R. E. ROTHE AND I. OH, "BENCHMARK CRITICAL EXPERIMENTS ON HIGHENRICHED URANYL NITRATE SOLUTION SYSTEMS," NUCL. TECHNOL. 41: 207225 (1978).

## EDITOR:

## ROY G. POST

Department of Nuclear Engineering
University of Arizona
Tucson, Arizona 85721

## ASSOCIATE EDITORS:

## KARL WIRTZ

Kernforschungszentrum Karlsruhe
Institut für Neutronenphysik und Reaktortechnik 75 Karlsruhe, Postfach 3640, Germany
RYOHEI KIYOSE
University of Tokyo
Department of Nuclear Engineering
7-3-1 Hongo, Bunkyo-ku
Tokyo, Japan

## VOLUME 41 <br> NOVEMBER, DECEMBER, MID-DECEMBER 1978

# BENCHMARK CRITICAL EXPERIMENTS ON HIGH-ENRICHED URANYL NITRATE SOLUTION SYSTEMS 

ROBERT E. ROTHE and INKI OH Rockwell International Atomics International Division, Rocky Flats Plant, P. O. Box 464<br>Golden, Colorado 80401

KEYWORDS: uranyl nitrates, solutions, configuration, critical size, tanks, highly enriched uranium, data, height, neutron reflectors

Received February 21, 1978
Accepted May 18, 1978

Seventy-six benchmark critical conditions are reported. Both material and geometry properties are so well determined as to reduce greatly any contribution to a theoretical/experimental discrepancy attributable to the experiment. The program uses uranyl nitrate solution with the uranium enriched to $93.17 \%{ }^{235} U$. The concentration ranges from 54.89 to 369.96 g U/l. Unreflected experiments are reported, as well as measurements within thick-walled cubical reflector shells composed of such common materials as concrete and plastic.

For experiments using a single tank, the diameter of the tank ranged from 27.88 to 50.69 cm , and arrays of up to 16 cylinders have containers of two diameters: 16.12 and 21.12 cm . Containers composed of aluminum or stainless steel are studied. For all these parameters, the critical heights range from 17.13 to 110.20 cm .

## I. INTRODUCTION

Precise criticality data on systems whose geometry and material compositions are well known can be used as reference experiments against which both present-day and future calculational techniques can be tested. The need for such reference (benchmark) data, especially for certain ranges of parameters, has been pointed out many times in the literature. ${ }^{1-6}$ Specifically, one frequently used calculational method consistently underestimates $k_{\text {eff }}$ by a considerable amount for unreflected systems containing concentrated, highly enriched uranyl nitrate solution. ${ }^{6-12}$ Another objective of reference experiments is to describe experiments so completely and accurately that no part of a discrepancy between calculated
and experimental results can be laid to that information.

The present paper answers these needs by reporting criticality data for high-enriched uranyl nitrate solution systems under minimally reflected conditions and fully reflected by such common materials as concrete and methyl methracrylate plastic. Other parameters include the uranium concentration within the solution, the diameter of the aluminum or stainless-steel tank containing the solution, and the number of containers-one or an array. The geometrical placement and elemental compositions of all materials within considerable distance of the fissile solution are accurately specified.

The data reported here are the first from a series of programs designed to provide reference criticality data for a wide variety of parameters of interest to the nuclear industry. Planned future programs will use low-enriched uranium in differing forms, so the combined programs will provide a broad parameter base for testing calculational models against experimental evidence.

## II. SUMMARY OF EXPERIMENTAL RESULTS

A total of 76 experimentally determined roomtemperature critical heights are reported in Tables I and II for high-enriched uranium solution in various containers and under various conditions of neutron reflection. Table I pertains to single tanks, while the other gives results for square or rectangular arrays of tall cylinders.

These principal-result tables are keyed to other tables throughout the paper, wherein equipment and materials are detailed to an extent necessary for benchmark data. For example, a complete geometrical description of the $33.01-\mathrm{cm}$-diam aluminum tank, found in the first columns of Table I, can be obtained from Table VII using the diameter

TABLE
Critical Height (in centimetres) of a Single

| Tank |  | Minimally Reflected ${ }^{\text {a }}$ |  |  | 25.7-cm-thick Concrete Shell ( $\sim 122-\mathrm{cm}$ inside dimension) |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | At Center ${ }^{\text {b }}$ | In Corner ${ }^{\text {c }}$ |  |
| Material |  <br> (cm) |  |  |  | $\begin{gathered} \text { Concentration }{ }^{\text {e }} \\ (\mathrm{g} \mathrm{U/} \mathrm{\ell}) \end{gathered}$ | Critical Height (cm) | $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ | $\begin{gathered} \text { Concentration }^{\text {e }} \\ (\mathrm{g} \mathrm{U} / \mathrm{\ell}) \end{gathered}$ | Critical <br> Height <br> (cm) | $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ | $\begin{aligned} & \text { Concentration }{ }^{\text {e }} \\ & (\mathrm{g} \mathrm{U} / \mathrm{\ell}) \end{aligned}$ | Critical <br> Height <br> (cm) |
| Stainless steel | 27.92 | $\begin{aligned} & 145.68 \\ & 346.73 \end{aligned}$ | $\begin{aligned} & 31.20 \pm 0.04 \\ & 28.93 \pm 0.09 \end{aligned}$ | $\begin{aligned} & +0.10 \\ & +0.10 \end{aligned}$ | $\begin{aligned} & 144.38 \\ & 334.77 \end{aligned}$ | $\begin{aligned} & 29.79 \pm 0.03 \\ & 27.23 \pm 0.03 \end{aligned}$ | $\begin{aligned} & +0.16 \\ & +0.11 \end{aligned}$ | $\begin{aligned} & 144.38 \\ & 334.77 \end{aligned}$ | $\begin{aligned} & 24.19 \pm 0.01 \\ & 21.79 \pm 0.01 \end{aligned}$ |
| Aluminum | 28.01 | $\begin{aligned} & 142.92 \\ & 357.71 \end{aligned}$ | $\begin{aligned} & 33.55 \pm 0.03 \\ & 30.91 \pm 0.04^{4} \end{aligned}$ | $\begin{array}{\|l} -0.11 \\ -0.11 \end{array}$ | $\begin{aligned} & 144.38 \\ & 334.77 \end{aligned}$ | $\begin{aligned} & 31.37 \pm 0.01 \\ & 28.60 \pm 0.03 \end{aligned}$ | $\begin{aligned} & +0.06 \\ & +0.02 \end{aligned}$ | $\begin{aligned} & 144.38 \\ & 334.77 \end{aligned}$ | $\begin{aligned} & 24.70 \pm 0.01 \\ & 22.33 \pm 0.01 \end{aligned}$ |
|  | 33.01 | 54.89 59.65 137.40 | $\begin{aligned} & 39.48 \pm 0.13^{3} \\ & 36.67 \pm 0.17^{3} \\ & 23.96 \pm 0.13^{3} \end{aligned}$ | $\begin{gathered} \mathrm{g} \\ -0.05 \\ \mathrm{~g} \end{gathered}$ | 59.65 | $34.10 \pm 0.02$ | +0.08 | 59.65 | $27.27^{1}$ |
|  |  | $\begin{aligned} & 145.68 \\ & 357.7 \end{aligned}$ | $\begin{aligned} & 23.67 \pm 0.03 \\ & 22.53 \pm 0.05 \end{aligned}$ | $-\underset{\mathrm{g}}{-0.05}$ | 334.77 | $\begin{aligned} & 22.85 \pm 0.03 \\ & 21.50 \pm 0.01 \end{aligned}$ | $\begin{aligned} & +0.12 \\ & +0.05 \end{aligned}$ | $\begin{aligned} & 144.38 \\ & 334.77 \end{aligned}$ | $\begin{aligned} & 18.24 \pm 0.01 \\ & 16.78 \pm 0.02 \end{aligned}$ |
|  | 50.69 | 63.95 | $20.48 \pm 0.05$ | -0.04 |  |  |  |  |  |

${ }^{\text {a }}$ Approximately centered in an $\sim 10-\mathrm{m}$ cubical room having thick concrete walls; see Table X .
${ }^{\mathrm{b}}$ Refer to Table X for distances to reflector.
${ }^{\text {c }}$ Refer to Table XI for distances to reflector.
${ }^{\mathrm{d}}$ These four tanks are completely specified in Table VII. Five entries in this table (at 60.32 g U/ $/$ ) used three taller tanks-also described in stainless-steel tanks were used for those critical heights reported at 50.52 and 67.48 cm , respectively.
${ }^{\mathrm{e}}$ Refer to Table IV for complete specification of uranium solution, and to Table VI for biases.
${ }^{\mathrm{f}}$ Obtained by an extrapolation of reciprocal multiplication curves.
${ }^{8}$ No wire height-check correction made.
itself as a key. Similarly, the $59.65 \mathrm{~g} / \mathrm{l}$ concentration used in that tank (next column) keys to the second line of Table IV wherein the solution is described completely. Finally, the $27.27-\mathrm{cm}$ critical height itself (several columns to the right) is a key to the first line of Table XI, which locates the tank relative to reflector walls.

