

REFERENCE CRITICAL EXPERIMENTS

Progress Report
January - March 1979

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Rockwell International

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SUMMARY

This is the fourteenth (14th) in the series of quarterly progress reports describing reference critical experiments being performed at the Critical Mass Laboratory at Rockwell International's Rocky Flats Plant for the U. S. Nuclear Regulatory Commission (NRC).

Eight critical experiments were performed using the low-enriched, damp (H/U ~ .77) uranium oxide (U₃O₈) in the concrete reflector. This concludes the interstitially-moderated experiments. Two critical experiments were repeated. One was repeated ten months after the original experiment to determine the combined effect of oxide weight gain and statistical error of measurements. The second repeat experiment was performed a few hours after the original experiment, so oxide weight gain would not be a factor, to see the effect on critical results of restacking and re-arranging the oxide cans.

Impurity content and particle size distribution of the oxide were determined by laboratory analysis.

Water has been added to the uranium oxide to increase the H/U atomic ratio from 0.77 to 1.25. The uranium oxide with an H/U ratio of 1.25 will be used in the next series of experiments.

Reweighing of the oxide cans indicates that the oxide continues to gain weight.

CRITICAL EXPERIMENTS AT ROCKY FLATS

Introduction to Critical Experiments

All experiments were performed on the horizontal split table, consisting of two halves which move toward each other. Each half of the table supports a portion of the oxide array and concrete reflector. The table is slowly closed until criticality is reached for the experimental array of oxide cans. A more detailed description of the horizontal split table is given in References 1, 2, and 3.

The uranium oxide (U_3O_8) is enriched to 4.46 wt-% ^{235}U and is packed in cubic aluminum cans 15.3 cm on each side. Each filled can weighs ~ 16 kg, and the oxide has a density of 4.68 g/cm^3 . Water was added to the oxide to achieve an H/U atomic ratio of 0.77.

The concrete reflector and oxide cans are the same ones used for an earlier program (Reference 4). The reflector consists of two "frames" plus two end reflector panels. One of each is shown in Figure 1. When the four are assembled and the table closed, a nearly cubical hollow shell, ~ 1.3 m outside and ~ 0.8 m inside, is formed. Precise dimensions for each critical configuration are given later in this report.

Figure 1

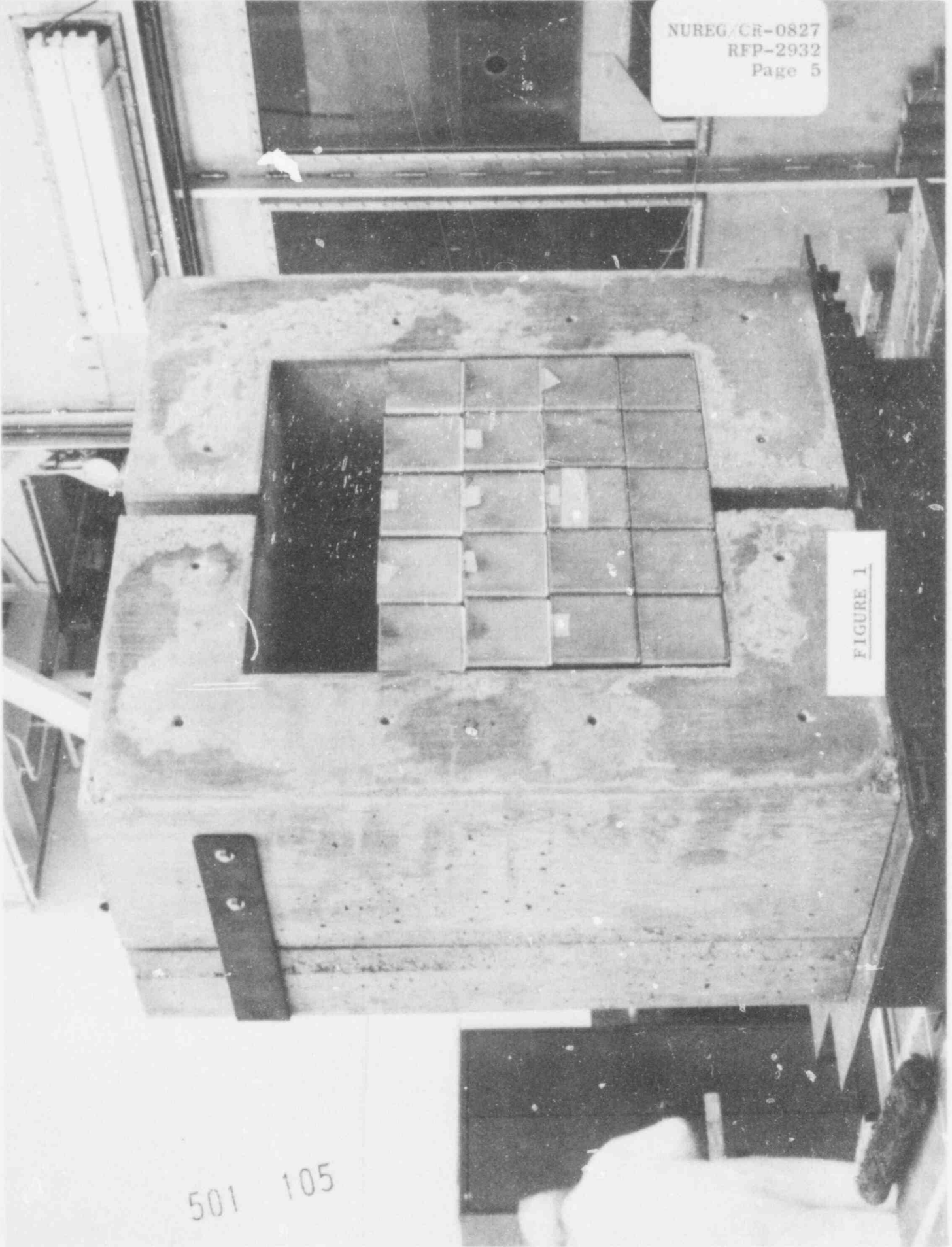
The south "frame" and its end reflector panel are shown assembled on the south portion of the horizontal split table (table closure is in the north-south direction). The cans shown inside the reflector are empty and do not correspond to any experimental array studied. The rectangular slots in the top and bottom of the reflector frame provide clearance for neutron source removal and the metal driver support rod, respectively.

