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CRITICAL MASS STUDIES, PART I

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This report is based on a series of experiments performed in the Pajarita Canyon Laboratory at Los Alamos, N.M. during March and April 1946.

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TABLE OF CONTENTS

ABSTRACT

Using 1108 one-inch cubes of mock-up UF_6, containing 54 kg of U-235 made from UF_4 (95% isotopic concentration of U-235) and $(CF_2)n$, and similar hydrogenous cubes of $(CH_2)n$, in various proportions and under various conditions of accumulation (all relevant to conditions in the K-25 plant), 26 assemblies were built to criticality and four other assemblies were built which did not become critical. From these experiments, at least some information was obtained on the effect on critical mass of (1) amount of intermixed hydrogen, (2) uniformity of hydrogen-uranium mixing, (3) density, (4) geometry, (5) shielding or reflector materials around the accumulation.

Method of preparation and properties of the materials used, description of experimental arrangements and procedures, and statement of data obtained, are given in this report.

A brief summary is given here of the pertinent results obtained in the experiments:

A cubical assembly of 54 kg of U-235, intimately mixed with fluorine and carbon to simulate UF_6C at a density of 4.7 gm/cm^3, containing no intermixed hydrogen but completely surrounded by 7 inches of paraffin, did not become critical. Best estimates from neutron multiplication measurements indicate about 100 kg as the critical mass under these conditions.

The critical mass drops sharply as small quantities of hydrogen are intermixed. A cubical assembly, surrounded by paraffin, having an H to U-235 atomic ratio of 5 became critical with 26 kg of U-235; one having H to U-235 atomic ratio of 10 was critical at 16 kg U-235.

In studying the effect of geometry, it was found that paraffin enclosed 6" x 6" rectangular parallelepipeds having H to U-235 atomic ratios of 10 and 20, could apparently be extended to indefinite lengths without becoming critical.

By stacking paraffin enclosed cubical assemblies of H to U-235 atomic ratios of 10 and 20 with different sized air gaps between the cubes, the critical mass was found to vary inversely approximately as the 1.7 power of the overall U-235 density.

The critical mass of paraffin enclosed cubical assemblies of H to U-235 atomic ratio of 10, when shielded by cadmium or boron, was found to be approximately double the critical mass of similar assemblies having no shielding.

I. INTRODUCTION

In order to determine and, where necessary, improve the safety of the K-25 plant from dangerous accumulations of fissionable material and to prepare for examination, from the standpoint of safety, possible proposals for altering the K-25 operating schedule, it was suggested that a series of experiments be undertaken for the determination of critical masses of U-235 under conditions similating those in the K-25 plant. Tennessee Eastman Corporation and Monsanto Chemical Company of Oak Ridge were invited to participate in these experiments since the results obtained would be expected to relate to operations performed by these Companies also. TEC was interested to the extent of sending a man from their organization to assist in the entire experimental program. Monsanto requested that they be kept informed on the experimental progress, and at the conclusion of the work, Dr. E. Greuling of that Company furnished considerable assistance in calculations and interpretation of the results.

The Oak Ridge group undertaking the experimental program had not worked previously with critical mass assemblies. Therefore, in view of certain hazardous aspects of these tests, arrangements were made for the first series of experiments to be performed at Los Alamos under the supervision of men there who had considerable experience in work of this kind. Thus a double objective was achieved:

(a) Considerable information on critical masses under conditions of interest to K-25 was established.

(b) Experience was gained by the Oak Ridge group, which could then continue into further investigations as necessary for the safety of the plants at Oak Ridge.

II K-25 PLANT CONDITIONS UNDER INVESTIGATION

In the K-25 plant, uranium hexafluoride in gaseous form is circulated through extensive equipment designed to separate the uranium 235 and 238 isotopes by the gaseous diffusion method. A small percentage of air is present in the system as the chief impurity, but small amounts of other impurities, including HF, are usually present also. The latter impurity is considered the most hazardous one. With all components in gaseous form, there is little danger of a sustained chain-reaction. But under certain conditions, there is a possibility of condensing the hexafluoride into liquid or solid form, and of condensing small amounts of HF in an intimate mixture with the uranium material. The experimental program, therefore, was designed to investigate critical masses of U-235, when in a form simulating uranium hexafluoride with none or only small amounts of hydrogen intermixed, and under other conditions, (density, reflector, etc.) at least as hazardous as would be expected to occur in the plant.

Density is an important factor in the amount of material required to support a sustained chain-reaction. Under ordinary condensing conditions, UF_6 deposits as a more or less fluffy solid, having overall densities up to about 4.5 gm/cm^3. (The Kellex Data Book states 2.1 gm/cm^3 as the normal packed density of the solid, 4.5 gm/cm^3 as the maximum liquid density). Under studied conditions, arranged to give maximum density of solid UF_6, a value of 5.1 gm/cm^3 was achieved, but it seems that these conditions would never inadvertently occur in the plant. For the present experiments, therefore, a density of 4.5 to 5.0 gm/cm^3 (in the absence of intermixed moderator) would simulate worse density conditions, producing lower critical masses, than would be expected to occur in plant operation.

It was assumed that no reflector conditions around an accumulation in the plant would be more effective in causing a sustained chain-reaction than complete enclosure in seven inches of paraffin. It is recognized that, for accumulations containing zero or little moderator, other materials at corresponding thicknesses are better reflectors than paraffin, but in the plant, thick layers of these better reflectors completely enclosing an accumulation would not occur. Most of the experimental assemblies, therefore, were arranged with complete casings of paraffin seven inches thick.

At the time these experiments were inaugurated K-25 was chiefly concerned with only partially separated isotopes of uranium (up to 30% U-235).

It was anticipated, however, that operating schedules might be changed so that more nearly pure U-235 would be present at certain points. For these reasons, it was proposed that a series of investigations be carried out on material at 30% U-235 isotopic concentration and at 95% concentration. The 95% material for the experiments was available immediately, hence experiments on this enriched material were performed first.

III. EXPERIMENTAL MATERIALS

The toxic and corrosive properties of UF_6 made it virtually impossible to perform the desired experiments directly on uranium in this chemical form. It was necessary to use a "mock-up" material having nuclear properties similar to UF_6 but having chemical and physical properties rendering it amenable to handling and control. The "mock-up" which appeared to meet these specifications most exactly was a mixture of UF_4 and enough polytetrafluoroethylene* $(CF_2)n$, to simulate UF_6C. The properties of UF_4 were acceptable: solid powder, high density, non-corrosive, non-hygroscopic, insoluble; the poly TFE (abbreviation for polytetrafluorethylene) was similarly inert and when added to UF_4 produced a plastic mixture which could be formed under pressure at room temperature into solid blocks. These two components, when mixed together, micropulverized, and pressed in suitable dies, formed a solid possessing quite satisfactory physical, chemical and nuclear properties.

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*Analysis of the polytetrafluoroethylene showed a fluorine content of 76.2% corresponding to a composition $CF_{2.02}$

Blocks of the UF_4--poly TEF mixture, referred to as U-cubes, contained no hydrogen. In experiments involving hydrogen, it was necessary to alternate U-cubes with hydrogen-containing blocks of the appropriate size. Polyethylene, $(CH_2)n$, was chosen as the material from which the "hydrogen blocks," referred to as H-cubes, were formed.

Special attention was given to the procedures by which the uranium-plastic cubes were prepared. Half-kilogram batches of finely powdered UF_4 were received from the stock of Y-12 product stored at Los Alamos. This material averaged about 95.3% U-235 isotopic concentration. Each batch was carefully weighed, mixed with an amount of poly TFE sufficient to give a mixture equivalent to UF_6 C and micropulverized to give a thoroughly mixed, finely ground powder. Appropriate batches of this powdered mixture were then weighed out, poured into molding dies, and cold-pressed at 40-50 tons per square inch pressure. The smooth-surfaced, regular cubes resulting from this operation were measured, weighed and stored in boron-lined buckets for subsequent use.

For the polyethylene cube-stock, Bakelite DE-3401 natural polyethylene, in the form of 1 1/2" cylindrical rods, was used. The rods were machined into one-inch rectangular rods and cut into either one-inch or one-half inch lengths. Analyses* of the polyethylene show 13.86% hydrogen content. Assuming the remainder to be carbon,

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*Credit is due Mr. C.O. Struther of the Linde Air Products Company and Mr. W.W. Harris of Carbide and Carbon Chemicals Corporation for these data.

one obtains $CH_{1.92}$ as the composition. Since, in the interpretation of the critical mass data, there is interest only in the hydrogen content, the validity of this assumption is not important. It does have a bearing, however, on the calculated critical masses.

Both the polyethylene and the poly TFE used in the cube fabrication were critically examined by chemical and spectrographic means for neutron absorbing impurities such as boron and cadmium which might affect the results of the experiments. In addition, independent neutron-absorbing tests were performed on samples of the material by Dr. E.O. Wollan of Monsanto and Dr. J.R. Dunning of Columbia University. None of these tests indicated the presence of impurities which would affect the experiments in any way.

Table 1A contains a quantitative description of the U-cubes, used in the experiments, and Table 1B a description of the two sizes of H-cubes.

TABLE 1

MATERIALS USED IN CRITICAL ASSEMBLIES

A - Properties of Uranium-Plastic Cubes - (U-Cubes)

Dimensions	1.004" 2.550 cm
Volume	1.012 in^3 16.584 cm^3
Mass	78.44 gm
Density	4.73 gm/cm^3
Total U Content	51.16 gm
U-235 Content	48.76 gm
Fluorine Content	24.71 gm
Carbon Content	2.57 gm

TABLE 1 (Cont.)

B - Properties of Polyethylene Blocks (H-Cubes)

	"One-inch" size	"One-half inch" size
Dimensions	1.005" cube 2.553 cm cube	1.006" x 1.006" x 0.502" 2.555 x 2.555 x 1.275 cm
Volume	1.015 in^3 16.632 cm^3	0.508 in^3 8.324 cm^3
Mass	15.11 gm	7.61 gm
Density	0.91 gm/cm^3	0.91 gm/cm^3
Hydrogen Content	2.09 gm	1.05 gm

These data represent average characteristics of a typical block of the respective materials. The blocks were made with sufficient geometrical regularity that overall stacking density of an assembly was only slightly lower than the absolute weighted density of the components.

IV. BRIEF DISCUSSION OF THEORY

In a recent report,(1) Beck has given an elementary discussion of the factors involved in a determination of critical mass by the method of adding small increments to a fissionable accumulation while noting the rise in neutron flux in the vicinity. The theory and experimental procedure described therein are applicable to the present case.

(1) Beck, Clifford K., "Preliminary Report on Critical Mass Experiments at X-10", December 5, 1945.

(a) - Neutron Density and Approach to Criticality

Briefly, the neutron density from background radiation or from a neutron source is multiplied in the vicinity of a fissionable accumulation by the factor $\frac{1}{1-k}$, where k, the "reproduction factor" relates the average number of neutron offspring to the number of parent neutrons in each generation in the accumulation. The accumulation is sub-critical, critical or supercritical as k is less than, equal to, or greater than unity. When k is much less than unity, i.e., the accumulation is far less than critical, the neutron density in the vicinity of the accumulation is essentially equal to background. As small quantities of fissionable materials are added to the sub-critical assembly, k increases slightly, and the neutron density increases, over a short transition period, to a new constant value. If small increments are added periodically, the neutron density assumes an increased equilibrium value after each addition until the total accumulation reaches criticality. When $k = 1$, assuming a constant number, S, of neutrons entering the assembly, the neutron density from the accumulation begins to increase at a constant rate, and continues to increase as long as conditions remain unchanged. If $k > 1$, the density increases exponentially with time, at a rate dependent on the amount by which k exceeds unity, (Fig. 1).

