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**OAK RIDGE
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**Critical Experiments With
Mixed Plutonium-Uranium
Nitrate Solutions Having Pu:(Pu + U)
Ratios Greater than 0.5**

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MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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Consolidated Fuel Reprocessing Program

**CRITICAL EXPERIMENTS WITH MIXED PLUTONIUM-URANIUM NITRATE
SOLUTIONS HAVING Pu:(Pu + U) RATIOS GREATER THAN 0.5**

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Contents

ABSTRACT	v
1. INTRODUCTION	1
2. EXPERIMENTAL APPARATUS	3
2.1 Cylindrical Vessels	3
2.1.1 Reflector Tank	4
2.1.2 Small Cylindrical Vessel	4
2.1.2.1 Reflectors for the small cylinder	4
2.1.3 Large Cylindrical Vessel	8
2.1.4 CFRP Cylindrical Assembly	11
2.2 Slab Tank	11
2.3 Location of Experimental Vessels in the Critical Assembly	
Room of the Critical Mass Laboratory	21
References for Section 2	22
3. EXPERIMENTAL MEASUREMENTS	23
4. CALCULATED κ -EFFECTIVES FOR CRITICAL EXPERIMENTS	27
4.1 Codes and Library	27
4.2 Computational Models	27
4.2.1 Cylinders	27
4.2.2 Slab	27
4.3 Calculated Values of κ -Effective	28
4.4 Tests for Trends and Biases	29
4.5 Tolerance Limits for Calculated κ -Effectives	30
References for Section 4	31
5. DERIVED CRITICALITY SAFETY LIMITS	33
5.1 Supplemental Investigations of the Effect on	
κ -Effective of Uncertainties in Experiments	33
References Section 5	36
6. CONCLUSIONS	37

Appendix A	39
Composition and Dimensions of Steel Reflectors	39
A.1. Physical Measurements of Steel Reflectors	39
A.2. Calculation of Atom Densities for Steel Reflectors	47
Appendix B	53
Composition of Concrete Reflector*	53
Appendix C	57
Analysis of Mixed Plutonium-Uranium Nitrate Solution Standards and Derived Critical Experiment Solution Parameters	57
C.1 Introduction	57
C.2 Standards Data	57
C.3 Statistical Analyses of Standards Data	58
C.4 Derivation of Corrections to the Reported Analytical Chemistry Data for Critical Experiment Solutions	62
C.4.1. Adjustments to the Reported Uranium Concentration and Specific Gravity	62
C.4.2. Adjustments to the Reported Plutonium Concentration	62
C.4.3. Adjustments to the Reported Free Acid Normality	64
C.4.3.1 Derivation of ideal solution equation	65
C.4.3.2 Experimentally determined free acid correlation	65
C.4.3.3 Use of experimentally determined free acid correlation with critical experiment solution data	66
C.5 Corrected Analytical Chemistry Data for Critical Experiment Solutions	66
References for Appendix C	67
Appendix D	71
Critical Experiment Measurements for Which Analytical Chemistry Data Were Judged to Be Inadequate	71
Appendix E	73
Analytical Chemistry Procedure for Determination of Free Acid Molarity of Mixed Plutonium-Uranium Nitrate Solutions	73
E.1 Summary	73
E.2 Application	73
E.3 Apparatus	73
E.4 Reagents	73
E.5 Standards	74
E.6 Safety	74
E.7 Quality Control	74
E.8 Analysis	74

Appendix F	77
Sample Code Input	77
Appendix G*	79
Method to Establish Subcriticality	79

v/vi

1. INTRODUCTION

During the late 1970s, breeder reactors using mixed plutonium-uranium oxide fuel were expected to be commercialized by the early 1990s. Consequently, the Consolidated Fuel Reprocessing Program (CFRP) was initiated to design a reprocessing plant capable of handling the spent fuel from several large [1000 MW(e)] liquid-metal fast breeder reactors (LMFBR).

During the early phases of the project, it was planned that a coprocessing scheme, in which the plutonium was never completely separated from the uranium, would be utilized. In the traditional Purex process, the plutonium is separated from the uranium and fission products. In the coprocessing scheme, the plutonium and uranium were to remain in the same solution throughout the processing operation. A maximum credible Pu:(Pu+U) ratio of 0.5 was theorized by process designers. A review of available critical experiment data revealed that no benchmark experiments were available for this particular Pu:(Pu+U) ratio. Consequently, a critical experiment program was initiated at the Pacific Northwest Laboratory (PNL) Critical Mass Laboratory (CML).

About 1980, the decision was made to return to the traditional Purex process. However, as a result of research conducted on the coprocessing scheme, it had been shown that mixed oxide produced by coprecipitating uranium and plutonium from the same solution appeared to offer dissolution qualities superior to mixed oxide produced by mechanically blending plutonium oxide with uranium oxide. Thus, even after the decision was made to design the reprocessing plant for separate uranium and plutonium streams, the possibility of using a coprecipitation process to produce mixed oxide meant that a need for the critical experiment data still existed.

In August 1983, the U.S. Department of Energy entered into a cooperative agreement with the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan to sponsor a criticality data development program. The PNC was engaged in the design of a LMFBR mixed-oxide fuel reprocessing plant. The Japanese design proposes to use a coprocessing scheme, and the Japanese thus identified a need for critical experiment data just as the personnel of the CFRP had done in the late 1970s.

This report contains a description of critical experiments conducted with mixed plutonium-uranium solutions having a Pu:(Pu+U) ratio >0.5 . Data obtained during May 1979 to August 1984 are included. Additional experiments with mixed plutonium-uranium solutions are planned.

2. EXPERIMENTAL APPARATUS

After consideration of the reprocessing plant design, it was deemed necessary to conduct experiments in both slab and cylindrical geometries. The slab experiments would serve as benchmarks for the proposed solution storage vessels. The cylindrical experiments would serve as benchmarks for pulse columns, surge tanks, and mixing tanks.

2.1 CYLINDRICAL VESSELS

Two cylindrical vessels with inside diameters of 35.39 and 68.68 cm (13.9 and 27 in.) were fabricated for use in the critical experiments. Several factors influenced the selection of these diameters.

Since the plutonium concentrations present in the reprocessing plant streams could vary from zero to a few hundred grams per liter, it was desirable to span this entire range with benchmark experiments. To minimize cost, as few vessels as possible were fabricated. The maximum possible height of the cylinders was limited to ~170 cm (67 in.) because it was necessary to mount control rod drive mechanisms above the cylinders and solution drain and fill lines below the cylinders. Only a limited quantity of plutonium (66 kg) was available for this experimental program. To address all potential reprocessing plant designs adequately, various reflectors were considered including water, concrete, and unreflected assemblies.

Considering these constraints, scoping calculations performed with the KENO-IV¹ computer code and its associated (Hansen-Roach) cross-section library showed that a cylinder with a 35.6-cm (14-in.) diam and a height of 100 cm (39 in.) would allow experiments to be conducted over almost the entire range of desired plutonium concentrations.

Criticality could not be achieved in the small-diameter cylinder (water reflected) with solutions having plutonium content less than ~50 g/L. Since data at low plutonium concentrations were desired (to determine maximum allowable plutonium concentrations in waste streams) and since it was planned in the future to conduct experiments with gadolinium-poisoned, Pu+U solutions, a second, larger cylinder was needed. Again, scoping calculations were performed, and it appeared that a vessel with a 76.2-cm (30-in.) diam [100-cm (39-in.) height] would allow experiments to be performed at all desired concentrations. This diameter was later reduced to 68.6 cm (27 in.) to provide sufficient leakage of neutrons for operational control. (The control rods were mounted external to the tank and provided shutdown margin by absorbing neutrons that would otherwise return to the fissile solution from the surrounding water reflector.)

2.1.1 Reflector Tank

Since criticality measurements were to be performed on both the small and large cylinders, it was necessary to construct a reflector tank around the two cylinders. To reduce costs, a common holding tank was constructed rather than two separate ones. A schematic of this reflector tank is shown in Fig. 2.1. The tank was fabricated from carbon steel and contained two Lucite port covers on the front of the vessel to allow for installation of the two cylindrical vessels. The "as-built" impurities of the carbon steel (ASTM carbon steel A36) used to fabricate the water reflector tank are listed in Table 2.1. The derived atom densities for the carbon steel (assuming a steel density of 7.82 g/cm^3) are listed in Table 2.2; those for Lucite are listed in Table 2.3. The base of the water reflector tank had a thickness of 0.25 in. (0.64 cm). The side walls supported by a frame structure had a thickness of 0.15 cm (0.06 in.) (16-gage steel).

2.1.2 Small Cylindrical Vessel

The engineering drawing of the small cylinder is shown in Fig. 2.2. The inside diameter was 35.39 cm (14 in.); inside height was 107.63 cm (42 in.); and the side, top, and bottom wall thicknesses were all 0.079 cm (0.031 in.). The vessel was fabricated from stainless steel 304 (SS304). The constituents of SS304 are given in Table 2.4. These constituents are typical for SS304 and were taken from Petrie and Cross.¹ A photograph of the vessel is shown in Fig. 2.3. The control blades, which were external to the cylinder, are shown fully inserted in Fig. 2.3.

The solution feed and dump lines enter the base of the cylinder through a common dump valve shown in Fig. 2.4. A third line, leading to a manometer that records solution height, also extends from the dump valve. With the piston closed, solution could be pumped into the cylinder. However, the dump line had a sufficiently large diameter to allow fissile solution to drain from the cylinder even if, for some reason, flow of solution into the cylinder could not be stopped when the piston opened.

2.1.2.1 Reflectors for the small cylinder

Four conditions of neutron reflection were examined. These included no reflection (empty water reflector tank), water, combined steel and water, and concrete reflection.

Bare and water-reflected experiments. The small cylindrical tank in Fig. 2.4 rested on the lip of the dump valve, 21.11 cm (8.3125 in.) above the base of the reflector tank. When bare experiments were conducted, the closest reflecting surface was this 0.64-cm- (0.25-in.) thick steel base. Note that this base was supported by steel bars 1.9 by 5 cm (0.75 by 2 in.) on 41-cm (16-in.) centers centered around each dump valve. Radially at the closest point, the 0.15-cm- (0.06-in.) thick carbon steel side walls of the reflector tank were 31.8 cm (12.5 in.) from the outer surface of the small cylinder. Lucite control and safety rods were used on the unreflected vessels. Control was achieved by the addition or removal of reflection. The large cylinder was closer to the reflector tank walls than the reflector tank base for a small solid angle. At the point of closest approach, the large cylinder was 15 cm (6 in.) from the reflector tank wall.

NOTE: DIMENSIONS
IN INCHES

Fig. 2.1. H₂O tank.

Table 2.1. "As-built" impurities in the carbon steel used for the water reflector tank

Element	Wt %
C	0.190
Mn	0.380
P	0.003
S	0.019
Si	0.190

Table 2.2. Constituents of carbon steel

Element	Atom density [atoms/(b·cm)]
Fe	8.376×10^{-2}
C	7.450×10^{-4}
Mn	3.258×10^{-4}
P	4.562×10^{-6}
S	2.791×10^{-5}
Si	3.186×10^{-4}

Table 2.3. Constituents of Lucite

Element	Atom density [atoms/(b·cm)]
H	5.6884×10^{-2}
C	3.5552×10^{-2}
O	1.4221×10^{-2}

When water-reflected experiments were conducted, the water level was at the top of the small cylindrical vessel. The reflection from the control rod drive mechanism above the cylinder and the effect on reactivity of the dump line below the cylinder are believed to be negligible.

Combination steel and water reflectors. During the planning stage of these experiments, a set of concentric, hemicylindrical steel shells was found at the Oak Ridge Critical

$\frac{1}{4}$ - 20 UNC - 28 x $\frac{3}{8}$ DEEP
(12) HOLES EQ SPACED

22 GA SHEET
(0.031 THICK)

NOTE: DIMENSIONS IN INCHES

Fig. 2.2. Small cylindrical vessel.

Table 2.4. Constituents of stainless steel 304

Element	Atom density [atoms/(b·cm)]
C	3.1691×10^{-4}
Cr	1.6471×10^{-2}
Mn	1.7321×10^{-3}
Fe	6.036×10^{-2}
Ni	6.4839×10^{-3}
Si	1.694×10^{-3}

Mass Laboratory. By coincidence, these shells had been constructed to fit snugly around a 35.5-cm- (14-in.) diam cylinder. The shells were retrieved, analyzed, and shipped to the PNL Critical Mass Laboratory. A description of each of the shells is provided in Appendix A. The composition of the steel reflectors was determined experimentally; the calculated atom densities of the constituents of the steel are given in Table 2.5 and are derived in Appendix A.

The steel shells rested on the base of the reflector tank in the the critical experiments using a composite steel and water reflector. In all steel and water experiments described in this report, the steel thickness was 1.27 cm. The water level in the experiments was at the top of the small cylinder.

Concrete reflector. A 25.4-cm- (10-in.) thick concrete reflector was fabricated to fit snugly [0.635-cm (0.25-in.) gap] around the 35.5-cm- (14-in.) diam cylinder. The dimensions of the reflector are provided in Fig. 2.5. An artist's conception of the position of the concrete reflector for the first concrete-reflected experiment (PNL experiment number 4R) is shown in Fig. 2.6. The concrete annulus rested on the base of the water reflector tank; the space between the bottom of the small cylinder and the base of the water reflector tank contained air, not concrete.

A detailed description of the concrete reflector is contained in Appendix B. The calculated atom densities for the constituents of the concrete are shown in Table 2.6.

2.1.3 Large Cylindrical Vessel

The dimensions of the large cylinder are shown in Fig. 2.7. The measured inside diameter of the cylinder was 68.68 cm (27 in.). As noted in the figure, the cylinder was fabricated from SS304. The solution feed and dump lines entered the large cylinder in the same manner as for the small cylinder (Fig. 2.4). All experiments conducted in the large cylinder were reflected by water. Reactor control was provided by two control rods that fit snugly around the outside surface of the tank. At the closest point, the water reflector tank wall was 15 cm (5.9 in.) from the outside edge of the large cylinder.

Fig. 2.3. Small cylindrical vessel with control blades inserted.

Fig. 2.4. Dump valve.

Table 2.5. Constituents of steel reflector shells

Element	Atom density [atoms/(b·cm)]
As	1.2636×10^{-5}
C	2.84022×10^{-3}
Cu	2.97970×10^{-5}
Fe	8.36490×10^{-2}
Mn	3.96356×10^{-4}
P	3.05648×10^{-5}

2.1.4 CFRP Cylindrical Assembly

Figure 2.8 is a diagram showing the two-vessel assembly. Figure 2.9 is a photograph of the completed assembly. At their closest point, the small and large cylinders were separated by a distance of 26.8 cm (10.5 in.).

2.2 SLAB TANK

A variable thickness slab tank had been constructed several years before the start of this series of experiments. This tank was used in critical experiments conducted with plutonium nitrate solutions. A description of the tank and the previous series of experiments can be found in refs. 2 and 3.

The following description of the slab tank is taken from ref. 2.