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FIGURE 1

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Interstitially-Moderated Critical Experiments

The arrays studied consisted of periodic arrangements of oxide-filled aluminum cans surrounded, in some cases, by thin sheets of neutron-absorbing materials on all six faces. In other cases, these neutron absorbers were omitted. Pieces of methyl methacrylate plastic (Plexiglas®) were inserted between the (unsheathed or absorber-sheathed) oxide cans as interstitial moderating material. There was no plastic between the cans and the reflector. Whichever materials were used in an array, the repeated laminations of these materials across the array were always pressed tightly against the floor and west wall of the concrete frames on both halves of the split table. This eliminated any gap between elements of the array and these surfaces. The elements of the array were positioned on both halves of the table such that the periodic arrangement of materials would be preserved, as precisely as possible, at the moment the two halves of the table were closed. After a core was assembled, any space between the array and the east wall of the reflector frame was filled with cuboidal pieces of concrete cast especially for that purpose. Several of the photographs later will show these pieces in position. Vacant space above an array was not filled with concrete because of the heavy weight which would be placed on the oxide cans. Instead, any array expected prior to assembly not to require the full vertical height of the reflector

shell was built on thick concrete blocks also cast especially for that purpose. This procedure minimized, but did not eliminate, the top gap between the array and the reflector frame. Similarly, cast concrete "plugs", just slightly smaller than the rectangular opening in the frame, were placed between the array and the reflector's end panel to minimize the gap to the reflector in this direction. Such plugs were used on the larger south portion of the reflector frame only. This was so because the array cans plus their interstitial moderator and absorber materials were already oversized (a few millimeters) for the smaller north portion of the reflector. This problem was solved by separating the north reflector end panel from the reflector frame a suitable distance, creating a gap a few millimeters wide in the nearly cubical reflector shell. This gap was filled, whenever it existed, by transite panels of suitable size to form an extension to the north reflector frame. Transite is a hard material composed of asbestos and Portland cement. The composition of this material and the amount used in different experiments is given in a later section.

Seven concrete-reflected critical cases are reported, and these spanned four different combinations of interstitial moderators between the cans and thin sheets of neutron absorbers surrounding the cans. These data complete the planned program for such experiments in both concrete and plastic reflector frames.

One combination studied had 2.4-cm-thick plastic between adjacent cans and no other material (e.g. strong neutron absorber) within the array. Figure 2 shows this combination looking at the south face of the split table. This is the optimum moderator thickness and is analogous to "category 1" as discussed for the plastic reflector in Reference 2. This configuration was taken to criticality twice in one day, the difference between the two being that the forty cans were reassembled in different locations (see Repeat Critical Moderated Experiment section of this report). Both experiments had identical array shapes, a two-layer-high assembly of sixteen cans per layer and a top layer of eight cans, for a total of forty cans. The sixteen cans were divided into identical 2 x 4 arrays on each half of the table, forming a 4 x 4 array per layer whenever the table became fully closed. The top layer was incomplete, missing the north and the south row of four cans each. The critical table separation for the first experiment was 0.57 cm, and that for the reassembled array was 0.62 cm.

