FAST AND THERMAL DATA TESTING OF U-233 CRITICAL ASSEMBLIES[1]

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Abstract

Data testing has been performed for U-233 fast and thermal benchmarks. Results are presented for both ENDF/B-VI and a modified JENDL-3.2 evaluation. The revised JENDL-3.2 evaluation is summarized and comparisons with ENDF/B-VI and measured values are discussed. Calculated results using both cross section sets are presented for 10 fast benchmarks (reflected and unreflected U-233 metal) and 38 thermal benchmarks (uranyl-nitrate solutions in spherical and cylindrical geometry). Using the revised JENDL-3.2 evaluation, very good results are obtained for the calculated k-effs for almost all of the 48 benchmarks considered in this study. Possible future work is discussed briefly.

Introduction

Many sources have been used to get U-233 benchmark descriptions. Unfortunately some of these are not reliable since a thorough and complete benchmark evaluation often has not been done. For 24 years a principal source for benchmarks has been the CSEWG (Cross Section Evaluation Working Group) Benchmark Specifications [1]. The CSEWG specifications included only two fast benchmarks and three thermal benchmarks. The thermal benchmarks were H2O moderated thorium-oxide exponential lattices. Since the thorium-oxide lattices were exponential experiments, they have not been widely used. CSEWG has also used the U-233 ORNL spheres [2] for many years. One advantage of the CSEWG fast benchmarks, JEZEBEL-23 and FLATTOP-23, is that experiments were done for central-reaction-rate ratios. These reaction rate ratios provide very valuable information to data testers and evaluators which would not otherwise be available.

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In recent years the International Handbook of Evaluated Criticality Safety Benchmark Experiments [3] has, in general, been a very useful and reliable source. The HANDBOOK does not include central-reaction-rate ratio experiments, however. New U-233 benchmark experiments have recently been evaluated and have been submitted for approval to the Criticality Safety Benchmark Experiment Program. These are paraffin-reflected cylinders of U-233 uranyl-nitrate solutions. Unfortunately the estimated benchmark uncertainties are on the order of 0.9 to 1.0% in k-eff. Benchmark testing has been done for 25 of these uranyl-nitrate cylinders. We have also discovered that the benchmark specifications for the J. T. Thomas uranyl-nitrate experiments [4] given in Ref. 5 are incorrect. One problem with the Ref. 5 specifications is that the excess acid was not included. As part of this work, we developed revised specifications which include an excess acid correlation based on information from the experimental log-book.

Uranyl-nitrate Spheres

Calculated results for the uranyl-nitrate spheres using SCALE (XSDRNPM) with ENDF/B-VI cross sections are given in Table 1. The calculated k-effs for the J. T. Thomas spheres, using the revised specifications which include the excess acid, are shown in Table 1. The average calculated k-eff for these spheres is 0.9969. The calculated k-eff for the ORNL spheres is shown in the bottom part of Table 1. The average calculated k-eff for these spheres is 0.9966. Results for the J. T. Thomas spheres and the ORNL spheres are in very good agreement but 3 of the J. T. Thomas spheres (JTS02, JTS04, and JTS13) are high relative to the other calculated k-effs in Table 1. The average for all 13 spheres in Table 1 is 0.9968.

Calculated k-effs using the AD-HOC-JENDL revised evaluation are also given in Table 1. Results using both the ENDF/B-VI and AD-HOC-JENDL revised evaluations are shown in Fig. 1. Using the revised evaluation, the ORNL spheres calculate higher by about 0.0027 and the average k-eff is 0.9993 (compared to 0.9966 for ENDF/B-VI). For the J. T. Thomas uranyl-nitrate spheres the change in the calculated k-eff varies from +0.0019 for JTS11 to +0.0025 for JTS04. Three of these spheres (JTS11, JTS12, and JTS13) are reflected by water, the others are unreflected. Using the revised cross sections the unreflected spheres are higher by an average delta-k of +0.0022. Since these are XSDRN calculations there are no statistical uncertainties.

The following comments can be made concerning the changes in Table 1 and Fig. 1:

1) The changes are entirely due to changes in the U-233 cross sections. At this point we have not analyzed the changes, this remains to be done. We expect to do this and the information will be used in the evaluation process for the next U-233 evaluation, planned for later in 1999.
2) The specifications for the J. T. Thomas spheres were determined by W. C. Jordan as part of this work. A complete benchmark evaluation has not been done at this point; it is possible that the specifications could change.

