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LIVERMORE PLUTONIUM ARRAY PROGRAM: EXPERIMENTS AND CALCULATIONS

FUEL CYCLES

KEYWORDS: *plutonium, criticality, safety, computer calculations, cylinders, Monte Carlo method*

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The first precision array measurements with high-density plutonium-metal cylindrical parts of 3 and 6 kg took place at the Livermore Critical Assembly Facility from 1965 to 1969. Cubic arrays of up to sixty-four 6-kg parts were measured. Mock high-explosive epoxy moderators were used in several measurements. Experiments observing the effects of simulated body reflectors provided personnel safety guidance for the construction of these arrays.

A comparison of Monte Carlo calculations and the experimental measurements indicated that the calculational method is sufficiently accurate to be used in nuclear safety guidance for arrays of these elements. Included for comparison are calculations for arrays comprised solely of the plutonium parts. Also included are calculations for 6-kg-part arrays in which a 0.479-cm-wide gap at the midplane has been eliminated and where the spacing was varied for each idealized array.

INTRODUCTION

During 1965 to 1969 a program was carried out at the Lawrence Livermore Laboratory (LLL) to study neutron-interacting plutonium arrays. The program included both experimental determinations of critical configurations and a study of the applicability of calculational techniques to such experimental results.

The primary purpose of the Livermore Array Program (LAP) was to develop a reliable calculational technique for answering questions about criticality safety, which arise in the design, transportation, and storage of arrays of fissionable material. The secondary purpose of the program was to generate experimental data that could be used directly for criticality safety analysis. These program objectives complement each other. The experimental data provided the only practical method of assessing the reliability of the calculational techniques. On the other hand, calculations were important for design of the experiments and for estimating the effect and relationship of experimental parameters.

We intended our studies to be specific to arrays of plutonium parts. Accordingly we studied arrays of plutonium metal parts rather than of some other more convenient fissionable material so the critical data obtained could be applied directly to some cases of interest. Also, anticipating that the critical spacings of moderated arrays would be strongly affected by the detailed character of the plutonium cross sections, we wished to study arrays under varying conditions of moderation.

In the program we determined the critical spacings of a number of regularly spaced approximately cubical arrays. The basic array part was a solid right-circular plutonium cylinder of density 19.53 g/cm^3 (α-phase), weighing 3 kg. The following parameters were varied:

1. size of array ($2 \times 2 \times 2$, $3 \times 3 \times 3$, etc.)
2. amount of moderator material interspersed among units
3. mass of array element (3 or 6 kg)
4. reflector material and thickness.

AVAILABLE EXPERIMENTAL DATA

Schuske et al. have reported measurements of planar and three-dimensional (3-D) arrays of 2-kg plutonium units in which array multiplication was

determined as a function of the number of array elements.[1] Most of the arrays were subcritical with low multiplication, but two column arrays ($4 \times 4 \times N$ and $5 \times 5 \times N$) were assembled to high enough multiplications to permit extrapolation to the critical configuration.

Other experiments with metal array units have been reported. Mihalczo and Lynn determined the critical number of units in arrays of 20.32- × 25.40- × 2.54-cm slabs of U(93.4) metal.[2] All arrays were reflected and results were given for both moderated and unmoderated conditions. The critical spacings for two U(93.2) metal units[3,4] were reported by Mihalczo. The units were either slabs, 20.32 × 25.40 × 2.54 cm, or disks ranging in diameter from 17.78 to 38.10 cm. Also reported is the Rossi-α for each critical configuration.

The most complete work to date involving metal units is the excellent series of measurements by Thomas, first reported in 1964 (Ref. 5). Thomas determined the critical spacings between elements in 3-D arrays of up to 64 units. The basic elements were U(93.2) metal cylinders ranging in mass from 10 to 26 kg, with height-to-diameter ratios from 0.47 to 1.90. Other variables were (a) amount of reflection, (b) array geometry, (c) geometry of array element, and (d) amount of moderation.

OVERVIEW OF THE LIVERMORE ARRAY PROGRAM

Experiments

Events divided the experiments into three phases. Phase I consisted of experiments performed on a small assembly machine originally constructed for another program. When experimental sizes became too large for this machine, a much larger one was constructed specifically for the array program and the immediately succeeding experiments were designated Phase II. When a safety rule was found to be invalid, temporarily suspending the regular experiments, a special set of linear-array experiments was performed. These experiments, called Phase III, were done with a single loading unit cut into two halves and turned horizontally on the Phase II machine.

Although many critical configurations were determined, only the basic arrays and the most interesting variations will be reported here. The critical point was generally determined by subcritical extrapolations. Most arrays attained a maximum multiplication >100 and four arrays achieved the critical state, that is, a small, positive power-level period. Arrays with lower multiplications naturally had greater uncertainties in their critical spacings.

Calculations

The Monte Carlo calculation technique was adapted as the working tool for this program. This type of calculation is the only one capable of handling the geometries of the systems of interest. It also lends itself well to describing and systematically studying detailed changes within a system.

We examined three Monte Carlo codes and selected the 05R code developed at the Oak Ridge National Laboratory (ORNL) (Ref. 6). The other two candidates, GEM (Ref. 7) and SORS (Ref. 8), were found to be less desirable for reasons such as limitations on geometry options and cross-section treatment. Calculational results using the 05R code were quite satisfactory and some of the results are presented below.

Along with the problem of determining the merits of various Monte Carlo techniques already in use (or not yet devised), there is the problem of evaluating existing (or providing new) cross sections suitable for safety calculations. The cross-section data used in LAP have been taken from the Howerton Evaluated Library (HEL) (Ref. 9). At the time we undertook this program, the Howerton Library appeared to provide the best starting point among existing sets of cross-section data.

SUMMARY OF EXPERIMENTAL DETAILS

Plutonium Cylinders

The geometry of the basic plutonium part is a solid right-circular cylinder. A total of 130 of these parts were fabricated at the Hanford facility. Each part was weighed before it was canned, but its dimensions were not recorded. The parts varied between 3.004 and 3.043 kg, with an average of 3.026 ± 0.008 kg.

The data of Table I are inferred from our measurements with canned parts and empty cans. These data are recommended as working values. Table II lists the average isotopic distribution of 32 plutonium parts as determined by mass spectrometer measurements. Calculated nuclear number densities are also given. Impurities were determined spectrographically for all 130 parts. Results given in Table III represent average values of these data.

Moderator Cells

The moderator cells, provided in three thicknesses, were designed to enclose either the 3-kg

TABLE I

Plutonium Part Specifications

Mass (kg)	3.026 ± 0.008
Diameter (cm)	6.525 ± 0.005
Height (cm)	4.633 ± 0.015
Density (g/cm^3)	19.53 ± 0.08

TABLE II

Plutonium Isotopic Composition

Isotope	Composition (wt%)	Calculated Number Densities (10^{22}/cm^3)
239	93.56	4.608
240	5.97	0.2925
241	0.46[a]	0.0225[a]
242	0.01	0.00068

[a]As of July 1965, this isotope was decaying with a 13.2-yr half-life.