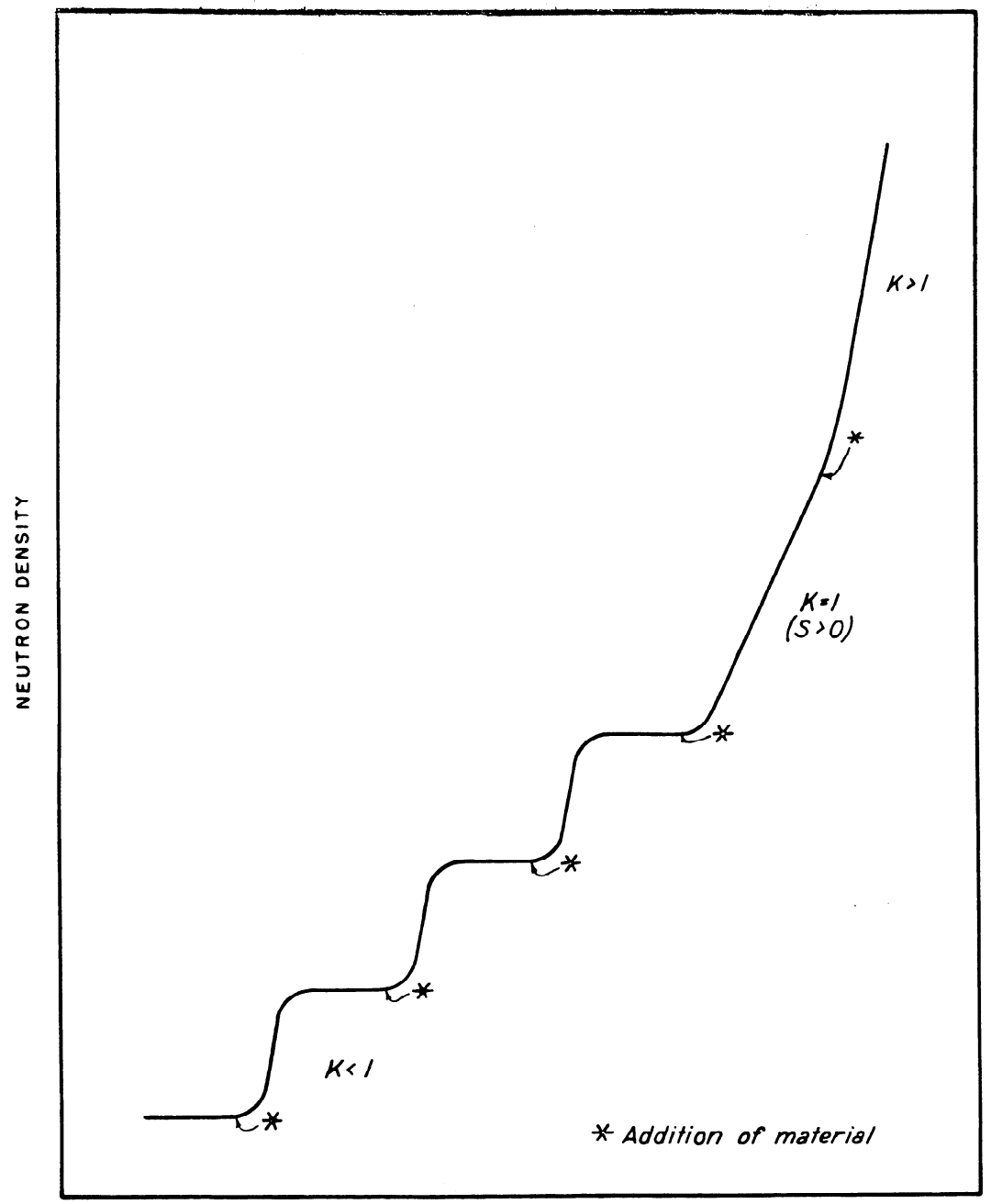


FIGURE 1

DWG. NO. LD-A-90

(b) - Delayed Neutrons

It is extremely fortunate that a portion of the neutrons from fission are delayed. This prevents instantaneous adjustment of the neutron density in response to a change in k, and permits work to be done on near-critical assemblies with much less danger. The existence of delayed neutrons also permits use of the so-called "source-jerk" method of determining qualitative nearness of approach to criticality: With a neutron source, S, nearby, the neutron density from a particular assembly of fissionable material has a value $n_1 = \frac{S}{1 - k}$ which is considerably above background. The response of the neutron density to a sudden removal of the source indicates the nearness of the assembly to criticality. Immediately after removal of the source, the density becomes

$$n_2 = n_1 \frac{f}{1-k(1-f)}$$

$$= \frac{S}{1 - k} \cdot \frac{f}{1 - k(1 - f)}$$

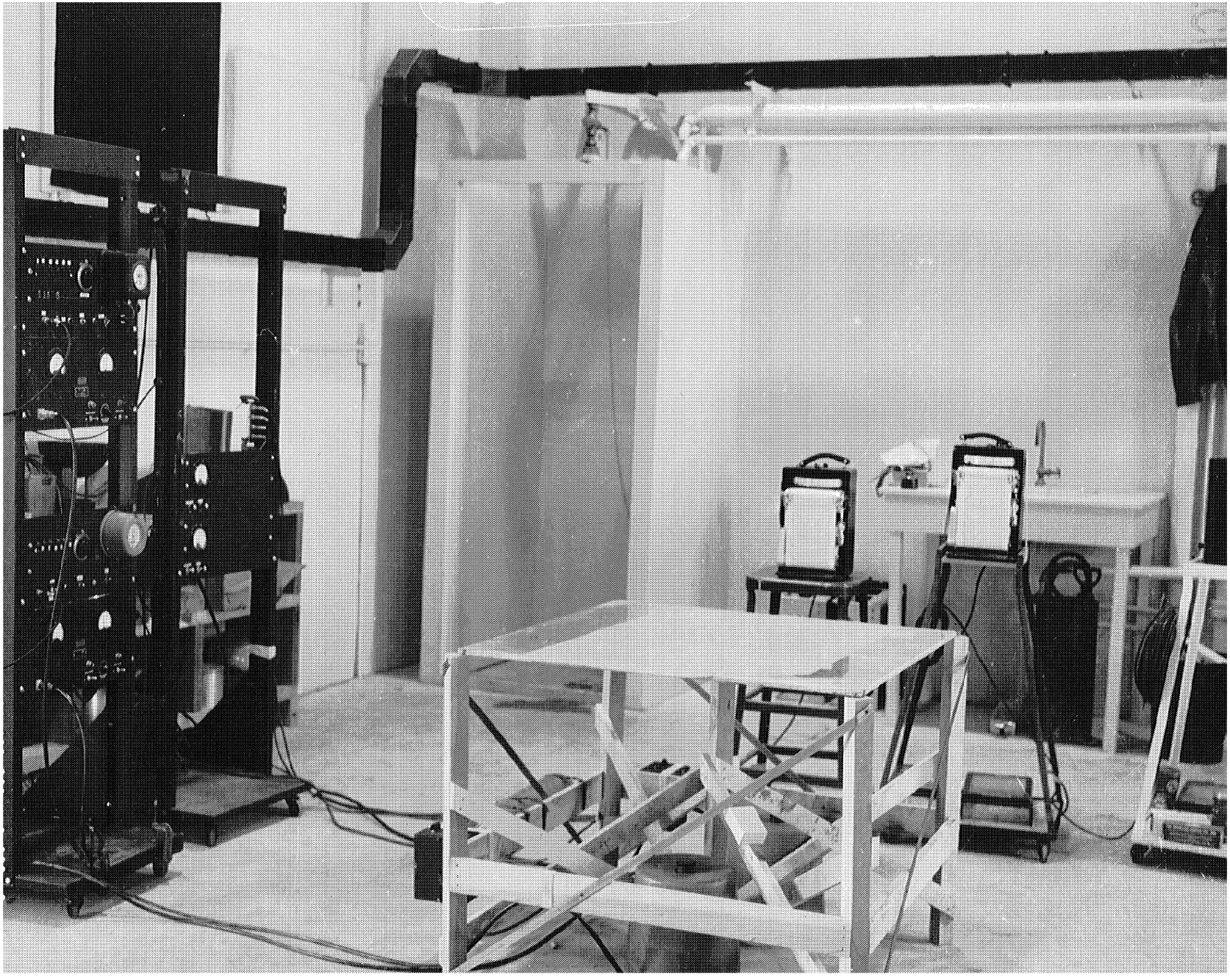
where f is the fraction of delayed neutrons. If n_1 is larger than n_2 by a factor of 3 or 4, then k is 2f or 3f less than 1. But if $k \cong 1$, then n_1 and n_2 are almost equal, i.e., the neutron density decreases slowly from the value n_1 when the source is removed. Thus, the rate at which the neutron density decreases after the removal of the source indicates the nearness of the assembly to criticality.

(c) - Neutron Multiplication: "Extrapolated" Critical Mass

If circumstances render it impossible or undesirable to build an accumulation to criticality, it is possible to obtain a more or less accurate value of the critical mass, depending on the nearness of approach to criticality, by the method of neutron multiplication. If S neutrons from a source are directed into a fissionable mass, they will be multiplied into a density $n = \frac{S}{1-k}$. If a neutron detector, say a G-M counter, arranged to measure this neutron density (or in practice, to indicate a counting rate proportional to the density) is used to obtain counting rates, C, for several different values of k, the data may be used to obtain a value of critical mass by extrapolation. The reciprocal of the counting rate plotted against the mass in the accumulation gives a more or less linear curve in the near-critical region, which may be extrapolated to the intercept corresponding to infinite counting rate for the value of the critical mass. It is desirable of course, to make the measurements as near critical as possible, since linear extrapolation is reliable only over relatively short distances below critical.

V. APPARATUS

In Figure 2 may be seen the table on which all the critical assemblies were built, and the monitoring instruments and recording meters. On the floor underneath the table may be seen two cylindrical blocks of paraffin, each surrounding a boron-lined proportional counter



for measuring neutron density. These (independent) detectors were connected to amplifying circuits (at the left of the table) and to indicating meters and mechanical counters mounted on the panel-boards. On the horizontal leg supports of the table may be seen two additional (also independent) neutron detectors consisting of paraffin-encased boron trifluoride filled cylindrical ionization chambers. These also were connected to amplifying circuits and, in turn, to indicating and recording meters mounted on stands directly behind the table. Thus, immediately underneath the critical assemblies four independent neutron detecting and measuring instruments were located.

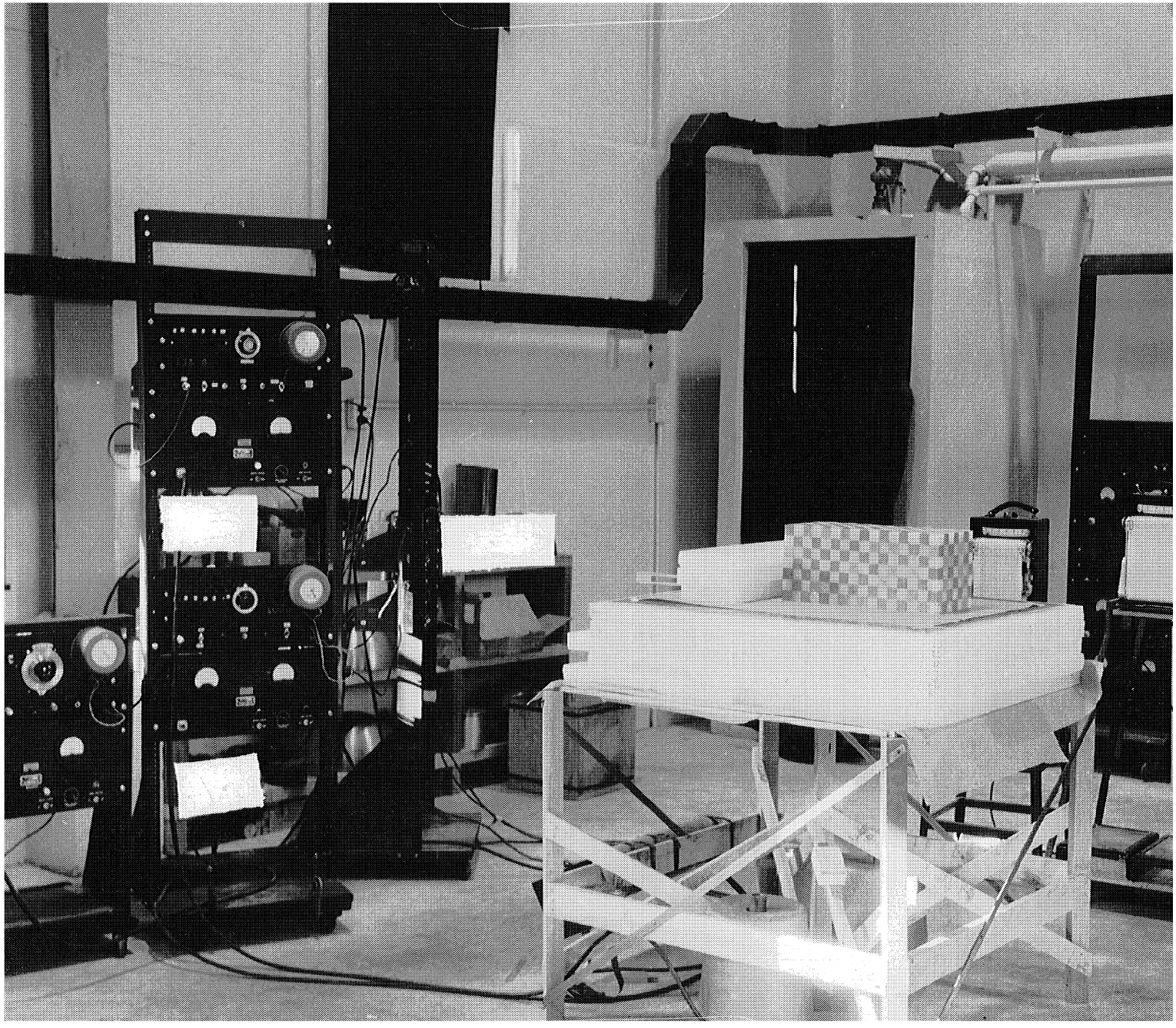
A piece of 1/2 inch aluminum served as the top of the table on which the assemblies were mounted. For paraffin-enclosed assemblies, (all except "untamped" assemblies) a 7-inch layer of paraffin, topped by an eighth-inch sheet of aluminum were placed on the table and on this as a base, the critical assemblies were built (Figs. 3 and 4). "Untamped" assemblies were built directly on the aluminum table top with no paraffin underneath (Fig. 5).

