INL/CON-05-01002
PREPRINT

Analysis of High Enriched Uranyl Nitrate Solution Containing Cadmium

PHYSOR-2006 Topical Meeting

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September 2006

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Battelle Energy Alliance

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Abstract

A benchmark evaluation has been performed for a set of twenty-one critical experiments involving high enriched uranyl nitrate solution with and without cadmium nitrate as a soluble neutron absorber. The critical experiments analyzed include two types of cylindrical vessels with 24.18 and 29.16 cm in diameters. The vessels were reflected with water and in some cases with water containing dissolved cadmium nitrate. The uranium concentration ranged from 482 to 529 g/l, and cadmium concentration in the uranyl nitrate solution ranged from 0.0 to 11.31 g/l. The cadmium concentration in the reflector solution ranged from 0.0 to 15.16 g/l.

Using MCNP and KENO-V.a, complete three-dimensional models were created for the two vessels filled with the uranyl nitrate solution and reflector solution. A series of criticality calculations was performed with KENO-V.a, MCNP4b (with ENDF/B-V data), and MCNP5 (with ENDF/B-VI data). In general, good agreement between KENO-V.a and MCNP4b was observed. However, MCNP5 results show consistently lower values compared with MCNP4b results with the maximum difference of 1.2 %. This ICSBEP supported evaluation provides valuable data for the effect of soluble neutron absorber (cadmium nitrate) on the criticality safety of high-enriched uranyl nitrate solution. These data can also be used in determining critical controls and for validation of the calculation methods.

KEYWORDS: Criticality Safety, High-Enriched Uranium, Uranyl Nitrate Solution, Cadmium Nitrate

1. Introduction

Twenty-one critical experiments involving high-enriched (~85 wt% ^{235}U) uranyl nitrate solution containing soluble cadmium nitrate absorber were performed [1] in 1978 and 1979 at the Pacific Northwest Laboratory. The experiments were conducted to validate criticality safety calculations by Allied Chemical Corporation (later known as Westinghouse Idaho Nuclear Company) in the design of proposed fluorinel process dissolver. The critical experiments consisted of a cylindrical experimental vessel filled with high-enriched uranyl nitrate solution with and without cadmium nitrate as a soluble neutron absorber. Water with and without cadmium nitrate was used as the reflector. Evaluation of this set of experiments was accepted by the International Criticality Safety Benchmark Evaluation Project (ICSBEP), as a benchmark [2].