TABLE III

Plutonium Impurities

Element	Amount (ppm)	Element	Amount (ppm)
Ag	<1	Mn	5
Al	5	Na	1
B	1	Ni	50
Ca	100	Pb	<1
Cr	20	Si	15
Cu	5	Sn	<2
Fe	35	Ti	5
Mg	20	(Total metallic, 265)	
C	180		

basic plutonium part or the 6-kg double part. They are comprised of disks inside the support column and rings on the outside. There are two inner disks per cell (immediately adjacent to the top and bottom heat sinks), and from one to three outer rings. Table IV lists the physical parameters of the six different-sized moderator cells. The total height of the outer rings does not exactly coincide with the height of the stacked inner parts of the cell. However, the bottom of the lowest ring is always positioned to be even with the bottom of the lowest disk.

Simplifying the description of each cell size for working purposes, we can consider the moderator density as 1.559 ± 0.018 g/cm^3 for all parts, the outside diameter of the disks as 6.795 cm, the inside diameter of the rings as 7.628 cm, and the three moderator thicknesses as 1.271, 2.549, and 3.816 cm, respectively. Finally, the height of the outer rings should be set equal to the distance between the outer faces of the inner disks.

The parts were made of a powdered mock high-explosive formulation that was cast in an epoxy binder. A chemical analysis of some representative parts is given in Table V. Moderator impurities are listed in Table VI.

Containers, Heat Sinks, and Spacers

Each plutonium part is sealed in a close-fitting deep-drawn seamless aluminum can fitted with a steel lid. The sides of the cans are 0.037 cm thick, and the bottom, which appears at the top of the part in the experimental configuration because the cans are loaded in an inverted position (Fig. 1), is 0.087 cm thick. The mass of the aluminum can is 19.2 g; its composition appears in Table VII. The steel lid is 0.021 cm thick and has a mass of 8.28 g; its composition is itemized in Table VIII.

TABLE IV

Physical Parameters of Moderator Parts

Average Cell Thickness (cm)	Inner Disk Size (cm)[a]	Average Inner Disk Mass (g)	3-kg Cell Parameters		6-kg Cell Parameters	
			Outer Ring Size (cm)[b]	Average Total Cell Mass (g)[c]	Outer Ring Size (cm)[b]	Average Total Cell Mass (g)[c]
1.27	6.795 × 1.278	71.51 ± 0.75	8.407 × 7.620 × 1.267	603.8 ± 6.5	14.267 × 7.625 × 1.267	930.8 ± 10.6
2.54	6.795 × 2.555	144.7 ± 1.7	10.932 × 7.635 × 2.543	1662.7 ± 9.5	16.746 × 7.633 × 2.548	2401.2 ± 8.8
3.81	6.795 × 3.813	214.6 ± 2.7	13.480 × 7.628 × 3.818	3354 ± 30	19.334 × 7.628 × 3.818	4614 ± 26

[a]o.d. × thickness.
[b]Height × i.d. × thickness.
[c]Includes two inner disks.

TABLE V

Moderator Composition

Elements	Composition (wt%)	Calculated Number Densities (10^{22}/cm^3)
C	30.56	2.398
H	3.14	2.89
N	31.57	2.123
O plus impurities	34.73	2.042

TABLE VI

Moderator Impurities

Element	Amount (ppm)	Element	Amount (ppm)
Al	3	Mo	0.2
B	1	Na	20
Ba	5	Ni	2
Ca	10	Pb	0.4
Cr	2	Si	2
Cu	0.3	Sn	0.2
Fe	25	Sr	0.2
K	200	Ti	0.5
Mg	3	Zn	0.5
Mn	0.5		

Aluminum heat sinks were used above and below all plutonium parts. When two parts were stacked vertically to form a 6-kg double part, a 16.10-g, 0.356-cm-thick heat sink was placed between the lid of one part and the bottom of the other. A 26.97-g, 0.479-cm-thick heat sink

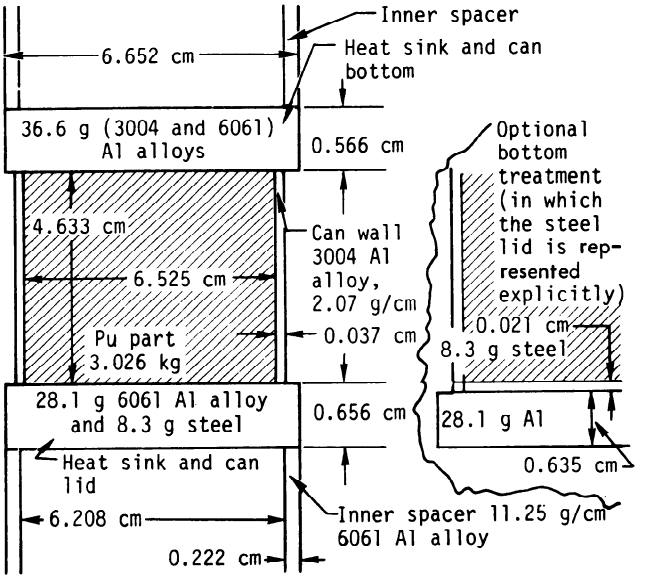

Fig. 1. Schematic drawing of array unit containing single plutonium part (support tube not shown).

TABLE VII

Aluminum Alloy Compositions

Element	Nominal Composition (%)			
	2024 Alloy	2219 Alloy	3004 Alloy	6061 Alloy
Cr	0.10	---	---	0.20
Cu	4.35	6.3	0.25	0.25
Fe	0.50	0.30	0.7	0.7
Mg	1.5	0.02	1.05	1.0
Mn	0.6	0.3	1.25	0.15
Si	0.50	0.20	0.30	0.6
Ti	---	0.06	---	0.15
Zn	0.25	0.10	0.25	0.25
All others	0.15 max	0.15 max	0.15 max	0.15 max
Al (remainder)	~92.05	~92.6	~96.05	~96.55
	Uses			
	Column, Phase I shoes,[a] all unspecified structural materials	Phase II table top	Container[b]	Heat sinks, inner spacers

[a]Shoes are used to fasten the column to the table.
[b]9.57 g to be homogenized with upper heat sinks.

TABLE VIII

Steel Alloy Compositions

Element	Nominal Composition (%)			
	Plutonium Can Lid	Bolts and Washers in Bottom Shoe[a]	Stainless-Steel Clamps in Phase II Upper Structure[b]	Angle Iron Under Tables[c]
C	0.08	0.38	0.15 max	0.21
Cr	---	4.67	18.00	---
Mn	0.37	0.35	---	0.43
Mo	---	1.21	---	---
Ni	---	---	5.00	---
P	0.015	0.02	---	0.04 max
Pb	---	0.02	---	---
S	0.025	0.04	---	0.05 max
Si	0.01 max	0.85	1.00	---
Sn	0.30[d]	---	---	---
V	---	0.47	---	---
Fe (remainder)	~99.2	~92.0	~75.9	~99.3

[a]Shoe fastens bottom end of Phase II column to table.
[b]Clamp fastens top end of Phase II column to table upper structure.
[c]Full density is ~7.85 g/cm^3.
[d]Tin plate.

appeared at the top of each part or double part and a 28.08-g, 0.635-cm-thick sink at the bottom.