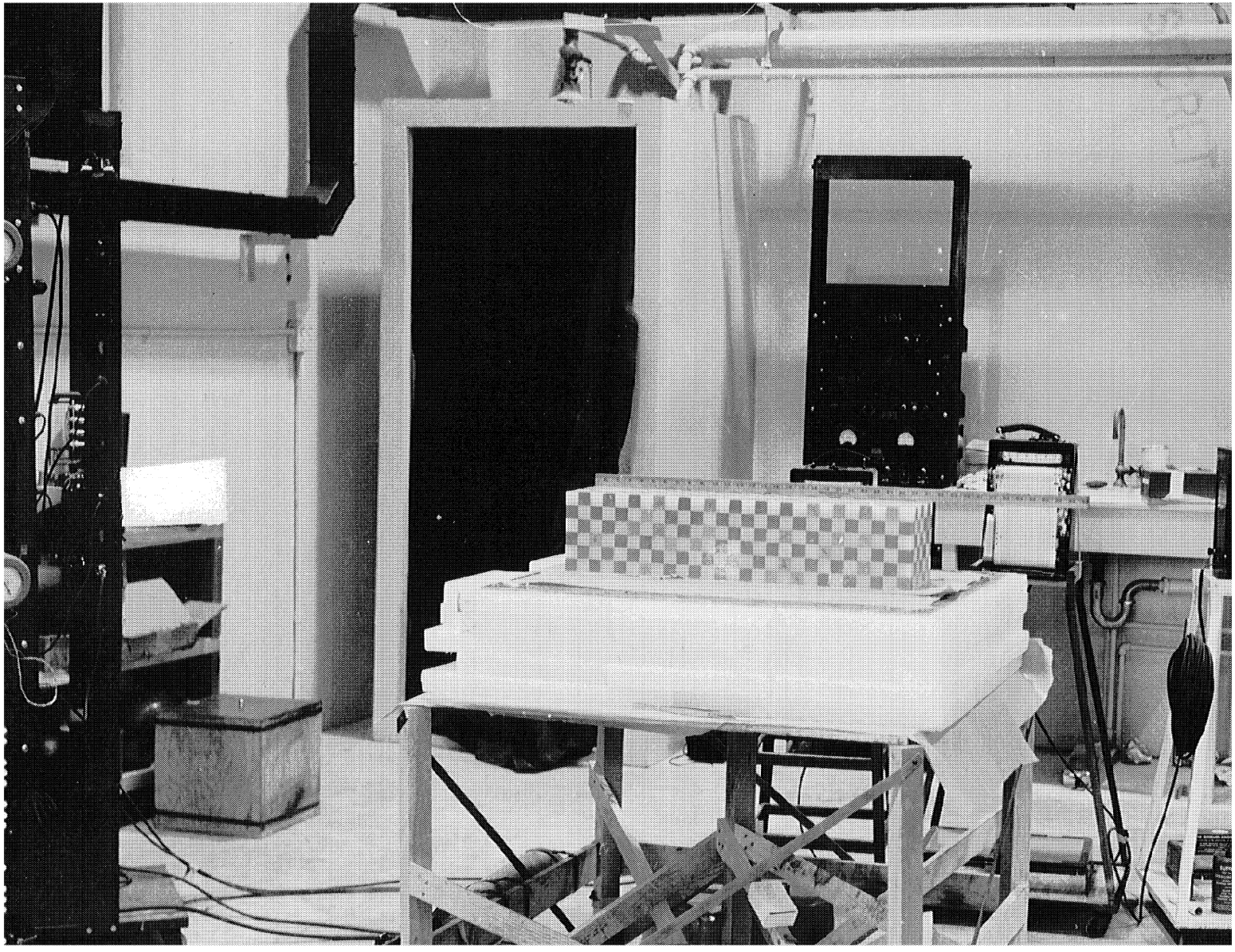
VI. OUTLINE OF TYPICAL EXPERIMENT

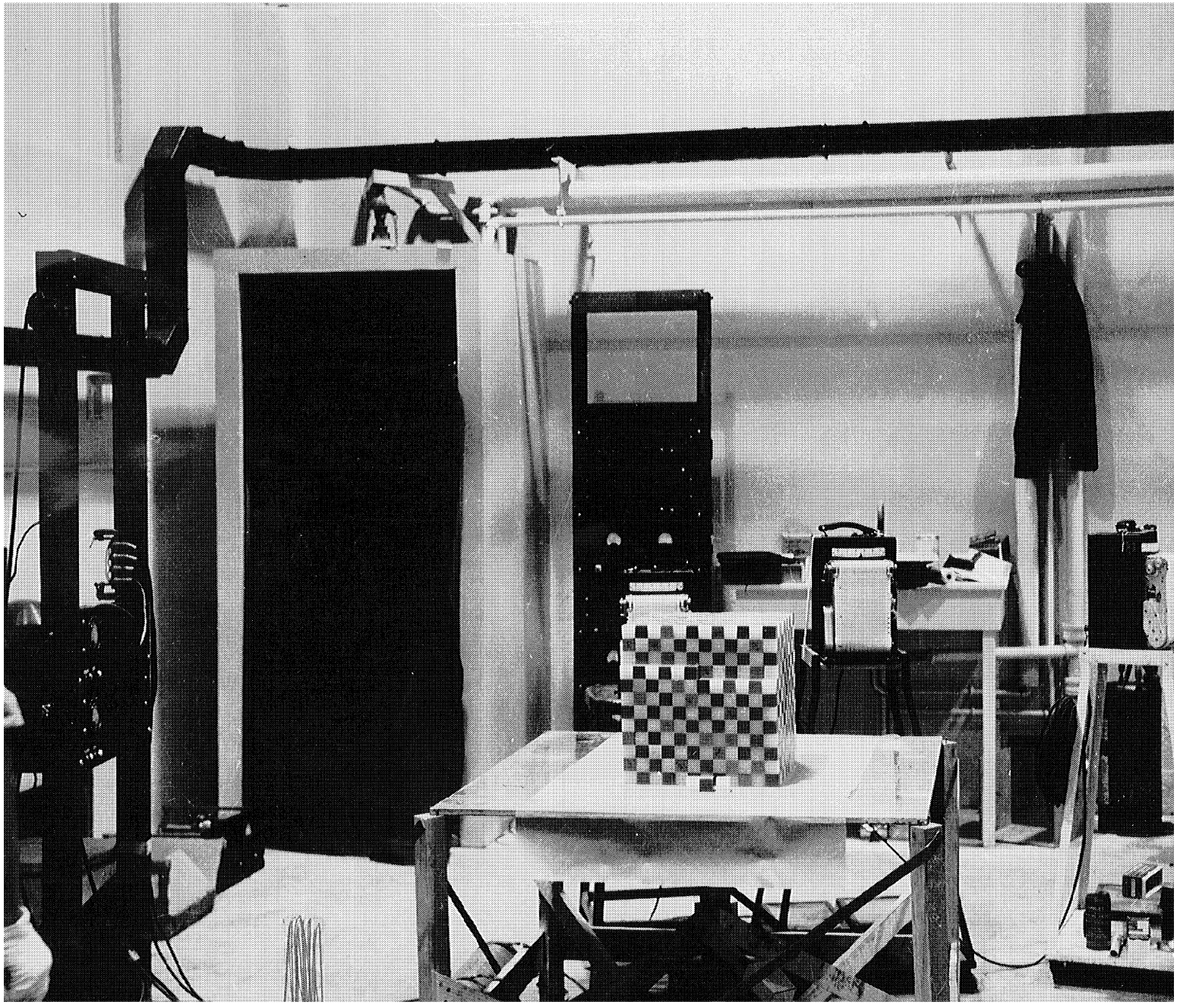
To determine the critical mass of the chosen materials under a given set of conditions, a standard procedure was adopted.

(a) Prior Planning

The persons performing the investigation agreed in advance on the plan of the experiment, its objective, and the responsibility of each person. Only one person was involved in building a given assembly. He was in absolute charge throughout the experiment.







Assistance could be asked of others present, but the number of assistants was kept to a minimum. The remaining persons kept at a considerable distance and were usually seated during the experiment.

(b) Instrument Check

Before beginning an assembly, the person in charge checked the sensitivity of the various neutron detectors by bringing a small neutron source near each detector in turn and noting the response of the connected meters, counters and recorders. Similar checks were made at intervals throughout the experiment.

(c) Accumulation

A quantity of active material, estimated to be near critical under the final conditions to be arranged, was slowly assembled on the table in a predetermined configuration, with a polonium-beryllium neutron source (1-100 curies) nearby. Each block was added to the assembly from the side, not from above, so that it would fall beside the assembly if dropped, not on it. Each cube was added slowly, to afford observation of the resulting density changes and to permit reversal of the action if desired.

(d) Addition of Reflector

The accumulation of active material was slowly surrounded with paraffin (in the "tamped" experiments). Paraffin slabs were placed around the assembly first. If criticality was not approached the paraffin was removed from the sides and a layer was placed on top.

(It is obviously more hazardous to approach criticality by tamping on top than on the side; if the assembly was not critical with the sides enclosed, it should not become critical with the top covered instead). With paraffin on top, the operator then proceeded slowly to enclose the sides of the assembly.

If criticality was not reached when fully enclosed in paraffin, or if the assembly became critical before fully enclosed, the paraffin was removed and the quantity of active material increased or decreased. The paraffin was then replaced by the same procedure as described above. By repeated trials, the quantity of active material was so adjusted that the assembly was just critical when fully enclosed in paraffin. The amount of material present then represented the critical mass under the particular conditions existing at the moment.

(e) Disassembly

As soon as the value was established, the paraffin and part of the active material were removed so that the assembly was sufficiently sub-critical that movement of personnel and material in the vicinity could not inadvertently render the assembly critical again. This concluded the experimental determination of critical mass for the particular set of conditions.

VII. EXPERIMENTAL RESULTS

Following the procedures outlined above, determinations of critical mass were made for various conditions of accumulation. The one-inch cubes of mock-up UF at 95% U-235 isotopic concentration, were alternated

with appropriate numbers of polyethylene cubes $(CH_2)n$, in regular lattice structures (Figs. 13,14, 15, 16) of cubical or rectangular shape, and the size of the assembly was adjusted to criticality under various conditions of interest: no reflector, paraffin reflector, cadmium or boron shielded, etc. The effect on critical mass of several factors were at least partially investigated:

Effect of concentration of intermixed hydrogen.

Effect of reflector or shielding around the accumulation.

Effect of geometry.

Effect of density.

Effect of non-uniform mixing of U and H .

The pertinent experimental arrangements and the results obtained are described in the following sections:

(A) Effect on Critical Mass of Concentration of Intermixed Hydrogen

1. Paraffin Enclosed

Completely paraffin-encased assemblies were built to criticality with H-cubes intermixed with U-cubes in various ratios from 1/2 H : 2 U to 7 H:1U. Extrapolated values of critical mass were obtained from neutron multiplication measurements at still lower values of H-cube, U-cube ratios of 1:7 and 0:1.(atomic ratios of 1.4 and 0). The data are contained in Table 2 and Fig. 6.

TABLE 2

EFFECT OF INTERMIXED HYDROGEN ON CRITICAL MASS

(a) Cubical assembly

(b) Completely paraffin enclosed

(c) Stacking lattice configurations refer to diagrams in Figs. 13, 14 and 15.

H-cubes: U-cubes	0:1	1:7	1/2:2	1/2:1	1:1	2:1	4:1	7:1
Lattice configuration	A	H	D	F	B	E	G	I
Size of assembly (cube units)**	10x10x11*	11x11x10+*	11x11x10+	9x9x10+	9x9x8	8x9x8+	10x9x9+	11x12x12
Overall density, gm/cm^3	4.7	4.3	4.0	3.5	2.8	2.2	1.7	1.4
H-cubes	0	158	242.5	263.5	324	396	672	1421
U-cubes	1108	1108	970	527	324	198	168	203
Total cubes	1108	1266	1212.5	790.5	648	594	840	1624
Atoms H/atoms U-235	0	1.4	2.5	5	10	20	40	70
Critical mass U, kg	105***	63***	49.6	27.0	16.6	10.2	8.6	10.4
Critical mass U-235, kg	100***	60***	47.3	25.7	15.8	9.7	8.2	9.9

*Not critical

**These assembly dimensions are given in the number of cubes (H and U) along an edge. The actual dimensions, in inches, are slightly larger.

***Extrapolated value from neutron multiplication in 54 kg of U-235

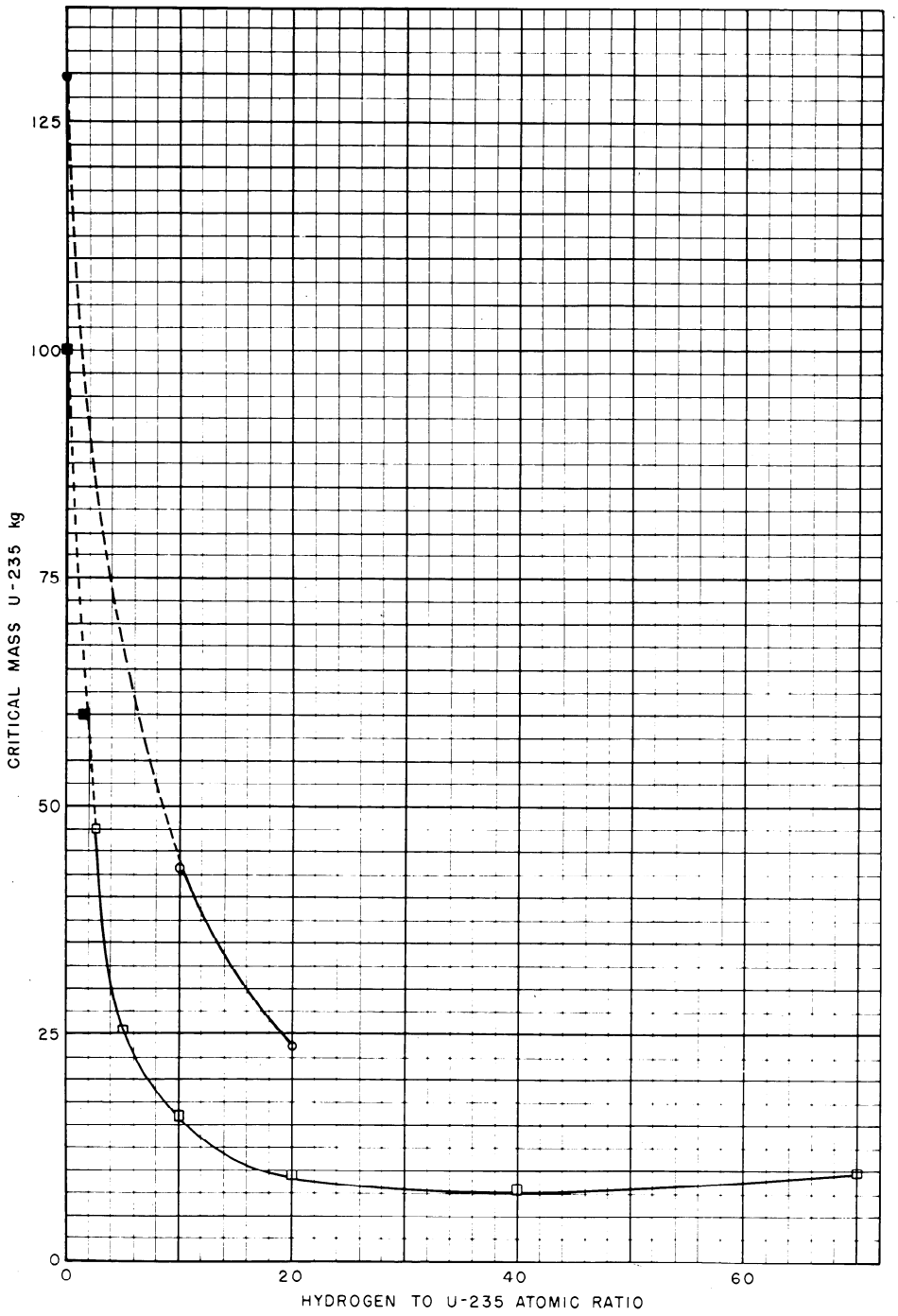


FIGURE 6

VARIATION OF CRITICAL MASS U-235
WITH CONCENTRATION OF INTERMIXED HYDROGEN

In this series of experiments, there are three factors which vary: overall density, uniformity of H-U mixing, and percentage of contained hydrogen. The effect of overall density variation on critical mass is discussed separately in Section E. It may be pointed out here, however, that this simultaneous change in density and in concentration of moderator is a realistic analogue of the conditions which would exist in the K-25 plant, say, if varying amounts of a low-density moderator should accumulate with a given compound of fissionable material. The effect of variation in one factor cannot be separated from that of the other.