The slab assembly [Fig. 2.10] used in these experiments was of rather unique design in that its thickness could be adjusted over a range of 3 to 9 in. This range of adjustments was made possible by stainless-steel bellows fabricated around the periphery of the tank. The slab thickness change was accomplished by means of adjustment screws, located at the corners of the vessel, between the opposite sides of the slab; slab thickness was measured by means of a dial caliper. The height and width of the tank was 42.6 in., based on the average of the bellows variation. The stainless-steel sides (0.062 in.) were reinforced with an egg-crate-type structure that was effective in maintaining the side position to within ~ 0.010 in. on filling the tank. The assembly and its parts were of Type-304-L stainless steel. A reflected assembly was achieved by ... attaching gasketed side plates and filling with water. The assembly was positioned in a large hood for contamination control.

The grid structure was composed of 0.3175-cm- (0.125-in.) thick steel plate. The measured average center-to-center spacing of the support grid structure was 10.6 cm (4.2 in.). The bellows (Fig. 2.10) was constructed with a tip-to-trough distance of 5.08 cm (2 in.) at minimum slab thickness. The bellows was composed of two sets of four V-shaped structures, the sets separated by a U-shaped structure having a diameter of 2.22 cm (0.87 in.) and a depth of ~ 2.5 cm (1 in.). The corners of the slab tank were rounded with a 14.44-cm (5.69-in.) radius measured from the outside of the tank.

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Fig. 2.5. Concrete reflector.

Fig. 2.6. Concrete-reflected cylindrical vessel.

Table 2.6. Calculated atom densities for the constituents of the concrete reflector

Element	Atom density [atoms/(b·cm)]
O	4.553×10^{-2}
Si	1.154×10^{-2}
Ca	4.201×10^{-3}
Al	2.491×10^{-3}
Fe	8.468×10^{-4}
Na	8.728×10^{-4}
H	1.462×10^{-2}
Mg	5.310×10^{-4}
K	2.584×10^{-4}
S	1.663×10^{-4}
Ti	9.667×10^{-5}

The slab tank was positioned inside a metal frame (Fig. 2.11) to which gasketed side plates could be attached. In both water-reflected and bare experiments these side plates *were* present. In water-reflected experiments, the water level was set at the center of the top bellows of the slab tank. The side plates for the broad faces of the water reflector tanks were 0.635 cm (0.25 in.) thick and fabricated from stainless steel. The side plates for the narrow faces of the water reflector tank were 0.476 cm (0.1875 in.) thick and were also fabricated from stainless steel.

The inside dimensions of the water reflector tank were 68.6 cm (27.0 in.) wide by 142.2 cm (56.0 in.) long by 144.8 cm (57.0 in.) high. The slab tank was positioned such that the tip of the bellows on the lower edge of the slab was 15.9 cm (6.3 in.) above the inside face of the lower water reflector side plate. The tank was positioned such that the narrow side faces were ~15.2 cm from the inside wall of the water reflector tank as measured from the bellows tips. The distances from the broad faces of the slab tank to the water reflector tank walls varied according to the critical configuration being examined, but in no instances were less than 15 cm (5.9 in.).

The slab tank and associated water reflector tank were surrounded by a containment hood (Fig. 2.12). The hood has a stainless steel structure with 0.95-cm- (0.37-in.) thick Plexiglas windows. The steel structural material was ~0.63 cm (0.25 in.) thick.

The bottom of the water reflector tank was located 100.6 cm (39.6 in.) above the concrete floor of the experimental cell. The broad faces of the water reflector tank were located 34.29 and 138.43 cm (13.5 and 54.5 in.), respectively, from the inside of the hood walls. The narrow faces of the tank were located 24.58 and 109.5 cm (9.7 and 43 in.), respectively, from the inside of the hood walls.

The slab tank was equipped with a control rod drive mechanism that provided for insertion of the control blade directly into the solution. The control rod drive mechanism was mounted above the slab tank. A 2.5-cm- (1-in.) thick steel grate platform was

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NOTE: DIMENSIONS IN INCHES

Fig. 2.7. Large cylindrical vessel.

Fig. 2.8. Dual cylinder tank assembly.

Fig. 2.9. CFRP experimental assembly.

Fig. 2.10. Experimental slab tank assembly.

Fig. 2.11. Critical experiments adjustable slab assembly.

Fig. 2.12. Containment hood surrounding slab tank assembly.

mounted inside the hood, 230 cm (90.5 in.) above the floor. The grate provided a working platform for the installation of the control rod drive mechanism.

2.3 LOCATION OF EXPERIMENTAL VESSELS IN THE CRITICAL ASSEMBLY ROOM OF THE CRITICAL MASS LABORATORY

As is apparent from Fig. 2.9, the CFRP experimental assembly was located approximately 30 cm (11.8 in.) from the containment hood that enclosed the slab tank. A floor plan of the experimental cell (not to scale) is shown in Fig. 2.13. A schematic showing the piping connections between the three experimental vessels is shown in Fig. 2.14. This piping arrangement allows critical experiments to be conducted with the same solution in each of three vessels.

NOTE: DRAWING NOT TO SCALE

Fig. 2.13. Floor plan of experimental cell.

Fig. 2.14. Piping schematic for three experimental vessels.

The Critical Assembly Room is 10.67 m^2 (35 ft^2) and has a ceiling height of 6.4 m (25 ft). The side walls are composed of 1.524-m - (5-ft) thick concrete. The concrete ceiling and floor are each 0.61 m (2.4 ft) thick.

The north side of the containment hood facing the wall with the uranium solution storage tanks was located 1.83 m (6 ft) from that wall. The side of the hood facing the wall containing the M-S and M-D tanks was located 1.52 m (5 ft) from that wall.

REFERENCES FOR SECTION 2

1. L. M. Petrie and N. F. Cross, *KENO-IV—An Improved Monte Carlo Criticality Program*, ORNL-4938, November 1975.
2. R. C. Lloyd et al., "Criticality of Pu Nitrate Solutions in Slab Geometry," *Nucl. Technol.* **18**, 225–30 (June 1973).
3. L. E. Hansen et al., "Critical Parameters of Plutonium Systems. Part 1: Analysis of Experiments," *Nucl. Appl.* **6**, 371–80 (April 1969).

3. EXPERIMENTAL MEASUREMENTS

The first CFRP-sponsored, solution-critical experiment was conducted in February 1981. In mid-March 1982, it was disclosed that some of the analytical chemistry procedures were in error. A study of analytical procedures was conducted, and the results are documented in Appendix C. As a result of that investigation, ten critical experiments were found to be inappropriate for use as benchmarks. These experiments do, however, provide some qualitative data regarding mixed Pu+U solutions and hence, data from these ten are reported in Appendix D. A revised analytical chemistry procedure for measuring the free acid content of mixed plutonium and uranium nitrate solutions was developed, and a summary of this procedure is presented in Appendix E.

The measured critical experiment parameters for those experiments judged to be of acceptable quality are shown in Table 3.1. The corrected analytical chemistry measurement results for the experimental solutions are shown in Table 3.2 (see Appendix C for derivation of the data in Table 3.2). The isotopic analyses of the plutonium and uranium in the solutions are given in Table 3.3.

Perhaps the most troublesome aspect of the data presented in Table 3.2 is that the nine measured free acid values are consistently lower than free acid values derived from (Eq.) C.5. Even though the uncertainty in the derived free acid value is large, the consistently low measured values raise the suspicion that some other factor, such as sample degradation, should be considered.

**Table 3.1. Mixed uranium-plutonium nitrate solution
critical experiment measurements**

PNL experiment number	Solution identification number	Geometry	Reflector ^a	Diameter or thickness ^b (cm)	Height ^c (cm)
9	929	slab ^d	water	14.90	56.30
10	930	cylinder	concrete	35.56	33.00
22	934	slab ^d	water	14.90	56.60
23	941	slab ^d	water	14.90	44.96
24	942	cylinder	water	35.56	28.04
25	943	cylinder	steel and water	35.56	30.45
26	944	cylinder	bare	35.56	56.57
27	945	cylinder	concrete	35.56	30.20
28	963	cylinder	concrete	35.56	32.54
29	964	cylinder	steel and water	35.56	33.92
30	965	cylinder	water	35.56	30.33
31	970	cylinder	water	35.56	25.29
32	971	cylinder	water	35.56	25.31
33	972	cylinder	water	35.56	25.32
34	972	cylinder	water	35.56	25.18
35	972	cylinder	water	35.56	25.06
36	972	cylinder	water	35.56	25.25
37	973	slab ^d	water	14.92	39.01
38	974	slab ^d	water	14.92	35.29
39	975	slab ^d	bare ^e	17.46	76.70
40	976	cylinder	water	35.56	23.66
41	977	cylinder	steel and water	35.56	25.38
42	983	cylinder	bare	35.56	40.80
43	984	cylinder	water	35.56	24.82
44	985	cylinder	water	68.58	16.40
45	994	cylinder	concrete	35.56	26.41

^aFor all steel and water experiments, the steel thickness was 1.27 cm (0.5 in.).

^b1 σ uncertainty in critical radius is ± 0.079 cm (0.0031 in.). 1 σ uncertainty in slab thickness is ± 0.025 cm (0.010 in.).

^c1 σ uncertainty is ± 0.159 cm (0.062 in.).

^dWidth of slab is 106.7 cm (42 in.).

^eThe 0.95-cm- (0.37-in.) thick Plexiglas containment hood walls provide significant reflection.

Table 3.2. Final analytical chemistry analyses for solutions utilized in critical experiments

Solution identification number	Pu concentration ^a (g/L)	U concentration ^b (g/L)	Specific gravity ^c (g/cm ³)	Free acid normality ^d (mol/L)
929	331.0 ^e	187.0 ^e	1.8260	2.00
930	346.0 ^e	190.0 ^e	1.8450	1.70
934	352.0 ^e	195.0 ^e	1.8640	1.80
941	297.0 ^e	160.0 ^e	1.7370	2.00
942	301.0 ^e	167.0 ^e	1.7430	1.63
943	303.0 ^e	168.0 ^e	1.7470	1.61
944	304.0 ^e	170.0 ^e	1.7480	1.50
945	307.0 ^e	171.0 ^e	1.7560	1.57
963	356.1	194.2	1.8615	1.55 (1.29)
964	356.2	194.8	1.8620	1.54 (1.21)
965	356.3	195.2	1.8615	1.50 (1.29)
970	245.1	135.0	1.6145	1.59
971	245.1	136.0	1.6145	1.55
972	244.8	135.7	1.6142	1.57
973	245.7	137.4	1.6168	1.53
974	199.3	111.9	1.5104	1.48 (1.32)
975	201.2	113.8	1.5161	1.48 (1.27)
976	201.1	114.2	1.5155	1.45 (1.23)
977	201.5	113.7	1.5167	1.50
983	174.0	153.6	1.5236	1.39 (1.08)
984	174.3	153.2	1.5239	1.40 (1.10)
985	173.6	153.3	1.5171	1.20 (1.12)
994	174.1	153.8	1.5236	1.37

^aUnless otherwise noted, 1 σ uncertainty is 0.55 g/L.

^bUnless otherwise noted, 1 σ uncertainty is 0.36 g/L.

^c1 σ uncertainty is 1.27×10^{-3} g/cm³.

^d1 σ uncertainty is 0.28 mol/L. Values in parentheses are the measured free acid values using the procedure in Appendix E and the correction factor in Table C.5 and have a 1 σ uncertainty of 0.013 mol/L.

^e1 σ uncertainty is 1 g/L.

Table 3.3. Plutonium and uranium isotopic analyses

	Experiments 001 to 041 (wt %) ^a	Experiments 042 to 045 (wt %) ^b
Plutonium		
238	0.030 ± 0.004	0.030 ± 0.003
239	91.500 ± 0.005	91.190 ± 0.050
240	7.891 ± 0.005	8.180 ± 0.040
241	0.519 ± 0.005	0.514 ± 0.005
242	0.060 ± 0.003	0.085 ± 0.001
Uranium		
234	0.011 ± 0.002	0.009 ± 0.002
235	0.724 ± 0.007	0.699 ± 0.006
236	0.029 ± 0.002	0.025 ± 0.001
238	99.223 ± 0.010	99.260 ± 0.007
233	0.013 ± 0.002	0.007 ± 0.002

^a $^{241}\text{Am} = 2.09 \times 10^{-3}$ of americium per gram of plutonium (December 30, 1981).

^b $^{241}\text{Am} = 2.86 \times 10^{-3}$ of americium per gram of plutonium (January 24, 1983).

4. CALCULATED k -EFFECTIVES FOR CRITICAL EXPERIMENTS

4.1 CODES AND LIBRARY

Values of k -effectives for the experiments were calculated using the CSAS2 module of the SCALE¹ code system. This module acts as a "driver," using one set of input data to provide input for the cross-section processing code NITAWL² and the Monte Carlo transport code KENO-IV³ (which is used to calculate k -effective). Cross sections were taken from a 27-group library derived from the ENDF/B-IV.^{1,4}

In processing the cross-section data, the infinite homogeneous medium option in CSAS2 was selected. In this option, the assumption is made that the neutrons are thermalized in the same medium in which they are born. This assumption is valid for all experiments described in this report since the minimum critical dimension of any assembly [14.9 cm (5.8 in.)] is several times greater than the neutron mean free path [~ 1 cm (2.5 in.)]. The calculations reported in this section are based on diameters for the small and large cylinders of 35.56 and 68.68 cm (14 and 27 in.) respectively. These values were taken from the engineering drawings and are slightly different from measured values.

4.2 COMPUTATIONAL MODELS

4.2.1 Cylinders

The half of the reflector tank, which contained the experimental vessel, the dump valve, and the experimental vessel were included in both of the computational models for the small and large cylinders. Some calculations were performed with a model containing both cylinders, which showed that interactions between the small and large cylinders were nonexistent. Sample input for each cylinder is contained in Appendix F.

4.2.2 Slab

The experimental vessel, supporting grid structure, support frame with steel side plates, Plexiglas contamination hood, and concrete floor were included in the computational model. The grid structure was modeled exactly by dividing the slab into individual boxes, each one corresponding to a single square grid (see Fig. 2.10). These boxes were then "stacked" to give structure to the entire slab.

The reported critical heights include the solution contained in the bellows. In order to correctly model the relation of solution height to grid structure, it was necessary to calculate an effective thickness for the bellows. In all cases, the thickness was calculated to give

an added 2.3 cm (0.9 in.) of solution (Sect. 2.2). Sample input for a water-reflected slab case is given in Appendix F.

4.3 CALCULATED VALUES OF k -EFFECTIVES

The calculated values of k -effectives and associated uncertainties are shown in Table 4.1. The measured free acid values were used in the calculational study. Most of these cases are the average of 100 generations of neutrons with 300 neutrons per generation. However, each case was inspected to ensure that the calculation had converged and that a normal distribution of k -effectives was generated. For those cases in which a measured free acid value was available, that value was used in the calculation. Microfiche copies of the output are provided at the end of this report.