Internal spacing between plutonium units was accomplished by suitable combinations of thin aluminum cylinders, 6.208 cm i.d. and 0.222 cm thick, fitted inside the support tube. The mass per unit length of spacers averaged 11.25 g/cm. Both the internal spacers and heat sinks described above are made of 6061 aluminum alloy (see Table VII for its composition).

Minor features, such as curves in the plutonium cans and heat sinks, exist in the actual apparatus but have not been described above. Furthermore, many air spaces exist that are not detailed. In Figs. 1 and 2, the single and double parts, as well as related components, are shown schematically. Some sections have been homogenized, but there is sufficient detail to prepare calculational input. Of course, further homogenization is always possible.

Assembly Machines and Support Structure

A photograph of the Phase I assembly machine is shown in Fig. 3. Figure 4 schematically depicts this machine and its support structure. The part of the column surrounding the plutonium is 6.850 cm i.d. and 0.184 cm thick; it has a mass per unit length of 11.01 g/cm. The material is 2024 aluminum alloy; its composition is found in Table VII.

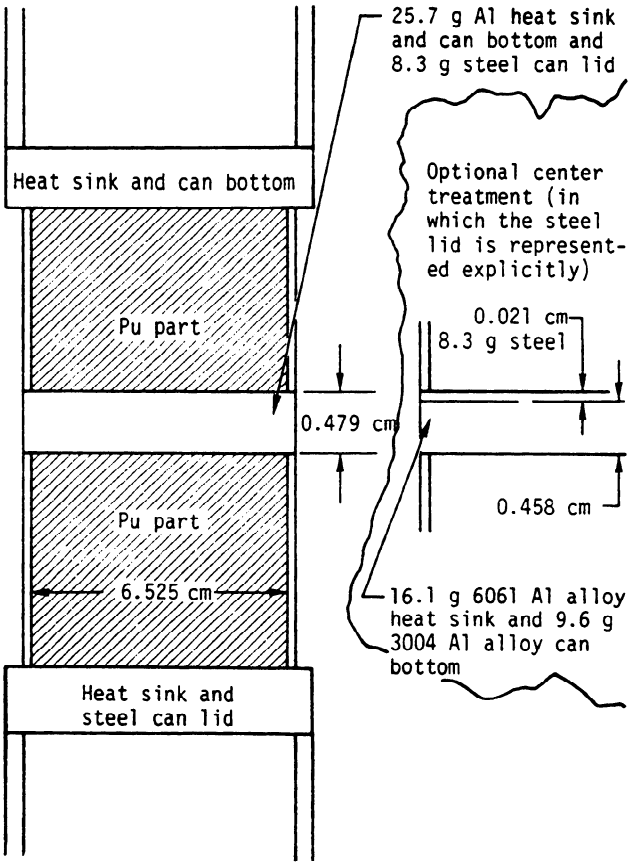

Fig. 2. Schematic drawing of array unit containing two plutonium parts (support tube not shown).

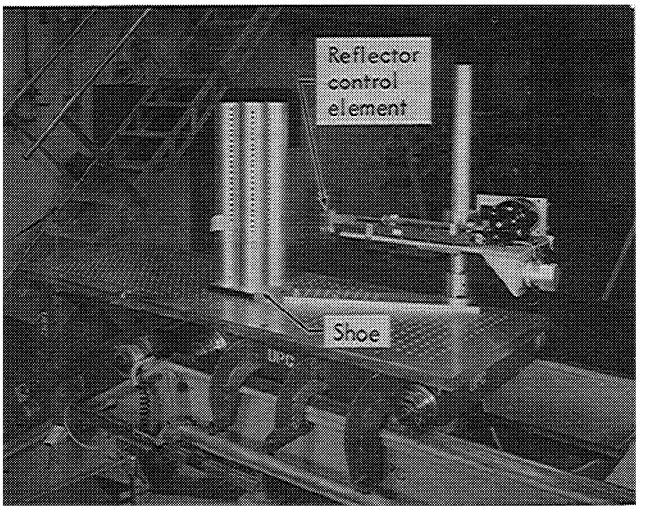

Fig. 3. Phase I assembly machine with mockup of 2 X 2 X 2 array.

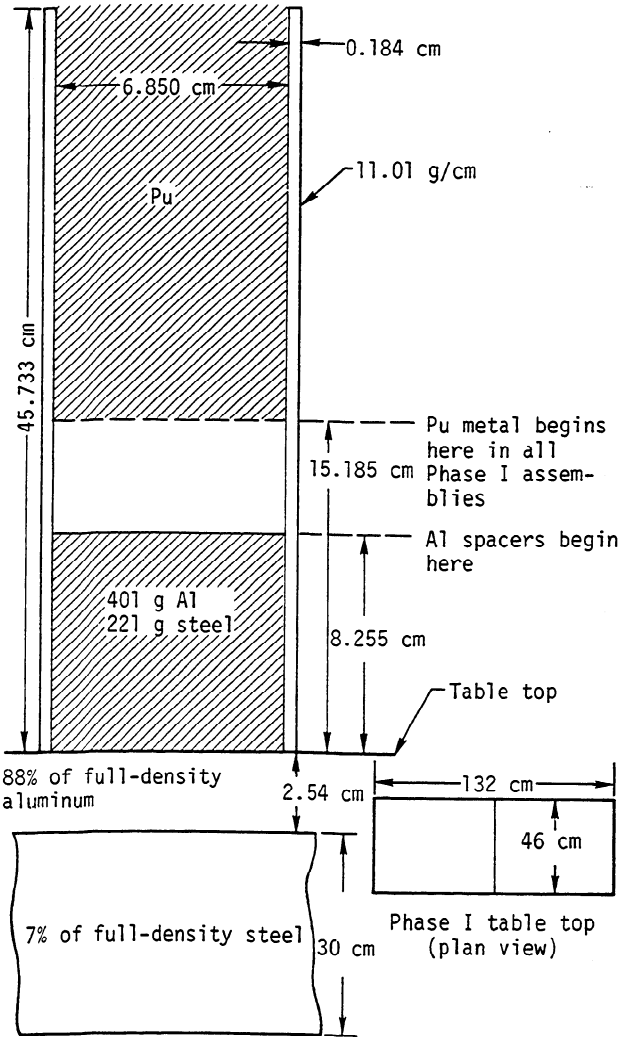

Fig. 4. Schematic drawing of Phase I support apparatus.

The shoes that fasten the columns to the assembly table are also made of 2024 alloy.