Critical solution heights in Tables I and II are the average of two critical experiments in most cases. Otherwise, a small superscript to the height indicates the number of experiments contributing to the average. Two critical heights were determined by extrapolating reciprocal multiplication curves because the tanks were slightly too short to achieve criticality. These are indicated in Table I by square brackets. The extrapolated data are presented in Fig. 11 because critical heights obtained in this manner are more subject to interpretation. Where two or more experiments were performed, the uncertainty assigned to the critical height equals one-half the range between heights measured on repeated experiments.

Tabled critical heights include a small bias adjustment resulting from the wire height-check procedure discussed in Sec. III. The critical height in Tables I and II equals the critical height indicated
by the level sampler during the experiment minus the bias shown, all three expressed in centimetres.

The uranium solution concentration associated with each critical height also includes a small bias correction. This is discussed in Sec. IV, and the biases are listed in Table VI.

## III. PROCEDURE

The critical data given in Tables I and II are averaged from two or more critical approach experiments. The critical height for each experiment was linearly interpolated between a slightly supercritical and a slightly subcritical height. Very few exceptions to these two statements are identified in the tables.

The reciprocal neutron multiplication technique was used for all approaches to criticality. By the time the multiplication of the system reached 50 or more, the critical height was fairly well defined, and a small ${ }^{252} \mathrm{Cf}$ neutron source, used in that technique, could be withdrawn safely.

Uranium solution was added in alternating incremental steps until the source was so far withdrawn as to have no further influence and the solution height yielded a long (approximately a few minutes)

Enriched Uranium Solution Cylinder

|  | 20.6-cm-thick Plastic Shell ( $\sim 122-\mathrm{cm}$ inside dimension) |  |  |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  | At Center ${ }^{\text {b }}$ |  |  | In Corner ${ }^{\text {c }}$ |  |  | Centered on Floor ${ }^{\text {b }}$ |  |  |
| $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ | $\begin{aligned} & \text { Concentration }{ }^{\mathrm{e}} \\ & (\mathrm{~g} \mathrm{U} / \mathrm{Q}) \end{aligned}$ | Critical <br> Height <br> (cm) | $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ | $\begin{aligned} & \text { Concentration }^{\mathrm{c}} \\ & (\mathrm{~g} \mathrm{U/X}) \end{aligned}$ | Critical Height (cm) | $\begin{gathered} \text { Bias } \\ (\mathrm{cm}) \end{gathered}$ | $\begin{aligned} & \text { Concentration }^{\text {e }} \\ & (\mathrm{g} \mathrm{U/} \mathrm{\ell}) \end{aligned}$ | Critical <br> Height <br> (cm) | $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ |
| $\begin{aligned} & +0.01 \\ & +0.01 \end{aligned}$ | $\begin{aligned} & 147.66 \\ & 345.33 \end{aligned}$ | $\begin{aligned} & 29.71 \pm 0.05 \\ & 27.60 \pm 0.01 \end{aligned}$ | $\begin{aligned} & -0.07 \\ & -0.03 \end{aligned}$ | $\begin{array}{r} 60.32 \\ 147.66 \\ 345.33 \end{array}$ | $\begin{aligned} & 50.52 \pm 0.09^{\mathrm{d}} \\ & 25.03 \pm 0.03 \\ & 22.75 \pm 0.01 \end{aligned}$ | $\begin{aligned} & +0.04 \\ & -0.04 \\ & -0.07 \end{aligned}$ | 60.32 | $67.48 \pm 0.20^{\text {d }}$ | -0.08 |
| $\begin{aligned} & -0.03 \\ & -0.03 \end{aligned}$ | $\begin{array}{r} 60.32 \\ 147.66 \\ 345.33 \end{array}$ | $\begin{gathered} {\left[78.1^{1}\right]^{\mathrm{d}, \mathrm{f}}} \\ 31.26 \pm 0.01 \\ 28.84 \pm 0.02 \end{gathered}$ | $\begin{aligned} & -0.08 \\ & -0.09 \\ & -0.08 \end{aligned}$ | $\begin{array}{r} 60.32 \\ 147.66 \\ 345.33 \end{array}$ | $\begin{aligned} & 51.67 \pm 0.05^{\mathrm{d}} \\ & 25.26 \pm 0.00 \\ & 22.87 \pm 0.01 \end{aligned}$ | $\begin{aligned} & -0.04 \\ & -0.10 \\ & -0.07 \end{aligned}$ | 60.32 | $\left[77.1^{1}\right]^{\text {d,f }}$ | +0.07 |
| 0.00 -0.02 -0.02 | 60.32 147.66 345.33 | $34.33 \pm 0.02$ $22.78 \pm 0.01$ $21.67 \pm 0.00$ | -0.05 -0.08 -0.10 | $\begin{array}{r} 60.32 \\ 66.33 \\ 147.66 \\ 345.33 \end{array}$ | $\begin{aligned} & 27.70 \pm 0.27^{3} \\ & 25.10^{1} \\ & 18.49 \pm 0.04 \\ & 17.20 \pm 0.03^{3} \end{aligned}$ | $\begin{aligned} & -0.05 \\ & -0.06 \\ & -0.09 \\ & -0.12 \end{aligned}$ | 60.32 | $31.75 \pm 0.03$ | -0.01 |

Table VII. The $76-9-\mathrm{cm}$-tall aluminum tank was used for those critical heights reported at $78.1,51.67$, and 77.1 cm . The $91.5-$ and $76.6-\mathrm{cm}-\mathrm{tall}$
positive reactor period. About 5 min later, a small amount of solution was drained, establishing a subcritical height and a negative reactor period of about the same magnitude as the positive period. The critical height was interpolated between the reciprocal periods at the two heights. The validity of this interpolation has been demonstrated for reactivities close to unity. ${ }^{6}$ Figure 1 illustrates these procedures for one experiment as recorded by one of several radiation detectors.

Solution heights throughout every experiment were determined by an electromechanical device ${ }^{13}$ that periodically "sampled" the liquid level with a precision of $\pm 0.05 \mathrm{~mm}$. This device measured the height accurately near the center of a tank, but the possibility remained that the tank was not perfectly level or the bottom was bowed or uneven. Either case would bias critical heights obtained by the device, so a wire height-check procedure was used to measure the effect. When the solution touched a wire, a preset height above the tank bottom, a feeble current turned on a transistor switch, lighting a lamp corresponding to that wire. Comparing the preset height with that indicated on the level sampler at that time measured the bias in the sampler readings. The wires were thin ( 0.08 -cm-diam) stain-
less-steel rods supported at the top of the tank by a small block of nonconducting material clamped to the tank. The solution-filling rate during this procedure was always slowed sufficiently that waves did not cause premature indications.

This bias measurement was not made on the earliest experiments, and that fact is footnoted in Table I. A single wire was used for most of the remaining unreflected single-tank experiments; however, the procedure proved so useful that four wires equally spaced around the tank perimeter were used to give multiple measures of the bias on all reflected and on the $50.69-\mathrm{cm}$-diam unreflected single-tank experiments.

Critical heights of Table I have been corrected for this small bias. The correction given in the table represents the average over the number of wires used on each experiment. In $63 \%$ of the individual lamp lightings, the bias was 10.051 cm or less. The largest bias was 0.25 cm in one case.

A similar height-check procedure was used in array experiments. Here, the purpose was not to measure variations within one tank but to relate the solution height in each cylinder of an array to that measured by the level sampler in one cylinder.

All cylinders of an array rested on the floor

TABLE
Critical Height (in centimetres) of an Array of

${ }^{\text {a }}$ Refer to Fig. 3 for identification of cylinders. Do not confuse lettered cylinder locations in this column with footnotes given
${ }^{\mathrm{b}}$ Refer to Table V for complete specification of uranium solution and to Table VI for biases.
${ }^{c}$ The adjustment was -0.07 cm for one experiment and +0.09 cm for the other two because two different wire configurations
${ }^{\mathrm{d}}$ The adjustment was +0.08 cm for one experiment and +0.27 cm for the other two because two different wire configurations
of the reflector, which had been installed as nearly level as possible. Still, minor imperfections or unevenness could allow one cylinder to rest a small amount above or below another. Two wires were installed at different preset heights in each cylinder, providing duplicate measures of such possible vertical displacement. The average of these two displacement measurements gave the "bias" for that cylinder relative to the cylinder having the level sampler. The average bias over all cylinders in the array equaled the correction term that was added to the critical height measured in one cylinder to obtain the average critical height over all cylinders. This last critical height is that published in Table II, and the adjustment included in the table is the average bias defined above. The biases of Table II are larger than those of Table I because a greater opportunity exists for members of an array to rest at slightly different
levels than for height variations to exist within a relatively small diameter tank. Some $60 \%$ of the individual lamp lightings occurred within 10.21 cm of the preset height, and an additional $34 \%$ occurred between there and 10.51 cm . The largest single bias was 0.9 cm in one case. Setting wires a known small distance above the floor of tall slender cylinders was not an easy task. The estimated accuracy of such settings is $\sim 0.1 \mathrm{~cm}$.

