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Criticality Experiments with Mixed Plutonium and Uranium Nitrate Solution at a Plutonium Fraction of 0.2 and 1.0 in Annular Cylindrical Geometry

R. C. Lloyd

April 1988

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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CRITICALITY EXPERIMENTS WITH MIXED PLUTONIUM AND
URANIUM NITRATE SOLUTION AT A PLUTONIUM FRACTION
OF 0.2 AND 1.0 IN ANNULAR CYLINDRICAL GEOMETRY

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Data Development

Pacific Northwest Laboratory
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SUMMARY

A series of critical experiments was completed with mixed plutonium-uranium solutions having $\text{Pu}/(\text{Pu} + \text{U})$ ratios of approximately 0.2 and 1.0. These experiments were a part of the Criticality Data Development Program between the United States Department of Energy (USDOE), and the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan. A complete description of, and data from, the experiments are included in this report. The experiments were performed with mixed plutonium-uranium solutions in annular cylindrical geometry. The measurements were made with a water reflector. The central region included a concrete, polyethylene or void annular cylindrical insert. Interior to the insert was a stainless steel bottle containing plutonium-uranium solution or a void region. In one experiment the central region was filled with a solid cadmium-covered polyethylene insert. The concentration of the solution in the annular region was varied from 61 to 489 g (Pu + U)/liter. The ratio of plutonium to total heavy metal (plutonium plus uranium) was 0.22 or 0.97 for all experiments.

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CRITICALITY EXPERIMENTS WITH MIXED PLUTONIUM AND URANIUM NITRATE SOLUTION AT A PLUTONIUM FRACTION OF 0.2 AND 1.0 IN ANNULAR CYLINDRICAL GEOMETRY

1.0 INTRODUCTION

The design and operation of facilities for recycling Fast Breeder Reactor (FBR) fuels involves criticality conditions which are much different from those encountered in the light water reactor fuel cycle. Conditions are encountered in plant operations with fissionable materials that involve complex equipment shapes, high plutonium content in solution with uranium, and neutron absorbing materials that affect criticality. Experimental criticality data are required for validation of the calculations and nuclear data used in facility design, operational procedures and related licensing activities to ensure freedom from criticality accidents.

In August, 1983 the U. S. Department of Energy (DOE) and the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan entered into an agreement to study such nuclear criticality aspects as they related to the development of fast breeder reactor recycling technology. This arrangement was developed through the DOE and PNC Agreement in the Field of Liquid Metal-Cooled Fast Breeder Reactors. Prior to this Joint Memorandum of Agreement (MOA) for Nuclear Criticality Data Development Programs, DOE had initiated an experimental program at the DOE Hanford Critical Mass Laboratory to provide basic criticality data on plutonium-uranium systems in support of the U. S. Liquid Metal Fast Breeder Reactor Program. Under this MOA, PNC has promoted and enlarged the DOE Program to cover areas of mutual interest as well as areas of specific interest to PNC.

Some computer codes for criticality calculations have been developed and applied to FBR fuel cycle facility designs. Application of these codes, however, and the associated cross-section libraries, result in uncertainties on the particular conditions for FBR fuel and need further verification based on actual critical experimental data. Therefore, experimental data are needed which will allow verification of codes and cross-section data to

minimize the uncertainties so that facility safety, efficiency, and reliability can be enhanced. The verification of criticality evaluation methods is the subject of regulatory licensing activity. These data have application whenever mixtures of plutonium and uranium exist, in the head-end of a fuel reprocessing plant through the first solvent extraction cycle, in storage vessels and during product conversion when a coprocessing scheme is used.

This report contains a description of, and data from, the criticality experiments conducted with mixed plutonium-uranium solutions at $\text{Pu}/(\text{Pu} + \text{U})$ ratios of approximately 0.2 and 1.0. The experiments were performed in annular cylindrical geometry. The measurements were made with a water reflector. The central region included a concrete, polyethylene or void annular cylindrical insert. Interior to the insert was a stainless steel bottle containing plutonium-uranium solution or void region. In one critical experiment the central region was filled with a solid cadmium-covered polyethylene insert. The concentration of the solution in the annular region was varied from 61 to 489 g (Pu + U)/liter.

2.0 DESCRIPTION OF EXPERIMENTAL ASSEMBLIES

This section includes the general description of the experimental assemblies used in obtaining the criticality data for this report.

2.1 GENERAL DESCRIPTION OF THE SOLUTION SYSTEM

An existing experimental system, previously used for solution experiments at the Critical Mass Laboratory, was used for conducting the measurements and providing the data for this report. The solution system is located in the critical assembly room. The addition of solution to the experimental vessel is remotely made from the control room. The layout of equipment in the critical assembly room is shown in Figure 2.1.

The critical assembly room is 10.67 meters square and has a ceiling height of 6.4 meters. The side walls are composed of 1.52 meters thick concrete. The concrete ceiling and floor are each 0.61 meters thick.

The containment hood (Hood 1) was located 1.83 meters from the north wall of the room. The west side of the hood, which faces the wall containing the DS and DM tanks was located 1.52 meters from that wall. The cylindrical vessel assembly positioned south of Hood 1 will be discussed in Section 2.2.

A schematic showing the piping connections between the three experimental vessels is shown in Figure 2.2. This piping arrangement allows critical experiments to be conducted with the same solution in each of three vessels without changing vessels, when desired. The annular cylinder was used in this series of experiments.

The fill, dump and manometer lines enter the bottom of the vessel through the dump valve system. The dump valve and lines are designed to drain the fissile solution even if the addition were continued. The vessel is connected to the dump valve pedestal by a Marmon flange connection which provides a leak tight connection.

