INTEGRATION OF SEVERAL ELEMENTS OF THE DOE NUCLEAR CRITICALITY SAFETY PROGRAM

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Abstract

The U. S. Department of Energy established the Nuclear Criticality Safety Program (NCSP) to maintain the infrastructure and expertise in nuclear criticality safety to support line criticality safety programs at various DOE sites. The seven tasks of the NCSP include critical experiments, benchmarking, nuclear data, analytical methods, applicable ranges of bounding curves and data, information preservation and dissemination, and training and qualification. The goals of this program are to improve the knowledge, tools, data, guidance, and information available to the nuclear criticality safety community. In addition various elements of the NCSP are integrated together to provide the nuclear criticality safety community with the most precise nuclear data for criticality safety analyses.

This paper describes how several elements of the NCSP were integrated together in the evaluation of the silicon nuclear data. Silicon is frequently encountered in decontamination and decommissioning efforts, process sludge and settling tanks, in situ vitrification, and waste remediation efforts (including waste storage, retrieval, characterization, volume reduction, and stabilization). Silicon was also identified as an important isotope for addressing concerns associated with the storage of spent nuclear fuels in a geologic repository. The inadequacy of the silicon nuclear data in the intermediate energy region mandated that additional neutron capture cross-section measurements had to be performed that encompassed the resolved resonance region. An evaluation was performed that included analysis of the most recent neutron capture and existing transmission cross-section measurements performed at the Oak Ridge Electron Linear Accelerator. Critical experiments were performed at the Institute of Physics and Power Engineering in Obninsk, Russia because of the lack of critical experiment data for analysis of storage of nuclear material in a geologic repository. These critical experiments were evaluated and benchmark models were developed and submitted to the International Criticality Safety Benchmark Evaluation Project for review and publication in the "International Handbook of Evaluated Criticality Safety Benchmark Experiments". Sensitivity analyses were performed as a part of the benchmark evaluation to determine the sensitivity of the critical experiments to the various constituents of the assembly. The benchmark models were then used to determine the computed k_{eff} for various cross section data sets. The variation in the computed k_{eff} value for the new evaluated data set was then used as an indicator to adjust the negative energy capture widths for the capture cross section data. Furthermore, the changes in k_{eff} were used as an indicator to the inadequacy of previous measured data in the unresolved resonance region. The result of the efforts of the NCSP provided the most precise set of nuclear data for silicon. The resulting ORNL evaluation produced the most consistent evaluation for silicon. This result could only be achieved through integration of many components of the NCSP.

Introduction

The U. S. Department of Energy established the Nuclear Criticality Safety Program (NCSP) to maintain the infrastructure and expertise in nuclear criticality safety to support line criticality safety programs at various DOE sites. The seven task of the NCSP include critical experiments, benchmarking, nuclear data, analytical methods, applicable ranges of bounding curves and data, information preservation and dissemination, and training and qualification. The goals of this program are to improve the knowledge, tools, data, guidance, and information available to the nuclear criticality safety community. In addition various elements of the NCSP are integrated together to provide the nuclear criticality safety community with the most precise nuclear data for criticality safety analyses.

The objective of this paper is to describe how several elements of the NCSP were integrated together in the evaluation of the silicon nuclear data. Silicon is frequently encountered in decontamination and decommissioning efforts, process sludge and settling tanks, in situ vitrification, and waste remediation efforts (including waste storage, retrieval, characterization, volume reduction, and stabilization). Silicon was also identified as an important isotope for addressing concerns associated with the storage of spent nuclear fuels in a geologic repository. The inadequacy of the silicon data in the intermediate energy region required that neutron capture cross-section measurements that extended well into the intermediate energy region be performed. The evaluation included the analysis of the most recent neutron capture and existing transmission cross-section measurements performed at the Oak Ridge Electron Linear Accelerator. In addition benchmark models were developed of critical experiments performed to address the need for critical experiments with significant amounts of silicon. These benchmark models were subsequently used to test the nuclear data and provide slight adjustments to the thermal neutron capture cross section for ^{28}Si. The computational methods that are supported by the NCSP were used for performing the benchmark calculations.