A photograph of the Phase II assembly machine appears in Fig. 5. A schematic drawing of the machine top and support apparatus is given in Fig. 6. Phase II columns are taller than the Phase I columns, have a 6.858-cm-i.d., 0.178-cm-thick wall, and a mass per unit length of 10.70 g/cm. These columns are also 2024 aluminum alloy, but the bottom shoes are made of naval brass, whose composition includes 60.00 wt% tin. Apart from its size, the support structure for the Phase II arrays is also distinguished by an upper support arrangement involving plugs, clamps, beams, and rails.

Because of the hundreds of bolt holes used for fastening the shoes, the density of both tables is reduced from solid aluminum density by ~12%.

The Phase III experiments were conducted using a modified Phase II outer tube suspended horizontally with its centerline 17.8 cm above the Phase II assembly table. Interior spacers and moderator cells were the same as those used in Phases I and II. A photograph of the Phase III apparatus appears in Fig. 7.

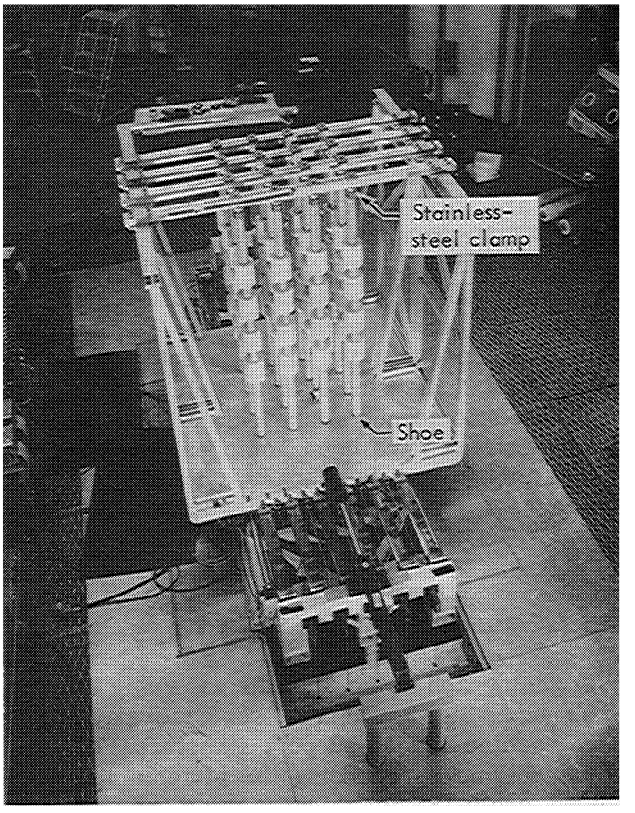

Fig. 5. Phase II assembly machine with mockup of 4 X 4 X 4 moderated array.

Reflector

The reflector used in the Phase I program was fabricated from polyethylene $(CH_2{:}CH_2)_n$ slabs and had a density of 0.92 ± 0.01 g/cm^3. It was 45.08 cm high × 35.03 cm wide × 20.64 cm thick. When in place, the bottom was flush with the assembly table top; the reflector was centered horizontally with the array and was touching the surfaces of the outer support columns.

The reflectors used in Phase III were intended to simulate the human body and were fabricated from polyethylene (density 0.92 ± 0.01 g/cm^3). The large torso simulators were each 152.4 × 35.6 × 15.2 cm, and the two arm simulators were 152.4 × 7.6 × 7.6 cm.

Experimental Procedures

Critical configurations were extrapolated from external neutron flux measurements and precise spacing determinations, using the standard inverse multiplication technique.[10] In several cases, we confirmed the extrapolated critical spacing by using a small external reflecting control element to bring the assembly to the critical state. In only five arrays was the maximum multiplication <50; in all other configurations the multiplication exceeded 100.

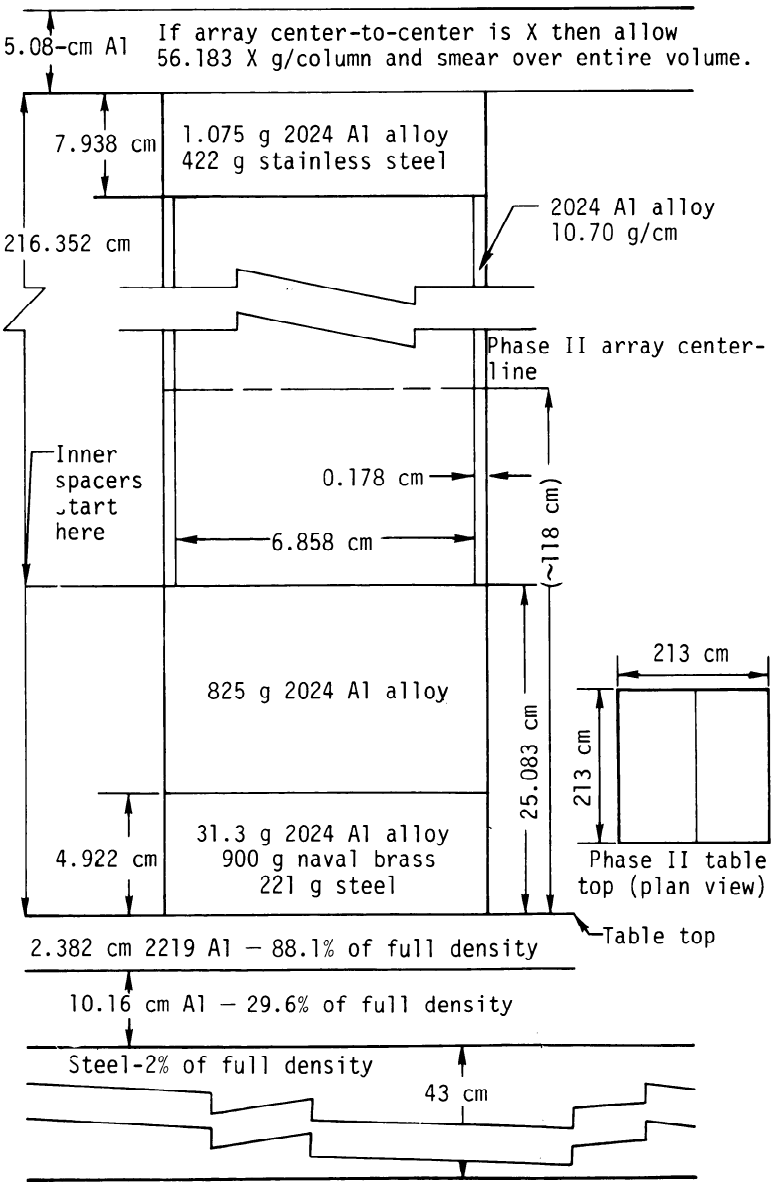

Fig. 6. Schematic drawing of Phase II support apparatus.

