Massive Subcritical Compact Array of Plutonium Metal

By
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MASSIVE SUBCRITICAL
COMPACT ARRAYS

of

PLUTONIUM METAL

by

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Consultant

reviewed by

Donald R. Ferguson
Lead Experimenter

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ABSTRACT

Two experimental critical-approach programs are reported. Both were performed at the Rocky Flats Plant near Denver, Colorado; and both date back to the late 1960s. Both involve very large arrays of massive plutonium ingots. These ingots had been cast in the foundry at the Rocky Flats Plant as part of their routine production operations; they were not specially prepared for either study. Consequently, considerable variation in ingot mass is encountered. This mass varied between approximately 7 kg and a little more than 10 kg. One program, performed in the spring of 1969, involved stacked arrays of ingots contained within cylindrical, disk-shaped, thin, steel cans. This program studied four arrays defined by the pattern of steel cans in a single layer. The four were: $1 \times N$, $3 \times N$, $2 \times 2 \times N$, and $3 \times 3 \times N$. The second was a tightly-packed, triangular-pitched pattern; the last two were square-pitched patterns. The other program, performed about a year earlier, involved similar ingots also contained in similar steel cans, but these canned plutonium ingots were placed in commercial steel drums. This study pertained to one-, two-, and three-layered horizontal arrays of drums. All cases proved to be well subcritical. Most would have remained subcritical had the parameters of the array under study been continued infinitely beyond the reciprocal multiplication safety limit. In one case for the drum arrays, an uncertain extrapolation of the data of the earlier program suggests that criticality might have eventually been attained had several thousand additional kilograms of plutonium been available for use.
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INTRODUCTION

This document is the sixth in a series of seven peer-reviewed papers written under the International Criticality Safety Benchmark Evaluation Project. Originally, only six were planned, but the value of the seventh became apparent in 1997. All seven place into the public domain previously unpublished or inadequately documented experimental data generated at the Rocky Flats, Colorado, Critical Mass Laboratory (CML). The benchmark evaluation project is administered for the Department of Energy by J. Blair Briggs of the Lockheed Idaho Technologies Company, Idaho National Engineering and Environmental Laboratory (INEEL). The previous five\textsuperscript{1,2,3,4,5} papers were published between 1994 and 1997.

In this sixth paper, two experimental programs are reported. One pertains to only four array configurations of massive ingots of plutonium metal, and these data have never before been published in any form. The metal was contained in cylindrical, disk-shaped, thin, commercial steel cans designed for storage of bulk photographic film. These four configurations consisted of different patterns laid out on the lowest layer, and the array assembly of the same pattern was built vertically. Experiments were performed at the Rocky Flats CML in 1969, and all were terminated while the array was still well subcritical. Reasonable extrapolations of the data in most cases revealed that a continued assembly of the array would have remained subcritical regardless of the height of the array. Only one array pattern, the largest,\textit{may} have yielded criticality had sufficient plutonium been available and if the reciprocal multiplication safety limit had not been approached.

The other program pertains to the same canned massive ingots of plutonium, but these were placed within commercial 55-gallon steel drums. Then, horizontal arrays of these drums were assembled in an effort to assess the array's proximity to criticality. Both one-, two-, and three-layered arrays of drums were measured. All configurations remained well subcritical and probably would have remained so if the array could have been extended to infinity. An extrapolation of the three-layer array to an even larger horizontal array suggests that criticality may have been possible near the completion of a third layer. Results of this program have been reported previously in an internal Rocky Flats report\(^6\). They will not be repeated here because that short report is appended to this document with no modifications. This author can add little to its content.

Experiments from both programs were terminated at well subcritical configurations. This was an intended safety consideration. The safety issue was the presence of personnel during the experiment - configurations were manually assembled and reasonable safety precautions precluded construction all the way to criticality.

These experiments are inadequately documented for benchmark purposes. The value of this paper lies in two facts: (1) A very large mass of plutonium metal was assembled in a relatively compact region and still remained assuredly subcritical. Almost a thousand kilograms in a space little more than a meter square by two meters high is still subcritical by a significant margin! The value of this information to the benchmark evaluation project lies in this limiting parameter boundary - plutonium arrays do not always achieve criticality simply because the metal mass in a confined area becomes very large. (2) These data may now be dismissed from further consideration by the benchmark evaluation group because the data is not sufficiently quantitative. The information does possess qualitative value in assessing very large massive arrays of plutonium metal.

Experiments in the vertical program included four configurations of plutonium ingots laid out on the floor. Each ingot weighed from about 7 to a little over ten kilograms, and each was contained in a commercial film can designed for the storage of bulk quantities of photographic film. These cans were not much larger than the ingots themselves. The four configuration patterns of ingots in film cans were:

1) a simple stack of single ingots, called a 1x1xN array,
2) a vertical stack of ingots arrayed in a tight triangular pattern of 3 ingots per layer.
3) a vertical stack of ingots arrayed in a 2x2xN pattern, and
4) a vertical stack of ingots arrayed in a 3x3xN pattern.

Adjacent layers of film cans always contained plutonium; no empty layers of film cans were inserted between fissile layers. The plutonium within the arrays was relatively compact.

This study, like all experiments performed at Rocky Flats, were used to provide nuclear criticality safety data to ensure continued safety of plant operations. Data were usually used two ways in those early days of nuclear criticality safety. First, they were applied directly to plant operations if those operations were deemed suitably similar to experiments. The degree of similarity required was often left to the discretion of the Criticality Safety Engineer. Secondly, they were used to compare experimental results with calculations from then state-of-the-art computational methods in vogue at the time. Again, details of this comparison were often left to the discretion of the Safety Engineer. For this study, the latter assessment was never performed, partly because computational methods before the advent of Monte Carlo codes required certain conditions of geometrical symmetry which did not exist in this program.

These experiments, even though subcritical and quite old, should prove especially useful to the International Criticality Benchmark Evaluation Project because of the very large masses considered. Experimental data are readily available for computer validation for masses often encountered in a production plant - up to several kilograms. Little experimental data exists,
however, for significantly larger masses. It would be comforting to have these same computer
codes predict that these massive stacks of plutonium in a confined space was substantially
subcritical. Confidence in any computational model used over any given range of masses would
certainly be improved if that same model were found to predict subcriticality correctly for
significantly larger masses equally well. These results may provide that missing experimental data.
In summary, then, the special value of these results lie in their very large mass, far outside the
normal range encountered in production.

This paper required considerable historical research and personal recollection of important
information. This defect exists for the following reasons: (1) This author was not involved in
these experiments, although the lead experimenter is still available for consultation and he
reviewed the text before publishing it. (2) The entire program, itself, was somewhat arbitrarily
chosen simply because it seemed interesting and useful. It was never an official program of the
laboratory; and it never fell under the auspices of a formal, written, plan. (3) The eventual value
of these results was not fully recognized at the time. (4) Documentation of such casual studies
was not required in the 1960s. Finally, (5) The wealth of experimental detail required for Monte
Carlo codes of the 1990s was not realized.

7 This reason is totally inconsistent with policies and procedures in place throughout the industry
after about 1970.
Theories

Approaches to criticality for both experimental programs were monitored by a method called the **Reciprocal Multiplication** technique. This was done to ensure safety. Here, the fact is used that the instantaneous neutron flux at any and every point within a system increases as criticality is approached. This flux is proportional to a neutron count rate measured at some fixed distance from the assembly being built using one or more neutron detectors. The previously unreported vertical array study provides an example. At any height, \( H \), of a growing assembly, this count rate, \( C(H) \), will be some factor greater than at the start of an experiment, \( C_0 \). Most of this increase is due to the increase in the nuclear fission density within the array. Some is due to the mere addition of plutonium as discussed below. This ratio, \( C(H)/C_0 \), is called the **Multiplication** of the system\(^8\). The inverse of this is the reciprocal multiplication already mentioned: \( C_0/C(H) \).

At the critical height, \( H_c \), never actually attained in these subcritical experiments, \( C(H_c) \) would be essentially infinite relative to that initial count rate, and \( C_0/C(H) \) would, obviously, approach zero. This produces an attractive feature for graphing safe critical approaches. Extrapolating graphs to infinity is nebulous whereas extrapolations to zero are clearly defined.

For safety, especially when humans are present, all parameters of critical approach experiments should be fixed save one. Otherwise, changes in the reciprocal multiplication curve could be attributed to either true increased multiplication or to changes in the manner other parameters respond to neutrons. In this vertical array program, the one variable changed was the height of the array being assembled. All other features remained fixed.

\(^8\) Actually, this empirical ratio is only an approximation to the true multiplication because of other complications to the theory.
One complication to the reciprocal multiplication technique appears when plutonium is the
fissile material. Plutonium is naturally a copious emitter of neutrons; and these are usually
sufficient to provide that initial neutron flux safely. An external source - such as that used in
uranium experiments - is not needed. The problem is that the addition of a 2\textsuperscript{nd}, 3\textsuperscript{rd}, ... N\textsuperscript{th} layer of
plutonium introduces additional source neutrons as well as any increased multiplication neutrons
caused by the closer proximity to criticality. In other words, each reciprocal multiplication value
presented in later tables is incorrectly estimated because it is formed from the following ratio:

\[
\begin{align*}
too \text{ few source neutrons obtained by ignoring source neutrons from upper layers,} \\
true \text{ multiplication neutrons + those same ignored source neutrons}
\end{align*}
\]

instead of the following better ratio:

\[
\begin{align*}
\text{all source neutrons regardless of which layer they come from,} \\
true \text{ multiplication neutrons only}
\end{align*}
\]

Fortunately, the finite number of neutrons associated with any one ingot is negligibly small
compared to the nearly infinite count rate that would exist near criticality. Therefore, the error is
insignificant and does not invalidate any of the results presented in this paper.

