Validation and Independent Uncertainty Analysis of the MIX-SOL-THERM-003 ICSBEP Benchmark

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INTRODUCTION

The International Criticality Safety Benchmark Evaluation Project (ICSBEP) was started in 1992 by the United States Department of Energy and later in 1995 became an international project with contributions from 22 countries. The project is now organized by the OECD (Organisation for Economic Co-operation and Development) Nuclear Energy Agency (NEA). In its most recent iteration, the ICSBEP handbook contains over five thousand evaluations of critical, near-critical, and subcritical experiments conducted in facilities all around the world. These benchmarks serve as valuable information for criticality safety engineers who can use them to validate calculation techniques and establish minimum subcritical margins for operations with fissionable materials.

The benchmarks in the handbook are categorized by their fissile material composition, material form (oxide, solution, or metal), and fission energy spectra. This is especially useful for those looking for benchmarks similar to a system they are working on to compare methods and identify trends.

The ICSBEP Handbook Uncertainty Guide [1] is document outlining recommended practices and methods for determining uncertainties in these benchmarks. Quantifying these uncertainties thoroughly is crucial as it allows a higher degree of confidence that data used from them is valid and relevant. The guide stresses the importance of a thorough and well documented uncertainty analysis when evaluating an experiment. All measured values of a system, whether they be dimensions or material compositions, have a certain amount of uncertainty associated with them and can be analyzed one by one to determine their effects on the system. Many evaluated benchmarks in the handbook present this in detail, however some do not, mostly earlier evaluations performed in the 1990's and early 2000's.

Recently at Los Alamos National Laboratory (LANL), the Nuclear Criticality Safety Division (NCSD) of LANL has been validating MCNP6.2® input files of criticality benchmarks for use by Whisper [2], a criticality code developed at LANL. This effort is also part of the OECD NEA Working Party on International Nuclear Data Evaluation Co-operation (WPEC) Subgroup 45 [3], also known as Validation of Nuclear Data Libraries (VaNDaL). The goal of VaNDaL is to compile a set of validated simulation inputs for use in validating nuclear data and simulation codes. As part of these efforts, one of the benchmarks reviewed was the MIX-SOL-THERM-003 ICSBEP benchmark [4]. This paper provides an independent uncertainty analysis of this benchmark experiment.

EXPERIMENT OVERVIEW

The experiments that served as the basis for MIX-SOL-THERM-003 were conducted in the early 1970's by the United Kingdom Atomic Energy Agency (UKAEA) at the AWRE Aldermaston site using the SCAMP (Solution Criticality Assembly Machine – Plutonium) facility. The assembly consisted of interchangeable stainless steel vessels of differing size. A vessel was submerged in water and filled with different fissile solutions of Pu and natural U. On top of the solution rested a 15 cm thick polyethylene plug to imitate an infinite hydrogenous reflector. A 1/M plot was made by measuring reciprocal neutron counts at incremental fissile solutions heights, this data could then be used to project the true critical height.

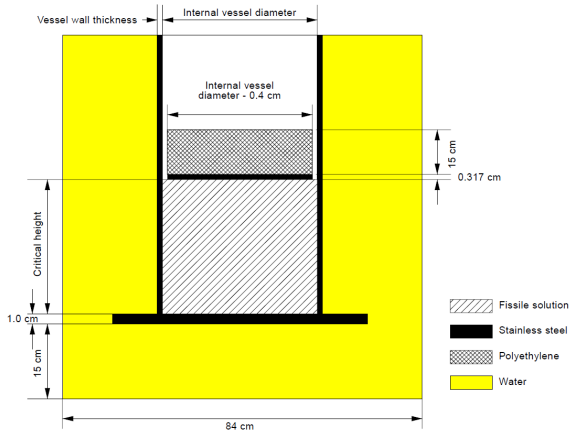

Figure 1 – Simplified experiment geometry [4]

Two variables were used to provide a range of experimental configuration: the size of the stainless steel vessels and the Pu/U concentration of the fissile solution. A total of eight combinations of four different vessels and four different solutions were reviewed in the benchmark.

Along with uncertainties in the physical dimensions of the system, the experimenters also

recorded uncertainties in the material composition of the fissile solutions used.

COMPARISON OF CALCULATIONS WITH EXPERIMENTS

The experimenters recorded a near critical solution height for Case 1 to be 56.31cm, corresponding to a k_{eff}=0.9985. When modeled by the ICSBEP evaluators using the MONK [5] simulation code and the UKNDL-2 nuclear data library the k_{eff} of that same system was calculated to be 1.0125±0.0011. This computational result provided by the evaluators is in close agreement with simulations run by LANL NCSD. Using MCNP6.2® [6] and ENDF-B/VIII.0 nuclear data library [7] the k_{eff} was found to be 1.01279±0.00004.

