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Benchmark Evaluation of Plutonium Nitrate Solution Arrays

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BENCHMARK EVALUATION OF PLUTONIUM NITRATE SOLUTION ARRAYS

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ABSTRACT

In October and November of 1981 thirteen approach-to-critical experiments were performed on a remote split table machine in the Critical Mass Laboratory of Pacific Northwest Laboratory in Richland, Washington, using planar arrays of polyethylene bottles filled with plutonium (Pu) nitrate solution. Arrays of up to sixteen bottles were used to measure the critical number of bottles and critical array spacing with a tight fitting Plexiglas® reflector on all sides of the arrays except the top. Some experiments used Plexiglas shells fitted around each bottle to determine the effect of moderation on criticality. Each bottle contained approximately 2.4 L of $Pu(NO_3)_4$ solution with a Pu content of 105 g Pu/L and a free acid molarity, H+, of 5.1. The plutonium was of low ^{240}Pu (2.9 wt.%) content. [1]

Of the thirteen approach-to-critical experiments eleven resulted in extrapolations to critical configurations. Four of the approaches were extrapolated to the critical number of bottles and seven of the approaches were extrapolated to critical array spacing for 3×4 and 4×4 arrays. Detailed and simple models were created and evaluated for these seven critical configurations using MCNP5 and KENO-VI. A thorough uncertainty analysis of the critical array spacings and all geometric and material parameters was performed using parameter perturbation methods. The acceptance of these experiments as criticality safety benchmarks is pending further investigation into the solution composition, the parameter with the largest uncertainty effect. Results of this study will be included in the International Criticality Safety Benchmark Evaluation Project Handbook [2].

Key words: critical experiment, plutonium solution array, benchmark evaluation

1 INTRODUCTION

Thirteen approach-to-critical experiments were sponsored by Rockwell Hanford Operations in October and November of 1981 in order to fill a lack of experimental data on the criticality of arrays of bottles of Pu solution such as might be found in storage and handling at the Purex Facility at Hanford. The results of these experiments were “to provide benchmark data to validate calculational codes used in criticality safety assessments of plant configurations” [1]. In order to fill the data deficit, thirteen experiments were carried out to determine the critical number of bottles and critical array spacing for up to sixteen 3-L-bottles partially filled with $Pu(NO_3)_4$. Tight fitting Plexiglas® reflectors were on all sides of the arrays except the top and some experiments used Plexiglas shells fitted around each bottle to determine the effect of interstitial moderation on criticality. During the thirteen experiments eleven critical configurations were estimated using extrapolations to critical. Four of the configurations determined critical number of bottles and seven determined critical array spacing for 3×4 and 4×4 arrays.

Great effort has been taken to create Monte Carlo models of the critical configurations and evaluate them for use as criticality safety benchmark experiments. This process included the creation of detailed experiment models in MCNP5 using ENDF/B-VII.0 neutron cross section libraries and then simplifying the geometry and calculating the associated bias. An uncertainty analysis was completed for the critical array spacings and all geometry and material parameters. Further investigation into the solution composition is in progress and thus the acceptability of the experiments as benchmarks cannot yet be determined. The complete evaluation of these experiments will be included in the International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook [2].

2 THE EXPERIMENT

The experiments were carried out on a remote split table machine (RSTM) located in one of the large walk-in experimental fume hoods in the reactor room of the critical mass laboratory (CML) [3,4,5]. Sixteen bottles were used in the experiments (see Fig. 1). Table I summarizes data given for the bottles in both the published report and the experimenter's logbook.