**Table 4.1. Calculated k -effectives for PNL experiments
(27 groups, ENDF/B-IV, KENO-IV)**

PNL experiment number	Geometry	Reflector	k -effective $\pm 1 \sigma$
9	slab	water	1.00719 $\pm$ 0.00481
10	cylinder	concrete	1.00363 $\pm$ 0.00509
22	slab	water	1.00354 $\pm$ 0.00509
23	slab	water	0.99893 $\pm$ 0.00503
24	cylinder	water	1.01321 $\pm$ 0.00444
25	cylinder	steel and water	0.99762 $\pm$ 0.00484
26	cylinder	bare	0.99655 $\pm$ 0.00248
27	cylinder	concrete	0.99606 $\pm$ 0.00486
28	cylinder	concrete	1.01590 $\pm$ 0.00244
29	cylinder	steel and water	1.02238 $\pm$ 0.00460
30	cylinder	water	1.01402 $\pm$ 0.00226
31	cylinder	water	0.99466 $\pm$ 0.00499
32	cylinder	water	0.98699 $\pm$ 0.00464
33	cylinder	water	0.99744 $\pm$ 0.00564
34	cylinder	water	0.99973 $\pm$ 0.00456
35	cylinder	water	0.99851 $\pm$ 0.00544
36	cylinder	water	0.99555 $\pm$ 0.00567
37	slab	water	0.99268 $\pm$ 0.00587
38	slab	water	1.00580 $\pm$ 0.00460
39	slab	bare	0.99210 $\pm$ 0.00198
40	cylinder	water	1.00459 $\pm$ 0.00513
41	cylinder	steel and water	0.99463 $\pm$ 0.00570
42	cylinder	bare	1.00269 $\pm$ 0.00511
43	cylinder	water	1.02197 $\pm$ 0.00530
44	cylinder	water	1.00751 $\pm$ 0.00237
45	cylinder	concrete	0.98703 $\pm$ 0.00519
Mean			1.00310

Since only one bare slab experiment (39) and one water-reflected, large cylinder experiment (44) were conducted, five k -effectives using five different random number sequences were calculated for each of these experiments. The average for each set of these five cases is listed in Table 4.1. Note that experiments 31 through 36 are the same configuration and were performed to check experiment repeatability.

The weighted (by standard deviation) mean value for all of the experiments was 1.00310. For those experiments conducted with solution having a Pu:(Pu+U) ratio of 0.64, the weighted mean value was 1.00254. For those experiments in which the solution Pu:(Pu+U) ratio was 0.53, the weighted mean value was 1.00602.

4.4 TESTS FOR TRENDS AND BIASES

To aid in understanding the data, some statistical analyses were performed with the SAS procedure CORR.⁵ A search for a linear relationship between a given variable and calculated k -effective was performed by calculating the Pearson product-moment correlation value and its level of significance. If a variable x can be expressed exactly as a linear function of another variable y , then the correlation value is 1 or -1 , depending on whether the two variables are directly related or inversely related. As the value of the significance level decreases, the degree of confidence that the computed correlation value represents a true phenomenon increases.

The values of the correlation coefficients and significance levels are shown in Table 4.2. No strong correlations were found. A linear regression against the three variables having the largest coefficients could explain only 34% of the variation in the data ($R^2 = 0.34$), and thus calculated k -effective was judged to be independent of the experimental parameters.

Table 4.2. Calculated Pearson product moment correlation coefficients and significance levels

Variable	Correlation coefficient	Significance level
Geometrical shape	-0.192	0.348
Reflecting conditions	0.129	0.529
Critical thickness ^a	0.198	0.333
Critical height	-0.251	0.216
Plutonium concentration	0.356	0.074
Uranium concentration	0.542	0.004
Specific gravity	0.413	0.036
Free acid normality	-0.261	0.198

^aDiameter for cylinders.

4.5 TOLERANCE LIMITS FOR CALCULATED k -EFFECTIVES

Since the k -effectives are averages of many simulated values, it is reasonable to assume that the parent distribution of the observed k -effectives is the normal or Gaussian distribution. Given this assumption, one can produce lower tolerance bounds on the assumed distribution by use of the methods proposed by Owen.⁶ However, before Owen's methodology can be applied, an estimate of the variability associated with the distribution must be determined.

There are two sources of variability in the data, the variability within a k -effective estimate (the KENO-IV Monte Carlo uncertainty) and the variability between the estimates (due to experimental uncertainties and other unknown factors). This pattern can be described by the model

$$k\text{-effective} = \mu + b + w ,$$

where μ is the mean value of k -effective, b is the variance component representing the variation between the estimates, and w is the variance component representing the variation within an estimate.

Given that the expected values of b and w are

$$E(b) = E(w) = 0 ,$$

$$V(b) = \sigma_b^2 ,$$

$$V(w) = \sigma_w^2 ,$$

and

$$V(k\text{-effective}) = \sigma_T^2 = \sigma_b^2 + \sigma_w^2 ,$$

an approximate estimate of σ_b^2 can be computed by calculating the sample variance of the 26 k -effectives. The formula for the sample variance is

$$\hat{\sigma}_b^2 = \Sigma(y_i - \bar{y})^2 / 25 = 9.109 \times 10^{-5} ,$$

where $y_i = 1, \dots, 26$, represent the k -effective estimates. Thus the quantity ΔK_u from Appendix G is calculated to be 0.00953.

An estimate of σ_w^2 can be found by calculating the average of the sums of squares of the k -effective uncertainties, that is

$$\sigma_w^2 \approx [(0.0048)^2 + (0.00509)^2 + \dots + (0.00237)^2 + (0.00519)^2] / 25 \approx 2.197 \times 10^{-5} .$$

Thus, the estimate of the variance of the distribution of the k -effective is approximately given by

$$\hat{\sigma}_T^2 \approx \hat{\sigma}_b^2 + \hat{\sigma}_w^2 = 9.109 \times 10^{-5} + 2.197 \times 10^{-5} = 11.306 \times 10^{-5} .$$

Since the degrees of freedom of σ_w^2 are not known, it is assumed to be zero, and σ_T^2 is assigned the degrees of freedom $\sigma_b^2, f = 25$.

Then the lower tolerance limit for the distribution of the k -effectives can be approximated by

$$L(f, P, \gamma) = 1.00310 - K_{f, P, \gamma} \sqrt{(1.131 \times 10^{-4})},$$

where the interval $[L(f, P, \gamma), +\infty]$ contains $100 \times P\%$ of the distribution of the k -effectives with $100 \times \gamma\%$ confidence, and 1.00310 represents the mean of the k -effectives.

Using the tables in Sect. 2 of Owen's report, one finds that

$$L(25, 0.99, 0.99) = 0.9652.$$

Thus if the normality assumption is correct, one can say that 99% of the k -effectives produced by the KENO-IV, 27-group ENDF/B-IV combination for the class of uranium and plutonium nitrate solution experiments of the type considered are larger than 0.965 with 99% confidence. If the confidence and inclusion limits are reduced to the 95%, 95% levels, the lower tolerance limit is calculated to be 0.9789. If the confidence and inclusion limits are increased to 99.9 and 99.5, the lower tolerance limit is calculated to be 0.9511.

REFERENCES FOR SECTION 4

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ABSTRACT

A series of critical experiments was conducted with mixed plutonium-uranium nitrate solutions having Pu:(Pu+U) ratios >0.5 . Three geometries and four conditions of reflection were examined. The plutonium concentrations ranged from 170 to 350 g/L. The value of k -effective for each experiment was calculated using the KENO-IV code and 27-group cross sections derived from the Evaluated Nuclear Data File B—version IV (ENDF/B-IV). The mean value for the set of 26 experiments was 1.003, with a minimum value of 0.987 and a maximum of 1.022. The spread in the distribution of calculated k -effectives is believed to be the result of uncertainties in analytical chemistry measurements. No correlation between condition of reflection and calculated k -effective was found. An allowable multiplication factor to be used in the evaluation of reprocessing equipment at conditions that have been investigated was calculated to be 0.945.

5. DERIVED CRITICALITY SAFETY LIMITS

As noted previously, Appendix G contains a procedure that can be used to determine an allowable multiplication factor for designing equipment to contain solutions with Pu:(Pu+U) ratios ranging from 0.50 to 0.65. The value K_c (Sect. 4), which is the k -effective that results from the calculation of benchmark experiments, has been determined to be 1.00310. By definition, the conduction of critical experiments justifies the use of a small margin of subcriticality for design purposes when the design is within the range of conditions studied in the experiments. Hence the value of ΔK_m , the margin of subcriticality, can be assumed to be 0.02. The quantity $K_c - \Delta K_u$ was determined in Sect. 4.5 to be 0.965 for 99% inclusion at 99% level of confidence and 0.98 for 95%, 95% levels of inclusion and confidence.

At this point, one must make a subjective judgment as to the degree of conservatism that should be incorporated in the derivation of safe limits. For areas where personnel are likely to be in contact with equipment containing mixed plutonium and uranium solutions of the type described in this report, the authors believe that the 99%, 99% levels are justified. Consequently, the maximum allowable multiplication factor, K_a from Appendix G, is determined to be 0.945.

5.1 SUPPLEMENTAL INVESTIGATIONS OF THE EFFECT ON k -EFFECTIVE OF UNCERTAINTIES IN EXPERIMENTS

While the discussion in Sect. 4.5 provides an evaluation of ΔK_u , some additional studies were performed to estimate the effects of individual experimental uncertainties on calculated k -effective. While these studies were not used in the derivation of safe limits, they are believed to be useful guides to future research directed towards reducing uncertainties in critical experiments.

Practically, it is impossible to assess individually the effects of all the uncertainties in all of the experimental measurements. Realistically, it is only necessary to examine those variables or combinations of variables that might have a reactivity effect, which is a significant fraction of the typical uncertainty in a particular KENO calculation (i.e., $\Delta K_u \sim 0.001$). The one-dimensional, discrete ordinates code XSDRNPM¹ was used with the 27-group, ENDF/B-IV cross sections to calculate reactivity changes for experimental uncertainties. The code XSDRNPM was used instead of KENO in order to eliminate statistical uncertainty.

The uncertainty in the critical height was 0.6 cm (0.06 in.), thickness was 0.025 cm (0.01 in.), and radius was 0.08 cm (0.03 in.). Experiments conducted in the bare slab tank should be the most sensitive to the uncertainty in critical geometry measurement. The code

XSDRNPM was used to calculate k -effectives for the reported critical thickness of experiment 39 and for the same configuration with a thickness of 17.62 cm (6.9 in.). The calculated difference in k -effective was 0.005. Adjusting this value by a ratio of 0.025 to 0.16 yields a k -effective difference of 0.001.

The uncertainty in the plutonium measurement was calculated to be 0.55 g/L. The effect of this uncertainty should be most pronounced in the experiment conducted with solution containing the lowest plutonium concentration (44). Consequently, k -effectives were calculated using XSDRNPM for experiment 44 with a plutonium concentration of 173.6 and 174.2. The change in k -effective from the increase in plutonium was -0.001 .

The uncertainty in the uranium measurement was calculated to be 0.36 g/L. To estimate the effect of this uncertainty, two XSDRNPM cases were executed. k -Effective for experiment 38 was calculated for a uranium concentration of 111.9 g/L and a second time with a concentration of 112.3. The change in k -effective due to this increase in uranium was -0.0004 .

The uncertainty in the density was calculated to be 1.27×10^{-3} g/cm³. Since experiment 38 had the lowest solution density, a model of that experiment was prepared in which the solution density was lowered to 1.509. The reactivity change because of this decrease was -0.0008 .

The uncertainty in the free acid measurement was the largest percentage error of any of the solution parameters, 0.28 mol/L for those acid values that were derived from the correlation developed from the mixed nitrate standards. To assess the effect of this uncertainty, two sets of XSDRNPM calculations were performed. Experiments 9 and 45 were each modeled with the free acid values given in Table 3.2 and those values less 0.28. The changes in k -effective from the reduction in free acid normality were 0.0095 and 0.0083 for experiments 45 and 9 respectively.

The uncertainties in the atom densities of the steel and water reflectors were felt to be sufficiently small to have an inconsequential effect on the calculated value of k -effective. An investigation was conducted on the effect of the uncertainties in the concrete reflector measurements for the small cylinder.

In order to study the effect of the concrete reflector, sensitivity coefficients were generated for all of the nuclides in the concrete. A sensitivity coefficient is defined as the percent change in k -effective due to a percent change in a particular cross section [coefficient = $(\Delta K/K)(\Delta \sigma/\sigma)$]. The coefficients were generated by performing both forward and adjoint XSDRNPM calculations of a concrete-reflected experiment, 4R, and then feeding the fluxes generated to the FORSS² computer code system. This system generates sensitivity coefficients for the cross sections selected.

The system was validated for this study by using FORSS to calculate a sensitivity coefficient for the hydrogen total cross section for the plutonium-uranium nitrate solution. This number was then compared to a sensitivity coefficient generated by performing two XSDRNPM calculations (one calculation had a solution hydrogen atom density that was 1% larger than the other calculation). The FORSS-generated coefficient was 0.35804; the XSDRNPM coefficient was 0.39378.

Calculated sensitivity coefficients are shown in Table 5.1. If the assumption is made that the cross-section values in the library are exact, then the quantity $\Delta \sigma/\sigma$ can be set equal to the experimental uncertainty in the measurement of a particular nuclide. Thus,

Table 5.1. Sensitivity of calculated k -effective for experiment 4R to uncertainties in the concentration measurements of the constituents of the concrete reflector^a

Element	Calculated sensitivity coefficient ^b [($\Delta K/K$)($\Delta\sigma/\sigma$)]	Positive k -effective sensitivity $\Delta K/K$
Al	8.5237×10^{-4}	1.77627×10^{-4}
Ca	9.0093×10^{-4}	1.63948×10^{-4}
Fe	1.7583×10^{-3}	1.35321×10^{-4}
Si	3.9468×10^{-3}	3.65377×10^{-4}
Mg	2.6148×10^{-4}	4.38282×10^{-5}
K	-5.1605×10^{-5}	8.94239×10^{-6}
Na	5.4158×10^{-4}	3.90429×10^{-5}
H	-1.6680×10^{-3}	8.80369×10^{-5}
S	1.3557×10^{-5}	1.69805×10^{-3}
Ti	-7.6410×10^{-3}	1.77684×10^{-3}
O	2.3125×10^{-2}	1.92063×10^{-3}
$\Sigma \Delta K/K $		4.7213×10^{-3}

^aCalculated with CSRL-IV, NITAWL, XSDRNPM, FORSS.

^bSensitivity to change in total cross section.

the maximum positive change in k -effective because of 1 σ uncertainties in the measured concrete nuclide concentrations can be determined by assuming that the uncertainty in the atom density is in the direction that provides positive reactivity. These $\Delta K/K$ values are also shown in Table 5.1.

The final entry in the table is the sum of the $\Delta K/K$ entries. The value 4.7213×10^{-3} is the maximum positive change in k -effective corresponding to a 1 σ uncertainty in the physical measurements.

The uncertainty in the value of calculated κ -effective from uncertainties in the isotopic analyses for plutonium and uranium would be largest for experiments 42 through 45. Specifically the ^{239}Pu and ^{240}Pu isotopic analyses have the largest uncertainties. The values of k -effective for experiment 45 were calculated for two cases. Case one was for nominal isotopic analyses, and case two was for ^{239}Pu content decreased by 0.05 wt % and ^{240}Pu content increased by 0.05 wt %. The change in κ -effective because of these changes was 0.001.

A summary of the maximum, 1 σ uncertainty in calculated k -effective due to the uncertainties in the experimental measurements is given in Table 5.2.

It is recognized by the authors that the procedure used in this section to derive uncertainties as a result of experimental measurements is likely to overestimate the value of that parameter. However, it is felt that these studies show that continued monitoring of free acid analyses should be an important part of future critical experiments programs. Analytical chemistry laboratories should be routinely tested with known uranium-plutonium nitrate standards; separate analytical chemistry laboratories should analyze samples from each experiment set. (See Appendixes C and E.)

