LA-UR-24-20963

Approved for public release; distribution is unlimited.

Title:	Chlorine Worth Study and Nuclear Data
Author(s):	Thompson, Nicholas William; Cutler, Theresa Elizabeth; Amundson, Kelsey Marie; Favorite, Jeffrey A.; Grove, Travis Justin; Hutchinson, Jesson D.; Kleedtke, Noah Andrew
Intended for:	American Nuclear Society (ANS) Annual Meeting, 2024-06-09 (Las Vegas, Nevada, United States)
Issued:	2024-02-02

Chlorine Worth Study and Nuclear Data

Nicholas W. Thompson,[a,*] Theresa Cutler,[a] Kelsey Amundson,[a] Jeffrey A. Favorite[b], Travis Grove,[a] Jesson Hutchinson,[a] and Noah Kleedtke[c]

[a]*Advanced Nuclear Technology Group, Nuclear Engineering and Nonproliferation Division, Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545*

[b]*Radiation Transport Applications Group, X-Theoretical Design Division, Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545*

[c]*Monte Carlo Methods, Codes, and Applications Group, X-Computational Physics Division, Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545*

[*]*nthompson@lanl.gov*

INTRODUCTION

The Chlorine Worth Study (CWS) was a series of experiments that took place at the National Criticality Experiments Research Center (NCERC), operated by Los Alamos National Laboratory (LANL). The focus of the experiments was to develop new integral benchmarks for the International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook [1] with high sensitivity to chlorine in the thermal neutron energy region and which match sensitivities of aqueous chloride operations at LANL. This work discusses the experiment and how the experimental and simulated results using different nuclear data libraries compare to each other[2, 3].

BACKGROUND

NCERC is the only general purpose critical experiments facility in the western world. It is located at the Device Assembly Facility at the Nevada National Security Site. NCERC has four critical assemblies; Comet, Planet, Flattop, and Godiva IV. Comet and Planet are vertical lift assemblies, Flattop is a spherical assembly with an HEU or Pu core and a 1000kg natural uranium reflector, and Godiva IV is a fast burst assembly. These assemblies, and the diverse nuclear material types and forms at NCERC allow for an incredible amount of flexibility in the types of experiments that can be performed. More information on each assembly can be found in the recent Special Issue of Nuclear Science and Engineering[4, 5, 6, 7].

The CWS experiment used plutonium-metal alloy plates from the Zero Power Physics Reactor (ZPPR) experiments at Argonne-West (now Idaho National Laboratory) with sheets of polyvinyl chloride (PVC), chlorinated polyvinyl chloride (CPVC), and high density polyethylene (HDPE)[8]. By using these materials in different combinations, three configurations were built. These cases were designed to match the sensitivity profiles of ^{35}Cl(n,γ) reaction in aqueous chloride operations at the plutonium facility at LANL. The three cases were designed to cover a range of 20 g/L $PuCl_3$ to 1000 g/L $PuCl_3$. The target design for Case 1 was an equivalent of 30 g/L $PuCl_3$, Case 2 had a target of 300 g/L $PuCl_3$, and Case 3 had a target of 600 g/L $PuCl_3$. The experiment was performed in late 2021 and has since been accepted as a benchmark by the ICSBEP with the "PU-MET-THERM-005" identifier [9]. Case 1 had ZZZ units stacked where each unit was one tray of Pu, and 0.062 in. of PVC sandwiched between two 0.394 in. HDPE moderator plates. Case 2 had ZZZ units stacked where each unit was one tray of Pu and a 0.71 in. thick, 7.9 in. diameter PVC cylinder with HDPE moderator surrounding the PVC. Case 3 had ZZZ units stacked where each unit was one tray of Pu and a 0.87 in. thick, 7.9 in. diameter CPVC cylinder with HDPE moderator surrounding the PVC. All cases had a HDPE reflector on the top, bottom and sides that was 2.9 in thick. Figure 1 shows a CPVC fuel tray position while setting up the experiment and Figure 2 shows a fuel tray with ZPPR plates.

