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CALCULATIONAL STUDY OF ARRAYS OF CYLINDERS
OF FISSILE SOLUTION

Deanne Dickinson

DOW CHEMICAL U.S.A.
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401

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Research and Ecology

NUCLEAR SAFETY GROUP

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CONTENTS

Abstract	1
Introduction	1
Methods of Calculation	1
Computer Codes	1
Material Properties	1
Calculation of Critical Parameters	2
Calculations of k_{eff} for Experimentally Critical Systems	2
Calculated Critical Parameters for Bare and Reflected Fissile Solution Systems	4
Effect of a Plexiglas Reflector around a Sphere of Fissile Solution	4
Unreflected Linear Arrays of Cylinders of Uranyl Fluoride (UO_2F_2) Solution	5
Plexiglas Reflected Arrays of Cylinders	5
Concrete Reflected Arrays of Cylinders	7
Two Proposed Designs Involving Double-Line Arrays	7
Ion-Column Array	9
Solution Process Area	9
References	9

CALCULATIONAL STUDY OF ARRAYS OF CYLINDERS OF FISSILE SOLUTION

Deanne Dickinson

Abstract. Values of k_{eff} for experimentally critical systems of enriched uranium or plutonium solution are calculated using the DTF-II, KENO, and KENO II computer codes. Both KENO and KENO II underestimate k_{eff} (effective neutron multiplication factor) for minimally reflected uranium-solution systems, indicating that some correction is needed if these codes are to be used conservatively in nuclear safety calculations. Calculated critical parameters are presented for line arrays of long cylinders of fissile solution reflected by Plexiglas or concrete. The reflector is a cuboidal shell, simulating a room, and the distance from the sides of the shell to the array is varied. Two examples of double-line arrays are presented.

INTRODUCTION

The storage and processing of fissile solution are often done in annular cylinders or in long cylinders of small diameter to satisfy nuclear safety requirements. In this report, only the case of long (*effectively infinite* length) cylinders will be considered. For a safe long cylinder, a necessary requirement would be to have a diameter smaller than the critical diameter for an infinite cylinder containing the same fissile solution and having the same external reflection. One-dimensional transport theory can be used to estimate the critical infinite cylinder diameter, and some data are available from Paxton *et al.* (7).¹

In actual situations, one usually has a one- or two-dimensional array of cylinders with a variety of reflectors at different distances from the array. These reflectors include the walls, floor, and ceiling of the room containing the cylinders and, in some cases, glove boxes and biological shielding.

Some of the parameters which affect the reactivity of a fissile-solution system are the solution composition, the container shape and material, the number of units, the arrangement of units (configuration and spacing), and the presence of moderators or reflectors. In this report, the number of parameters has been reduced by considering linear arrays of long cylinders. Both the cylinder length and the array length are effectively infinite; i.e., increasing the cylinder height or extending the array by adding more units would have a negligible effect on system reactivity. The variables studied include cylinder diameter and spacing, and the effect of the position of an external reflector.

The first section of the report is concerned with establishing the validity of the computer codes and cross-section data used by performing calculations on experimentally determined critical systems. The next section presents calculated critical parameters for line arrays, both bare and reflected, for uranium and plutonium solutions. The information has application in preliminary design calculations involving line arrays. Finally, two examples of double-line arrays considered in the planning of a new building are discussed.

METHODS OF CALCULATION

Computer Codes:

The Monte Carlo codes KENO (11) and KENO II (12) were used for most of the calculations, since a three-dimensional geometry description was necessary. KENO II is an improved version of the KENO code having a similar neutronics calculation and a more versatile geometry routine. Some calculations on spherical systems were done with the one-dimensional transport code, DTF II (3). All three codes use the Hansen-Roach sixteen group cross sections (5).

Calculated values of k_{eff} , the effective neutron multiplication factor, are given with error bars of one standard error, denoted as sigma (σ); i.e., the 68-percent confidence interval. In recent versions of KENO and KENO II, the Shapiro-Wilk test for normality (9) is used to determine if the k_{eff} values are approximately normally distributed, and hence if the sample standard error is a reliable estimate of the error in the average k_{eff} . Calculations which fail the Shapiro-Wilk test are indicated.