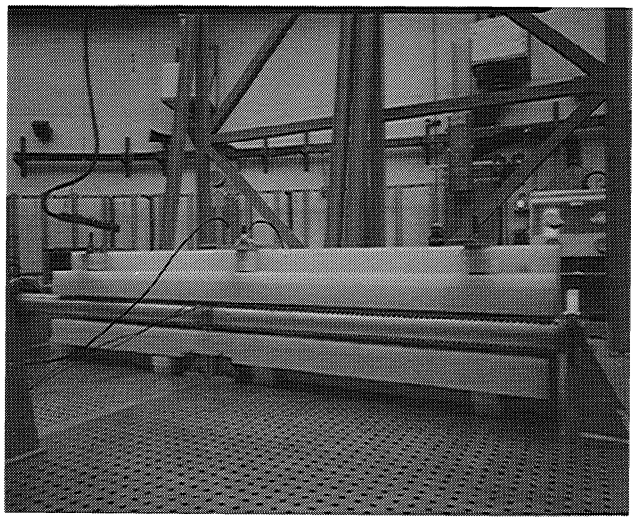

Fig. 7. Phase III apparatus on the Phase II assembly machine.

EXPERIMENTAL RESULTS

In the tabulation of experimental results that follows, the order of presentation is based on the degree of complexity, without regard to their order of performance. They have been numbered, however, in chronological order of completion. The Phase I arrays are the 100 series, the Phase II arrays the 200 series, and the Phase III arrays the 300 series.

Perturbation studies, which are described in detail in Ref. 10, are the arrays numbered 102, 104, 106, and 107. Array number 213 was the subject of a study in which the critical spacing was determined as a function of the height above the assembly table and is described in detail in Ref. 11. Reference 12 includes schematic diagrams of all the arrays.

Table IX lists the experimentally determined critical spacings of the 1-, 2-, and 3-D arrays. Unit cell volumes, the product of the three center-to-center dimensions in Table IX, are included in the tabulation. The temperatures that appear in Table IX were measured with a thermocouple in contact with one of the plutonium parts on the outer boundary of the array.

The arrays referred to in footnote e of Table IX deserve a brief comment. One might anticipate that any departure from an equilateral configuration would require the average fissile material density to be increased to maintain criticality. However, if the individual parts are sufficiently reactive (as they are here) there is a horizontal spacing at which a single layer of parts will be critical. In this case, additional layers may be introduced and criticality can be maintained by increasing the horizontal spacing even though the vertical spacing is relatively large. The unit cell volumes of such critical systems will therefore be relatively large and the average fissile material density will be relatively small. A small average fissile material density for criticality is also obtained where a single column of parts can achieve criticality. Thomas reports on this effect.[5]

The experimental uncertainties in Table IX arise from the uncertainties in the multiplication values and the spacings. The parts were assembled with an accuracy of ±0.003 cm, but ancillary measurements showed an average uncertainty in the location of the individual parts about four times that value, due to bowing of the support columns and spacing tolerances within them. The supercritical runs showed that the error due to uncertainties in multiplication was less than the uncertainty due to positioning.

Overall, we are confident that an array constructed with exactly the same arrangement and parts as ours, but with a spacing greater than the allowed range, will be subcritical. With a spacing smaller than the allowed range, we are sure it will be supercritical.

COMPUTATIONAL TECHNIQUES

The 05R neutron Monte Carlo code is capable of handling complicated geometries such as the plutonium arrays described here. At the same time the calculation provides for a very detailed energy and angular description of the neutron cross sections and the neutron physics processes that take place. The neutron reactions treated explicitly in the 05R calculation are elastic scattering, inelastic scattering, the $(n, 2n)$ reaction, and fission.

Neutrons with energies <1 eV are scattered isotropically in the laboratory system with no change in energy and with reaction probabilities specified by a one energy group set of parameters. For the calculational results given in this paper these one-group parameters are based on the Hansen-Roach cross sections.[13] This simple treatment is known to introduce serious errors into 05R results for some types of systems. An improved model based directly on a quantum mechanical formulation is needed to account for molecular binding effects and the thermal motion of the target nuclei.

The cross-section data used in all 05R calculations to date have been taken from HEL.[9] This library of neutron cross sections has been developed by Howerton and co-workers at LLL. It is described in a series of reports available upon request to LLL. Calculational results reported here refer to the library in 1965. In all the calculations described here, there were 200 neutrons

TABLE IX

Summary of Critical Array Spacings (Ref. 12)

Array Number	Description	Critical Center-to-Center Separation (cm)		Unit Cell Volume (cm^3)	Outer Can Temperature (K)
		Vertical	Horizontal		
		3.026-kg units with height-to-diameter ratio 0.710			
304	1 × 1 × 4 unmoderated, four sides reflected[a]	5.56 ± 0.03	---	---	311
301	1 × 1 × 5 unmoderated, four sides reflected[a]	6.18 ± 0.03	---	---	303
303	1 × 1 × 7 unmoderated, four sides reflected[a]	6.75 ± 0.03	---	---	313
302	1 × 1 × 11 unmoderated, four sides reflected[a]	7.25 ± 0.03	---	---	322
101[b]	2 × 2 × 2 unmoderated, bare	5.40 ± 0.03	7.30 ± 0.03	287.7	315
102[b]	2 × 2 × 2 unmoderated, one side reflected[c]	5.74 ± 0.03	7.64 ± 0.03	334.8	309
103[b]	3 × 3 × 3 unmoderated, bare	7.71 ± 0.03	9.60 ± 0.03	710.1	306
104[b]	3 × 3 × 3 unmoderated, one side reflected	8.24 ± 0.03	10.15 ± 0.03	848.4	315
107	3 × 3 × 3 unmoderated, except 1.27-cm moderator cell around central unit, bare	7.72 ± 0.03	9.61 ± 0.03	712.5	306
106	3 × 3 × 3 unmoderated, except 1.27-cm-thick ^{6}LiD slab between two rows, bare	7.73 ± 0.03	9.62 ± 0.03	715.4	306
207	4 × 4 × 4 unmoderated, bare	7.86 ± 0.01	12.51 ± 0.01	1230	304
204	4 × 4 × 4, 1.27-cm moderator cells, bare	9.63 ± 0.01	14.19 ± 0.01	1939	307
205	4 × 4 × 4, 2.54-cm moderator cells, bare	13.64 ± 0.01	14.55 ± 0.01	2888	305
206	4 × 4 × 4, 3.81-cm moderator cells, bare	13.63 ± 0.01	14.62 ± 0.35[d]	2913[d]	318

[a]Two sides were reflected by 152.4- × 35.6- × 15.2-cm polyethylene body simulators and two sides by 152.4- × 7.6- × 7.6-cm arm simulators.
[b]Arrays 101, 102, 103, and 104 were actually taken to the critical state.
[c]The polyethylene reflector slab was 45.1 cm high × 35.1 cm wide × 20.5 cm thick.
[d]Extrapolated estimate; array was terminated at 15.25-cm horizontal center-to-center spacing when moderator cells touched.