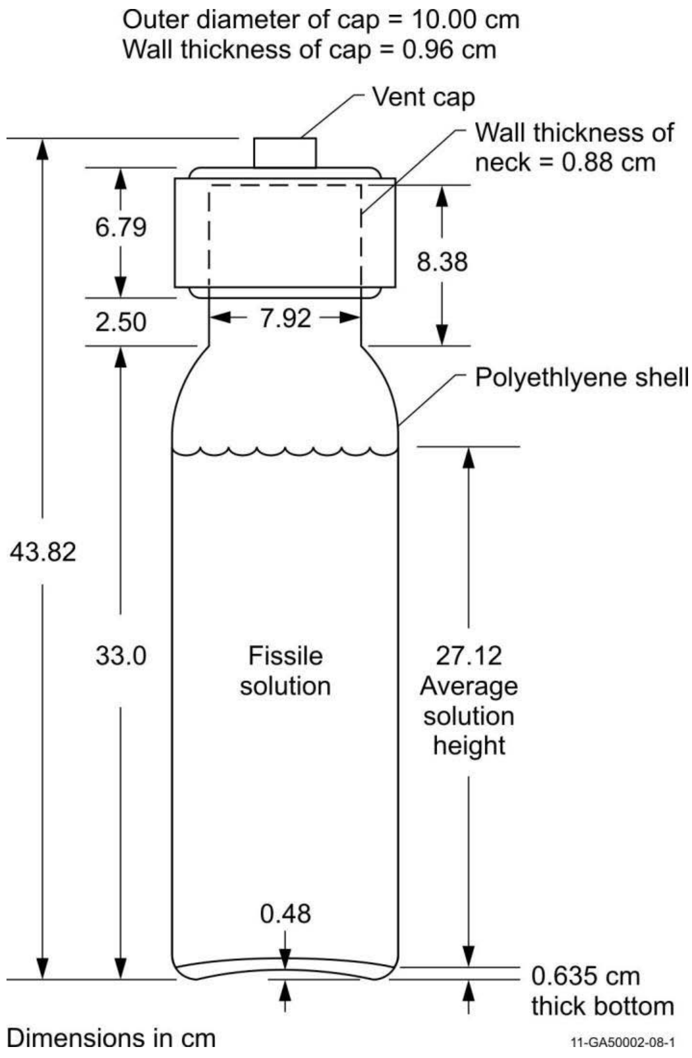

Figure 1. Bottle Dimensions

Table I. Bottle and Solution Measurements

Bottle #	Bottle OD (cm)	Bottle ID (cm)	Empty Bottle Weight (g)	Full Bottle Weight. (g)	Solution Weight (g)	Volume of Solution (L)	Solution Height in Bottle (cm)[(a)]
1	11.740	10.533	1008.9	4418.6	3409.7	2.3995	27.538
2		10.571	1010.8	4419.7	3408.9	2.3989	27.333
3		10.604	1007.7	4468.8	3461.1	2.4357	27.580
4		10.535	1019.2	4443.4	3424.2	2.4097	27.644
6		10.645	1001.6	4431.8	3430.2	2.4139	27.123
7		10.605	1001.6	4435.9	3434.3	2.4168	27.361
8		10.640	970.4	4396.5	3426.1	2.4110	27.116
10		10.681	972.2	4375.0	3402.8	2.3946	26.725[(b)]
11		10.681	972.0	4415.0	3443.0	2.4229	27.041[(c)]
12	11.826	10.678	960.1	4373.7	3413.6	2.4022	26.825
13	11.766	10.643	977.6	4385.2	3407.6	2.3980	26.954
14		10.681	970.3	4385.7	3415.4	2.4035	26.824
15		10.681	978.9	4391.3	3412.4	2.4014	26.801
16		10.678	971.3	4385.5	3414.2	2.4027	26.831
17		10.602	988.5	4400.1	3411.6	2.4008	27.195
18		10.640	980.8	4391.3	3410.5	2.4001	26.993
Average	11.777 ± 0.044	10.631 ± 0.051	987.0 ± 18.4	4407.3 ± 27.2	3420.4 ± 15.4	2.4070 ± 0.0109	27.118 ± 0.298

(a) Solution heights were calculated, not measured, using solution volume and the inside diameter of the bottle.
(b) Value was published as 27.725 but recreation of calculation showed this value to be a typo.
(c) Value was published as 26.041 but recreation of calculation showed this value to be a typo.