Uranyl-nitrate Cylinders

Calculated results for the uranyl-nitrate cylinders using the 199-group ENDF/B-VI cross sections (VITAMIN-B6) with the U-233 AD-HOC-REVISED evaluation are given in Table 2. Table 2 gives the experiment number, diameter of the cylinder (inches), the hydrogen-to-U233 ratio (H/X) and the calculated k-eff, using the SCALE/KENOV.a program. The range of H/X is from 57 to 754 which is a range of interest both for applications and for data testing. The benchmark k-eff for these experiments is 1.0040 ± 0.0090. The average calculated k-eff in Table 2 is 1.0021 which gives a C/E ratio of 0.9981. Considering the benchmark uncertainty, ± 0.0090, the results obtained in Table 2 are quite acceptable. Results for the uranyl-nitrate cylinders are also shown in Fig. 2.

Calculated results for the uranyl-nitrate cylinders using the 199-group ENDF/B-VI cross sections with the ENDF/B-VI U-233 evaluation (all ENDF/B-VI) are shown in Fig. 3. The average calculated k-eff in Fig. 3 is 1.0015 which gives a C/E ratio of 0.9975. These results differ only slightly from the previous results using the U-233 AD-HOC-REVISED evaluation given in Table 2 and shown in Fig. 3. The slope of the regression line in Fig. 2 is slightly greater than the corresponding line shown in Fig. 3. The average C/E ratios differ by only 0.06%; thus both calculations give very satisfactory results for the uranyl-nitrate cylinders. It may be noted that the results in Fig. 2 appear to have a “tighter pattern” and to be somewhat more “consistent” than the corresponding results in Fig. 3.

Revised U-233 Evaluation

The revised U-233 evaluation is a relatively minor revision to the JENDL-3.2, MAT 9222, evaluation. The evaluation, done initially in 1996, is described in previous benchmark data testing reports (Refs. 6 and 7). In the initial evaluation, a uniform reduction of 2.0% to the JENDL-3.2 fission cross section between 0.1 and 3.6 MeV was made. The resulting fission cross section was very similar to the ENDF/B-VI fission cross section. For this work we decided to base the revised fission cross section on the ENDF/B-VI fission cross section. The changes for the revised evaluation are summarized in Table 3. It was felt that even though the differences between this evaluation and the previous one are quite small, that it was preferable to base the revised fission cross section on a slightly modifed ENDF/B-VI fission cross section. The elastic cross section is also revised as described in Table 3.

The ENDF/B-VI fission cross sections for U-233 and U-235 are shown in Fig. 4. The U-233 fission cross section is higher by a factor of 1.7 at 0.6 MeV and about 1.45 at 6.6 MeV. Actually the U-233 fission is usually measured as a ratio to the U-235 fission. The measured U-233/U-235 fission ratio as measured by Kanda et al .[8] is shown in Fig. 5. Also shown in Fig. 5, as the dashed line, is the evaluated ratio from this work.. It may be noted that the uncertainty in the measured data is about 1.8 to 2.0% and that the evaluation is in very good agreement with the measured fission ratio as given by Kanda. Benchmark data testing results given in the next section indicate that the revised evaluation performs quite well for fast benchmarks. Results for the thermal benchmarks shown in this paper were also very acceptable.

Fast Benchmarks

Calculated results for 10 fast benchmarks using SCALE(XSDRNPM) with ENDF/B-VI, JENDL-3, and the AD-HOC-JENDL revised cross sections are given in Table 2. The first benchmark in Table 2 is the well known JEZEBEL-23 benchmark [1]. Descriptions of the other nine fast benchmarks may be found in Ref. 3. The calculated k-eff for the JEZEBEL-23 benchmark using ENDF/B-VI cross sections is 0.9929, a rather low value. On the other hand the calculated k-eff using the JENDL-3 cross sections is much too high. The calculated k-eff using the AD-HOC-JENDL revised cross sections is very close to unity. Two of the fast benchmarks (U233-MAT-FAST-004a and -b) have calculated k-eff values which are significantly higher than the other fast benchmarks.