This large bias as well as a lack of a detailed uncertainty analysis presented in the ICSBEP handbook evaluation are the basis for performing this independent uncertainty analysis. Typically, handbook evaluations will present the effects of uncertainties of measured values on k_{eff} at one standard deviation. While a total uncertainty in k_{eff} is presented by the evaluators there is very little documentation on the methods used to achieve that value. The evaluators only assessed uncertainties from the critical height, vessel radius, and ^{239}Pu and ^{235}U concentrations. A well documented analysis consistent with the ICSBEP Uncertainty Guide [1] analyzes as many sources of uncertainty as possible while making conservative assumptions when needed. In this case, much more information was provided by the experimenters that appears to have been neglected by the handbook evaluators. The aim of this independent uncertainty analysis is to better document these values to potentially gain insight into the cause of the large discrepancy between the experimental k_{eff} and the computational k_{eff}.

METHODS OF EVALUATION

The geometries and material compositions of experiments can never be known with perfect accuracy, therefore modeling the neutronic behavior of the system will always have uncertainty associated with it. The principle neutronic parameter analyzed by criticality benchmarks evaluated in the ICSBEP handbook is the k_{eff} of the system, the effective multiplication factor, henceforth referred to as k. A first order estimate of the uncertainty of k can be made by varying the experimental parameters in computational simulations.

In some cases, the upper and lower bounds of each parameter given by the experimenters do not represent the standard deviation, rather the maximum and minimum values. An assumption made for this analysis is that the true value of the parameter is equally probable to be anywhere within those bounds, rather than the probability following a Gaussian distribution. This assumption is recommended by the ICSBEP Uncertainty Guide for values given without a standard deviation [1].

With this assumption that the probability is equal within the bounds we can derive the standard uncertainty. This standard uncertainty is equal to one standard deviation of uncertainty.

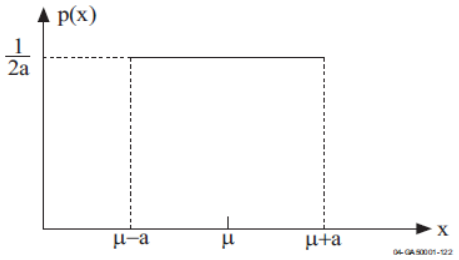

Figure 2 – Probability Density Function p(x) for arbitrary parameter [1]

For the total probability over the interval to be 1 and the probability to be equal at all points p(x) must equal 1/(2a). Therefore the variance is as follows:

Eq. (1) $$\sigma^2(x) = \int_{\text{all } x} (x-\mu)^2 p(x)dx = \int_{\mu-a}^{\mu+a} (x-\mu)^2 \left(\frac{1}{2a}\right) dx$$

Evaluating the integral yields $\sigma(x)=a/\sqrt{3}$. This means to the standard uncertainty, u, of a measurement is equal to the variation divided by the square root of three. Running simulations at these bounds will yield:

Eq. (2) $$k_u = k_{ref} \pm \Delta k_u$$

Here, k_{ref} is the k of the system using the reference values and Δk_u is the change in k using variation of one standard deviation, the standard uncertainty.

Occasionally, the Δk_u found using the perturbation, u, is too small to properly discern from statistical noise and to account for numerical convergence limits. To compensate, a larger perturbation can be used, δx, where $\delta x > u$. It is important to ensure that the response in k to this larger perturbation is linear, a first order effect. Then $\Delta k_u = u\, \delta k/\delta x$ can be used to find the desired change in k associated with the standard uncertainty.

Parameters lacking additional information were assumed to have bounds represented by one sigma (see Table 1). For these cases the given bounds are already equal to the standard uncertainty. This assumption results in a conservative estimation of the actual uncertainty and was used due to a lack of information.

Table 1: Uncertainty Analysis

Parameter	Measured Value	Variation	Standard Uncertainty (*u*)	Perturbation Δk (pcm) -	+	Sensitivity Δk/Δx (pcm)	Δk(1σ) (pcm)
Internal Radius (cm)	12.7125	0.01	0.01	-93	96	9470	**95**
Critical Height (cm)	56.31	0.06	0.06	-17	18	292	**18**
H/Pu (by atoms)	224	1	1	-77	72	75	**75**
Pu/(Pu+U)Wt%	30.7	0.2	0.2	-48	49	242	**48**
Density (g/mL)	1.524	0.001	0.001	-36	33	34500	**34**
Pu (mg/mL)	101.3	0.2	0.2	-16	17	83	**17**
U (mg/mL)	228.5	1	1	-23	24	24	**24**
NO3 (mg/mL)	319	3	3	-50	47	16	**48**
H2O (mg/mL)	857	2	2	-78	75	38	**76**
N/Pu (by atoms)	12.1	0.1	0.1	3	-1	-23	-2
(atoms per mlx10^20)							
Pu239	2.396	0.006	0.006	-32	32	5383	32
Pu240	0.144	0.001	0.001	32	-34	-32800	-33
Pu241	0.011	0.001	0.001	-12	8	10000	10
U235	0.042	0.01	0.01	-84	92	8800	88
U238	5.739	0.02	0.02	-17	18	880	18
H	573	2	2	-59	53	28	56
N	21	0.3	0.3	2	-2	-7	-2
O	386	1	1	-57	57	57	57
Temperature (˚C)	26	3	1.73205	74	-49	-21	**-36**
Am241	5	5	5	94	-94	19	**-94**