FIGURE 2.1 Floor Plan Schematic of the Critical Assembly Room

FIGURE 2.2 Piping Schematic for the Three Experimental Vessels

2.2 CYLINDRICAL VESSEL ASSEMBLY

A photograph of the cylindrical vessel system is shown in Figure 2.3. This system contains two cylindrical vessels. The vessel used for the experiments in this report was an annular cylinder and it was located on the right side of the system. The control and safety blade mechanisms are mounted above the vessel and can be seen in the figure. The reflector tank serves to contain water for the water reflected experiments. Windows of polycarbonate (Lexan) were installed on the front for access to the tank and for visual inspection. This reflector tank was fabricated of carbon steel (CS). A schematic of the experimental vessel arrangement giving dimensions is shown in Figure 2.4 for all experiments except experiment 108 where a 53 cm diameter boiler tube-type vessel replaced the small cylindrical vessel.

The annular cylindrical vessel was fabricated of Type 304L stainless steel. The vessel had an outside diameter of 53.3 cm and an inside diameter of 25.4 cm. The wall thickness was 0.074 cm. The control and safety blades are external to the vessel and are withdrawn during the neutron flux determination during the critical approach measurement. A schematic of the annular cylindrical vessel is shown in Figure 2.5 and a photograph of the top of the system is shown in Figure 2.6. This shows the annular concrete insert and the bottle of solution located in the central region. Figure 2.7 is a photograph showing the annular tank and bottle insert prior to installation for critical experiments.

A schematic of the concrete insert is shown in Figure 2.8. Four concrete inserts were used in the critical experiments with each insert having a different amount of B_4C mixed with the concrete. The as-built dimensions for the inside and outside diameters of the four inserts were not statistically different from each other so the average of the diameters is shown in the figure. The figure also includes other dimensions based on the fabrication specifications.

FIGURE 2.3 Photograph of the Cylindrical Vessel System

FIGURE 2.4 Schematic of Dual Cylinder Tank with Annular and Small Cylinder Mounted

FIGURE 2.5 Schematic of the Annular Cylindrical Vessel with Concrete Insert and Bottle

FIGURE 2.6 Photograph of the Annular Assembly

FIGURE 2.7 Photograph Showing Annular Tank and Bottle

FIGURE 2.8 Schematic of Annular Concrete Insert

ORNL-DWG 88-5557A

FIGURE 2.9 Schematic of Solid Cadmium-Covered Polyethylene Insert

Note: Dimensions in cm

FIGURE 2.10 Schematic of Annular Cadmium-Covered Polyethylene Insert

Schematics of the solid cadmium-covered polyethylene insert and the annular cadmium-covered polyethylene insert are shown in Figures 2.9 and 2.10. The figures include as-built dimensions for the diameters and cadmium thicknesses as well as other dimensions based on fabrication specifications. The polyethylene used in the fabrication of these inserts was considered "pure polyethylene." Typical elemental analysis for this material indicates hydrogen and carbon weight percentages of 14.28 and 85.72 respectively and a density of 0.90 g/cm^3 .

The experiments with the annular cylinder were conducted with the reflector tank containing water. The reflector tank was filled to the level below the top of the annular vessel as given in Section 3.0. The distance between the annular cylinder (portion below the annular region) and the bottom of the reflector tank (inside surface) is $40.1 \pm 0.1 \text{ cm}$.

Engineering drawings were provided in PNL-5768 (Lloyd 1986) for the cylindrical vessel system and in PNL-6119 (Lloyd 1988) for the annular tank assembly. These drawings contain detailed dimensions used for fabrication.

A listing of the engineering drawings are provided in Appendix A for the cylindrical vessel system. A listing of the engineering drawings are provided in Appendix B for the annular tank assembly.

3.0 EXPERIMENTAL RESULTS

This section provides the results of the experiments including a description of the measurement techniques involved in obtaining the data.

3.1 CRITICALITY MEASUREMENT TECHNIQUES

The critical heights for the experiments reported herein were determined using the critical approach method (Clayton 1985). In this critical approach method, neutron flux measurements are made as the height of solution is incrementally increased. Inverse count rate is plotted versus solution height. At delayed critical condition the neutron count rate approaches infinity so that the inverse count rate approaches zero. By extrapolation of the inverse multiplication curves to zero value, the critical height is determined for the system. The neutron flux is routinely taken on three boron-lined proportional detectors located near the experimental vessel. The data from the three counters extrapolate to essentially identical values for solution height at near critical values. The computer calculated least square fits of the inverse multiplication curves, used in determining the critical value for the height in each experimental assembly, are included in Appendix C.

3.2 CRITICALITY DATA

The criticality data for this report were obtained from June 1986 - November 1987, when fourteen experiments were completed using the annular cylindrical vessel. The data are summarized in Table 3.1. The chemical analyses data for the (Pu + U) nitrate solutions given in these tables were provided by the Chemical and Analysis Section of the Westinghouse Hanford Company, Hanford Engineering Development Laboratory (HEDL) from samples of solution supplied to them. The sample analyses methods and descriptive titles are given in Table 3.2. The critical heights were calculated by a least squares fit to the inverse neutron multiplication data from three neutron detectors. (Computer printout provided in Appendix C). The ²⁴¹Am content for each sample analyzed, the analysis date and the experiments covered by that sample are given in Table 3.3. The isotopic analyses values

TABLE 3.1 Criticality Measurements with (Pu + U)
Nitrate Solution in the Annular Cylinder

