	0.974	0.9752
MIT-3	34.59	0.9826	0.9996	0.975	0.9788

Slide 5

RATIO OF EPITHERMAL-TO-THERMAL ^{238}U CAPTURES (ENDF/B-III)

	ρ^{28}						
BENCHMARK	EXP	ANC	BAPL	CRNL	GGA	ORNL	SRL
TRX-1	1.311 ±0.020	1.438	1.422	1.419	1.416	1.44	1.454
TRX-2	0.830 ±0.015	0.906	0.899	0.874	0.877	0.91	0.890
MIT-1	0.498 ±0.008			0.5319	0.534	0.535	0.5683
MIT-2	0.394 ±0.002			0.4365	0.435	0.430	0.4659
MIT-3	0.305 ±0.004			0.3400	0.334	0.346	0.3624

Slide 6

RATIO OF EPITHERMAL-TO-THERMAL ^{235}U FISSIONS (ENDF/B-III)

	δ^{25}						
BENCHMARK	EXP	ANC	BAPL	CRNL	GGA	ORNL	SRL
TRX-1	0.0981 ±0.001	0.1019	0.1031	0.1100	0.0981	0.10	0.1024
TRX-2	0.0608 ±0.0007	0.0619	0.0649	0.0665	0.0602	0.06	0.0621
MIT-1	0.0447 ±0.0019			0.0520	0.0473	0.049	0.0538
MIT-2	0.031 ±0.003			0.0424	0.0385	0.042	0.0439
MIT-3	0.0248 ±0.0010			0.0327	0.0295	0.029	0.0338

Slide 7

RATIO OF ^{238}U FISSIONS TO ^{235}U FISSIONS (ENDF/B-III)

	δ^{28}						
BENCHMARK	EXP	ANC	BAPL	CRNL	GGA	ORNL	SRL
TRX-1	0.0914 ±0.0020	0.0901	0.0894	0.0878	0.0910	0.093	0.0912
TRX-2	0.0667 ±0.002	0.0651	0.0654	0.0627	0.0659	0.069	0.0651
MIT-1	0.0597 ±0.0020			0.0529	0.0584	0.058	0.0600
MIT-2	0.0596 ±0.0017			0.0515	0.0562	0.056	0.0580
MIT-3	0.0583 ±0.0012			0.0504	0.0537	0.055	0.0561

Slide 8

PLUTONIUM NITRATE SPHERES (ENDF/B-III)

BENCHMARK	RADIUS, cm	$H/^{239}Pu$	k_{eff} GGA	k_{eff} SRL
PNL-1	19.509	698	1.0212	1.0239
PNL-2	19.509	124		1.0324
PNL-3	22.700	1204	1.0012	0.9996
PNL-4	22.700	911	1.0089	1.0111
PNL-5	20.126	578	1.0150	1.0197

Slide 9

ENDF/B-IV CROSS SECTIONS

- ^{235}U
  - NEW LEAST SQUARES ADJUSTMENT OF 2200-M/SEC DATA
  - ENDF/B-III SHAPES RETAINED AT THERMAL ENERGIES
  - $\bar{E}_m$ FOR FISSION SPECTRUM INCREASED FROM 1.95 TO 1.985 MeV
- ^{238}U
  - 2200-M/SEC CAPTURE CROSS SECTION REDUCED FROM 2.72 TO 2.70 b
  - EFFECTIVE RESONANCE INTEGRAL ∿0.3 b LOWER THAN ENDF/B-III

Slide 10

URANYL NITRATE SPHERES (PRELIMINARY ENDF/B-IV)

BENCHMARK	AVG. k_{eff} ENDF/B-III	ESTIMATED Δk_{eff}	k_{eff} ENDF/B-IV
ORNL-1	0.9979	0.0012	0.9991
ORNL-4	0.9962	0.0012	0.9974
ORNL-10	0.9941	0.0027	0.9968

Slide 11

CRITICALITY OF URANIUM LATTICES (PRELIMINARY ENDF/B-IV)

BENCHMARK	AVG. k_{eff} ENDF/B-III	ESTIMATED Δk_{eff}	k_{eff} ENDF/B-IV
TRX-1	0.9805	0.0082	0.9887
TRX-2	0.9896	0.0059	0.9955
MIT-1	0.9816	0.0111	0.9927
MIT-2	0.9805	0.0102	0.9907
MIT-3	0.9840	0.0093	0.9933

Slide 12

ρ^{28} FOR URANIUM LATTICES (PRELIMINARY ENDF/B-IV)

	ρ^{28}			
BENCHMARK	EXPERIMENT	AVERAGE ENDF/B-III	ESTIMATED % REDUCTION	ESTIMATED ENDF/B-IV
TRX-1	1.311 ±0.020	1.431	1.75	1.406
TRX-2	0.830 ±0.015	0.893	1.68	0.878
MIT-1	0.498 ±0.008	0.542	1.84	0.532
MIT-2	0.394 ±0.002	0.442	1.58	0.435
MIT-3	0.305 ±0.004	0.346	1.68	0.340

Slide 13

CONCLUSIONS AND PROGRAM

- GOOD PREDICTION OF k_{eff} FOR ORNL SPHERES
- k_{eff} UNDERPREDICTED FOR U LATTICES
  - ^{238}U RESONANCE CAPTURE OVERPREDICTED
  - SEMINAR ON RESONANCE METHODS
- k_{eff} OVERPREDICTED FOR PLUTONIUM NITRATE SOLUTIONS
  - MORE PLUTONIUM BENCHMARKS (HOMOGENEOUS; LATTICE)
  - ACTIVATION PARAMETERS
- BROADEN SCOPE
  - PARTICIPATION
  - BENCHMARKS