This paper presents a brief description of the critical experiments in Section 2 followed by a brief description of the benchmark model and the results of the sensitivity studies performed for the benchmark analysis in Section 3. A discussion of the nuclear data measurements and analyses are provided in Section 4 along with the results of the calculations of the critical experiment benchmark model. Finally, the paper is summarized in Section 5.

Critical Experiments at IPPE

Five critical configurations with heterogeneous combinations of highly enriched uranium, silicon dioxide, and polyethylene in a large critical facility at the Institute for Physics and Power Engineering (IPPE), Obninsk, Russia were performed to provide benchmark data for criticality safety applications.[1] Silicon dioxide filled aluminum tubes were interspersed among uranium/silicon dioxide filled tubes to form an assembly of fuel rods. Additionally, quartz-sand filled tubes were placed in the periphery of the assembly in the reflector region. In some of the measurements, polyethylene dowels were positioned between the fuel tubes to moderate the neutron flux. The configurations varied in the number and type of fuel tubes and silicon dioxide tubes. Table 1 contains a brief description of each configuration and the excess reactivity for each configuration. A sketch of one of the fuel configurations is provided in Figure 1.

Table 1. Description of BFS Critical Experiments with Silicon Dioxide.

Experiment	Excess Reactivity	Description
BFS-79/1	$+0.09\beta_{eff}$	175 fuel tubes, compact fuel tube arrangement, silicon moderator and reflector, polyethylene dowels included
BFS-79/2	$+0.04\beta_{eff}$	220 fuel tubes, spaced fuel tube arrangement, silicon moderator and reflector, polyethylene dowels included
BFS-79/3	$+0.16\beta_{eff}$	172 fuel tubes, spaced fuel tube arrangement, silicon and polyethylene moderated, silicon reflected, polyethylene dowels included
BFS-79/4	$+0.21\beta_{eff}$	237 fuel tubes, compact fuel tube arrangement, silicon moderator and reflector, fewer polyethylene dowels included
BFS-79/5	$+0.06\beta_{eff}$	504 fuel tubes, compact fuel tube arrangement, silicon moderator and reflector, no polyethylene dowels

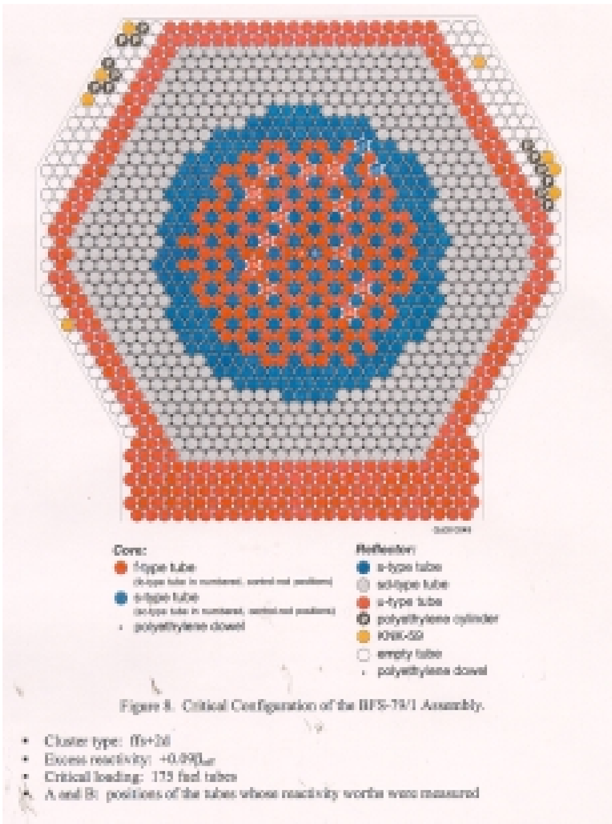

Figure 1. Sketch of BFS-79/1 Critical Configuration.[1]

Benchmark Model

A benchmark model was developed by Tsiboulia et al[1] for the IPPE critical experiments and has been documented in the Handbook of the International Criticality Safety Benchmark Evaluation Project (ICSBEP). The spectral characteristics of these experiments were calculated by the evaluators and are summarized in Tables 2 and 3. Furthermore the evaluators computed the fission and capture events in the predominant isotopes in the core. This information is summarized in Table 4. The percentage of captures in silicon increases as the average fission group energy (AFGE) decreases. However, the capture events in silicon predominantly occur at thermal energies except for experiment BFS-79/5 that has a harder neutron spectrum.