<u>Run Date</u>	<u>Case Number</u>	<u>Experiment Number</u>	<u>Insert^(a) Description</u>	<u>Reflector^(b)</u>	<u>Sample Number</u>	<u>Pu (g/liter)</u>	<u>U (g/liter)</u>	<u>Density^(c) (g/cm³)</u>	<u>Free Acid (M)</u>	<u>Critical Height (cm)</u>
06/20/86	31	087	Part 26	Water	1188	102.19	365.20	1.6803	0.77	48.55
06/27/86	31	087S	Part 26	Water	1191	102.69	364.88	1.6780	0.77	48.99
07/02/86	38	088	Part 28	Water	1192	102.65	364.59	1.6781	0.77	(d)
07/22/86	33	091	Part 24/B-2	Water	1197	103.37	363.66	1.6776	0.78	27.67
06/18/87	34	092	Part 25/B-2	Water	1202	106.30	373.33	1.6953	0.65	37.19
06/26/87	35	093	Part 27/B-2	Water	1203	107.91	379.55	1.7034	0.66	51.10
07/08/87	36	094	B-2	Water	1208	108.27	380.41	1.7040	0.62	32.86
07/15/87	60	095	Part 26/B-3	Water	1210	195.61	6.5	1.4390	3.28	27.51
07/20/87	61	096	Part 26/B-3	Water	1211	110.13	3.8	1.2598	1.95	25.69
07/22/87	62	097	Part 26/B-3	Water	1212	58.30	2.3	1.1467	1.14	28.94
07/28/87	46	098	Part 26/B-2	Water	1213	72.74	247.33	1.4739	0.53	39.58
08/05/87	39	099	Part 29/B-2	Water	1214	73.64	250.30	1.4788	0.52	79.18
08/12/87	53	100	Part 28	Water	1215	74.25	251.64	1.4828	0.53	104.62
11/12/87	50	108	Part 26/B-2	Water	1232	47.08	161.72	1.3134	0.52	45.09

(a) Part 24 Annular Concrete Insert 0 wt% B₄C
Part 25 Annular Concrete Insert 1 wt% B₄C
Part 26 Annular Concrete Insert 2 wt% B₄C

Part 27 Annular Concrete Insert 6 wt% B₄C
Part 28 Solid Polyethylene with Cd Cover
Part 29 Annular Polyethylene with Cd Cover

Bottle B-2, contained 9.86 ± 0.02 liters of (Pu + U) solution (sample number 1195). The Pu concentration was 103.36 g/liter, the U concentration was 363.30 g/liter. The density was 1.6777 g/cm³. The free acid was 0.77 M. The bottle was filled to 59.4 ± 0.2 cm.

Bottle B-3, contained 6.11 ± 0.02 liters of (Pu + U) solution (sample number 1209). The Pu concentration was 194.92 g/liter. The U concentration was 5.1 g/liter. The density was 1.4362 g/cm³. The free acid was 3.31 M. The bottle was filled to 36.8 ± 0.2 cm.

(b) The water level was 24.1 cm below the top of the vessel for experiments 087, 087S, 091 - 099 and 1.9 cm for experiments 088, 100 and 108.

(c) Density measured at 23°C.

(d) Criticality could not be achieved within vessel height limitations.

TABLE 3.2 Chemical Analyses Methods

<u>Measurement</u>	<u>Method Title</u>	<u>Date of Approval</u>
Plutonium	Plutonium by Automated Amperometric Titration. (30.3)	03/18/85
Uranium	Uranium by Automated Potentiometric Titration. (30.8)	02/05/86
Impurities	Impurities by Emission Spectroscopy: Direct Reader. (40.13)	06/11/85
^{241}Am	Americium-241 by Anion Exchange and Alpha Analysis. (40.16)	05/14/75
Free Acid	Determination of Free Acid in Uranium/Plutonium Solutions. (Using an improved oxalate method) (40.22)	02/04/86
Density	Density of Solutions. (Using Mettler/ Paar Density Meter) (40.23)	02/05/86
Isotopic	Isotopic Composition of Plutonium and Uranium by Mass Spectroscopy. (30.6)	09/27/78
Impurities	Impurities by Spark Source Mass Spectrometer. (40.15)	05/22/75
Impurities	ICP Analysis (SP-7B)	04/01/86

(a) The numbers in brackets are HEDL's method numbers.

TABLE 3.3 Chemical Analysis Values for Americium-241

<u>Sample Number</u>	<u>^{241}Am (ug/ml)</u>	<u>Analysis Date</u>
1195	553	08/22/86
1197	561	08/22/86
1202	595	07/08/87
1209	919	08/13/87
1212	300	08/13/87
1214	406	08/27/87
1232	265	12/16/87

Sample 1195 is the analysis for solution in the bottle B-2

Sample 1197 covers experiments 087, 087S, 088, 091

Sample 1202 covers experiments 092 - 094

Sample 1209 is the analysis for solution in the bottle B-3 and covers experiment 095. The result can be applied to experiment 096 following adjustment based on Pu ratio

Sample 1212 covers experiment 097

Sample 1214 covers experiments 098 - 100

Sample 1232 covers experiment 108.

for the plutonium and uranium of the experiments are given in Table 3.4. Table 3.5 provides information on the temperatures of the critical assembly room (CAR), the dump mix tank (DM) and the water reflector. Also in Table 3.5, the reflector water level and the position of the bottom of the control and safety blades are given. (Reference is the vessel top).

Appendix D provides data on the chemical analyses for the impurities found in the (Pu + U) nitrate solutions.

The chemical analyses of the reflector water samples are given in Appendix E.

The composition of the concrete insert used in these experiments is given in Appendix F. Also provided are sample analyses for the boron and certificate of analysis provided by the vendor for the B_4C . Weight changes of the sample of concrete are also given in Appendix F. The weights of the ingredients used to make up the concrete insert are given.

3.3 SOURCES OF ERROR

It is practically 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 combination of variables which might have a reactivity effect which is a significant fraction of the typical uncertainty in a particular KENO calculation. This evaluation was done for the significant measurements involved in earlier experiments and reported (Primm 1986). From that analysis it was found that the primary uncertainty that caused significant error was from the free acid values. Since those measurements, a study was made and a free acid analysis method developed and reported (Ryan 1985). This has significantly reduced uncertainties in the analysis for free acid. Further work provided free acid standards so that the analyses could be confirmed.

The evaluation of uncertainties by Primm included the critical height, plutonium concentration, uranium concentration, density, free acid and composition of reflectors. It was recognized by Primm that the procedure used to derive uncertainties due to experimental and chemical analysis measurements were likely to over estimate the value of each parameter.

