Preliminary Chlorine Worth Study Benchmark Evaluation

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INTRODUCTION

The Chlorine Worth Studies (CWS) experiments with polyvinyl chloride (PVC with chemical formula $(C_2H_3Cl)_n$), chlorinated polyvinyl chloride (CPVC with chemical formula $(C_9H_{11}Cl_7)_n$), and high density polyethylene (HDPE with chemical formula $(CH_2)_n$) were a series of measurements performed at the National Criticality Experiments Research Center (NCERC). The purpose of the CWS experiments was to perform integral experiments that were highly sensitive to the thermal ^{35}Cl(n,γ) reaction and matched the sensitivities of aqueous chloride operations at the plutonium facility at Los Alamos National Laboratory (LANL). The CWS experiments were performed on the Planet critical assembly machine at NCERC and utilized weapons grade plutonium (WGPu) plates as fuel.

The design process and design of the CWS experiment were discussed previously [1]. This paper discusses the benchmark evaluation of the experiment, intended for the International Criticality Safety Benchmark Evaluation Project (ICSBEP). Criticality calculations were performed with MCNP version 6.3 [2]. Results presented here are preliminary, as the benchmark has not yet been submitted to the ICSBEP.

DESCRIPTION OF THE EXPERIMENT

There were three experimental configurations, each attempting to replicate the neutron spectrum and cross-section sensitivities of a specific concentration of plutonium in an aqueous chloride solution. The three concentrations were 30, 300, and 600 (g plutonium)/(L solution).

The experiments were fueled with the Plutonium Aluminum No Nickel (PANN) Zero Power Physics Reactor (ZPPR) plates [3, 4], which are steel-clad Pu-Al alloy plates nominally 2 in. × 3 in. × 0.125 in. The Pu-Al alloy is nominally 1.1 wt.% aluminum and has a density of approximately 15.09 g/cm^3. Each plate contains approximately 98.5 g ^{239}Pu.

Twenty ZPPR plates were arranged in a 4 × 5 horizontal array within an aluminum tray. HDPE moderators and either PVC or CPVC absorbers were stacked on the ZPPR plates. Walls built into the tray contained the fuel plates, while an aluminum frame stacked on the walls of the tray contained the moderators and absorbers. Each tray and frame of fuel plates, moderators, and absorbers comprised a *unit*. Units were stacked to form the critical configurations (or cases). The number of units and type of absorber in each configuration is given in Table I. The mass of HDPE moderator per unit and the mass of absorber per unit are also given in Table I. A rendering of Case 2 is shown in Fig. 1, and stacked trays and frames are shown in Fig. 2.

Nylon set screws were used to compress the rows and columns of the ZPPR plate array, both to reduce the possibility of gaps and to reduce the uncertainty associated with the location of each plate. Two of the nine set screws per unit are shown in Fig. 2.

The core of stacked units was reflected on the top, bottom, and sides by blocks of HDPE. Long aluminum bolts were used to compress the stacks of HDPE blocks to reduce the possibility of gaps and misalignment.

EVALUATED k_{eff}

Measured reactor periods were used in the Inhour equation to infer the experiment k_{eff} for each case. Results are shown in Table II. Delayed neutron decay constants and relative abundances for each of the delayed neutron groups for ^{239}Pu thermal fission [5] were used. Main uncertainties were due to the reproducibility of the experiment and the delayed neutron data.

TABLE II. Evaluated k_{eff} (1σ Uncertainty).

Case	k_{eff}
1	1.00050 (+0.00004 / –0.00005)
2	1.00047 (+0.00004 / –0.00005)
3	1.00045 (+0.00004 / –0.00005)

TABLE I. CWS Case Summaries.

Case	Number of Units	Absorber	Mass of Moderator(a) per Unit (g)	Mass of Absorber per Unit (g)
1	8	PVC	1500.24	174.81
2	14	PVC	794.57	802.41
3	18	CPVC	973.18	1015.83

(a) Moderator is HDPE in all cases.