## IV. URANIUM SOLUTION

The uranium solution was uranyl nitrate $\left[\mathrm{UO}_{2}\left(\mathrm{NO}_{3}\right)_{2}\right]$ dissolved in nitric acid and diluted to the desired concentration with water. Experiments with single-tank configurations are reported at three uranium concentrations, while those involving arrays of cylinders were performed at only two.

II
Enriched Uranium Solution Cylinders

| Concrete Steel dimension) |  |  | 20.6-cm-thick Plastic Shell <br> ( $\sim 122-\mathrm{cm}$ inside dimension) |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| High Concentration |  |  | $60.32 \mathrm{~g} \mathrm{U} / \mathrm{l}$ |  | $355.94 \mathrm{~g} \mathrm{U} / \ell$ |  |
| $\begin{aligned} & \text { Concentration }{ }^{\mathrm{b}} \\ & (\mathrm{~g} \mathrm{U/} \mathrm{\ell}) \end{aligned}$ | Critical Height (cm) | $\begin{aligned} & \text { Bias } \\ & (\mathrm{cm}) \end{aligned}$ | Critical Height (cm) | Bias (cm) | Critical Height (cm) | Bias <br> (cm) |
| 369.96 | $17.24^{4} \pm 0.10$ | +0.03 | $34.82 \pm 0.02$ | +0.09 | $19.27 \pm 0.01$ | +0.10 |
| 364.11 | $17.13 \pm 0.02$ | +0.13 | $31.76 \pm 0.00$ | +0.09 | $18.82 \pm 0.00$ | -0.02 |
| 360.37 | $29.49 \pm 0.01$ | -0.15 | $110.20^{1}$ | +0.02 | $31.93 \pm 0.01$ | +0.13 |
| 364.11 | $31.11 \pm 0.03$ | -0.17 | $102.29 \pm 0.09$ | +0.02 | $33.20 \pm 0.02$ | +0.02 |
| 360.37 | $32.32^{3} \pm 0.09$ | d | $105.85 \pm 0.03$ | +0.08 | $38.10 \pm 0.03$ | +0.13 |
| 359.55 | $31.82 \pm 0.01$ | +0.19 | $78.40 \pm 0.05$ | +0.08 | $35.56 \pm 0.02$ | +0.19 |
| 359.55 | $51.45{ }^{1}$ | -0.15 |  |  |  |  |
| 359.55 | $65.49^{1}$ | +0.06 |  |  |  |  |
|  |  |  |  |  | $95.20 \pm 0.04$ | +0.07 |
|  |  |  |  |  | $89.78 \pm 0.02$ | +0.09 |
| 359.55 | $101.45 \pm 0.03$ | -0.02 |  |  |  |  |
| 359.55 | $104.04 \pm 0.03$ | +0.14 |  |  |  |  |

elsewhere in the table. For arrays smaller than $4 \times 4$, unused cylinder locations were vacant.
were used.
were used.

The uranium was enriched to $\sim 93 \%{ }^{235} \mathrm{U}$. On five occasions during the program, composite samples were formed from samples taken over the preceding few weeks and analyzed by mass spectrographic methods for the isotopic weight percents of the various isotopes. These results are shown in Table III, and the average measured values are assumed to apply to all experiments.

A systematic sampling program was followed throughout the entire experimental program. The solution sampled was drawn from the line returning the solution from the experimental area to storage, assuring the sample to be representative of that in the just-completed critical experiment. During the course of every sequence of experiments using the same solution, samples were taken for every few experiments and were always drawn in pairs.

The results of this sampling program are given
in Tables IV and V. Solutions in Table IV were those used in the single tank experiments reported in Table I. The complete specifications of the solutions can be keyed to the appropriate critical height data through the concentration values appearing in both tables. Similarly, the solutions in Table V were those used in the array experiments of Table II, and the same key applies between these two tables.

Two separate measurement control programs were run during the 14 months of data accumulation. Solutions having known uranium concentrations were prepared by the Rocky Flats Standards Laboratory and submitted to the Rocky Flats Analytical Laboratory for analysis as unknowns. Standards in all three concentration ranges were submitted. Comparing the standard concentrations against those measured by the Analytical Laboratory measured


Fig. 1. Neutron detector response during a typical experiment.

TABLE III
Uranium Isotopic Enrichment of Solution Used in Both Single-Tank and Array Experiments

| Date <br> Reported | Isotope (\%) |  |  |  |
| :--- | :---: | :---: | :---: | :---: |
|  | ${ }^{234} \mathrm{U}$ | ${ }^{235} \mathrm{U}$ | ${ }^{236} \mathrm{U}$ | ${ }^{238} \mathrm{U}$ |
| July 7,1976 | 1.01 | 93.16 | 0.44 | 5.40 |
| Oct. 5,1976 | 1.10 | 93.08 | 0.44 | 5.38 |
| Dec. 21, 1976 | 0.99 | 93.17 | 0.43 | 5.41 |
| Mar. 29, 1977 | 1.01 | 93.23 | 0.43 | 5.33 |
| June 21,1977 | 1.01 | 93.22 | 0.43 | 5.34 |
| Average | $1.022 \pm 0.043$ | $93.172 \pm 0.060$ | $0.434 \pm 0.005$ | $5.372 \pm 0.036$ |

the bias in the results due to the individual technician, equipment, and method used. The results of this program are shown in Table VI. The concentrations of Tables IV and V include the average of these as a bias correction.

Elements contributing most to the total impurity given in the two tables are listed below. Each impurity is given in ppm (parts of impurity per million parts of uranium by weight); the sizable uncertainty reflects the difficulty of measuring such small contributions. Strong neutron absorbers, boron and cadmium, are included. The principal impurities are: aluminum ( $240 \pm 195$ ), boron ( $13 \pm 15$ ), ${ }^{\text {a }}$ calcium ( $200 \pm 160$ ), cadmium ( $37 \pm 22$ ), ${ }^{\text {a ch chromium }}$

[^0]$(48 \pm 56)$, copper $(33 \pm 26)$, iron $(440 \pm 240)$, potassium $(48 \pm 21)$, magnesium $(260 \pm 250)$, manganese ( $21 \pm 13$ ), nickel ( $76 \pm 56$ ), and silicon ( $140 \pm 100$ ).

## V. TANKS AND CYLINDERS

All containers used in these experiments were open-topped right circular cylinders. Each had an $\sim 30-\mathrm{cm}$-long coaxial "tailpipe" of the same material welded to the bottom as shown in Fig. 2. This tailpipe passed solution to and from the cylinder during experiments.

Aluminum cylinders of three diameters were used in the single-tank experiments. The smallest

TABLE IV
Properties of Uranyl Nitrate Solutions Used in Single-Tank Experiments of Table I*

| Uranium <br> Concentration <br> $(\mathrm{g} \mathrm{U/l})$ | Solution <br> Density <br> $\left(\mathrm{g} / \mathrm{cm}^{3}\right)$ | Excess <br> Nitric Acid <br> $(\mathrm{molar})$ | Total <br> Impurities <br> $(\mathrm{ppm})$ | $\mathrm{H}: \mathrm{U}$ |
| :---: | :---: | :---: | :---: | :---: |
| $54.89 \pm 0.25$ | $1.0758 \pm 0.0006$ | $0.105 \pm 0.001$ | 2340 | 465.6 |
| $59.65 \pm 0.42$ | $1.0825 \pm 0.0006$ | $0.114 \pm 0.004$ | $2150 \pm 680$ | 427.7 |
| $60.32 \pm 0.55$ | $1.0837 \pm 0.0007$ | $0.113 \pm 0.002$ | $2860 \pm 990$ | 423.0 |
| $63.95 \pm 0.34$ | $1.0883 \pm 0.0002$ | $0.111 \pm 0.003$ | 398.5 |  |
| $66.33 \pm 1.52$ | $1.0920 \pm 0.0025$ | $0.120 \pm 0.003$ | $2130 \pm 250$ | 383.9 |
| $137.40 \pm 0.63$ | $1.1923 \pm 0.0007$ | $0.287 \pm 0.002$ | 2210 | 180.2 |
| $142.92 \pm 0.52$ | $1.2007 \pm 0.0024$ | $0.283 \pm 0.003$ | $1960 \pm 580$ | 173.1 |
| $144.38 \pm 0.47$ | $1.2023 \pm 0.0006$ | $0.272 \pm 0.003$ | $1850 \pm 130$ | 171.3 |
| $145.68 \pm 1.04$ | $1.2038 \pm 0.0001$ | $0.294 \pm 0.002$ | $1240 \pm 110$ | 169.5 |
| $147.66 \pm 0.75$ | $1.2069 \pm 0.0009$ | $0.271 \pm 0.010$ | $1690 \pm 440$ | 167.4 |
| $334.77 \pm 1.27$ | $1.4636 \pm 0.0011$ | $0.521 \pm 0.004$ | $1390 \pm 30$ | 68.5 |
| $345.33 \pm 1.18$ | $1.4779 \pm 0.0011$ | $0.534 \pm 0.023$ | $1420 \pm 540$ | 66.1 |
| $346.73 \pm 0.95$ | $1.4800 \pm 0.0003$ | $0.542 \pm 0.005$ | $1360 \pm 190$ | 65.8 |
| $357.71 \pm 1.99$ | $1.4951 \pm 0.0006$ | $0.549 \pm 0.015$ | $1430 \pm 360$ | 63.5 |

*All uncertainties represent one standard deviation about the mean for multiple samples. All solution properties were measured at $23.0^{\circ} \mathrm{C}$.