TABLE 3.4 Isotopic Analyses Values of Pu and U^(a)

	Sample 1188 ^(b)	Sample 1192 ^(c)	Sample 1194 ^(d)	Sample 1202 ^(e)	Sample 1209 ^(f)	Sample 1214 ^(g)	Sample 1232 ^(h)
Pu ⁽ⁱ⁾	07/10/86	07/23/86	07/23/86	07/07/87	08/12/87	08/27/87	12/14/87
238	0.029 ± 0.001	0.029 ± 0.002	0.028 ± 0.002	0.025 ± 0.002	0.027 ± 0.002	0.031 ± 0.003	0.046 ± 0.005
239	91.12 ± 0.05	91.12 ± 0.04	91.13 ± 0.04	91.12 ± 0.04	91.57 ± 0.04	91.10 ± 0.04	91.11 ± 0.04
240	8.30 ± 0.05	8.32 ± 0.04	8.31 ± 0.04	8.34 ± 0.04	7.94 ± 0.04	8.35 ± 0.04	8.33 ± 0.04
241	0.449 ± 0.004	0.442 ± 0.004	0.445 ± 0.004	0.425 ± 0.008	0.393 ± 0.006	0.426 ± 0.006	0.419 ± 0.02
242	0.094 ± 0.001	0.091 ± 0.001	0.092 ± 0.001	0.089 ± 0.002	0.068 ± 0.001	0.095 ± 0.002	0.095 ± 0.001
U ^(j)	07/08/86	07/28/86	07/28/86	07/08/87	08/13/87	08/28/87	12/14/87
238	99.229 ± 0.007	99.236 ± 0.007	99.235 ± 0.007	99.242 ± 0.006	97.103 ± 0.010	99.238 ± 0.005	99.234 ± 0.008
236	0.053 ± 0.002	0.052 ± 0.002	0.052 ± 0.002	0.052 ± 0.002	0.467 ± 0.008	0.053 ± 0.002	0.053 ± 0.005
235	0.710 ± 0.006	0.706 ± 0.006	0.705 ± 0.006	0.698 ± 0.005	2.292 ± 0.010	0.702 ± 0.004	0.705 ± 0.008
234	0.007 ± 0.001	0.007 ± 0.001	0.007 ± 0.001	0.007 ± 0.002	0.138 ± 0.004	0.008 ± 0.001	0.008 ± 0.001

(a) All values given in wt%

(b) Sample 1188 is for experiments 087 and 087S

(c) Sample 1192 is for experiment 088

(d) Sample 1194 is for the solution in bottle B-2 and experiment 091

(e) Sample 1202 is for experiments 092 - 094

(f) Sample 1209 is for the solution in bottle B-3 and experiments 095 - 097

(g) Sample 1214 is for experiments 098 - 100

(h) Sample 1232 is for experiment 100

(i) Date of Pu analysis

(j) Date of U analysis.

TABLE 3.5 Information on Temperature, Reflector Level,
and Control and Safety Blade Position

Experiment Number	Temperature °C			Reflector Level Distance Below Vessel Top (cm)	Control and Safety Blade Distance Below Vessel Top (cm)
	CAR	DM Tank	Reflector		
087	22.8	28.4	21.4	24.1	5
087S	25.8	28.5	23.2	24.1	5
088	23.7	29.4	23.2	1.9	5
091	23.2	27.6	23.8	24.1	5
092	25.2	30.38	22.7	24.1	5
093	25.9	30.53	23.9	24.1	5
094	25.3	32.09	23.2	24.1	5
095	26.8	30.33	21.9	24.1	5
096	24.6	27.97	25.1	24.1	5
097	25.2	27.19	25.5	24.1	5
098	26.1	28.38	25.6	24.1	5
099	26.2	28.86	21.4	24.1	5
100	27.4	29.57	23.6	1.9	5
108	23.5	23.57	22.5	1.9	5

The latest estimated values of uncertainties are listed in the following table:

TABLE 3.6 Estimate of Measurement Uncertainties

Pu Concentration	$\pm 0.2\%$
U Concentration	$\pm 0.2\%$
Density	$\pm 0.0003 \text{ g/cm}^3$
Free Acid	$\pm 0.04 \text{ M}$
Critical Height	$\pm 1.6 \text{ mm}$

The uncertainty values for the chemical analyses was provided by M. C. Burt of the Chemical and Analysis Section of the Westinghouse Hanford Company, Hanford Engineering Development Laboratory. The critical height uncertainty is given as 1.6 mm though the least square fitting of approach data for three counting systems would indicate a smaller value as reasonable. The 1.6 mm is the smallest unit on the sight tube.

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- L. N. Terry for typing, proofreading and guidance in preparation of this report.

5.0 REFERENCES

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APPENDIX A

A LISTING OF ENGINEERING DRAWINGS FOR THE CYLINDRICAL VESSEL SYSTEM

APPENDIX A

A LISTING OF ENGINEERING DRAWINGS FOR THE CYLINDRICAL VESSEL SYSTEM

CFRP Assembly H-2-33856, Sheet 1 of 5

CFRP H₂O Tank and Cover H-2-33856, Sheet 2 of 5

CFRP Process Tanks H-2-33856, Sheet 3 of 5

CFRP Tank Covers and Shield H-2-33856, Sheet 4 of 5

CFRP Dump Valve H-2-33856, Sheet 5 of 5

APPENDIX B

A LISTING OF ENGINEERING DRAWINGS FOR THE ANNULAR TANK ASSEMBLY

APPENDIX B

A LISTING OF ENGINEERING DRAWINGS FOR THE ANNULAR TANK ASSEMBLY

Annular Tank with FTR Pin Lattices, H-2-95741, Sheet 1 of 6
Annular Tank with FTR Pin Lattices, H-2-95741, Sheet 2 of 6
Annular Tank with FTR Pin Lattices, H-2-95741, Sheet 3 of 6
Annular Tank with FTR Pin Lattices Details, H-2-95741, Sheet 4 of 6
Annular Tank with FTR Pin Lattices Inserts Details, H-2-95741, Sheet 5 of 6
Annular Tank with FTR Pin Lattices Details, H-2-95741, Sheet 6 of 6

APPENDIX C

LEAST SQUARE FITS OF THE CRITICAL APPROACH DATA

APPENDIX C

LEAST SQUARE FITS OF THE CRITICAL APPROACH DATA

The extrapolated values given are for solution height in the annular tank. The solution height values used in these plots are given in inches.

