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Criticality Experiments with Planar Arrays of Three-Liter Bottles Containing Plutonium Nitrate Solution

B. M. Durst
E. D. Clayton
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January 1985

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

Pacific Northwest Laboratory
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CRITICALITY EXPERIMENTS WITH PLANAR ARRAYS OF THREE-LITER BOTTLES CONTAINING PLUTONIUM NITRATE SOLUTION

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MASTER

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SUMMARY

The storage of nuclear materials in arrangements so as to preclude criticality by accident is an item for concern throughout the nuclear fuel cycle. In the reprocessing of nuclear fuels, such as in the Purex Facility at Hanford, plutonium nitrate solutions are commonly stored in stainless steel, on polyethylene, cylindrical bottles. Because experimental data on the criticality of interacting arrays of Pu solution containers, as might occur in storage and handling, were lacking, the Rockwell Hanford Operations sponsored a series of experiments at the Critical Mass Laboratory of PNL with interacting arrays of bottles containing plutonium nitrate solutions. The objective of these experiments was to provide benchmark data to validate calculational codes used in criticality safety assessments of plant configurations. Arrays containing up to as many as sixteen three-liter bottles filled with plutonium nitrate were used in the experiments. A split-table device was used in the final assembly of the arrays. The planar arrays were reflected with close fitting plexiglas on each side and on the bottom but not the top surface. The experiments addressed a number of factors effecting criticality, namely: 1) the critical air gap between bottles in an array of fixed number of bottles, 2) the number of bottles required for criticality if the bottles were touching, and 3) the effect on critical array spacing and critical bottle number due to the insertion of an hydrogeneous substance into the air gap between bottles. Each bottle contained about 2.4ℓ of $Pu(NO_3)_4$ solution at a Pu concentration of 105g Pu/ℓ, with the 240Pu content being 2.9 wt% at a free acid molarity H^+ of 5.1.

After the initial series of experiments were performed with bottles separated by air gaps, plexiglas shells of varying thicknesses were placed around each bottle to investigate how moderation between bottles effects both the number of bottles required for criticality and the critical spacing between each bottle.

The minimum number of bottles required for criticality was found to be 10.9 bottles, occurring for a square array with bottles in contact. As the bottles were spaced apart, the critical number increased. For sixteen bottles in a square array, the critical separation between bottle surfaces in both x and y directions was 0.96cm. The addition of plexiglas around each bottle decreased the critical bottle number, compared to those separated in air, but the critical bottle number, even with interstitial plastic in place was always greater than 10.9 bottles. The most reactive configuration was a tightly packed array of bottles with no intervening material.

As a result of these experiments, calculational benchmarks now exist for criticality analysis of bottle arrays of plutonium nitrate solutions.

CONTENTS

FIGURES

TABLES

INTRODUCTION

The safe handling of nuclear fuel is a prime concern of the nuclear fuel cycle, worldwide. The criticality of interacting arrays of fissionable material is important, particularly in fuel reprocessing, storage and shipment, but there remains a singular lack of critical experiment data on interacting arrays of solution containers of plutonium on which to validate calculations used in criticality safety assessments.

To provide such data, a series of critical experiments, funded by the Rockwell Hanford Operation, were performed at the Critical Mass Laboratory (CML), of the Pacific Northwest Laboratory (PNL) with planar arrays of three-liter polyethylene bottles containing plutonium nitrate solution. The experiments are somewhat similar to those performed at the CML on bottles of 233U solution reported in 1968.(1) Plexiglas reflected arrays containing up to sixteen bottles, each bottle containing about 2.4 liters of solution, at a concentration of 105 g Pu/ℓ were used in the experiments. The plutonium was of low 240Pu content (2.9 wt% 240Pu). The experiments provide data on various factors effecting criticality of the arrays including information on: 1) The critical number of bottles in various planar arrays as a function of the spacing between bottles, 2) Number of bottles required for criticality with bottles in contact, 3) Effect on critical array spacing and critical number of bottles due to insertion of an hydrogeneous substance into the air gap between bottles of an array.

EXPERIMENT DESIGN

The Fissile Solution

The fissile solution used in these experiments is described in Table 1. It had an average concentration of 105 g/ℓ Pu with the 240Pu content being 2.9 wt% at a free acid molarity (H^+) of 5.1. The solution specific gravity was determined to be 1.420. Prior to bottle filling, the solution was thoroughly mixed to ensure the same concentration of plutonium in each bottle.

TABLE 1. Description of Plutonium Nitrate Solution

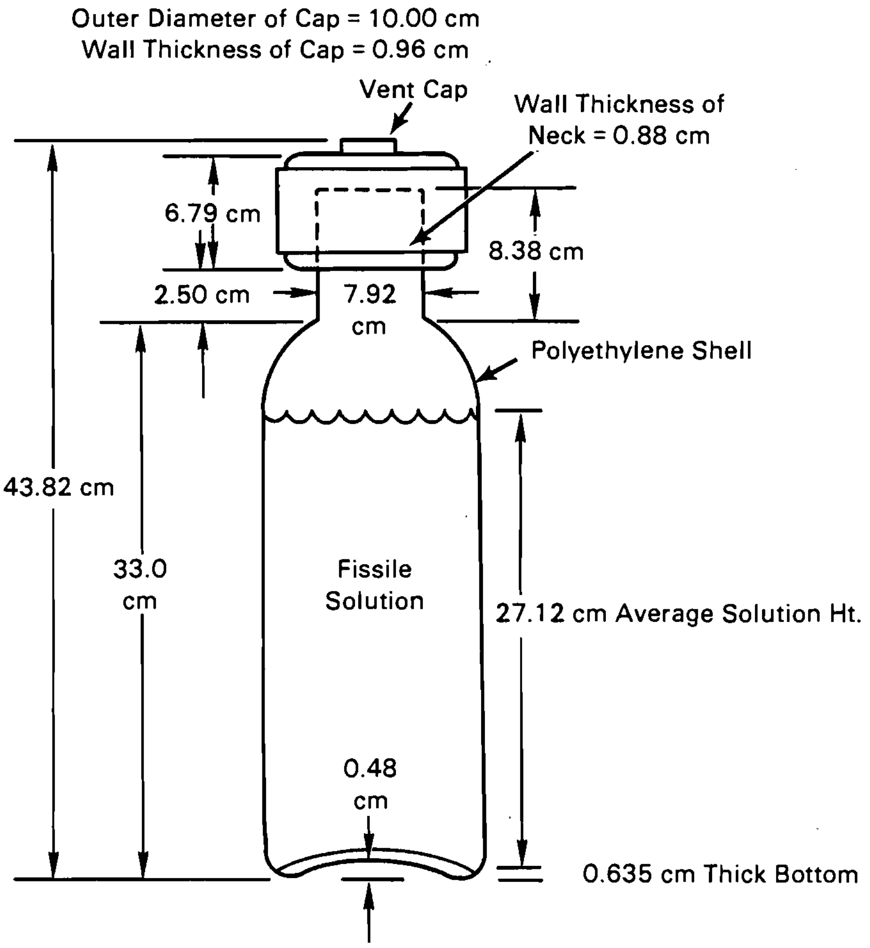
Component	Concentration (g/ℓ)
Plutonium	105
Uranium	3.1
Nitrate (NO_3)	505 (Total Nitrate)
Iron	3.2
Chromium	0.8
Nickel	0.6
Aluminum	8.0
Manganese	0.7
Cadmium	0.0005
Boron	0.005
Water	788.3 (obtained by difference)
^{241}Am (4/6/83)	0.18