TABLE V
Properties of Uranyl Nitrate Solutions Used in Array Experiments of Table II*

| Uranium <br> Concentration <br> $(\mathrm{g} \mathrm{U} / \ell)$ | Solution <br> Density <br> $\left(\mathrm{g} / \mathrm{cm}^{3}\right)$ | Excess <br> Nitric Acid <br> (molar) | Total <br> Impurities <br> (ppm) | $\mathrm{H}: \mathrm{U}$ |
| :---: | :---: | :---: | :---: | :---: |
| $60.32 \pm 0.55$ | $1.0837 \pm 0.0007$ | $0.113 \pm 0.002$ | $2860 \pm 990$ | 423.0 |
| $63.95 \pm 0.34$ | $1.0883 \pm 0.0002$ | $0.111 \pm 0.003$ | $780 \pm 320$ | 398.5 |
| $67.28 \pm 0.27$ | $1.0934 \pm 0.0003$ | $0.128 \pm 0.004$ | $2300 \pm 240$ | 378.2 |
| $76.09 \pm 0.21$ | $1.1057 \pm 0.0001$ | $0.137 \pm 0.002$ | $2190 \pm 210$ | 333.5 |
| $80.72 \pm 0.16$ | $1.1122 \pm 0.0000$ | $0.143 \pm 0.001$ | $2060 \pm 30$ | 313.8 |
| $83.49 \pm 0.47$ | $1.1164 \pm 0.0006$ | $0.151 \pm 0.002$ | $2610 \pm 250$ | 303.2 |
| $355.94 \pm 2.68$ | $1.4925 \pm 0.0029$ | $0.494 \pm 0.019$ | $1160 \pm 310$ | 64.1 |
| $359.55 \pm 1.38$ | $1.4984 \pm 0.0008$ | $0.578 \pm 0.019$ | $1610 \pm 120$ | 63.1 |
| $360.37 \pm 2.60$ | $1.4995 \pm 0.0037$ | $0.585 \pm 0.021$ | $1530 \pm 320$ | 62.9 |
| $364.11 \pm 1.78$ | $1.5054 \pm 0.0009$ | $0.584 \pm 0.016$ | $1420 \pm 20$ | 62.3 |
| $369.96 \pm 1.45$ | $1.5120 \pm 0.0017$ | $0.598 \pm 0.025$ | $1340 \pm 100$ | 61.0 |

*All uncertainties represent one standard deviation about the mean for multiple samples. All solution properties were measured at $23.0^{\circ} \mathrm{C}$.
was also fabricated of stainless steel to determine the effect of container material on critical heights. Array experiments used only two diameters of aluminum cylinders, and the material effect was determined by slipping close-fitting stainless-steel sleeves, rolled from sheet stock, over the aluminum on some experiments. Tailpipes on array experiments were of two diameters because two cylinders served as safety "scrams."

The inside diameter of each vessel was obtained by a water calibration prior to use. For each water increment, the corresponding increase in height was noted and the diameter averaged over

$$
D_{i}=\left[(\pi / 4)\left(\Delta H_{i} / \Delta V_{i}\right)\right]^{-1 / 2}
$$

Wherever uranium solution would come into contact with aluminum surfaces, a protective coating of acid-resistant paint was applied. The paint was

TABLE VI
Bias in Uranium Solution Concentrations Reported by the Analytical Laboratory*

| Date <br> Reported | Nominal Concentration Range |  |  |
| :--- | :---: | :---: | :---: |
|  | Low | Middle | High |
|  | $-0.03 \pm 0.14$ | $-0.70 \pm 0.20$ | $-2.07 \pm 0.81$ |
| Jan. 1977 | $+0.07 \pm 0.05$ | $-0.20 \pm 0.10$ | $-1.10 \pm 0.40$ |
| Average | $+0.02 \pm 0.15$ | $-0.45 \pm 0.22$ | $-1.58 \pm 0.90$ |

*True concentration $=$ reported concentration - bias. The uncertainty represents the standard deviation about the mean for three samples in each case.
a modified phenolic called "Phenoline 300."b The density of the fully cured paint is $1.505 \mathrm{~g} / \mathrm{cm}^{3}$. The thickness of this coating was calculated from the mass difference before and after painting divided by the density and the surface area covered.

A number of different thicknesses of paint are shown in Table VIII. These occurred because the mild acid eventually penetrated the paint and began to attack the aluminum. It became necessary to repaint or strip and repaint various portions of the cylinders at different times throughout the experimental program. The thicknesses shown for the condition given are reasonable ones to use in computer simulations of these experiments.

A complete dimensional specification of all containers and sleeves used in this program is given in

[^1]

Fig. 2. A single tank under minimally reflected conditions.

TABLE VII
Properties of Tanks Used in Single-Tank Experiments of Table I

| Material $^{\mathrm{a}}$ | Inside <br> Diameter <br> $(\mathrm{cm})$ | Inside <br> Height <br> $(\mathrm{cm})$ | Uncoated <br> Mass <br> $(\mathrm{g})$ | Mass of <br> Coating <br> $(\mathrm{g})$ | Thickness <br> of Coating <br> $(\mathrm{cm})$ | Tailpipe ${ }^{\mathrm{b}}$ <br> Length <br> $(\mathrm{cm})$ |
| :--- | :---: | :---: | :---: | :---: | :---: | :---: |
| Type 6061 | $50.69 \pm 1.25$ | 30.9 | 7834 | 323 | 0.030 | 29.9 |
| aluminum | $33.01 \pm 0.25$ | 49.5 | 6049 | 170 | 0.018 | 30.4 |
|  | $28.01 \pm 0.14$ | 41.9 | 4473 | 118 | 0.017 | 30.3 |
|  | $27.88 \pm 0.09$ | 76.9 | 7165 | 189 | 0.017 | 29.7 |
| Type 304 or $316^{\text {c }}$ | $27.92 \pm 0.38$ | 41.6 | 12326 |  |  |  |
| stainless steel | $27.93^{\mathrm{d}} \pm 0.16$ | 91.5 | 23739 | No coating on | $30.7^{\mathrm{e}}$ | $27.93^{\mathrm{d}} \pm 0.16$ |

[^2]TABLE VIII
Properties of Cylinders and Sleeves Used in the Array Experiments of Table II

| $\begin{aligned} & \text { Dimension } \\ & (\mathrm{cm}) \end{aligned}$ | $\sim 16$ - cm -diam Cylinders | $\sim 21-\mathrm{cm}$-diam Cylinders |
| :---: | :---: | :---: |
| Cylinder ${ }^{\text {a }}$ |  |  |
| inside diameter | $16.12 \pm 0.07$ | $21.12 \pm 0.01$ |
| wall thickness | $0.32 \pm 0.01$ | $0.40 \pm 0.01$ |
| outside diameter ${ }^{\text {b }}$ | 16.77 | 21.92 |
| inside length | $119.1 \pm 0.1$ | $119.1 \pm 0.1$ |
| bottom thickness ${ }^{\text {c }}$ | 0.32 | 0.32 |
| Coating (thickness) |  |  |
| inside wall, concrete reflector | $0.017 \pm 0.003$ | $0.014 \pm 0.002$ |
| bottom and tailpipe, concrete reflector | $0.157 \pm 0.030$ | $0.014 \pm 0.002$ |
| inside wall, plastic reflector | $0.017 \pm 0.003$ | $0.015 \pm 0.002$ |
| bottom and tailpipe, plastic reflector | $0.157 \pm 0.030$ | $0.077 \pm 0.030$ |
| Tailpipe |  |  |
| outside diameter, except cylinders $f$ and $j$ | 1.27 | 1.27 |
| outside diameter, cylinders $f$ and $j$ only | 2.54 | 2.54 |
| wall thickness | 0.13 | 0.13 |
| length | 30.4 | 30.4 |
| Sleeve ${ }^{\text {d }}$ |  |  |
| wall thickness | $0.31 \pm 0.01$ | $0.32 \pm 0.01$ |
| length | $(2 \times) 61.0 \pm 0.01$ | $122.0 \pm 0.1$ |


| Cylinder <br> Location ${ }^{\text {e }}$ | Mass of Individual Components (g) |  |  |  |
| :---: | :---: | :---: | :---: | :---: |
|  | Small-Diameter Cylinders |  | Large-Diameter Cylinders |  |
|  | Cylinder ${ }^{\text {f }}$ | Sleeve | Cylinder ${ }^{\text {f }}$ | Sleeve |
| a | 6086 | 16039 | 9154 | 20935 |
| b | 5708 | 16656 | 9048 | 20940 |
| c | 5695 | 16204 | 9015 | 20858 |
| d | 6091 | 16180 | 8970 | 20747 |
| e | 6115 | 16582 | 9106 | 20783 |
| f | 5659 | 16620 | 8980 | 20950 |
| g | 5591 | 16585 | 8829 | 20762 |
| h | 5695 | 15745 | 9209 | 20893 |
| i | 5700 | 16217 | 8766 | 20911 |
| j | 5670 | 16613 | 9108 | 20755 |
| k | 6095 | 16205 | 9194 | 20755 |
| 1 | 5595 | 16544 | 8800 | 20977 |
| m | 6093 | 16214 | 9295 | 20757 |
| n | 5590 | 16530 | 8850 | 20740 |
| 0 | 5597 | 15757 | 9012/9252 ${ }^{\text {g }}$ | 20940 |
| p | 5688 | 16599 | 9095/9116 ${ }^{\text {g }}$ | 20961 |
| Average, standard deviation | $5792 \pm 216$ | $16331 \pm 305$ | $\begin{aligned} & 9027 \pm 156 / \\ & 9043 \pm 166^{\mathrm{g}} \end{aligned}$ | $20854 \pm 93$ |