A description is given in Section B of a brief study, both experimental and theoretical, of the effect of non-uniform mixing of H and U in the accumulation.

2. Unenclosed

Three cubical assemblies were built with only air as a reflector. These contained no internal hydrogen, equal numbers of H and U cubes, and an H cube: U cube ratio of 2, respectively. The unmoderated assembly, containing 1000 U cubes (48.8 kg of U-235), failed by a wide margin to become critical. Enough material was available to make the other assemblies critical (Fig. 6, Table 3).

TABLE 3

EFFECT OF INTERMIXED HYDROGEN ON CRITICAL MASS

(a) Cubical assembly

(b) No reflector

(c) Stacking lattice configurations refer to Figs. 13 and 14.

H-cube: U-cube	0:1	1:1	2:1
Lattice configuration	A	B	E
Size of assembly (cube units)	10x10x10*	12x12x12-	12x10x12-
Overall density, gm/cm^3	4.7	2.8	2.2
H-cubes	0	844	926
U-cubes	1000	844	463
Total cubes	1000	1688	1389
Atoms H/atoms U-235	0	10	20
Critical mass U, kg	136**	43.2	23.7
Critical mass U-235, kg	130**	41.2	22.6

*Did not go critical

**Extrapolated value from neutron multiplication in 48.8 kg U-235

Reference to Fig. 6 shows the critical mass to be strongly dependent upon the degree of hydrogen moderation at values of the atomic ratio less than 10. It is desirable at this time to point out a cause for an error of possibly as much as 10% in some of the values of the critical masses under these conditions which might be read from the curve. The hydrogen to U-235 atomic ratio has been calculated in every case from the appropriate cube ratio within the body of the assembly. What might at first be considered to be minor irregularities have arisen in the cube stacking at the surfaces of the array because of the inflexibility of cube dimensions and shapes. Further, in the case of paraffin encased assemblies, there is the question of whether the paraffin immediately in contact with a surface U-cube is not, to some degree, moderator and, therefore, should be considered in the evaluation of the atomic ratio. These small effects, at low dilution, are believed sufficient to alter the nominal H to U-235 ratio by an amount which will displace the curve, thereby causing errors of the above magnitude.

(3) Effect on Critical Mass of Inhomogeniety of H:U Mixing in Moderated Assemblies

The stacking of unit quanities of active material and moderator in the core of an assembly according to some regular pattern other than uniform mixing, may result, according to the

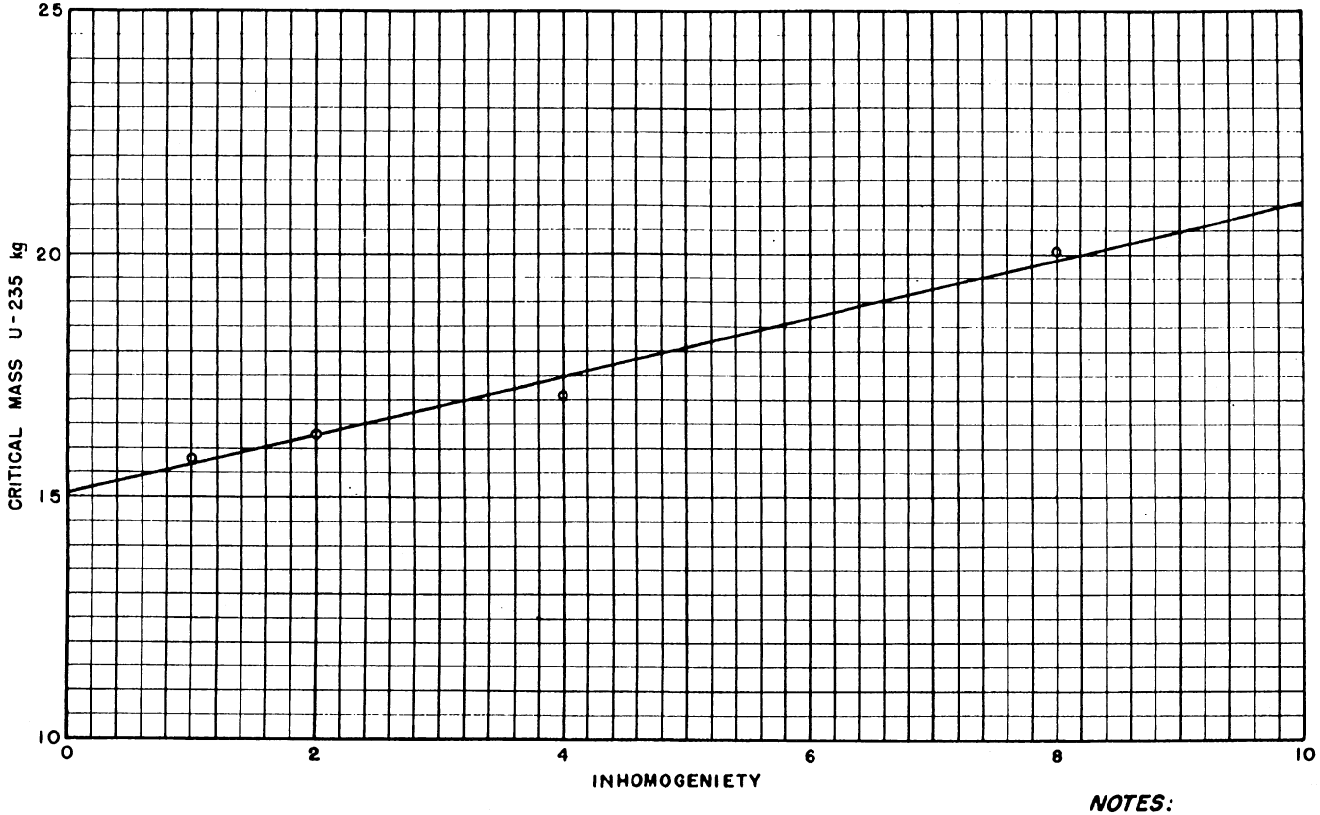
particular circumstances, either in larger or smaller critical masses than those for uniform mixing. In the case of a pile built of normal uranium and graphite, the careful spacing of blocks of uranium in the graphite resulted in a much smaller critical assembly than would have been possible with intimate mixing of the components (Smythe Report). In the case of the one-inch H and U cubes used in the experiments described in this report, the inhomogeniety effect when relatively small numbers of H cubes were used appears to be negligible. But for assemblies containing a 1:1 or larger ratio of H to U cubes, the effect is appreciable.

1. Experimental Study

An attempt was made to obtain some estimate of this inhomogeniety effect in assemblies having a 1:1 H-cube to U-cube ratio. Critical masses were determined for several cases in which the respective stacking patterns were different, but in all cases this nuclear ratio was maintained. In the first arrangement (Test 1, Table 4, and Fig. 13B) the cube pattern is similar along any edge of the assembly: H and U cubes alternate. In the second case (Test 2, Table 4, and Fig. 15L) a 2:2 ratio of H:U cubes is maintained in one dimension, while a 1:1 ratio holds in the other two dimensions. In the third case (Test 3, Table 4, and Fig. 15K) a 2:2 ratio holds in 2 dimensions, while H and U cubes alternate (1:1) in the third dimension. The fourth case (Test 4, Table 4, and Fig. 15J) is a

repetition of the first arrangement, except blocks of 8 H cubes alternate with blocks of 8 U cubes in an 8:8 ratio; or in other words, there is a 2:2 ratio in all three dimensions. Obviously, from the values of critical masses obtained, the effect of inhomogeniety in each successive arrangement of these four is considerably larger than that in the proceeding arrangement.

If one makes the rather arbitrary assumption that the inhomogeniety of mixing in these four respective arrangements varies in the proportions 1:2:4:8, it appears that the critical mass for uniform mixing obtained by extrapolation of the experimental data (Fig. 7), would not be greatly reduced below that for the 1:1 arrangement. Or, in other words, inhomogeniety in the 1:1 arrangement does not result in a critical mass substantially larger than the value which would result with the materials uniformly mixed. Theoretical considerations of inhomogeniety described in the following paragraphs, however, indicate that this assumption of inhomogeniety is not true, and that the uniformly mixed critical mass is actually about 14 kg instead of 15kg, as indicated in Fig. 7.



NOTES:
Cubical, moderated assembly with paraffin reflector. Hydrogen to U-235 Atomic Ratio = 10.

FIGURE 7

EFFECT OF INHOMOGENIETY ON CRITICAL MASS

DWG. NO. LD-A-92

TABLE 4

EFFECT ON CRITICAL MASS OF DIFFERENT STACKING ARRANGEMENTS OF COMPONENT MATERIALS

(a) Equal numbers of H and U cubes
(b) Overall density of 2.8 gm/cm^3
(c) Completely paraffin enclosed
(d) H to U-235 atomic ratio = 10
(e) Cubical assembly

Test number	(1)	(2)	(3)	(4)
Stacking lattice	B	L	K	J
"Inhomogeniety"	1	2	4	8
Size of assembly (cube units)	9x9x8	9x9x8+	9x9x9-	10x10x8+
H-cubes	324	334	350	412
U-cubes	324	334	350	412
Total cubes	648	668	700	824
Critical mass U, kg	16.6	17.1	17.9	21.1
Critical mass U-235, kg	15.8	16.3	17.1	20.1

2. Theoretical Consideration of Inhomogeniety Effects

Greuling[2] has derived an equation for computing the critical mass of spherical homogenous mixtures of enriched uranium and water surrounded by infinite water reflector. The critical condition

(2) Greuling, E., "Theory of Water-Tamped Water Boiler," LA-399

obtains, for these circumstances, when the total hydrogen absorption rate equals the net production rate of thermal neutrons:

$$N_H \sigma_H = N_{25} \sigma_f \nu H(d) - (N_U \sigma_U - N_U D \sigma_H) G(d) \quad (1)$$

Where N_H, N_{25}, N_U are respectively the atomic densities of hydrogen in the reflector and of U-235 and U in the core, D is the number of hydrogen atoms displaced by a molecule containing one uranium atom. The thermal neutron cross sections σ_H and σ_f are respectively those of $HO_{1/2}$ absorption and U-235 fission. σ_U is the combined thermal neutron cross section of a uranium atom and all the other atoms associated with it in the active material. The average number of neutrons emitted per fission is ν. H(d) and G(d) are leakage functions representing respectively (1) the probability that a fission neutron produced in the core, after slowing and diffusing, is absorbed as a thermal neutron in the spherical core of diameter d, and (2) the probability that a thermalized neutron is absorbed in the core after diffusing as a thermal neutron.

When the components of the spherical assembly are other than uranium and water, homogenized UF_6C and $CH_{1.92}$, for example, equation (1) can still be used to obtain the value of critical mass provided appropriate numerical values of the constants are used.

If the core components are arranged in a lattice configuration instead of being homogeneously mixed, equation (1) can be altered as follows to give the value of critical mass to be expected: the left side, which is a measure of the absorption rate common to core and reflector

is multiplied by $(n_o + vn_1)$ and the right side, which is the net production rate of thermal neutrons in the core, is multiplied by n_o. Here n_o and n_1 are respectively the average thermal densities in the uranium-containing lattice element and in the moderator lattice element and v is the volume ratio of hydrogen lattice element to uranium lattice element. Also the homogenous (assuming uniform distribution) uranium atomic density must be increased by the factor $(1 + v)$ which makes it equal to the uranium atomic density in the active lattice element.

The equation for calculating critical masses in latticed configuration of active element and moderator element then becomes:

$$\frac{\left(1 + \frac{n_1 v}{n_o}\right)}{1 + v} N_H \sigma_H = N_{25} \sigma_f \nu H(d') - (N_U \sigma_U - N_U D \sigma_H) G(d') \qquad (2)$$

The difficult operation in using this equation is the evaluation of the average neutron densities n_o in the active element and n_1 in the moderator element. The evaluation is made by assuming that the total core of the assembly is composed of small cells, each consisting of an active element surrounded by a concentric moderator shell of appropriate thickness, and applying standard diffusion equations to the neutron behavior in this simplified conception of the lattice assembly.

None of the above considerations includes the effect of epi-thermal neutrons on critical mass. Hence for unmoderated or little moderated assemblies where fast neutrons play a considerable part, the calculations would not give valid results.

Theoretical values of critical mass, computed by Dr. Grueling from equations (1) and (2), for the materials and conditions used in these experiments (a) for the experimental lattice configuration actually used, and (b) for a homogenous mixture of the core components, are given in Table 5 along with the values actually measured in the experiments.