This difficulty could easily have been avoided by “mapping” contributions of source
neutrons from each future ingot’s location in the N layers of the array to be built. This would be
done by placing a single, well-subcritical, plane of ingots at each level to be occupied later by
plutonium in building the experimental array. All other locations would be filled with empty film
cans. The contribution to source neutrons from each level, then, would be recorded for later use.
When an actual array of plutonium was built with fissile metal in all locations, the source term, \(C_o\),
would simply have been the mathematical sum of neutrons from ingots at each of the N layers as
expressed in the numerator of the second ratio, above.
That better source handling procedure was not employed in this experimental program. Instead, $C_0$ was defined as the neutron flux from an obviously subcritical arrangement of ingots. This consisted of either one or two layers of ingots of the pattern being studied. Then, as subsequent levels of the array were added, this no-longer-correct $C_0$ (too small because of omitted source neutrons) was divided by an also incorrect neutron flux (too large because it wrongly included those same source neutrons). The resulting reciprocal multiplication is wrong because both errors are non-conservative in the sense that they predict a multiplication somewhat smaller than the better procedure would have yielded. Conceivably, then, such an error could predict subcriticality when, in truth, the array under construction might have attained criticality had it been built far enough.

Some effort was made by the lead experimenter to adjust these reciprocal multiplication curves for the increased number of source neutrons introduced as the array was built. This is tersely recorded on the original data pages, but the details of his correction is not known three decades later. His corrected data is included in later tables but without explanation or further elaboration. A second attempt to correct for this effect is also offered by this author. Those results, too, are included in the tables.

Still, even at this late date, a theoretical correction to the existing reciprocal multiplication curves published in this paper is possible. At best it would be only a crude approximation. This procedure would be to assume that the contribution to $C_0$ from higher layers of plutonium would be the same as that from the bottom layer actually used but diminished inversely with the distance to the counter tubes. For example, if the $C_0$ actually used was obtained from just the bottom layer of plutonium which was a distance, $D$, above the counter tubes and if subsequent layers of plutonium were, say, $3D$, $6D$, $9D$, etc. away from the counters, then an approximate $C_0$ for the 4th layer would be:

$$C_0 \left[ 1 + 1/3 + 1/6 + 1/9 \right].$$

This correction has been attempted for the published data, but it may not be warranted.
DATA QUALITY

This paper was difficult to write for a number of reasons; and the usefulness of the data may be diminished somewhat because of that. Some factors contributing to that difficulty were:

1) The experiments were performed almost three decades ago.
2) The author of this paper was not involved in them; his recollection is of no value.
3) The studies were not well documented. The lead experimenter was a criticality safety engineer and not trained in critical mass physics. He was well-intentioned and had taken part in other *in situ* experiments; but he failed to record some important information.\(^9\)
4) The studies served an immediate purpose related to a specific safety issue and was not viewed useful beyond that. They were considered a "short", casual, study and not even regarded as one of the laboratory’s experimental programs.

On the positive side, the lead experimenter, Donald R. Ferguson, is still living in the greater Denver area and is available for private communication. He has also agreed to review this paper critically before publication. Although retired, he still serves the industry as a consultant on safety issues at Rocky Flats. In addition, a number of other retired Rocky Flats employees live nearby and have contributed their knowledge and recollections to this paper. They are well-acquainted with the plutonium ingots cast at Rocky Flats, although most of them were in no way connected with the experiments themselves.

If these experiments reported *critical* data, the dearth of technical detail would render them useless for the purposes of benchmark validation. Circumstances which render these results useful to some degree are:

---

\(^9\) Reciprocal multiplication curves are sometimes ill-defined or ambiguous; and important dimensions were not recorded in many cases and must be inferred from other data or recalled from memories. No written experimental plan could be found.
1) They represent a very large mass of plutonium metal in a relatively confined space that proved to be well subcritical.

2) Approximations or estimates of missing dimensions are possible through tersely written notes and 3-decade-old recollections.

Another problem was the large variability in the mass of the ingots used. These were production-line ingots borrowed from the plant's normal production stream for this brief study. They were not specifically made for this experiment. Masses varied by a few kilograms from an average mass of several kilograms. Specific masses used are given later in the tables. Reciprocal multiplication curves were graphed as a function of the number of plutonium ingots in an array. They were not graphed as a function of plutonium mass, although the needed information is provided such that these other graphs could be constructed if desired.

All available original data has been collected into one location for this paper. Sometime in 1998, it will be contributed to the Archives of Criticality Experiments which are maintained at the Los Alamos National Laboratory (LANL). The two files are expected to be identified simply as "Plutonium Ingot Studies". This transfer into the archives will await publication of this document.
FILM CANS

Each plutonium ingot was stored in a commercial steel film can. These cans are intended for the storage of bulk quantities of photographic film. There was only one ingot housed in each can even though ingots varied considerably in mass. These film cans were large-diameter, thin-walled, two-piece canisters probably spun or drawn from a single sheet of steel per piece. The bottom piece consisted of a flat, circular bottom with a raised cylindrical wall. The height of this wall defined the overall height of the finished film can because the top piece rested on this wall’s upper edge. The top piece was similar to the bottom except that its circular diameter was just enough larger that its cylindrical side slipped easily down over the bottom piece.

A familiar analogy would be a coffee can and its plastic lid intended to retain freshness. The analogy differs in that the plastic lid fits the can more tightly than the film can’s top fit the bottom. That fit may best be described as a loose, easy fit. Another difference in the analogy is the height-to-diameter ratio of the two pieces. The coffee can is much taller than its diameter, whereas the film can was much larger in diameter than the height of its cylindrical walls. The coffee can’s plastic lid also has the wrong aspect ratio. The heights of the sides of the film can’s top and bottom pieces were almost equal. Figure 1 shows that a finished film can had the outside wall extend about a quarter of the way down the outside toward the bottom of the can. One retired Rocky Flats employee, who had spent his career preparing and canning these castings, revealed in a private communication\textsuperscript{10} that these cans were usually taped shut with a 50-mm-wide vinyl plastic tape after being loaded with a plutonium ingot.

The exact packaging of the plutonium metal within the film can is not recalled nor documented. Some plastic wrapping would have been used; but the amount is not certain. A reasonable guess would be that each ingot would have been wrapped two to four times on all sides with thin plastic sheet and vinyl tape. The wrapped ingot was loose within its can; so its exact location or orientation can never be know.

\textsuperscript{10}Louis DiGiallonardo, private communication, November 22, 1995.
Figure 1. The author holds one of the film cans used for another purpose. A rectangular hole in the top lid positioned a plastic block used to hold, in turn, uranium solution standards. Aside from this hole, the can is representative of those used to contain plutonium ingots.
The specific film cans to be used in this study were tersely dimensioned in two documents from Rocky Flats, both generated in the late 1960s. The two do not agree with one another, although the difference is not large. The Rocky Flats Plant internal report, RFP-1242, dated 1968, describes these film cans as "...19-inch diameter by 23\(\frac{3}{8}\) inches high". The other document, a Rocky Flats internal plant memo dated February 24, 1969, described the cans as "...17 inch diameter by 22 inch steel cans". The former document pertained to the drum experiment; the later, the vertical stacked array. The lead experimenter stated most confidently in a telephone conference\(^\text{11}\) that the larger diameter can is correct and the smaller is an error. The error, he said, was the result of dimensions provided prior to actual receipt of material, but they proved to be wrong when the material arrived. Therefore, the 19" diameter can is assumed for the vertical stack program in spite of the February 24, 1969, memo. Several long-time Rocky Flats employees do not recall ever seeing multiple sizes of film cans in use at the plant, although that may still have been the case.

Whichever dimensions were actually correct, they are suspected to be nominal outside measurements, although that fact is nowhere recorded. It is assumed because that would be the conventional manner of defining such a commercial product, there would be little purpose in specifying inside dimensions. The departure from nominal dimensions is also not known, although commercial products are generally fairly true to their nominal dimensions.

The metal was fairly thin. Mr. DiGiallonardo described the cans as being "flimsy", although this author recalls the metal as being stout enough not to cause concern over their containing massive pieces of plutonium. In 1995, Mr. DiGiallonardo estimated the thickness of the steel as "about a sixteenth of an inch". This thickness is not inconsistent with the author's recollection, but some uncertainty should be assigned because this dimension is not documented. The thickness suggested for this film can stock is \(1.6 \pm 0.5\) mm.

\(^{11}\) Donald R. Ferguson, Private Communication, February 12, 1998.
If all these observations are correct, then the outside diameter of the film can would be about $483 \pm 5$ mm, the outside height on the two-piece can would be $60.3 \pm 2$ mm, and the thickness would be $1.6 \pm 0.5$ mm. These uncertainties are quite arbitrary. They are large because neither variations between cans nor the exact measure referred to by the “19 inch diameter” dimension are known.

In addition, a small possibility exists that the 17" diameter was correct. Even a third diameter seems possible. Mr. DiGiallonardo was asked by this author during the private communication if he recalled these film cans “as being about 24 inches by about 2 inches thick”. He agreed. Whether he was biased by the author’s preemptive statement of dimensions, just speaking in approximate terms, or more than one sized film can was used on occasion at Rocky Flats is not known.