${ }^{\text {a }}$ Constructed from schedule 10 S commercial aluminum (Type 6061-T6) pipe.
${ }^{6}$ Not measured, o.d. $=$ i.d. $+2 \times$ wall.
${ }^{\mathrm{d}}$ Not measured, nominal value.
${ }^{\mathrm{d}}$ Rolled from Type 304 stainless steel.
${ }^{\text {e }}$ Refer to Fig. 3 for identification of cylinders. Do not confuse lettered cylinder locations in this column with footnotes given elsewhere in the table.
${ }^{1}$ Before coating with paint.
${ }^{8}$ See text for explanation of double entries.

Tables VII and VIII. For both, the diameters themselves serve as a key identifying a specific tank with a particular critical height in Table I or II. Three tanks in Table VII do not appear explicitly in the critical height table. These are taller tanks constructed especially for experiments expected to have

TABLE IX
Composition of Containers in Weight Percent

| Element | Aluminum (Type 6061-T6) | Stainless Steel |  |
| :---: | :---: | :---: | :---: |
|  |  | (Type 304) | (Type 316) |
| Carbon |  | 0.066 | 0.042 |
| Magnesium | 1.00 |  |  |
| Aluminum | $97.35^{\text {a }}$ |  |  |
| Silicon | 0.60 | 0.81 | 0.45 |
| Phosphorus |  | 0.025 | 0.031 |
| Sulfur |  | 0.019 | 0.014 |
| Titanium | 0.03 |  |  |
| Chromium | 0.17 | 18.5 | 16.6 |
| Manganese | 0.07 | 1.29 | 1.25 |
| Iron | 0.47 | $70.02^{\text {a }}$ | $70.313^{\text {a }}$ |
| Nickel |  | 9.27 | 11.3 |
| Copper | 0.25 |  |  |
| Zinc | 0.06 |  |  |
| Molybdenum |  | 0.018 | 2.1 |
| Density $\left(\mathrm{g} / \mathrm{cm}^{3}\right)$ | 2.737 | 7.927 | $7.92{ }^{\text {b }}$ |

${ }^{2}$ Determined by difference.
${ }^{\mathrm{b}}$ Handbook value.
a critical height exceeding the capacity of the tanks on hand. These taller tanks were used in five configurations with the plastic reflector using $60.32 \mathrm{~g} \mathrm{U} / \ell$ uranium solution. All five cases are footnoted in Table I.

Some dimensional variation existed within the commercial aluminum pipe used. The five smalldiameter array cylinders labeled a, d, e, k , and m (locations specified on Fig. 3) apparently came from heavier stock. Their weights averaged $6092 \pm$ 17 g , while the remaining 11 averaged $5653 \pm 50 \mathrm{~g}$. Although the larger cylinders had a similar weight spread, no such clearly defined grouping is apparent.

Fabrication difficulties necessitated rolling the sleeves for the small cylinders in two pieces, which were then stacked to cover the full height of the cylinder. Ten of these pieces came from thinner stock (the top half-sleeve of cylinders $\mathrm{a}, \mathrm{c}, \mathrm{i}$, and k ; the bottom half-sleeve of cylinders d and m ; and both sleeve pieces for cylinders $h$ and $o$ ). The average weight of these 10 was $7884 \pm 21 \mathrm{~g}$, while the remaining 22 averaged $8293 \pm 39 \mathrm{~g}$.

Large-diameter cylinders $o$ and $p$ were damaged between experiments in the two different reflectors. Replacements were fabricated and given one coat of paint on the interior. Entries to the left/right of the slash near the bottom of Table VIII' correspond to experiments within the concrete/plastic reflector.

The elemental compositions of materials used in fabricating tanks, cylinders, and sleeves are given in Table IX. These results were determined by laboratory analysis of scrap salvaged during fabrica-

TABLE
Interior Dimensions of Reflectors and Location* of Tanks

| Description of Experiment Category | Table $^{\text {a }}$ | North | South | Range | Total |
| :--- | :---: | :---: | :---: | :---: | :---: |
| Minimally reflected ${ }^{\text {b }}$ |  |  |  |  |  |
| Most cases | I | 556 | 511 | 10 | 1067 |
| $50.69-c m-d i a m ~ t a n k ~ o n l y ~$ | I | 535 | 532 | 0 | 1067 |
| Concrete reflected |  |  |  |  |  |
| Single tank, centered | I | 57.4 | 64.8 | 1.4 | 122.2 |
| Single tank, in corner | I | 16.6 | 105.6 | 0.3 | 122.2 |
| Arrays, all cases | II |  | (See Fig. 3) |  | 122.2 |
| Plastic reflected |  |  |  |  |  |
| Single tank, centered | I | 60.4 | 62.5 | 0.5 | 122.9 |
| Single tank, in corner | I | 17.3 | 105.6 | 0.9 | 122.9 |
| Single tank, centered on floor | I | 61.1 | 61.8 | 0.5 | 122.9 |
| Arrays, no sleeves | II |  | (See Fig. 3) |  | 122.9 |
| Arrays, with sleeves | II |  | (See Fig. 3) |  | 122.9 |

[^3]tion. Type 316 stainless steel was inadvertently used to make the bottom of the tall stainless-steel tank, instead of the Type 304 material requested. Table IX gives the analyses of both materials.

The approximate locations of the minimally reflected single tanks within the large thick-walled ${ }^{c}$ concrete assembly room are given in the upper third of Table X. Entries in the columns labeled "total" give the interior dimensions of the room. Several different setups were made with slight variations between them but, since the range of these variations was small compared to the distances to the room walls, only the range is given. Similarly, the locations of single tanks within the concrete and plastic reflectors and their ranges are given in the middle and lower third of Table X, respectively. Again, the totals give the interior dimensions of the reflector shells. Small variations in the interior height of the reflector occurred because of the need to rest the lid on small pads of various thickness to obtain ample clearance for equipment. The height of the side walls, of course, did not change. Single tanks located in one corner of either reflector are more highly affected by the two closest adjacent walls, so these distances are given for individual measurements in Table XI.

Holes were cast or drilled into the floors of the reflector shells to receive the tailpipes. The

[^4]holes were positioned on $30.48-\mathrm{cm}$ centers and thereby determined the lattice spacing for the arrays of cylinders. A lightweight aluminum angle framework, visible in Fig. 3, held the tops of all cylinders at this same spacing and kept them vertical. The nominal distance from the reflector walls to the centers of the perimeter cylinders of a $4 \times 4$ array was half the lattice spacing.

## VI. CONCRETE REFLECTOR

A type of concrete representative of that used in the nuclear industry was selected for this program. The one designated " 03 " in Ref. 14 met that goal. This concrete has a greater carbon content than many, so limestone was selected for the aggregate material, along with the necessary amounts of sand, cement, and water to yield the desired composition.