Bottles were arranged on a Plexiglas reflector in 3×4 and 4×4 arrays and were surrounded on four sides by a Plexiglas reflector. Aluminum slider plates, dovetails, support rods, and spacers were used to hold bottles in place and maintain array spacing. The size of the bottom reflector was not explicitly given but assumed to match the dimensions of the table on the RSTM. For some experiments Plexiglas shells were placed around the bottles to study the effect of interstitial moderation. The shells contained grooves to allow for the aluminum support rods and had dimensions as shown in Table II. Figure 2 shows the array setup and gives the dimensions of the reflectors and aluminum structure.

Table II. Shell Dimensions

Nominal thickness	Measured OD (cm)	Measured Thickness (cm)
1/8 in.	12.70	0.33
3/8 in.	13.94	0.99
1/4 in.	15.25	0.64
Shell Height (cm)		30.48

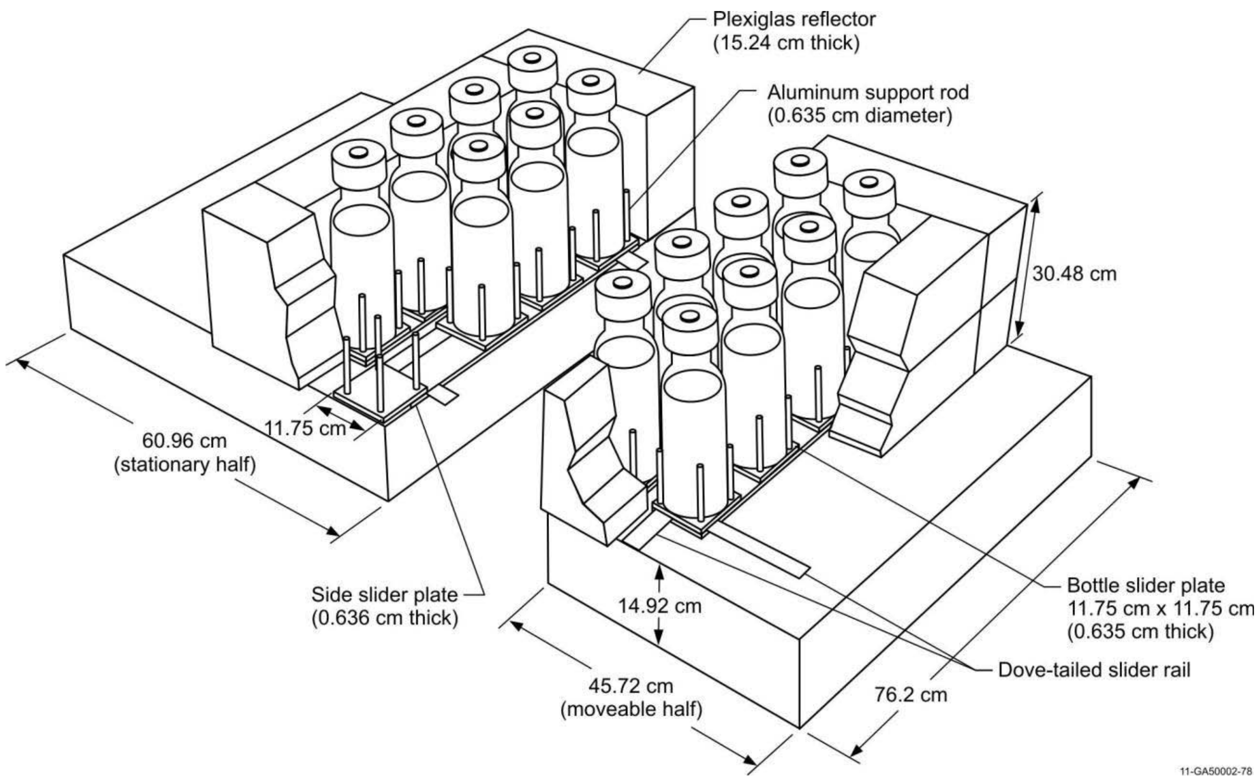

Figure 2: Experimental Setup.