Some detective work into film can dimensions further clouds the question. (1) A hand-drawn drawing of the floor pattern for the 3x3 array of ingots had the false floor beneath the array drawn in and dimensioned. If this drawing was drawn to scale, these cans appeared to be 439 mm in diameter, closer to the 17" size than the 19". (2) Some loose leaf papers that seem to pertain to a criticality safety evaluation of these same film cans containing plutonium implies that they fit into lattice spaces 457.2 mm square (18") and also fit onto a rolling cart with a deck about 432 mm by 483 mm (17" x 19"). Neither of these points are consistent with either a 19" or 24" diameter. (3) Even the can’s height is in question. The 63.5 mm dimension is supported by a hand-written note on a reproduced\textsuperscript{12} copy of an original reciprocal multiplication curve for the 3x3xN array. The text of that note is: “stored in 22-inch thick film cans”. The original page was dated March 17, 1969; but the date of this entry is not known for certain because it is written in a different colored ink. The handwriting is that of the lead experimenter’s, and even he would not likely have made that entry after about 1970.

\textsuperscript{12} Images left by the reproducing machine identify the copy as one made before about the mid 1970s.
The metal stock was certainly steel; that fact has been substantiated by everyone with any knowledge of the question. The precise composition is not known, but the composition of any inexpensive, readily available, steel alloy commonly used in spinning and/or drawing operations may be assumed with little risk of significant error. This low risk is especially true considering the qualitative nature of this program.
PLUTONIUM

Rocky Flats continually cast plutonium ingots as part of their contribution to the United States' military preparedness between the 1950s and the 1980s. These castings were an early stage in the production of weapon's components. Castings were later rolled, punched, annealed, and machined into their final shape. Ingots used in this experimental program were routine castings temporarily diverted from the production stream for this program; they were not especially cast for this study.

Still, these early rectangular castings were the ones used in both criticality safety studies reported here. Several people recall that the rectangular castings were made in a variety of sizes. Two common ones were 203.2 mm x 254.0 mm and 228.6 mm x 304.8 mm. The largest castings, according to one retired Rocky Flats employee\(^\text{13}\), were 305 mm square and weighed up to 13 kg each! Rocky Flats' employees who had been intimately involved with such casting over many decades recalled average ingot masses at between 10 kg and 12 kg during the early years (1960s and 1970s). A nominal average casting was said to weigh about 10.5 kg.

Considerable variation in final masses of cast ingots was observed. For any given size, variations in mass were the consequence of variations in just one of the two larger dimensions of the casting. The thickness was always fixed because of the manner in which ingots were cast. The graphite mold used to receive the molten metal stood on its edge; so the thickness and one other dimension were fixed by that orientation. The other dimension depended on the height of the particular pour, and this, in turn, depended on the amount of plutonium in the furnace's charge.

\[^{13}\] Jack Weaver, Private Communication, November 22, 1995.
The ingot thickness has been recalled differently by different persons. The lead experimenter remembers all castings coming from molds at Rocky Flats were 10.16-mm thick. One retired employee\(^4\), on the other hand, recalls the thickness at 12.065 mm.

Most plutonium metal at Rocky Flats was cast in its delta phase. Some ingots over the years were cast as alpha-phase metal, but none of these are believed to have been used in these two programs. The delta-phase metal was probably (but not certainly) stabilized in that phase by being alloyed with Gallium. The density of the metal castings was about 15.8 mg/mm\(^3\). However, no density information specific to the ingots used was discovered on any of the loose leaf pages associated with these programs.

The masses of the two most common sizes of ingots, assuming the most likely thickness, at their minimum and maximum densities would be:

\[(203.2 \text{ mm}) \times (254.0 \text{ mm}) \times (10.16 \text{ mm}) \times (15.8 \text{ mg/mm}^3) = 8.3 \text{ kg}\]
\[(203.2 \text{ mm}) \times (254.0 \text{ mm}) \times (10.16 \text{ mm}) \times (15.8 \text{ mg/mm}^3) = 8.8 \text{ kg}\]
\[(228.6 \text{ mm}) \times (304.8 \text{ mm}) \times (10.16 \text{ mm}) \times (15.8 \text{ mg/mm}^3) = 11.2 \text{ kg}\]
\[(228.6 \text{ mm}) \times (304.8 \text{ mm}) \times (10.16 \text{ mm}) \times (15.8 \text{ mg/mm}^3) = 11.4 \text{ kg}\]

Few of the ingots actually used exceeded the smallest of these masses, although several came close. Most were much lighter. The conclusion is that most - if not all - ingots were the smaller size and, even then, not the full 254.0 mm tall. They could have been even shorter versions of the larger size, but the lead experimenter recalls that probably all ingots were cast in the smaller mold. Scanning ingot masses actually used reveals that the smallest ingot was a little over 5 kg. The lead experimenter recalls that the earlier (drum) program used production ingots which he had the opportunity to select to be close to the maximum possible mass. The later study did not have that option, so production ingots were accepted for use as available. This accounts for the much wider range of ingot masses used in this second program.

Ingots used in the 1 x N array ranged from 8.360 kg to 10.219 kg; the average and standard deviation of those 15 ingots was 9.354 ± 0.829 kg. Masses for the triangular 3 x N array ranged from 5.552 kg to 10.313 kg; the average and standard deviation of those 37 ingots were 8.470 ± 1.362 kg. Masses for the square-pitched 2 x 2 x N array also ranged from 5.552 kg to 10.313 kg (the heaviest ingot), but the average and standard deviation of those 52 ingots were different: 8.616 ± 1.298 kg. Masses for the square-pitched 3 x 3 x N array ranged from 5.110 kg (the lightest ingot) to 10.313 kg; the average of those 91 ingots was 8.425 kg. Actually, data was taken for only ten layers of nine ingots, but a 91st ingot had been added to the array before the experiment was terminated. The standard deviation for this array was not calculated. The specific mass of each ingot in all four arrays is identified in later tables, although which ingot appeared where in a layer is not recorded.

Not only did the masses vary widely throughout the array, but three other factors complicate any attempt to describe these assemblies at all precisely.

(1) Ingots were selected randomly for placement within the array. Variations in mass were in no way distributed evenly. For example, four consecutive ingots in the 3 x N triangular array weighed in excess of 10 kg each, and seven of ten ingots in that same general region exceeded 10 kg. Elsewhere, strings of 6, 7, and 8 kg ingots can be found. Reciprocal multiplication graphs were drawn as a function of the number of layers in the array without consideration of variations in the average mass from layer to layer.

(2) Relative orientations of ingots within cans is not at all known. The best assumption that can be made is that ingots were randomly oriented relative to one another. On occasion, reciprocal multiplication data was repeated with one to several cans in the top layer of the array under construction rotated 45°. This rotation never changed the observed count rate outside the expected statistical reproducibility of the original rate; so this orientation may be unimportant.
(3) Some plutonium ingots may have slid to one side of the film can rather than being centered. If so, adjacent ingots may have been horizontally bunched as close together as possible or as far apart as possible. The lead experimenter recalls that a sponge rubber was placed inside each film can to preclude this. Considering this rubber and the large amount of plastic wrapping material enclosing each ingot, ingots are believed to have remained relatively well centered. Furthermore, plutonium metal would have been handled with considerable care considering the implications of careless handling. Film cans were almost certainly maintained in a horizontal orientation at all times, so sliding about would not be very likely.

Each ingot was wrapped in at least one plastic bag sealed with vinyl tape for contamination control, although some recall additional plastic bags being used to ensure that control. RFP-1242 describes an amazing 1.1 kg of plastic being used per film can such that “If the plastic were distributed evenly around the ingot, the thickness of the plastic would be about 0.2 inches.” Two private communications\textsuperscript{15,16} describe an ingot as being at least double bagged. Plastic bags and vinyl tape contained considerable hydrogen because they were plastic. Still others recalled that sponge rubber (also, containing hydrogen) was used to protect the ingot and to keep it centered.

\textsuperscript{15} Donald R. Ferguson, Private Communication in response to the author’s note of November 20, 1995.

TEMPERATURE

These experiments were conducted in a typical temperature-controlled room common to most scientific laboratories in that era. The entire building was heated in the winter and cooled in the summertime to provide a comfortable work environment. Room #102 was quite comfortable to work in most of the time.

These plutonium arrays, however, were anything but room temperature. Plutonium spontaneously emits a considerable amount of radiation - a wide variety of nuclear particles at a variety of energies. When these particles lose any or all their energy in the fissile material, that energy appears as heat. The larger the mass, the more heat is generated, and plastic-wrapped metal pieces do not allow the easy escape paths for heat found in powders and other loose forms. Even the 3 kg plutonium metal cylinders of some earlier reports in this series were generally regarded as “hot to the touch”. They were uncomfortable to be held in the hands for long periods of time.

Older plutonium worsened the problem still further because this material inbreeds Americium as a result of the normal nuclear decay of plutonium. Americium is a much greater source of radiation than even plutonium, and this element’s radiation would also contribute heat to the metal piece. This probably was not a problem as explained in the next paragraph.