These two benchmarks have tungsten reflectors; it has been suggested that the higher calculated k-eff values may be due to problems with the tungsten cross sections. The AD-HOC-JENDL revised evaluation is clearly improved, relative to the original JENDL-3.2 evaluation, for prediction of the criticality of fast benchmarks. Cross section differences for the AD-HOC-JENDL revised evaluation, relative to the ENDF/B-VI and JENDL-3.2 evaluations will be discussed. The most important differences are for the elastic, inelastic, and fission cross sections.

Conclusions

Benchmark calculations for 10 fast and 38 thermal benchmarks were performed and the results are given in Tables 1, 2, and 4. Using the revised JENDL-3 U-233 evaluation described in Table 3, very good results are obtained for the calculated k-eff for almost all of the 48 benchmarks considered in this study. Results obtained represent a big improvement relative to either the ENDF/B-VI or JENDL-3.2 evaluations. The revised U-233 fission cross section is in agreement with the measured U-233/U-235 fission ratio.[8] This is absolutely essential; any evaluation which did not meet this criterion would be unacceptable.

The lack of critical benchmark experiments with spectra peaking in the intermediate energy range (with average energy of fission ~ 2 to 200 eV) is a serious limitation which prevents testing of the U-233 cross sections in this energy range. Measurements of the U-233 cross sections from 0.36 eV to ~700 keV have recently been done [9] at ORELA (the Oak Ridge Electron Linear Accelerator). The SAMMY code system [10] will be used to determine new Reich-Moore, resolved resonance parameters which will be part of a revised ENDF/B-VI evaluation which will then be available for use in applications which have spectra in the intermediate energy range.

References

1. *Cross Section Evaluation Working Group (CSEWG) Benchmark Specifications*, BNL-19302 (ENDF-202), National Nuclear Data Center, Brookhaven National Laboratory (1974).
2. R. Gwin and D. W. Magnuson, The Measurement of Eta and Other Nuclear Properties of U233 and U235 in Critical Aqueous Solutions, *Nucl. Sci. Eng.* 12, 364-380 (1962); see also U233-SOL-THERM-001 (Ref. 3).
3. *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/DOC(95)03, Nuclear Energy Agency, OECD, Paris (September 1998 Edition).
4. J. T. Thomas, *U-233 Uranyl-Nitrate Experiments, Neutron Physics Division Annual Progress Report*, ORNL-4280 (MAY 31, 1968).
5. S. R. McNeany and J. D. Jenkins, An Evaluation of ENDF/B-IV and Hansen-Roach Uranium-233 Cross Sections for Use in Criticality Calculations, *Nucl. Sci. Eng.* 65, 441-453 (1978).
6. L. C. Leal and R. Q. Wright, *Assessment of the Available 233-U Cross-Section Evaluations in the Calculation of Critical Benchmark Experiments*, ORNL/TM-13313, Oak Ridge National Laboratory (1996).
7. R. Q. Wright and L. C. Leal, "Benchmark Testing of 233-U Evaluations," pp. 371-377, in Proc.Conf. Criticality Safety Challenges in the Next Decade, Chelan, WA (September 1997).
8. K. Kanda et al., "Measurement of Fast Neutron Induced Fission Cross Sections of ^{232}Th, ^{233}U, and ^{234}U Relative to ^{235}U," *Nuclear Data for Basic and Applied Science*, Vol 1, Santa Fe, New Mexico (May 1985).

9. K. H. Guber, et al., "Fission, Total and Neutron Capture Cross Section Measurements at ORELA for 233-U, 27-Al and Natural Chlorine," pp. 1726-29, Vol. 2, in Proc. Intnatl. Conf Phys. of Nucl. Sci. & Tech, Long Island, NY, (October 1998).
10. Nancy M. Larson, *Updated Users' Guide for SAMMY Multilevel R-matrix Fits to Neutron Data Using Bayes' Equation*, Oak Ridge National Laboratory report ORNL/TM-9179/R4 (December 1998).