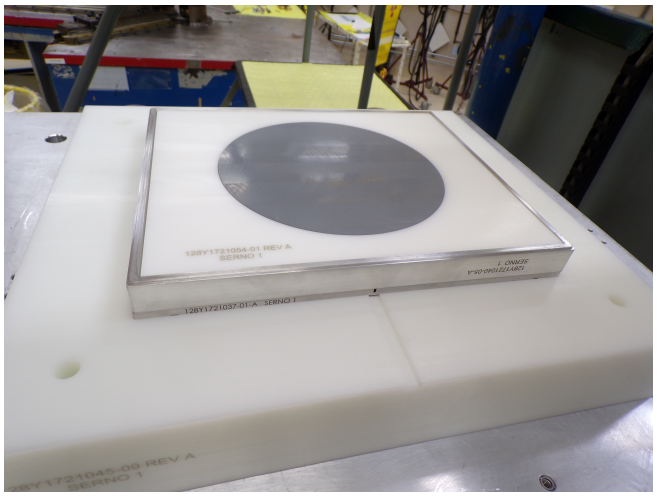

Fig. 1. CPVC cylinder in the CWS experiment.

NUCLEAR DATA COMPARISONS

In order to investigate differences among nuclear data libraries that the CWS experiment is sensitive to, simulations were run using MCNP® Code Version 6.2[1] [10] with the detailed benchmark model using the ENDF/B-VIII.0, ENDF/B-VII.1, JEFF-3.3, JENDL-5.0, and CENDL-3.2 nuclear data libraries [11, 12, 13, 14, 15]. All simulations were run with 200,000 neutrons per cycle, 200 discard batches, and 8000 active batches and had an uncertainty of 2 pcm.

[1]MCNP® and Monte Carlo N-Particle® are registered trademarks owned by Triad National Security, LLC, manager and operator of Los Alamos National Laboratory. Any third party use of such registered marks should be properly attributed to Triad National Security, LLC, including the use of the designation as appropriate. For the purposes of visual clarity, the registered trademark symbol is assumed for all references to MCNP within the remainder of this paper.

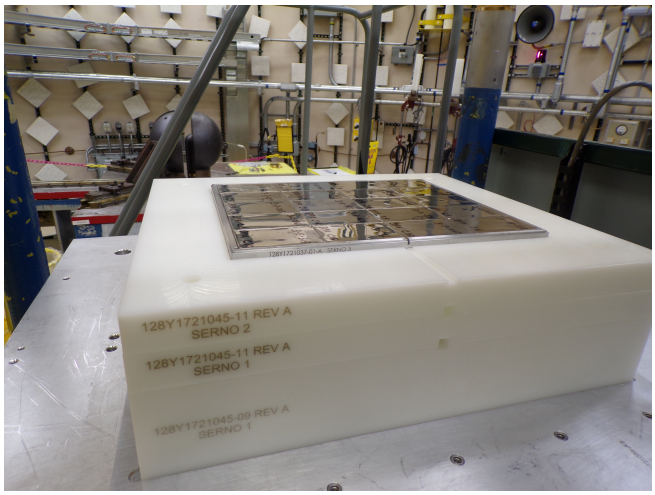

Fig. 2. ZPPR fuel tray in the experiment.

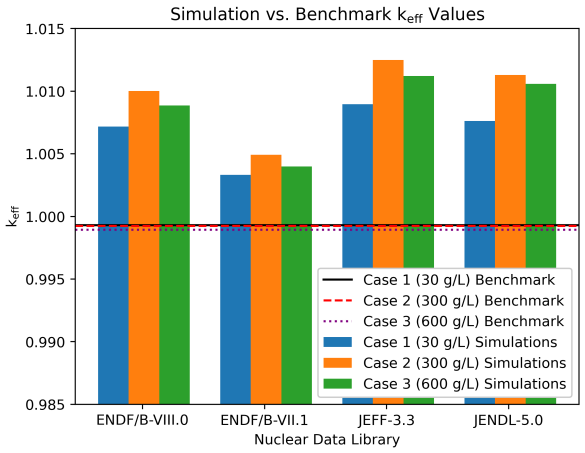

Fig. 3. Benchmark and Simulated k_{eff} values for different nuclear data libraries. g/L values are g/L $PuCl_3$.