Acid Molarity (H^+)	Specific Gravity
5.1	1.420 (g/cc)

Isotope	Wt%
^{238}Pu	0.011
^{239}Pu	96.942
^{240}Pu	2.882
^{241}Pu	0.119
^{242}Pu	0.046

The Experiment Equipment

The experiments were performed with sixteen three-liter polyethylene bottles (density of polyethylene 0.98 g/cm^3) filled with $Pu(NO_3)_4$. The bottles are described in Figure 1. The volume of solution in each individual container, shown in Table 2, ranged from 2.39 to 2.42 with an average of 2.407 ± 0.011. The average outside diameter of the bottles was 11.78 ± 0.04cm. The inner bottle diameters, also shown in Table 2, ranged from 10.53 to 10.68cm with the average being 10.63 ± 0.05cm. Based on these data, the average solution height per bottle was 27.12 ± 0.30cm. In every instance, the solution height was limited to levels below the curved neck of the L-3 bottle (see Figure 1).



Average Bottle Dimensions (Over the Portion of Bottle Filled with Solution)

Average Solution Ht.	- 27.12 ± 0.30 cm
Average Inner Diameter	- 10.63 ± 0.05 cm
Average Outer Diameter	- 11.78 ± 0.04 cm
Average Volume	- 2.407 ± 0.011 ℓ

Composition of Polyethylene Bottle

Chemical Form: $(CH_2)_n$

Isotope	Wt%
C	85.63
H	14.37

Composition of Plexiglas Reflector

Isotope	Wt%
H	8.0
C	60.0
O	32.0

Density of Polyethylene Bottle: 0.98 ± 0.04 g/cm^3

Weight of Empty Bottle: 987.0 ± 18.4 g

Density of Plexiglas: 1.185 g/cm^3

FIGURE 1. Description of L-3 Bottle

TABLE 2. Description of L-3 Bottles Used in Experiments

Bottle Number	Outside Diameter (cm)	Inside Diameter (cm)	Volume of(a) Solution (ℓ)	Solution Height(b) in Bottle (cm)
1	11.740	10.533	2.399	27.538
2		10.571	2.399	27.333
3		10.604	2.436	27.580
4		10.535	2.410	27.644
6		10.645	2.414	27.123
7		10.605	2.417	27.361
8		10.640	2.411	27.116
10		10.681	2.395	27.725
11		10.681	2.423	26.041
12	11.826	10.678	2.402	26.825
13	11.766	10.643	2.398	26.954
14		10.681	2.403	26.824
15		10.681	2.401	26.801
16		10.678	2.403	26.831
17		10.602	2.401	27.195
18		10.640	2.400	26.993
Average	11.777 ±0.044cm	10.631 ±0.051cm	2.407 ±0.011ℓ	27.118 ±0.298cm

---

(a) Based on specific gravity of solution and net weight of solution per bottle. Also, volumes were checked against a known volume.

(b) Calculated depending on volume per bottle and inside bottle diameter.

All arrays were reflected with plexiglas on all sides and the bottom as shown in Figure 2. The top was unreflected. Bottles were accurately positioned with the help of an aluminum framework, shown in both Figures 2 and 3. Bottle spacing could be adjusted by sliding bottles along dove-tailed grooves in both the x and y directions.