Another combination of materials studied used the same thickness plastic between each can, but each can was further sheathed on all faces by 0.054-cm-thick sheets of polyvinylchloride (see Figure 3). This is analogous to "category 4" as used in Reference 2. The neutron absorption added by this material, principally by the chlorine, increased the size of the array needed for criticality from forty to

Figure 2

Criticality occurred with three layers of cans when the 2.4-cm-thick plastic optimum moderator thickness was used. The cans were not sheathed by any strong neutron absorbers. The array was built on concrete block set within the frame because prior calculations indicated the full height of the frame would not be required. Similarly, a pre-cast concrete piece to the left reduces the gap to the reflector in that direction. Some gaps still existed, requiring the definition of two cuboids: the core cuboid and the inner reflector cuboid.

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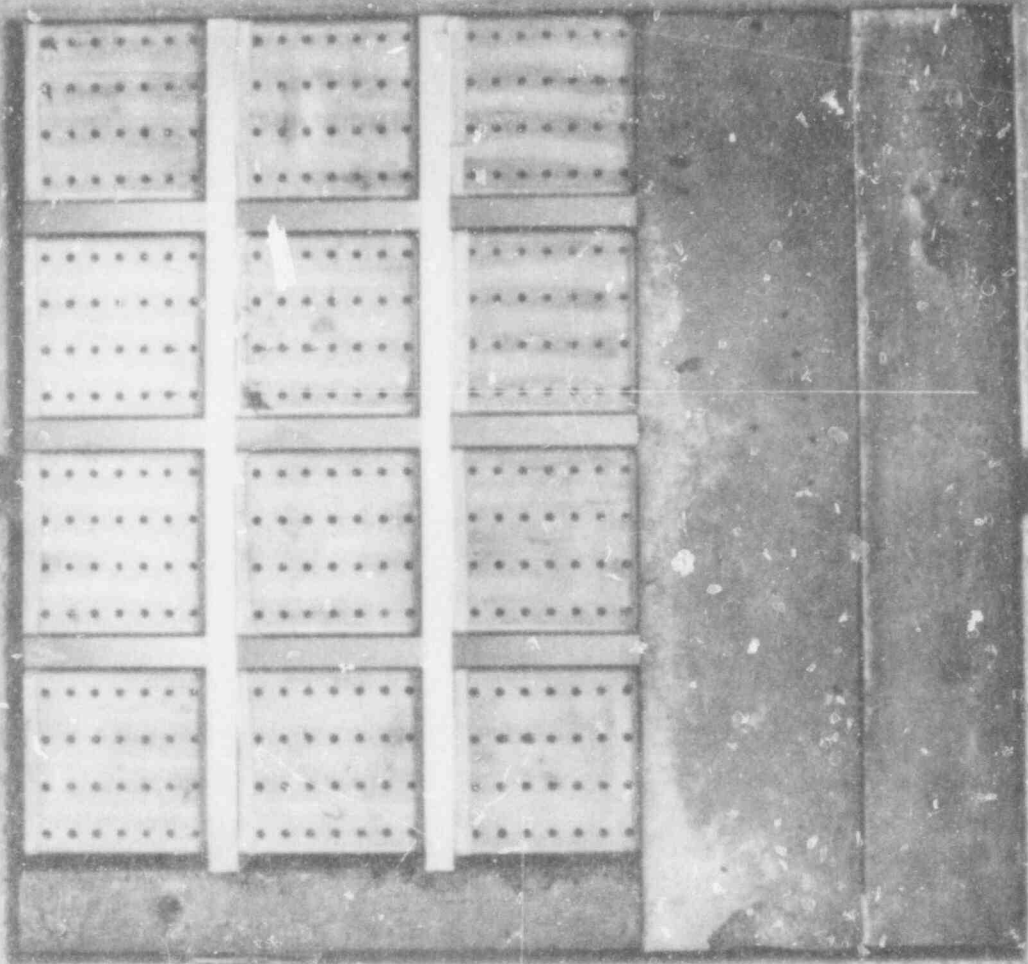


FIGURE 2

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Figure 3

The oxide cans are sheathed on all six faces with 0.054 cm polyvinylchloride sheets. Plastic (2.4-cm-thick) is used for the interstitial moderator.