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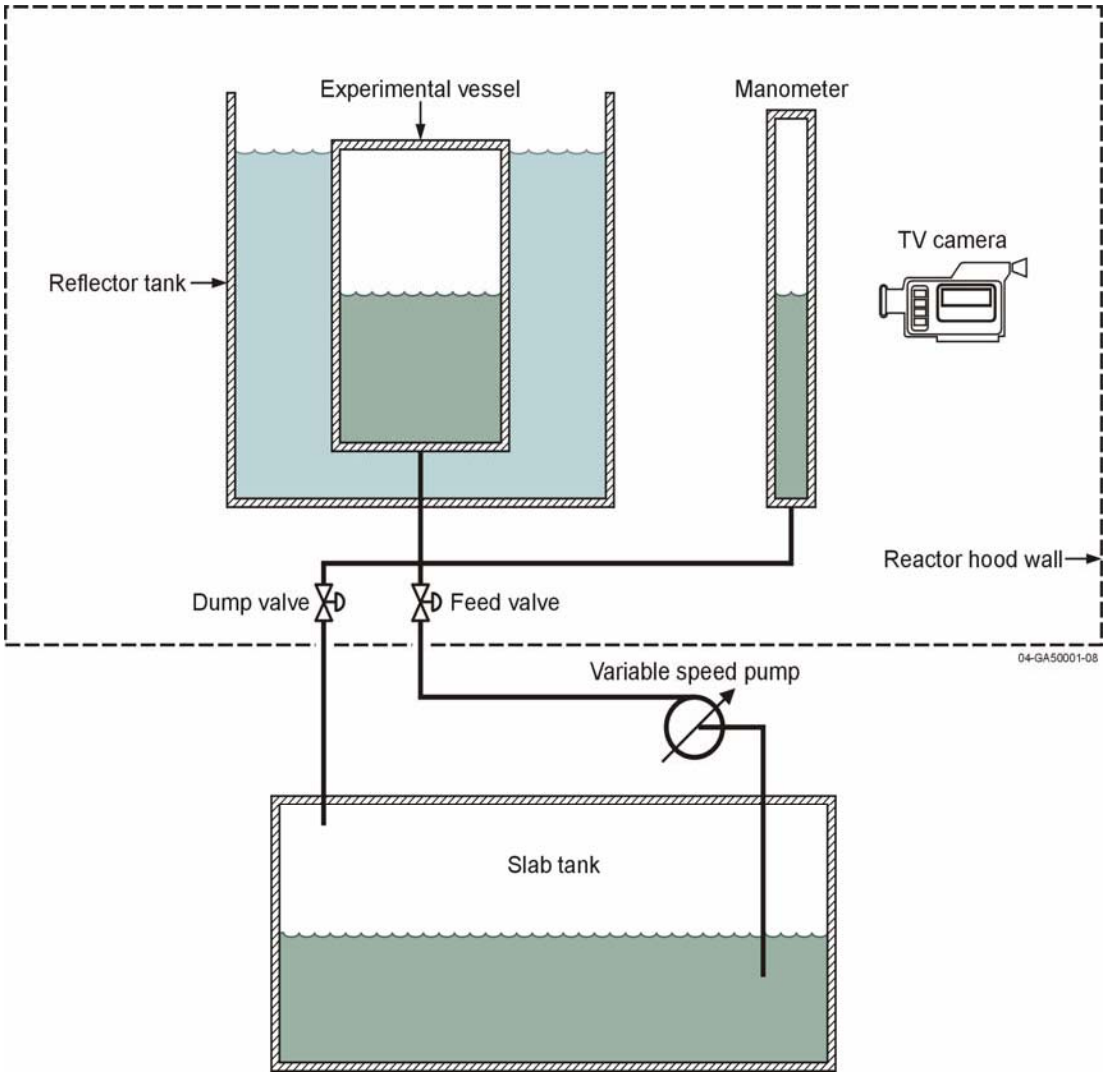
2. Critical Experiments

The critical experiments were conducted inside the Critical Mass Laboratory at the Pacific Northwest Laboratory. A simplified diagram of the experiment is shown in Fig. 1.

Figure 1: A simplified diagram of experiments.



The experiments were performed using two cylindrical vessels. Experiments 001 through 008 were performed using a 24.18-cm-ID vessel (see Fig. 2). Experiments 009 to 021 were performed using a 29.16-cm-ID vessel (Fig. 3).

Figure 2: Experimental diagram with 24.18-cm ID vessel.