Table 2. Percentage of Fissions in Thermal, Intermediate, and Fast Energy Regions

Case	AFGE (eV)	Fissions %		
		<0.625 eV	0.625 eV – 100 keV	>100 keV
BFS-79/1	$4.97\ 10^{1}$	35.4	45.9	18.6
BFS-79/2	$1.47\ 10^{1}$	47.9	38.3	13.9
BFS-79/3	$2.81\ 10^{0}$	68.4	20.5	11.1
BFS-79/4	$1.85\ 10^{2}$	23.8	54.9	21.2
BFS-79/5	$4.71\ 10^{3}$	4.4	68.4	27.2

Table 3. Percentage of Captures in Thermal, Intermediate, and Fast Energy Regions

Case	AFGE (eV)	Captures %		
		<0.625 eV	0.625 eV – 100 keV	>100 keV
BFS-79/1	$4.97\ 10^{1}$	37.2	52.3	10.4
BFS-79/2	$1.47\ 10^{1}$	55.2	37.3	7.5
BFS-79/3	$2.81\ 10^{0}$	82.0	14.1	3.9
BFS-79/4	$1.85\ 10^{2}$	26.4	61.8	11.8
BFS-79/5	$4.71\ 10^{3}$	3.0	79.8	17.2

Table 4. Balance of Fissions and Captures by Isotopes Over the Core.

	Isotope	BFS-79/1	BFS-79/2	BFS-79/3	BFS-79/4	BFS-79/5
Percent of Fissions	^{234}U	0.1	-	-	0.1	0.1
	^{235}U	69.4	64.6	54.6	69.5	72.2
	^{238}U	0.2	0.2	0.1	0.2	0.3
Percent of Captures	^{234}U	0.6	0.5	0.3	0.6	0.5
	^{235}U	18.5	16.3	11.4	19.9	21.9
	^{238}U	1.3	1.0	0.5	1.5	1.6
	Al	2.3	2.8	3.9	2.0	0.5
	Si	3.2	7.0	8.8	2.4	1.4
	H	2.2	5.0	17.0	2.0	-
	O	1.1	1.1	0.8	1.1	1.3
	^{10}B	0.1	0.3	0.4	-	-
	Other	0.7	1.0	1.8	0.4	-

The measured and MCNP[2,TM] calculated k_{eff} values for these experiments as presented in the ICSBEP evaluation are provided in Table 5. The results depended significantly on the neutron cross-section data set. Results for the ENDF/B-VI cross-section data sets were provided for two different releases of the ^{235}U cross section data. The results are more sensitive to the ^{235}U cross section data than any other isotope as indicated in Table 5 and as was indicated in the fission and capture percentages presented in Table 4. The computed k_{eff} values typically differ from the measured values by more than one percent.

Table 5. Measured and MCNP Computed k_{eff} Values from the ICSBEP Evaluation.

Case	Measured	ENDF/B-V	ENDF/B-VI (Release 2)	ENDF/B-VI (Release 4)
BFS-79/1	1.0007 ± 0.0027	1.0129 ± 0.0005	1.0127 ± 0.0006	1.0051 ± 0.0006
BFS-79/2	1.0003 ± 0.0028	1.0229 ± 0.0005	1.0234 ± 0.0006	1.0153 ± 0.0005
BFS-79/3	1.0012 ± 0.0029	1.0189 ± 0.0005	1.0182 ± 0.0006	1.0135 ± 0.0005
BFS-79/4	1.0016 ± 0.0030	1.0158 ± 0.0006	1.0172 ± 0.0006	1.0073 ± 0.0005
BFS-79/5	1.0005 ± 0.0040	1.0064 ± 0.0005	1.0115 ± 0.0005	1.0016 ± 0.0005