FIGURE C.1 Least Square Fits for CFRP-PNC 087

FIGURE C.2 Least Square Fits for CFRP-PNC 087S

FIGURE C.3 Least Square Fits for CFRP-PNC 091

FIGURE C.4 Least Square Fits for CFRP-PNC 092

FIGURE C.5 Least Square Fits for CFRP-PNC 093

FIGURE C.6 Least Square Fits for CFRP-PNC 094

FIGURE C.7 Least Square Fits for CFRP-PNC 095

FIGURE C.8 Least Square Fits for CFRP-PNC 096

FIGURE C.9 Least Square Fits for CFRP-PNC 097

FIGURE C.10 Least Square Fits for CFRP-PNC 098

FIGURE C.11 Least Square Fits for CFRP-PNC 099

FIGURE C.12 Least Square Fits for CFRP-PNC 100

FIGURE C.13 Least Square Fits for CFRP-PNC 103

APPENDIX D

CHEMICAL ANALYSES DATA OF THE IMPURITIES IN (Pu + U) NITRATE SOLUTIONS

APPENDIX D

CHEMICAL ANALYSES DATA OF THE IMPURITIES IN (Pu + U) NITRATE SOLUTIONS

The measurement uncertainty for the Spark Source Mass Spectrographic analyses is very large. These measurements are performed primarily for qualitative purposes. The measurement uncertainty for the Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP) method is about ± 25 percent.

The Spark Source Mass Spectrographic analysis results for samples 1192, 1194, 1195, 1197, and 1202 are included. These sample results cover experiments 087, 087S, 088, 091 - 094, and bottle B-2. Sample 1208 was analyzed by the more accurate ICP method and the results apply to experiments 087, 087S, 088, 091 - 094, and bottle B-2. Sample 1212 was analyzed by the ICP method and the results apply directly to experiment 097. The ICP results for sample 1212 can be applied to experiments 095, 096, and bottle B-3 by multiplying the results by the ratio of plutonium concentrations. Sample 1215 was analyzed by the ICP method and the results apply to experiments 098 - 100, and 108.

TABLE D.1 Spectrographic Analysis Report - 1192 and 1194

[illegible]

LOG NO. <u>55,56</u>		Hanford Engineering Development Laboratory		SPARK SOURCE MASS SPECTROGRAPHIC ANALYSIS REPORT			
MATERIAL <u>Mixed Oxide</u>							
SUBMITTED BY <u>J. SPASOFF</u>		SUBMITTER'S NO. <u>SEE BELOW</u>		ANALYZED BY <u>RK</u>		DATE REPORTED <u>9/9/66</u>	
ELE- MENT	<u>55</u>	<u>56</u>	ELE- MENT	<u>55</u>	<u>56</u>	ELE- MENT	<u>55</u>
Li	<u>0.2</u>	<u>0.6</u>	Ga	<u>80</u>	<u>80</u>	Pm	
Be	<u>*</u>	<u>*</u>	Ge	<u><5</u>	<u><5</u>	Sm	
B	<u>1</u>	<u>1</u>	As	<u><5</u>	<u><5</u>	Eu	
F			Rb			Gd	
Na	<u>30</u>	<u>30</u>	Sr	<u>8</u>	<u>5</u>	Tb	
Mg	<u><2</u>	<u>2</u>	Y			Dy	
Al	<u>100</u>	<u>100</u>	Zr	<u>10</u>	<u>7</u>	Ho	
Si	<u>20</u>	<u>20</u>	Nb			Er	
P	<u>3</u>	<u>3</u>	Mo	<u>2</u>	<u>1</u>	Tm	
S			Ru ^{Tc}	<u>20</u>	<u>60</u>	Yb	
Cl			Rh			Lu	
K	<u>10</u>	<u>10</u>	Pd			Hf	
Ca	<u>20</u>	<u>60</u>	Ag	<u>*</u>	<u>*</u>	Ta	
Sc			Cd			W	
Ti	<u>10</u>	<u>10</u>	In			Re	
V	<u><3</u>	<u><3</u>	Sn			Os	
Cr	<u>70</u>	<u>70</u>	Sb			Ir	
Mn	<u>20</u>	<u>7</u>	Cs			Pt	
Fe	<u>100</u>	<u>100</u>	Ba	<u>40</u>	<u>80</u>	Au	
Co	<u>0.5</u>	<u>0.3</u>	La	<u><3</u>	<u><3</u>	Hg	
Ni	<u>30</u>	<u>10</u>	Ce			Th	<u>20</u> <u>60</u>
Cu	<u>60</u>	<u>40</u>	Pr			Pb	<u>2</u> <u>1</u>
Zn	<u>10</u>	<u>10</u>	Nd			Bi	

TYPE OF ANALYSIS		
☐ QUALITATIVE ☐ PARTS PER MILLION ☐ PERCENT ☐ _____ APPROX PRECISION $\pm$ FACTOR _____	☒ SEMIQUANTITATIVE ☒ PARTS PER MILLION ☐ PERCENT ☐ _____ APPROX PRECISION $\pm$ <u>FACTOR 3</u>	☐ QUANTITATIVE ☐ PARTS PER MILLION ☐ PERCENT ☐ _____ APPROX PRECISION $\pm$ FACTOR _____