From the data presented in Table 5A, it is apparent that the experimentally measured critical mass at H-cube: U cube ratios above 1:1 is considerably larger than would be obtained with uniform mixing. At H to U volume ratios of 1/2 : 1 (and lower) the calculated value is larger than the experimental. This, one would expect, because at this small degree of moderation, fast neutrons begin to affect appreciably the value of the critical mass, hence the experimental value should be lower than the calculated value which is based on the effects of thermal neutrons only. The experimental values and those calculated for the latticed array are in fair agreement in the region for which the assumptions in the calculations are valid.

From these considerations it is clear that the apparent minimum at an atomic ratio of 40 in the experimental curve of critical mass versus H to U-235 atomic ratio, Fig. 6, is the result of inhomogeniety

in H and U mixing in the assemblies. (Both the calculated and experimental curves are plotted in Fig. 12). The calculated values of the critical mass for homogeneous mixing of the components do not exhibit a minimum at this low value of the atomic ratio, and, furthermore, the calculated values are in general compatible with, and may be extrapolated to, the "Water Boiler" critical masses, which were obtained for H to U-235 ratios far larger than those considered here. (For example at H:U ≅ 500, Mc ≅ 1 kg). It is to be recalled that there is a real minimum in the critical mass curve at an atomic ratio of about 500.

TABLE 5.

COMPARISON OF EXPERIMENTAL AND CALCULATED VALUES OF CRITICAL MASS.

A. Variation in Hydrogen Content

H-cubes / U-cubes	Atoms H / Atoms U-235	Critical mass U-235 kg.		
		Experimental	Calculated	
			Latticed	Homogeneous
1/2:1*	5	25.7	32.2	30.1
1:1	10	15.8	16.4	14.1
2:1	20	9.7	9.68	6.9
4:1	40	8.2	7.78	3.53
7:1	70	9.9	11.05	2.15

* Low H content in this case renders assumption invalid and results undependable.

B. Variation in Lattice Arrangement

H-cubes / U-cubes	Atoms H / Atoms U-235	Critical mass U-235 kg	
		Experimental	Calculated Latticed
Homogeneous	10	—	14.1
1:1	10	15.8	16.4
2:2	10	16.3	17.6
4:4	10	17.1	19.2
8:8	10	20.1	22.0

The calculated and experimental values of critical mass for a 1:1 cube ratio arranged in various configurations which are listed in Table 5B differ in some cases by several percent. The calculated values of the critical mass for lattice configurations when plotted on the arbitrary inhomogeniety scale mentioned above, i.e., 0, 1, 2, 4 and 8 for the homogeneous, 1:1, 2:2, 4:4 and 8:8 lattice arrangements, respectively, define a smooth curve. One would expect that experimentally, the unsymmetrical arrangements 2:2 and 4:4 (lattice configurations L and K, Fig. 15) would give lower values of the critical mass than would be obtained with elements cubical in shape but of the same volume. Extrapolation of the experimental values to the homogeneous case gives, systematically, too high a value of the critical mass. Hence the calculated value of 14.1 kg for critical mass of uniformly mixed material in 1:1 volume ratio is believed more reliable than the 15kg value which is indicated in Fig. 7 by the extrapolation of the experimental data to zero inhomogeniety.

In other words, the experimental value, 15.8 kg for the critical mass of the 1:1 H to U cube array appears to be about 13% higher than the critical mass, 14 kg, which would have been obtained had the components of the array been intimately mixed.

(C) Effect of a Shield or Reflector on Critical Mass

A series of experiments was designed and performed with the object of determining the effect on critical mass of different materials around an assembly of fissionable material. In all cases, a 1:1 ratio of H to U cubes was used, in cubical configuration, surrounded by the shielding material being investigated. Except in the "untamped" case, the shielding material was surrounded in turn by 7" of paraffin.

The details of the arrangement and the results obtained are presented in Table 6.

TABLE 6

EFFECT OF REFLECTOR ON CRITICAL MASS

(a) 1:1 ratio of H-cubes to U-cubes

(b) Cubical assembly

(c) Lattice configuration B

(d) H to U-235 atomic ratio = 10

(e) Overall density = 2.8 gm/cm^3

Shielding	Paraffin	Paraffin Outside 1/4" Plywood	Paraffin Outside 3/4"Boron Plastic* 0.124 gm B/cm^2	Paraffin Outside 0.356 gm Cd/cm^2	Paraffin Outside 1.14 gm B/cm^2 as B_4C	Air (untamped)
Dimensions (cube units)	9x9x8	9x9x8½	11x11x10-	11x11x10½	11x11x10½	12x12x12-
H-cubes	324	342	583	619	651	844
U-cubes	324	342	583	619	651	844
Total cubes	648	684	1166	1238	1302	1688
Critical mass U, kg	16.6	17.5	29.8	31.7	33.3	43.2
Critical mass U-235, kg	15.8	16.7	28.4	30.2	31.7	41.2

*4.3% boron by weight (in form of $Na_2B_4O_7 \cdot 10 H_2O$), 34 1/2% cellulose acetate, 25% polystyrene; density 1.6 gm/cm^3.

From these data, it is seen that, of the shielding or reflector materials investigated, paraffin produces the lowest critical mass, and no reflector at all (air only) results in the highest critical mass.

Use of a boron impregnated plastic, cadmium sheet and boron carbide powder results in critical masses approximately double the paraffin tamped critical mass. The small increase in critical mass due to use of paraffin surrounded plywood is probably due in large part to the spacing between the assembly and the paraffin caused by the presence of the plywood. (The plywood was used to help support the early assemblies; it was not used after the discovery that its presence had an appreciable effect on the critical mass.)

(D) Effect of Configuration of Assembly on Critical Mass

With other conditions fixed, the smallest critical mass is obtained when a given assembly has a spherical shape since the ratio of surface (through which neutrons can escape) to volume is smaller in this case than in any other. For practical reasons, it would be desirable to know the relation between critical mass in a spherical shape to that in a cylindrical shape. Experimentally, efforts were directed towards a comparison of critical masses when the material was arranged in a cube and in a long rectangular parallelepiped. Investigation was made at each of the two H-cube to U-cube ratios. The results are given in Table 7 and Fig. 8.

TABLE 7

EFFECT OF GEOMETRIC SHAPE ON CRITICAL MASS

A- 1:1 ratio of H-cubes to U-cubes

(a) Lattice configuration B

(b) H to U-235 atomic ratio = 10

(c) Overall density = 2.8 gm/cm^3

(d) Completely paraffin enclosed

Dimensions, cube units	9x9x8	7x7x16½	6x7x27
H-cubes	324	394	567
U-cubes	324	394	567
Total cubes	648	788	1134
Critical mass U, kg	16.6	20.2	29.0
Critical mass U-235, kg	15.8	19.2	27.6

B - 2:1 ratio of H-cubes to U-cubes

(a) Lattice configuration E

(b) H to U-235 atomic ratio = 20

(c) Overall density = 2.2 gm/cm^3

(d) Paraffin reflector

Dimensions (cube units)	9x8x8½	7x7x14½	6x7x18½	6x6x53*
H-cubes	396	462	510	1272
U-cubes	198	231	255	636
Total cubes	594	693	765	1908
Critical mass U, kg	10.2	11.9	13.0	[32.5*
Critical mass U-235, kg	9.7	11.3	12.4	31.0*]

* This assembly containing 31 kg U-235, was not critical and apparently would not have become so even at infinite length.

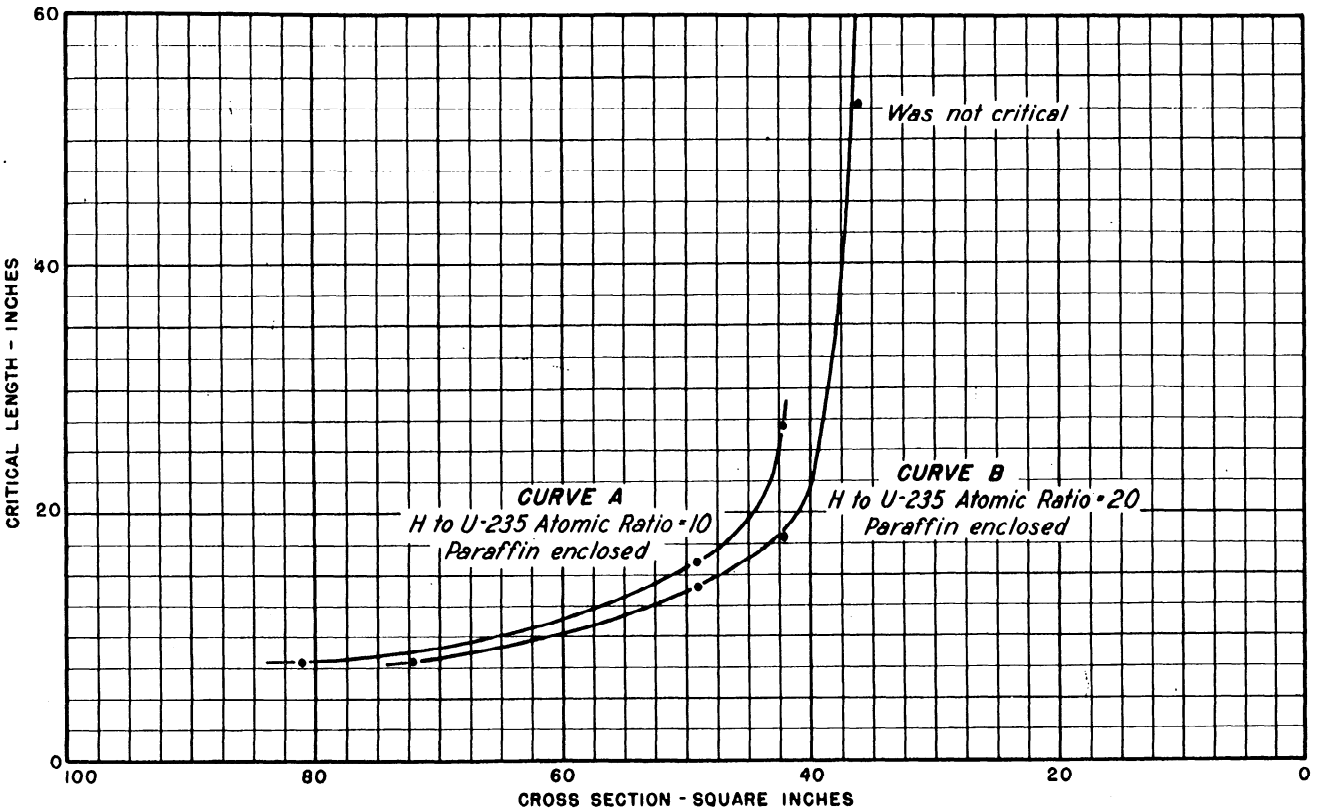


FIGURE 8

EFFECT OF GEOMETRY ON CRITICAL DIMENSIONS

DWG. NO. LD-A-93

From these two series of tests, it is likely that, under the stated conditions, an assembly of 6" x 6" cross-section or less could be extended indefinitely, without becoming critical, (Fig. 8). From these two cases, however, it cannot be stated as a general rule that 6" x 6" assemblies (or even 6-inch diameter circular cylinders) may be indefinitely extended without reaching criticality.

(E) Effect of Density on Critical Mass

A brief investigation was made of the effect of density on critical mass by stacking the N and U cubes in a cubical assembly with various sized air-spaces between the cubes. The arrangement is shown in Fig. 16 where it is seen that altering the amount of space between the cubes resulted in assemblies having different overall U-235 densities, though the densities of the component cubes remained unchanged. In the following summary of results, (Table 8) the proportions of the respective components of an assembly are express as the ratio: Number of one-inch N-cubes: number of one-inch U-cubes: number of one-inch air spaces.

TABLE 8

EFFECT OF DENSITY ON CRITICAL MASS

A. 1:1 ratio of H-cubes to U-cubes

(a) Completely paraffin enclosed

(b) H to U-235 atomic ratio = 10

(c) Cubical assembly

Volume Ratio of Materials	H:U:A 1:1:0	H:U:A 1:1:1/2	H:U:A 1:1:1
Lattice configuration	B	N	M
Dimensions (cube units)	9x9x8	10x10x12	12x13x13
Density, gm U-235/cm^3 (ρ)	1.39	1.15	0.96
H-cubes	324	464	676
U-cubes	324	464	676
Air spaces (cube units)	0	232	676
Total "cubes" (including air)	648	1160	2028
Critical mass U, kg	16.6	23.7	34.6
Critical mass U-235, kg (M_c)	15.8	22.6	33.0
x*	—	1.54	1.58**

*$M_c \propto (\rho)^{-x}$; the values of x are determined by comparison with the 1:1:0 array.

** Assembly stacked in plywood box and yields this value when compared with 1:1:0 assembly in plywood box.

B. 2:1 ratio of H-cubes to U-cubes

(a) Completely paraffin enclosed

(b) H to U-235 atomic ratio = 20

(c) Cubical assembly

Volume Ratio of Materials	H:U:A 2:1:0	H:U:A 2:1:1/2	H:U:A 2:1:1
Lattice configuration	I	P	O
Dimensions (cube units)	9x9x8+	10x10x9+	11x11x12
Density gm U-235/cm^3 (ρ)	1.00	0.84	0.71
H-cubes	396	538	726
U-cubes	198	269	363
Air spaces (cube units)	0	135	363
Total "cubes" (including air)	594	942	1452
Critical mass U, kg	10.2	13.8	18.6
Critical mass U-235, kg (M_c)	9.7	13.1	17.7
κ^*	---	1.80	1.87

$M_c \propto (\rho)^{-\kappa}$

Dr. R.D. Feynman[3] has pointed out that, by the following argument, the critical mass, in the ideal case, would be expected to vary inversely as the square of the density:

(3) Private Communication

(a) Assume an arbitrarily tamped assembly of fissionable material which is exactly critical. All the characteristics of this assembly, mean free path, cross-sections, etc., are so adjusted that criticality just obtains.