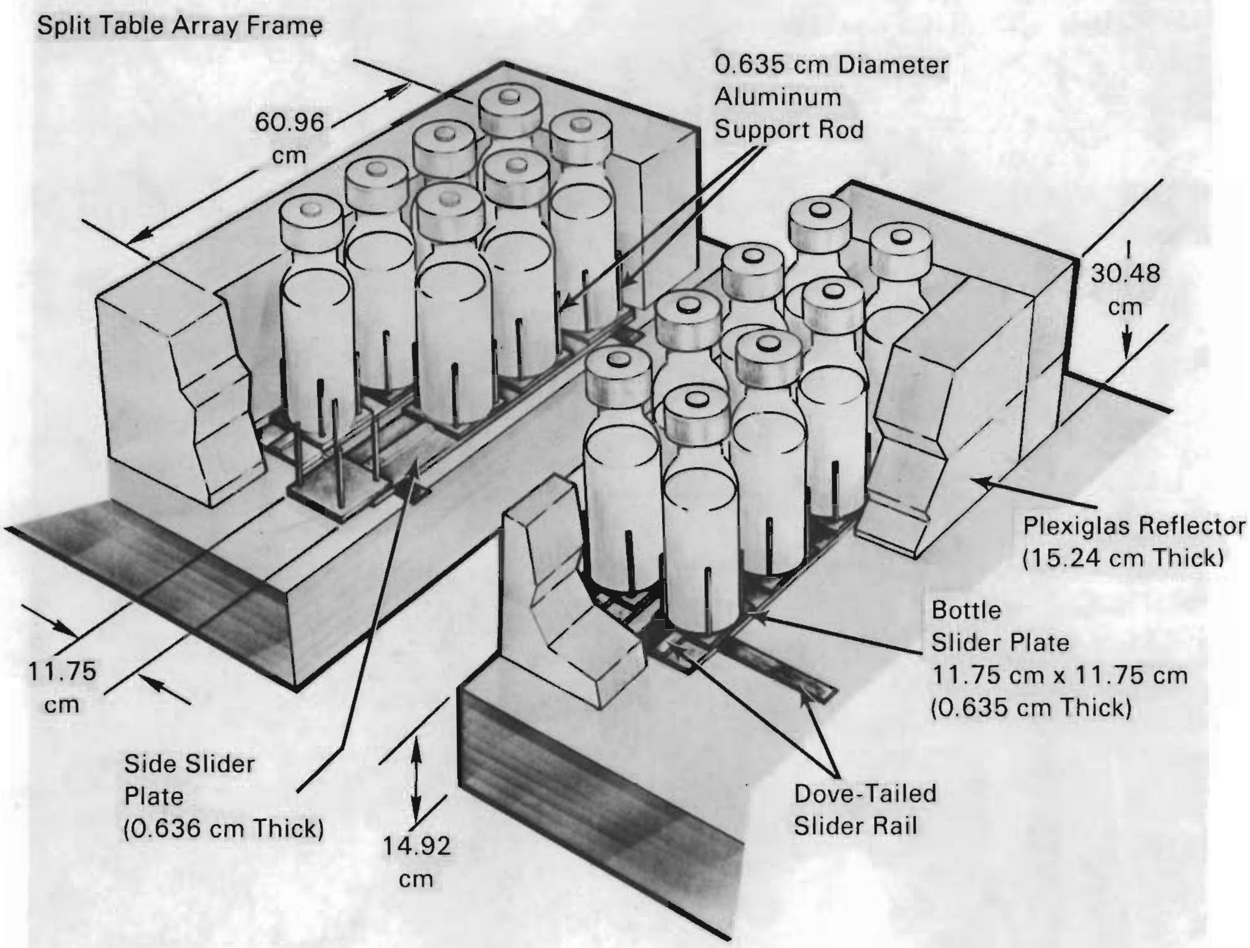


FIGURE 2. Isometric View of Interacting L-3 Bottles

Figure 3. Photograph of Experiment Showing Split Table Arrangement

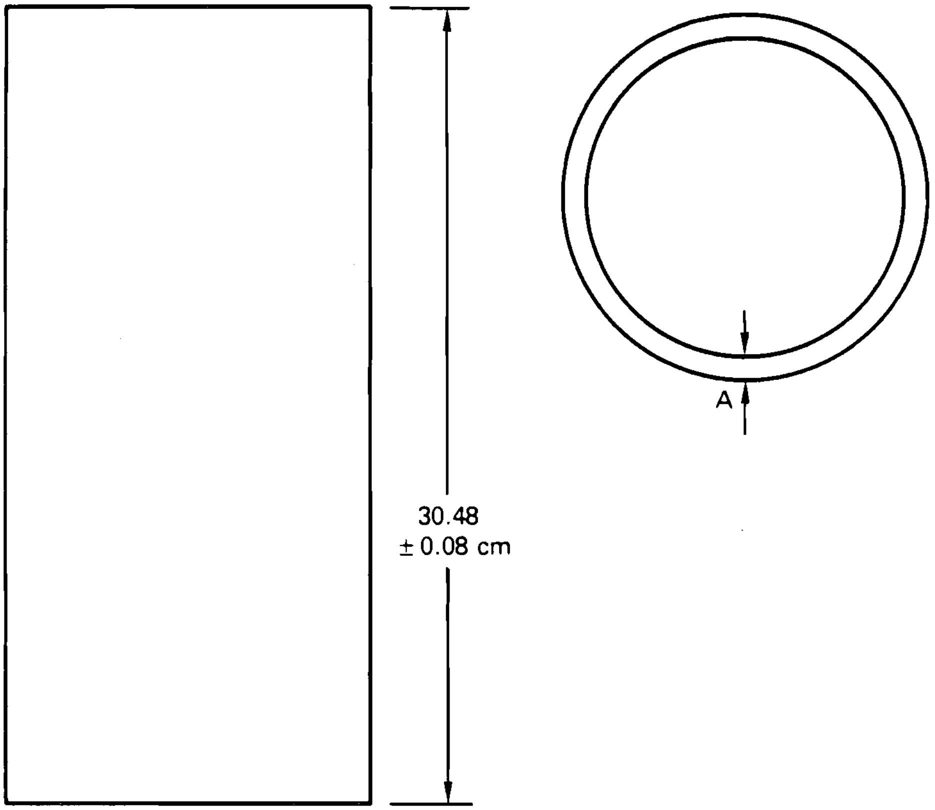
Four small aluminum pins securely held each bottle in place to prevent tipping and bottle movement. The pins were shown to have essentially no effect on the critical experiment data, as the same critical values were determined with and without the pins in place for several experiments.

Criticality was approached in steps by loading about half of the bottles on each side of the split table device with the faces separated. (See Figure 3). The fuel loading adjustment was made by hand and, after each subsequent change to the system the table was closed remotely, completing the array geometry desired. The critical approach method was used to measure the critical spacing or critical bottle number, depending upon the experiment. That is, small incremental changes were made to the arrays for a subcritical loading and the inverse count rates obtained were extrapolated to the critical condition.

After the initial series of experiments was performed with bottles separated by air gaps, plexiglas shells of varying thickness were positioned around each bottle to determine the effect of adding a moderating material in the space between bottles. The plexiglas serves to internally moderate the neutrons and effects both the neutron leakage from, and the neutron interaction between, the bottles making up the array. The shells are described in Figure 4.

EXPERIMENTAL RESULTS

The experimental results are summarized in Table 3 (Experiments 8-12 with plastic shells, Figure 4, on the bottles). Critical array configurations for given numbers of bottles are graphically illustrated in Figures 5 through 11. Critical numbers that appear in the table that are not integral (whole numbers of bottles) were obtained from extrapolation of the multiplication curves of bottles required for criticality for a given spacing. By this means the smallest critical number of bottles was found to occur with bottles in contact (no surface-to-surface separation), the extrapolated number being ~10.9. The manner in which this number might be used in a calculation is somewhat questionable, but the data establish a limit on the smallest critical number which is very useful in criticality control considerations. As the number of bottles was increased to twelve (3 x 4 array), with the bottles in contact in the x direction (see Figure 5), the critical spacing between rows



SHELL OUTSIDE DIAMETER (cm)	SHELL THICKNESS, A (cm)
15.25 ± 0.03	1.62 ± 0.03
13.94 ± 0.04	0.99 ± 0.01
12.72 ± 0.07	0.33 ± 0.01

COMPOSITION OF SHELL	WT%
H	8.0
C	60.0
O	32.0

DENSITY OF 1.185 g/cm^3

Figure 4. Description of Plexiglas Shells

TABLE 3. Results of Experiments (Plexiglas Reflected Arrays; see Figure 2 for Illustration)

Experiment Number	Described in Figure Number	Critical(a) Number of Bottles	X Spacing(b) (cm)	Y Spacing(c) (cm)	Shell Thickness (cm)
RSTM-L3-02	--	10.89±0.10	0.0	0.0	0.0
-03	5	12	0.0	0.49±0.03	0.0
-04	6	12	0.33±0.01	0.36±0.03	0.0
-05	7	12	0.59±0.01	0.0	0.0
-06	8	16	0.0	1.92±0.04	0.0
-07	9	16	1.26±0.01	0.66±0.05	0.0
8(d)	--	>25	3.47±0.05	3.47±0.05	1.62±0.03
9(d)	--	16.89±0.16	2.16±0.06	2.16±0.06	0.99±0.01
10(d)	--	13.1 ±0.3(e)	0.95±0.09	0.95±0.09	0.33±0.01
11(d)	--	13.1 ±0.3(f)	0.95±0.09	0.95±0.09	0.33±0.01
12	10	16	0.95±0.09	2.19±0.10	0.33±0.01
13	11	16	0.68±0.02	1.20±0.02	0.0

(a) The aluminum positioning pins or grooves cut in plastic shells had no appreciable effect on the extrapolated values shown (see footnote f).
(b) Measured from bottle surface to bottle surface.
(c) Measured from bottle surface to bottle surface.
(d) In these experiments, the outer plexiglas shells were touching in the X and Y directions. The X and Y spacings shown are measured from bottle surface to bottle surface. Any difference in spacing is due to void around each bottle.
(e) Average of two experiments, one where the 13th bottle is placed in the center of the array and one where the 13th bottle is on the array edge.
(f) Repeat of Experiment 10 with twice as many grooves in shells to establish the worth of the reflector displaced by support pegs.

became 0.49 cm. Figures 5 through 7 show the different critical configurations that were obtained with a 3 x 4 array of bottles as the surface-to-surface separation was varied between rows and columns in the x and y directions. By utilizing these data (see Figure 12) the surface-to-surface separation for a 3 x 4 array with equal separation in the x and y directions is estimated to be 0.35 cm.