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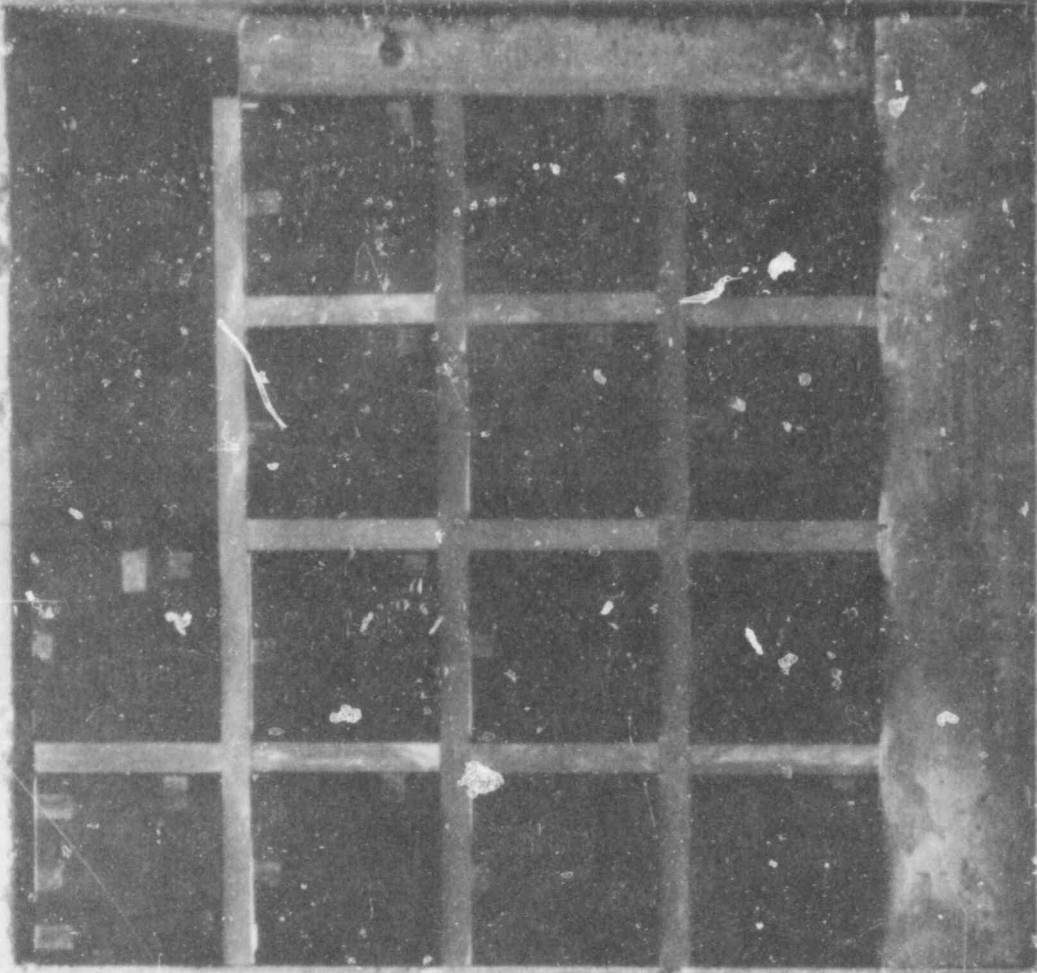


FIGURE 3

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fifty-three cans. The critical table separation was 1.05 cm. A previous table closure with fifty-two cans had also achieved criticality (0.40 cm) but, upon disassembly of the core, eight PVC pieces which should have covered the tops of the bottom layer of cans on the south table were discovered to have been inadvertently omitted.

The thin but rigid sheets of PVC material were held in place with twelve grams of ordinary office tape (Scotch[®] Magic Mending tape).

A third combination used the same 2.4-cm-thick plastic moderator. However, the cans were surrounded on all six faces by 0.117-cm-thick plates of mild steel as shown in Figure 4. This configuration is referred to as "category 2" in Reference 2. This 59-can array was so reactive that the critical table separation was 1.18 cm. One can was removed and the experiment repeated, but a positive reactor period could not be obtained. The configuration was so close to criticality that a very long negative reactor period (164 minutes) was obtained with the table closed. Nineteen grams of office tape was used to hold the steel plates in position during assembly of the 59-can array.

The final configuration involved no neutron absorber and did not have plastic between every can. Oxide cans were grouped into subarrays having no interspersed material, and then these "super-cans" (analogous to "category 7" of

Figure 4

The oxide cans are sheathed on all six faces by 0.117-cm-thick mild steel plates. The plastic moderator was 2.4-cm-thick.