Americium was probably a lesser problem because these castings were most likely recently cleansed of Americium as part of normal production operations. Wrapping them in plastic reduced convection cooling and increased the temperature. Placing them in a metal can further contained the heat, although it also provided some cooling fins for radiating heat. Placing two or more loaded film cans together worsened the problem as each tended to heat the other.
Ingots in the larger arrays became very hot. Arrays were massive. The lead experimenter described larger arrays as “extremely hot”! The heat was so great, in fact, that film cans in the middle of the array could not easily be handled even with thick workmen’s gloves. Thermocouples placed near the center of larger arrays indicated temperatures which caused workers to worry about melting the plastic wrapping used for contamination control. Recorded temperatures noted during construction of the $3 \times 3 \times N$ array were $158^\circ\text{F}$ after 72 ingots had been stacked and $160^\circ\text{F}$ after 81 ingots.

Experimenters did not remain close to the array for two reasons. Heat was only one problem. The radiation flux from that much plutonium - increased by the multiplication factor of the assembly - was not considered safe to be around for too long a period of time. Data suggests that thermal neutron fluxes approaching one-and-a-quarter million neutrons per minute were being recorded.

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Workers sometimes approached about the same distance from an array as the thermal neutron radiation detectors were. These detectors were about 50 mm by about 250 mm in cross section, and they may be thought of as being about 25% efficient. Those detectors would attain count rates close to their saturation point of about a million neutrons a minute for arrays near the multiplication limit. A simple estimate reveals that workers may have been exposed to a considerable radiation dose during this program.
ENVIRONMENT

The environmental conditions for the horizontal drum arrays containing plutonium metal ingots, the 1968 program, are described in RFP-1242. This report describes the apparatus used and the rooms involved. These experiments, however, were performed in Building 991 (then, just Building 91) at Rocky Flats - not in the Critical Mass Laboratory. Therefore, further details about the place experiments were carried out, such as their room number and detailed construction, has been lost these three decades later. No additional detail can be added by this author.

Experiments involving vertical stacks of film cans - but no drums - took place at the Rocky Flats plant site in Building 886, then just called Building 86. This building contained the Critical Mass Laboratory and was the building in which all previous experiments in this series were performed. The specific room in which these experiments were performed was Room #102. It was the fissile metal storage room and was located only a few meters from the Assembly Room (Room #101) in which all critical experiments spanning 25 years had been performed at Rocky Flats. The room was also called the “Vault Room” because of the high control over personnel access.

Room #102 measured 6.71 m east/west by 4.72 m north/south. The lowest point of the wall was 3.81 m high and the roof had a gentle slope up from that. The ceiling was a thin sheet steel corrugated deck covered outside with a composite roofing material. Because of its thinness, no further description seems necessary. The south wall was 1.52 m thick and composed of strongly reinforced concrete because the other side was the Assembly Room, itself. The north wall was 0.4-m thick (also concrete) to provide additional radiation shielding for personnel operating the critical experiment assembly controls for other programs. The interior west wall was also reinforced concrete; it, too, was 0.4 m thick. The exterior east wall was only about 0.2-m thick and was constructed of light-weight, expanded concrete block and mortar. Holes in these blocks were aligned such that steel rebar could be passed vertically through them to strengthen this load bearing wall. All holes were also back filled with mortar, so the wall was essentially
solid. The construction resulted in a wall very similar in composition to other reinforced concrete walls.

That exterior wall, at the time of these experiments, also contained a pair of light-weight, steel, exterior doors. These doors were intended for the routine receipt and shipments of plutonium and enriched uranium in metal, solution, and other forms as needed by the laboratory for experimental programs. The interior west wall contained a similar light-weight door.

Years later, this room was upgraded to enhance the safe and secure storage of fissionable materials. This upgrade took place in two phases. The first upgrade was simple. Both exterior doors were removed, and a solid wall built in its place. This probably happened about 1970. Several years later, the room was about doubled in size by expanding it eastward, increasing exterior walls to the same 0.4 m thickness, integrating a thick cast concrete ceiling into the walls, and installing a bank type vault door. These experiments were certainly performed before the second upgrade. Whether performed before or just after the first is not recalled for certain. Since the experiments did not take place close to the east wall or its doors, the answer to this question is probably not important.

Experiments were performed adjacent to the thick (1.52 m) south wall of the room. The exact location along this wall is not known, but it probably would have been somewhere near the center because experimenters needed to move around the array area and to store the loaded film cans in an approved, critically safe, manner.

The actual vertical stacks of plutonium-filled film cans were not built directly upon the floor of the room. One hand written comment on a loose leaf data page describes the experiment as being “reflected by the floor and one concrete wall”, but the floor referred to there was a “false floor”. The design of the subcritical reciprocal approach experiments called for a pair of radiation detectors be placed under the array and roughly centered. This false floor accommodated these detectors. It was composed of high-density cast concrete blocks laid on one edge and touching
one another except for a narrow channel for the detectors. The concrete blocks were each approximately 102 mm thick by 204 mm high. The length of each block was either 305 mm or 408 mm; that detail is not recalled for certain. The false floor, however, was solid except for the detector channel, so that individual length is not important.

The size of this false floor apparently varied a little for some of the array patterns. One loose leaf page describes the floor for the $3 \times 3 \times N$ array as being 45" by 48" (about 1.14 m by 1.22 m). That drawing suggests that perimeter cans would have hung over the sides of the false floor. If the film cans were 19" in diameter as believed, the overhang would have been about 6" (150 mm) on the shorter side and 4.5" (114 mm) on the longer. A similar drawing for the smaller $2 \times 2 \times N$ array showed a similar - but larger - overhang. Scaling from these drawings reveals that the false floor for the smaller square array may have been about 28" by 32" (0.71 m by 0.81). Scaling such pencil drawings is always a risky thing to do, but it is the best evidence available. No such drawings were found for the $3 \times N$ or the $1 \times N$ arrays, but the reciprocal multiplication data page shows a hand-drawn elevation view that includes the false floor. That false floor is sketched too large for either array. It is not known if this false floor is the same as for the $2 \times 2 \times N$ array or a little smaller.

The exact distance from the array to the thick south wall is not known. Drawings and recollections imply that the near face of the array was “built up against the wall”. Whether a space of a few millimeters existed or the cans literally touched the wall is not known.

No other equipment is known at this time to have existed in that room at the time of these experiments.
RESULTS

Experimental results for the horizontal drum arrays containing plutonium metal ingots, the 1968 program, are described in RFP-1242. This report describes these results as completely as possible. No additional detail can be added by this author. The reader is referred to that document which is appended to this report for those results.

Four relatively compact vertical arrays of massive slabs of plutonium metal were assembled in an effort to determine the height of each array where criticality might be attained. Each of the four was characterized by the arrangement of plutonium in a typical layer. The metal was contained in steel cans only a little larger than the diameter of the rectangular ingots of plutonium. These approaches toward criticality were manually assembled, so experiments were restricted to remain well subcritical. The critical parameter, if any, would have to be extrapolated from the subcritical assembly data.

The four arrays were characterized by the configuration of each layer. The simplest was merely a vertical stack of loaded film cans. The second was a tightly-packed triangular array of three cans per layer. The third was a square array of four cans touching one another. The last was the largest, each layer was a tightly packed $3 \times 3$ square array of film cans. Each layer of this last array would contain between 70 and 80 kg of plutonium metal.

Results were quite surprising. Three of the four configurations were assembled past the point necessary to state confidently that the array would have remained well subcritical even if it had been constructed infinitely high. The fourth is the only one where the possible criticality of a taller stack remains in question. It, too, would probably have been subcritical at an infinite height; but that statement cannot be made with great confidence.
The untreated data for the four reciprocal multiplication approaches are presented in Tables I to IV for the four configuration patterns studied. Untreated data is presented because any attempt to adjust this data for other experimental considerations is quite arbitrary and subject to criticism. Graphs of these unmodified data with the reciprocal multiplication along the vertical axis and three different parameters of the growing array plotted horizontally are presented in Figures 2 to 5. Neutron counts for both $C_o$ and $C(N)$ are assumed to have been collected over one minute.\(^{18}\)

Each configuration will be discussed separately below, although the first table will be described in greater detail because much of the discussion applied to it also applies to the other tables. Two points are found at each reciprocal multiplication. One (•) corresponds to numbered parameters such as the number of layers which equals the number of ingots in the first case only; the other (x) corresponds to the plutonium mass in the array along the upper axis.

In Table I only, the number of layers equals the number of ingots because it is a 1 x N array. Likewise, the first two columns of masses are the same for the same reason; the third gives the total mass of plutonium in the array. The initial condition to start the reciprocal multiplication curve, $C_o$, was arbitrarily selected to be that for two loaded film cans stacked one upon the other. Two radiation detectors were always used, and the observed neutron counts for both, #1 and #2, over the same interval of time is recorded in adjacent columns. The next two columns present the corresponding reciprocal multiplications, calculated simply by dividing the just-counted flux

\(^{18}\) The interval used to count neutrons is believed to be 60 seconds; but that fact is not documented anywhere. It is assumed because it is a “natural” unit of time and because it was a common interval used at the Rocky Flats laboratory for such data collections. Furthermore, with such high fluxes, counting longer than one minute would have seemed to unnecessarily expose workers to excessive radiation. On the other hand, the same high total counts again suggest that they would not have been collected in a very much shorter period of time - say, 10 seconds. This is so, because the radiation flux within the room between counting intervals (graphing data, discussing the next safe increment, etc.) would have been too high to allow personnel to remain in the room for long periods. The only other reasonable assumption would be that this counting period was 30 seconds.
Table I. Neutron Count Rates for the 1 x N Array of Plutonium Ingots As Measured on March 12, 1969 Using the Reciprocal Multiplication Technique.