(b) If all linear dimensions are of the above assembly (of both active material and reflector) are doubled, with no change in the total masses involved, the only effect would be doubling of the neutron mean free path everywhere in the assembly. Hence the assembly would be exactly onehalf critical.

(c) If now in the enlarged assembly, one additional atom is added for every one present, in core and reflector, i.e., the total mass is double the original, then the mean free path is reduced to the original value and the assembly is again exactly critical. The mass has been doubled; the density decreased to one-fourth; hence, the mass is inversely proportional to the density squared.

In actual cases, in which only the core density is varied, it would be expected that greatest deviations from this "inverse square law" would be found in assemblies relatively largely affected by the reflector, i.e., small unmoderated assemblies. Here, one would expect the numerical value of the negative exponent of density to be somewhat less than 2. Large well-moderated assemblies would be expected to follow more nearly the inverse-square rule.

The results obtained from the experiments described above are in agreement with these arguments: in the 1:1 arrangement, critical mass varies inversely approximately with the 1.6 power of density, in the 2:1 arrangement with the 1.8 power. There are too few data, however, to insure a complete verification of the argument.

(F) Critical Mass by Extrapolation of Neutron Multiplication

As pointed out earlier, it should be possible to measure the neutron multiplication for sub-critical assemblies at a few different values of k and from those data obtain an estimate of the mass which would become critical. In several experiments, it was found that an insufficient quantity of U-235 was available to reach criticality under the conditions of the experiments. In these cases, attempts were made to obtain an estimate of critical mass by neutron multiplication. In one experiment, a comparison was made between the values of a critical mass obtained by direct measurement in a critical assembly and by neutron multiplication measurement on sub-critical assemblies. These experiments are described below.

1. Comparison of Directly Measured and Extrapolated Values of Critical Mass.

It was found that a 10x10x9+ assembly of cubes in the ratio 1H:2U, surrounded by paraffin, became critical. Neutron

multiplication measurements were made on similar 9x9x9, 8x8x8, and 7x7x7 assemblies of cubes arranged under the same conditions and with a neutron source at the center. A "background" count was made for each assembly by placing the source in the same position that it occupied during the test but with the active material removed. The ratio of the counting rates with and without the active material present is the multiplication. By direct measurement, a critical mass of 29.4 kg of U-235 was obtained; extrapolation of neutron multiplication measurements on the sub-critical assemblies gave a value of 28 kg.

The pertinent data are listed in Table 9, and the graph of neutron multiplication, extrapolated to criticality, is shown in Fig.9.

TABLE 9

COMPARISON OF VALUE OF CRITICAL MASS DIRECTLY DETERMINED WITH THAT OBTAINED FROM EXTRAPOLATION OF NEUTRON MULTIPLICATION DATA

(a) Cubical assembly

(b) Completely paraffin enclosed

(c) H to U-235 atomic ratio = 5

(d) Lattice configuration C, Fig. 13

Dimensions (cube units)	10x10x9+	9x9x9	8x8x8	7x7x7
H-cubes	301	242	170	112
U-cubes	602	486	341	230
Total cubes	903	728	511	342
Counts/min	-	1410	556	386
Background, counts/min	-	281	287	294
$(\text{Multiplication})^{-1}$	-	0.199	0.516	0.762
Mass U-235, kg	29.4	23.7	16.6	11.2
Critical Mass U-235, kg	29.4 (Experimental)	←	28	→ (Extrapolated)

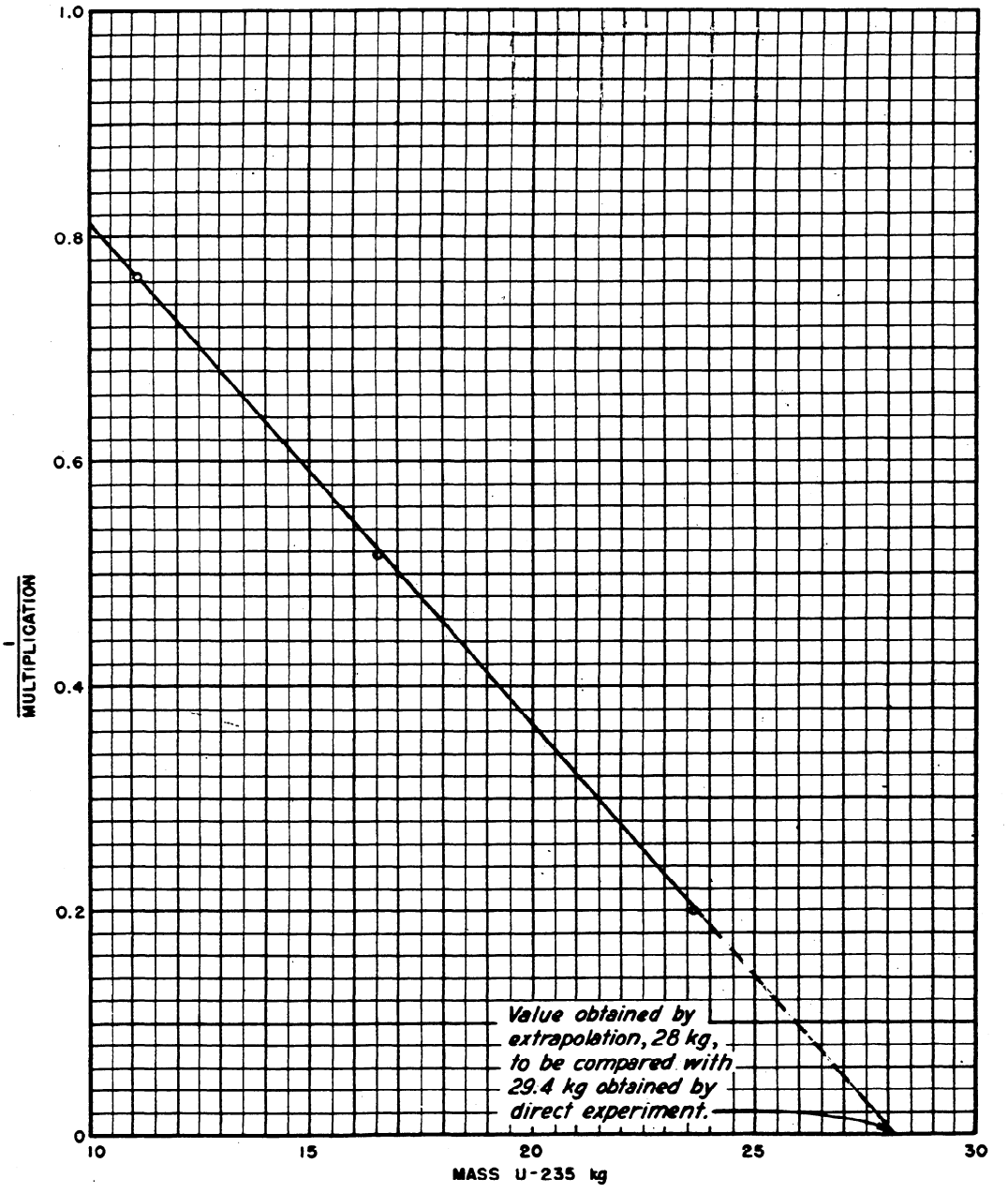


FIGURE 9

INVESTIGATION OF VALIDITY OF MULTIPLICATION METHOD OF CRITICAL MASS DETERMINATION

DWG. NO. LD-A-94

2. Unmoderated, Paraffin Enclosed Assembly

With no internal hydrogen, but complete enclosure in paraffin, a cubical assembly of the total amount of material available (54 kg U-235) was far from critical. Neutron multiplication measurements were made on assemblies in which the U-235 mass was varied up to the maximum quantity available, and an estimate of critical mass was obtained by extrapolation of the data, (Table 10A, and Fig. 10). The estimate of critical mass thus obtained, about 100 kg U-235, is quite uncertain. The true value may be 10% lower, or 25% or more higher. It is simply very clear from the low values of neutron multiplication that the maximum accumulation of 54 kg U-235 under these conditions, was far below critical.

3. Moderated, Untamped Assembly

The experiment described above was repeated with no paraffin around the assembly. The degree of criticality, with all available material in the assembly was less than in the former case, hence the estimate of critical mass obtained by extrapolation of neutron multiplications is even more unreliable, (Table 10-B, Fig. 11).

TABLE 10

CRITICAL MASS DETERMINATION FROM EXTRAPOLATION OF NEUTRON MULTIPLICATION DATA

A. Fully paraffin enclosed

(a) No intermixed hydrogen

(b) Cubical assembly

(c) Lattice configuration A

Dimensions (cube units)	10x10x10	9x9x9	8x8x8
U-cubes	1000	729	512
Mass U-235, kg	48.8	35.6	25.0
Counts/min	488	406	313
Background counts/min	253	245	238
$(\text{Multiplication})^{-1}$	0.518	0.603	0.760
Critical mass U-235, kg	100	(highly uncertain value obtained by extrapolation)	

B. Air reflector only

(a) No intermixed hydrogen

(b) Cubical assembly

(c) Lattice configuration A

Dimensions (cube units)	10x10x10	9x9x9	8x8x8
U-cubes	1000	729	512
Mass U-235, kg	48.8	35.6	25.0
Counts/min	5005	4429	3917
Background counts/min	← 2470 →		
$(\text{Multiplication})^{-1}$	0.494	0.558	0.631
Critical mass U-235, kg	130 (highly uncertain value obtained by extrapolation)		