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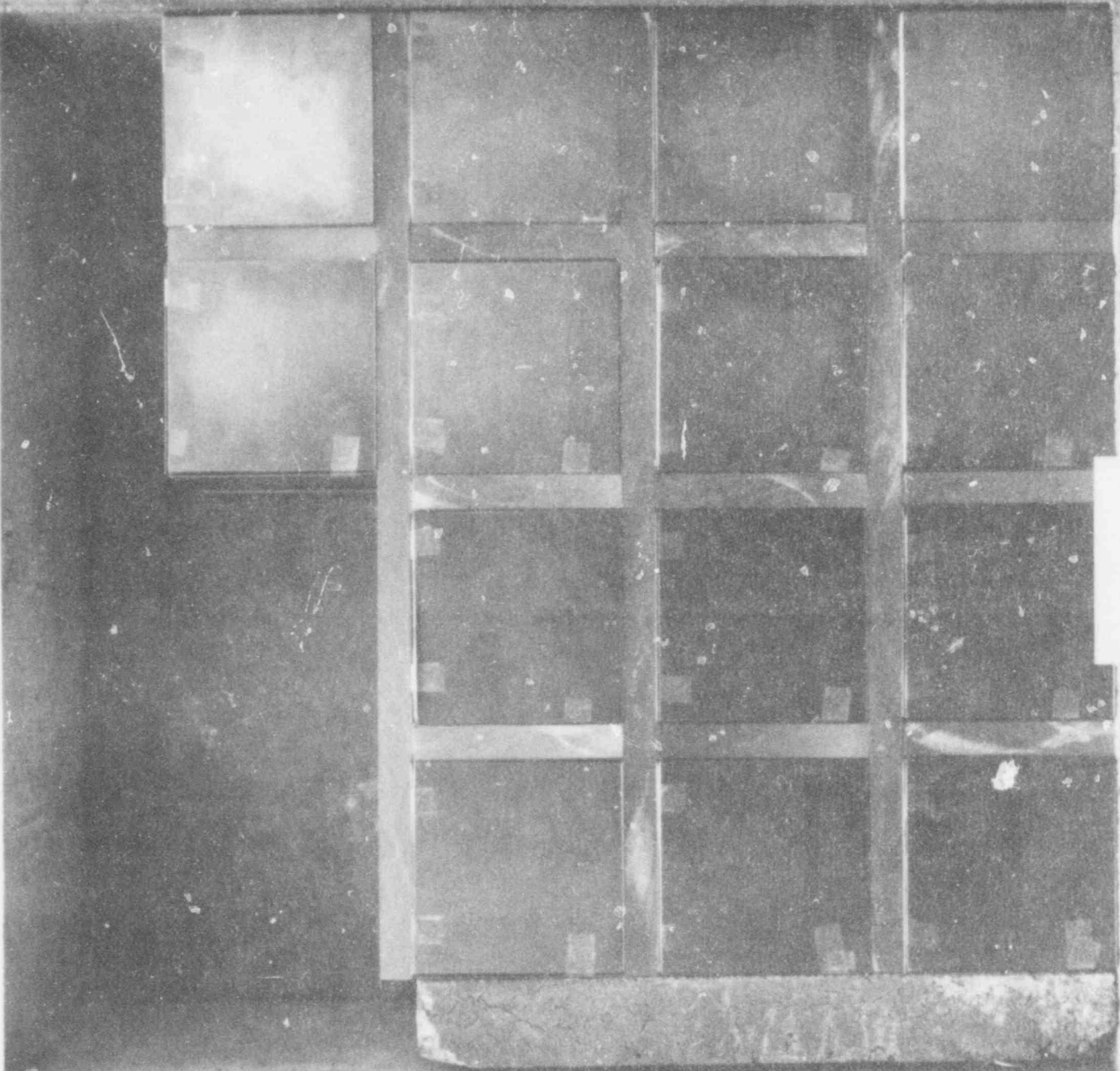


FIGURE 4

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Reference 2) were interstitially moderated by 2.5-cm-thick plastic. The specific super-can array reported here (see Figure 5) contained a $2 \times 1 \times 2\frac{1}{2}$ high array of $2 \times 2 \times 2$ super-cans on the north half of the split table. The south half of the split table was different and somewhat more complicated because the top layer was used to "shim" (by varying the number of cans) the reactor to attain criticality. The south table contained a full $2 \times 1 \times 2$ high array of $2 \times 2 \times 3$ super-cans (three cans in the direction of table movement) plus eight additional cans on the fifth layer. These eight cans were built as though they were the start of two new super-cans being built on top of the four already present on the south table. Criticality for the 96-can array (total), composed of four and two-halves $2 \times 2 \times 2$ super-cans on the north table and four and a fraction of two more $2 \times 2 \times 3$ super-cans on the south table, occurred for a table separation of 0.59 cm.