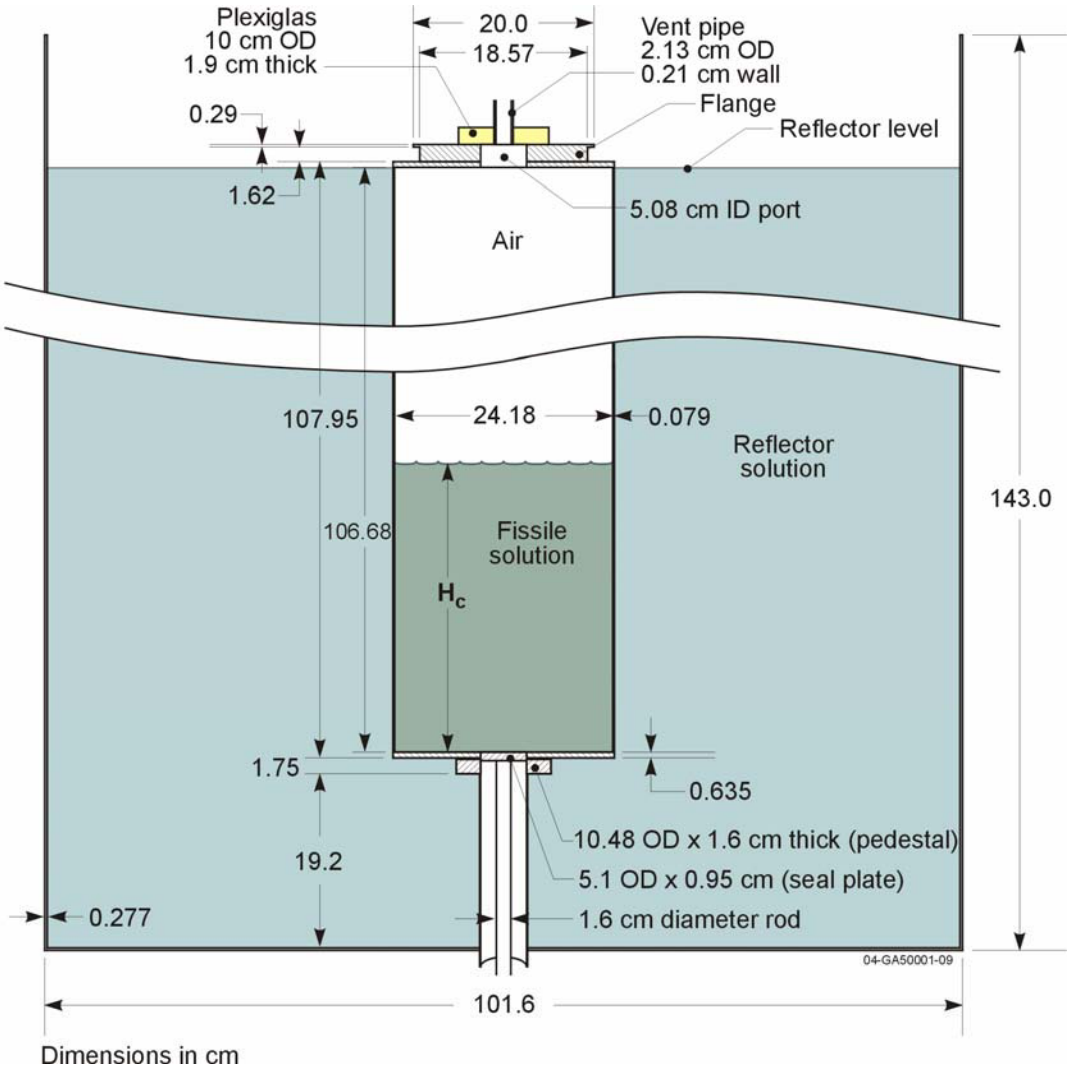