Table 1. Calculated Results for Uranyl-Nitrate Spheres

CASE	H/U-233	ENDF/B-VI	AD-HOC JENDL-REV
JTS11	189.40	0.9949	0.9968
JTS01	190.70	0.9957	0.9979
JTS02	248.70	1.0000	1.0022
JTS12	268.10	0.9949	0.9970
JTS03	344.50	0.9951	0.9974
JTS13	542.70	0.9990	1.0014
JTS04	575.80	0.9986	1.0011
JTS-AVERAGE		0.9969	0.9991
ORNL-9	1324.00	0.9963	0.9990
ORNL-8	1369.00	0.9969	0.9997
ORNL-7	1417.00	0.9970	0.9997
ORNL-6	1470.00	0.9972	1.0000
ORNL-5	1533.00	0.9968	0.9995
ORNL-11	1986.00	0.9954	0.9980
ORNL-AVERAGE		0.9966	0.9993
AVERAGE		0.9968±.0016	0.9992±.0017

Table 2. Uranyl nitrate cylinders ENDF/B-VI cross sections, U-233 ad-hoc-revised evaluation			
EXP	DIAM.	H/X	CALC. K-EFF
4	8"	119	1.0081 ± 0.0033
5	8"	149	0.9944 ± 0.0033
8	8"	192	1.0062 ± 0.0031
10	8.5	246	1.0045 ± 0.0030
11	8.5	296	1.0082 ± 0.0027
12	9	355	1.0014 ± 0.0028
14	10	393	0.9852 ± 0.0029
15	10	459	1.0050 ± 0.0030
17	12	580	0.9902 ± 0.0029
18	12	628	1.0072 ± 0.0026
19	12	754	1.0107 ± 0.0024
22	8	84	0.9923 ± 0.0031
24	8	57	0.9900 ± 0.0030
34	8	144	1.0003 ± 0.0036
35	8	212	1.0106 ± 0.0028
36	9	378	1.0061 ± 0.0025
38	10	513	1.0080 ± 0.0029
3	6	119	1.0074 ± 0.0035
6	6	149	1.0076 ± 0.0031
20	6	84	1.0004 ± 0.0034
25	6	57	0.9909 ± 0.0032
30	6	66	0.9996 ± 0.0033
27	7.5	57	1.0040 ± 0.0032
28	7.5	66	1.0034 ± 0.0031
33	7.5	144	1.0100 ± 0.0031
AVERAGE			1.0021 ± 0.0080

Table 3. Revised U-233 Evaluation

The JENDL-3.2, MAT 9222 evaluation, is revised as follows--

1. The fission cross section is revised from 0.4 to 6.1 MeV. From 0.4 to 0.8 Mev the revised fission is the same as the ENDF/B-VI evaluation. From 1.0 to 5.5 MeV the revised fission is equal to a factor of 1.005 times the ENDF/B-VI evaluation (0.5 percent increase). Between 0.8 and 1.0 MeV and between 5.5 and 6.1 MeV, there is a transition between 0.5 percent increase and no change. Above 6.1 MeV and below 0.4 MeV the fission cross section is unchanged from JENDL-3.2

2. The elastic scattering cross section is changed between 0.4 and 6.1 MeV to reflect the change in the fission cross section, so as to keep the total cross section unchanged. The total cross section is the same as for JENDL-3.2.

Table 4. Benchmark calculations for ^{233}U benchmarks
199-group cross sections

Benchmark	ENDF/B-VI	JENDL-3	AD HOC JENDL-REV
U233-MET-FAST-001	0.9929	1.0125	1.0007
U233-MET-FAST-002a	0.9952	1.0093	0.9989
U233-MET-FAST-002b	0.9975	1.0088	0.9995
U233-MET-FAST-003a	0.9958	1.0100	0.9988
U233-MET-FAST-003b	0.9971	1.0085	0.9976
U233-MET-FAST-004a	1.0027	1.0172	1.0068
U233-MET-FAST-004b	1.0061	1.0181	1.0057
U233-MET-FAST-005a	0.9949	1.0094	0.9980
U233-MET-FAST-005b	0.9974	1.0097	0.9988
FLATTOP-23	1.0004	1.0088	0.9985
AVERAGE	0.9980 ± .0040	1.0112 ± .0036	1.0003± .0032

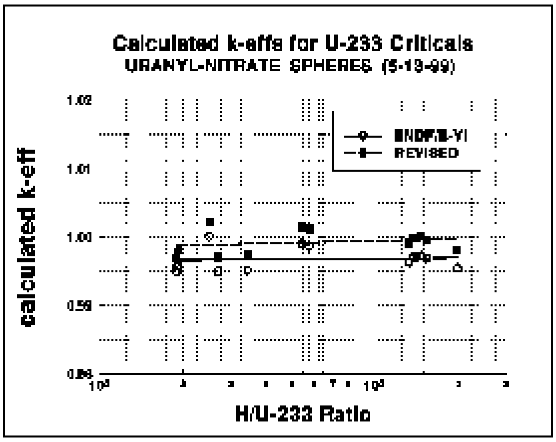

Figure 1. Calculated K-effs for uranyl-nitrate spheres.