Table 5.2. Maximum estimates of uncertainty in calculated k -effective due to uncertainties in experimental data

Parameter	Maximum 1σ uncertainty
Variation in critical dimensions	0.001
Plutonium concentration	0.001
Uranium concentration	
Specific gravity	0.001
Free acid	0.010
Concrete analyses	0.005
Isotopic analyses	0.001

REFERENCES FOR SECTION 5

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Appendix A

COMPOSITION AND DIMENSIONS OF STEEL REFLECTORS

This appendix describes the steel reflectors used in some of the experiments conducted in the small [35.6-cm- (14-in.) diam] cylinder. All reflectors were previously fabricated from the same unknown type of steel.¹ The composition of the steel was determined experimentally and found to be AISI-SAE 1020. Detailed measurements of the radii, thicknesses, and lengths of the reflectors are discussed. Each reflector's weight is provided along with calculated steel densities. The elemental components of the reflector are also identified and atom densities computed. Since the reflectors were painted for rust protection, the paint composition is provided. The density and thickness of the coat of paint were determined, and this information is provided.

A.1 PHYSICAL MEASUREMENTS OF STEEL REFLECTORS

The steel reflectors used with the small [35.6-cm- (14-in.) diam] cylinder were a series of concentric half cylinders; the innermost fits snugly around the solution containment cylinder. The set of steel shells is illustrated in Fig. A.1. Twenty-four shells were available for use as reflectors. All shells had a thickness of either 0.635 or 1.27 cm (0.25 or 0.5 in.). Table A.1 lists the possible combinations of reflector thickness (achievable by stacking shells). More than one combination exists for some total reflector thicknesses because, for some radii, two sets of shells were fabricated.

Measurements of the inside and outside radius of each of the 24 shells were obtained at several different points on each shell. Several points were measured so that some estimate of the cylindrical eccentricity could be obtained.

Figure A.2 identifies various points on the shell surface where measurements were taken. Initially, the heights AE, FJ, F^1J^1 , and A^1E^1 were measured. Then the points B, C, D, B^1 , C^1 , and D^1 were identified by spacing these points at equal axial intervals; that is, the distances AB, CD, B^1C^1 , D^1E^1 , etc., were all equal. Next, the inside and outside diameters were measured between the points BB^1 , CC^1 , and DD^1 . The centerline or axis of the cylindrical shell was then identified. Points G, H, I, G^1 , H^1 , and I^1 were then selected by spacing these points at equal axial intervals and at 60° spacings with respect to the centerline of the cylinder. The inside radius and the thickness of each shell were measured at points G, H, I, G^1 , H^1 , and I^1 .

The length and radius measurements were made with a calibrated wire. The thickness measurements were determined by the use of ultrasonics. The instrumental error for the thickness measurements was ± 0.1 mm (0.004 in.). Radii and diameter measurements had

Fig. A.1. Steel reflectors for 35.6-cm-ID cylinder.

Table A.1. Possible steel reflector thicknesses

Reflector thickness [cm (in.)]	Combination of shells required to achieve thickness (number assigned to each half shell)
0.635 (0.25)	(21, 24)
1.270 (0.50)	(21, 24) + (22, 23)
1.905 (0.75)	(21, 24) + (22, 23) + (6, 9)
2.540 (1.00)	(21, 24) + (22, 23) + (12, 13) ^a
3.175 (1.25)	(21, 24) + (22, 23) + (12, 13) + (5, 7)
3.810 (1.50)	(21, 24) + (22, 23) + (12, 13) + (11, 14) ^b
	(21, 24) + (22, 23) + (12, 13) + (5, 7) + (4, 10)
4.445 (1.75)	(21, 24) + (22, 23) + (12, 13) + (11, 14) + (3, 8)
5.715 (2.25)	(21, 24) + (22, 23) + (12, 13) + (11, 14) + (3, 8) + (2, 20)
6.985 (2.75)	(21, 24) + (22, 23) + (12, 13) + (11, 14) + (3, 8) + (2, 20) + (1, 19)

^a(15, 17) same thickness and radii as (12, 13).^b(16, 18) same thickness and radii as (11, 14).

Fig. A.2. Points on steel reflector at which measurements were taken.

an instrumental error of ± 0.1 mm (0.004 in.); height measurements, an error of ± 0.5 mm (0.010 in.). Table A.2 reports the measured parameters for each shell. The letters used to designate points in Table A.2 refer to the locations shown in Fig. A.2 (C is centerline).

Ideally, it would have been desirable to weigh the shells immediately after measuring their dimensions. Since the shells were not stainless steel, rapid oxidation (rusting) of the shells necessitated painting the shells with an epoxy paint before weighing. Seven shells were weighed both before and after painting to determine the quantity of paint applied to the surface (see Table A.3). The instrumental error of the scales was ± 0.1 g. The weights of the remaining shells are reported in Table A.4. These shells were all weighed after painting.

Table A.2. Physical dimensions for steel reflectors

	Reflector shell							
	1	2	3	4	5	6	7	8
Thickness at point (mm)								
G	13.1	13.0	6.4	6.5	6.4	6.4	6.4	6.4
H	13.1	13.0	6.4	6.4	6.4	6.4	6.4	6.4
I	13.2	13.0	6.4	6.4	6.4	6.4	6.4	6.4
G ^I	13.1	13.1	6.4	6.4	6.4	6.4	6.4	6.4
H ^I	13.1	13.1	6.4	6.5	6.4	6.4	6.4	6.4
I ^I	13.2	13.1	6.4	6.5	6.4	6.4	6.4	6.5
Radius (mm) ($\bar{r}$ to point)								
G	237.2	223.2	217.3	209.9	203.5	197.7	204.1	217.5
H	237.3	223.5	217.1	210.0	203.3	197.8	204.3	217.1
I	237.3	223.6	217.4	210.2	203.9	197.7	204.4	217.4
G ^I	237.3	223.7	217.3	203.3	203.3	197.5	204.9	217.9
H ^I	237.3	223.5	217.6	202.6	202.6	197.5	204.1	216.7
I ^I	237.2	223.4	217.1	203.2	203.2	197.5	204.1	216.9
Inside diameter at point (mm)								
BB ^I	473.5	447.0	434.4	421.2	407.6	394.5	408.2	433.7
CC ^I	473.5	446.9	434.4	421.1	407.1	394.6	408.2	433.0
DD ^I	473.6	447.0	434.4	421.0	407.4	394.7	407.8	433.3
Outside diameter at point (mm)								
BB ^I	499.7	473.3	447.3	434.1	420.4	407.3	421.1	446.6
CC ^I	499.5	473.3	447.3	434.0	419.9	407.5	421.1	445.9
DD ^I	500.0	473.4	447.2	434.0	420.3	407.5	420.7	446.1
Height (mm)								
AE	1206.5	1206.5	1206.5	1206.5	1206.5	1206.9	1206.9	1206.7
FJ	1206.0	1206.5	1206.5	1206.5	1206.5	1206.9	1206.9	1206.8
F ^I J ^I	1206.5	1206.5	1206.5	1207.0	1206.5	1206.9	1206.9	1206.8
A ^I E ^I	1206.0	1206.5	1206.0	1206.5	1206.5	1206.9	1206.9	1206.0

Table A.2. (Continued)

	Reflector shell							
	9	10	11	12	13	14	15	16
Thickness at point (mm)								
G	6.4	6.4	13.5	13.2	13.2	13.4	13.5	13.36
H	6.4	6.4	13.5	13.2	13.1	13.4	13.4	13.41
I	6.4	6.4	13.4	13.2	13.1	13.3	13.4	13.49
G ¹	6.4	6.4	13.4	13.3	13.2	13.4	13.4	13.86
H ¹	6.4	6.4	13.4	13.2	13.3	13.3	13.4	13.36
I ¹	6.4	6.4	13.4	13.2	13.2	13.3	13.4	13.36
Radius (mm) (ζ to point)								
G	197.5	210.2	209.5	194.7	194.5	207.9	193.6	208.15
H	197.2	210.0	209.4	194.8	196.0	207.9	194.2	209.18
I	197.5	210.1	210.5	195.4	195.5	208.4	194.7	208.69
G ¹	197.9	210.4	207.4	194.8	192.4	207.9	193.9	209.84
H ¹	196.9	209.2	208.4	195.6	192.5	208.7	194.2	210.60
I ¹	197.4	209.6	213.8	196.0	192.7	208.8	194.2	210.90
Inside diameter at point (mm)								
BB ¹	394.9	419.7	415.9	389.3	385.9	414.4	387.3	415.95
CC ¹	394.1	418.9	416.5	390.3	388.3	415.4	385.5	407.93
DD ¹	394.5	419.2	416.9	390.3	387.9	415.8	388.7	417.80
Outside diameter at point (mm)								
BB ¹	407.6	432.5	442.5	415.8	412.1	440.9	414.3	442.67
CC ¹	407.1	431.8	443.3	416.8	414.5	441.9	412.6	444.60
DD ¹	407.3	432.0	443.8	417.5	414.0	442.4	415.7	444.55
Height (mm)								
AE	1207.1	1206.7	1196.8	1199.6	1194.1	1194.2	1194.0	1194.43
FJ	1206.8	1206.8	1193.8	1193.7	1193.8	1193.7	1193.7	1193.93
F ¹ J ¹	1206.9	1206.9	1193.8	1193.6	1193.8	1193.8	1193.8	1193.80
A ¹ E ¹	1206.9	1207.1	1194.2	1194.2	1194.1	1194.0	1016.4	1194.33

Table A.2. (Continued)

	Reflector shell							
	17	18	19	20	21	22	23	24
Thickness at point (mm)								
G	13.39	13.28	13.26	13.26	6.4	6.37	6.43	6.37
H	13.34	13.26	13.26	13.21	6.4	6.37	6.45	6.37
I	13.39	13.23	13.26	13.23	6.4	6.35	6.45	6.43
G ¹	13.36	13.23	13.21	13.18	6.4	6.37	6.45	6.48
H ¹	13.31	13.36	13.22	13.28	6.4	6.40	6.48	6.45
I ¹	13.36	13.31	13.23	13.23	6.4	6.37	6.45	6.35
Radius (mm) (ζ to point)								
G	193.26	208.47	237.3	223.64	179.64	186.23	185.85	178.97
H	195.55	209.30	237.6	224.20	180.96	187.81	186.44	179.45
I	195.06	210.40	237.3	223.92	179.73	187.06	186.51	179.39
G ¹	193.99	207.10	237.1	223.17	178.83	185.86	185.62	178.69
H ¹	194.56	207.50	236.8	223.16	179.02	185.32	185.79	178.69
I ¹	195.1	207.70	237.3	223.58	178.77	184.75	185.75	178.60
Inside diameter at point (mm)								
BB ¹	386.72	414.05	473.0	446.15	358.0	371.22	371.04	357.35
CC ¹	388.37	414.44	472.82	445.80	359.8	370.20	371.91	357.83
DD ¹	389.31	417.14	473.71	446.74	358.4	371.20	369.82	357.38
Outside diameter at point (mm)								
BB ¹	413.41	440.44	499.39	472.54	378.0	384.18	383.92	370.20
CC ¹	415.19	441.81	499.19	472.24	372.8	386.18	384.81	370.95
DD ¹	416.08	443.64	550.15	473.12	371.3	384.16	383.95	370.43
Height (mm)								
AE	1194.33	1194.44	1206.75	1206.91	1193.9	1193.98	1193.85	1193.60
FJ	1194.1	1194.05	1206.75	1206.80	1193.9	1193.85	1193.62	1193.72
F ¹ J ¹	1193.8	1193.93	1206.96	1206.91	1194.0	1193.85	1193.67	1193.70
A ¹ E ¹	1194.33	1194.16	1207.01	1206.98	1194.0	1194.00	1193.85	1193.90

Table A.3. Weights of seven unpainted steel reflectors

Reflector identification number	Weight before painting (kg)	Weight after painting (kg)
1	95.0406	95.2428
2	89.8351	90.0708
3	41.6992	41.9192
4	40.5420	40.7480
5	39.2068	39.4102
6	38.0308	38.2288
13	76.8290	77.0338

Table A.4. Final weights of painted steel reflectors

Reflector identification number	Weight after painting (kg)
7	37.6940
8	42.1106
9	38.2368
10	40.8346
11	84.0384
12	77.7096
14	83.2915
15	79.4012
16	83.7744
17	78.6706
18	83.0508
19	95.4508
20	90.2774
21	33.1164
22	38.2640
23	36.1528
24	35.3580

Before cleaning and repainting, a small sample was cut from one of the shells, and a spark source mass spectrometry analysis was performed to determine the steel composition. The precision of the spark source spectrometer is $\pm 100\%$, $\pm 50\%$. Upon examination, the sample was found to contain two regions, the main steel region and a thin surface layer that was not soluble in $\text{HCl} + \text{HNO}_3$. The insoluble layer was white and powdery and constituted $\sim 2\%$ of the total reflector material by weight.

The constituents of the steel layer are reported in Table A.5. The constituents of the white, powdery layer are reported in Table A.6. It is speculated that the white layer is the residual paint, which was present on the surface of the reflectors before they were cleaned

Table A.5. Measured constituents of steel reflectors

Element	Reported weight parts per million of steel
As	200
B	0.7
C	7200
Ca	0.6
Cd	<2
Co	100
Cr	70
Cu	400
Dy	<2
Eu	<2
Fe	Major
Ga	20
Gd	<2
Mn ^a	4600
Mo	30
Ni	150
P	200
Sb	40
Sm	<2
Sn	70
V	9
W	30
Zr	0.4

^aPhotoelectric analysis, precision ± 5 .**Table A.6. Measured constituents of surface layer found on the steel reflector sample before cleaning**

Element	Surface layer (wt %)
Al	~5
Ba	0.5
Ca	0.04
Fe	~10
K	10
Mg	0.2
Mn	0.05
Ni	0.03
P	~2
Rb	0.01
Si	~30
Ti	~50
Zn	0.3

and repainted. If that is true, then this layer would have been removed by the cleaning (sandblasting) process.

Subsequent to cleaning and measurement of thickness, length, and radii, the shells were coated with a grey epoxy paint. This paint had a density, when dry, of 1.62 g/cm^3 . A spark source mass spectrometry analysis was performed on a dry chip to determine the constituents of the paint. The results of the analysis are shown in Table A.7.

A.2 CALCULATION OF ATOM DENSITIES FOR STEEL REFLECTORS

Mean values for the radii of each of the steel shells were calculated and are reported in Table A.8. The measurements of the inner diameter at points B, C, and D as well as the measured radii at other points were used to determine the average radii. The standard deviation of the radii distribution is also reported.

Table A.7. Measured constituents of epoxy paint used to coat steel reflectors

Element	Weight of solid (ppm)
Al	5000
As	10
B	20
Ba	300
Br	50
Ca	1000
Cd	50
Co	20
Cr	4
Cu	700
Fe	100
Ga	3
K	200
Mg	~3000
Mn	100
Na	200
Nb	1
Ni	4
P	30
Pb	80
Sb	600
Si	5000
Sr	10
Ti ^a	Major
Zn ^a	Major

^aEstimated by optimal emission spectrometry to constitute ~40% of solid material.