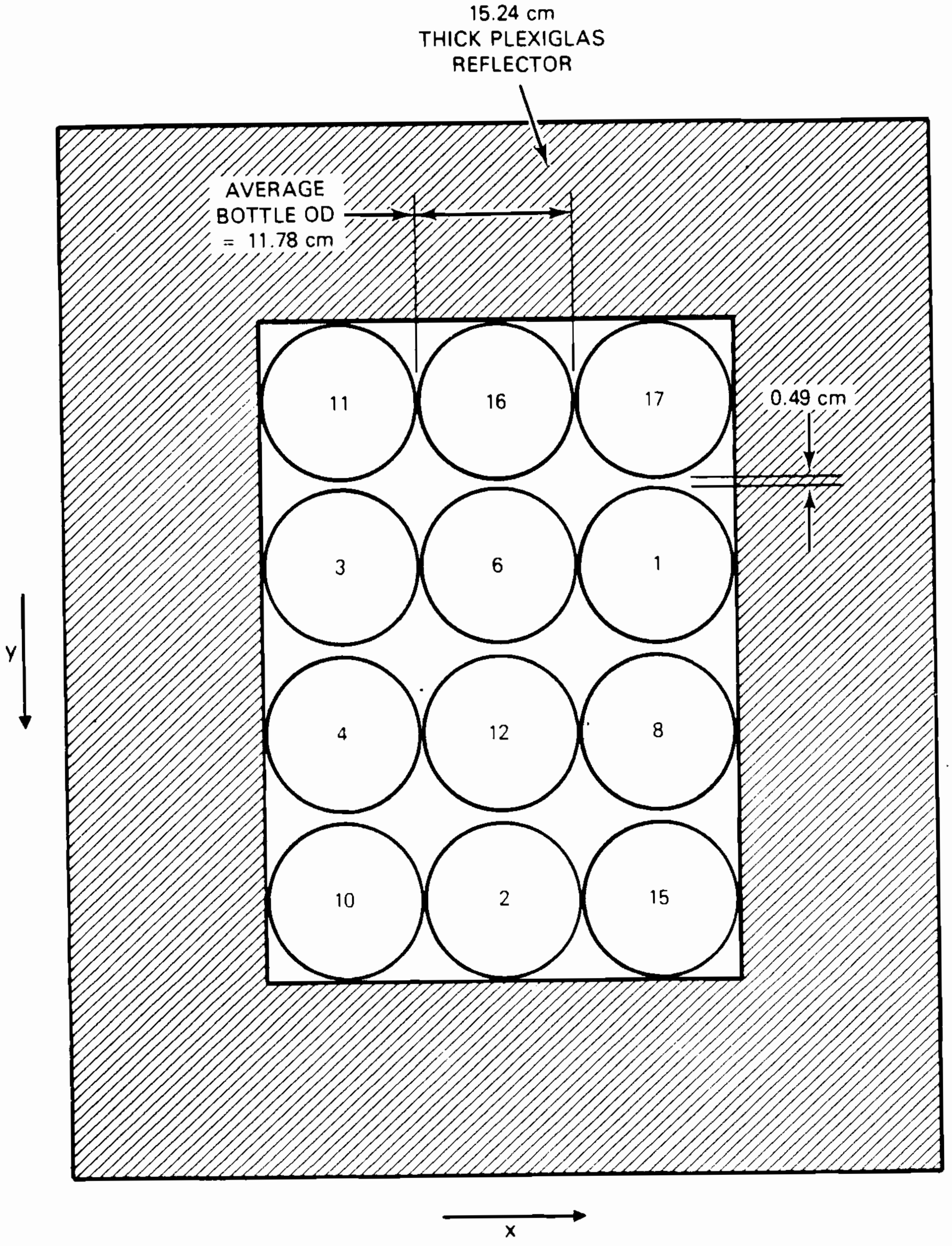


Figure 5. Experiment RSTM-L3-03

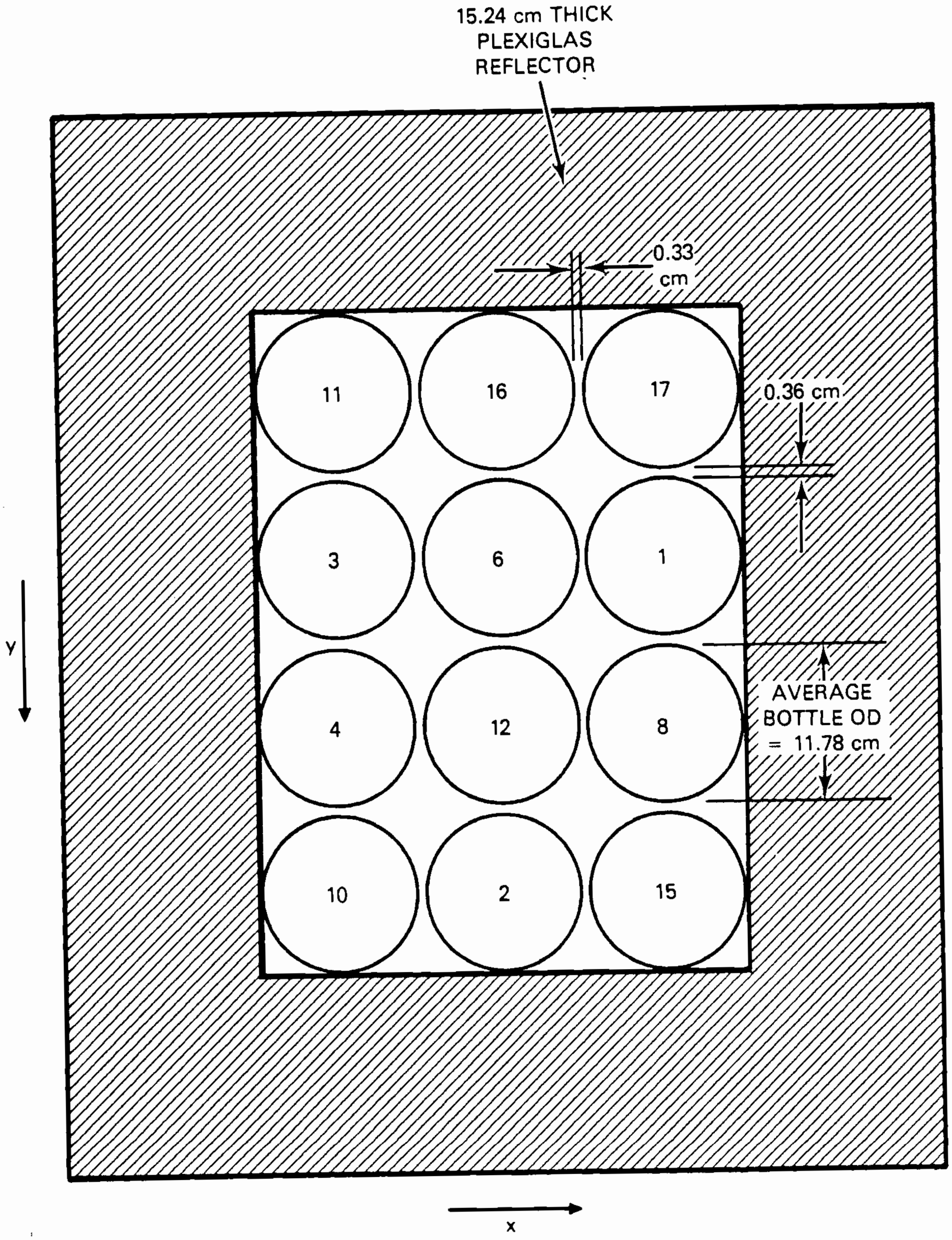


Figure 6. Experiment RSTM-L3-04

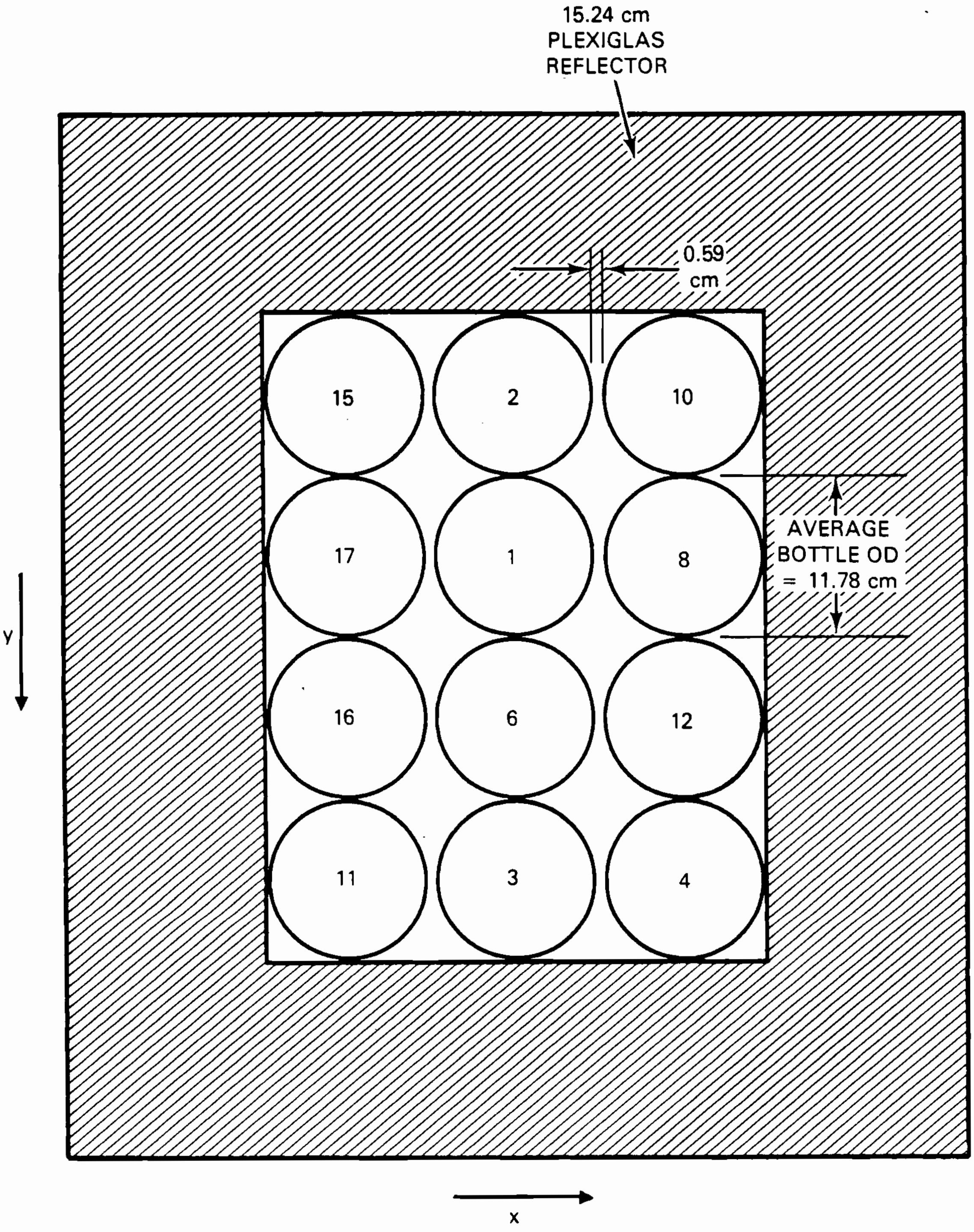


Figure 7. Experiment RSTM-L3-05

Different critical configurations obtained with 16 bottles (4 x 4 array) are shown in Figures 8 through 11. With the bottles in contact in the x direction, the critical surface-to-surface separation between rows of bottles in the y direction is 1.92 cm (see Figure 8). The data from the interacting arrays of 12 bottles and 16 bottles are plotted in Figure 12. These curves may be used to predict the critical separations for data points not included in the experiments. The curve for the 3 x 4 array implies that three separated rows of bottles, four abreast, constitute a more reactive configuration than four separated rows, containing three bottles each. In the first case the spatial density is lower than in the second at the point of criticality.

The inclusion of plastic around each bottle reduces the number of L-3 bottles required for criticality (see Figures 4, 10, 13). Although the plexiglas shell increases the bottle to bottle surface spacing and also the distance of each bottle from the side reflectors, the critical number of bottles is reduced. For a surface-to-surface spacing near one cm, this reduction amounts to almost 3 bottles or ~19% in critical mass. The minimum number of bottles required for criticality still occurs, however, for bottles arranged in a tightly packed array (see Figure 13).

The decrease in critical number of bottles, on inclusion of plastic shells around each bottle may be equated to an increase in spacing required to produce the same effect in an air spaced array without the extra plastic. For example, a 4 x 4 array of bottles with equal surface-to-surface separation in the x, y directions (0.96 cm in air) would require an increase in the y direction to about 2.2 cm if plastic shells of 0.33 cm thickness were positioned over each of the 16 bottles in the array.