(Continued)

TABLE IX (Continued)

Array Number	Description	Critical Center-to-Center Separation (cm)		Unit Cell Volume (cm^3)	Outer Can Temperature (K)
		Vertical	Horizontal		
		6.052-kg units with height-to-diameter ratio 1.493			
305	1 × 1 × 2 unmoderated, four sides reflected[a]	11.49 ± 0.03	---	---	300
306	1 × 1 × 5, 1.27-cm moderator cells, four sides reflected[a]	13.54 ± 0.03	---	---	319
105	2 × 2 × 1 unmoderated, bare	---	7.59 ± 0.03	---	308
215	2 × 2 × 2 unmoderated, bare	11.98 ± 0.02	9.76 ± 0.03	1 141	303
214	3 × 3 × 3 unmoderated, bare	13.68 ± 0.02	14.51 ± 0.03	2 878	308
213	4 × 4 × 1 unmoderated, bare	---	10.91 ± 0.02[e]	---	304
210	4 × 4 × 4 unmoderated, bare	47.12 ± 0.02[f]	11.93 ± 0.01	6 707	305
209	4 × 4 × 4 unmoderated, bare	32.12 ± 0.02[f]	13.09 ± 0.01	5 505	304
211	4 × 4 × 4 unmoderated, bare	22.12 ± 0.02[f]	15.23 ± 0.01	5 131	306
208	4 × 4 × 4 unmoderated, bare	17.12 ± 0.02[f]	17.28 ± 0.01	5 111	307
212	4 × 4 × 4 unmoderated, bare	13.12 ± 0.02	20.19 ± 0.02	5 349	314
201	4 × 4 × 4, 1.27-cm moderator cells, bare	25.79 ± 0.02	17.50 ± 0.01	7 897	311
202	4 × 4 × 4, 2.54-cm moderator cells, bare	25.82 ± 0.02	21.24 ± 0.01	11 653	312
203	4 × 4 × 4, 3.81-cm moderator cells, bare	25.82 ± 0.02	24.52 ± 0.02	15 526	311

[e]With this planar array located at five heights between 28.3 and 113.4 cm above the assembly table, the critical horizontal center-to-center separation varied between 11.12 ± 0.02 cm and 10.90 ± 0.02 cm. The nominal critical spacing is listed above.

[f]This set of measurements is referred to as the unit cell series; i.e., each array was 4 × 4 × 4, but the unit cell configuration was progressively distorted.

per batch. The neutrons in the first batch originated at a point close to the geometrical center of the array.

COMPUTATIONAL RESULTS

We performed an extensive series of 05R calculations, as described above, to investigate various aspects of the program. The calculations, though extensive, are not offered as representative of an exhaustive calculational study. We considered only those features that suggested themselves to us as of particular interest. In this section, we briefly present results concerning calculational uncertainties and the dependence of reactivity on various parameters. In addition, we provide complete tabulation of all computational results. The results of all calculations are summarized in Table X.

TABLE X

Summary of Calculated Results

System Description			k_{calc} [a]
2 × 2 × 2, 6-kg plutonium parts, unmoderated, unit cell model, 9.765-cm center-to-center (CTC), computational analog of critical array no. 215. (This is the Series 1 calculation referred to in the text.)			0.9872 ± 0.0005
2 × 2 × 2, 6-kg plutonium parts, unmoderated, *complete* geometry (including assembly machine) included, 9.76-cm CTC, computational analog of critical array no. 215. (This is the Series 2 calculation referred to in the text.)			0.9968 ± 0.0004
2 × 2 × 2, 6-kg plutonium parts, unmoderated, array composed solely of plutonium cylinders with 0.479-cm separation at midplane, computational idealization of array no. 215. (This is the Series 50 calculation referred to in the text.)			
Spacing =	6.525-cm CTC	0 surface-to-surface (STS)	1.161 ± 0.002
	7.525	1.0	1.077 ± 0.002
	8.725	2.2	1.007 ± 0.002
	9.65	3.125	0.974 ± 0.002
	10.025	3.5	0.960 ± 0.002
	10.525	4.0	0.947 ± 0.002
	11.525	5.0	0.922 ± 0.002
2 × 2 × 2, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 215. (This is the Series 7 calculation referred to in the text.)			
Spacing =	6.525-cm CTC	0 STS	1.198 ± 0.002
	7.525	1.0	1.101 ± 0.002
	8.725	2.2	1.039 ± 0.002
	9.325	2.8	1.020 ± 0.002
	9.65	3.125	1.005 ± 0.002
	10.025	3.5	0.991 ± 0.002
	10.525	4.0	0.982 ± 0.002
	11.525	5.0	0.950 ± 0.002
3 × 3 × 3, 6-kg plutonium parts, unmoderated, unit cell model, 14.505-cm CTC, computational analog of critical array no. 214.			0.986 ± 0.004
3 × 3 × 3, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 214.			
Spacing =	14.505-cm CTC		1.003 ± 0.004
	14.925		0.990 ± 0.006
	15.325		0.99 ± 0.01
4 × 4 × 1, 6-kg plutonium parts, unmoderated, unit cell model with 1-cm aluminum overhang, 10.925-cm CTC, computational analog of critical array no. 213.			0.988 ± 0.006
4 × 4 × 1, 6-kg plutonium parts, unmoderated, unit cell model with 10-cm aluminum overhang, 10.925-cm CTC, computational analog of critical array no. 213.			0.996 ± 0.003
4 × 4 × 1, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 213.			
Spacing =	10.125-cm CTC		1.024 ± 0.004
	10.525		1.019 ± 0.005
	10.925		1.004 ± 0.005
	11.325		0.977 ± 0.006
	11.725		0.972 ± 0.005
4 × 4 × 4, 6-kg plutonium parts, unmoderated, unit cell model, 11.935-cm CTC, computational analog of critical array no. 210.			0.981 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 210.			
Spacing =	11.525-cm CTC		1.004 ± 0.006
	11.935		0.995 ± 0.005
	12.325		0.982 ± 0.006

(Continued)

TABLE X (Continued)