Case 1 - Total Uncertainty (Δk_{eff} (1σ)): 194pcm

Once these values are found for each parameter recorded with uncertainty, they can be summed in quadrature to find the total uncertainty in k in the experiment. Assuming parameters are independent, they can be summed as follows where i represents the i^{th} parameter varied up to n. The parameters not being summed are the atoms per milliliter values as they were derived from the other parameters. Counting both would result in double counting. The bold values in Table 1 represent the values summed.

Eq. (3) $$\Delta k_{eff}(1\sigma)=\sqrt{\sum_{i=1}^{n} \Delta k^2{}_{u,i}}$$

The reported uncertainty of the vessel radius in the ICSBEP Handbook for Case 1 is reported at ±0.005cm, however, the other uncertainty for other comparable vessel radii range from two to eight times this uncertainty. Measurements appear to be made to the nearest eight of a millimeter, this does not correspond to an uncertainty of 0.005cm. For this reason a more conservative estimate of the uncertainty was set at 0.01cm.

DISCUSSION OF UNCERTAINTIES

The ISCBEP evaluators recorded a total uncertainty of 200pcm at one sigma for Case 1 of the benchmark. However, it is very unclear how they ascertained this value. Only four variables were evaluated: critical height, vessel radius, and ^{239}Pu and ^{235}U concentrations. The sum of the uncertainties in quadrature they found in these parameters is only 37pcm.

In our independent analysis we also began by solely looking at Case 1. Some of the major individual uncertainties in k unveiled include the vessel internal radius, the H/Pu ratio, and the H_2O and ^{235}U concentrations. These factors can largely be attributed to the fact that this system is highly thermal with over 90% of fissions caused by neutrons of less than 0.625eV. With this being the case we would expect any changes to the moderator as well as fissile materials with large fission cross sections in the thermal range to have a sizable impact on the k.

In addition to analyzing uncertainties of measured parameters, there was a need to determine the uncertainty associated with the age of the Pu used in the solution. No information was found in the ICSBEP evaluation nor the original experiment [8] concerning the age of the Pu used. This is an important factor as ^{241}Pu has a half-life of 14.35 years β^- decaying into ^{241}Am. It was therefore assumed the separation of Pu was performed five years prior to the experiment where the ^{241}Am concentration was set to zero. This had an effect of -94 pcm on the *k* of the system. Additional scenarios of one year and ten years of age resulted in -20 pcm and -208 pcm respectively.

The ICSBEP evaluators additionally did not analyze the effects of impurities on the system. The experimenters presented the results of a spectrographic analysis of the solution in their report giving concentrations of impurities in the solution. After running simulations with the highest possible level of impurities (Table 2) indicated by the spectrograph, a change in *k* of -71pcm was found.

Table 2: Impurities from Spectrograph [8]

Element	mg/l
Be	5
B	5
Mg	5
Al	40
Ca	40
Ti	20
V	5
Cr	20
Mn	5
Fe	190
Co	5
Ni	10
Cu	30
Ga	5
Ge	5
Mo	5
Cd	5
In	5
Sn	5
Sb	5
Au	5
Tl	5
Pb	5
Bi	5

Bias from impurities: -71pcm

FINAL BENCHMARK k_{eff}

All simulations used MCNP6.2® [6] and ENDF-B/VIII.0 nuclear data library [7]. The total uncertainty in *k* at one sigma was found to be 193pcm.

SUMMARY AND CONCLUSIONS

The uncertainty found from summing parameters independent of one another in quadrature was found to be 194pcm, 6pcm less than the 200pcm uncertainty reported in the handbook. However, as discussed earlier, the handbook evaluation does not state what most of the uncertainty is due to nor the methods used to derive that uncertainty. Some important sources of uncertainty were neglected in the handbook, including ^{241}Am build up and changes in temperature.

The bias found due to impurities is a partial contributor to the remaining 1000pcm disparity between the MCNP6.2® *k* found by LANL NCSD and unity. This information could be used in further evaluations of biases not addressed here such as the extrapolation methods used to ascertain the critical height of the solution.

ACKNOWLEDGEMENTS

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