Table A.8. Mean values of physical parameters for steel reflectors

Reflector identification number	Inner radius (mm)	Thickness (mm)	Outer radius (mm)
1	237.0 $\pm$ 0.3	13.1	250.1 $\pm$ 0.3
2	223.5 $\pm$ 0.1	13.0	236.5 $\pm$ 0.1
3	217.2 $\pm$ 0.1	6.4	223.6 $\pm$ 0.1
4	210.3 $\pm$ 0.3	6.4	216.7 $\pm$ 0.3
5	203.5 $\pm$ 0.4	6.4	209.9 $\pm$ 0.4
6	197.5 $\pm$ 0.2	6.4	203.9 $\pm$ 0.2
7	204.2 $\pm$ 0.3	6.4	210.6 $\pm$ 0.3
8	217.0 $\pm$ 0.4	6.4	223.4 $\pm$ 0.4
9	197.3 $\pm$ 0.3	6.4	203.7 $\pm$ 0.3
10	209.8 $\pm$ 0.4	6.4	216.2 $\pm$ 0.4
11	209.0 $\pm$ 1.7	13.4	222.4 $\pm$ 1.7
12	195.2 $\pm$ 0.4	13.2	208.4 $\pm$ 0.4
13	193.8 $\pm$ 1.2	13.2	207.0 $\pm$ 1.2
14	207.9 $\pm$ 0.5	13.4	221.3 $\pm$ 0.5
15	193.9 $\pm$ 0.6	13.4	207.3 $\pm$ 0.6
16	208.3 $\pm$ 2.2	13.5	221.8 $\pm$ 2.2
17	194.3 $\pm$ 0.7	13.4	207.7 $\pm$ 0.7
18	208.0 $\pm$ 1.1	13.3	221.3 $\pm$ 1.1
19	236.9 $\pm$ 0.4	13.2	250.1 $\pm$ 0.4
20	223.4 $\pm$ 0.4	13.2	236.6 $\pm$ 0.4
21	179.4 $\pm$ 0.6	6.4	185.8 $\pm$ 0.6
22	185.8 $\pm$ 0.9	6.4	192.2 $\pm$ 0.9
23	185.7 $\pm$ 0.5	6.5	192.2 $\pm$ 0.5
24	178.9 $\pm$ 0.3	6.4	185.3 $\pm$ 0.3

The average thickness and computed outer radius for each reflector shell are also shown in Table A.8. The distribution of the thickness measurements was small and, in many cases, zero. Therefore, it can be assumed that the same quantity was being measured at each point and that the resulting error in the final determination of the thickness is an instrumental uncertainty and is therefore equal to $0.1 \text{ mm} / \sqrt{6} = 0.041 \text{ mm}$. The reported uncertainty for the outer radius is simply the square root of the sum of the variances of the distribution of the inner radii and thicknesses; that is, the standard deviation of the distributions.

As mentioned previously, some steel reflectors were weighed both before and after painting. The quantity of paint on each shell, calculated from the values shown in Table A.3, is reported in Table A.9. It should be noted that the reflectors listed in Table A.9 span almost the entire range of radii and thicknesses, yet approximately the same amount of paint is contained on each shell.

Assuming a value for the weight of paint of 210 g, a corrected, unpainted weight for the steel reflectors was calculated. A listing of the measured and calculated unpainted weights of all the reflectors is provided in Table A.10.

Table A.9. Weight of paint on reflectors

Reflector identification number	Weight of paint (g)
1	202.2
2	235.7
3	220.0
4	206.0
5	203.0
6	198.0
13	204.8
Average value	210.0 ± 13.3

Table A.10. Unpainted weights of steel reflectors

Reflector identification number	Unpainted ^a weight (kg)
1	95.0406 ^b
2	89.8351 ^b
3	41.6992 ^b
4	40.5420 ^b
5	39.2068 ^b
6	38.0308 ^b
7	37.4840
8	41.9006
9	38.0268
10	40.6246
11	83.8284
12	77.4996
13	76.8290 ^b
14	83.0815
15	79.1912
16	83.5644
17	78.4606
18	82.8408
19	95.2408
20	90.0674
21	32.9064
22	38.0540
23	35.9428
24	35.1480

^aUnless noted otherwise, uncertainty = 1.3×10^{-2} kg.

^bUncertainty = 1×10^{-4} kg.

Examination of the length measurements revealed that all lengths were either 1194 or 1206.5 mm (47 or 47.5 in.) and were within the instrumental uncertainties. Since four measurements were made for each shell, the uncertainty associated with the length measurement can be assumed to be an instrumental uncertainty and equal to $0.5 \text{ mm} / \sqrt{4} = 0.25 \text{ mm}$.

Using values of radii from Table A.8 and weights from Table A.10 along with the appropriate length measurement, the density of each shell can be calculated. These densities are reported in Table A.11.

The calculated density and carbon and manganese content identify the reflector material as a carbon steel. The AISI-SAE number is 1020.

Atom densities for the six major elements present in the steel are reported in Table A.12. The values were calculated based on the concentrations reported in Table A.5 and the mean density reported in Table A.11.

Atom densities for the two major elements of the epoxy paint are $\text{Ti} = 1.00164 \times 10^{-2}$ and $\text{Zn} = 7.33839 \times 10^{-3} \text{ atoms}/(\text{b} \cdot \text{cm})$. The maximum possible paint thickness

Table A.11. Calculated densities of steel reflectors

Reflector identification number	Density (g/cm^3)
1	7.859 ± 0.255
2	7.927 ± 0.086
3	7.799 ± 0.172
4	7.828 ± 0.519
5	7.819 ± 0.691
6	7.811 ± 0.345
7	7.450 ± 0.494
8	7.844 ± 0.693
9	7.817 ± 0.518
10	7.862 ± 0.695
11	7.732 ± 1.388
12	7.756 ± 0.333
13	7.743 ± 0.996
14	7.702 ± 0.407
15	7.854 ± 0.498
16	7.674 ± 1.769
17	7.766 ± 0.574
18	7.736 ± 0.905
19	7.818 ± 0.335
20	7.827 ± 0.336
21	7.507 ± 0.995
22	8.387 ± 1.668
23	7.802 ± 0.849
24	8.040 ± 0.533
Mean value	7.866 ± 0.062

Table A.12. Calculated atom densities for major constituents of steel reflectors

Element	Atom density [atoms/(b·cm)]
As	1.26364×10^{-5}
C	2.84022×10^{-3}
Cu	2.97970×10^{-5}
Fe	8.36490×10^{-2}
Mn	3.96356×10^{-4}
P	3.05658×10^{-5}

can be computed by assuming 200 g of paint applied to the shell with the smallest radii. The paint thickness is thus computed to be 0.0894 mm (3.5 mills). Due to this small thickness, the coating of paint should not affect the neutronics of the critical experiments.

REFERENCES FOR APPENDIX A

1. E. B. Johnson and C. E. Newton, "The Effect of Steel-Water Reflectors on the Criticality of Low-Enriched Uranyl Fluoride Solution," *Trans. Am. Nucl. Soc.* **2**, 383-84 (June 1968).

6. CONCLUSIONS

Critical experiments were performed with plutonium-uranium nitrate solutions having Pu:(Pu+U) ratios greater than 0.5. The value of k -effective was calculated for each experiment, and a mean value of 1.003 was obtained for the set of 26 experiments. The variation in calculated k -effectives is likely a result of variation in the analytical chemistry measurements of the solution composition. A new procedure for analyzing the free acid normality of mixed plutonium-uranium nitrate solutions was developed as a part of the experimental program. An allowable multiplication factor of 0.945 (to be used in evaluating reprocessing equipment) was derived from the experimental data. Additional experiments, both poisoned and unpoisoned, in an unreflected slab tank and a large cylinder are recommended.

Appendix B

COMPOSITION OF CONCRETE REFLECTOR*

Two aliquots of the concrete reflector were analyzed for material composition by two techniques—X-ray fluorescence (XRF) and isotope neutron activation analysis (INAA). The procedure involved obtaining a section of the center of the core and reducing the material so that it would pass through a 140-mesh sieve. Two aliquots were thus obtained.

The INAA procedure involved a 5-min irradiation in the neutron multiplier facility, a 5-min delay, and then a counting of 600 s to obtain Al, V, Ti, Mg, and Ca. Approximately 2 h later the samples were recounted for 1000 s to obtain data for Na, K, and Mn. A summary of this data is found in Table B.1.

The XRF procedure involved pelletizing the material to form a thin wafer. The samples were then analyzed by both a Zr and then an Ag secondary source. These data are also shown in Table B.1. The reported error is the 1σ value for each sample and the weighted standard deviation for the mean value $[1/\Sigma(1/\sigma_i^2)]$.

Each sample was then separated into two aliquots. The water content was determined by the following procedure. Each of the samples was weighed, then heated to 100°C for 1 h, cooled, and reweighed. The samples were then heated to 1000°C for 1 h, cooled, and reweighed. The results of the analyses are shown in Table B.2. Based on the data in Table B.2, the hydrogen content of the concrete is determined to be 1.05 ± 0.03 wt %.

The major constituents of the concrete are listed in Table B.3. The unidentified mass from the analysis was assumed to be oxygen. The density of the concrete was determined to be 2.33 ± 0.1 g/cm³. The computed atom densities for each element are also listed in Table B.3.

*Analyses performed by E. Lepel, Pacific Northwest Laboratory, documented in a memorandum to M. Durst, PNL, May 27, 1981.

Table B.1. Concentration of elements in concrete reflector^a

Element	Unit of measure (wt)	INAA ^b		XRF ^c		Average
		1	2	1	2	
Na	%	1.40 ± 0.01	1.46 ± 0.01			1.43 ± 0.007
Mg	%	0.84 ± 0.46	1.00 ± 0.50			0.92 ± 0.34
Al	%	5.27 ± 0.03	5.58 ± 0.03	4.0 ± 1.4	4.3 ± 1.4	4.79 ± 0.02
Si	%			22.3 ± 1.5	23.9 ± 1.6	23.1 ± 1.1
P	%			<0.6	<0.6	<0.6
S	%			0.40 ± 0.09	0.36 ± 0.09	0.38 ± 0.06
K	%	0.86 ± 0.14	0.70 ± 0.15	0.67 ± 0.04	0.67 ± 0.04	0.72 ± 0.3
Ca	%	10.8 ± 0.5	10.4 ± 0.5	13.8 ± 0.7	12.9 ± 0.7	12.0 ± 0.3
Ti	ppm	3910 ± 340	3770 ± 340	2900 ± 200	2700 ± 200	3320 ± 122
V	ppm	91.3 ± 3.6	85.5 ± 3.8	91 ± 31	140 ± 30	102 ± 3
Cr	ppm			196 ± 21	134 ± 19	165 ± 14
Mn	ppm	557 ± 4	520 ± 4	606 ± 36	577 ± 34	565 ± 3
Fe	%			3.45 ± 0.17	3.29 ± 0.16	3.37 ± 0.12
Co	ppm			<49	<49	<49
Ni	ppm			57 ± 4	52 ± 6	54 ± 3
Cu	ppm			42 ± 3	39 ± 3	40 ± 2
Zn	ppm			176 ± 9	146 ± 8	161 ± 6
Ga	ppm			12 ± 1	11 ± 1	11.5 ± 0.7
Pb	ppm			34 ± 2	23 ± 2	28 ± 1
As	ppm			33 ± 2	29 ± 2	31 ± 1
Rb	ppm			21 ± 1	20 ± 1	20.5 ± 0.7
Sr	ppm			280 ± 20	300 ± 30	290 ± 17
Y	ppm			15 ± 1	15 ± 1	15 ± 0.7
Zr	ppm			85 ± 7	87 ± 7	86 ± 5
Nb	ppm			5.4 ± 1.0	5.3 ± 0.7	5.4 ± 0.6
Mo	ppm			6.9 ± 0.8	5.6 ± 0.8	6.2 ± 0.6

^aReported error is the 1- σ value for each sample. The weighted standard deviation is reported for the average.

^bIsotope neutron activation analysis method.

^cX-ray fluorescence method.

Appendix C

ANALYSIS OF MIXED PLUTONIUM-URANIUM NITRATE SOLUTION STANDARDS AND DERIVED CRITICAL EXPERIMENT SOLUTION PARAMETERS

C.1 INTRODUCTION

During CY 1981, 17 critical experiments were conducted. The experimental parameters were reported to ORNL in December 1981. Using these data, *k*-effectives were calculated, and by mid-January 1982 it was apparent that significant errors were present either in the experimental data or in the computational methods. After two months of study, the problem was isolated as being the analytical determination of the free acid normality.

Several methods exist for determining the free acid normality of plutonium-bearing solutions. Chemists at Pacific Northwest Laboratory, Hanford Engineering Development Laboratory (HEDL), and ORNL jointly investigated the various available methods. The results of their work were documented in a technical journal article.¹ The conclusion reached by all parties was that a revised analytical procedure was necessary. The new procedure is listed in Appendix E. This procedure is to be used in all future free acid determinations sponsored by the CFRP.

C.2 STANDARDS DATA

A set of 18 plutonium-uranium standards was prepared. Analyses of plutonium concentration, uranium concentration, specific gravity, and free acid normality were conducted. Uranium analyses were done by potentiometric titration, and plutonium analyses were done by amperometric titration. The density measurement was performed by weighing the solution in a tared 50-mL volumetric flask. The procedure described in Appendix E was utilized for free acid determination with the exception that 0.10 *M* oxalate was utilized. After the analysis of these standards, it was discovered that 0.15 *M* oxalate provided improved agreement between makeup and analysis. However, a complete set of standard data is not available for the 0.15 *M* oxalate analyses.

The makeup and analyzed values for the 18 standards are reported in Table C.1. Makeup values are calculated on the basis that the standard reagents, uranyl nitrate and nitric acid, have the exact molarities stated by the manufacturer and that a highly accurate weighed quantity of $\text{Pu}(\text{NO}_3)_4$ crystals dissolves in the uranyl nitrate-nitric acid reagent to form an ideal solution.

Table C.1. Plutonium-uranium nitrate solution standards

Standard ^a	Makeup				Analyzed			
	Pu (g/L)	U (g/L)	Specific gravity (g/cm ³)	Free acid (N)	Pu (g/L)	U (g/L)	Specific gravity (g/cm ³)	Free acid (N) ^b
1	19.98	80.05	1.1713	0.930	19.46	82.40	1.1702	0.97
2	20.06	79.97	1.2295	2.756	19.44	85.18	1.2288	2.79
3	19.99	80.05	1.3105	5.472	19.37	81.15	1.3101	5.60
4	40.03	59.96	1.1771	0.931	39.00	61.99	1.1783	0.94
5	39.97	59.97	1.2343	2.755	38.95	61.20	1.2346	2.82
6	39.99	59.96	1.3172	5.480	38.73	61.00	1.3149	5.56
7	59.97	39.99	1.1837	0.932	59.02	42.57	1.1843	0.95
8	60.03	40.04	1.2404	2.756	57.27	41.23	1.2403	2.79
9	60.02	40.07	1.3221	5.476	58.25	41.23	1.3202	5.54
10	80.00	320.03	1.5795	0.696	77.99	322.17	1.5769	0.72
11	79.98	319.96	1.6192	2.052	77.12	322.04	1.6161	2.06
13	160.06	240.01	1.6023	0.697	155.84	240.32	1.6027	0.72
14	159.99	240.02	1.6373	2.043	155.87	242.94	1.6400	2.05
15	159.99	240.02	1.6776	3.957	156.68	244.99	1.6901	4.13
16	240.07	160.04	1.6232	0.697	233.92	165.04	1.6274	0.73
17	240.01	159.98	1.6618	2.052	232.89	163.67	1.6598	2.09
18	240.01	159.96	1.6920	3.923	233.99	163.78	1.7092	4.05
19	400.03	0.00	1.6969	2.031	390.91	0.00	1.7032	2.22

^aStandard 12 was not analyzed due to obvious uranium insolubility.