Cross Section Evaluation

Neutron cross-section data are typically parameterized to describe the energy dependent structure. This parameterization simplifies the description of the cross section data and allows recreation of the various reactions without requiring the energy dependent reactions to be stored for each isotope. Furthermore, the parameterization allows temperature dependent data to be created without requiring neutron cross-section measurements at different temperatures. The parameterization typically involves expressing the neutron cross section as a function of the spin of the neutron (s=1/2), the spin of the target nucleus (S), the spin of the compound nucleus, the total angular momentum (J), the nuclear radius (R), the atomic number (A), the energy of the resonance, the partial widths (Γ_i), and other parameters. The partial widths simply characterize the probability of a particular reaction type such as capture, inelastic scatter, and fission.

The current resonance evaluation in the ENDF/B-VI data files for the silicon neutron cross section data were performed at ORNL by Hetrick et al.[3] This evaluation was based on transmission measurements with natural and enriched silicon dioxide samples performed at the Oak Ridge Electron Linear Accelerator (ORELA). The neutron capture widths were obtained from the recommended values provided by Mughabghab et al.[4] The evaluation of Hetrick et al was used as the starting point for the current ORNL evaluation that is presented in this work.

[TM] MCNP is a trademark of the Regents of the University of California, Los Alamos National Laboratory.

Guber et al[5] recently performed neutron capture measurements with natural silicon samples over the energy range from 1 keV to 700 keV. These measurements were performed to address concerns with the current ENDF/B-VI evaluation because of uncertainties in the capture widths in the resolved resonance region. The latest ORNL evaluation[6] is based on transmission measurements with natural and enriched silicon dioxide samples and the capture measurements performed by Guber et al. The measured data were analyzed with the computer code SAMMY[7] to determine the Reich-Moore resonance parameters.

Comparisons of the ENDF/B-VI and final ORNL processed capture cross sections for ^{28}Si are provided in Figs. 2 through 4. The ORNL and ENDF/B-VI capture cross-sections in the energy range from 10 keV to 100 keV are compared in Fig. 2. The most significant difference between these two data sets occurs for the resonance near 56 keV. The latest ORNL evaluation is much lower than the ENDF/B-VI evaluation. Furthermore, the peak cross-sections and the cross sections between resonances are much smaller in the ORNL evaluation than in the ENDF/B-VI evaluation in the resonance region. A comparison of the ENDF/B-VI and ORNL evaluations in the energy range from 100 keV to 250 keV is provided in Fig. 3. As can be seen in this figure, a very narrow resonance for ^{28}Si around 148 keV is missing in the ENDF/B-VI evaluation. This is most likely due to the poor resolution of the capture measurements that were used in the ENDF/B-VI evaluation. This very narrow resonance can be observed in the ORNL evaluation because of the high resolution of the ORELA measurements. A comparison of the capture cross sections from the two evaluations in the energy region from 250 keV to 310 keV is provided in Fig. 4. In the ENDF/B-VI evaluation, the resonance around 300 keV has been represented by two resonances whereas actually only one resonance is present around 300 keV. The second resonance in the ENDF/B-VI evaluation is actually a resonance in ^{30}Si that has been assigned to ^{28}Si in error. As noted previously, the capture cross section from the ORNL evaluation is consistently lower than that from the ENDF/B-VI evaluation in the resonance region.

The initial ORNL evaluation produced a thermal neutron capture cross section of 166.4 mb for ^{28}Si. The thermal neutron capture cross section is mostly determined by the capture width of the first negative energy resonance. Negative energy resonances are added to the evaluation to determine the proper shape and value of the cross section at low energies and to mock up the negative bound levels. The thermal capture cross section was deemed to be too low as compared to the measured thermal capture cross sections.