Geometrically, the reflector was a thick-walled cubical shell $\sim 173 \mathrm{~cm}$ along an exterior side with an $\sim 122-\mathrm{cm}$ interior cavity. The reflector was cast in six panels. The square bottom panel was large enough to support the four identical side panels, arranged such that each wall consisted of the side of one panel and the end of another. Figure 4 shows an elevation of the reflector shell and its supporting structure. All side panels stood $121.9 \cdot \mathrm{~cm}$ high but rested on rubber pads designed to subject the bottom to compressive loading only. These pads placed the top of the four sides 123.1 cm above the floor. Figure 5 shows the reflector at this stage of

X
Within Them for Various Categories of Experiments

| East | West | Range | Total | Up | Down | Range | Total |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  |  |  |  |  |  |
| 451 | 677 | 5 | 1128 | 480 | 495 | 9 |  |
| 445 | 683 | 0 | 1128 | 469 | 506 | 0 | 975 |
|  |  |  |  |  |  |  |  |
| 64.6 | 57.2 | 2.4 | 121.8 | 82.0 | 41.7 | 0.2 | 123.7 |
| 104.1 | 17.7 | 0.8 | 121.8 | 123.7 | 0 | 0 | 123.7 |
|  | (See Fig. 3) |  | 121.8 | 124.4 | 0 | 0 | 124.4 |
|  |  |  |  |  |  |  |  |
| 61.2 | 61.7 | 1.7 | 122.9 | 81.4 | 41.5 | 0.2 | 122.9 |
| 105.0 | 17.9 | 0.6 | 122.9 | 122.9 | 0 | 0 | 122.9 |
| 61.3 | 61.6 | 0.2 | 122.9 | 122.9 | 0 | 0 | 122.9 |
|  | (See Fig. 3) |  | 122.9 | 122.9 | 0 | 0 | 122.9 |
|  | (See Fig. 3) |  | 122.9 | 123.9 | 0 | 0 | 123.9 |

measures the extremes between cases within each category, and the totals give the interior dimensions of the reflectors in centi-

TABLE XI
Precise Locations for Single Tanks on the Floor in the Corner of the Two Reflectors

| Reflector | Key to Table I |  |  | Distance ${ }^{\text {a }}$ to Reflector (cm) |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | Tank Diameter (cm) and Material | SolutionConcentration$(\mathrm{g} / \mathrm{\ell})$ | Critical Height (cm) |  |  |
|  |  |  |  | North | West |
| Concrete | 33.01 aluminum | 59.65 | 27.27 | 16.50 | 17.92 |
|  | 33.01 aluminum | 144.38 | 18.24 | 16.50 | 17.92 |
|  | 28.01 aluminum | 144.38 | 24.70 | 16.50 | 17.80 |
|  | 27.92 stainless steel | 144.38 | 24.19 | 16.76 | 17.31 |
|  | 27.92 stainless steel | 334.77 | 21.79 | 16.76 | 17.31 |
|  | 28.01 aluminum | 334.77 | 22.33 | 16.65 | 18.05 |
|  | 33.01 aluminum | 334.77 | 16.78 | 16.50 | 17.25 |
| Plastic | 33.01 aluminum | 345.33 | 17.20 | 17.30 | 17.70 |
|  | 28.01 aluminum | 345.33 | 22.87 | 17.50 | 17.80 |
|  | 27.92 stainless steel | 345.33 | 22.75 | 16.81 | 18.06 |
|  | 27.92 stainless steel | 147.66 | 25.03 | 17.06 | 17.91 |
|  | 28.01 aluminum | 147.66 | 25.26 | 17.52 | 17.95 |
|  | 33.01 aluminum | 147.66 | 18.49 | 17.10 | 17.72 |
|  | 33.01 aluminum | 66.33 | 25.10 | 17.10 | 17.72 |
|  | 33.01 aluminum | 60.32 | 27.70 | 17.10 | 17.72 |
|  | 27.92 stainless steel | 60.32 | 50.52 | 17.71 | 18.26 |
|  | 28.01 aluminum | 60.32 | 51.67 | 17.51 | 17.75 |

${ }^{\mathrm{a}}$ From centerline of tank.


Fig. 3. An array of cylinders in the concrete reflector.
construction. The top panel, which was the same size as the bottom, was supported by these sides but rested on other pads that provided clearance for instrumentation cables. The actual interior dimensions for each measurement are given in later tables. The completed concrete reflector rested on a $173-\mathrm{cm}-$ square $X \cdot 1.3-\mathrm{cm}$-thick steel plate for better load distribution and this, in turn, was supported 40 cm above a $2.5-\mathrm{cm}$-thick sturdy steel table by eight jacks (see Fig. 6).

The total weight of concrete in all six panels 16 months after they were poured was 7790.8 kg , and the cured concrete density was $2.321 \pm 0.017$ $\mathrm{g} / \mathrm{cm}^{3}$. The average thickness of the four walls was 25.7 cm , but the top and bottom were slightly thinner at 25.4 cm . In addition to the concrete, the six pancls combined contained 11.9 kg of steel reinforcing wire and 3.9 kg of other imbedded steel pieces. The "rebar" consisted of $0.48-\mathrm{cm}$-diam steel rod welded in a rectangular gridwork placed in the mid-plane of each panel during pouring. The top and bottom panels each contained $\sim 22 \mathrm{~m}$ of this wire weighing $\sim 3 \mathrm{~kg}$, while each side panel contained $\sim 10.8 \mathrm{~m}$ of the same wire.

Both top and bottom pancls contained a number of small holes serving various purposes. The top had twenty-seven $2.5-\mathrm{cm}$-diam holes, and the bottom had four of that diameter plus fourteen half that size (see Fig. 7). Each side panel contained only


Fig. 4. Drawing of the concrete reflector in elevation.
one 3.8 -cm-square hole at one corner used as a safety drain in the event of a uranium solution leak. (The interior had been lined with vinyl sheet for contamination control.) All holes and imbedded materials consumed only $\frac{1}{4} \%$ of the reflector volume; the density given above is for concrete only, having been corrected for these small mass and volume perturbations.

The top panel cracked during construction but was made safe by surrounding it with a compression belt of steel 10 cm wide $\times 0.6 \mathrm{~cm}$ thick. This belt weighed $\sim 36 \mathrm{~kg}$ and is not included in the weight given above, although it was present during experiments.

The composition of cured concrete was determined two ways. Having an elemental analysis of the sand, cement, and limestone, the amount of each element in the overall composition was calculated by multiplying the weight fraction of the element within the ingredient by the weight fraction of the ingredient within the concrete. The second method was an elemental analysis of the well-cured concrete by a private laboratory. They were given a large piece broken from a sample block, cast along with the experimental panels. The sample block ex-


Fig. 5. A single tank in one corner of the concrete reflector.


Fig. 6. Reflector support showing all nearby environmental reflectors.
perienced the same environment and had the same thickness as the panels used; thus, the two are assumed to have dried out at the same rate.

The weight of all materials put into the concrete at the time of mixing is shown in Table XII. Both the cement and limestone aggregate (average chip size, 1.6 cm ) were assumed to contain no absorbed water because considerable effort was expended to keep them dry. The sand, however, came from an


Fig. 7. Concrete reflector top and bottom panels during fabrication, showing all perturbing holes.

TABLE XII
Concrete Reflector Ingredients

| Ingredient | Wet Mix (kg) | Cured 16 Months |  |
| :---: | :---: | :---: | :---: |
|  |  | (kg) | (wt\%) |
| Cement | 1029.51 |  | 13.22 |
| Dry sand | 2591.10 |  | 33.27 |
| Limestone ${ }^{\text {a }}$ | 3646.33 |  | 46.81 |
| Total water | 737.51 | 521.99 | 6.70 |
| Pozzolith |  | 1.91 | b |
| Totals | 8006.36 | 7790.84 | 100.00 |
| Density | 2.384 | $2.321 \pm 0.017$ |  |

${ }^{\text {a }}$ The average volume of a limestone chip was $0.385 \mathrm{~cm}^{3}$.
${ }^{\mathrm{b}}$ Pozzolith is a lignin that serves as a water-reducing agent. Its small weight percent was distributed among the three dry ingredients in arriving at the last column.
outdoor loading bin and was subsequently shown to contain $7.38 \%$ moisture. This moisture has been included with the water added at the cement plant in arriving at the total water content given in the first column of the table.

The elimination of water during the curing process is assumed to be the only change in the concrete reflector throughout the entire experiment. The first experiments using this reflector were performed four months after it was cast, and they were completed in another four months. The total weight of water eliminated over 16 months was 215.52 kg ; however, Fig. 8 shows that most of this weight loss occurred before the first experiments began.

The elemental analyses of the four principal constituents of concrete are presented in Table XIII. No analysis of pozzolith, a lignin that serves as a


Fig. 8. Water retention of a test concrete block over a curing time of several months. The three arrows along the axis, left to right, give the first and last days for experiments with this reflector and the time at which a private laboratory made their elemental analysis.
water-reducing agent, was made because of its small amount. Two self-consistency checks show good agreement when applied to the limestone analysis. Dolomite limestone is principally $\mathrm{CaCO}_{3}$ and $\mathrm{MgCO}_{3}$. If all calcium and magnesium observed were in the form of carbonates, then these two, other metals (aluminum, silicon, iron, and titanium), and a measured $1.41 \%$ of oxygen (assumed to be the oxides of these metals) sum to $100.13 \%$, well within analytical uncertainties. Another check is obtained by subtracting the measured calcium and magnesium weights from the calculated weights of their carbonates. This suggests that $59.50 \%$ of the limestone

TABLE XIII
Composition of Concrete Ingredients in Weight Percent

| Element | Portland <br> Cement | Ordinary <br> Sand | Limestone <br> Aggregate | Water $^{\mathbf{a}}$ |
| :--- | :---: | :---: | :---: | :---: |
| Hydrogen |  |  |  | 11.19 |
| Carbon |  |  | 11.9 |  |
| Oxygen | 38.41 | 48.63 | 48.88 | 88.81 |
| Sodium |  | 0.62 |  |  |
| Magnesium | 3.6 | 0.17 | 2.51 |  |
| Aluminum | 1.75 | 5.1 | 0.03 |  |
| Silicon | 9.9 | 42.5 | 0.99 |  |
| Sulfur | 1.1 |  |  |  |
| Potassium |  | 0.38 | 35.6 |  |
| Calcium | 44.5 | 1.1 | 3.6 |  |
| Titanium |  |  | 0.01 |  |
| Iron | 0.74 | 1.5 | 0.08 |  |
| Totals | 100.0 | 100.0 | 100.0 | 100.0 |

${ }^{\text {a }}$ Not analyzed.
boxygen determined "by difference."
weight is $\mathrm{CO}_{3}$, in good agreement with the assumption that all carbon observed is in the carbonate form: 59.45\%.