^b0.10 M oxalate.

C.3 STATISTICAL ANALYSES OF STANDARDS DATA

A variety of statistics related to the standards data are shown in Table C.2. Some of these statistical data are necessary inputs to the procedure for deriving an acceptable design level of k -effective (Appendix F). Examining the makeup and analyzed values for plutonium, uranium, and free acid, it is immediately apparent that biases exist. The quantification of these biases is necessary in order to correctly adjust the experimental data.

Before examining Table C.2, it is useful to review some statistical assumptions. Any set of measured quantities will have a mean value and associated uncertainty. The mean value may be an accurate estimate of the true but unknown measured quantity or the measurement technique may be biased.

An idealized measurement is shown on the left side of Fig. C.1. Neither μ , s , nor b can be determined. They can, however, be estimated. The right side of Fig. C.1 shows the procedure used in this report to estimate the values of the bias and uncertainty. The bias b is assumed to be estimated by the quantity B , the difference between 0 and the mean of makeup and analyzed values with the implicit assumption being that the makeup value represents the true value. The standard error of the analyzed value ($s/\sqrt{N}$) (where N is the number of observations) is assumed to be estimated by ($\sigma/\sqrt{N}$).

A comparison of Tables C.1 and C.2 reveals that for plutonium concentration the difference between makeup and analyzed values may be correlated with the makeup value.

Table C.2. Statistical analyses of plutonium-uranium standards

Standard	(Makeup-analyzed)				100 × (Makeup-analyzed)/makeup			
	Pu (g/L)	U (g/L)	Specific gravity (10 ⁻⁴ g/cm ³)	Free acid (10 ⁻³ N)	Pu	U	Specific gravity	Free acid
1	0.52	-2.35	11	-40	2.603	-2.936	0.094	-4.301
2	0.62	-5.21	7	-34	3.091	-6.515	0.057	-1.233
3	0.62	-1.10	4	-128	3.102	-1.374	0.030	-2.334
4	1.03	-2.03	-12	-9	2.573	-3.386	-0.102	-0.967
5	1.02	-1.23	-3	-65	2.552	-2.051	-0.0243	-2.359
6	1.26	-1.04	23	-80	3.151	-1.734	0.175	-1.460
7	0.95	-2.58	-6	-18	1.584	-6.452	-0.051	-1.931
8	2.76	-1.19	1	-34	4.598	-2.972	0.008	-1.233
9	1.77	-1.16	19	-64	2.949	-2.895	0.144	-1.169
10	2.01	-2.14	26	-24	2.512	-0.669	0.165	-3.448
11	2.86	-2.08	31	-8	3.575	-0.650	0.191	-0.390
13	4.22	-0.31	-4	-23	2.637	-0.129	-0.025	-3.300
14	4.12	-0.92	-27	-7	2.575	-0.380	-0.165	-0.343
15	3.31	-0.97	-125	-173	2.069	-0.398	-0.745	-4.372
16	6.15	-5.00	-42	-33	2.562	-3.124	-0.259	-4.735
17	7.12	-3.69	20	-38	2.967	-2.306	0.120	-1.852
18	6.02	-3.82	-172	-127	2.508	-2.388	-1.016	-3.237
19	9.12	0.00	-63	-189	2.280		-0.371	-9.306
Mean	3.082	-2.166	-17.3	-60.8	2.77	-2.37	-9.86 × 10 ⁻²	-2.67
Standard error of mean	0.603	0.355	-12.7	1.33	1.50	4.56 × 10 ⁻¹	7.67 × 10 ⁻²	5.05 × 10 ⁻¹
Standard deviation	2.558	1.463	5.40 × 10 ⁻³	5.66 × 10 ⁻²	6.34 × 10 ⁻³	1.88 × 10 ⁻²	3.26 × 10 ⁻³	2.14 × 10 ⁻²

Fig. C.1. Comparison of statistics for idealized measurement with statistics available from standards data.

Linear regression analyses of the form $\text{makeup} + \text{analyzed} = a + b(\text{makeup})$ were examined. Using this formulation, the bias in each variable is no longer the constant, B , as shown in the right side of Fig. C.1 but is instead a linear function of the makeup value. The quantity, σ , in Fig. C.1 is now defined to be the square root of the residual sum of squares error. The correlation coefficients (R^2) for each of the four linear regressions are shown in Table C.3. From these values it is apparent that only the plutonium concentration exhibits a linear bias. The analysis of variance for the plutonium concentration error model is shown in Table C.4.

Table C.3. Correlation coefficients for linear regression of makeup-analysis difference versus makeup values

Measured quantity	R^2
Plutonium	0.957
Uranium	0.003
Specific gravity	0.229
Free acid	0.288

Table C.4. Analysis of variance for linear plutonium concentration error model^a

Source of variance	Degrees of freedom	Sum of squares	Mean square
Model	1	106.538	106.538
Error	16	4.731	0.296
Total	17	111.270	

$$^a R^2 = 0.957477.$$

$$\text{Intercept of model} = 0.28972.$$

$$\text{Standard error of intercept estimate} = 0.1951.$$

$$\text{Coefficient of plutonium makeup concentration variable} = 0.0237.$$

$$\text{Standard error of coefficient estimate} = 1.249 \times 10^{-3}.$$

$$\text{Root mean square error} = 0.5438.$$

Using the data contained in Tables C.2 and C.4, the biases and uncertainties in each of the experimentally measured quantities can be determined. These values are reported in Table C.5.

It is questionable whether a bias for specific gravity actually exists. The uncertainty in the measurement accounts for a significant fraction of the bias. However, the reported chemical analyses for specific gravity were corrected for this assumed bias.

Table C.5. Biases and uncertainties for analytical analyses of mixed plutonium-uranium nitrate solutions

Variable	Bias ^a	Standard error of mean	Units
Plutonium concentration	$(0.024 \times A) + 0.2967^b$	0.5438	g/L
Uranium concentration	-2.166	0.355	g/L
Specific gravity	-1.73×10^{-3}	1.27×10^{-3}	g/cm ³
Free acid	-6.08×10^{-2}	1.33×10^{-2}	mol/L

^aDefined as the quantity that must be added to the value reported by the analytical chemist in order to obtain the "true" value.

^bA is the reported value of plutonium concentration.

It is clear that there is a bias for plutonium concentration. The chemist who analyzed the standards stated that this is not surprising in view of the fact that the makeup values are based on a molecular weight of $\text{Pu}(\text{NO}_3)_4 \times \text{H}_2\text{O}$ obtained by direct firing to PuO_2 . Any nonvolatile impurities present in the plutonium stock solution from which the nitrate was made would appear to be plutonium by this method.² Since the impurities concentration would vary as the plutonium concentration varied, the linear equation shown in Table C.5 would be understandable. The constant term (0.2967) is believed to be due to the analytical method.² Additional discussion of this subject will be presented in Sect. C.3.2.

Regarding uranium concentration, there appears to be a definite bias. The chemist performing the analyses stated that this is somewhat surprising and is likely due to the analytical procedure.²

C.4 DERIVATION OF CORRECTIONS TO THE REPORTED ANALYTICAL CHEMISTRY DATA FOR CRITICAL EXPERIMENT SOLUTIONS

Table C.6 contains the analytical chemistry data for the solutions utilized in the critical experiments. These reported data must be adjusted to reflect the biases in the analytical techniques discussed in the previous section.

C.4.1 Adjustments to the Reported Uranium Concentration and Specific Gravity

Both the uranium concentration and the specific gravity were adjusted according to the biases shown in Table C.5.

C.4.2 Adjustments to the Reported Plutonium Concentration

According to the chemist who analyzed the solution standards, the linear bias in the plutonium concentration ($0.024 \times A$ from Table C.5) is due to impurities in the solution. This hypothesis was tested by analyzing three of the solution standards for impurities. The results of the analyses are shown in Table C.7. The total quantity of impurities was 0.972, 0.971, and 0.990 g/L for standards 16, 17, and 18 respectively. From Table C.1, it is seen

Table C.6. Reported analytical chemistry analyses for solutions utilized in critical experiments

Solution identification number	Pu concentration (g/L)	U concentration (g/L)	Specific gravity (g/cm ³)	Free acid normality ^a (mol/L)
918	65.00	32.00	1.1760	1.74
920	65.00	32.00	1.1760	1.74
921	65.00	32.00	1.1760	1.74
929	324.00	189.00	1.8280	7.00
930	339.00	192.00	1.8470	7.00
931	357.00	178.00	1.8530	7.50
932	280.00	188.00	1.7540	5.90
934	345.00	197.00	1.8660	6.90
941	291.00	162.00	1.7390	1.64
942	295.00	169.00	1.7450	1.49
943	297.00	170.00	1.7490	1.52
944	298.00	172.00	1.7500	1.46
945	301.00	173.00	1.7580	1.50
963	348.86	196.34	1.8632	1.28
964	348.95	196.94	1.8637	1.20
965	349.02	197.39	1.8632	1.28
970	240.04	137.18	1.6162	2.51
971	240.00	138.16	1.6162	1.34
972	239.73	137.85	1.6159	1.32
973	240.60	139.60	1.6185	1.21
974	195.10	114.10	1.5121	1.31
975	197.00	116.00	1.5178	1.26
976	196.90	116.40	1.5172	1.22
977	197.22	115.91	1.5184	1.15
983	170.30	155.80	1.5253	1.07
984	170.60	155.40	1.5256	1.09
985	169.90	155.50	1.5188	1.11
994	170.40	156.00	1.5253	1.17

^aSamples 918 to 934 analyzed using Hanford Engineering Development Laboratory oxalate procedure without complexing agent; samples 941 to 945, 971 to 973, 977, and 994 analyzed using Rockwell HVO3C procedure; 970 potassium iodate procedure; 963 to 965, 974 to 976, and 983 to 985 Appendix E procedure.

that these impurities account for 15% of the difference between makeup and analyzed values. It appears that the linear bias shown in Table C.5 is because of the analytical procedure rather than the presence of impurities.

The plutonium concentration data in Table C.1 was reanalyzed to take into account the presence of impurities. Each of the makeup plutonium concentration values was reduced by the quantity $(\text{Pu conc}/240.24) \times 0.978$ to account for impurities. The revised

Table C.7. Spark source mass spectrometry analyses of impurities in three solution standards

Element	Concentration (10 ⁻³ g/L)		
	Standard 16	Standard 17	Standard 18
Ba			≤10
Zr		≤5	
Zn		10	10
Cu		≤4	≤5
Ni	50	60	90
Co			≤4
Fe	200	300	300
Mn	10	20	20
Cr	70	100	100
Ti	≤30	≤30	≤30
Ca	100	100	100
K	20	20	20
Cl	100	40	30
S	100	50	70
P	10	10	10
Si	≤100	≤50	≤30
Al	100	100	100
Mg	50	30	20
Na	30	40	40
B	≤2	≤2	≤0.6

Source: Analyses approved by W. H. Chester, ORNL, and sent to R. T. Primm, III, ORNL, Log No. 5051, Report No. 32896, Nov. 21, 1983.

makeup values were then correlated against the analyzed plutonium values and the following relation was found:

$$\text{True Pu conc} = 1.02 \times \text{Pu reported} + 0.29 \quad (R^2 = 0.99997, \quad \text{Root mean square error} = 0.5547) . \quad (\text{C.1})$$

(Equation) C.1, *not* the bias in Table C.5, was used to correct the plutonium concentration measurements for the solutions used in the critical experiments.

C.4.3 Adjustments to the Reported Free Acid Normality

The analysis techniques for uranium, plutonium, and specific gravity did not change throughout the sequence of criticality experiments. Likewise, these same procedures were utilized for the uranium-plutonium nitrate solution standards. However, the free acid evaluation techniques used for most of the critical experiments were determined to be inappropriate. Therefore, those data cannot be adjusted in the manner used for the plutonium, uranium, and other specific gravity data [i.e., use of the bias factors from Table C.5 and

(Eq.) C.1]. The standards data must be used to develop a correlation of free acid as a function of plutonium, uranium, and specific gravity, and this correlation is then used to estimate the free acid content of the experimental solutions.

C.4.3.1 Derivation of ideal solution equation

If plutonium-uranium nitrate solutions exhibit ideal behavior, then the total volume of a solution sample would be equal to the sum of the volumes of its constituents. Likewise, the density of the solution would be equal to the sum of the concentrations of the constituents, and an equation of the form of (Eq.) C.2 could be derived as

$$\text{Density} = (\text{Pu} + \text{U}) + \text{free acid} + \text{water} + \text{impurities} . \quad (\text{C.2})$$

In previous sections of this appendix, it has been shown that density, plutonium, and uranium can be accurately measured. The data in Table C.7 indicate that total impurities can be accurately measured although the uncertainties of individual elements are quite large. The quantity of water in the acid solutions is unknown; however, the relationship of water and acid molarity can be determined by examining nitric acid chemical properties.³ Examining the density and molarity of standard nitric acid solutions, it is possible to derive (using linear regression techniques) (Eq.) C.3, which relates water content to acid molarity at a temperature of 20°C.

$$\text{Water (g/L)} = 1000.57 - 31.2594 [\text{free acid (mol/L)}] . \quad (\text{C.3})$$

Substituting (Eq.) C.3 into C.2 and inserting the proper constants to allow for variation in units, (Eq.) C.4 can be derived.

$$\begin{aligned} \text{Free acid (mol/L)} = & 31.4926 [\text{density (g/cm}^3) \\ & - \text{impurities (g/cm}^3)] - (6.41737 \times 10^{-2}) \\ & \times [\text{Pu (g/L)}] - (5.21359 \times 10^{-2}) \times [\text{U (g/L)}] \\ & - 31.5105 . \end{aligned} \quad (\text{C.4})$$

C.4.3.2 Experimentally determined free acid correlation

The makeup values in Table C.1 were used to derive an experimental correlation corresponding to (Eq.) C.4. However, the impurities concentration was subtracted from the density and plutonium concentration values so that a clean correlation could be derived. Since most of the impurities exist in solution as nitrates, some adjustment in impurity concentration has to be made for the associated nitrate ions. An investigation of the valence states of the various impurities revealed that the total concentration of impurities should be multiplied by a factor of 2 (exact estimate 2.035 rounded to 2) in order to correctly adjust the solution density for the nitrate ions associated with the impurities. The experimentally derived correlation is shown as (Eq.) C.5.

$$\begin{aligned} \text{Free acid (mol/L)} = & 34.7146 [\text{density (g/cm}^3\text{)}] \\ & - 5.33612 \times 10^{-2} \times [\text{Pu (g/L)}] - 4.5444 \times 10^{-2} \\ & \times [\text{U (g/L)}] - 35.2291 \end{aligned} \quad (\text{C.5})$$

The R^2 value for this correlation is 0.979; the standard errors of the density, plutonium, and uranium coefficient are 1.49, 2.46×10^{-2} , and 1.90×10^{-3} , respectively, and the root mean square error (which is the uncertainty in the derived free acid value) is 0.280 mol/L. Note that the uncertainty in the derived free acid value is ~ 20 times larger than the uncertainty obtained when using the procedure in Appendix E (1.33×10^{-2} from Table C.5).