System Description	k_{calc} [a]
4 × 4 × 4, 6-kg plutonium parts, unmoderated, unit cell model, 13.095-cm CTC, computational analog of critical array no. 209.	0.989 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 209.	
Spacing = 12.525-cm CTC	1.003 ± 0.006
13.085	0.998 ± 0.005
13.725	0.981 ± 0.007
4 × 4 × 4, 6-kg parts, unmoderated, unit cell model, 15.245-cm CTC, computational analog of critical array no. 211.	0.979 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 211.	
Spacing = 15.235-cm CTC	0.993 ± 0.004
15.525	0.991 ± 0.004
4 × 4 × 4, 6-kg plutonium parts, unmoderated, unit cell model, 17.275-cm CTC, computational analog of critical array no. 208.	0.981 ± 0.002
4 × 4 × 4, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 208.	
Spacing = 17.275-cm CTC	0.997 ± 0.002
17.525	0.991 ± 0.002
17.725	0.986 ± 0.002
4 × 4 × 4, 6-kg plutonium parts, unmoderated, unit cell model, 20.185-cm CTC, computational analog of critical array no. 212.	0.970 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 212.	
Spacing = 19.725-cm CTC	1.000 ± 0.006
20.185	0.998 ± 0.006
20.925	0.98 ± 0.01
2 × 2 × 1, 6-kg plutonium parts, unmoderated, unit cell model, 7.595-cm CTC, computational analog of critical array no. 105.	0.982 ± 0.003
2 × 2 × 1, 6-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 105.	
Spacing = 7.595-cm CTC	0.998 ± 0.004
2 × 2 × 2, 3-kg plutonium parts, unmoderated, unit cell model, 7.295-cm CTC, computational analog of critical array no. 101.	0.996 ± 0.002
2 × 2 × 2, 3-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 101.	
Spacing = 7.295-cm CTC	0.979 ± 0.008
3 × 3 × 3, 3-kg plutonium parts, unmoderated, unit cell model, 9.595-cm CTC, computational analog of critical array no. 103.	0.982 ± 0.002
3 × 3 × 3, 3-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 103.	
Spacing = 9.595-cm CTC	0.959 ± 0.005
4 × 4 × 4, 3-kg plutonium parts, unmoderated, unit cell model, 12.515-cm CTC, computational analog of critical array no. 207.	0.981 ± 0.002
4 × 4 × 4, 3-kg plutonium parts, unmoderated, array composed solely of solid plutonium cylinders, computational idealization of array no. 207.	
Spacing = 12.515-cm CTC	0.961 ± 0.005
4 × 4 × 4, 3-kg plutonium parts, 1.27-cm moderator thickness, unit cell model, 14.21-cm CTC, computational analog of critical array no. 204.	0.998 ± 0.005
4 × 4 × 4, 3-kg plutonium parts, 2.54-cm moderator thickness, unit cell model, 14.56-cm CTC, computational analog of critical array no. 205.	1.000 ± 0.006

(Continued)

TABLE X (Continued)

System Description	k_{calc} [a]
4 × 4 × 4, 3-kg plutonium parts, 3.81-cm moderator thickness, unit cell model, 14.62-cm CTC, computational analog of critical array no. 206.	1.010 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, 1.27-cm moderator thickness, unit cell model, 17.51-cm CTC, computational analog of critical array no. 201.	0.991 ± 0.006
4 × 4 × 4, 6-kg plutonium parts, 2.54-cm moderator thickness, unit cell model, 21.24-cm CTC, computational analog of critical array no. 202.	0.996 ± 0.003
4 × 4 × 4, 6-kg plutonium parts, 3.81-cm moderator thickness, unit cell model, 24.52-cm CTC, computational analog of critical array no. 203.	0.998 ± 0.004

[a]The uncertainties quoted are only the statistical uncertainties calculated by O5R.

Calculational Uncertainties

Effect of Neutron Setting

We wished to assess the effect on reactivity of possible incomplete achievement of an equilibrium neutron distribution. We considered two systems in our attempt to accomplish this assessment. Both were idealizations of experimental array number 215 (see Table IX).

In the first (Series 50), all material external to the plutonium parts was omitted. The 6-kg parts thus had a 0.479-cm gap at their midplane. In the second system (Series 7), everything was as in the first system, only the two halves of the 6-kg parts were joined at the midplane.

For reasons that will become apparent, k was calculated for various spacings between the parts. The spacings were always uniform in the sense that separation between curved surfaces (in both horizontal directions) was maintained equal to the vertical separation between the flat ends.

In most of the calculations, 250 neutron batches were processed. The 05R calculations produce an estimate of the effective multiplication constant $\bar{k}(N)$ at the conclusion of each batch history. For both Series 50 and Series 7, we determined the quantities $\Delta k_s(N)$, where

$$\Delta k_s(N) = \bar{k}(N) - \bar{k}(100)$$

and N, the batch number, assumed the values 120, 200, and 250.

For each series we ran a total of 42 problems. We determined six distributions of $\Delta k_s(N)$ [number of cases in a 0.001 range versus $\Delta k_s(N)$ for N = 120, 200, 250, for Series 50 and Series 7]. This procedure was performed without regard to the variations in spacing already mentioned. (The Series 50 problems were run with seven different spacings, Series 7 with eight different spacings.)

We then calculated $\Delta\bar{k}_s$ as the mean for each of the six sets of data. In all cases this nearly vanished ($\Delta\bar{k}_s < 0.001$ or $\Delta\bar{k}_s \approx 0.001$). Accordingly, we estimated a probable error σ_s as that value of Δk_s such that half the cases were contained in the interval $-\sigma_s \leq \Delta k_s \leq \sigma_s$. The results are tabulated in Table XI.

We note that in all cases $|\Delta\bar{k}_s|$ is considerably smaller than σ_s and σ_s does not decrease significantly as N increases. We take this to indicate that (for these all-metal systems, at least) equilibrium has been attained essentially by the time 100 neutron batches have been processed.

Effect of Structure Outside the Unit Cell

Again, we ran two series of problems relevant to experimental array number 215. In the Series 2 problems, all structure was represented in the calculation insofar as practical. We did omit all structure outside the assembly machine itself. Included however, were the support columns, both above and below the array, and the table, clamping devices, and upper support structure. The columns were represented exactly, whereas the other structural elements were homogenized over regions (squat parallelepipeds) expected to provide a fair approximation to the actual neutronic effect.

In the Series 1 problems, we represented the array solely in terms of the corresponding unit cell. In this representation an array unit is surrounded by a fictitious box. The faces of the box

TABLE XI

The Average Change in the Multiplication Constant from N = 100 and the Probable Error for Six Sets of Data

N=		120	200	250
Series 50	$\Delta\bar{k}_s$	-0.0004	0.0014	0.0015
	σ_s	0.0015	0.0045	0.0045
Series 7	$\Delta\bar{k}_s$	-0.0005	-0.0002	-0.0006
	σ_s	0.0025	0.0030	0.0030

are located at the midplanes between adjacent array elements. Within the unit cell the support structure with the support tube, etc. included is represented in the detail of Fig. 2. The homogenized version of the space between the plutonium parts was used. The array is then approximated as an array of these unit cells. It should be apparent that no structure outside the unit cell can affect the results of these calculations.

We wanted to obtain these comparison results because in all other calculations the arrays were approximated by their unit cell representation. An estimate of the effect of this approximation on our calculated results was therefore desired.

In both the Series 1 and Series 2 problems, the array elements were spaced at the experimentally determined critical spacing. In the Series 1 calculations, we processed 11 443 batches of 200 neutrons each. In the Series 2 calculations, we processed 23 496 batches of 200 neutrons each. The results were

$$\bar{k} \text{ (Series 1)} = 0.9872 \pm 0.0005$$

$$\bar{k} \text{ (Series 2)} = 0.9968 \pm 0.0004 \ .$$

Because we (almost) never processed fewer than 250 batches in a given run, and sometimes as many as 2500, we are of the belief that the uncertainties in the $\bar{k}$ values due to settling are negligible. Also, because the effect we wish to evaluate is obtained as the difference between two $\bar{k}$ values, we believe the uncertainty in it due to cross sections is a second-order effect.