Material Properties:

All references to uranyl fluoride (UO_2F_2) solution are to the uranium solution described in Table I. The solution properties are taken from Fox *et al.* (4), where it is stated that this concentration gives the minimum critical volume for reflected systems. Similarly, all references to 200 grams per liter (g/l) plutonium nitrate [$\text{Pu}(\text{NO}_3)_4$] solution are to the plutonium solution described also in Table I. This is an *artificial* solution, since it contains no excess nitric acid, and hence is more reactive than real solutions, which always contain some excess nitric acid. The 200-g/l concentration is believed to be close to the minimum critical volume concentration for aqueous solutions of plutonium nitrate containing 5 percent of plutonium-240 (^{240}Pu) and no excess acid.

¹ Numerals in parentheses relate to references given at end of text.

TABLE I. Fissile Solution Compositions.

	Uranyl Fluoride (UO_2F_2) Solution	Plutonium Nitrate ($\text{Pu}(\text{NO}_3)_4$) Solution
Concentration	576.82 grams of uranium per liter	200 grams of plutonium per liter
Enrichment	93.2 weight percent of uranium-235 (^{235}U)	95 weight percent of plutonium-239 (^{239}Pu)
Specific Gravity	1.661	1.316
$^a\text{H/X}$	$\text{H}/^{235}\text{U} = 44.3$	$\text{H}/^{239}\text{Pu} = 126.9$

^aH/X = ratio of number of hydrogen atoms per unit volume to number of fissile atoms per unit volume

Calculation of Critical Parameters:

Critical parameters are calculated by linear interpolation, using points close enough to critical that any departure from linearity is negligible compared to statistical errors. If only two data points were available, the critical value of the parameter was calculated by linear interpolation between them, and errors were estimated by the formula² for propagation of errors (1). If more than two data points were found, a linear least squares fit was made, and the resulting equation was solved for the critical value of the parameter. The error is estimated by the method described in Kenney *et al.* (6, Section 8.10). *It is important to note that, because the random variable involved is not normally distributed, one cannot assume any relation between the error bars for the 68-percent confidence interval and the error bars for any other confidence interval.*

Calculations of k_{eff} for Experimentally Critical Systems:

All three computer codes used in this report were checked against experimentally determined critical systems for both uranium and plutonium solutions, bare and reflected.

Several KENO and KENO II calculations were done on arrays of cylinders of UO_2F_2 solution (as described in Table I) using the critical data of Fox *et al.* (4). The calculational results shown in Table II imply that both codes give values of k_{eff} about 1 to 2-percent low for minimally reflected systems. Further calculations showed

that this is equivalent to overestimating the critical diameter by 3 to 4 percent. Calculations for water-moderated and reflected arrays were in good agreement with experiment, with the exception of one case for which k_{eff} was too high by about 3 percent.

Since the 200-g/l plutonium solution is artificial, no comparisons with experiment are possible. However, transport and diffusion-theory calculations have been done (8) which show that Hansen-Roach cross sections give conservative results for spherical $\text{Pu}(\text{NO}_3)_4$ solution systems, both bare and reflected, for plutonium concentrations ranging from 24.4 to 268 g/l and ^{240}Pu content ranging from 0.54 to 4.6 percent.

Several cases from Richey (8) and Bouly *et al.* (2) were checked using KENO and KENO II. The results are presented in Tables III and IV. The KENO code gives $k_{\text{eff}} > 1$ for all the plutonium solutions, both bare and reflected. KENO II gives satisfactory results for water-reflected plutonium solution systems, but low values of k_{eff} for two of the six bare systems. The reason for the low values of k_{eff} for the bare solution spheres (Table III) is not known, but it may be the omission of the room and surrounding apparatus.

Most of the calculations in this report are for arrays reflected by a cuboidal shell of Plexiglas or concrete. Data on uranium-solution cylinder arrays interacting with a solution slab and reflected by Plexiglas are given by Tuck and Clark (10), and Monte Carlo calculations for these systems are presented in Table V. The results are in excellent agreement with the experimental data.

The results of all calculations on experimental data are summarized in Table VI. In two cases, the average k_{eff} is less than one (bare uranium solution with KENO or KENO II), indicating that some correction is needed if calculations are to be used conservatively for nuclear safety considerations. The magnitude of these corrections is not known and would probably have to be determined separately for each specific geometry. Also, no experimental data are available for units and arrays reflected by a cuboidal shell of concrete, and it is exactly this case which occurs in practice for solution systems contained in a concrete room. A comparison of the energy spectra of neutrons causing fissions for the various cases shows that the concrete-shell reflector results in a spectrum intermediate between that of the bare case and the Plexiglas-shell reflected system. However, it is impossible to determine from the available data whether calculations on solution arrays in a concrete shell reflector are conservative.