RESULTS

Table I contains the benchmark k_{eff} values for the three experimental configurations. Note, the experimental k_{eff} values were all critical, but in generating the detailed benchmark model, there are some modeling biases which need to be taken into account (for example, omitting the floor, ceiling, and walls from the model). Therefore it is more accurate to compare the simulated k_{eff} values with benchmark k_{eff} values instead of experimental k_{eff} values. First, simulations were performed where all isotopes changed to a different nuclear data library (e.g. all isotopes used the ENDF/B-VII.1 library). Results of these simulations can be seen in Table II (where C-E means "computational result minus experimental result") and plotted in Figure 3. Note, simulations for CENDL-3.2 were not run for this as CENDL-3.2 contains many more natural element cross sections and it would have taken a significant amount of time to change the input file to run this case. This case can be run in the future, but at present time it was not deemed worth the effort.

TABLE I. Benchmark k_{eff} Values

	k_{eff}	uncertainty
Case 1	0.99930	0.00255
Case 2	0.99924	0.00253
Case 3	0.99894	0.00435

As can be seen in Table II and Figure 3, all libraries simulate higher k_{eff} values than the benchmark values for all three cases. This indicates there is significant room for improvement in the nuclear data this benchmark is sensitive to. At first glance there doesn't seem to be a clear trend with Cl concentration, since for each of the libraries the 600 g/L $PuCl_3$ simulation result is between the 30 and 300 g/L $PuCl_3$ results. However, there still may be a trend with Cl concentration that is hidden due to the fact that the 600 g/L $PuCl_3$ case uses CPVC instead of PVC, which has a higher concentration of Cl, but may also be adding another variable.

To investigate this further, simulations were run where the ENDF/B-VIII.0 nuclear data library was used for all cross sections except for one element or isotope was changed to a different library (e.g. everything ENDF/B-VIII.0 except ^{239}Pu was JEFF-3.3). By doing this for all the most important elements and isotopes, individual changes between libraries can be readily identified and quantified. Table III shows the results of these simulations, where the ENDF/B-VIII.0 k_{eff} value is subtracted and the difference is expressed in terms of pcm (e.g. JEFF-3.3 ^{239}Pu - ENDF/B-VIII.0 base case). For these simulations, CENDL-3.2 was included except for ^{35}Cl as CENDL-3.2 just has a natural Cl evaluation and the polyethylene TSL as it does not have any TSL evaluations.

The results show (somewhat unsurprisingly) that the isotope that makes the biggest impact on k_{eff} when comparing nuclear data libraries is ^{239}Pu. There are large differences (>500 pcm) between ENDF/B-VII.1 and ENDF/B-VIII.0, significant differences between JEFF-3.3 and CENDL-3.2 compared to ENDF/B-VIII.0 (up to 250 pcm) and minor differences between ENDF/B-VIII.0 and JENDL-5.0. The minor differences between ENDF/B-VIII.0 and JENDL-5.0 are due to the fact that both evaluations are based on the CIELO project[16]. It's clear that somewhat minor changes in plutonium cross sections between nuclear data libraries lead to large differences in simulated k_{eff} values for this system.

The main focus of this project was Cl, specifically ^{35}Cl, so focusing on Cl next, we see large differences between ENDF/B-VIII.0 and JENDL-5.0, but only for Cases 1 and 2. This would suggest there is some trend with Cl for these two libraries, with the vast majority of that difference coming from ^{35}Cl. Figure 4 is a plot of ^{35}Cl (n,γ) cross sections for the different libraries. When looking at the underlying cross sections, below 1.2 MeV, ENDF/B-VIII.0, ENDF/B-VII.1, and JEFF-3.3 all use virtually the same evaluation. JENDL-5.0 is also virtually the same until 100 keV, where JENDL-5.0 is significantly different compared to the other libraries. This seems to be leading to the large differences between it and ENDF/B-VIII.0 in Cases 2 and 3. Given that ENDF/B-VIII.0, ENDF/B-VII.1, and JEFF-3.3 essentially are identical evaluations below 1.2 MeV, it's likely that ^{35}Cl is one of the major

TABLE II. Simulated k_{eff} values with different nuclear data libraries

Library	Case 1	C-E	Case 2	C-E	Case 3	C-E
ENDF/B-VII.1	1.00332 +/- 0.00002	402	1.00492 +/- 0.00002	568	1.00398 +/- 0.00002	504
ENDF/B-VIII.0	1.00717 +/- 0.00002	787	1.01002 +/- 0.00002	1078	1.00886 +/- 0.00002	992
JEFF-3.3	1.00896 +/- 0.00002	966	1.01249 +/- 0.00002	1325	1.01121 +/- 0.00002	1227
JENDL-5.0	1.00763 +/- 0.00002	833	1.01130 +/- 0.00002	1206	1.01058 +/- 0.00002	1164

TABLE III. Simulated k_{eff} values - ENDF/B-VIII.0 values with one isotope replaced, in pcm.