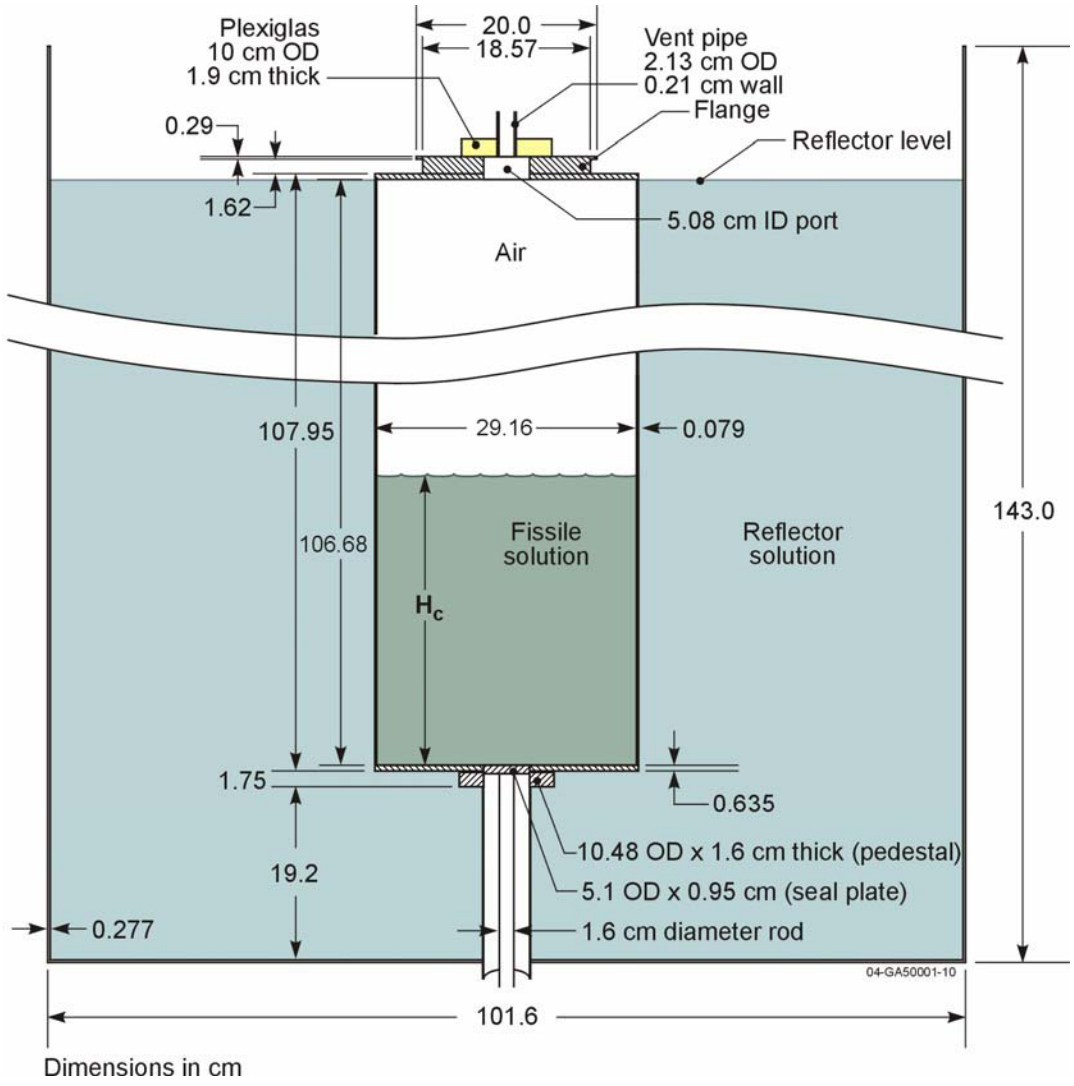