²The method was pointed out in discussion with Robert E. Rothe of Nuclear Safety, who also derived the specific formula used.

TABLE II. Results of Monte Carlo Calculations for Experimentally Critical Arrays of Cylinders Containing Uranyl Fluoride Solution.

Number of Cylinders	Pattern	Solution Height (centimeters)	Center-to-Center Separation (centimeters)	Reflector	$k_{eff} \pm \sigma$	Computer Code
5	Line	78.74	28.24	Tank, wall	0.989 ± 0.007	KENO
5	Line	40.13	21.01	Tank, wall	0.984 ± 0.007	KENO
5	Line	40.13	21.01	Tank, wall	0.983 ± 0.008	KENO II
3	Line	45.72	21.01	Tank, wall	0.975 ± 0.009	KENO II
7	Hexagon	55.88	43.48	Tank, wall	0.979 ± 0.008	KENO II
2	Line	21.08	28.24	Water	0.991 ± 0.013	KENO
2	Line	17.53	21.01	Water	1.009 ± 0.009	KENO
3	Equilateral Triangle	21.34	30.78	Water	0.994 ± 0.011	KENO
7	Hexagon	14.99	25.70	Water	1.034 ± 0.010	KENO
4	Line	20.83	28.24	Water	^a 1.007 ± 0.009	KENO II

NOTE: The experimental data were given by Fox *et al.*, see Reference 4. The fissile solution is uranyl fluoride (UO_2F_2) as described in Table I. The cylinders are aluminum with an inner diameter of 8 inches and a wall thickness of 0.06 inches. The cylinders are in a tank 9.5 feet in diameter, 10 feet high, lateral wall of 0.375-inch mild steel clad with 0.063-inch stainless steel, bottom 1-inch mild steel clad with 0.063-inch stainless steel. The bottoms of the solution cylinders are 12 inches above the tank bottom. For the reflected arrays, the tank is filled with water to the level of the solution in the cylinders. For the arrays not reflected by water, one concrete wall was placed 6 feet from the tank.

^aThis case fails the Shapiro-Wilk test at the 1-percent significance level (see Reference 9).

TABLE III. Results of Transport and Monte Carlo Calculations for Critical Spheres of Plutonium Nitrate Solution.

Sphere Diameter (centimeters)	20-Centimeter Water Reflected	Plutonium Concentration (grams per liter)	Weight Percent of Plutonium-240	H ⁺ Normality	$k_{eff} \pm \sigma$	Computer Code
29.21	Yes	132.	4.6	2.25	1.027 ± 0.008	KENO
29.21	Yes	132.	4.6	2.25	^a 0.991 ± 0.009	KENO II
29.21	Yes	268.7	4.6	1.08	1.046	DTF II
29.21	Yes	268.7	4.6	1.08	1.052 ± 0.008	KENO
29.21	Yes	268.7	4.6	1.08	1.019 ± 0.011	KENO II
35.56	Yes	27.91	0.54	1.72	1.000 ± 0.010	KENO II
38.61	No	39.	4.6	0.363	1.015	DTF II
38.6	No	39.	4.6	0.363	1.029 ± 0.008	KENO
38.6	No	39.	4.6	0.363	0.987 ± 0.008	KENO II
38.6	No	172.8	4.6	4.77	1.001 ± 0.007	KENO
38.61	No	172.8	4.6	4.77	0.979 ± 0.007	KENO II

NOTE: Critical parameters were obtained from Tables I and III of Reference 8, C. R. Richey.

^aTest significant at 10 percent level, see Reference 9, Shapiro and Wilk.

TABLE IV. Results of Monte Carlo Calculations for Experimentally Critical Arrays of Plutonium Nitrate Solution in Air.