<table>
<thead>
<tr>
<th>Number of Layers</th>
<th>Number of Ingots</th>
<th>Mass of Plutonium Ingots in Layer [individual(^a)] (Kg)</th>
<th>Mass of Plutonium Ingots in Layer [total] (Kg)</th>
<th>Total Plutonium Mass (Kg)</th>
<th>Neutron Count Rates(^{ab}) #1</th>
<th>Neutron Count Rates(^{ab}) #2</th>
<th>Reciprocal Multiplication #1</th>
<th>Reciprocal Multiplication #2</th>
<th>Source Corrected Reciprocal Multiplication (counter #1) 1969</th>
<th>Source Corrected Reciprocal Multiplication (counter #1) 1998</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>8.360</td>
<td>8.360</td>
<td>8.360</td>
<td>7630</td>
<td>6925</td>
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<td>1.0</td>
<td>1.0</td>
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<tr>
<td>2</td>
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<td>9.827</td>
<td>9.827</td>
<td>18.187</td>
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<td>10.219</td>
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<td>93318</td>
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<td>102101</td>
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<td>0.186</td>
<td>0.236</td>
<td>0.362</td>
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<td>10</td>
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<td>7.688</td>
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<td>0.195</td>
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<tr>
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<td>10.061</td>
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<td>9.902</td>
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<td>0.153</td>
<td>0.144</td>
<td>lost</td>
<td>lost</td>
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</table>

\(^a\) Neutrons counted per minute.

\(^b\) Initial count rate taken at 2 layer of cans: \(C_0(#1) = 23,548\) and \(C_0(#2) = 20,480\) neutrons per minute.
Table II. Neutron Count Rates for the 3 x N Array of Plutonium Ingots As Measured on March 17, 1969 Using the Reciprocal Multiplication Technique.

<table>
<thead>
<tr>
<th>Number of Layers</th>
<th>Mass of Plutonium Ingots in Layer</th>
<th>Total Plutonium Mass</th>
<th>Neutron Count Rates(^a)</th>
<th>Reciprocal Multiplication</th>
<th>Source Corrected Reciprocal Multiplication (counter #1)</th>
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<tr>
<td></td>
<td>[individual(^b)] (g)</td>
<td>[total] (g)</td>
<td>#1</td>
<td>#2</td>
<td>1969</td>
</tr>
<tr>
<td>1</td>
<td>3</td>
<td>8764.5728,8330</td>
<td>22822</td>
<td>54895,53993</td>
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<tr>
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<td>6</td>
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<tr>
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<td>9</td>
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</table>

\(^a\) Neutrons counted per minute.

\(^b\) Initial count rate taken at 1 layer of cans: \(C_0(#1) = 54,895\) and \(C_0(#2) = 53,993\) neutrons per minute.
Table III. Neutron Count Rates for the $2 \times 2 \times N$ Array of Plutonium Ingots As Measured on March 17, 1969 Using the Reciprocal Multiplication Technique.

<table>
<thead>
<tr>
<th>Number of Layers</th>
<th>Ingots</th>
<th>Mass of Plutonium Ingots in Layer [individual] (g)</th>
<th>Mass of Plutonium Ingots in Layer [total] (g)</th>
<th>Total Plutonium Mass (g)</th>
<th>Neutron Count Rates$^a$ #1</th>
<th>Neutron Count Rates$^a$ #2</th>
<th>Reciprocal Multiplication #1</th>
<th>Reciprocal Multiplication #2</th>
<th>Source Corrected Reciprocal Multiplication (counter #1)</th>
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</thead>
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<td>4</td>
<td>8357.5728,8764,8330</td>
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<td>31179</td>
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<td>1.0</td>
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<td>139370</td>
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<td>1026606</td>
<td>0.073</td>
<td>0.093</td>
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<tr>
<td>11</td>
<td>44</td>
<td>8469.7166,9089,10077</td>
<td>34801</td>
<td>377295</td>
<td>1124468</td>
<td>0.067</td>
<td>1157187</td>
<td>0.064</td>
<td>0.081</td>
</tr>
<tr>
<td>12</td>
<td>48</td>
<td>7592.9559,9666,8118</td>
<td>34935</td>
<td>412230</td>
<td>1255856</td>
<td>0.060</td>
<td>1293361</td>
<td>0.058</td>
<td>0.072</td>
</tr>
<tr>
<td>13</td>
<td>52</td>
<td>9957.8078,7904,9958</td>
<td>35897</td>
<td>448127</td>
<td>1401767</td>
<td>0.053</td>
<td>1444217</td>
<td>0.052</td>
<td>0.063</td>
</tr>
</tbody>
</table>

$^a$ Neutrons counted per minute.

$^b$ Initial count rate taken at 1 layer of cans: $C_o(#1) = 74,994$ and $C_o(#2) = 74,494$ neutrons per minute.
Table IV. Neutron Count Rates for the 3 x 3 x N Array of Plutonium Ingots As Measured on March 17, 1969 Using the Reciprocal Multiplication Technique.

<table>
<thead>
<tr>
<th>Number of Layers</th>
<th>Number of Plutonium Ingots in Layer</th>
<th>Mass of Plutonium Ingots in Layer (g)</th>
<th>Total Plutonium Mass (g)</th>
<th>Neutron Count Rates (^{ab})</th>
<th>Reciprocal Multiplication</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>[individual]</td>
<td>[total]</td>
<td>#1</td>
<td>#2</td>
</tr>
<tr>
<td>1</td>
<td>9</td>
<td>9829,9901,9902,9559,10022,9707,10027,9887</td>
<td>88083</td>
<td>65602</td>
<td>62641</td>
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<tr>
<td>2</td>
<td>18</td>
<td>10201,8197,7904,9666,7578,7132,9558,8357,8330</td>
<td>77628</td>
<td>131697</td>
<td>125493</td>
</tr>
<tr>
<td>3</td>
<td>23</td>
<td>8469,8372,8611,8404,8360</td>
<td>42216</td>
<td>185889</td>
<td>180025</td>
</tr>
<tr>
<td>4</td>
<td>36</td>
<td>7668,8500,7595</td>
<td>23783</td>
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<td>318607</td>
</tr>
<tr>
<td>5</td>
<td>45</td>
<td>8133,10219,10068</td>
<td>28420</td>
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<td>376760</td>
</tr>
<tr>
<td>6</td>
<td>51</td>
<td>8250,7911,7850</td>
<td>24111</td>
<td>577796</td>
<td>577796</td>
</tr>
<tr>
<td>7</td>
<td>60</td>
<td>7166,10313,8129</td>
<td>25608</td>
<td>720178</td>
<td>715156</td>
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<td>9</td>
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<td>24911</td>
<td>839797</td>
<td>833535</td>
</tr>
<tr>
<td>10</td>
<td>90</td>
<td>8000,8283,8118</td>
<td>24401</td>
<td>934000</td>
<td>896400</td>
</tr>
</tbody>
</table>

\(^{a}\) Neutrons counted per minute.

\(^{b}\) Initial count rate taken at 2 layer of cans: \(C_0(#1) = 131,697\) and \(C_0(#2) = 125,493\) neutrons per minute.
Figure 2. The reciprocal multiplication curve without source neutron correction for the 1 x N array of plutonium ingots suggests that an infinitely tall assembly would remain subcritical with an asymptotic multiplication of about 7.
Figure 3. The reciprocal multiplication curve without source neutron correction for the 3 x N array of plutonium ingots suggests that an infinitely tall assembly would remain subcritical with an asymptotic multiplication of about 12.
Figure 4. The reciprocal multiplication curve without source neutron correction for the $2 \times 2 \times N$ array of plutonium ingots suggests that an infinitely tall assembly would remain subcritical with an asymptotic multiplication of about 20.
Figure 5. The reciprocal multiplication curve without source neutron correction for the $3 \times 3 \times N$ array of plutonium ingots suggests that an infinitely tall assembly would probably remain subcritical, although the asymptotic multiplication of about 10 shown here may be erroneously high due to dead time losses.
level\textsuperscript{19} into the initial count, \(C_0\). These data for this single stack of film cans are displayed graphically in Fig. 2. Although the curve does not appear to have quite attained its lowest asymptotic reciprocal multiplication, an infinitely high stack of similar film cans would appear to achieve a multiplication of about 6.7. This vertical stack, then, would be well subcritical under any stretch of the imagination.

In Table II, the number of ingots is three times the number of layers because it is a 3 x N array. A wide column of ingot masses simply lists the three masses in each layer, but no information exists as to which of the three positions contained which ingot. The next column gives the total mass in the layer, and the last mass column gives the total plutonium mass in the array. This time, \(C_0\) was arbitrarily selected to correspond to just the bottom full layer of three loaded film cans. Two radiation detectors were always used, and the actual neutron count observed for the same interval of time for both detectors is recorded as a function of the number of layers. The next two columns present the corresponding reciprocal multiplications, calculated simply by dividing the just-counted\textsuperscript{20} flux level into the initial \(C_0\). These data are displayed graphically in Fig. 3. Two data points are again found at each reciprocal multiplication. Again, one (*) corresponds to numbered parameters such as the number of layers, the upper of the two axes along the bottom, and the number of ingots (bottom axis); the other (x) corresponds to the total mass along the upper axis. This curve appears to be further from the asymptotic reciprocal multiplication value than the 1 x N array, but it still appears to predict subcriticality regardless of height. An infinitely high stack of triangularly pitched film cans would produce a multiplication of, perhaps, 12 to 15.