Figure 3: Experimental diagram with 29.16-cm ID vessel.



The solution was pumped from a slab storage tank to the experimental vessel. Cadmium nitrate was added in measured amounts to the uranyl nitrate in the slab tank, and the solution was thoroughly mixed prior to use in each of the experiments. Neutron absorbing safety and control blades or rods (not shown in the figures) were provided. In the case of the 24.18-cm-ID vessel, curved safety and control blades (304L stainless steel clad cadmium sheet) were positioned in the reflector region at the interface of the vessel wall. For the larger (29.16-cm-ID) vessel, Zircaloy clad cadmium-Lucite rods were used for control and safety rods. These rods were placed directly in the uranyl nitrate solution for increased effectiveness [1].

With the control rod fully inserted, uranyl nitrate solution was pumped to the experimental vessel to a desired height, then the control rod was withdrawn until it no longer influenced the reactivity of the experiment. The process of adding solution to the cylinder, then withdrawing

the control rod was repeated until the critical height was determined. The neutron count rate from three neutron detectors was recorded following each solution addition. As the solution level was raised, the inverse count rate as a function of solution height was plotted. The inverse count rate plot was then extrapolated to determine the critical height. The size of the extrapolation was less than a half-inch, which corresponds to less than 0.028 in reactivity. The system was secured by inserting both rods and draining the solution to the slab tank.

Table 1 summarizes the experimental data from the twenty-one critical configurations. The uranium concentration ranged from 481.99 to 528.76 g/l of uranium as uranyl nitrate. The cadmium concentration in the uranyl nitrate solution ranged from 0.0 to 11.33 g/l. The cadmium concentration in the reflector solution ranged from 0.0 to 15.18 g/l. Experiment 018 was the same as Experiment 017, and therefore, not considered as separate experiment. For all of the experiments, the reflector solution was maintained at the lower level of the upper end cap of the experimental vessel. The uranium isotope composition of the solution is shown in Table 2. The uranyl nitrate solution used in the experiments came from six reserved product canisters. The enrichments of the six canisters ranged from 83.90 to 85.96 wt.% U-235.

Table 1: Composition of twenty-one critical configurations.