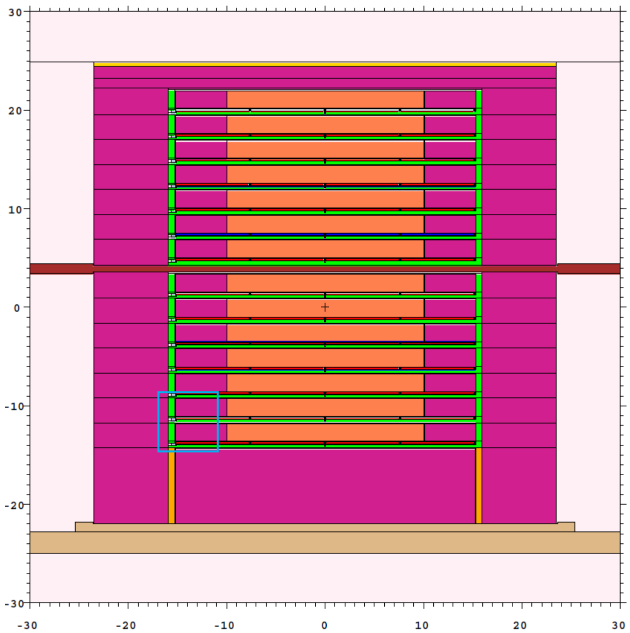

Figure 1. Midplane slice through Case 2. Trays and frames are green; HDPE is magenta; PVC is orange; fuel is red. The blue square indicates the region shown in Fig. 2. Dimensions in centimeters.

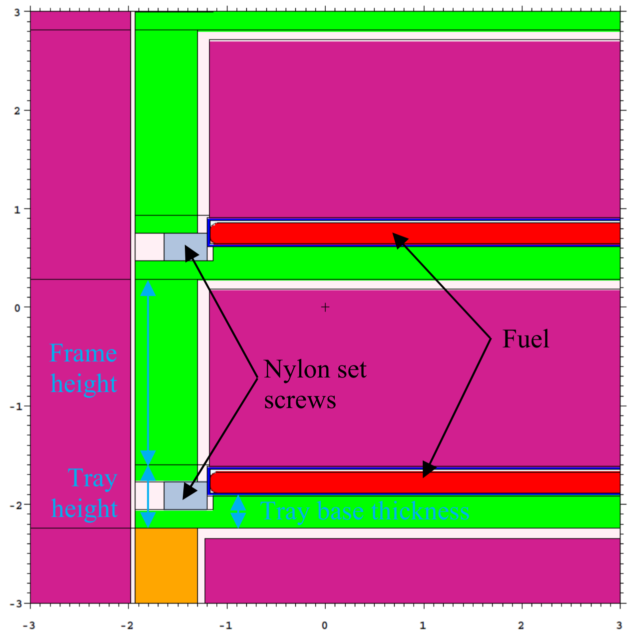

Figure 2. Close-up on the lowest two units of Case 2 (see Fig. 1) showing how the tray and frame heights affect the fuel separation distance. Nylon set screws for fixing the fuel plates in place are also shown. Dimensions in centimeters.

UNCERTAINTY ANALYSIS

Uncertainty Due to PANN Cores

The fuel plates used in these experiments, the PANN cores within ZPPR plates, are not well characterized. The ADEN database [4] gives the mass of ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and Al in each core as well as the "core" mass, but the sum of the given nuclide masses is ~0.007 g less than the given core mass. The composition of the missing mass is unknown.

In 2018, staff at Lawrence Livermore National Laboratory (LLNL) sampled the PANN core of a ZPPR plate, choosing plate number J18, and destructively analyzed its composition. Decay products were present in the fuel that are not accounted for by the decay of the composition given in the ADEN database, and some actinides are present in discrepant quantities. There was clearly ^{238}U, ^{237}Np, and ^{238}Pu in the initial material. There was evidently about 10 % more ^{241}Pu than was measured in 1960 and about 2 % more ^{240}Pu.

J18 also contained impurities that weighed ~0.165 g, too large by a factor of ~20 to explain the 0.007-g difference in the ADEN database.