\textsuperscript{19} Neutron flux levels in excess of 150,000 neutrons per minute were found about three times the distance from the center of the completed array as the experimenters might approach the array. Assuming the detectors are about 25% efficient and 5% the size of an experimenter's body suggests that the one placing the cans on the stack might be exposed to up to 15 million neutrons. No record is made as to what, if any, protective clothing was worn.

\textsuperscript{20} The neutron flux levels at comparable multiplications (about 6.5) was about 370,000 counts per minute. This is about 2.5 times the flux encountered at the final 1 x 1 x 15 array. The flux level at twice the number of cans (30) was about 3.3 times as great.
In Table III, the number of ingots is four times the number of layers because it is a square $2 \times 2 \times N$ array. The wide column of ingot masses, again, simply lists the four masses in each layer, but no information exists as to which position contained which ingot. The next column gives the total mass in the layer, and the last column of masses gives the total plutonium mass in the whole array. The initial count rate to start the reciprocal multiplication curve, $C_o$, was arbitrarily selected to be that for the first full layer of four loaded film cans. Two radiation detectors were always used, and the actual neutron count for the same standard interval of time is recorded for both detectors as a function of the number of layers in adjacent columns. The next two columns present the corresponding reciprocal multiplications, calculated simply by dividing the just-counted flux level\textsuperscript{21} into the initial $C_o$. These data are displayed graphically in Fig. 4. As before, two data points are found at each reciprocal multiplication with similar meanings to the (⋆) and the (x). As in the case of the $1 \times 3$ array, this array does not appear to have achieved the asymptotic reciprocal multiplication. The multiplication for an infinitely high stack of $2 \times 2$ loaded film cans appears to be between 50 and, say, 75. Even this array would clearly remain subcritical regardless of height.

In Table IV, the number of ingots is *nine* times the number of layers because it is a square $3 \times 3 \times N$ array. Each layer spans almost 1.5 m on a side. The wide column of masses, again, simply lists the ingot masses in each layer, but no information exists as to which position contained which ingot. Reciprocal multiplication data was taken at some partially filled layers, so the nine masses of a given layer are spread over two or three lines in the table in some cases. The second mass column gives the total mass in the layer or partial layer, but the last column of masses gives the total plutonium mass in the whole array. The initial count rate to start the reciprocal multiplication curve, $C_o$, was arbitrarily selected to be that for the first *two* full layers of *nine* loaded film cans. Why 18 film cans were selected for this starting point is not known these three decades later. Two radiation detectors were always used, and the actual neutron count for the same standard interval of time is recorded for both detectors as a function of the number of

\textsuperscript{21} The maximum flux levels encountered were in excess of 1.4 million neutrons per second. The same number of ingots (36) in the square array produced only about 50% more neutrons than in the triangular array.

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layers or partial layers in adjacent columns. The next two columns present the corresponding reciprocal multiplications, calculated simply by dividing the just-counted flux level\textsuperscript{22} into the initial $C_0$. These data are also displayed graphically in Fig. 5. As before, two data points, (? ) and (x), are found at each reciprocal multiplication with similar meanings as before.

Unlike the previous three array patterns, the eventual condition of a much taller assembly does not appear to be well determined at the point the experiment ended. Reasonable arguments could be made that this array would eventually attain criticality or that it, too, would become asymptotic at some subcritical level. This experiment was terminated because the construction of the 10\textsuperscript{th} layer actually produced a \textit{decrease} in the observed neutron flux. The decrease was not immediately explicable; so the experiment was terminated at that point as prescribed by laboratory policies\textsuperscript{23}. One possible explanation - suggested three decades later - has to do with the very high count rates observed. The 9\textsuperscript{th} layer registered about 1.3 million neutrons per minute, and this very high count rate may have introduced some dead time losses into the neutron detection apparatus. Then, the addition of plutonium onto the 10\textsuperscript{th} layer may actually have begun to "paralyze" the detectors. If so, the count rate would, in fact, decrease as observed. If such losses did occur, then the observed reciprocal multiplication for the 9\textsuperscript{th} layer (and, quite possibly the 8\textsuperscript{th}) would have been lower than shown. This argument, then, \textit{increases} the possibility that this largest array \textit{might} have eventually attained criticality had it been constructed further. The very lowest that criticality might have occurred would appear to be somewhere around the 12\textsuperscript{th} to 16\textsuperscript{th} layer, but this guess is the result of a very lengthy extrapolation.

A careful examination to the reciprocal multiplication values obtained for partially-filled layers of the $3 \times 3 \times N$ array shows that data points appear to fall reasonably close to the curve generated by integral layers only. This same feature was also observed during the construction of

\textsuperscript{22} The maximum flux levels encountered were only slightly in excess of 1.2 million neutrons per second. This is surprisingly smaller than the count rate for the $2 \times 2 \times N$ array. This point will be discussed more later.

\textsuperscript{23} Any experiment was required to be terminated if any aspect of it was not fully understood by the experimenters.
the 3 x N triangular array, although there, an electronic meter\textsuperscript{24}, used for an instantaneous readout of the approximate reciprocal multiplication, revealed that partially filled layers yielded data falling on the same curve formed with only full layers. This evidence is shown in Fig. 6. This phenomenon suggests that this functional relationship was much more dependent on the mass of plutonium metal in the array than on the exact location of the metal within a given layer. Ingot masses varied by about a factor of two, but even that shows up only as small departures from an otherwise smooth curve in Fig. 7. This apparent insensitivity to position within a layer and even to fairly large variations in mass would seem to imply that some kind of correction for added source neutrons might be possible.

\textit{Source Neutron Correction}

Each plutonium ingot added beyond the initial "source count configuration" introduced both true reactivity as well as additional source neutrons. All "source" neutrons came from spontaneous fission of the plutonium\textsuperscript{25}. The curves of Figs. 2 to 5 make no effort to correct for these extra source neutrons, they are wrongly excluded from the numerator and wrongly included in the denominator of the reciprocal multiplication expression \( C_0/C \). Indeed, no correction is immediately obvious and any is subject to question. One attempt to evaluate that correction was made empirically by the lead experimenter almost 30 years ago. This author offers another model using the same empirical data. The two yield very different results.

The 1969 measurements simply "mapped" count rates for just a single layer of loaded film cans as a function of height. One layer of plutonium-loaded film cans were separated from the bottom layer by 0, 1, 2, 3, or more layers of empty film cans. These data yielded an estimate of

\textsuperscript{24}This electronic information is not quoted elsewhere because of its low precision.

\textsuperscript{25}In contrast, a similar experiment involving uranium would have had the initial source neutrons introduced via an external source. The only difference between the two fissile metals is that source neutron are intrinsic in one and added externally in the other. The external source is fixed in its emission rate; but the plutonium case is more complicated. Polonium-beryllium sources were used until the advent of the element Californium.

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Figure 6. The reciprocal multiplication curve for the $3 \times N$ array of plutonium ingots shows that data for partially filled layers fall on the same curve as that generated by only completely filled layers. The same was noted for the $3 \times 3 \times N$ square array.
Figure 7. Additional source neutrons from plutonium ingots above the layer(s) selected for the $C_0$ count rate are to be accounted for when calculating a corrected reciprocal multiplication curve. The actual extra source neutrons for each of the four layer configuration patterns are shown by dots. The dashed curves merely connect dots and represent no theoretical consideration. The scale for three of the four is to the left; that for the $1 \times N$ array is to the right. The theoretical shape of this inverse linear decrease is shown by the solid curve; the scale is arbitrary.
the source count from the N\textsuperscript{th} layer. This procedure was repeated for all four layer configuration patterns. The assumptions here are that no single layer contributed any significant reactivity so that all neutrons from any one layer may be considered source neutrons. Another implicit assumption is that the initial configuration, \( C_o \), of lowest one or two layers never contributes reactivity. The validity of this assumption is not certain, especially when 18 film cans (2 layers) was used for the 3 x 3 x N array.

Evidently, the single-layer study was made under a significantly different environment than the critical approach experiments. The two were obtained several days apart, and the neutron fluxes differed markedly. The background count rate for the single-layer study ranged from 40,000 to 52,000 neutrons per minute, whereas count rates for the critical approach experiments had as few as 7600 neutrons per minute. This could only be so if large numbers of ingots had been in the vicinity of the single-layer study but not for the critical approaches. That is assumed to be the case.

The 1 x N array had one loaded film can raised by placing up to 13 empty film cans under it, one at a time. The mass of this one ingot was probably 10175 g although that information had to indirectly inferred. In like manner, one layer of three loaded film cans (10175, 9666, and 6776 g) in the triangular pitched array were raised by up to 11 layers of empty cans. Four loaded film cans in the 2 x 2 pattern (10175, 9666, 6776, and 9703 g) were raised by up to 9 empty layers; and, finally, nine filled cans in the largest pattern (10206, 5552, 10077, 9666, 7386, 10084, 7904, 8557, and 9958 g\textsuperscript{26}) were raised by up to 4 empty layers. Count rates for one of the two neutron detectors are presented in Table V and illustrated in Fig 6 for all four layer patterns.