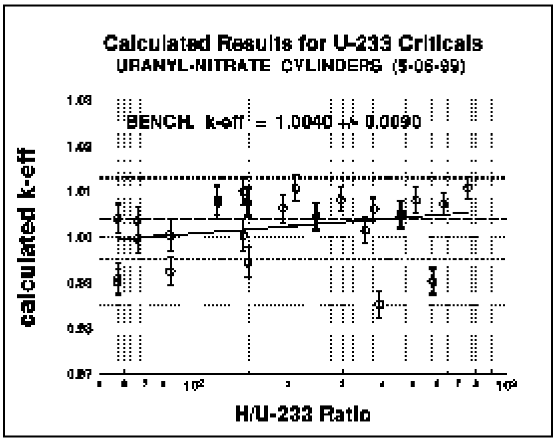

Figure 2. Uranyl-nitrate cylinders, revised evaluation.

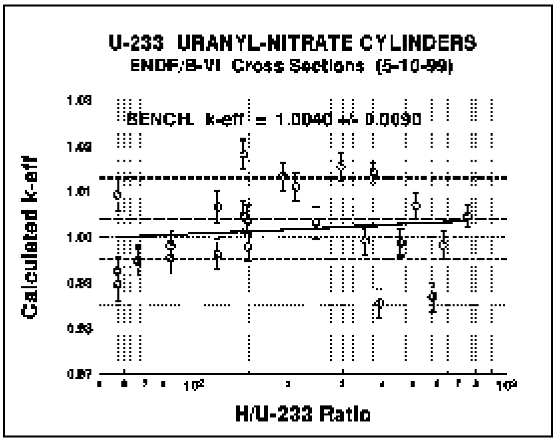

Figure 3. Uranyl-nitrate cylinders, ENDF/B-VI evaluation.

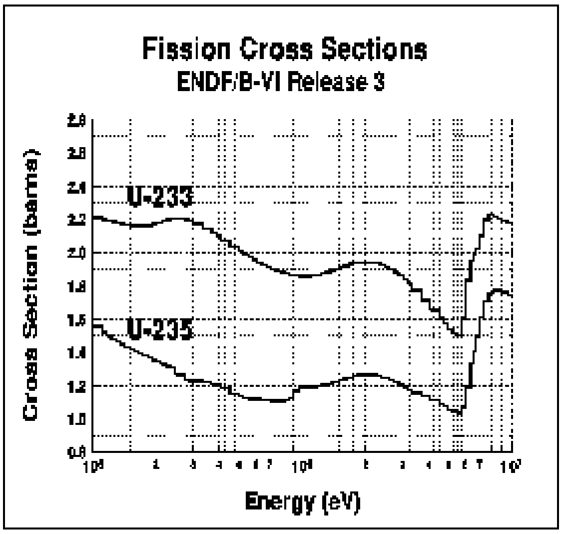

Figure 4. U-233 and U-235 fission cross-sections.

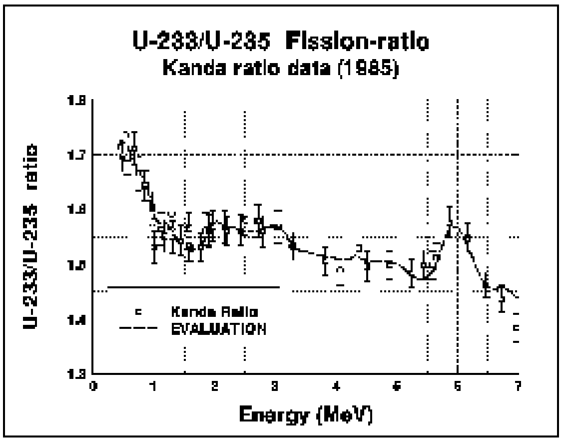

Figure 5. U-233/U-235 fission-ratio.