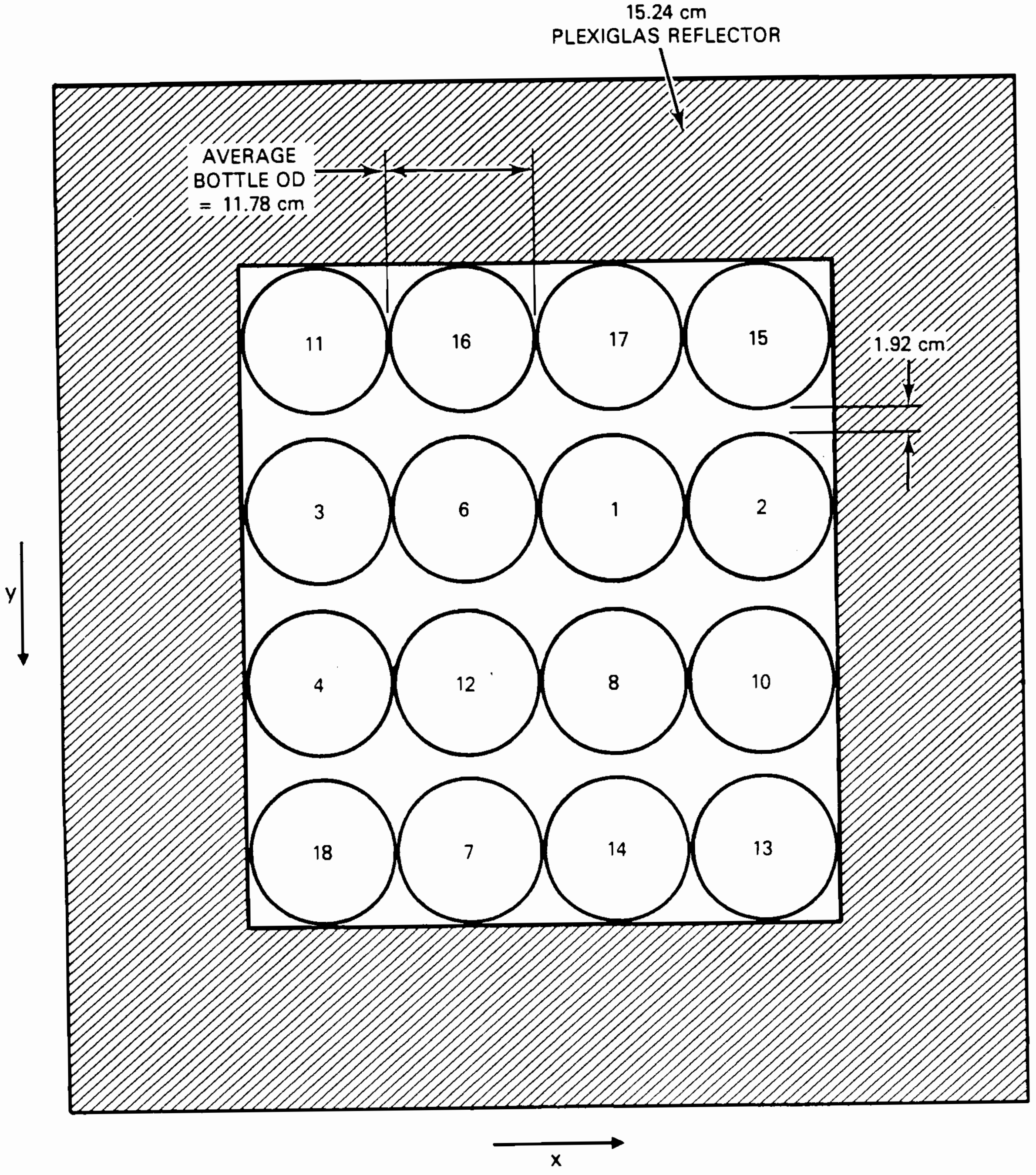


Figure 8. Experiment RSTM-L3-06

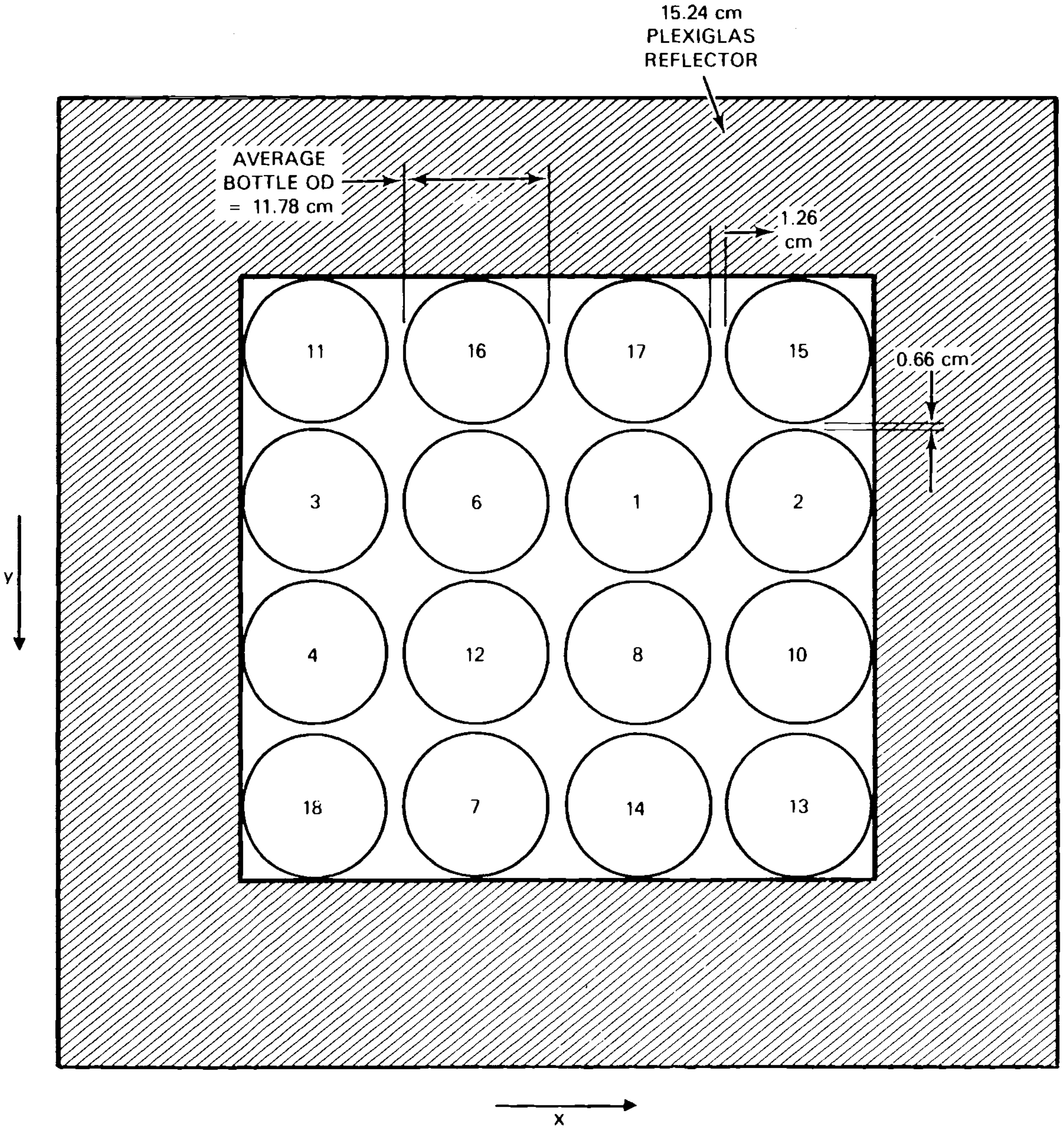


Figure 9. Experiment RSTM-L3-07

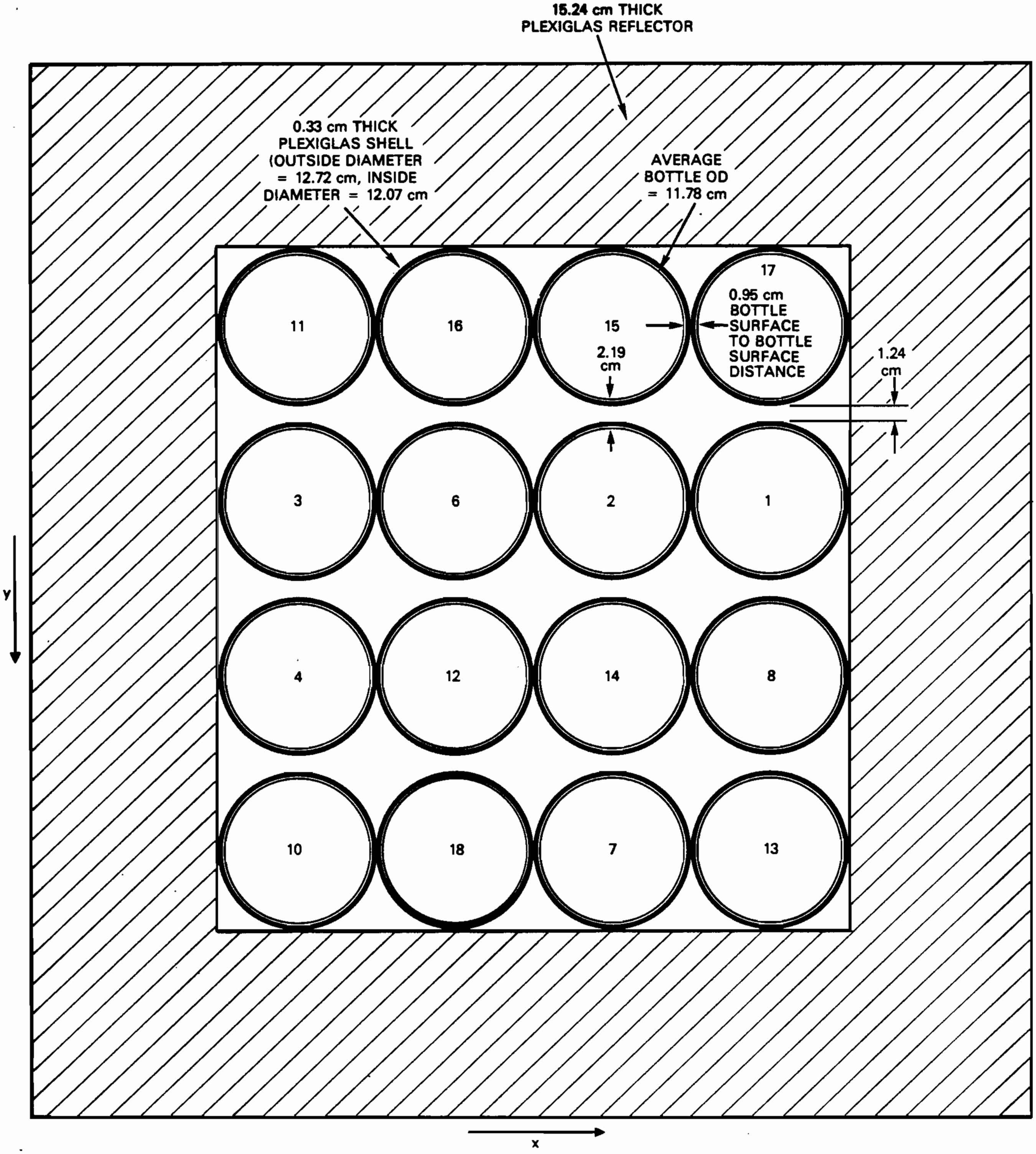


Figure 10. Experiment RSTM-L3-12

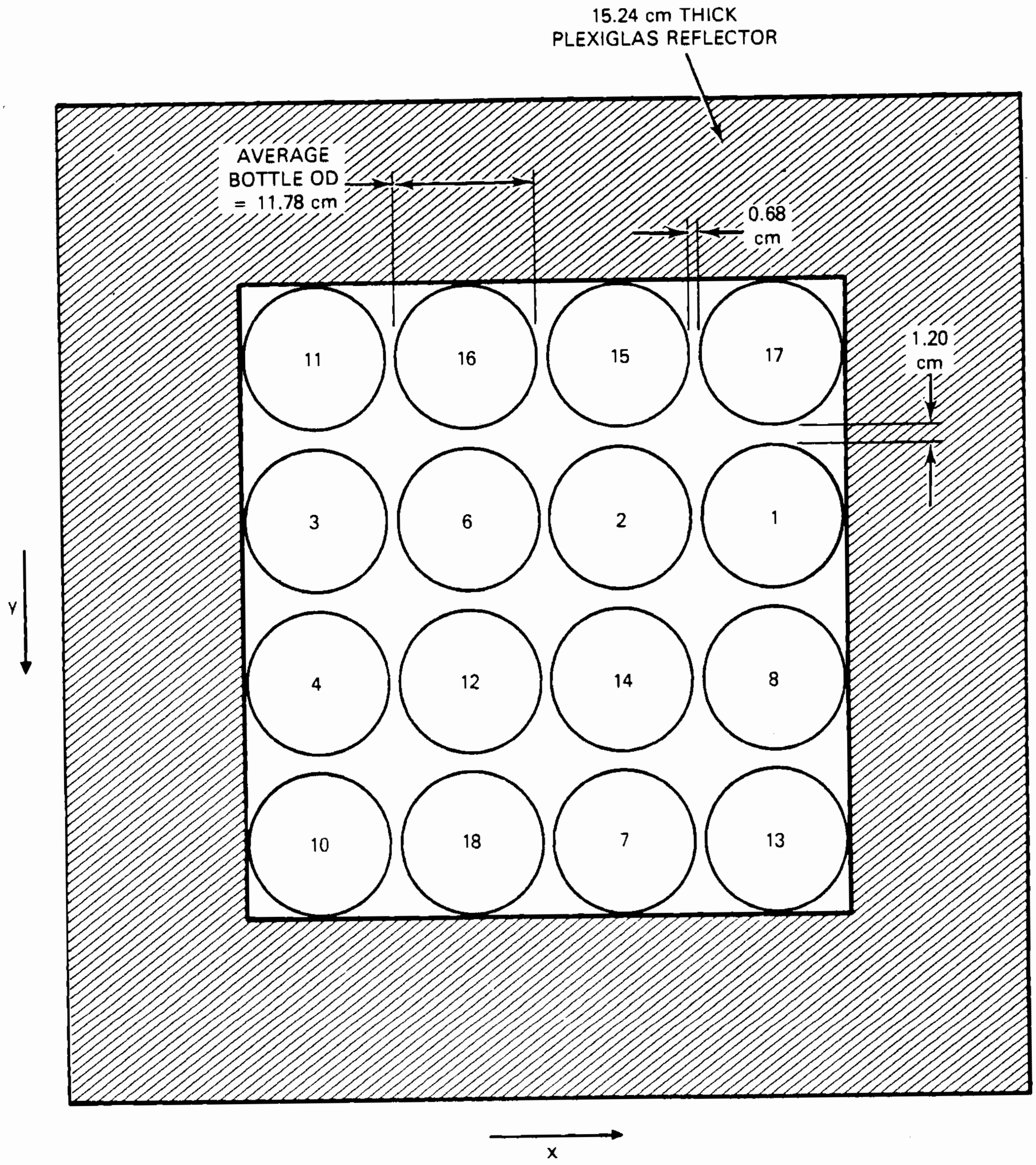


Figure 11. Experiment RSTM-L3-13

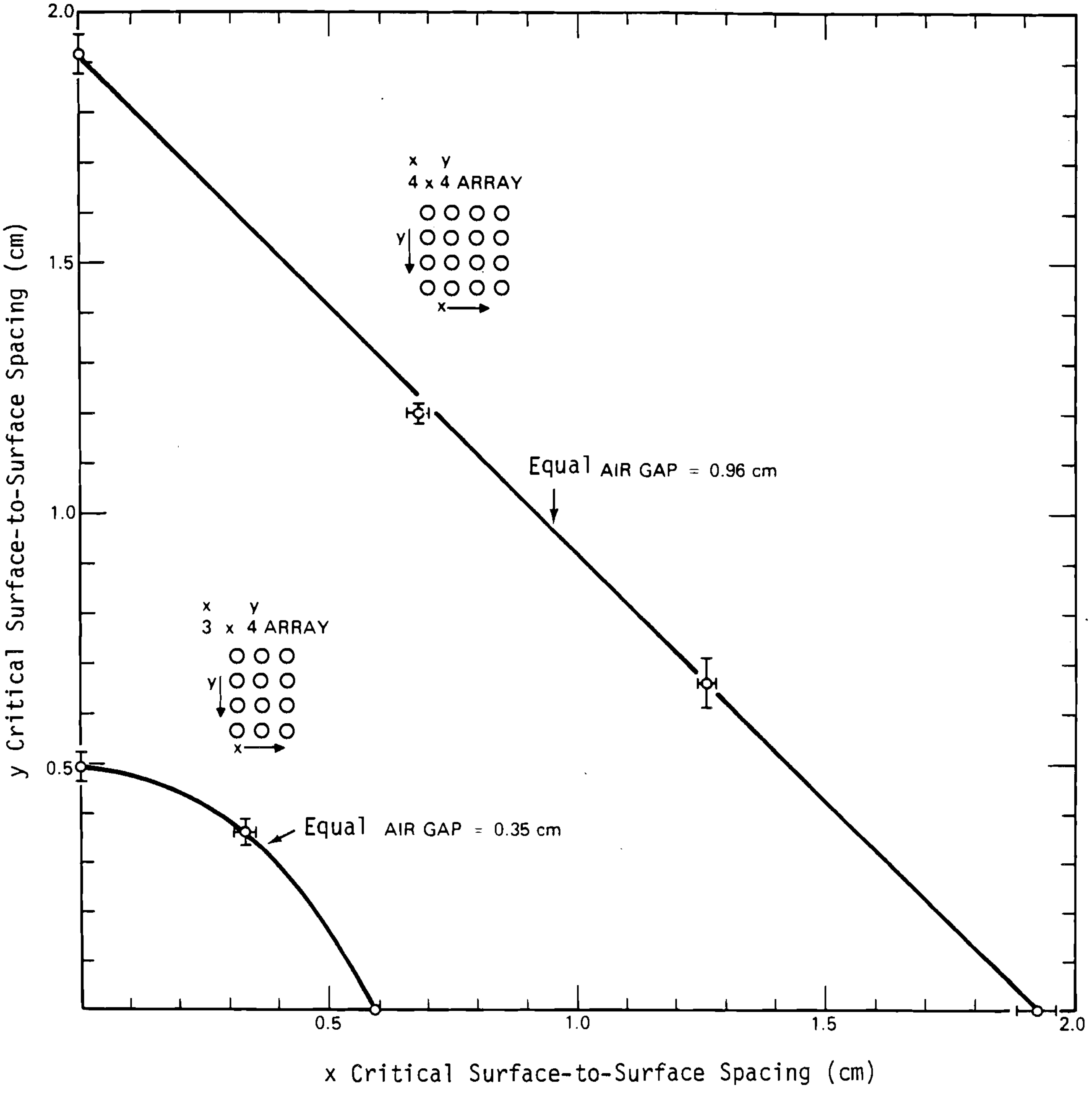


Figure 12. Critical Surface-to-Surface Spacing Between L-3 Bottles as a Function of Array Size

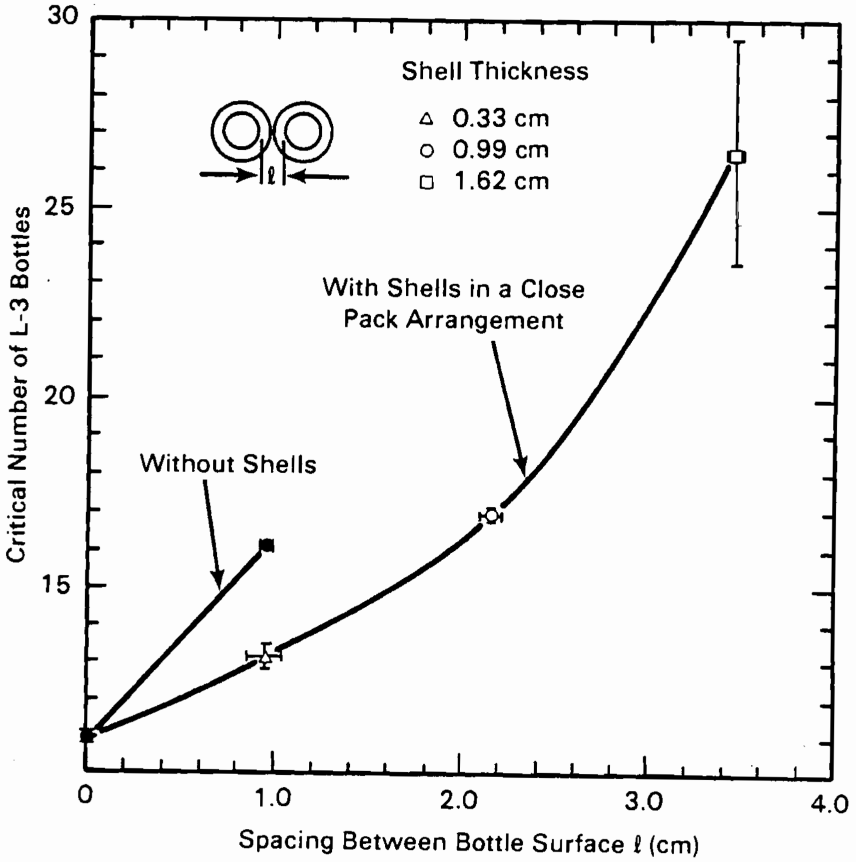


FIGURE 13. Critical Number vs Plastic Shell Thickness and Spacing

CONCLUSIONS

Data on the criticality of arrays of plutonium solutions have previously been unavailable. The critical experiments reported here on planar arrays of containers filled with plutonium nitrate solution provide the only known data for validation of calculational models on interacting arrays of this type. The