For completeness, the critical parameters of the concrete-reflected undermoderated array experiments, reported in Reference 3 and defined in Reference 2 as "category 6", are included in all appropriate tables.

Experimental parameters for nine concrete-reflected critical experiments in five categories of interstitial moderation are presented in Table I. Repeated experiments within a category are designated by letters A and B. The principal experimental parameter is the "Critical Table

Figure 5

The north table for the "super-can" configuration.

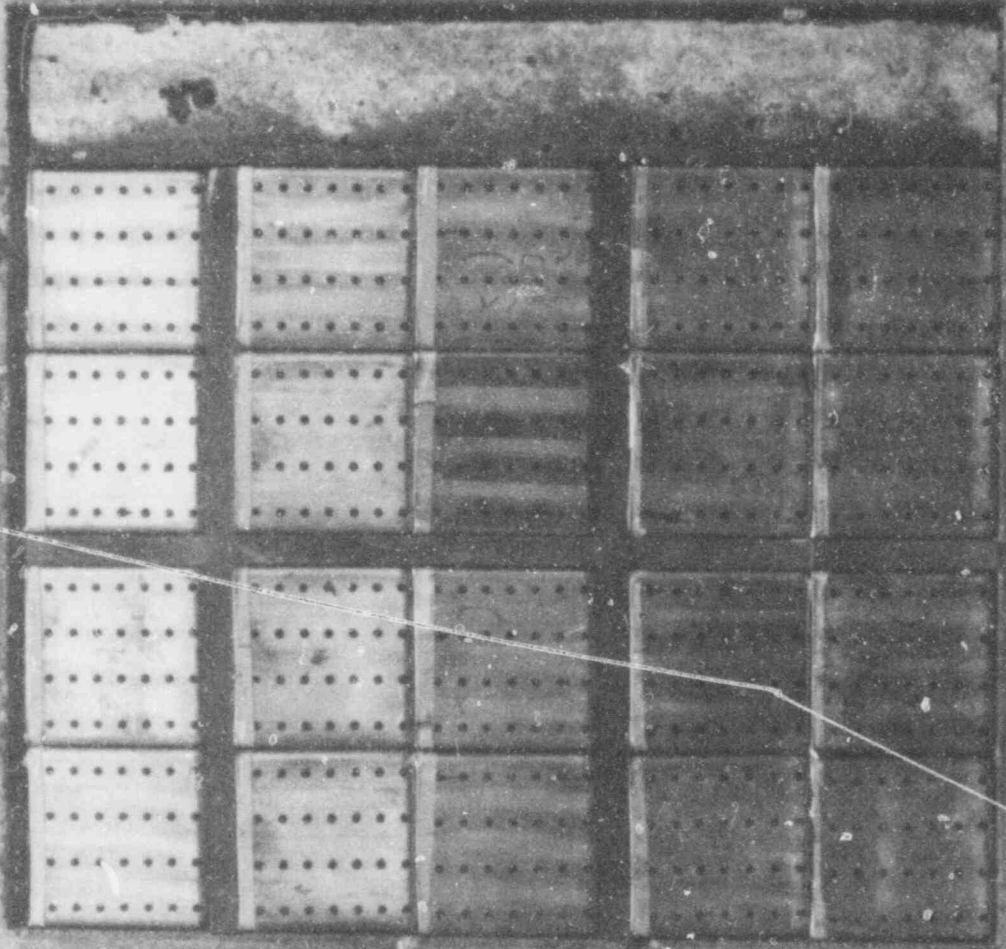


FIGURE 5

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TABLE I
Principal Critical Parameters of Nine
Interstitially-Moderated Arrays of Damp, Compacted,
Low-Enriched Uranium Oxide Reflected by Concrete

Description	Category	Target Array Size ^a	Critical Parameters			
			Number of Cans	Weight of Cans ^b (g)	Table Separation ^c (cm)	Configuration of Top Layer of Cans ^d (North) (South)
optimum moderator	1A	4 x 4 x 3	40	639,783	0.567	
optimum moderator	1B	4 x 4 x 3	40	639,783	0.617	same as above
steel sheathed	2A	4 x 4 x 4	59	943,800	1.184	
steel sheathed	2B	4 x 4 x 4	58	927,790	0.251 ^e	
polyvinyl-chloride sheathed	4A	4 x 4 x 4	53	847,819	1.047	
polyvinyl-chloride sheathed	4B	4 x 4 x 4	52	831,804	0.400	
minimal moderator	6A	4 x 4 x 5	100	1599,501	1.187	FULL
minimal moderator	6B	4 x 5 x 5	98	1567,602	0.484	
super-cans	7	4 x 5 x 5	96	1535,692	0.583	

^aThe smallest completely-filled cuboidal array predicted to have a neutron reproduction factor exceeding one with the table closed.