Number of Cylinders	Pattern	Center-to-Center Separation		$k_{eff} \pm \sigma$	Computer Code
		Solution Height (centimeters)			
1	—	42.53	—	1.018 ± 0.009	KENO
2	Line	39.72	60.	1.027 ± 0.008	KENO
3	Equilateral Triangle	37.30	60.	1.014 ± 0.009	KENO II
3	Equilateral Triangle	39.90	90.	1.001 ± 0.008	KENO II
3	Right Triangle	26.58	^a 30.	1.005 ± 0.009	KENO II
4	Square	22.96	30.	1.002 ± 0.010	KENO II

NOTE: The critical experiments were reported in Reference 2. The fissile solution is 152.5 grams per liter plutonium nitrate, 2.24 normal, 95.47 weight percent plutonium-239. The cylinders are 29.4-centimeters (cm) inside diameter with 0.3-cm thick stainless steel walls. The hood and room were included in the geometry description, but several calculations without them showed that their effect on k_{eff} is small (≈ 2 percent maximum).

^aThe separation refers to distances along the legs of the right triangle only.

TABLE V. Monte Carlo Calculations for Uranyl Nitrate $[UO_2(NO_3)_2]$ Solution Cylinder Arrays in a Solution Slab Reflected by a Cuboidal Shell of Plexiglas.

Number of Cylinders in Array	Solution Slab Thickness (centimeters)	^a Height of Solution in Cylinders (centimeters)	Uranium Concentration (grams per liter)	Top and Side Reflector Thickness (centimeters)	$k_{eff} \pm \sigma$	Computer Code
9	9.5	20.	505.	10.2	1.007 ± 0.010	KENO
16	0.	92.	505.	10.2	1.015 ± 0.010	KENO II
16	3.8	61.	505.	10.2	0.999 ± 0.009	KENO
16	0.	96.	510.	7.6	0.996 ± 0.011	KENO II
16	0.	105.	510.	5.1	1.005 ± 0.011	KENO II

NOTE: Experimental critical data are from Table VI of Reference 10, Tuck and Clark. The cylinder diameter is 13.6 centimeters (cm). The cylinder walls were of 0.28-cm stainless steel. The bottom reflector thickness is 10.2-cm in all cases.

^aMeasured from top of solution slab.

TABLE VI. Average k_{eff} for each Type of System Represented in Tables II, III, IV, and V.

System		KENO	KENO II
Uranium Solution	Bare	0.987 ± 0.004	0.979 ± 0.004
	Plexiglas Shell Reflected	1.003 ± 0.006	1.005 ± 0.010
	Reflected	1.007 ± 0.020	^a 1.007
Plutonium Solution	Bare	1.019 ± 0.013	0.998 ± 0.013
	Reflected	1.040 ± 0.018	1.003 ± 0.014

^aOnly one data point fits in this category.

CALCULATED CRITICAL PARAMETERS FOR BARE AND REFLECTED FISSILE SOLUTION SYSTEMS

The purpose of this section is to provide calculated values of critical parameters for linear arrays of cylinders of fissile solution. While no design problem is expected to fit exactly the situations considered, the data provided can serve as a starting point for calculations.

Effect of a Plexiglas Reflector around a Sphere of Fissile Solution: Before studying the effect of reflector position on an array, consider first the simpler problem of a single sphere of fissile solution reflected by a 15.24 centimeter (cm) thick shell of Plexiglas and vary the inner radius of

TABLE VII. Calculations for a Sphere of Uranium or Plutonium Solution Reflected by a Shell of Plexiglas.

Gap (centimeters)	Critical Radius (centimeters)		Reflector Savings (centimeters)		Ratio of Plutonium Critical Radius to Uranium Critical Radius
	Plutonium	Uranium	Plutonium	Uranium	
0	13.1	11.0	4.6	4.3	1.19
30.48	16.9	14.6	0.8	0.7	1.16
60.96	17.4	15.0	0.3	0.3	1.16
182.88	17.7	15.3	0.0	0.0	1.16
∞	17.7	15.3	0.0	0.0	1.16

NOTE: The solution properties are given in Table I. The calculational geometry is shown as follows:

the shell. Later, the same problem will be treated for the case of a Plexiglas-shell reflector around a line array of solution cylinders.

The case of a single sphere may be calculated using the DTF-II code (3). The results are shown in Table VII and refer also to Figure 1. For the reflector at a distance of 30 cm or more, the critical radius of the plutonium-solution sphere is larger than the critical radius of the uranium-solution sphere by a constant 16 percent. No such constant relationship has been found for the case of a line array of cylinders.