Material properties for the solution, polyethylene bottles and Plexiglas were given and are summarized in Table III. The aluminum was typical aluminum 6061.

Table III. Material Properties

Solution Properties				Plastics Properties	
Component	Concentration (g/L)	Isotope	Wt.%		
Plutonium	105	Pu-238	0.011	Polyethylene	
Uranium	3.1	Pu-239	96.942		Wt %
Nitrate (Total)	505	Pu-240	2.882	H	14.37
Iron	3.2	Pu-241	0.119	C	85.63
Chromium	0.8	Pu-242	0.046	Density (g/cc)	0.98
Nickel	0.6				
Aluminum	8			Plexiglas	
Manganese	0.7				Wt %
Cadmium	0.0005			H	8
Boron	0.005			C	60
Water	788.3[a]			O	32
Am-241 (4/6/83)	0.18			Density (g/cc)	1.185
Acid Molarity (H+) (mol/L)	5.1				
Density	1.420 (g/cc)				

(a) Obtained by difference.

Critical configurations were found by averaging the results of two extrapolation methods. The first plotted inverse count rate versus number of bottles or array spacing. The second method plotted number of bottles or array spacing over count rate versus number of bottles or array spacing. The second method was believed to give a more accurate critical value earlier in the approach to critical [6]. The following critical configurations were found. Results of extrapolations to critical are boldface and highlighted.

Table IV. Experimental Results[(a)]

Experiment Number		Critical Number of Bottles[(a)]	*x* S-S Spacing [(c)] (cm)	*y* S-S-Spacing [(c)] (cm)	Shell Thickness[(b)] (cm)
RSTM-L3-	1	**>2x5**	0	0	-
	2	**10.89**	0	0	-
	3	12	0	**0.492**	-
	4	12	0.330	**0.361**	-
	5	12	**0.585**	0	-
	6	16	0	**1.923**	-
	7	16	1.257	**0.709**	-
	08	**>25**	0	0	0.64
	09	**16.89**	0	0	0.99
	10[(d)]	**13.1**	0	0	0.33
	11[(e)]	**13.1**	0	0	0.33
	12	16	0	**1.240**	0.33
	13	16	0.686	**1.204**	-

(a) Data is compiled from logbooks and Reference 1. Data from the logbook was considered more accurate and precise and was preferred to data from Reference 1

(b) The aluminum support rods or grooves cut into plastics shells had no appreciable effect on the extrapolated values shown (see footnote e).

(c) Spacings were surface-to-surface (S-S) spacings. For RSTM-L3-8 through 12 this was shell S-S and bottle S-S for the rest.

(d) Average of two experiments, one where the 13th bottle is placed in the center of the array and one where the 13th bottle is on the array edge.

(e) Repeat of Experiment 10 with twice as many grooves in shells to establish the worth of the reflector displaced by support rods.

3 THE MODEL

In order to study the effects of uncertainty on the experiment system a simple model of the setup was first created. For the simple model to be useful the bias of all simplifications also had to be determined. Simplifications included: removal of the surrounding room, RSTM structure, and fume hood; simplifications of the bottle and averaging of bottle dimensions; simplification of aluminum support structure; and removal of impurities and air from model. Since geometry could only be defined for a discrete number of bottles only RSTM-L3-3, 4, 5, 6, 7, 12, and 13 could be modeled and studied. Figures 3, 4 and 5 show the bottle specifications and array setup for the simple models.