The lead experimenter in 1969 chose to correct the reciprocal multiplication data by reducing the observed count rate for each layer after the initial one or two by the wrongly

\textsuperscript{26} Some confusion exists here. A different set of nine ingots are recorded on the same data sheet dated March 21, 1969: 10175, 8197, 6776, 10188, 7132, 9703, 5762, 9884, and 8060 g. The first set are assumed because of the placement on the page of the list of nine masses. No explanation is offered for the second set of masses.
Table V. Additional Source Neutron Count Rates for the Four Layer Configuration Patterns Studied

<table>
<thead>
<tr>
<th>one Layer of Plutonium Ingots in Nth Layer</th>
<th>Neutron Count Rates* with one Layer of Plutonium Ingots in Nth Layer After Subtracting Background</th>
</tr>
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<tr>
<td></td>
<td>1 x N Array</td>
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<tr>
<td>1</td>
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<tr>
<td>2</td>
<td>5589</td>
</tr>
<tr>
<td>3</td>
<td>4462</td>
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<td>3147</td>
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<td>6</td>
<td>2652</td>
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<tr>
<td>7</td>
<td>1977</td>
</tr>
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<td>8</td>
<td>1890</td>
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<td>2 x 2 x N Array</td>
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</tr>
<tr>
<td>5</td>
<td>44547</td>
</tr>
<tr>
<td>a</td>
<td>Neutron per minute</td>
</tr>
</tbody>
</table>

included "new" source neutrons from higher layers. Mathematically, this early model may be expressed as:

\[
\frac{C_0(l)}{C_{obs}(N) - C_0(N)}
\]

where:

C_0(l) is the source neutron count for the reciprocal multiplication approach for l layers, either 1 or 2,

C_0(N) is the sum of all source neutrons from all layers above l as obtained through the ancillary study involving one layer at a time, and
C_{obs}(N) is the actual neutron count obtained for the reciprocal multiplication approach at the N^{th} layer.

The author's model views the same empirical data differently. For any array containing N layers, the total number of source neutrons would be the sum of such source neutrons from each and every one of the N layers but taken only one layer at a time, as though each, alone, contributed source neutrons. The plutonium itself was believed to contribute essentially no reactivity (although the validity of that assumption is unclear for the largest array containing 18 ingots). This ever-increasing numerator is divided by the observed neutron count for N layers, C_{obs}(N). Mathematically, this newer approach may be expressed as:

\[
\frac{C_0(\ell) + C_0(N)}{C_{obs}(N)}
\]

The same definition of terms is used here as above. Obviously, the two expressions must yield different results. Both contain the same three terms, but one of them appears in the numerator in one model and the denominator in the other.

Two other ways of viewing the effect of source neutrons can be derived from first principals. These are expressed verbally and then described mathematically.

(1) **The multiplication of the array at the N^{th} layer equals the observed count rate at that layer less the multiplication of the system at that layer times the sum of contributions of "new" source neutrons for the 2^{nd} through N^{th} layers divided by the initial count rate, C_0.**

\[
M(N) = \left\{ C_N(\text{obs}) - M \sum_{k=1}^{N} "C_0(n)" \right\} / C_0(\text{initial}).
\]

Solving this equation for M(N) and then forming the reciprocal yields:

\[
1/M(N) = \sum_{n=1}^{N} C_0(n) / C_N(\text{obs})
\]
(2) The reciprocal multiplication of the array at the \( N^{th} \) layer equals the count rate for the \( N^{th} \) layer that would have existed had just one layer (only) of plutonium been positioned at each layer and these \( N \) contributions summed over the \( N \) layers before dividing that by the observed count rate. Mathematically:

\[
1/M(N) = \left( C_0(\text{nat}) + \sum_{t=1}^{N} "C_0(t)" \right) / C_N(\text{obs}) = \sum_{i=1}^{N} C_0(i) / C_N(\text{obs}).
\]

Both interpretations lead to the author's formulation, so this is thought to be the better one. Still, even this expression is not without question. The proper treatment of source neutrons is far from trivial. Because this complex matter is so subject to interpretation, all available data is presented in this paper. This enables others to draw their own conclusions free of any bias. Both “corrected” reciprocal multiplication values discussed above are presented in italics font in the far right columns of Tables I to IV.

This high background flux during the single-layer study, which was absent in the earlier measurements, complicated the determination of the term \( C_0(N) \) used in both of the above expressions. The following terms are defined for this calculation:

- \( B \) = room background count;
- \( K(N^{th}) \) = count for plutonium ingots only in the \( N^{th} \) layer (no other plutonium in array);
- \( K(N^{th}) - B \) = source neutron contribution due to plutonium only in the \( N^{th} \) layer;
- \( l = 1 \) or \( 2 \) = number of layers arbitrarily selected to produce \( C_0 \); and,

\[ C_0(N) = \sum_{t=1}^{N} [K(t^{th}) - B] = \text{total source contribution due to plutonium in layers } l+1 \text{ to } N. \]

Figure 7 also shows the source correction for each term of the equation immediately above. The total contribution of source neutrons for an array \( N \) layers high would be the sum over the first \( N \) layers as expressed in the equation.
The decrease in source neutrons per layer may also be estimated theoretically as a function of level number. A similar theoretical calculation could have been made per ingot or per kilogram of plutonium metal, but this was not done for this paper. This calculation is left to the interested party. In this theoretical model, plutonium ingots are imagined to exist in one layer at a time with layers of empty film cans below the loaded one. A drawing of this assumption is shown in Fig. 8 (not to scale) where the distance between counters and the lowest layer of film cans is D; and the vertical distance between successive layers is the thickness of the film cans, T. Each layer would then contribute source neutrons represented by each term of the following equation:

\[ C_0(\text{corrected}) = C_0 \left[ 1 + \left(1 + \frac{T}{D}\right)^{-n} + \left(1 + \frac{2T}{D}\right)^{-n} + \left(1 + \frac{3T}{D}\right)^{-n} + \ldots + \left(1 + \frac{NT}{D}\right)^{-n} \right] \]

or

\[ C_0(\text{corrected}) = \sum_{1}^{N} \left(1 + \frac{NT}{D}\right)^{-n} \]

where \( n = 1 \) or 2 depending on the geometry assumed.

A linear decrease, a “1/R” dependence, would seem more appropriate if the geometry of the counters and the film cans resembled two planes separated by increasingly greater distances. Here, \( n=1 \) in the above equation. If the geometry were more “spherical”, an inverse quadratic dependence, “1/R^2”, would better describe the physics. Both were calculated and the former seemed to fit the shape of the observed data quite well.

The thickness of the film can is known to be \( 2^{3/8} \) inches (about 60.3 mm), but the parameter D is nowhere specified except that it obviously must be thicker than the concrete blocks used to create the false floor. Assuming a T to D ratio of 0.15 (a pure guess at a
Figure 8. Additional source neutrons from plutonium ingot layers above those layer(s) selected for the $C_0$ count rate (the bottom one or two) were measured by placing empty film cans above one loaded layer. This procedure was repeated for all four layer configuration patterns although only one is represented in this not-to-scale elevation view. Neutron detectors were located under the array beneath a false floor; the distance $D$ was never recorded.
reasonable D), the linear form of this theory appears to match the shape of the empirical data quite well. The quadratic form did not. The theoretical prediction for the linear assumption is also presented in Fig. 7 as a solid curve. The absolute value of this curve is relative, and its scale factor is shown to the right.

This theoretical model, too, is subject to argument. The only valuable observation is that the general shape of the functional dependence seems to match empirical curves quite well.

All this discussion about source neutron corrections is probably superfluous. The fact remains that all four reciprocal multiplication curves for different layer configurations probably still predict subcriticality. In fact, the inclusion of source neutron effects under any of the assumptions diminishes the confidence with which any conclusion concerning the eventual criticality of a very tall array can be drawn. The character of the reciprocal multiplication curves are less well developed. Ending values are less reactive, and the ultimate asymptotic values of this parameter appears to be correspondingly uncertain.

No experimental results from the other program wherein plutonium metal ingots were placed within film cans which were, in turn, placed within large drums are presented in this paper. They are presented in the Appendix to this document. That Appendix is a direct copy of the earlier report written on the study in December of 1968. This author can add no further information to the data published there.
UNCERTAINTIES

This experimental program is fraught with uncertainties. In addition, it contains some parameters whose value is altogether unknown. In spite of these experimental limitations, the study has value to the benchmark evaluation project because of the very large mass of plutonium metal that was confined to a relatively small space and still remained well subcritical. It forms a qualitative limiting case.

The ingots themselves varied in mass by about a factor of two from the lightest to the heaviest. Even this weakness did not seem to present problems. The reciprocal multiplication curves seemed to predict the about the same final condition regardless of whether graphed against the mass of the array or simply the number of ingots in it. The latter is independent of mass. Although masses in each layer were recorded, no connection is possible between which ingot appeared in which position in the layer. Again, experimental evidence suggests that this information, too, is not really all that important. Dimensions of the ingots, too, was uncertain within about three or four different ingot mold possibilities in use at Rocky Flats. Although only one mold thickness was used at the plant, some question exists as to the thickness quoted throughout this paper. Finally, the orientation of an ingot within its container was not known nor was the amount of plastic wrapping material recorded.

The size of the film can container for the plutonium is believed to be correct in the body of this report, but some troubling evidence hints at a somewhat different size. Its composition is unknown except that it was “steel”. The exact size and precise composition of the false floor under the arrays is equally vague.

All these shortcomings would render these results useless to the benchmark criticality safety project if the experiments reported critical configurations. Neither the geometry nor the composition of the arrays under consideration could be specified with any degree of confidence.
These results are not useless. Their value lies in the clear subcriticality of four different arrays containing many hundreds of kilograms of plutonium metal in ingot form.
ACKNOWLEDGMENTS

This paper is written under the auspices of the International Criticality Safety Benchmark Evaluation Project, funded by the United States Department of Energy. The project is administered by J. Blair Briggs of Lockheed Idaho Technologies Company. This author is grateful to Mr. Briggs for the opportunity to publish these data in the peer-reviewed literature before the author's full retirement. This is the sixth of seven papers presenting previously unpublished or inadequately documented results from criticality experiments performed at the Rocky Flats Critical Mass Laboratory. These seven documents will mark a satisfying closure to this author's long career in the arena of critical experiments.