experiments provide data on 1) critical spacings for fixed numbers of bottles, 12 and 16 in planar arrays, and also 2) estimated critical numbers of bottles as a function of spacing.

The minimum critical number of L-3 bottles (~10.9) was found to occur with the bottles in contact. Adding plastic shells to each of the bottles always resulted in a larger critical number than 10.9. Separating the bottles significantly increased the number required for criticality, but if plastic shells were then positioned over each of these bottles the critical number was then substantially reduced. For example, with a bottle surface-to-surface separation of 0.96 cm in air, the critical number is about 16. Positioning 0.33 cm thick plastic shells over each bottle at this same surface-to-surface separation reduces the critical number to ~13.1. Thus, the effect on criticality of the array caused by intervening hydrogenous material, such as might occur from plastic bags or other containers, flooding from sprinkler systems, etc., must be considered in safety evaluations.

Utilizing the data on critical surface-to-surface separations for fixed numbers of bottles, a 3 x 4 array in one case and a 4 x 4 array, it is possible to evaluate the condition for criticality for equal surface-to-surface spacing. In the case of the 16 bottles the spacing is ~0.96 cm, and for the 12 bottle array ~0.35 cm.

ACKNOWLEDGMENTS

The authors wish to acknowledge the contribution of Sr. Technician Joseph Lauby, who was instrumental in designing the load in system for filling the bottles with the plutonium solution used in these experiments. This resulted in a filling accuracy on volume of less than 1/2% per bottle.

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