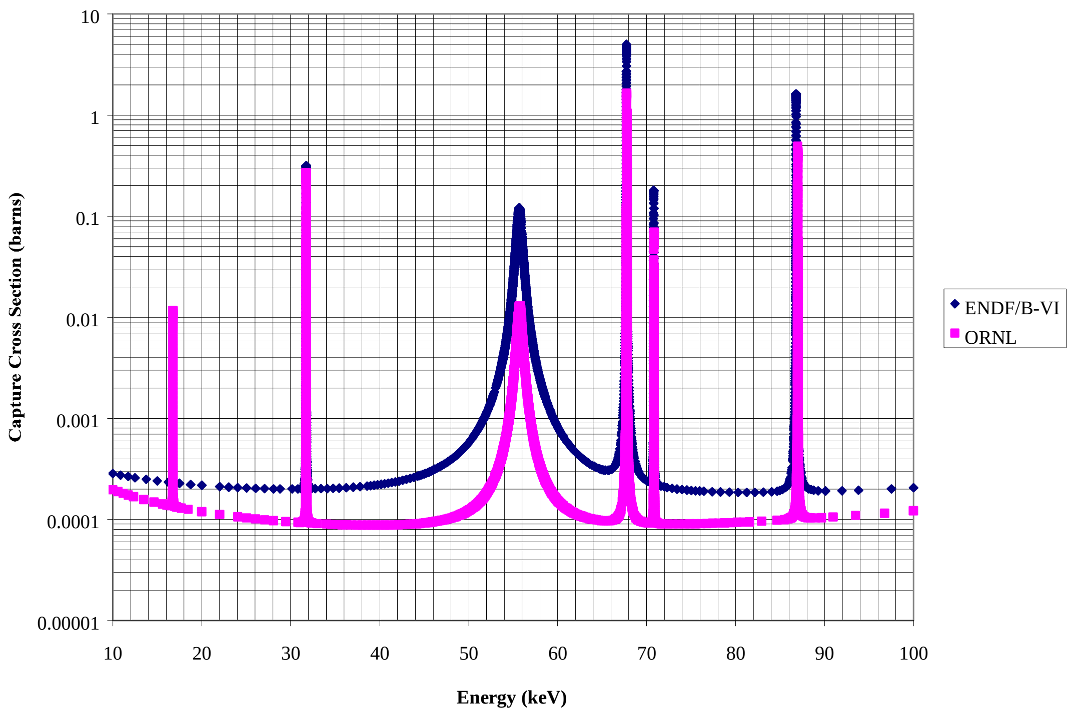

Figure 2. Comparison of the ENDF/B-VI and ORNL ^{28}Si Capture Cross Section from 10 keV to 100 keV.