Unreflected Linear Arrays of Cylinders of Uranyl Fluoride (UO_2F_2) Solution: A series of KENO calculations was done to find the variation in critical separation as a function of the number of cylinders in a bare, linear array of

20.32-cm diameter, 250-cm long cylinders of uranyl fluoride (UO_2F_2). The cylinders have 0.15-cm thick aluminum walls. These results are shown in Figure 2. The critical center-to-center separation is approximately constant (at 33.4 cm) for any number of cylinders greater than 20. Later calculations, using the KENO II code (12), showed that an infinite line of 20.32-cm diameter cylinders at 33.3-cm center-to-center spacing had a k_{eff} of 0.986 ± 0.006 , compared to $k_{\text{eff}} = 0.981 \pm 0.006$ for a line of 30 cylinders. (The KENO code cannot handle infinite arrays.)

As a result of these calculations, further calculations on line arrays were done for a line of 30 cylinders. According to Figure 2 and the KENO II calculations, 30 cylinders are *effectively infinite*.

FIGURE 1. Critical Radius of a Uranium or Plutonium Solution Sphere as a Function of the Gap between the Sphere and a Plexiglas Shell Reflector (15.24 centimeters thick).

FIGURE 2. Critical Center-to-Center Spacing for Unreflected Line Arrays of 20.32-Centimeter Diameter Cylinders of Uranyl Fluoride Solution versus Number of Cylinders in Array.

FIGURE 3. Leakage Spectrum for Critical Line Array of Thirty 20.32-Centimeter Diameter Cylinders of Uranyl Fluoride with a Center-to-Center Separation of 33.3 Centimeters.

The leakage spectrum by energy group for an unreflected line array of 30 cylinders is shown in Figure 3. The ordinate of the graph is the neutron leakage fraction. The leakage fraction for an energy group is the probability that a neutron produced in the system will leave the system with an energy in the specified range.

Plexiglas Reflected Arrays of Cylinders: The effect of a 15.24-cm thick cuboidal shell Plexiglas reflector was studied for a line array of 30 cylinders, each cylinder having a height of 250 cm. The center-to-center spacing of the cylinders was fixed, and the critical cylinder diameter was calculated using the KENO code for several reflector positions. The reflector was always kept in contact with the top and bottom of the cylinder array, and the distance of the lateral walls of the reflector from the lateral walls of the cylinders was varied. The results of these calculations are shown in Table VIII.

For the 33.02-cm center-to-center spacing, the plutonium critical diameter is always greater than the corresponding value for uranium but, unlike the case of the reflected single sphere, the increase varies from 6 to 18 percent. Some of this variation may be the result of statistical uncertainty in the calculated critical diameters.

Comparing the critical diameters for uranium solution at different center-to-center spacings, one finds that the critical diameters for the 60.96-cm spacing are larger than those for the 33.02-cm spacing by 6 to 14 percent. Again, some of this variation may be the result of statistical uncertainty.

Concrete Reflected Arrays of Cylinders: The KENO II code was used to calculate the critical spacing for an infinitely long line of cylinders of UO_2F_2 solution of fixed diameter reflected by a shell of concrete 30.48 cm thick. The concrete was 30.48 cm from the lateral wall of the cylinder. The results are given in Table IX. These data can be used conservatively to estimate the critical spacing for a line array of cylinders in a concrete room, as long as the nearest wall is at least 30.48 cm away.

TWO PROPOSED DESIGNS INVOLVING DOUBLE-LINE ARRAYS

Two examples of double-line arrays of long cylinders of interest for use at Rocky Flats are considered. Monte Carlo calculations are given for normal and abnormal conditions.

TABLE VIII. Critical Diameters and Reflector Savings for a Line of Thirty Cylinders (250-centimeter height) of Uranium or Plutonium Solution Reflected by 15.24 centimeters of Plexiglas.