C.4.3.3 Use of experimentally determined free acid correlation with critical experiment solution data

In order to use (Eq.) C.5 with the data shown in Table C.6, the plutonium concentrations, and specific gravities were first adjusted according to the procedures described in Sects. C.3.1 and C.3.2. The revised specific gravities then had to be adjusted for the concentration of impurities present in the critical experiment solutions.

Table C.8 contains the impurity concentrations for four of the critical experiment solutions. The total impurity concentrations were 4514, 4940, 4151, and 3810 $\mu\text{g/g}$ ($\text{PuO}_2 + \text{U}_3\text{O}_8$) for solution numbers 920, 941, 974, and 984 respectively. Using the data in Table A.9 and (Eq.) C.1, the corresponding concentrations of 7929, 9030, 7733, and 8596 $\mu\text{g/g}$ (Pu) are obtained. The average of these four values, 8322, was used to adjust the corrected solution densities when deriving the free acid normalities.

C.5 CORRECTED ANALYTICAL CHEMISTRY DATA FOR CRITICAL EXPERIMENT SOLUTIONS

Utilizing the correction factors described in Sect. C.3, the raw analytical chemistry values were adjusted to yield the corrected values shown in Table C.9. As a final test of the accuracy of the solution analyses, the Pu:(Pu+U) ratio was computed for each of the solutions. The results are shown in Table C.10. Solutions 983 through 994 were expected to have a significantly lower plutonium content than the other solutions. However, solutions 918 through 977 should all have the same Pu:(Pu+U) ratio. The large variations in the first seven samples indicate that either the plutonium concentration measurement, the uranium concentration measurement, or both were in error for solutions 918, 920, 921, 931, and 932. Because of this variation, the critical experiments conducted with these solutions are judged to be invalid as benchmarks. Hence data for those experiments were excluded from the main body of this report.

Table C.8. Spark source mass spectrometry analyses of impurities in four critical experiment solutions

Element	Concentration [10 ⁻⁶ g/g (PuO ₂ + U ₃ O ₈)]			
	920	941	974	984
Ag			<10	<10
Al	60	700	300	600
B	0.3	20	6	>40
Be	1		<2	<2
Ca	300	400	100	300
Cd			<2	7
Co		20	>20	20
Cr	300	200	400	300
Cu	20		<20	<20
Fe	2000	1000	1000	800
K	200	40	200	
Li	10		<1	1
Mg	300		150	100
Mn	30		90	90
Mo			60	30
Na	20		90	200
Ni	200	300	300	300
Pb			<10	<10
Si	100		<40	200
Sn			60	20
Ti	600	300	200	300
V	9	2000	>1000	60
Zn	6		250	200

Source: Personal communication from R. C. Lloyd, Pacific Northwest Laboratory, to R. T. Primm, III, ORNL May 3, 1985.

REFERENCES FOR APPENDIX C

1. J. L. Ryan et al., "Preparation of Standards for and Determination of Free Acid in Concentrated Plutonium-Uranium Solutions," *Analytical Chemistry*, pp. 1423-27 (June 1985).
2. Personal communication to R. C. Lloyd, Pacific Northwest Laboratory, from J. L. Ryan, Pacific Northwest Laboratory, Dec. 2, 1983.
3. R. C. Weast, ed., *Handbook of Chemistry and Physics*, 53rd ed., p. D-199, the Chemical Rubber Co., Cleveland, 1972.

Table C.9. Corrected analytical chemistry analyses for solutions utilized in critical experiments

Solution identification number	Pu concentration ^a (g/L)	U concentration ^b (g/L)	Specific gravity ^c (g/cm ³)	Free acid normality ^d (mol/L)
918	66.60	29.80	1.1740	0.62
920	66.60	29.80	1.1740	0.62
921	66.60	29.80	1.1740	0.62
929	331.00	187.00	1.8260	2.00
930	346.00	190.00	1.8450	1.70
931	364.00	176.00	1.8510	1.60
932	286.00	186.00	1.7520	1.90
934	352.00	195.00	1.8640	1.80
941	297.00	160.00	1.7370	2.00
942	301.00	167.00	1.7430	1.63
943	303.00	168.00	1.7470	1.61
944	304.00	170.00	1.7480	1.50
945	307.00	171.00	1.7560	1.57
963	356.14	194.17	1.8615	1.55 (1.29)
964	356.23	194.77	1.8620	1.54 (1.21)
965	356.30	195.22	1.8615	1.50 (1.29)
970	245.14	135.01	1.6145	1.59
971	245.10	135.99	1.6145	1.55
972	244.82	135.68	1.6142	1.57
973	245.70	137.40	1.6168	1.53
974	199.30	111.90	1.5104	1.48 (1.32)
975	201.20	113.80	1.5161	1.48 (1.27)
976	201.14	114.23	1.5155	1.45 (1.23)
977	201.46	113.74	1.5167	1.50
983	174.00	153.60	1.5236	1.39 (1.08)
984	174.30	153.20	1.5239	1.40 (1.10)
985	173.60	153.30	1.5171	1.20 (1.12)
994	174.10	153.80	1.5236	1.37

^a1- σ uncertainty is 0.55 g/L.

^b1- σ uncertainty is 0.36 g/L.

^c1- σ uncertainty is 1.27×10^{-3} g/cm³.

^d1- σ uncertainty is 0.28 mol/L. Values in parentheses are the measured free acid values using the procedure in Appendix E and the correction factor in Table C.5 and have a 1- σ uncertainty of 0.013 mol/L.

Table B.2. Water content of concrete reflector

Sample number	Weight loss after heating to 100°C (wt %) ^a	Weight loss after heating to 1000°C (wt %) ^a
A1	3.0	9.2
A2	2.9	9.2
B3	3.1	9.6
B4	3.3	9.8
Average	3.1 ± 0.2	9.4 ± 0.3

^aReferenced to original sample weight.**Table B.3. Calculated atom densities for the concrete reflector**

Element	Weight percent	Atom density [atoms/(b·cm)]
O	51.91	4.553×10^{-2}
Si	23.10	1.154×10^{-2}
Ca	12.00	4.201×10^{-3}
Al	4.79	2.491×10^{-3}
Fe	3.37	8.468×10^{-4}
Na	1.43	8.728×10^{-4}
H	1.05	1.462×10^{-2}
Mg	0.92	5.310×10^{-4}
K	0.72	2.584×10^{-4}
S	0.38	1.663×10^{-4}
Ti	0.33	9.667×10^{-5}

**Table C.10. Calculated Pu:(Pu+U) ratios for solutions
utilized in critical experiments^a**

Solution identification number	Pu:(Pu+U)
918	0.691
920	0.691
921	0.691
929	0.639
930	0.646
931	0.674
932	0.605
934	0.644
941	0.650
942	0.643
943	0.643
944	0.641
945	0.642
963	0.647
964	0.647
965	0.646
970	0.645
971	0.643
972	0.643
973	0.641
974	0.640
975	0.639
976	0.638
977	0.639
983	0.531
984	0.532
985	0.531
994	0.531

^aBased on values reported in Table A.10.

Appendix E

ANALYTICAL CHEMISTRY PROCEDURE FOR DETERMINATION OF FREE ACID MOLARITY OF MIXED PLUTONIUM-URANIUM NITRATE SOLUTIONS

E.1 SUMMARY

Solutions containing hydrolyzable ions (plutonium and uranium) are titrated to an equivalence end point in a solution of ammonium oxalate. The oxalate ion forms a complex with uranium and plutonium to reduce their interference in the titration. An automatic potentiometric titrator is used to record the titration curve.

E.2 APPLICATION

1. Material: acid solutions containing plutonium-uranium.
2. Range: 0.5 to 6 *M* HNO₃, 0 to 400 g/L HM, Pu:(Pu+U) of 0.2 to 0.6
3. Reliability: see Appendix A.
4. Interferences: ions that form equilibria with hydrogen and hydrolyzable ions not complexed by oxalate.

E.3 APPARATUS

1. Automatic potentiometric titration system, Brinkmann Instruments Model E636.
2. Combination electrode, Orion Model 81-02.
3. Pipets, Eppendorf, adjustable 25 to 100 μ L with disposable tips.
4. Magnetic stirrer.
5. Stir bars, Teflon covered.

E.4 REAGENTS

1. Ammonium oxalate solution, 0.15 *M*.
2. Standardized NaOH, $\sim$ 0.1 *N*.
3. Standardized HNO₃ for spike, 1 and 6 *N*.

E.5 STANDARDS

1. Uranyl nitrate standard, 300 g/L uranium and $\sim 3\text{ N HNO}_3$.
2. Uranium-plutonium standard series as described in Appendix A.

E.6 SAFETY

1. Plutonium-bearing materials are radioactive and toxic.
2. Precautions to avoid contamination of the laboratory and personnel.
3. Observe general laboratory safety rules.
4. Follow manufacturer's instructions for operation of titrator.

E.7 QUALITY CONTROL

1. Calibration includes, but is not limited to, titration of standard nitric acid solutions versus standardized sodium hydroxide using multiple aliquot sizes.
2. Control is established by analysis of prepared uranium-plutonium acid standards and a uranyl nitrate standard.

E.8 ANALYSIS

1. Set up titration system according to manufacturer's instructions, including calibration of electrode with pH 4 and/or pH 7 buffers. Do not change instrument settings once optimum conditions are established.
2. Analyze standard acid utilizing three aliquot sizes (see no. 4). Determine slope and intercept for these titrations.
3. Determine reagent blank by titrating oxalate solution (see no. 4).
4. Titration of sample.
 - a. Add 20 mL of oxalate solution to titration vessel containing stirring bar.
 - b. Immerse electrode and delivery tip in solution and start stirrer.
 - c. Pipet sample into vessel and initiate titration. Use at least three different aliquot sizes. (Maximum allowable plutonium-uranium 400 g/L.) A measured acid spike may be added prior to sample addition to check recovery.
 - d. After titration is complete, remove electrode and delivery tip and wash with oxalate solution. Rinse titration vessel and replace. Proceed to next sample.
 - e. Record volumes of sample added, spike, and titrant consumed.

Appendix D

CRITICAL EXPERIMENT MEASUREMENTS FOR WHICH ANALYTICAL CHEMISTRY DATA WERE JUDGED TO BE INADEQUATE

Table D.1. Mixed uranium-plutonium nitrate solution critical
experiment measurements
(Previous analytical chemistry)

PNL experiment number	Solution identification number	Geometry	Reflector ^a	Diameter or thickness ^b (cm)	Height (cm) ^b
1	918	cylinder	water	35.39	22.3
2	920	cylinder	steel and water	35.39	23.5
3	920	cylinder	water	68.68	15.7
4	920	cylinder	concrete	35.39	23.3
5	921	slab	bare ^c	17.50	55.4
6	921	slab	water	14.90	34.5
7	921	slab	water	12.40	61.7
12	931	cylinder	water	35.39	32.5
14	932	cylinder	bare	35.39	57.4
18	932	cylinder	water	35.39	29.7

^aFor the steel and water experiment, the steel thickness was 1.27 cm (0.5 in.).

^b1- σ uncertainty is ± 0.16 cm (0.06 in.).

^cThe 0.95-cm- (0.37-in.) thick Plexiglas decontamination hood walls provide significant reflection.

5. Calculations. (Calculations are performed by the titrator after insertion of appropriate information.)

$$N_a = \frac{(V_b)(N_b)}{V_s} , \quad (E.1)$$

where

N_a = normality of acid (sample)
 V_b = volume standard base used, in mL
 N_b = normality of standard NaOH
 V_s = volume sample used, in mL.

The volume of base used for the titration as given by the titrator is the amount required to titrate to the inflection (equivalence) point. A recording of the shape of the curve (pH or mV versus volume base) is also produced and should be recorded.

Computer program input for small, concrete reflected cylinder

```

=CSAS2
PNL EXPT NUMBER 10
27GROUPNDF4 4 29 1 INFHOMMEDIUM 0 0
H 1 0.0 .488322E-01 END
N 1 0.0 .547170E-02 END
O 1 0.0 .412810E-01 END
FE 1 0.0 .664294E-05 END
U-233 1 0.0 .638438E-07 END
U-234 1 0.0 .537909E-07 END
U-235 1 0.0 .352535E-05 END
U-236 1 0.0 .140611E-06 END
U-238 1 0.0 .477054E-03 END
PU-238 1 0.0 .262663E-06 END
PU-239 1 0.0 .797770E-03 END
PU-240 1 0.0 .685133E-04 END
PU-241 1 0.0 .448750E-05 END
PU-242 1 0.0 .516643E-06 END
AM-241 1 0.0 .180711E-05 END
N 2 0.0 3.97877E-5 END
O 2 0.0 1.06742E-5 END
SS304 3 1.0 END
O 4 0.0 4.553E-2 END
SI 4 0.0 1.154E-2 END
CA 4 0.0 4.201E-3 END
AL 4 0.0 2.491E-3 END
FE 4 0.0 8.468E-4 END
NA 4 0.0 8.728E-4 END
H 4 0.0 1.462E-2 END
MG 4 0.0 5.310E-4 END
K 4 0.0 2.584E-4 END
S 4 0.0 1.663E-4 END
TI 4 0.0 9.667E-5 END
KENO INPUT
30.0 103 300 3 3 1 1 3 0
BOX TYPE 1
CYLINDER 3 0.4 0.635 0.0 -0.5
CYLINDER 2 1.0 0.635 0.0 -0.5
CYLINDER 3 1.3 0.635 0.0 -0.5
CYLINDER 2 2.6 0.635 0.0 -0.5
CUBOID 3 44.77 -44.92 49.68 -49.68 0.635 0.0 -0.5
BOX TYPE 2
CYLINDER 3 0.4 21.11 0.0 -0.5
CYLINDER 2 1.0 21.11 0.0 -0.5
CYLINDER 3 1.3 21.11 0.0 -0.5
CYLINDER 2 2.6 21.11 0.0 -0.5
CYLINDER 3 3.05 21.11 0.0 -0.5
CYLINDER 2 18.415 21.11 0.0 -0.5
CYLINDER 4 43.815 21.11 0.0 -0.5
CUBOID 2 44.77 -44.77 49.53 -49.53 21.11 0.0 -0.5

```

Computer program input for small, concrete reflected cylinder (continued)

```

CUBOID 3 44.77 -44.92 49.68 -49.68 21.11 0.0 -0.5
BOX TYPE 3
CYLINDER 1 17.78 0.0 -0.5
CYLINDER 2 17.78 107.63 0.0 -0.5
CYLINDER 3 17.859 107.709 -0.079 -0.5
CYLINDER 2 18.415 107.709 -0.079 -0.5
CYLINDER 4 43.815 107.709 -0.079 -0.5
CUBOID 2 44.77 -44.77 49.53 -49.53 107.709 -0.079 -0.5
CUBOID 3 44.77 -44.92 49.68 -49.68 107.709 -0.079 -0.5
END GEOMETRY
1 1 1 1 1 1 1 1 0
2 1 1 1 1 1 2 2 1 0
3 1 1 1 1 1 3 3 1 1
END