Furthermore, the mass of the PANN core of ZPPR plate J18 was 105.4 g [6]. In the ADEN database, the mass of the core is 105.621 g and the sum of the nuclide masses is 105.608 g. Thus, there is a 0.2-g difference (0.19 %) between the historic records for this PANN core and the new measurement. The mass uncertainty for all PANN cores was taken to be 0.19 %.

In addition, the dimensions of the PANN cores and therefore their densities are extremely uncertain. The uncertainty in the volume (as calculated using design drawing dimensions) is +2.9 %/–2.4 %. Pu-Al alloy was found to have a density of 15.11 to 15.27 g/cm^3 [7], but the modeled volume yields densities around 15.09 g/cm^3.

Despite the large uncertainties associated with the PANN core masses and dimensions, their contribution to the CWS experiment uncertainties are surprisingly small. These are shown in Table III. The uncertainty of the PANN core composition is the largest contributor.

Uncertainty Due to Temperature

Most of the units in each case were instrumented with RTDs (resistance temperature detectors). The temperature of a unit varied from the ensemble average by as much as –1.5 °C, –2.8 °C, and +4.7 °C in Cases 1, 2, and 3, respectively. The uncertainty of the ensemble average temperature was assumed to be ±5 °C in all cases; this value was intended to account for the measurement uncertainty as well as the temperature variation within the assemblies.

The effect of temperature was evaluated by changing the temperature of the S(α,β) thermal scattering data; changing the free-gas thermal temperature; changing the nuclear data temperature by computing cross sections for

TABLE III. k_{eff} Uncertainties (with 1σ Relative Uncertainties of the Uncertainties) Due to PANN Cores.

Parameter	Case	k_{eff} Uncertainty
Mass of ZPPR Plate PANN Cores	1	± 0.00011 ± 0.16 %
	2	± 0.00011 ± 0.13 %
	3	± 0.00010 ± 0.13 %
ZPPR PANN Core Dimensions	1	± 0.00009 ± 1.80 %
	2	± 0.00007 ± 1.62 %
	3	± 0.00007 ± 1.54 %
Composition of ZPPR Plate PANN Cores	1	± 0.00027 ± 0.13 %
	2	± 0.00031 ± 0.12 %
	3	± 0.00031 ± 0.12 %
Location of PANN Cores Within ZPPR Plate	1	Negligible
	2	Negligible
	3	Negligible
Total Due to PANN Cores	1	± 0.00031 ± 0.19 %
	2	± 0.00034 ± 0.12 %
	3	± 0.00033 ± 0.12 %

^{239}Pu, ^{240}Pu, ^{35}Cl, and ^{37}Cl using NJOY [8]; and applying thermal contraction and expansion to the dimensions in each assembly. Results are shown in Table IV. By far the largest contribution was due to the effect of thermal scattering.

Dimensional Uncertainties

The other important uncertainty is due to uncertainty in the height of the walls of the trays and frames and the thickness of the bottom of the trays. These dimensions are important because the trays and frames stack together, dictating the spacing between fuel layers (see Fig. 2). These uncertainties are shown in Table V.

Total Experimental Uncertainty

Only the largest sources of uncertainty are presented here. The total experimental uncertainty for Cases 1, 2, and 3 was evaluated to be ±0.00087, ±0.00061, and ±0.00066, respectively. The statistical uncertainty of these uncertainties is estimated to be ±2 %.

MODELING BIAS

A very detailed MCNP6.3 model of the CWS experiments has been constructed. Within the assemblies, many simplifications were made that were judged to have a negligible effect on the calculated k_{eff}.

Four features of the surroundings were evaluated explicitly: 1) the presence of Flattop in the room with Planet; 2) the presence of a crane in the room; 3) the presence of walls, ceiling, and floor; and 4) the presence of some Planet assembly components not included in the benchmark. The first two of these had a negligible effect

TABLE IV. k_{eff} Uncertainties (with 1σ Relative Uncertainties of the Uncertainties) Due to Temperature.

Case	Slope (/°C)	k_{eff} Uncertainty[(a)]
1	0.00014 ± 0.00000	± 0.00070 ± 2.53 %
2	0.00006 ± 0.00000	± 0.00030 ± 5.91 %
3	0.00009 ± 0.00000	± 0.00045 ± 3.94 %

(a) Using a temperature uncertainty of ±5 °C.