Case 1 Library	C	Cl	^{35}Cl	H	Al	Pu	^{239}Pu	^{240}Pu	U+Np+Am	Poly TSL
ENDF/B-VII.1	29	-2	-3	35	-4	-548	-486	-63	-1	99
JEFF-3.3	31	-5	-8	33	0	141	148	-8	-72	68
JENDL-5.0	-9	19	18	-5	-55	15	70	-60	-63	127
CENDL-3.2	36	-150	N/A	98	30	-20	163	-185	51	N/A
Case 2 Library	C	Cl	^{35}Cl	H	Al	Pu	^{239}Pu	^{240}Pu	U+Np+Am	Poly TSL
ENDF/B-VII.1	30	2	0	39	2	-642	-577	-68	0	79
JEFF-3.3	35	-2	-8	34	0	239	257	-10	-73	47
JENDL-5.0	-4	143	129	-1	-56	27	83	-61	-67	98
CENDL-3.2	35	-70	N/A	90	34	-245	-64	-181	50	N/A
Case 3 Library	C	Cl	^{35}Cl	H	Al	Pu	^{239}Pu	^{240}Pu	U+Np+Am	Poly TSL
ENDF/B-VII.1	25	-4	-1	31	-6	-628	-571	-68	0	88
JEFF-3.3	25	-14	-9	32	-3	215	222	-13	-70	56
JENDL-5.0	-10	159	148	4	-43	10	63	-54	-67	114
CENDL-3.2	36	-89	N/A	77	31	-272	-98	-179	45	N/A

culprits causing the significant differences between experimental and simulated k_{eff} values. New differential cross section measurements of Cl would be useful in determining if this is indeed a cause of the large discrepancy between experimental and simulated results. This also highlights the importance of independent evaluations: it's possible there was an issue with the ENDF/B-VII.1 evaluation the other libraries are based on, which might have been caught if two evaluations significantly disagreed with each other. Additionally, often different evaluators will make different choices in which data to incorporate that can result in significantly different evaluations which may perform better. In the case of CENDL-3.2, it's significantly different compared to the other evaluations (differences up to 150 pcm), and it also results in lower k_{eff} values that the other evaluations - another sign that chlorine might be one of the main culprits in the C-E discrepancy. Another important note here is the CPVC was very difficult to characterize. Multiple samples were sent to multiple labs using different techniques for quantifying composition and impurities and the results were not consistent. Much work was done to figure out the composition as precisely as possible[3].

Another one of the biggest differences between the libraries is the polyethylene thermal scattering law (TSL) libraries. ENDF/B-VII.1, JEFF-3.3, and JENDL-5.0 all have

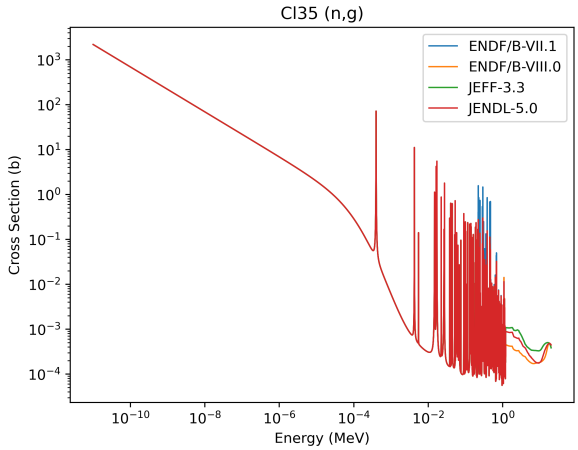

Fig. 4. ^{35}Cl (n,γ) cross sections.

polyethylene TSL libraries, and all result in a higher simulated k_{eff} when compared to ENDF/B-VIII.0. There are other differences between libraries that result in k_{eff} changes of less than 100 pcm, for example, Carbon, Hydrogen, Aluminum, Uranium, Neptunium, and Americium. These are statistically

significant, but unlikely to make up the bulk of the C-E discrepancy.