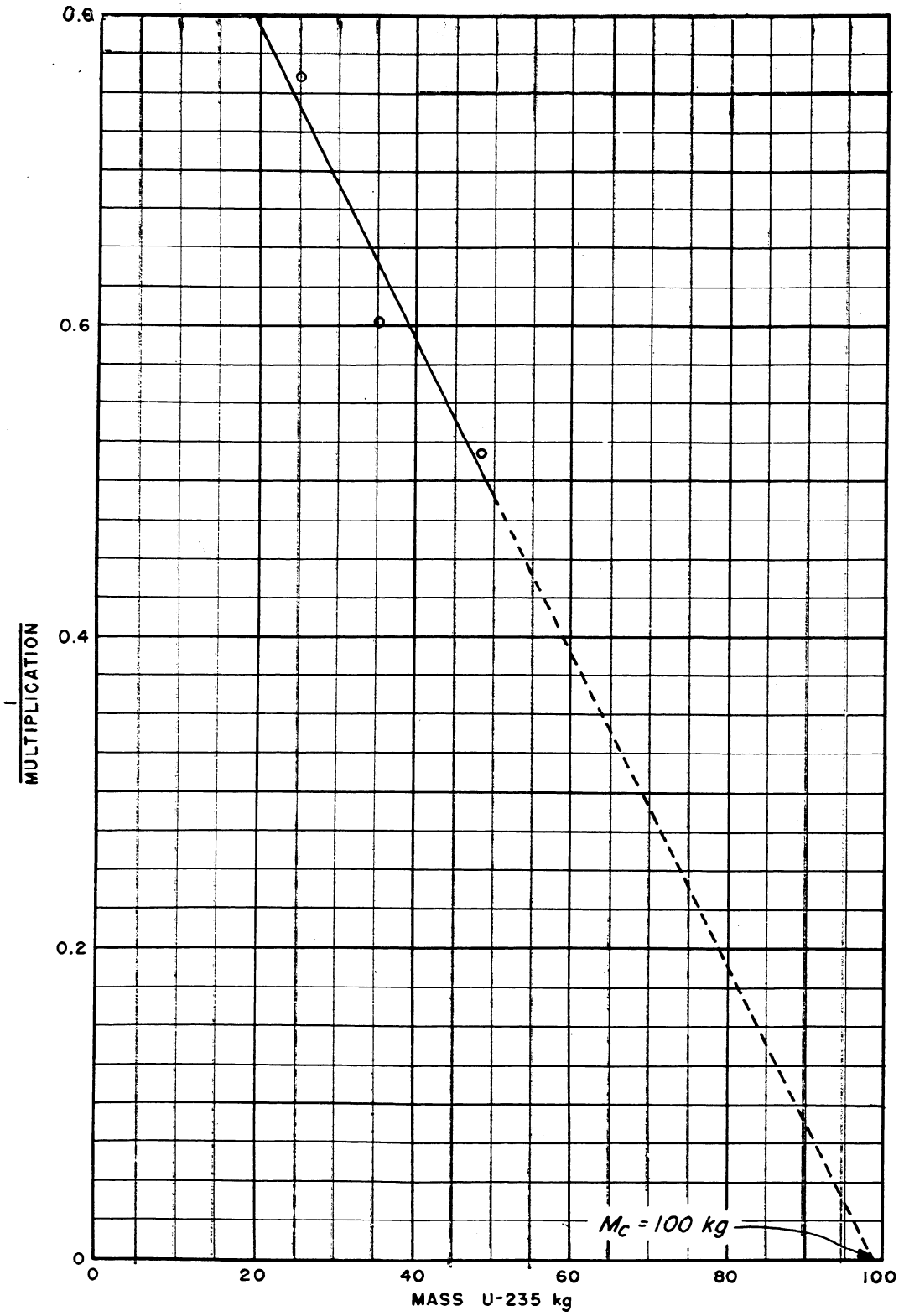


FIGURE 10

APPROXIMATION OF CRITICAL MASS OF UNMODERATED CUBICAL ASSEMBLY ENCLOSED IN PARAFFIN

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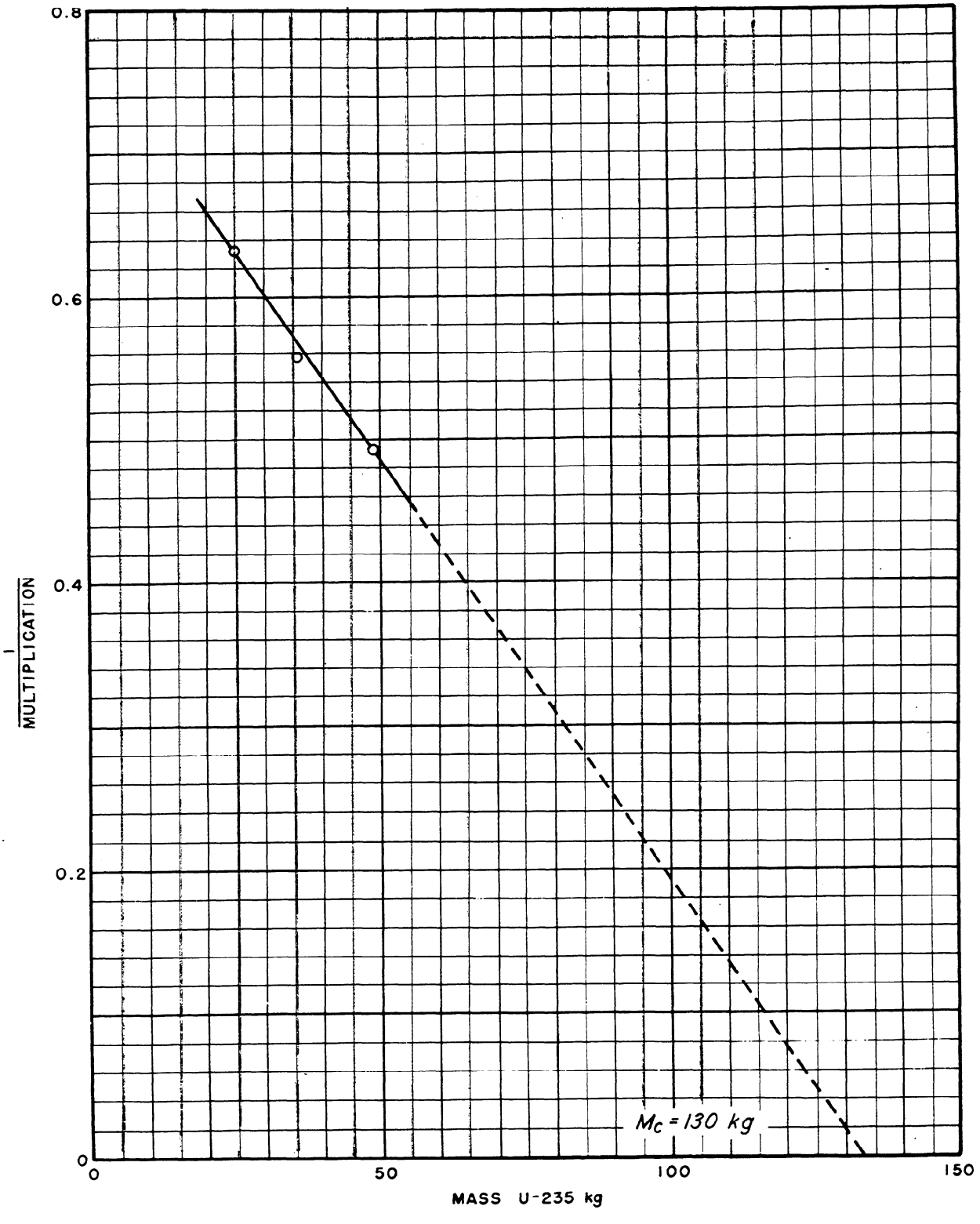

FIGURE 11

APPROXIMATION OF CRITICAL MASS OF UNMODERATED CUBICAL ASSEMBLY WITH NO REFLECTOR

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VIII. SUMMARY

The complete summary of results obtained in the experiments is best shown by a graph of critical mass as a function of the H to U-235 atomic ratio. In Fig. 12, where such a graph is presented, as much description as possible of the experimental conditions is given beside each plotted point. Curves have been drawn showing how the critical mass of bare and paraffin enclosed latticed assemblies varies with the amount of intermixed hydrogen. A curve has also been drawn through the calculated critical mass values of homogeneous, paraffin enclosed arrays. Only two experimental points on the bare assembly curve were definitely established. The values of the critical mass for UW moderated assemblies either with or without reflector are highly uncertain.

IX. ACKNOWLEDGMENTS

Dr. N.E. Bradbury, Dr. E.R. Jette and other members of the staff at Los Alamos were very cooperative in setting up this program. Acknowledgment is made of the generous assistance given by Dr. Louis Slotin, also of the Los Alamos Laboratory, in directing these experimental procedures and in the training of the Oak Ridge personnel. The preparation of the materials for the experiments and their performance was greatly aided by the efforts of Mr. H.F. Priest, Mr. Joe Schaffner, Mr. S.D. Snyder and Mr. W.G. Kirby of Carbide and Carbon Chemicals Corporation, and Mr. James Taub and Mr. Dwight Young of Los Alamos.

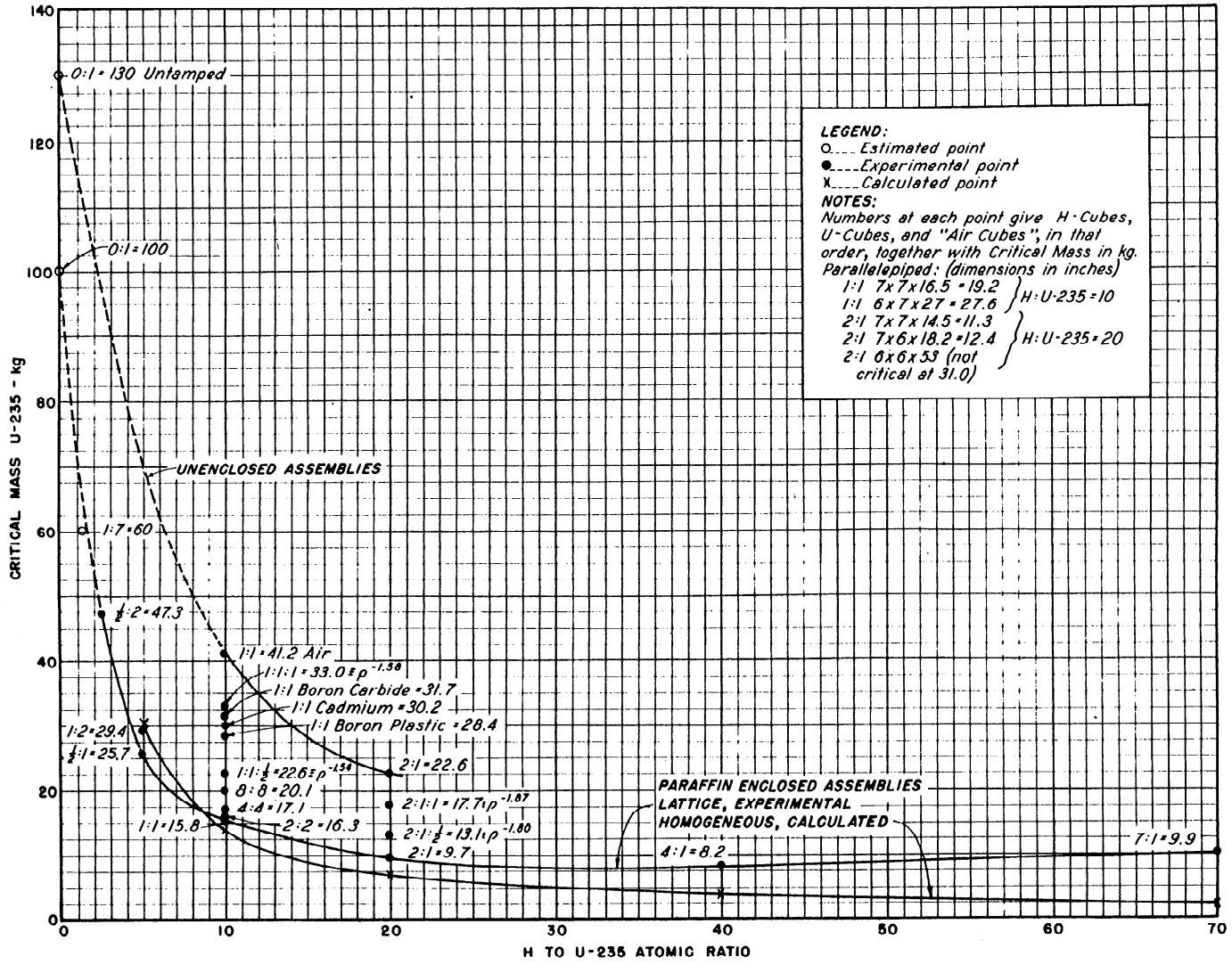
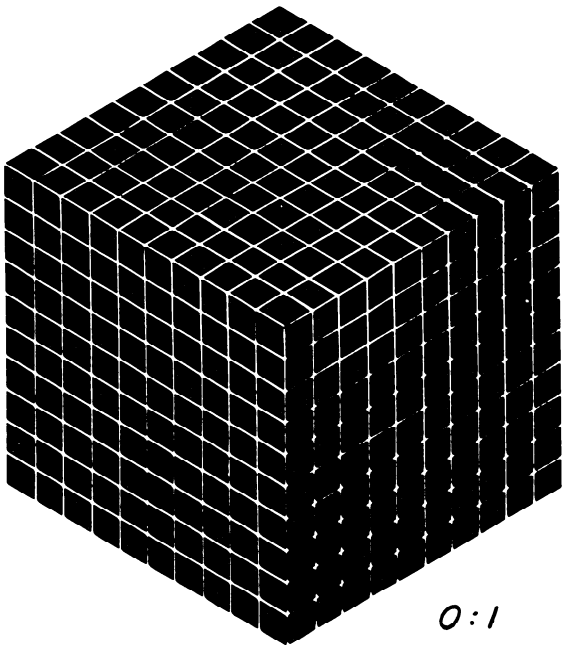


FIGURE 12

SUMMARY OF CRITICAL MASSES OF U-235 OBTAINED UNDER THE CONDITIONS INDICATED



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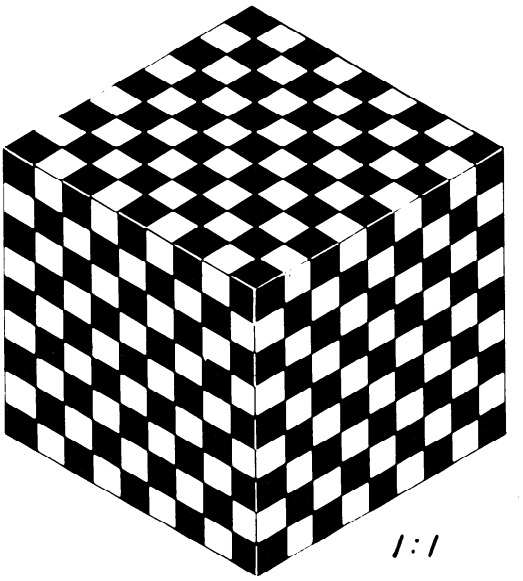

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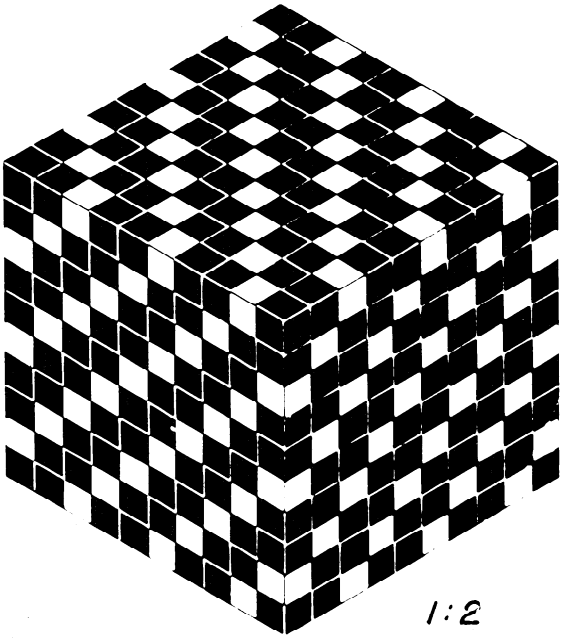

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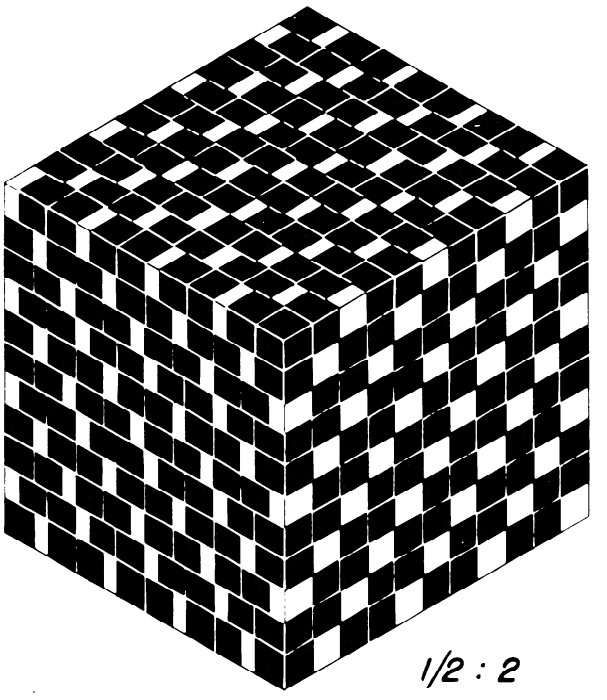

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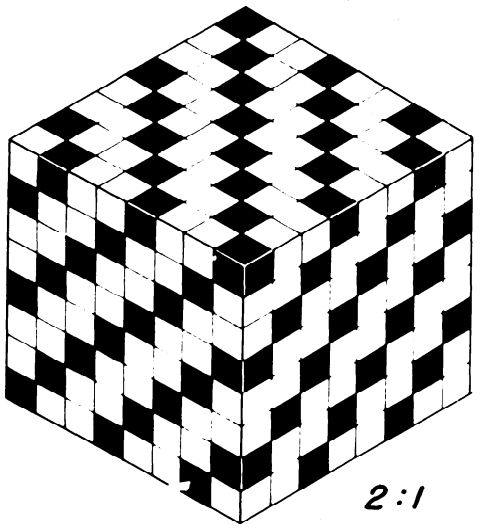
D