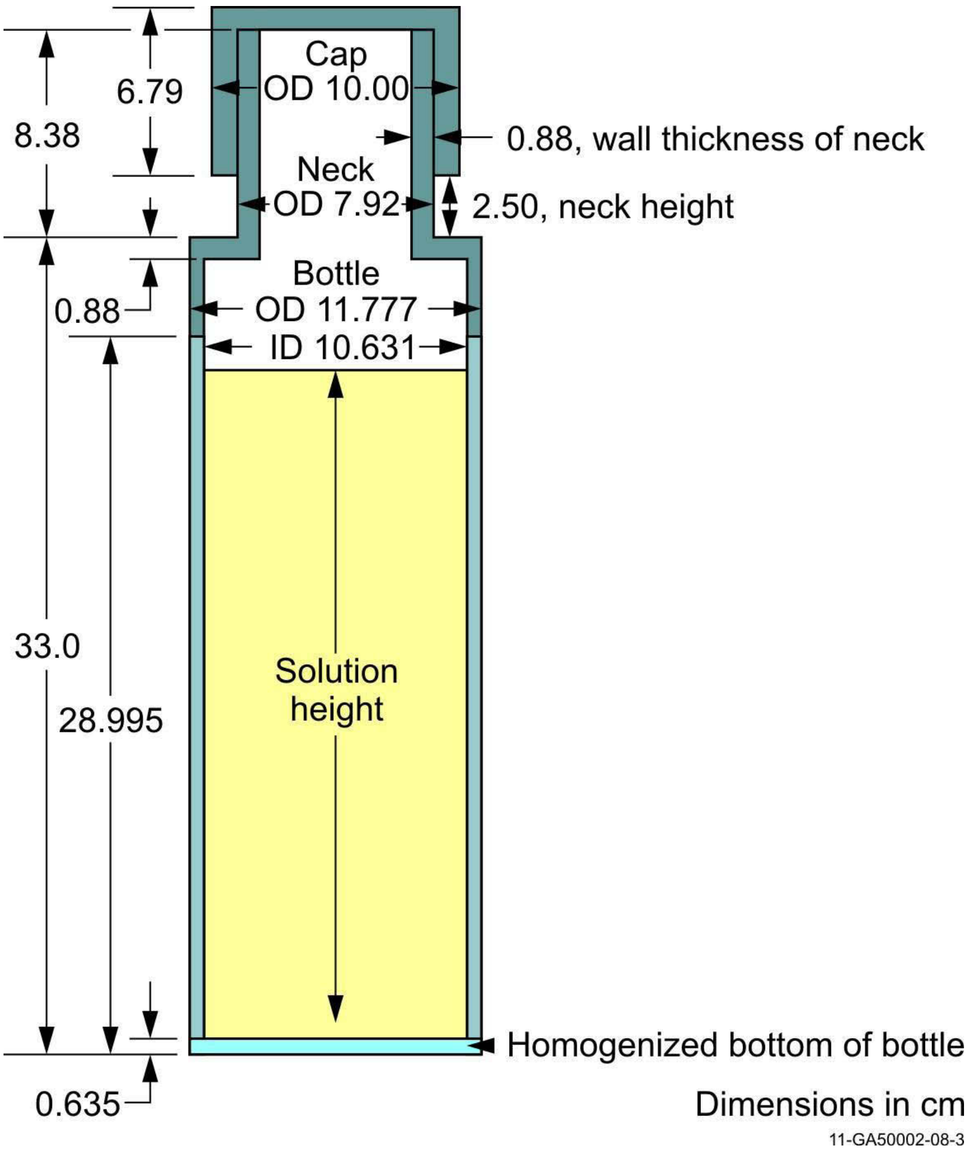

Figure 3: Bottle Specifications for Simple Model

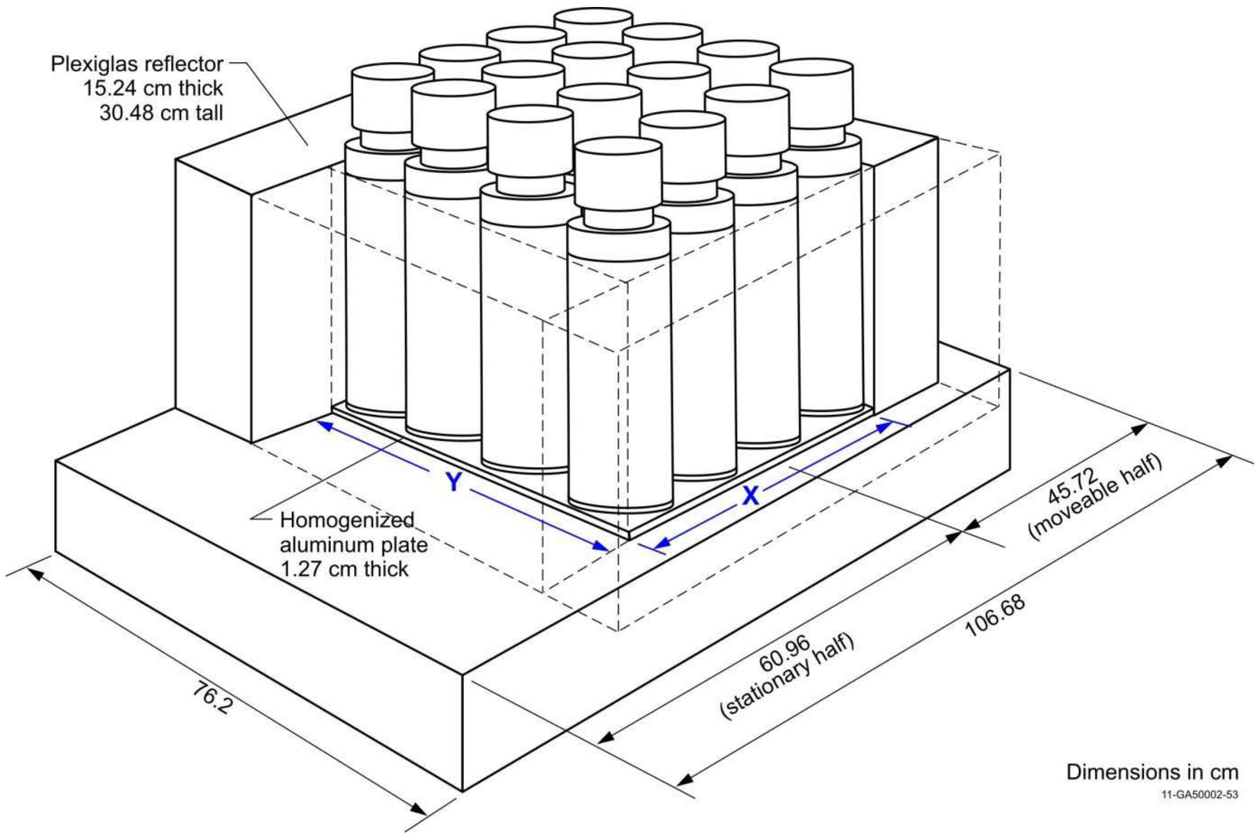

Figure 4: Array Setup for Simple Model[1]

The average bias of these simplifications for all seven modeled experiments was -0.00219 Δk_{eff}. A more thorough discussion of the derivations of the detailed and simple models is available in Reference 2.

4 UNCERTAINTY ANALYSIS

Significant effort was put forth to characterize the uncertainty in all parameters of the experiment and the effect on k_{eff}. Table V contains the Δk_{eff} for grouped uncertainties with discussion of the major contributors following. For uncertainties and their effects for each individual parameter see Reference 2.