Table XIV gives the elemental analysis of the cured concrete as determined by these two methods, with the average used in calculating atomic number densities. This concrete may be compared with three others listed in Ref. 15. The analytical methods
are listed in the table. For most methods, the nominal accuracy claimed is about $\pm 5 \%$ of the amount present except for the Keldahl method at this nitrogen level: $\pm 100 \%$. The agreement between the two methods appears to be much better than that. The private laboratory employed emission spectroscopy to measure impurities, finding a total 2746 ppm distributed over 53 elements. Strong neutron absorbers boron, chlorine, and cadmium were 24,42 , and 0.28 ppm , respectively.

## VII. PLASTIC REFLECTOR

The methyl methacrylate plastic ${ }^{\text {d }}$ reflector resembled the concrete reflector in size and shape but differed in several important respects. Each panel of the thick-walled cubical shell was laminated of two thick plastic sheets bolted together. The average thickness for the six panels was $20.6 \mathrm{~cm}, \sim 5 \mathrm{~cm}$ thinner than the concrete. The interior cavity remained $\sim 122 \mathrm{~cm}$ on a side, so the exterior side length was $\sim 162 \mathrm{~cm}$. Accurate interior dimensions for each arrangement can be found in the tables.

The completely assembled reflector, as seen in Fig. 9, is not a perfect cube, having small corner blocks absent. This occurred because the maximum width of available material dictated the assembly

[^5]TABLE XIV
Composition of Concrete in Weight Percent

| Element | Analysis of <br> Ingredients | Analysis of <br> Cured Concrete | Average | Method $^{\mathbf{a}}$ | Number Densityb <br> (atom/b $\cdot \mathrm{cm})$ |
| :--- | :---: | :---: | :---: | :---: | :---: |
| Hydrogen | 0.75 | 0.75 | 0.75 | CH | 0.0104008 |
| Carbon | 5.57 | 5.52 | 5.55 | CH | 0.0064590 |
| Nitrogen | 0.00 | 0.02 | 0.01 | K | 0.0000100 |
| Oxygen | 50.09 | 48.49 | 49.29 | Difference | 0.0430634 |
| Sodium | 0.21 | 0.63 | 0.42 | AA | 0.0002554 |
| Magnesium | 1.71 | 1.25 | 1.48 | AA | 0.0008509 |
| Aluminum | 1.94 | 2.17 | 2.06 | AA,C | 0.0010672 |
| Silicon | 15.91 | 15.50 | 15.70 | AA,C | 0.0078138 |
| Sulfur | 0.15 | 0.19 | 0.17 | Eschka | 0.0000741 |
| Potassium | 0.13 | 1.37 | 0.75 | AA | 0.0002681 |
| Calcium | 22.91 | 23.00 | 22.95 | AA | 0.0080040 |
| Titanium | 0.00 | 0.10 | 0.05 | AA | 0.0000146 |
| Iron | 0.63 | 100 | 0.82 | AA | 0.0002052 |
| Total | 100.0 |  |  |  |  |

${ }^{\mathrm{a}} \mathrm{AA}=$ atomic absorbtion
$\mathrm{CH}=$ carbon/hydrogen analyzer $\mathrm{K}=\mathrm{k}$ jeldahl $\mathrm{C}=$ calorimetric.
${ }^{\mathrm{b}}$ Based on a density of $2.321 \mathrm{~g} / \mathrm{cm}^{3}$.


Fig. 9. A single tank centered in the plastic reflector.
pattern. Unfortunately, filler blocks to complete a truly cubical geometry, of no consequence experimentally but desirable from a computational point of view, were not used.

The six panels were assembled as shown, somewhat simplified, in Fig. 10. The bottom panel consisted of two $121.9-\times 162.6-\mathrm{cm}$ sheets bolted together with their long axes at right angles to one another, providing lips to support the side panels. All five other panels were ordinary parallelepipeds. The two walls with long axes horizontal measured $121.9 \times 162.6 \mathrm{~cm}$, and the other two walls were 121.9 cm wide $\times 152.4 \mathrm{~cm}$ high. The removable top panel was $121.9 \times 162.6 \mathrm{~cm}$. The top rested on small pads to provide needed clearance for wires, so precise interior dimensions for various configurations must be obtained from the tables.

A surprisingly large variability in thickness existed among the 12 nominally $10.2-\mathrm{cm}$-thick sheets composing the six panels. The material used in the bottom and all four sides apparently belonged to one group centered at $10.39 \pm 0.24 \mathrm{~cm}$, while the average thickness of the two top sheets was $10.09 \pm$ 0.19 cm . In summary, the total plastic thickness for five panels was $20.8 \pm 0.3 \mathrm{~cm}$, while the top was $20.2 \pm 0.3 \mathrm{~cm}$ thick.

The plastic reflector was supported on the same $1.3-\mathrm{cm}$-thick steel plate placed under the concrete. The same jacks and support, shown in Fig. 6, were used except that the jacks were raised to locate the floor at the same elevation established for the concrete ( 65.4 cm above the steel table top). Essentially the same hole pattern existed in the top and bottom panels as in the corresponding concrete pieces;


Fig. 10. Assembly drawing of the plastic reflector, showing perturbations from a complete cube.
however, no drain holes were needed in the sides because the interior was not lined with vinyl sheet.

During postexperiment analyses, two kinds of plastic material were discovered. All plastic used in the four walls was common Plexiglas $\left[\mathrm{CH}_{2}: \mathrm{C}\left(\mathrm{CH}_{3}\right) \mathrm{CO}_{2} \mathrm{CH}_{3}\right]$. The material used in the top and bottom panels was fire-retardant plastic designated "Plexiglas FI-3." This latter plastic has the same chemical formulation to which a bromine compound has been added as a fire inhibitor. The additive increases the density of the plastic. This parameter, the amount of material used in the reflector, and the elemental composition of both plastics are given in Table XV.

## VIII. ENVIRONMENTAL REFLECTORS

The centers of both reflectors were at about the same location within a large stainless-steel hoodlike enclosure. The hood measured 4.9 m long $X$ 3.0 m wide $X 5.7 \mathrm{~m}$ high. The reflectors were centered in the width, but the reflector center was $\sim 1.5 \mathrm{~m}$ from one end. The metal was 0.16 cm thick, but $\sim 25 \%$ of each wall contained $1.3-\mathrm{cm}$-thick plastic windows. The other half of the hood contained an aluminum tubular structure (a split table), partially visible in Figs. 3 and 5.

A stainless-steel tank a few centimetres below the reflectors served as a distribution manifold directing solution to one tank or many cylinders as needed. The inside dimensions of this tank were 38.4 cm in diameter $\times 3.8 \mathrm{~cm}$ thick. The support

TABLE XV
Properties of the Plastic Reflector and Composition of Materials

|  | Total, All Six Panels | Top and Bottom Panels (Fire-Retardant Plastic) | All Four Side Panels (Nonfire-Retardant Plastic) |
| :---: | :---: | :---: | :---: |
| Density $\left(\mathrm{g} / \mathrm{cm}^{3}\right)$ Weight (kg) <br> Volume ( $10^{6} \mathrm{~cm}^{3}$ ) | $\begin{array}{r} 2920.1 \\ 2.3938 \end{array}$ | $\begin{gathered} 1.286 \\ 1040.8 \\ 0.8096 \end{gathered}$ | $\begin{gathered} 1.186 \\ 1879.3 \\ 1.5842 \end{gathered}$ |
| Composition (wt\%) |  |  |  |
| Hydrogen <br> Carbon <br> Nitrogen <br> Oxygen <br> Phosphorus <br> Chlorine <br> Bromine <br> Ash ${ }^{\text {b }}$ <br> Totals |  | $\begin{gathered} 7.18 \pm 0.16 \\ 52.68 \pm 0.10 \\ 0.10 \\ 29.40 \pm 0.32 \\ 1.54 \\ 1.63 \\ 6.50 \\ 0.71 \\ 99.74 \end{gathered}$ | $\begin{aligned} & 8.03 \pm 0.07 \\ & 59.72 \pm 0.05 \\ & \mathrm{a} \\ & 32.14 \pm 0.14 \\ & \mathrm{a} \\ & \mathrm{a} \\ & \mathrm{a} \\ & \mathrm{a} \\ & 99.89 \pm 0.16 \end{aligned}$ |

${ }^{\text {a Not measured. }}$
${ }^{\text {b }}$ Products of combustion at high temperature for a long time.
table (see Fig. 4) consisted of a $2.5-\mathrm{cm}$-thick $\times 155$ cm -square steel top supported by a heavy frame consisting of steel angle and channel pieces. The top weighed $\sim 470 \mathrm{~kg}$, and the remainder of the table weighed $\sim 400 \mathrm{~kg}$.