^a Lateral Reflector Separation (centimeters)	^b Critical Cylinder Diameter (centimeters)			^b Reflector Savings on Diameter (centimeters)		
	Uranium Solution		^c Plutonium Solution	Uranium Solution		Plutonium Solution
	33.02-centimeters Center-to-Center Spacing	60.96-centimeters Center-to-Center Spacing	33.02-centimeters Center-to-Center Spacing	33.02-centimeters Center-to-Center Spacing	60.96-centimeters Center-to-Center Spacing	33.02-centimeters Center-to-Center Spacing
0	13.31 ±0.49	14.83 ±0.05	14.46 ±0.40	6.91 ±0.51	6.63 ±0.09	8.81 ±0.44
30.48	14.55 ±0.71	16.36 ±0.08	16.31 ±0.61	5.67 ±0.72	5.10 ±0.11	6.96 ±0.64
60.96	16.05 ±1.14	18.19 ±0.15	17.04 ±0.13	4.17 ±1.15	3.27 ±0.17	6.23 ±0.22
182.88	17.86 ±0.13	18.77 ±0.05	20.09 ±0.51	2.36 ±0.18	2.69 ±0.09	3.18 ±0.54
∞	18.80 ±0.08	20.42 ±0.13	22.20 ±0.05	1.42 ±0.15	1.04 ±0.15	1.07 ±0.19
^d No Reflector	20.22 ±0.13	21.46 ±0.08	23.27 ±0.18	0.0	0.0	0.0

^aThe reflector is in contact with the tops and bottoms of the cylinders.

^bError bars are about 70-percent confidence limits.

^cThese calculations were performed by a staff member, Robert E. Rothe.

^dTop and bottom reflectors removed also.

TABLE IX. Critical Center-to-Center Separation for an Infinite Line of Cylinders of Uranyl Fluoride (UO₂F₂) Solution Reflected by 30.48 Centimeters of Concrete (KENO II Calculations).

Cylinder Diameter (centimeters)	^a Critical Center-to-Center Spacing (centimeters)
13.97	47.9 ±2.3
15.24	58.6 ±1.3
17.78	117.5 ±6.4

Note: Cylinders are 250 centimeters (cm) long and have 0.32-cm thick stainless steel walls. Concrete is in contact with cylinder top and bottom and is 30.48 cm away from lateral surface.

^aErrors are approximately 70-percent confidence limits.

TABLE X. Results of KENO Calculations on Ion-Column Arrays.

Center Wall	^a Solution	Boron in Glass	$k_{eff} \pm \sigma$
Present	Plutonium	Yes	0.660 ±0.010
^b Absent	Plutonium	Yes	0.645 ±0.009
Present	Plutonium	^c No	0.672 ±0.009
Present	Uranium	Yes	0.722 ±0.010

^aThe solution properties are given in Table I.

^bThe center wall might be destroyed in case of an accident.

^cSince the boron content of the glass is not part of the design specification, it cannot be relied on to poison the system.

Ion-Column Array:

A proposed ion-column array consists of a double line of cylinders separated by a water wall for biological shielding. Each line contains 6 ion columns, and the entire array is reflected below and on all 4 sides by a 5.1 cm thick cuboidal shell of water. The geometry of the array and the dimensions are shown in Figure 4. The ion columns normally contain plutonium nitrate solution at a concentration of less than 150 grams per liter (g/l). Calculated results are shown in Table X.

For the particular geometry chosen, the array is slightly more reactive with the 10.2-cm thick center wall than without it, but the difference is not significant at the 95-percent confidence level. The boron content of the cylinder glass reduces the reactivity slightly, but again the difference is not statistically significant. Replacing the plutonium solution by uranium solution increases k_{eff} by 9 percent. One can estimate from Table VIII that the critical diameter for UO_2F_2 solution in this configuration is greater than 15.5 cm.

Solution Process Area:

A proposed area for plutonium-solution processing consisted of a concrete room, 975 cm long and 305 cm wide. Included were a line of cylinders containing concentrated (200 g/l maximum) solution along one 975-cm long wall and a line of cylinders containing dilute (7 g/l maximum) solution along the opposite wall. Details are noted in Figure 5. KENO II was used to calculate k_{eff} for three cases. For the normal case, as described above, $k_{eff} = 0.694 \pm 0.008$. For 200-g/l plutonium solution in both rows, $k_{eff} = 0.835 \pm 0.008$. Finally, for comparison, both rows of cylinders were filled with UO_2F_2 solution (as described in Table I, Page 2), resulting in $k_{eff} = 0.972 \pm 0.010$.

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FIGURE 4: Ion-Column Array.

FIGURE 5. Double-Line Array of Cylinders. (All dimension in centimeters.) (Each cylinder has a 13.34-cm inner diameter; 609.6-cm inner height; and wall, top, and bottom of Type-304 stainless steel, 0.635-cm thick. Floor and ceiling are 30.48-cm thick concrete.

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