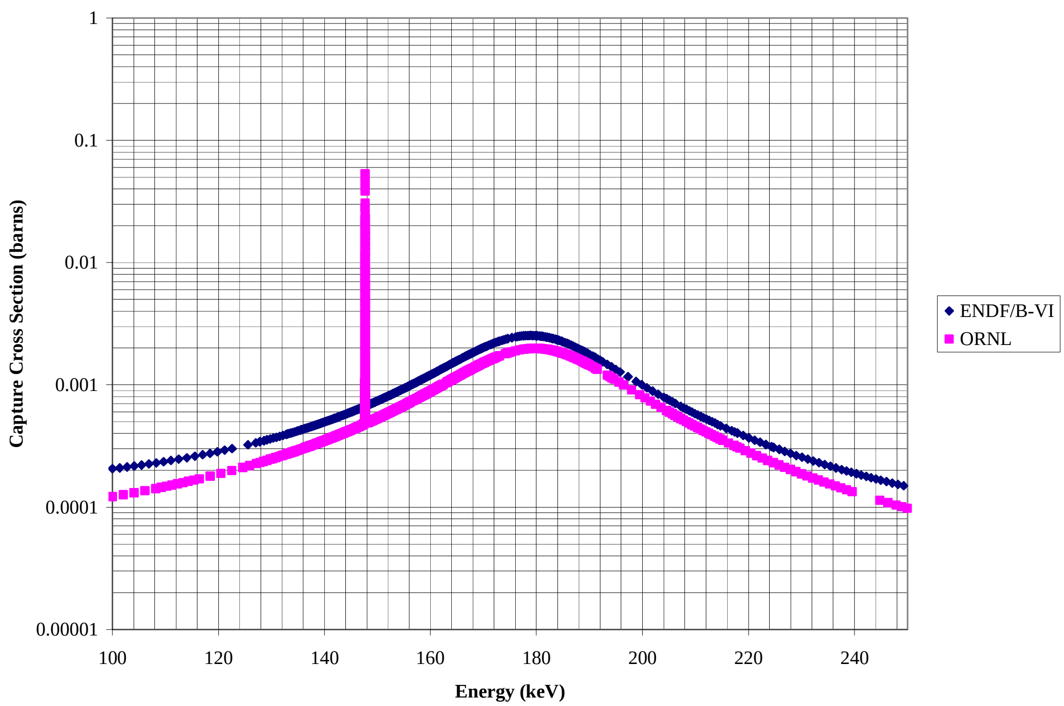

Figure 3. Comparison of the ENDF/B-VI and ORNL ^{28}Si Capture Cross Section from 100 keV to 250 keV.

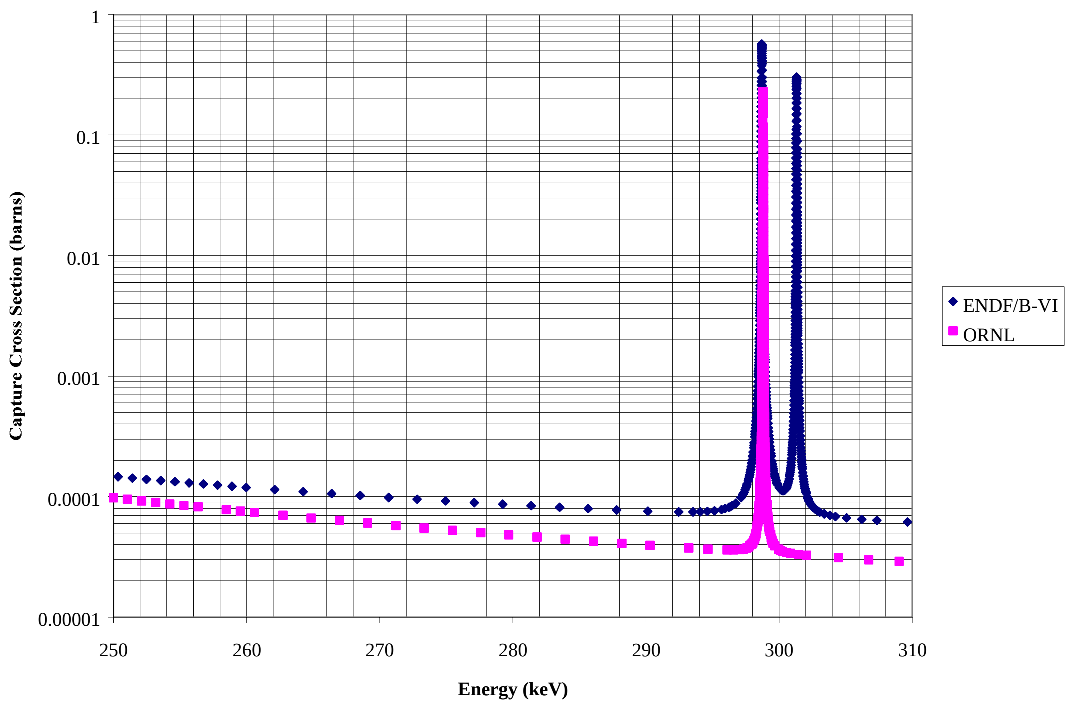

Figure 4. Comparison of the ENDF/B-VI and ORNL
^{28}Si Capture Cross Section from 250 keV to 310 keV.