FIGURE 13

LATTICE CONFIGURATIONS OF
H CUBES (WHITE) AND U CUBES (BLACK)



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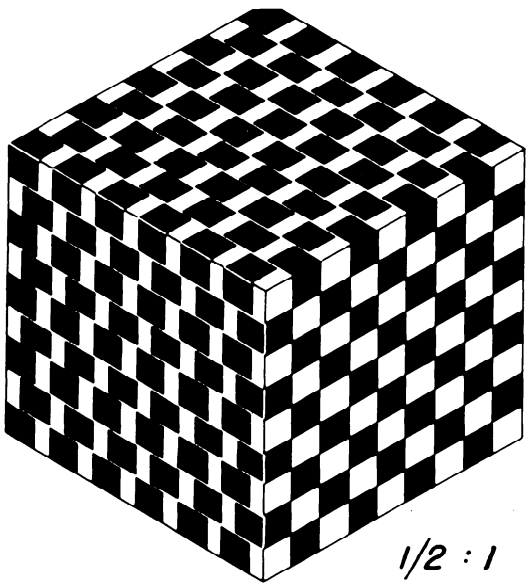

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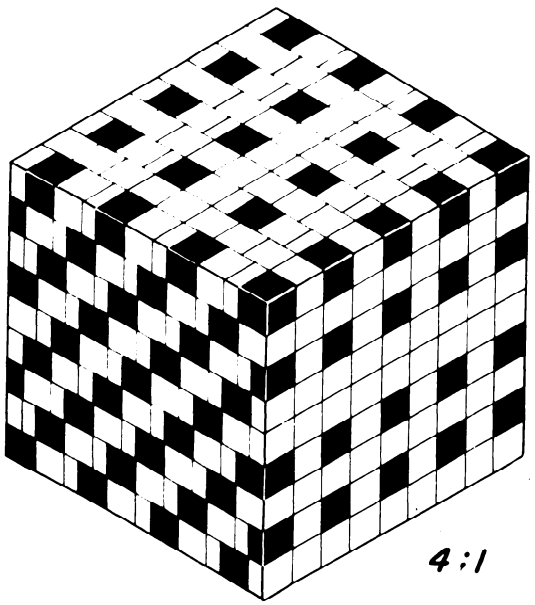

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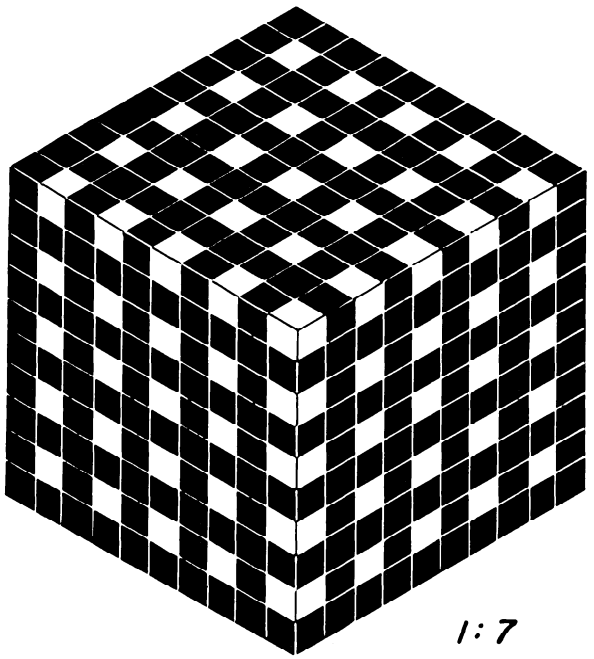

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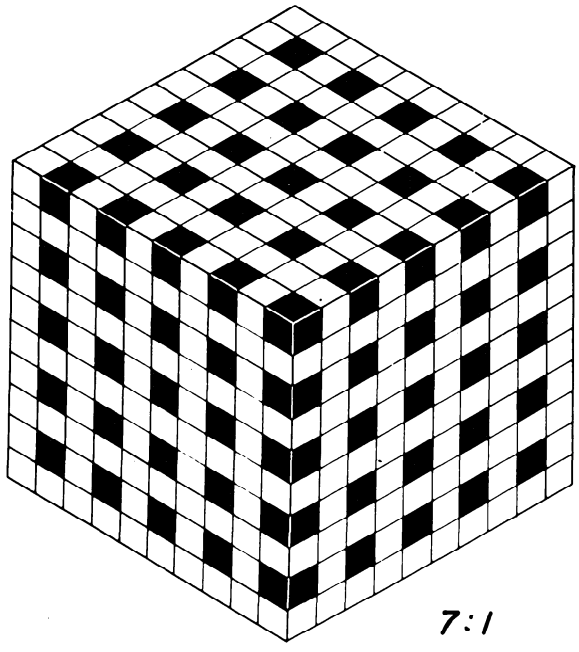
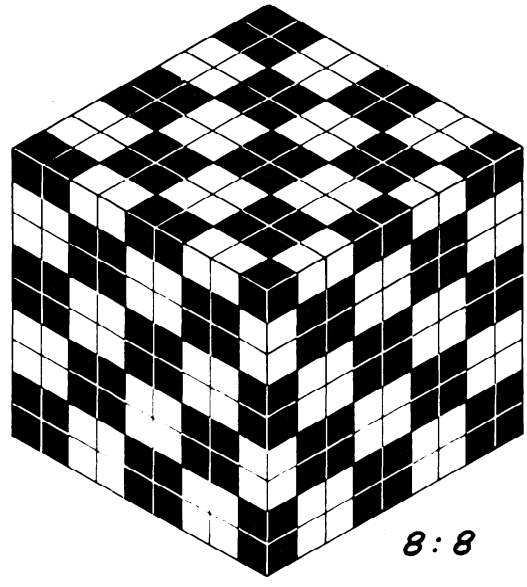
FIGURE 14

LATTICE CONFIGURATIONS OF
H CUBES (WHITE) AND U CUBES (BLACK)



I



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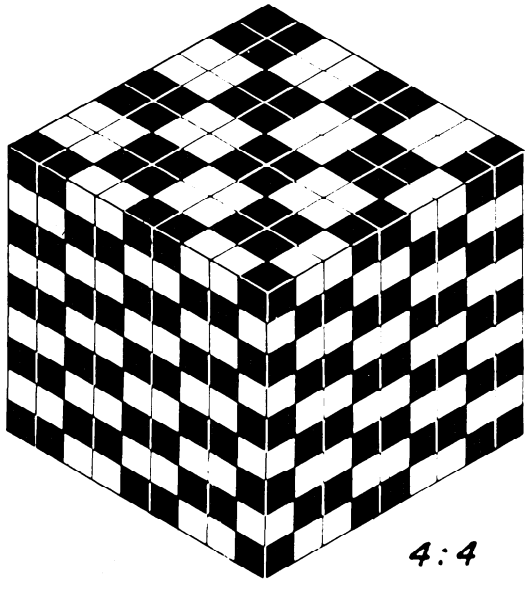
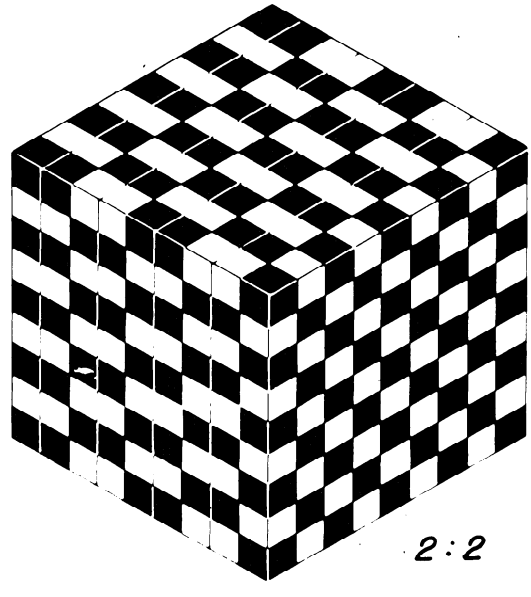


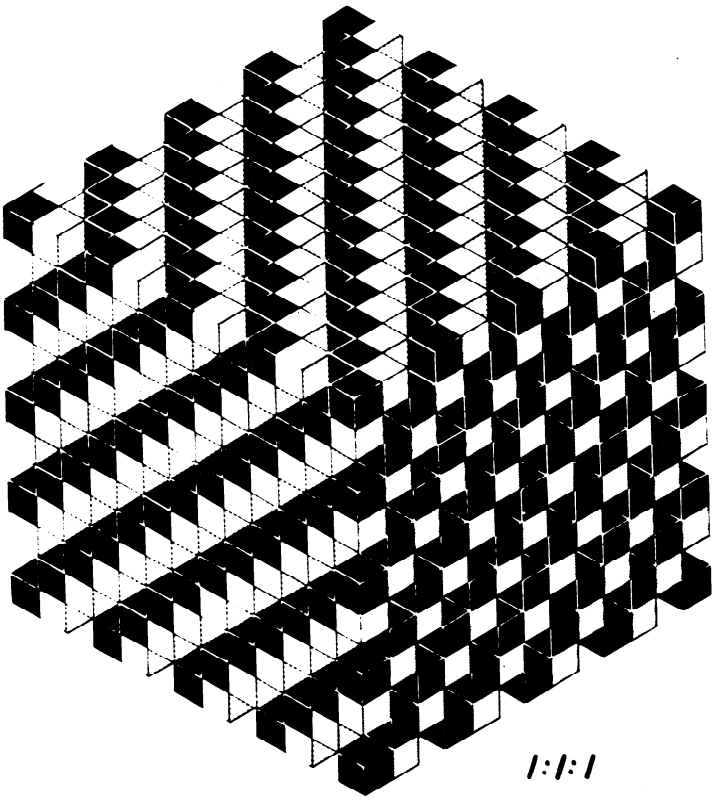
K



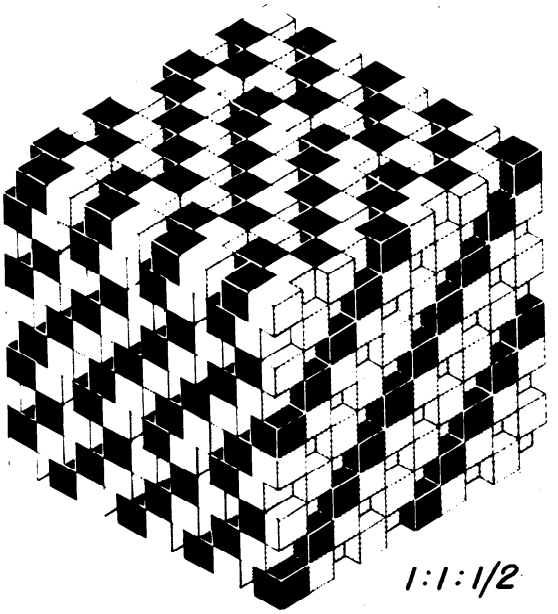
L

FIGURE 15

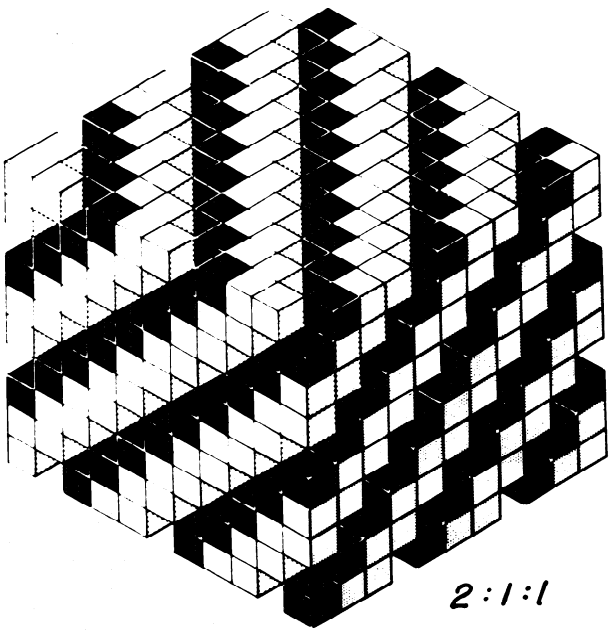
LATTICE CONFIGURATIONS OF
H CUBES (WHITE) AND U CUBES (BLACK)



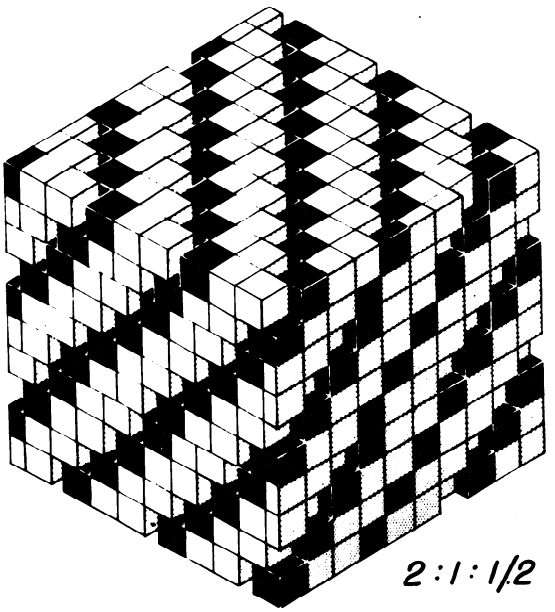
M



N



O



P

FIGURE 16

LATTICE CONFIGURATIONS OF H CUBES (WHITE), U CUBES (BLACK), AND AIR SPACES (BLANK)