REMARKS: <u>55 = 1195 - A</u> <u>56 = 1197 - A</u>	REPORT APPROVED <u>[Signature]</u>
--	------------------------------------

* INTERFERENCE

TABLE D.3 Spectrographic Analysis Report - 1202

LOG NO. <u>#53</u>		Hanford Engineering Development Laboratory		SPARK SOURCE MASS SPECTROGRAPHIC ANALYSIS REPORT	
MATERIAL <u>P₄O₁₀</u>					
SUBMITTED BY <u>J. SPASOFF</u>		SUBMITTER'S NO. <u>1202 AB</u>		ANALYZED BY <u>JK</u>	
				DATE REPORTED <u>7/10/87</u>	
ELE- MENT		ELE- MENT		ELE- MENT	
Li		Ga		Pm	
Be	*	Ge		Sm	
B		As		Eu	
F	<u>6</u>	Rb		Gd	
Na	<u>30</u>	Sr		Tb	
Mg	<u>50</u>	Y		Dy	
Al	<u>100</u>	Zr		Ho	
Si	<u>200</u>	Nb		Er	
P	<u>30</u>	Mo		Tm	
S	<u>300</u>	Ru		Yb	
Cl	<u>10</u>	Rh		Lu	
K		Pd		Hf	
Ca	<u>20</u>	Ag	*	Ta	
Sc		Cd	<u>10</u>	W	
Ti	<u>100</u>	In		Re	
V		Sn		Os	
Cr	<u>30</u>	Sb		Ir	
Mn	<u>30</u>	Cs		Pt	
Fe	<u>300</u>	Ba		Au	
Co		La		Hg	
Ni	<u>80</u>	Ce		Th	<u>30</u>
Cu	<u>30</u>	Pr		Pb	
Zn		Nd		Bi	
TYPE OF ANALYSIS					
☐ QUALITATIVE		☒ SEMIQUANTITATIVE		☐ QUANTITATIVE	
☐ PARTS PER MILLION		☒ PARTS PER MILLION		☐ PARTS PER MILLION	
☐ PERCENT		☐ PERCENT		☐ PERCENT	
☐ _____		☐ _____		☐ _____	
APPR'X PRECISION ± FACTOR _____		APPR'X PRECISION ± <u>Factor 3</u>		APPROX PRECISION ± FACTOR _____	
REMARKS:					
<u>* Interference</u>					
REPORT APPROVED

TABLE D.4 Inductively Coupled Plasma Spectroscopy
Analysis (mg/liter)

<u>Element</u>	<u>Sample Number</u>		
	<u>1208</u>	<u>1212</u>	<u>1215</u>
Al	86.2	87.9	88.7
B	3.9	1.3	3.0
Ba	2.5	2.6	2.0
Ca	50.8	30.2	41.0
Cd	4.8	2.1	4.3
Ce	3.2	0.7	4.2
Cr	70.0	45.1	57.1
Cu	11.2	2.6	12.0
Dy	0.7	0.3	0.8
Fe	297.7	150.7	248.7
Gd	<1	<1	<1
K	11.0	4.0	5.0
La	0.6	0.4	0.7
Li	0.2	0.2	0.2
Mg	10.4	6.9	8.6
Mn	13.5	6.6	11.4
Mo	1.6	1.1	1.2
Na	33.6	19.6	20.6
Nd	1.2	0.7	1.3
Ni	50.7	29.6	41.4
Rh	13.4	2.8	14.0
Ru	4.6	1.9	4.2
Si	8.3	14.6	16.5
Sr	0.3	0.2	0.2
Te	(a)	(a)	(a)
Ti	21.8	9.2	17.4
Zn	4.0	<1	3.6
Zr	4.8	0.6	3.9

(a) Element not detected

APPENDIX E

CHEMICAL ANALYSES DATA OF THE REFLECTOR WATER SAMPLES

APPENDIX E

CHEMICAL ANALYSES DATA OF THE REFLECTOR WATER SAMPLES

The sample for experiment 087 also covers experiment 087S. The sample for experiment 090 also covers experiment 091.

TABLE E.1 Water Sample Analyses-087, 088 and 090

HANFORD ENVIRONMENTAL
HEALTH FOUNDATION

August 25, 1986

CO-10801

Pacific Northwest Laboratory
209E/14
200 East Area

Attn: R. C. Lloyd

WATER SAMPLES ANALYSES

The results of the analysis of the three water samples received July 11, 1986, are following. Analyses were done in accordance with Standard Methods for the Analysis of Water and Wastewater, 16th Ed.

Parameter	SAMPLES		
	Exp 086,087	088	090
pH	7.13	7.46	7.97
Total Alkalinity (mg/L)	35.8	40.1	40.9
HCO ₃ Alkalinity [as CaCO ₃] (mg/L)	29	37	37
CO ₃ Alkalinity [as CaCO ₃] (mg/L)	<0.5	<0.5	<0.5
Total dissolved solids (mg/L)	83	62	66
Fluoride (mg/L)	<0.1	<0.1	<0.1
Chloride (mg/L)	3.6	2.1	2.5
Nitrate [as N] (mg/L)	<0.05	<0.05	<0.05
Sulfate (mg/L)	18.5	11.7	12.8
Cadmium (mg/L)	0.0006	0.0014	0.0010
Copper (mg/L)	<0.05	0.52	<0.05
Chromium (mg/L)	<0.005	<0.005	<0.005
Iron (mg/L)	0.109	0.089	0.080
Lead (mg/L)	<0.005	0.015	<0.005
Manganese (mg/L)	<0.01	<0.01	<0.01
Zinc (mg/L)	0.79	0.82	0.65

If you have any questions concerning this report, please contact Environmental Health Sciences.