CONCLUSIONS

This paper investigates the large difference between computational and benchmark k_{eff} results for the CWS experiment series. All nuclear data libraries tested calculate much higher than the benchmark k_{eff} value. To investigate further, MCNP was used to isolate how changes in cross sections between nuclear data libraries results in changes in k_{eff}. While it's unclear what the exact cause of the C-E difference is, it's possible that plutonium and chlorine are the two main causes.

ACKNOWLEDGMENTS

Special thanks to everyone who helped with the experiment, including Joetta Goda, Jessie Walker, Kenneth Valdez, Justin Martin, and the NCERC-FO team.

REFERENCES

1. NUCLEAR ENERGY AGENCY, editor, *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, OECD Nuclear Energy Agency, Paris, France (2021).
2. T. CUTLER, K. AMUNDSON, J. HUTCHINSON, N. KLEEDTKE, and N. WYNNE, "The CWS Experiments - An Experimental Study of the Effects of Chlorine on Thermal Neutron Absorption," *American Nuclear Society Annual Meeting* (2022).
3. T. CUTLER, T. GROVE, and J. HUTCHINSON, "Polymer Compositions in Critical Experiments: Possible Not What You Think," *Transactions of the American Nuclear Society*, **129**, *1*, 646–648 (November 2023).
4. N. THOMPSON ET AL., "A New Era of Nuclear Criticality Experiments: The First 10 Years of Comet Operations at NCERC," *Nuclear Science and Engineering*, **195**, *sup1*, S17–S36 (2021).
5. R. SANCHEZ ET AL., "A New Era of Nuclear Criticality Experiments: The First 10 Years of Planet Operations at NCERC," *Nuclear Science and Engineering*, **195**, *sup1*, S1–S16 (2021).
6. D. HAYES ET AL., "A New Era of Nuclear Criticality Experiments: The First 10 Years of Flattop Operations at NCERC," *Nuclear Science and Engineering*, **195**, *sup1*, S37–S54 (2021).
7. J. GODA ET AL., "A New Era of Nuclear Criticality Experiments: The First 10 Years of Godiva IV Operations at NCERC," *Nuclear Science and Engineering*, **195**, *sup1*, S55–S79 (2021).
8. A. B. SHUCK, H. F. JELINEK, A. G. HINS, N. J. CARSON JR, A. A. DENST, and T. A. STEELE, "The Development of a Design and Fabrication Method for Plutonium-Bearing Zero-Power Reactor Fuel Elements," Tech. Rep. ANL-7313, Argonne National Laboratory (1967).
9. J. FAVORITE and T. CUTLER, "Thermal-Spectrum Critical Assemblies with a Polyvinyl Chloride and Chlorinated Polyvinyl Chloride-Plutonium-Aluminum Metal Alloy Core Surrounded by a Polyethylene Reflector," *In: International Handbook of Evaluated Criticality Safety Benchmark Experiments*, **Submitted - [DVD]/Nuclear Energy Agency. - Paris : OECD Nuclear Energy Agency**, (NEA;7328).
10. C. WERNER ET AL., "MCNP Users Manual - Code Version 6.2," *Los Alamos National Laboratory*, **LA-UR-17-29981** (2017).
11. D. BROWN ET AL., "ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data," *Nuclear Data Sheets*, **148**, 1 – 142 (2018), special Issue on Nuclear Reaction Data.
12. M. CHADWICK ET AL., "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data," *Nuclear Data Sheets*, **112**, 2887–2996 (2011).
13. A. J. M. PLOMPEN ET AL., "The joint evaluated fission and fusion nuclear data library, JEFF-3.3," *The European Physical Journal A*, **56**, *7*, 181 (2020).
14. O. IWAMOTO ET AL., "Japanese evaluated nuclear data library version 5: JENDL-5," *Journal of Nuclear Science and Technology*, **60**, *1*, 1–60 (2023).
15. GE, ZHIGANG ET AL., "CENDL-3.2: The new version of Chinese general purpose evaluated nuclear data library," *EPJ Web Conf.*, **239**, 09001 (2020).
16. M. CHADWICK ET AL., "CIELO Collaboration Summary Results: International Evaluations of Neutron Reactions on Uranium, Plutonium, Iron, Oxygen and Hydrogen," *Nuclear Data Sheets*, **148**, 189 – 213 (2018), special Issue on Nuclear Reaction Data.