TABLE V. k_{eff} Uncertainties (with 1σ Relative Uncertainties of the Uncertainties) Due to Tray and Frame Dimensions.

Case	k_{eff} Uncertainty
1	± 0.00031 ± 0.28 %
2	± 0.00030 ± 0.22 %
3	± 0.00025 ± 0.23 %

on k_{eff}. The effect of the second two together (walls, ceiling, and floor and Planet components) is shown as the simplification bias in Table VI. The walls, ceiling, and floor contributed –-0.00003, –0.00009, and –0.00004 for Cases 1, 2, and 3, respectively, very little compared to the Planet assembly, but not negligible.

The benchmark temperature is 20 °C, but the assembly average experiment temperatures were 4.0, 10.1, and 11.1 °C greater for Cases 1, 2, and 3, respectively. The temperature bias was calculated considering the four temperature effects discussed previously. The results are shown in Table VI.

The total modeling bias is shown in Table VI.

BENCHMARK k_{eff} AND COMPARISON WITH SAMPLE CALCULATION

The experiment k_{eff} and its evaluated uncertainty, the modeling bias and its uncertainty, and the final benchmark k_{eff} and its uncertainty are shown in Table VII. There were small asymmetric uncertainties (such as those shown in Table II) that were not large enough to cause the total uncertainty to be asymmetric.

The k_{eff}'s calculated using MCNP6.3 and ENDF/B-

TABLE VI. Modeling Biases.

Type	Case	Value
Simplification Bias	1	–0.00042 ± 0.00004
	2	–0.00032 ± 0.00004
	3	–0.00019 ± 0.00003
Temperature Bias	1	–0.00056 ± 0.00001
	2	–0.00061 ± 0.00004
	3	–0.00100 ± 0.00004
Total Bias[(a)]	1	–0.00098 ± 0.00008
	2	–0.00093 ± 0.00006
	3	–0.00119 ± 0.00005

(a) The uncertainty includes a small component associated with the distribution of fuel mass.

TABLE VII. Benchmark k_{eff}.

Case	Experiment k_{eff}	Modeling Bias[(a)]	Benchmark Model k_{eff}
1	1.00050 ± 0.00087	–0.00098 ± 0.00008	0.99952 ± 0.00087
2	1.00047± 0.00061	–0.00093 ± 0.00006	0.99954 ± 0.00061
3	1.00045± 0.00066	–0.00119 ± 0.00005	0.99926 ± 0.00066

(a) From Table VI.

VIII.0 nuclear data are compared with the benchmark k_{eff}'s in Table VIII. The calculated k_{eff}'s are far outside 3σ for Cases 1 and 2 but inside 1σ for Case 3.

TABLE VIII Sample Calculation Results.

Case	Calculated k_{eff}	Calc.–Expt. (pcm)
1	1.00257 ± 0.00002	305
2	1.00487 ± 0.00002	533
3	0.99985 ± 0.00002	59

SUMMARY AND CONCLUSIONS

This paper presents the preliminary evaluation of the CWS experiments that were performed to provide new integral measurements by which to validate nuclear data for chlorine at thermal neutron energies.

These experiments were designed to minimize or eliminate uncertainties due to gaps and part positioning. That goal was met. A large uncertainty now was due to the uncertain dimensions that set the separation distance of the fuel layers. Future designs may target reduction in those uncertainties in addition to the improvements in gaps and positioning.

Another large uncertainty is due to the composition of the fuel, the PANN cores of the ZPPR plates. Destructive analysis of one PANN core in 2018 provided new data that will improve the benchmarks using these plates, but the new data's inconsistency with the original data has increased rather than decreased the uncertainties associated with the ZPPR plates.

The CWS experiments were quite sensitive to temperature, and both the uncertainty due to uncertain and varying temperature and the bias due to elevated operating temperature were large.

A simplified model is also being developed.

The results presented here will undergo several thorough reviews before being published in the ICSBEP Handbook.

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