P. A. Thurman

P. A. Thurman
Environmental Health Sciences

kw

TABLE E.2 Water Sample Analyses-092, 093 and 094

HANFORD ENVIRONMENTAL
HEALTH FOUNDATION

CO 11769

December 2, 1987

Pacific Northwest Laboratory
209-E Building, 200-E Area

Attn: R. C. Lloyd

WATER SAMPLES ANALYSES

The results of the analyses of the three water samples received July 17, 1987, are following. Analyses were done in accordance with Standard Methods for the Analysis of Water and Wastewater, 16th Ed.

Parameter	092	093	094
pH	7.0	7.3	7.5
Total alkalinity (mg/L)	45	47	31
HCO ₃ alkalinity (as CaCO ₃) (mg/L)	42	42	30
CO ₃ alkalinity (as CaCO ₃) (mg/L)	<0.5	<0.5	<0.5
Total dissolved solids (mg/L)	67	63	85
Fluoride (mg/L)	0.19	0.16	0.14
Chloride (mg/L)	2.4	1.1	0.97
Nitrate (as N) (mg/L)	<0.05	<0.05	<0.05
Sulfate (mg/L)	12.5	8.8	8.8
Cadmium (mg/L)	0.001	<0.0005	<0.0005
Copper (mg/L)	<0.05	<0.05	<0.05
Chromium (mg/L)	<0.005	<0.005	<0.005
Iron (mg/L)	<0.03	<0.03	0.07
Lead (mg/L)	<0.005	<0.005	<0.005
Manganese (mg/L)	0.012	<0.01	<0.01
Zinc (mg/L)	0.75	<0.05	<0.05

If there are questions concerning this report, please contact us.

P. A. Thurman

P. A. Thurman
Environmental Health Sciences

jt

TABLE E.3 Water Sample Analyses-095,
096, 097, 098, 099 and 100

HANFORD ENVIRONMENTAL
HEALTH FOUNDATION

CO 11769

December 2, 1987

Pacific Northwest Laboratory
209-E Building, 200-E Area

Attn: R. C. Lloyd

WATER SAMPLES ANALYSES

The results of the analyses of the water samples received August 11 and 27, 1987, are attached. Analyses were done in accordance with Standard Methods for the Analysis of Water and Wastewater, 16th Ed.

If there are questions concerning this report, please contact us.

P. A. Thurman

P. A. Thurman
Environmental Health Sciences

jt

TABLE E.3 (contd)

R. C. Lloyd, PNL

CO 11769

Parameter	095	096	097	098	099	100	101	103
pH	7.4	7.5	7.7	7.2	8.3	7.3	7.6	7.7
Total alkalinity (mg/L)	63	66	63	51	49	51	52	51
HCO ₃ alkalinity (as CaCO ₃) (mg/L)	62	65	62	48	44	46	50	48
CO ₃ alkalinity (as CaCO ₃) (mg/L)	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Total dissolved solids (mg/L)	53	52	76	142	142	140	137	136
Fluoride (mg/L)	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Chloride (mg/L)	0.70	0.76	0.56	1.12	1.14	1.15	1.12	1.00
Nitrate (as N)(mg/L)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Sulfate (mg/L)	8.6	9.2	8.8	9.3	8.2	9.4	11.1	9.2
Cadmium (mg/L)	<0.0005	<0.0005	<0.0005	0.0019	0.0006	<0.0005	0.0008	0.0007
Copper (mg/L)	<0.05	<0.05	0.12	<0.05	<0.05	<0.05	<0.05	<0.05
Chromium (mg/L)	0.010	<0.005	<0.005	<0.005	0.009	<0.005	<0.005	<0.005
Iron (mg/L)	<0.03	0.103	0.148	<0.03	0.064	<0.03	0.181	0.279
Lead (mg/L)	<0.005	0.006	0.028	<0.005	0.080	<0.005	0.040	<0.005
Manganese (mg/L)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.011	0.011
Zinc (mg/L)	0.055	2.10	1.20	<0.05	2.41	0.071	3.13	0.071

TABLE E.4 Water Sample Analysis-108

HANFORD ENVIRONMENTAL
HEALTH FOUNDATION

CO 12325

February 11, 1988

Pacific Northwest Laboratory
209-E Building, 200-E Area

Attn: R. C. Lloyd

WATER SAMPLES ANALYSES

The results of the analyses of the seven water samples received January 6, 1988, are attached. Analyses were done in accordance with Standard Methods for the Analysis of Water and Wastewater, 16th ed.

If there are questions concerning this report, please contact us.

P. A. Thurman
Environmental Health Sciences

jt

Attach.

TABLE E.4 (contd)

R. C. Lloyd, PNL

CO 12325

Parameter	Exp.106, 107, 108	Exp. 109, 110	Exp. 111	Exp. 112, 113	Exp. 114	Exp.115	Exp.116
pH	7.0	7.0	7.5	7.2	7.3	7.1	7.4
Total alkalinity (mg/L)	55	55	56	56	54	57	56
HCO ₃ alkalinity (as CaCO ₃) (mg/L)	50	50	56	52	50	52	51
CO ₃ alkalinity (as CaCO ₃) (mg/L)	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Total dissolved solids (mg/L)	86	85	89	79	76	101	105
Fluoride (mg/L)	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Chloride (mg/L)	0.91	0.91	0.92	0.97	0.98	0.95	0.94
Nitrate (as N) (mg/L)	<0.05	<0.05	.088	<0.05	<0.05	<0.05	<0.05
Sulfate (mg/L)	10.6	10.7	10.7	10.7	11.7	11.5	10.5
Cadmium (mg/L)	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005	<0.0005
Copper (mg/L)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
Chromium (mg/L)	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Iron (mg/L)	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03
Lead (mg/L)	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Manganese (mg/L)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Zinc (mg/L)	0.08	0.06	0.08	0.07	0.42	0.38	0.08

APPENDIX F

COMPOSITION OF THE CONCRETE INSERTS

APPENDIX F

COMPOSITION OF THE CONCRETE INSERTS

In March 1985, four annular concrete inserts were fabricated for use in critical experiments at the CML in conjunction with an annular cylinder. The four inserts are nearly identical except for varying amounts of B_4C . The components used in making each of the inserts are shown in Table F.1. It should be recognized that the fraction of each component in the cured concrete will change since the water content of the concrete will change as the concrete sets. Also, the water content shown only represents the pure water added, and does not include the amount of water intrinsically part of the other components, especially the aggregate.