Special recognition is due Mr. Donald R. Ferguson, the Lead Experimenter as he is referred to frequently throughout this paper. He conceived these two experimental programs, convinced the necessary management of their merit, designed them, arranged for the simultaneous collection of a significant fraction of the world's supply of plutonium metal at that time, performed the experiments with the assistance of a few colleagues, analyzed the data for nuclear safety purposes at Rocky Flats, and authored one internal report (see Appendix). Mr. Ferguson still lives in the Denver area and willingly contributed all loose leaf documents as well as his recollections to this author's efforts to report Ferguson's work. This contribution was so vital that his name also is included on the title page of this paper. Although retired, Mr. Ferguson continues to serve the nuclear industry as a consultant on nuclear safety matters at Rocky Flats.

Several colleagues participated in at least one experiment in one way or another. These persons were all associated with the nuclear safety program at Rocky Flats in those years. Participants included: Merlyn L. Boss, Harold E. Clark, Douglas C. Hunt, Howard W. King, Douglas E. Payne, Warren R. Sheets, and the laboratory's director, Clarence Lee Schuske. None of these were "Certified Experimenters"; that designation had not yet been introduced by the late-1960s. Any individual who understood the physics involved was both welcome and invited to assist.
Others at Rocky Flats also contributed to this document through their knowledge of plutonium processing. Some are retired, some still working; but all were knowledgeable persons in the arena of plutonium production in the 1960s. These included Louis DiGiallonardo, James C. Docktor, Robert L. Fiore, Paul Sasa, and Jack D. Weaver.

The patient and capable efforts of Christine White and Peggy Shiffer who work at Lockheed Martin in Idaho are, as in past papers under this program, gratefully recognized. They prepared figures and tables, respectively. This was rendered more difficult because of separation; they work in Idaho, the author, Colorado.
APPENDIX

This paper naturally falls into two closely allied programs. Both involve manually assembled arrays of massive plutonium ingots. Both were performed as \textit{in situ} experiments. Both are collected in this paper for a sense of completeness. Truly new results are presented for otherwise uncontained film cans housing these ingots. An even earlier study involved similar ingots in similar film cans, but these cans were, in turn, contained in large steel drums. That study was described in an early internal Rocky Flats report. This author can add nothing new to that program beyond that recorded in that report. Consequently, that report is reproduced in this appendix in its entirety.
NEUTRON MULTIPLICATION MEASUREMENTS OF PLUTONIUM INGOTS IN ARRAYS

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NEUTRON MULTIPLICATION MEASUREMENTS OF PLUTONIUM INGOTS IN ARRAYS

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Appreciation is expressed to the following for work done in performing the reported measurements: H. Clark, H. W. King, R. J. Kosizek, D. Payne, W. R. Sheets, C. A. Stevens, and K. O. Zumwalt.
NEUTRON MULTIPLICATION MEASUREMENTS OF PLUTONIUM INGOTS IN ARRAYS

Donald R. Ferguson

Abstract. Included in the report are a series of neutron multiplication measurements on partially moderated and partially reflected arrays of 55 gallon drums. Each drum contains a plutonium ingot of about 12 kilograms with a density of 15.8 grams per cubic centimeter.

INTRODUCTION

Two sets of neutron multiplication measurements were made for the dual purpose of examining the safety margin of the present mode of plutonium-ingot storage which was based on calculations; and secondly to obtain data which could be used in developing safe storage rules, hopefully to increase current storage capabilities for the ingots.

EXPERIMENTAL SETUP

The first set of measurements was made in a high vault of 15 by 25 by 13.5 feet with thick concrete walls. A false floor was laid consisting of 4-inch thick solid concrete blocks. An opening was left in the center of the room to place two boron trifluoride (BF$_3$) counter tubes for permanently locating them beneath the center of the array as shown in Figure 1.

Measurements:

Measurements were made on three types of arrays in the vault:

1. A planar close-packed hexagonal array of 46 drums.
2. A two-layer high hexagonal array with 92 drums.
3. A three-wide by two-high hexagonal array around the perimeter of the vault with 136 drums.

A second set of measurements made in a tunnel connecting to the vault. The tunnel is 7.5 feet wide by 10 feet high by 225 feet long with thick concrete walls. Measurements were made also on three different arrays:

1. One row, two high by 23 drums long.
2. Two rows, two high by 23 drums long.
3. Three rows two high by 23 drums long.

Two BF$_3$ counters were placed against the wall on the end of these arrays to minimize source-effect distortion of the reciprocal multiplication curves.

Fuel and Equipment:

The fuel used for the measurements was plutonium ingots having a density of 15.8 grams per cubic centimeter (g/cm$^3$). The weight of the ingots varied from 11.4 minimum to 12.0 kilograms (kg) maximum. The average weight of an ingot in the array was 11.8 kg. The ingot dimensions are approximately 10 by 12 by 0.4 inches thick. Each ingot was wrapped in a polyethylene bag and plastic tape which weighs about 2.5 pounds.$^1$ The wrapped ingot is then contained in a 19-inch diameter by 21$\frac{1}{2}$-inch high film can.

The outside container is a 55-gallon drum of 24-inch diameter by 35 inches high. A Fibre Pak®,$^2$ which is a cardboard annulus having a 0.25-inch wall thickness, 15-inch diameter, 16-inch height, and a weight of 7 pounds, is placed in the bottom of each 55-gallon drum. On top of the Fibre-Pak

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$^1$If the plastic were distributed evenly around the ingot the thickness of the plastic would be about 0.2 inches.

Numerals on drums indicate the sequence in which the drums were added to the array.

Drums added to only one side of array because of limiting dimensions of room. The addition of these drums indicated that the array was close to being infinite. Thus, the further addition of drums was unnecessary.

FIGURE 1. Diagram of First Array Measurement in the Room.

is placed the film can containing the ingot. On top of the film can, another Fibre-Pak spacer is placed which is 13.5 inches high and weighs 5 pounds. The drum cover is then sealed on the drum. When the drums were stacked, the ingots had a 35-inch minimum vertical center-to-center separation.

The drums when placed together in a close-packed hexagonal array occupy an area lattice of 499 square inches and a lattice volume of 10.1 cubic feet. The plutonium density in the arrays was 0.041 g/cm³.

EXPERIMENTAL PROCEDURE

In the first set of measurements, one drum from the center of the array was placed on the false concrete floor directly over the two detectors. A 60-second count was taken and used as a
base count for calculating the reciprocal neutron multiplication for the rest of the measurements.

Additional drums were added two at a time on opposite sides of the center drum to maintain a symmetric hexagonal array. A 60-second count was taken and the reciprocal multiplication (1/M) data were calculated and plotted after each fuel addition.

After assembling the 37-drum hexagonal array (Figure 1) additional drums could only be added to three sides of the array because the array blocked off the other side of the room.

The drums were then arranged in an array, 3-drums wide and 2-layers high around the perimeter of the room as noted in Figure 2 and were found to be less reactive than the array in Figure 1.

In the set of measurements done in the tunnel, the detectors were placed on the floor at the end of the array (Figure 3). Drums were added in a row along the tunnel wall and counts were taken periodically. After 15 drums were placed in a row, the 1/M plot clearly became asymptotic. The array was extended to 23 drums in length just to make certain the plot stayed flat.

Then a second layer of drums was added to the first row in a similar manner. A second row was started in the same manner as the first and similarly a third row.

FIGURE 2. Diagram of Array Measurement in the Room.
The dimensions of the tunnel permitted only 3 rows of drums stacked 2-layers high.

RESULTS AND DATA

Figures 4 and 5, respectively, are graphs of reciprocal multiplication data (1/M) versus total number of drums for planar, and 2-layer, hexagonal arrays. Extrapolations were made to the number of drums needed to fill the room.

Figure 6 is a graph of extrapolated values of 1/M versus layers of drums stacked vertically. Each layer contained 97 drums which would completely fill a 15 by 25-foot room. Clearly, for large arrays the drums could only be stacked two high, as a straight-line extrapolation of the data indicates 2.75 layers to be critical.

Figure 7 is a graph of measured values of 1/M versus layers of drums in a critical array. The 7-, 19-, and 37-drum arrays are hexagonal in shape. The 46-drum array, which was the largest assembled, had drums added only to 3 sides of the 37-drum hexagon and was therefore not symmetric. Straight-line extrapolations of 1/M values show that the 19-drum hexagonal base extrapolated to 3.6 layers being critical and most nearly gives a cubic critical array, approximately 10 feet wide by 10 feet high.

CONCLUSIONS

The findings indicate that it was safe to more than double the present storage capabilities for plutonium ingots. The former mode of storage, which was calculationally based, called for only a planar rectangularly packed array of drums. The measurements demonstrated that a two-layer hexagonally packed array was subcritical. Consequently an increase was made in storage limits to a 2-wide by 2-high array of drums around the perimeter of a 15 by 25-foot room and a 2-wide by 2-high array down the length of the connecting 225-foot tunnel with additional provision for pass-through of drums, past the existing array.

FIGURE 5. Hexagonal Close-Packed Array of Drums Two Layers High.

FIGURE 7. Number of Drums in a Symmetric Base.