Expr. Date	Expr. No.	Uranyl Nitrate Solution					Reflector Solution			Solution Height Critical Height (cm)
		Solution Sample No.	Specific Gravity	Excess Nitric Acid	Uranium (mg/g)	Cadmium (mg/g)	Solution Sample No.	Specific Gravity	Cadmium (mg/g)	
24.18-cm-ID Vessel										
9/26/78	001	830	1.65919	0.22773	293.845	0.0	-	1.0	0.0	22.3520
9/27/78	002	831	1.66006	0.21369	294.175	0.0	854	1.02257	14.848	31.2674
9/28/78	003	832	1.66092	0.23976	292.98	1.208	-	1.0	0.0	28.1940
9/29/78	004	833	1.66092	0.23374	291.34	2.393	-	1.0	0.0	37.8460
9/29/78	005	834	1.66284	0.30312	290.38	3.897	-	1.0	0.0	76.3524
1/29/79	006	837	1.72322	0.23575	306.435	4.069	-	1.0	0.0	82.2198
1/30/79	007	838	1.72532	0.23374	306.79	4.196	-	1.0	0.0	92.4814
2/2/79	008	839	1.72606	0.32660	306.935	4.279	-	1.0	0.0	98.6282
		839[(a)]	1.72597	0.23374	306.86	4.262				
29.16-cm-ID Vessel										
2/16/79	009	842	1.68007	0.30393	298.38	0.0	-	1.0	0.0	17.8308
2/22/79	010	843	1.68041	0.30192	298.76	0.0	855	1.0151	10.596	21.4884
							855[(a)]	-	10.600	
2/23/79	011	844	1.69471	0.27385	301.175	1.240	855	1.0151	10.598	26.5938
2/26/79	012	845	1.70589	0.26583	303.38	2.250	855	1.0151	10.598	32.7406
2/27/79	013	846	1.70729	0.25781	302.03	3.362	855	1.0151	10.598	42.9006
2/28/79	014	847	1.70781	0.25380	301.84	4.189	855	1.0151	10.598	57.9374
3/1/79	015	848	1.70869	0.24778	300.945	4.577	855	1.0151	10.598	71.6280
3/2/79	016	849	1.70851	0.24979	300.775	4.897	855	1.0151	10.598	87.0204
3/5/79	017	850	1.70921	0.25380	301.065	5.049	856	1.01175	9.519	98.1456
3/7/79	018	850[(a)]	1.70848	0.35621	301.095	5.044	856[(a)]	-	9.511	98.0948
3/12/79	019	851	1.71041	0.35621	301.555	5.032	-	1.0	0.0	46.4312
3/13/79	020	852	1.70956	0.26583	300.33	5.937	-	1.0	0.0	64.2874
3/13/79	021	853	1.71021	0.36744	302.06	6.626	-	1.0	0.0	102.362

[(a)] Second Analysis of the Sample.

Table 2: Uranium isotopic composition (wt%).

Expr. No.	Sample No.	^{234}U	^{235}U	^{236}U	^{238}U
24.18–cm-ID Vessel					
001	830	0.94	85.27	3.47	10.32
002	831	0.94	85.27	3.47	10.32
003	832	0.94	85.24	3.48	10.34
004	833	0.93	85.28	3.47	10.32
005	834	0.94	85.26	3.47	10.33
006	837	0.94	85.21	3.47	10.38
007	838	0.94	85.16	3.47	10.43
008	839	0.93	85.19	3.47	10.41
008	839(b)	0.94	85.17	3.47	10.42
29.16-cm-ID Vessel					
009	842	0.85	84.29	1.40	13.46
010	843	0.86	84.28	1.39	13.47
011	844	0.87	84.55	2.00	12.58
012	845	0.89	84.76	2.49	11.86
013	846	0.89	84.74	2.49	11.88
014	847	0.89	84.74	2.49	11.88
015	848	0.90	84.77	2.49	11.84
016	849	0.89	84.72	2.50	11.89
017	850	0.89	84.77	2.50	11.84
018	850[(a)]	0.90	84.75	2.49	11.86
019	851	0.90	84.78	2.49	11.83
020	852	0.90	84.76	2.49	11.85
021	853	0.90	84.72	2.49	11.89

[(a)] Second analysis of the sample.

3. Experiment Modeling

Using MCNP [3, 4] and KENO-V.a from SCALE [5], a complete model of the experimental system was created for the 24.18-cm-ID vessel (see Fig. 4). A similar model was created for the 29.16-cm-ID vessel (see Fig. 5). The top flange, Plexiglas cap, and seal plate were explicitly modeled. Twenty individual critical solution heights with different solution densities were modeled. For all of the experiments, the top of the reflector solution level was positioned at 107.315 cm from the bottom of the experimental vessel.

Figure 4: XZ view showing the 24.18-cm-ID experimental vessel.