[1] This figure shows a 4×4 array as was present in RSTM-L3-6, 7, and 13. RSTM-L3-3, 4, and 5 had only a 3×4 array. RSTM-L3-12 had a 4×4 array but also had Plexiglas shells around each bottle.

Table V. Summary of Uncertainties

	RSTM-L3-						
Source of Uncertainty	3	4	5	6	7	12	13
Critical Array Spacing Uncertainties	0.00221	0.00456	0.00066	0.00131	0.00771	0.00059	0.00867
Bottle Dimension Uncertainties	0.00077	0.00094	0.00077	0.00094	0.00184	0.00122	0.00093
Solution Measurements Uncertainties	0.00115	0.00110	0.00110	0.00102	0.00100	0.00094	0.00101
Reflector Measurements Uncertainties	0.00047	0.00046	0.00046	0.00039	0.00037	0.00034	0.00038
Shell Measurements Uncertainties	-[(a)]	-[(a)]	-[(a)]	-[(a)]	-[(a)]	0.00089	-[(a)]
Solution Composition Uncertainties Except Pu and H+	0.00151	0.00137	0.00138	0.00138	0.00141	0.00130	0.00138
Uncertainty in Pu and H+ in Nitrate Solution	0.00949	0.00964	0.00966	0.01023	0.01032	0.01327	0.01028
Solution Temperature Uncertainties	0.00023	0.00011	NEG	NEG	NEG	NEG	NEG
Polyethylene Uncertainties	0.00462	0.00450	0.00456	0.00424	0.00430	0.00401	0.00449
Plexiglas Uncertainties	0.00026	0.00023	0.00026	0.00024	0.00027	0.00042	0.00018
Support Structure Uncertainties	0.00012	0.00013	0.00013	0.00017	0.00016	0.00012	0.00013
Total	0.01099	0.01176	0.01089	0.01133	0.01382	0.01406	0.01432

(a) No shells were used for this configuration.

The largest contributors to the uncertainty were the critical array spacing and the solution properties. The critical array spacing uncertainty was a function of random variation in the bottle surfaces (0.0254 cm), uncertainty in the array spacers (0.00254 cm), and uncertainty in the extrapolation which was assumed to be the difference between the two extrapolation methods [6].

Two entries in Table V apply to uncertainty in the solution composition. The first includes half the last significant digit for all elements in Table III except Pu and free acid molality (H+) as well as possible Ca, Co, Ti, and V impurities based on information in the logbooks. The second entry is due to the disagreement between the reported solution density and the solution density predicted using Sakurai's equation [7]. A solution density of 1.421 g/cc was reported for the solution. Sakurai's equation gives a density of 1.321 g/cc based on the the Pu, U and H+ concentration given in Table III and a temperature of 22 ^{o}C. Because the solution density was measured and also calculated using solution mass and volume it is believed that 1.421 g/cc is the correct solution density therefore at least one of the solution parameters, Pu, U, H+ and/or temperature, which are used in Sakurai's equation must be incorrect. Because temperature must be varied by an unrealistic amount it is not believed to be the cause of the discrepancy. Likewise, the uranium content is also believed to not be the cause of the discrepancy because the plutonium feedstock was known to have very low uranium concentrations. Thus either the Pu and/or the H+ must be incorrect. To find the effect of the uncertainty in these parameters each one was varied independently until Sakurai equation's density agreed with the measured density. The two parameters are correlated and the uncertainties were combined to get the uncertainty shown in Table V. This uncertainty is the largest contribution to the total uncertainty. Further effort is being made to verify the solution composition.

5 SAMPLE CALCULATION RESULTS

Models were created with MCNP5 using ENDF/B-V.0 and ENDF/B-VII.0 neutron cross sections and KENO-VI using ENDF/B-VII.0. Calculation results for the simple model as well as expected k_{eff} values are given in Table VI. It is not known why the calculated results are so much higher than the expected values.

Table VI. Sample Calculation Results.