The ENDF/B-VI and preliminary ORNL evaluations for the silicon isotopes were processed with NJOY[8] to create point wise cross section data for MCNP. For consistency, the same NJOY tolerance limits were used for both evaluations. Furthermore, the latest release of ^{235}U (release 5) was obtained from the National Nuclear Data Center and processed using NJOY. These libraries were used in the MCNP calculations. The preliminary ORNL evaluation and the ENDF/B-VI evaluation were used to compute the k_{eff} values for the critical experiments that are presented in Table 6. Experiments BFS-79/2 and BFS-79/3 have a very thermal neutron spectrum and are most sensitive to thermal neutron capture in silicon. Only the thermal value for ^{28}Si needed to be adjusted because ^{28}Si is the major isotope of silicon. Therefore, these two experiments were the basis for adjusting of the thermal capture width for ^{28}Si. The calculated k_{eff} values obtained with the preliminary ORNL evaluation were higher than those obtained with the ENDF/B-VI cross-section libraries. The first negative energy resonance for ^{28}Si was adjusted such that the computed k_{eff} values for experiments BFS-79/2 and BFS-79/3 were consistent with the results obtained from the ENDF/B-VI calculations. Although the resolved resonance region of the ENDF/B-VI evaluation is inadequate, the thermal capture cross-sections agree well with the measured thermal values. As stated in the report by Hetrick et al, the ENDF/B-VI evaluation for ^{28}Si had been normalized to the thermal value of Raman.[9] Thus, the thermal benchmarks should be consistent between the ENDF/B-VI evaluation and the ORNL evaluation.

Table 6. MCNP Computed k_{eff} Values with
ENDF/B-VI (Release 5) and the Initial ORNL Evaluation

Case	ENDF/B-VI (Release 5)	Initial ORNL Evaluation
BFS-79/1	1.0024 ± 0.0004	1.0039 ± 0.0004
BFS-79/2	1.0123 ± 0.0004	1.0143 ± 0.0004
BFS-79/3	1.0121 ± 0.0004	1.0145 ± 0.0004
BFS-79/4	1.0044 ± 0.0004	1.0060 ± 0.0004
BFS-79/5	0.9975 ± 0.0004	1.0000 ± 0.0004

The first negative energy resonance for ^{28}Si was adjusted such that the ORNL evaluation would produce essentially the same computed k_{eff} values for experiments BFS-79/2 and BFS-79/3 while still providing an excellent representation of the differential data. The SAMMY code was then used to analyze the measured data with the adjusted negative energy region. The resulting thermal capture cross-section for ^{28}Si was 172 mb. This value agrees well with the measured values and is only 3% higher than the initial value that is well within the uncertainty in the measurement of this small capture cross section. The measured and evaluated thermal capture cross-sections are provided in Table 7. The ORNL evaluation is in statistical agreement with the measured values of Raman[9] and McMaster[10]. The resonance parameters for the resolved resonance region from the Guber et al data were used to predict the average cross sections in the unresolved resonance region. The estimated unresolved resonance cross section values from the Guber et al data were significantly lower than that in the Hetrick et al evaluation in ENDF/B-VI. This lower cross section in the unresolved resonance region from the Guber et al data is consistent with the lower cross section in the resolved resonance region. Therefore, the Guber et al data were used to normalize the Hetrick et al evaluation in the unresolved resonance region. The lower cross section in the unresolved resonance region from the Guber et al measurement as compared to the evaluated unresolved resonance data from the Hetrick et al evaluation most likely results from the sensitivity of the measurements to scattered neutrons in the data evaluated by Hetrick et al.

Table 7. Measured and Evaluated Thermal Capture Cross Sections.

Isotope	Raman[9] (mb)	McMaster[9] (mb)	Mughabghab[4] (mb)	ENDF/B-VI (mb)	ORNL (mb)
^{28}Si (92.23)[a]	169 ± 4	171 ± 3	177 ± 5	169.1	172.1
^{29}Si (4.67)[a]	119 ± 3	122 ± 4	101 ± 14	120.1	121.5
^{30}Si (3.10)[a]	108 ± 3	103 ± 4	107 ± 2	107.1	108.2

[a] Isotopic abundance.

The computed k_{eff} values for the critical experiments are provided in Table 8. The computed k_{eff} values obtained using the ENDF/B-VI cross-sections and the ORNL cross sections are statistically the same for the measurements with the most thermal spectrum. For experiment BFS-79/5, the computed k_{eff} value obtained with the ORNL cross sections is higher than that obtained with the ENDF/B-VI cross sections. Almost all of the capture in the BFS-79/5 is above thermal energies. Therefore, this calculation shows the greatest improvement in the computed k_{eff} value. As shown in Figs. 2 through 4, the ORNL capture cross section for ^{28}Si is significantly lower than that from the ENDF/B-VI cross section. Therefore, the computed k_{eff} should be higher for assembly BFS-79/5 as was demonstrated.