^bWeight of can, oxide, and injected water as of March 1, 1978. No allowance is made for the small weight gain phenomenon discussed elsewhere in the text.

^cThe separation between the two table segments at criticality averaged over the core region and the reflector region.

^dIncomplete layers for arrays from all categories here are shown as though viewed from above; hence, the figure is divided correspondingly to the two table segments.

^eNo positive reactor period achieved.

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Separation". This is an interpolated value of the average separation between table halves which would have existed had the system been precisely critical.

Figure 6 gives the reciprocal of the measured reactor periods for each case studied. This figure is used to interpolate critical table separation, that separation at which the reciprocal of the reactor period equals zero. This average critical table separation is obtained from data taken over the mating faces of the fuel region and the reflector region.

Repeat Critical Moderated Experiment

One interstitially-moderated critical configuration in the concrete reflector was repeated. The one selected was the 40-can optimum-moderated (2.4 cm plastic between cans, category 1) experiment. The purpose was to determine the effect on the critical table separation of small variations in gaps between adjacent cans resulting from small differences in can dimension and assembly technique combined with the effect of rearranging cans. The repeat experiment was done on the same day as the original experiment specifically to exclude any possible contribution due to the weight gain problem. The general philosophy for restacking was to trade the most centered cans with corner cans, and near-center cans with other cans along the edges of the original array. Critical assembly dimensions for the repeat experiment are

Figure 6

The positive and negative reactor periods can be obtained from this graph for eight of the nine cases presented in Table I. The remaining experiment never achieved a table separation yielding a positive period. The ordinate (y-axis) is the reciprocal of the reactor periods measured at the two table separations shown by dots. The line joining the two dots permits the critical table separation to be interpolated (reciprocal reactor period equals zero).