RSTM-L3-	Expected k_{eff} [(a)]	MCNP5 [(b)] ENDF/B-V.0	MCNP5 [(b)] (ENDF/B-VII.0)	KENO-VI[(c)] (continuous energy ENDF/B-VII.0)[(d)]
3	0.9978 ± 0.0110	1.03856 ± 0.00004	1.02954 ± 0.00005	1.02758 ± 0.00026
4	0.9979 ± 0.0118	1.03278 ± 0.00004	1.02387 ± 0.00005	1.02185 ± 0.00023
5	0.9980 ± 0.0109	1.03815 ± 0.00005	1.02916 ± 0.00005	1.02746 ± 0.00022
6	0.9978 ± 0.0113	1.03696 ± 0.00005	1.02839 ± 0.00005	1.02636 ± 0.00026
7	0.9975 ± 0.0138	1.03338 ± 0.00004	1.02478 ± 0.00005	1.02269 ± 0.00020
12	0.9979 ± 0.0141	1.02364 ± 0.00004	1.01490 ± 0.00004	1.01120 ± 0.00024
13	0.9977 ± 0.0143	1.03586 ± 0.00005	1.02731 ± 0.00005	1.02513 ± 0.00029

(a) Found using expected k_{eff} of 1.0000 for detailed model and bias for model simplifications. Uncertainties from uncertainty analysis described in Section 4.
(b) Run using 4000 cycles, skipping the first 100 cycles, with 100,000 histories or particles per cycle.
(c) Results provided by John D. Bess of Idaho National Laboratory.
(d) Run using 155 cycles, skipping the first 5 cycles, with 100,000 histories or particles per cycle.

6 CONCLUSIONS

These experiments have not yet been determined as being acceptable or unacceptable benchmark experiments. The disagreement between the reported solution density and the solution density calculated using Sakurai's equation has led to a large uncertainty in the Pu and H+ content of the solution. Attempts are being made to try to verify the correct solution composition. The acceptability of these experiments as benchmarks will be determined following the conclusion of that search for information. The final results of this evaluation will be published in the ICSBEP Handbook.

7 REFERENCE

1. B. M. Durst, E. D. Clayton, and J. H. Smith, "Criticality Experiments with Planar Arrays of Three-Liter Bottles Containing Plutonium Nitrate Solution," PNNL-5387, Pacific Northwest National Laboratory (January 1985).
2. M. A. Marshall, "Arrays of Bottles of Plutonium Nitrate Solution," PU-SOL-THERM-037, *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/Doc(95)03 (September 2012).
3. R.W. Bartholomay, "Lucite-Moderated and Unmoderated, Reflected and Non-Reflected Arrays of Bottles Containing Uranyl Nitrate (98.2wt.% ^{233}U) Solution," U233-SOL-THERM-014, *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/Doc(95)03 (September 2002).
4. W. A. Reardon, E. D. Clayton, C. L. Brown, R. H. Masterson, T. J. Powell, C. R. Richey, R. B. Smith, and J. W. Healy, "Hazards Summary Report For The Hanford Plutonium Critical Mass Laboratory," HW-66266, (August 1960).
5. C. R. Richey, E. D. Clayton, R. H. Odegaarden, J. D. White, and W. A. Reardon, "Hazards Summary Report For The Hanford Plutonium Critical Mass Laboratory, Supplement No. 1, The Remote Split-Table Machine," HW-6266 SUP1 REV, (October 1963).
6. R.C. Lloyd, E.D. Clayton, and J.H. Chalmers, "Criticality of Array of ^{233}U Solution,"*Nucl. Appl.,* **4,** pp. 136-141, (1968).
7. S. Sakurai and S. Tachimori, "Density Equation of Aqueous Solution Containing Plutonium (IV), Uranium (VI), and Nitric Acid," *Journal of Nuclear Science and Technology*, **Vol. 33, No. 2**, pp. 187-189 (February 1996).