Table 8. MCNP Computed k_{eff} Values with
ENDF/B-VI (Release 5) and the ORNL Evaluation

Case	ENDF/B-VI (Release 5)	ORNL Evaluation
BFS-79/1	1.0024 ± 0.0004	1.0030 ± 0.0004
BFS-79/2	1.0123 ± 0.0004	1.0127 ± 0.0004
BFS-79/3	1.0121 ± 0.0004	1.0117 ± 0.0004
BFS-79/4	1.0044 ± 0.0004	1.0043 ± 0.0004
BFS-79/5	0.9975 ± 0.0004	0.9997 ± 0.0004

Summary

Various elements of the NCSP were integrated together in the evaluation of the silicon nuclear data. The generation of the ORNL silicon evaluation involved not only the nuclear data component of the NCSP but also involved the methods, benchmark, and critical experiments components of the NCSP. The high-resolution capture cross sections measurements performed at ORELA demonstrated the inadequacies of the existing ENDF/B-VI for ^{28}Si in the intermediate energy region. A new evaluation was created by ORNL that included the latest ORELA measurements. In the evaluation process, the benchmark models of the critical experiments performed at the Institute of Physics and Power Engineering were used to evaluate the impact of the use of the ORNL cross section library on the MCNP computed k_{eff} values. Sensitivity analyses were performed as a part of the benchmark evaluation to determine the sensitivity of the critical experiments to the various constituents of the assembly. The benchmark models were then used to determine the computed k_{eff} for various cross section data sets. The variation in the computed k_{eff} value for the new evaluated data set was then used as an indicator to adjust the negative energy capture widths for the capture cross section data while still maintaining excellent agreement between the SAMMY fit and the measured differential data. The resulting ORNL evaluation produced the most consistent evaluation for silicon. This result could only be achieved through integration of many components of the NCSP.

References

1. "International Handbook of Evaluated Criticality Safety Benchmark Experiments," NEA/NSC/DOC (95) 03, Nuclear Energy Agency, Organization for Economic Cooperation and Development (Sep. 2000).
2. J. F. Briesmeister, ed., *MCNP-A General Monte Carlo N-Particle Transport Code*, LA-13709-M, Los Alamos National Laboratory (2000).

3. D. M. Hetrick, D. C. Larson, N. M. Larson, L. C. Leal, and S. J Epperson, "Evaluation of ^{28,29,30}Si Neutron Induced Cross Sections for ENDF/B-VI," ORNL/TM-11825, Oak Ridge National Laboratory (1997).
4. S. F. Mughabghab, M. Divadeenam, and N. E. Holden, "Neutron Cross Sections, Vol. 1, Neutron Resonance Parameters and Thermal Cross Sections, part A: Z=1,60," Academic Press, N. Y. (1981).
5. K. H. Guber, L. C. Leal, R. O. Sayer, R. R. Spencer, P. E. Koheler, T. E. Valentine, H. Derrien, and J. A. Harver, "Neutron Cross Section Measurements for Light Elements at ORELA and Their Application in Nuclear Criticality and Astrophysics," to be published in the Proceedings of the Bologna 2000 – Structure of the Nucleus at the Dawn of the Century, Bologna, Italy, 2000.
6. H. Derrien, L. C. Leal, K. H Guber, T. E. Valentine, and N. M. Larson, "Evalutaion of Silicon Neutron Resonance Paramters in the Energy Range Thermal to 1800 keV," ORNL-TM-to be published, Oak Ridge National Laboratory (2001).
7. N. M. Larson, "Updated Users' Guide for SAMMY: Multilevel R-Matrix Fits to Neutron Data Using Bayes' Equations," ORNL/TM-9179/R5, Oak Ridge National Laboratory (2000).
8. R. E. MacFarlane and D. W. Muir, "The NJOY Nuclear Data Process System Version91," LA-12740-M, Los Alamos National Laboratory (1994).
9. S. Raman, E. T. Jurney, J. W. Starner, and J. E. Lynn, "Thermal-Neutron Caputre by Silicon Isotopes," Phys. Rev**. C46**, 972 (1992).
10. McMaster's values as cited in reference 9.