The most significant environmental reflector for minimally reflected single-tank measurements was one face of the hood described above. The tank was $\sim 2 \mathrm{~m}$ from the $4.9-\times 5.7-\mathrm{m}$ face and slightly below the top of the hood. The circular steel band, shown in Fig. 2 supporting auxiliary equipment, was 95 cm above the top of the tank. A massive steel table, weighing $\sim 5400 \mathrm{~kg}$, sat on the floor $\sim 4 \mathrm{~m}$ below the suspended tank. The table measured $2.1 \times 5.2 \mathrm{~m}$.

## IX. UNCERTAINTIES

Measured or estimated uncertainties in parameters are given in the tables along with the parameters themselves. Uncertainties, $\sigma_{x}$, associated with averaged measured parameters are the standard deviation of the measurements, $x_{i}$, and are calculated by

$$
\sigma_{i}=(N-1)^{-1 / 2} \sum_{i=1}^{N}\left(x_{i}-\bar{x}\right)^{2}
$$

Some uncertainties, such as those relating to some elemental composition determinations, are only estimates of the precision of a method, and these uncertainties may be taken to have a similar meaning
$( \pm \sim 1 \sigma)$. The uncertainty in the critical height data of Tables I and II equals half the range between measurements. Here, the confidence that the true value falls within the uncertainty would be greater than the confidence associated with one standard deviation. The critical height bias adjustment (see also Tables I and II) is usually larger than the uncertainty in the height itself. The uncertainty in this bias correction is unknown but probably exceeds the repeatability uncertainty by a small amount.

Still, the parameters expressed in this report are known with a precision considerably better than that presently required for validating calculational methods. Every experimental case presented in this paper has been calculated by the authors, ${ }^{16}$ and the uncertainty in the calculated critical height ${ }^{d}$ always exceeds the experimental uncertainties presented in Tables I and II, with one exception. In fact, the calculated uncertainty exceeds the experimental by a factor of 10 or more in over $80 \%$ of the cases. Based on these calculations, the experimental data uncertainties are acceptably small compared to calculational uncertainties.
${ }^{\mathrm{d}}$ Critical heights are obtained by calculating neutron reproduction factors ( $k_{\text {eff }}$ ) for two heights near criticality and linearly interpolating between the two to $k_{\text {eff }}=1.0$. These reproduction factors have a statistical uncertainty because they are based on a finite number of neutron histories. These statistical uncertainties translate into an uncertainty in the critical height.


Fig. 11. Reciprocal multiplication graphs for two experiments for which criticality could not occur.

Two critical heights presented in Table I (enclosed in square brackets) were not determined from critical measurements because the tank was slightly too short to permit criticality. Instead, they were obtained by extrapolating reciprocal multiplication data. Such extrapolations are quite subjective, and the uncertainty in the resulting critical height may be large because of the subjectivity. The last four measured reciprocal multiplication data (for two neutron detection channels) are given in Fig. 11 for these two cases. The reader may use these data on his own to obtain these critical heights and their uncertainties.

## X. ADDENDUM

Boron and cadmium are strong neutron absorbers, so the impurity levels, given at the end of Sec. IV, are presented below (in parts of impurity per million parts of uranium) in greater detail for the three solution concentration ranges studied in this paper:
(a) 334.77 to $369.96 \mathrm{~g} \mathrm{U} / \mathrm{\ell}$ : boron ( $8 \pm 9$ ), cadmium ( $36 \pm 15$ )
(b) 137.40 to $147.66 \mathrm{~g} \mathrm{U} / \mathrm{Q}$ : boron ( $7 \pm 5$ ), cadmium ( $37 \pm 18$ )
(c) 54.89 to $83.49 \mathrm{~g} \mathrm{U} / \mathrm{Q}:$ boron $(18 \pm 16)$, cadmium ( $42 \pm 21$ ).

## ACKNOWLEDGMENTS

This paper is respectfully dedicated to Clarence Lee Schuske, who died in an automobile accident on July 12, 1977, in Boulder, Colorado. Mr. Schuske had been active in the field of critical mass physics since 1948, and had managed the nuclear safety program at Rockwell International's Rocky Flats Plant for 25 years.

Thanks are due to G. Tuck, W. R. Sheets, N. D. Gaylord (now deceased), and D. E. Payne for their assistance in data collection and equipment maintenance. Deanne (Dickinson) Pecora performed many computations important to the design of the experiment. Many helpful discussions with
her and G. Tuck during the preparation of this paper are also gratefully recognized.

This work was performed for the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, under U.S. Department of Energy Contract EY-76-C-04-3533.

## REFERENCES

1. D. DICKINSON and G. E. WHITESIDES, "The Monte Carlo Method for Array Criticality Calculations," Nucl. Technol., 30, 166 (1976).
2. C. L. SCHUSKE and H. C. PAXTON, "History of Fissile Array Measurements in the United States," Nucl. Technol., 30, 101 (1976).
3. C. L. SCHUSKE et al., "Reference Critical Experiments Progress Report for Period July 1, 1975, Through December 31, 1975," RFP-NUREG-2481, Rockwell International, Rocky Flats Plant (1976).
4. "Validation of Calculational Methods for Nuclear Criticality Safety," ANSI-N16.9, American National Standards Institute (1975).
5. H. NELTRUP, "Benchmark Calculations on Homogeneous Spheres," RISO-M-1526, Riso, Roskilde, Denmark (1972).
6. R. E. ROTHE, "Critical Measurements on an Enriched Uranium Solution System," Nucl. Sci. Eng., 35, 267 (1969).
7. D. C. HUNT and R. E. ROTHE, "Criticality Measurements on Uranium Metal Spheres Immersed in Uranium Solution," Nucl. Sci. Eng., 46, 76 (1971).
8. D. C. HUNT and R. E. ROTHE, "A Criticality Study of Fissile Metal and Fissile Solution Combinations," Nucl. Sci. Eng., 53, 79 (1974).
9. R. E. ROTHE and D. C. HUNT, "The Effect of Position on the Criticality of Uranium Spheres in Uranium Solution Cylinders," Nucl. Sci. Eng., 54, 360 (1974).
10. R. E. ROTHE, D. L. ALVAREZ, and H. E. CLARK, "The Criticality of Periodically Boron-Poisoned Enriched Uranium Solution Systems," Nucl. Technol., 25, 502 (1975).
11. D. DICKINSON, "Calculational Study of Arrays of Cylinders of Fissile Solution," RFP-1821, Dow Chemical U.S.A., Rocky Flats Plant (1972).
12. J. T. THOMAS, "Critical Three-Dimensional Arrays of Neutron-Interacting Units, Part I, Aqueous U(92.6) Solutions," ORNL-TM-719, Oak Ridge National Laboratory (1963).
13. W. R. SHEETS, "An Electromechanical Level Sampler," Instrum. Control Systems, 40 (1972).
14. "Reactor Physics Constants," ANL-5800, Argonne National Laboratory (1963).
15. L. M. PETRIE and N. F. CROSS, "KENO-IV-An Improved Monte Carlo Criticality Program," ORNL-4938, Oak Ridge National Laboratory (1975).
16. I. OH and R. E. ROTHE, "A Calculational Study of Benchmark Critical Experiments on High-Enriched Uranyl Nitrate Solution Systems," Nucl. Technol., 41, 226 (1978).

[^0]:    ${ }^{\text {a }}$ See Addendum on p. 224 of this paper.

[^1]:    ${ }^{\mathrm{b}}$ Manufactured by Carboline, St. Louis, Missouri.

[^2]:    ${ }^{\text {a }}$ Nominal wall and bottom thicknesses in all cases were 0.32 and 0.64 cm , respectively.
    ${ }^{\mathrm{b}}$ Nominal $2.54-\mathrm{cm}-\mathrm{o}$. tubing with $0.12-\mathrm{cm}$-thick wall except for footnote e.
    ${ }^{\text {c }}$ See text.
    ${ }^{\mathrm{d}}$ These two are actually the same tank. Only the height was changed for different experiments.
    ${ }^{\mathrm{e}}$ Nominal $2.22-\mathrm{cm}-\mathrm{o} . \mathrm{d}$. tubing with $0.12-\mathrm{cm}$-thick wall.

[^3]:    *Distance in centimetres from center of the underside of the bottom of a tank to each of the six reflecting surfaces. The range metres.
    ${ }^{\text {a }}$ Table containing the corresponding critical height data.
    ${ }^{\mathrm{b}}$ Approximately centered in a large room having thick concrete walls.

[^4]:    ${ }^{\text {c }}$ The north wall is 152 cm thick; the other three walls are 122 cm thick. The ceiling is 61 cm thick, and the floor is 20.3 cm thick, resting, of course, on earth.

[^5]:    ${ }^{\mathrm{d}}$ Plexiglas, trademark of Rohm and Haas Company, Philadelphia, Pennsylvania.