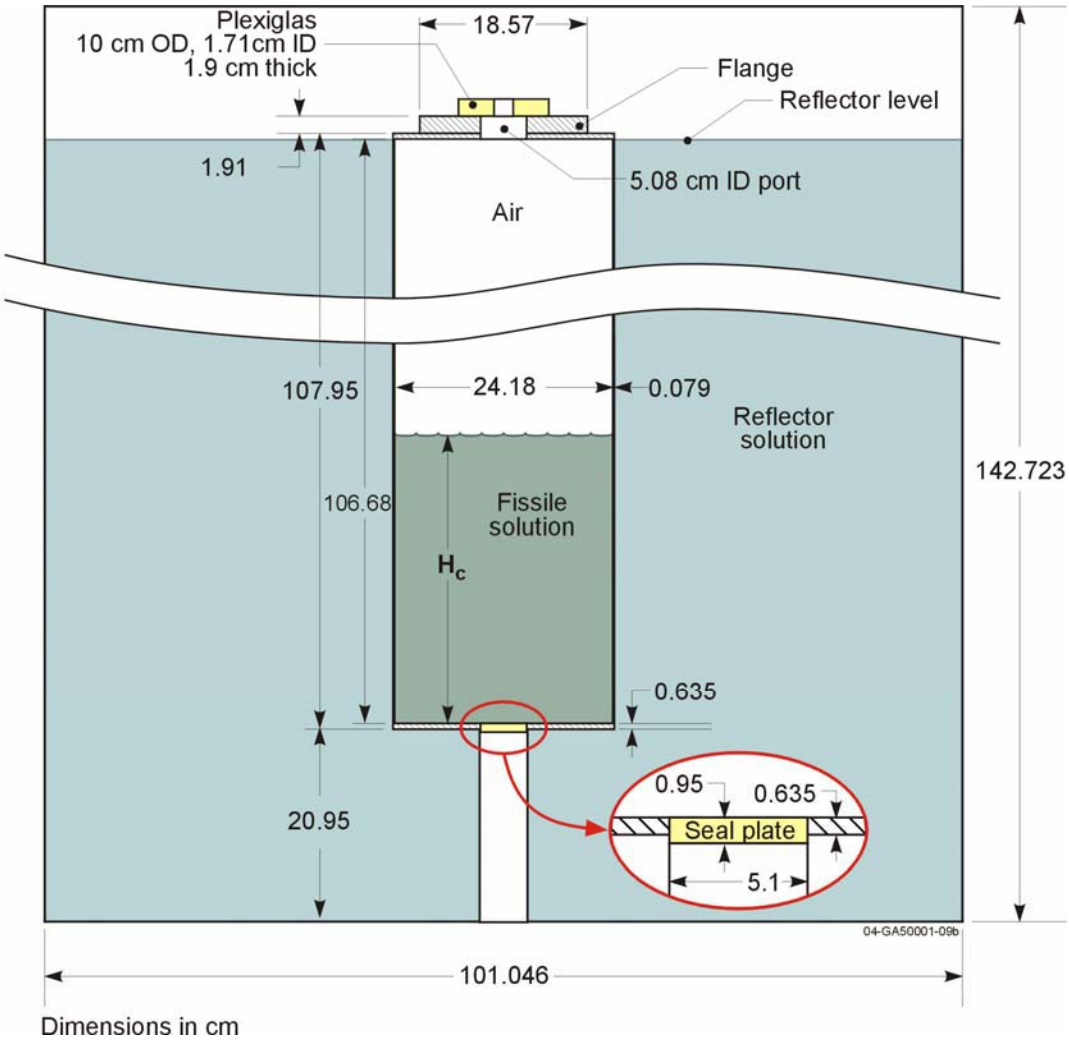
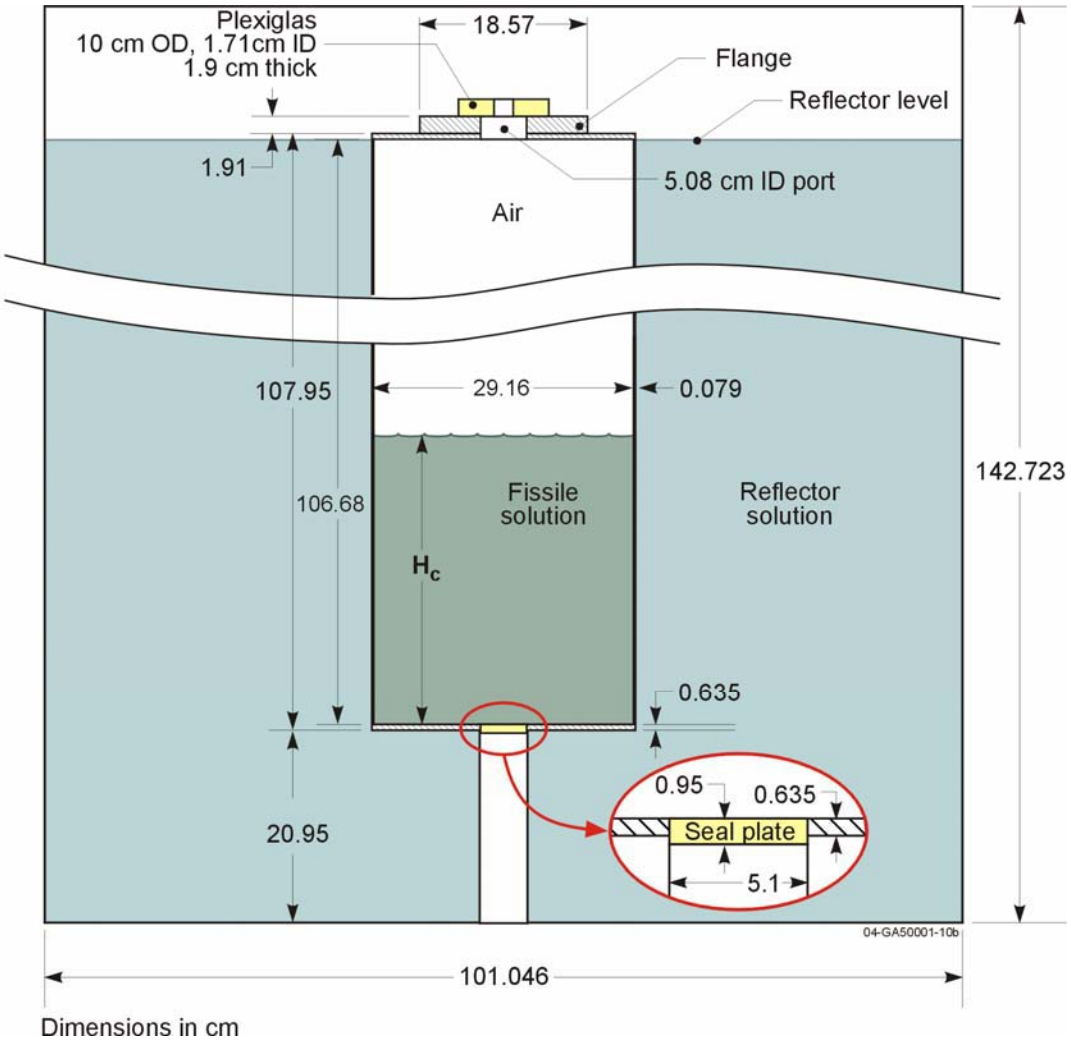