```

Computer program input for large, water reflected cylinder

```

=CSAS2
PNL EXPT NUMBER 44
27GROUPNDF4 4 19 1 INFHOMMEDIUM 0 0
H 1 0.0 .565869E-01 END
N 1 0.0 .319956E-02 END
O 1 0.0 .383308E-01 END
FE 1 0.0 .325677E-05 END
U-233 1 0.0 .277372E-07 END
U-234 1 0.0 .355097E-07 END
U-235 1 0.0 .274618E-05 END
U-236 1 0.0 .978022E-07 END
U-238 1 0.0 .385050E-03 END
PU-238 1 0.0 .131788E-06 END
PU-239 1 0.0 .398916E-03 END
PU-240 1 0.0 .356348E-04 END
PU-241 1 0.0 .222986E-05 END
PU-242 1 0.0 .367228E-06 END
AM-241 1 0.0 .124073E-05 END
N 2 0.0 3.97877E-5 END
O 2 0.0 1.06742E-5 END
SS304 3 1.0 END
H2O 4 1.0 END
KENO INPUT
30.0 103 300 3 3 1 1 3 0
BOX TYPE 1
CYLINDER 3 0.4 0.635 0.0 -0.5
CYLINDER 2 1.0 0.635 0.0 -0.5
CYLINDER 3 1.3 0.635 0.0 -0.5
CYLINDER 2 2.6 0.635 0.0 -0.5
CUBOID 3 44.77 -44.92 49.68 -49.68 0.635 0.0 -0.5
BOX TYPE 2
CYLINDER 3 0.4 21.11 0.0 -0.5
CYLINDER 2 1.0 21.11 0.0 -0.5
CYLINDER 3 1.3 21.11 0.0 -0.5
CYLINDER 2 2.6 21.11 0.0 -0.5
CYLINDER 3 3.05 21.11 0.0 -0.5
CUBOID 4 44.77 -44.77 49.53 -49.53 21.11 0.0 -0.5
CUBOID 3 44.77 -44.92 49.68 -49.68 21.11 0.0 -0.5
BOX TYPE 3
CYLINDER 1 34.29 0.0 -0.5
CYLINDER 2 34.29 107.63 0.0 -0.5
CYLINDER 3 34.3687 107.709 -0.079 -0.5
CUBOID 4 44.77 -44.77 49.53 -49.53 107.709 -0.079 -0.5
CUBOID 3 44.77 -44.92 49.68 -49.68 107.709 -0.079 -0.5
END GEOMETRY
1 1 1 1 1 1 1 1 1 0
2 1 1 1 1 1 1 2 2 1 0
3 1 1 1 1 1 1 3 3 1 1
END

```

Computer program input for water reflected slab

```

=CSAS2
PNL EXPT NUMBER 22
27GROUPNDF4 6 21 1 INFHOMMEDIUM 0 0
H 1 0.0 .483691E-01 END
N 1 0.0 .561769E-02 END
O 1 0.0 .414826E-01 END
FE 1 0.0 .678314E-05 END
U-233 1 0.0 .655239E-07 END
U-234 1 0.0 .552064E-07 END
U-235 1 0.0 .361813E-05 END
U-236 1 0.0 .144311E-06 END
U-238 1 0.0 .489608E-03 END
PU-238 1 0.0 .267218E-06 END
PU-239 1 0.0 .811604E-03 END
PU-240 1 0.0 .697014E-04 END
PU-241 1 0.0 .456532E-05 END
PU-242 1 0.0 .525602E-06 END
AM-241 1 0.0 .183844E-05 END
N 2 0.0 3.97877E-5 END
O 2 0.0 1.06742E-5 END
SS304 3 1.0 END
H2O 4 1.0 END
PLEXIGLASS 5 1.0 END
REG-CONCRETE 6 1.0 END
KENO INPUT
30.0 103 300 3 27 12 12 2 0
BOX TYPE 1
CUBOID 4 5.13715 -5.13715 5.13715 -5.13715 11.43000 0.0 -0.5
CUBOID 3 5.29590 -5.29590 5.29590 -5.29590 11.58750 0.0 -0.5
CUBOID 1 5.29590 -5.29590 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 2
CUBOID 4 5.13715 -5.13715 5.13715 -5.13715 11.58750 0.15748 27*0.5
CUBOID 3 5.29590 -5.29590 5.29590 -5.29590 11.58750 0.0 -0.5
BOX TYPE 3
CUBOID 4 5.13715 -5.13715 1.21738 0.0 11.43000 0.0 -0.5
CUBOID 3 5.29590 -5.29590 1.21738 -0.15748 11.58750 0.0 -0.5
CUBOID 1 5.29590 -5.29590 1.21738 -0.15748 26.48750 0.0 -0.5
BOX TYPE 4
CUBOID 4 5.13715 -5.13715 1.21738 0.0 11.58750 0.15748 -0.5
CUBOID 3 5.29590 -5.29590 1.21738 -0.15748 11.58750 0.0 -0.5
BOX TYPE 5
CUBOID 4 5.13715 -5.13715 10.27430 1.21738 11.43000 0.0 -0.5
CUBOID 3 5.29590 -5.29590 10.43178 1.21738 11.58750 0.0 -0.5
CUBOID 2 5.29590 -5.29590 10.43178 1.21738 26.48750 0.0 -0.5
BOX TYPE 6
CUBOID 4 5.13715 -5.13715 10.27430 1.21738 11.58750 0.15748 -0.5
CUBOID 3 5.29590 -5.29590 10.43178 1.21738 11.58750 0.0 -0.5
BOX TYPE 7
CUBOID 4 5.13715 -5.13715 5.13715 -5.13715 11.43000 0.0 -0.5
CUBOID 3 5.29590 -5.29590 5.29590 -5.29590 11.58750 0.0 -0.5

```

Computer program input for water reflected slab (continued)

```

CUBOID 2 5.29590 -5.29590 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 8
CUBOID 4 2.10612 0 5.29590 -5.29590 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 5.29590 -5.29590 11.58750 0.0 -0.5
CUBOID 1 2.26487 0 5.29590 -5.29590 26.48750 0.0 -0.5
CUBOID 3 2.26487 -0.15748 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 9
CUBOID 4 2.10612 0 5.29590 -5.29590 11.58750 0.15748 -0.5
CUBOID 3 2.26487 -0.15748 5.29590 -5.29590 11.58750 0.0 -0.5
BOX TYPE 10
CUBOID 4 2.10612 0 1.21738 -0.15748 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 1.21738 -0.15748 11.58750 0.0 -0.5
CUBOID 1 2.26487 0 1.21738 -0.15748 26.48750 0.0 -0.5
CUBOID 3 2.26487 -0.15748 1.21738 -0.15748 26.48750 0.0 -0.5
BOX TYPE 11
CUBOID 4 2.10612 0 1.21738 -0.15748 11.58750 1.21738 -0.5
CUBOID 3 2.26487 -0.15748 1.21738 -0.15748 11.58750 0.0 -0.5
BOX TYPE 12
CUBOID 4 2.10612 0 10.43178 1.21738 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 10.43178 1.21738 11.58750 0.0 -0.5
CUBOID 2 2.26487 0 10.43178 1.21738 26.48750 0.0 -0.5
CUBOID 3 2.26487 -0.15748 10.43178 1.21738 26.48750 0.0 -0.5
BOX TYPE 13
CUBOID 4 2.10612 0 10.43178 1.21738 11.58750 0.15748 -0.5
CUBOID 3 2.26487 -0.15748 10.43178 1.21738 11.58750 0.0 -0.5
BOX TYPE 14
CUBOID 4 2.10612 0 5.29590 -5.29590 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 5.29590 -5.29590 11.58750 0.0 -0.5
CUBOID 2 2.26487 0 5.29590 -5.29590 26.48750 0.0 -0.5
CUBOID 3 2.26487 -0.15748 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 15
CUBOID 4 2.26487 0.15748 5.29590 -5.29590 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 5.29590 -5.29590 11.58750 0.0 -0.5
CUBOID 1 2.26487 0 5.29590 -5.29590 26.48750 0.0 -0.5
CUBOID 3 2.42362 0 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 16
CUBOID 4 2.26487 0.15748 5.29590 -5.29590 11.58750 0.15748 -0.5
CUBOID 3 2.42362 0 5.29590 -5.29590 11.58750 0.0 -0.5
BOX TYPE 17
CUBOID 4 2.26487 0.15748 1.21738 -0.15748 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 1.21738 -0.15748 11.58750 0.0 -0.5
CUBOID 1 2.26487 0 1.21738 -0.15748 26.48750 0.0 -0.5
CUBOID 3 2.42362 0 1.21738 -0.15748 26.48750 0.0 -0.5
BOX TYPE 18
CUBOID 4 2.26487 0.15748 1.21738 -0.15748 11.58750 0.15748 -0.5
CUBOID 3 2.42362 0 1.21738 -0.15748 11.58750 0.0 -0.5
BOX TYPE 19
CUBOID 4 2.26487 0.15748 10.43178 1.21738 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 10.43178 1.21738 11.58750 0.0 -0.5
CUBOID 2 2.26487 0 10.43178 1.21738 26.48750 0.0 -0.5

```

Computer program input for water reflected slab (continued)

```

CUBOID 3 2.42362 0 10.43178 1.21738 26.48750 0.0 -0.5
BOX TYPE 20
CUBOID 4 2.26487 0.15748 10.43178 1.21738 11.58750 0.15748 -0.5
CUBOID 3 2.42362 0 10.43178 1.21738 11.58750 0.0 -0.5
BOX TYPE 21
CUBOID 4 2.26487 0.15748 5.29590 -5.29590 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 5.29590 -5.29590 11.58750 0.0 -0.5
CUBOID 2 2.26487 0 5.29590 -5.29590 26.48750 0.0 -0.5
CUBOID 3 2.42362 0 5.29590 -5.29590 26.48750 0.0 -0.5
BOX TYPE 22
CUBOID 4 5.29590 -5.29590 0 -2.26487 11.43000 0 -0.5
CUBOID 3 5.29590 -5.29590 0 -2.26487 11.58750 0 -0.5
CUBOID 1 5.29590 -5.29590 0 -2.26487 26.48750 0 -0.5
CUBOID 3 5.29590 -5.29590 0 -2.42362 26.48750 0 -0.5
BOX TYPE 23
CUBOID 4 5.29590 -5.29590 0 -2.26487 11.58750 0.15748 -0.5
CUBOID 3 5.29590 -5.29590 0 -2.42362 11.58750 0 -0.5
BOX TYPE 24
CUBOID 4 2.26487 0 0 -2.26487 11.43000 0 -0.5
CUBOID 3 2.26487 0 0 -2.26487 11.58750 0 -0.5
CUBOID 1 2.26487 0 0 -2.26487 26.48750 0 -0.5
CUBOID 3 2.26487 -0.15748 0 -2.42362 26.48750 0 -0.5
BOX TYPE 25
CUBOID 4 2.26487 -0.15748 0 -2.42362 11.58750 0.15748 -0.5
CUBOID 3 2.26487 -0.15748 0 -2.42362 11.58750 0 -0.5
BOX TYPE 26
CUBOID 4 2.26487 0 0 -2.26487 11.43000 0.0 -0.5
CUBOID 3 2.26487 0 0 -2.26487 11.58750 0.0 -0.5
CUBOID 1 2.26487 0 0 -2.26487 26.48750 0.0 -0.5
CUBOID 3 2.42362 0 0 -2.42362 26.48750 0.0 -0.5
BOX TYPE 27
CUBOID 4 2.42362 0 0 -2.42362 11.58750 0.15748 -0.5
CUBOID 3 2.42362 0 0 -2.42362 11.58750 0 -0.5
CORE BOUNDARY 0 134.21910 23.45513 123.85675 15.51513
53.96250 15.88750 -0.5
CUBOID 3 134.21910 23.45513 124.01423 15.51513
53.96250 15.88750 -0.5
CUBOID 4 141.92 0.0 124.01423 0.0 69.85 0.0 -0.5
CUBOID 2 141.92 0.0 143.51 0.0 69.85 0.0 -0.5
CUBOID 3 143.555 -0.635 143.51 -0.635 70.485 -0.635 -0.5
CUBOID 2 158.795 -86.0425 258.349 -101.283 173.99
-34.925 -0.5
CUBOID 5 159.748 -86.995 258.349 -101.283 174.942
-35.8775 -0.5
CUBOID 6 159.748 -86.995 258.349 -192.723 174.942
-35.8775 -0.5
END GEOMETRY
1 2 11 1 2 6 1 1 1 1 0
2 2 11 1 2 6 1 2 2 1 0
3 2 11 1 7 7 1 1 1 1 0
4 2 11 1 7 7 1 2 2 1 0
5 2 11 1 8 8 1 1 1 1 0
6 2 11 1 8 8 1 2 2 1 0
7 2 11 1 9 12 1 1 1 1 0
2 2 11 1 9 12 1 2 2 1 0

```


Appendix F
SAMPLE CODE INPUT

Computer program input for water reflected slab (continued)

8 1 1 1 2 6 1 1 1 1 0
9 1 1 1 2 6 1 2 2 1 0
10 1 1 1 7 7 1 1 1 1 0
11 1 1 1 7 7 1 2 2 1 0
12 1 1 1 8 8 1 1 1 1 0
13 1 1 1 8 8 1 2 2 1 0
14 1 1 1 9 12 1 1 1 1 0
9 1 1 1 9 12 1 2 2 1 0
15 12 12 1 2 6 1 1 1 1 0
16 12 12 1 2 6 1 2 2 1 0
17 12 12 1 7 7 1 1 1 1 0
18 12 12 1 7 7 1 2 2 1 0
19 12 12 1 8 8 1 1 1 1 0
20 12 12 1 8 8 1 2 2 1 0
21 12 12 1 9 12 1 1 1 1 0
16 12 12 1 9 12 1 2 2 1 0
22 2 11 1 1 1 1 1 1 1 0
23 2 11 1 1 1 1 2 2 1 0
24 1 1 1 1 1 1 1 1 1 0
25 1 1 1 1 1 1 2 2 1 0
26 12 12 1 1 1 1 1 1 1 0
27 12 12 1 1 1 1 2 2 1 1
END

Appendix G*

METHOD TO ESTABLISH SUBCRITICALITY

The evaluated multiplication factor of the system shall be equal to or less than an established maximum allowable multiplication factor k_a ,

$$k_s < k_a \quad (G.1)$$

where k_s = the evaluated multiplication factor including any necessary allowance for statistical uncertainties in the calculational technique such as in Monte Carlo calculations.

The maximum allowable multiplication factor shall be calculated from the expression

$$k_a = k_c - \Delta k_u - \Delta k_m \quad (G.2)$$

where $k_c = k_{\text{eff}}$ that results from the calculation of the benchmark experiments using a particular calculational method. The value represents a combination of theoretical technique and numerical data.

Δk_u = the uncertainty in the benchmark experiments.

Δk_m = the value required to assure an accepted margin of subcriticality.

Δk_u shall include both uncertainties in the benchmark experiments as well as uncertainties in the bias, which result from extrapolation of the benchmark experiments into the range of parameters encountered in the design.

Δk_m shall provide an adequate margin of subcriticality under the operating limitations and shall be no less than 0.02.

In the absence of information that justifies a small margin of subcriticality, a value of 0.05 shall be assumed for Δk_m for the design.

*From American National Standards Institute, Standard Number ANSI/ANS-57.7-1981, "Design Criteria for an Independent Spent Fuel Storage Installation (Water Pool Type)."

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