The Hanford Engineering Development Laboratory performed an isotopic analysis of the boron used in the B_4C . The analysis indicates that ^{10}B constitutes 20.02 ± 0.1 atom percent, with the remainder being ^{11}B . The certificate of analysis from the supplier of the B_4C is shown in Table F.2.

The density of each insert was determined to be 2.19 ± 0.05 g/cc, based on the weight (1.5 Kg) and volume (0.7 liter) of a sample of one of the concrete inserts. This value was confirmed by analysis of the total weight and volume of each insert. Also, the concrete inserts were weighed at various times between October 1985 and January 1988 and the weights did not change more than 0.2 percent.

The elemental composition of each insert was determined by the Inductively Coupled Plasma-Atomic Emission Spectra Method performed by PNL. Samples were fused with both K and Na/Zr hydroxides to obtain most of the elemental concentrations. The oxygen, hydrogen, and carbon contents could not be obtained with this analysis. The results of these analyses are shown in Table F.3.

TABLE F.1 Mixing Components Used to Fabricate Concrete Inserts

Component	Insert			
	0% B ₄ C (lb)	1% B ₄ C (lb)	2% B ₄ C (lb)	6% B ₄ C (lb)
Water	28.16	27.42	27.86	27.57
Cement	56.22	56.22	55.47	56.22
Aggregate	154.60	155.00	150.00	150.00
B ₄ C	0.00	1.55	4.64	15.45

TABLE F.2 Certificate of Analysis for B_4C

NOAH CHEMICAL DIV. NOAH INDUSTRIAL CORP.

87 GAZZA BOULEVARD, FARMINGDALE, NEW YORK 11735 TELEPHONE (516) 293-3336 TELEX 645175

Certificate of Analysis

PRODUCT: BORON CARBIDE, B_4C , 99.5%, -325 mesh

LOT: 37855

ANALYSIS: Average particle size by Fisher = 5.95 microns

C	(theo.)	=	21.74%
C	(found)	=	22.57%
B	(theo.)	=	78.26%
B	(found)	=	78.0%

Al	0.01%
Ca	0.01
Cu	0.005
Fe	0.06
Mg	5 ppm
Mn	5 ppm
Si	.04%
Sn	5 ppm
Zn	.004%

TABLE F.3 Concentration of Elements in
Annular Cylinder Concrete Inserts

Element	Insert Composition (wt%)			
	0% B ₄ C	1% B ₄ C	2% B ₄ C	6% B ₄ C
Al	4.82	4.90	4.82	4.53
B	0.02	0.58	1.56	5.02
Ba	0.07	0.07	0.07	0.06
Ca	15.4	15.3	13.9	14.0
Cu	0.01	0.01	0.01	0.01
Fe	4.39	4.47	4.35	4.16
K	0.54	0.64	1.52	1.36
Li	0.02	0.01	0.02	0.02
Mg	1.12	1.15	1.15	1.14
Mn	0.07	0.07	0.07	0.07
Na	1.89	1.59	1.48	1.45
Si	21.8	22.4	22.1	20.6
Sr	0.04	0.04	0.03	0.03
Ti	0.57	0.58	0.58	0.55
Zr	0.04	0.06	0.10	0.04

The carbon content can be inferred from the amount of boron present, since it can be assumed that this element normally is not present. From the boron contents shown in Table F.3, the B_4C content in the 0% B_4C , 1% B_4C , 2% B_4C , and 6% B_4C are 0.0, 0.72, 1.97 and 6.39 wt%, respectively. These evaluations accounted for the 0.02 wt% naturally occurring boron in concrete.

The remaining elements in the concrete inserts are oxygen and hydrogen. The hydrogen is assumed to be associated with the water content in the concrete. Approximately 7 g samples of pulverized concrete from each of the insets were dried in incremental steps to 1000°C. After heating to a given temperature, the sample was cooled in a desiccator and then weighed. The results of this analysis are shown in Table F.4. The PNL staff conducting the measurements observed the following during the analysis: At 750°C, the samples containing B_4C showed a reddish tint with the 6% B_4C sample showing a significant reddish color. At 1000°C, the sample with 6% B_4C had definitely glassified while the sample with 2% B_4C showed some characteristic of forming a glass. From the table of weight changes with temperature, the 2% B_4C and 6% B_4C samples showed a definite weight gain at 750°C. This weight change and color change is probably due to the decomposition of B_4C to B_2O_3 and CO_2 . The reddish color noted in the samples at this temperature is probably due to the formation of Fe_2O_3 . Also, if CO_2 is formed in cement, this gas is probably trapped as $CaCO_3$ by the CaO in the cement. With a source of B_2O_3 from the B_4C decomposition, a borosilicate-type glass forms at about 1000°C.

The amount of water in the concrete can be deduced from the data in Table F.4. Since the samples are both losing weight by water evaporation and gaining weight by material decomposition, some assumptions are necessary to evaluate the total water content in each of the inserts.

The amount of oxygen in the inserts can be deduced by subtracting the elemental compositions in Table F.3, and the carbon and hydrogen contents from 100.

TABLE F.4 Analysis for Water Content
of Concrete Inserts

Temperature (°C)	Concrete Sample Weight (g)			
	0% B ₄ C	1% B ₄ C	2% B ₄ C	6% B ₄ C
25	7.8207	8.8389	6.8866	7.4325
110	7.5881	8.6029	6.7275	7.3098
250	7.4781	8.4858	6.5091	7.2027
500	7.3459	8.4039	6.4549	7.2311
750	7.1731	8.2570	6.4736	7.6231
1000	7.1554	8.2312	6.4499	7.5940

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