Figure 5: XZ view showing the 29.16-cm-ID experimental vessel.



4. Results

A series of criticality calculations was performed for the 20 experiments with KENO-V.a, MCNP4b, and MCNP5. The 44-group ENDF/B-V cross-section data used in the KENO-V.a runs were based on 20°C. The continuous-energy ENDF/B-V cross-section data of the MCNP4b, and the ENDF/B-VI cross-section data of MCNP5 runs were based on 27°C. Table 3 summarizes these results, showing good agreement between KENO and MCNP4b. MCNP5 results show consistently lower values compared with MCNP4b results, with the maximum difference of 1.2 %.

Table 3: KENO-V.a, MCNP4b and MCNP5 results.

Expr No.	Critical Solution Height (cm)	Uranyl Nitrate Solution			Cadmium in Reflector (mg/g)	KENO-V.a (44-Group ENDF/B-V)	MCNP4b (Continuous Energy ENDF/B-V)	MCNP5 (Continuous Energy ENDF/B-VI)
		H/ ^{235}U	Cd/^{235}U	Cadmium (mg/g)				
01	22.3520	52.5	0.0	0.0	0.0	1.0043 ± 0.0004	1.0023 ± 0.0004	0.9972 ± 0.0004
02	31.2674	52.4	0.0	0.0	14.848	0.9972 ± 0.0004	1.0008 ± 0.0004	0.9948 ± 0.0004
03	28.1940	52.5	0.0101	1.208	0.0	1.0025 ± 0.0004	1.0005 ± 0.0004	0.9928 ± 0.0004
04	37.8460	52.8	0.0201	2.393	0.0	1.0044 ± 0.0005	1.0003 ± 0.0004	0.9917 ± 0.0004
05	76.3524	52.6	0.0329	3.897	0.0	1.0059 ± 0.0004	1.0024 ± 0.0004	0.9922 ± 0.0004
06	82.2198	47.4	0.0326	4.069	0.0	1.0101 ± 0.0004	1.0066 ± 0.0004	0.9956 ± 0.0004
07	92.4814	47.3	0.0336	4.196	0.0	1.0097 ± 0.0005	1.0064 ± 0.0004	0.9958 ± 0.0004
08	98.6282	47.1	0.0342	4.2705	0.0	1.0089 ± 0.0003	1.0054 ± 0.0004	0.9943 ± 0.0004
09	17.8308	51.3	0.0	0.0	0.0	1.0027 ± 0.0004	1.0006 ± 0.0004	0.9960 ± 0.0004
10	21.4884	51.2	0.0	0.0	10.598	0.9949 ± 0.0004	0.9942 ± 0.0004	0.9881 ± 0.0004
11	26.5938	50.0	0.0102	1.24	10.598	0.9999 ± 0.0004	0.9974 ± 0.0004	0.9893 ± 0.0004
12	32.7406	49.0	0.0183	2.25	10.598	1.0022 ± 0.0004	0.9997 ± 0.0004	0.9896 ± 0.0004
13	42.9006	49.2	0.0275	3.362	10.598	1.0032 ± 0.0003	1.0006 ± 0.0004	0.9894 ± 0.0004
14	57.9374	49.1	0.0342	4.189	10.598	1.0041 ± 0.0003	1.0007 ± 0.0004	0.9890 ± 0.0004
15	71.6280	49.3	0.0375	4.577	10.598	1.0054 ± 0.0003	1.0022 ± 0.0004	0.9908 ± 0.0004
16	87.0204	49.3	0.0402	4.897	10.598	1.0046 ± 0.0003	1.0014 ± 0.0004	0.9894 ± 0.0004
17	98.1202	49.0	0.0413	5.0465	9.515	1.0037 ± 0.0003	1.0011 ± 0.0004	0.9883 ± 0.0004
19	46.4312	48.7	0.0412	5.032	0.0	1.0048 ± 0.0005	1.0009 ± 0.0004	0.9895 ± 0.0004
20	64.2874	49.2	0.0488	5.937	0.0	1.0045 ± 0.0003	1.0018 ± 0.0004	0.9905 ± 0.0004
21	102.362	48.2	0.0541	6.626	0.0	1.0034 ± 0.0004	0.9994 ± 0.0004	0.9876 ± 0.0004

KENO-V.a calculations were also performed for the 24.18-cm-ID and the 29.16-cm-ID vessels to calculate the worth of cadmium in the uranyl nitrate solution. Table 4 summarizes results for Experiments 03, 08, 11, and 21.

Table 4: KENO-V.a cadmium worth results.

Experiment No.	Cd (mg/cm^3)	With Cd	Without Cd	%Δk_{eff}
03	2.003	1.0025 ± 0.0004	1.0633 ± 0.0006	6.08
08	7.358	1.0089 ± 0.0003	1.2024 ± 0.0006	19.35
11	2.098	0.9999 ± 0.0004	1.0604 ± 0.0004	6.05
21	11.312	1.0034 ± 0.0003	1.2968 ± 0.0004	29.34

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This ICSBEP supported evaluation provides valuable data for the effect of soluble neutron absorber (cadmium nitrate) on the criticality of high-enriched uranyl nitrate solution. These data can also be used in determining criticality controls and for validation of the calculation methods.

Acknowledgements

The author would like to thank Linda Sandoval of Los Alamos National Laboratory for help in retrieving experimental logbook. He is grateful to Blair Briggs and Todd Taylor of Idaho National Laboratory for providing notes and letter files that contain valuable information about analysis of the experiments. Special acknowledgment is given to B. M. Durst who provided answers for some key questions about the experiments.

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