Accordingly, for this system, we assess the structure outside the unit cell representation of the array to have a Δk value of 0.0096.

Uncertainty Due to Cross Sections

The Series 2 problems described in the preceding section could be used to estimate the uncertainty in our calculational results produced by cross-section error. We have already noted that the effect of settling should be negligible for this series. Also, the structure inside and external to the array was represented nearly exactly. Thus, the error in the calculated value of k should be due almost solely to cross-section errors Δk_c.

$$\bar{k} \text{ (Series 2)} = 0.9968 \pm 0.0004$$

$$\Delta k_c = 1 - \bar{k} \text{ (Series 2)}$$

$$= 0.0032 \pm 0.0004 \ .$$

The cross-section component of the uncertainty in the calculated results (all-metal systems only) is thus seen to be but a fraction of a percent.

A similar evaluation for arrays containing moderating materials was not done.

Dependence of Reactivity on Various Parameters

Reproduction Constant Versus Spacing

We have defined the 7- and 50-series problems in a preceding section (see Table X). Note that for these series, we calculated k versus spacing. Figure 8 summarizes this variation. (Note that the Series 7 calculations pertain to a denser array. Thus, for a given spacing, we expect the k value calculated for the Series 7 system to exceed that for the Series 50 system. Reference to the figure shows this expectation to be borne out.)

Worth of Structure in the Unit Cell

We have also defined the Series 1 and Series 50 systems (see Table X). The only difference between the two systems is that the structure

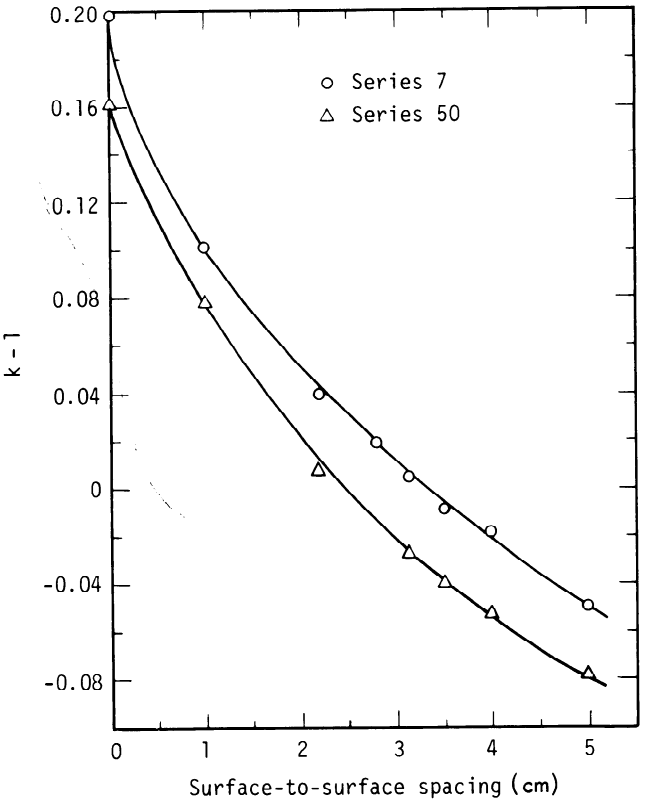

Fig. 8. Reproduction constant minus one versus surface-to-surface spacing for Series 7 and Series 50 problems. Series 7 is an idealization of array no. 215 where all material other than the plutonium is deleted and the 6-kg plutonium parts have no separation at their midplanes. Series 50 is an idealization of array no. 215 where all material other than the plutonium is deleted and the 6-kg plutonium parts are divided into two 3-kg parts with a 0.479-cm gap between their adjacent flat faces. The Monte Carlo calculated points are plotted to indicate a measure of the scatter in the calculations.

internal to the unit cell is present in Series 1 and deleted in Series 50. (In addition, all Series 1 problems were run with a constant plutonium surface-to-surface spacing of 3.24 cm. In Series 50, the spacing was varied and in fact was not calculated at a spacing of 3.24 cm. We can infer k for this spacing from Fig. 8.)

Thus, the worth of the material in the unit cell is

$$\Delta k_1 = \bar{k}_1\,(3.24\text{ cm}) - \bar{k}_{50}\,(3.24\text{ cm})$$
$$= 0.98720 - (1 - 0.031)$$
$$= +0.018\ .$$

Worth of Pairing the 3-kg Plutonium Parts

For this evaluation we refer to the already defined Series 7 and Series 50 problems. The only difference between these two systems is that in Series 50 the 6-kg plutonium parts are solid, whereas in Series 7 they are divided into two 3-kg parts with a 0.479-cm gap between their adjacent flat faces.

The worth of pairing the two 3-kg parts in the array is thus given by

$$\Delta k_2 = \bar{k}_7\,(S) - \bar{k}_{50}\,(S)\ ,$$

where S is the spacing between the parts as already defined. The results are summarized in Table XII. The worth Δk_2 is seen to be independent of spacing (although we note some random variability) and is ~0.03.

TABLE XII

Worth of Pairing the 3-kg Plutonium Parts in a 2 × 2 × 2 Array of 6-kg Plutonium Parts

(The 6-kg parts are each composed of two 3-kg parts.)

Surface-to-Surface Spacing (cm)	$\bar{k}$ (Series 7) ± 0.002[a]	$\bar{k}$ (Series 50) ± 0.002	Δk_2
0	1.198	1.161	+0.037
1.0	1.101	1.077	+0.024
2.2	1.039	1.007	+0.032
3.125	1.005	0.974	+0.031
3.5	0.991	0.960	+0.030
4.0	0.982	0.947	+0.034
5.0	0.950	0.922	+0.029

[a]The uncertainties quoted in Table XII are only the statistical uncertainties as calculated by 05R.

CONCLUSIONS AND SPECULATIONS

We have presented the results of a series of experimental determinations of the critical parameters of a large number of plutonium arrays. We have also presented the results of an extensive series of Monte Carlo calculations relevant to these arrays.

We believe that the following comments are of value:

1. Elementary array calculation methods will likely be displaced by Monte Carlo methods because of the better accuracy of Monte Carlo and the increasing availability of large computers.
2. Monte Carlo techniques are also important because clean experimental data generally cannot be extrapolated to real systems of interest.
3. We note a decrease in calculational accuracy as moderating materials are incorporated into the arrays. This arises from the approximation made to deal with thermal neutrons. An accurate treatment is difficult and this problem is likely to plague Monte Carlo results for a considerable time to come.
4. The effect of cross-section errors (in one case at least) is <1% in k. Thus we believe that improvement of Monte Carlo neutron cross sections is of secondary importance only.

References 14 through 24, along with Refs. 10, 11, and 12, provide a complete bibliography of Livermore Plutonium Array Program documentation.

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