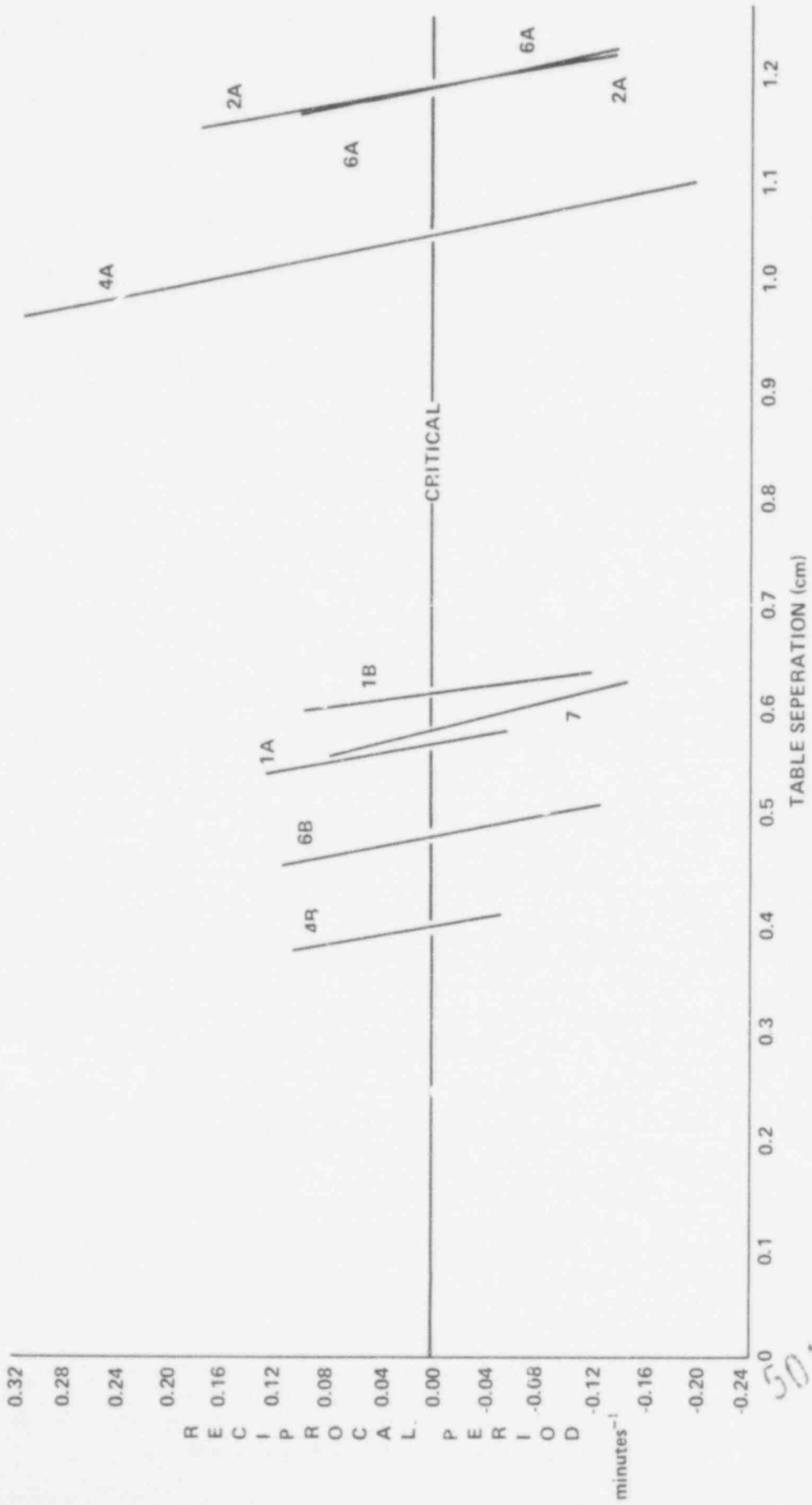


FIGURE 6

given in the Critical Assembly Dimensions section of this report. The dimensions did not vary significantly from the original experiment.

The original experiment was critical with a 0.57 cm table separation, compared to 0.62 cm separation for the repeated experiment. The importance of the 0.05 cm difference in table separation can be determined in terms of reactivity. The positive and negative reactor periods from which critical table separation is interpolated can be converted to excess reactivity (Reference 3) and an average reactivity gradient for table movement can be calculated. The average reactivity gradient for the original and repeat experiment is 55.7 ϕ /cm. If the table separation during critical of the repeat experiment was closed 0.05 cm closer (critical separation of original experiment), the repeat experiment would be 2.79 ϕ above delayed critical. Multiplying by $\frac{\beta}{100}$, where $\beta \sim 0.0065$, the 0.57 cm table separation would correspond to a Δk of 0.0002.

The repeat moderator experiment indicates that can placement and restacking have little effect on the reactivity of the system.

Repeat Critical Driver Experiment

The concrete-reflected experiment with the metal driver was repeated ten months after the original experiment to

determine the uncertainties due to the oxide weight gain and statistical error of measurements.

The repeat critical driver experiment used a 29.870 kg enriched uranium ($\sim 93\%$ ^{235}U) metal sphere driver in a $5 \times 5 \times 5$ (124 cans and a space for the driver) array of oxide cans. The experiment was performed on the horizontal split table using the concrete reflector. Oxide cans were placed in the same position in the array as in the original experiment. The total oxide can weight for the $5 \times 5 \times 5$ array was 1855 kg (based on March 1, 1978, weighings) for the original experiment. One different can was used in the repeat experiment, increasing the total weight by 35 grams. The original experiment was described in Reference 4.

The critical core separations of the repeated and original experiments were 1.82 and 1.26 cm, respectively, under the same conditions except for the oxide weight gain described in Reference 3 and a small perturbation of the core resulting from the reassembly of the oxide cans. The reactivity, $\frac{\Delta k}{\Delta x}$, in terms of change of the core separation near critical is 12.6 ϕ/mm for the original experiment and 15.1 ϕ/mm for the repeated experiment, averaging 13.9 ϕ/mm . The difference (5.56 mm) of two core separations corresponds to $\Delta k \approx 0.005$, which is approximately one sigma of the k_{eff} value calculated by the KENO-IV code using 100 batches of 300 neutrons per batch. Therefore, the repeated experiment implies that variations of the oxide weight gain and small perturbations

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of the can position have little effect on the reactivity of the system.

Restacked core cuboid dimensions nearly duplicate the cuboid dimensions of the original experiment and are given in another section of this report.

Critical Assembly Dimensions

Three north-south pairs of cuboids (table closure was in the north-south direction) are defined to describe the dimensions of the critical assemblies. These pairs are the outer reflector cuboids, inner reflector cuboids, and core cuboids. All cuboid dimensions, north-south (N-S), east-west (E-W), and vertical are given in reference to the position of the horizontal table. The cuboids are defined in more detail in Reference 3. Cuboid dimensions for the entire series of interstitially-moderated, concrete-reflected critical approaches and the concrete-reflected repeat driver experiment are given in Table II. Figure 7 is the key to Table II. The thickness of transite used (if any) between the north reflector end panel and north reflector frame is also given in Table II. Total weights and thicknesses of plastic and absorber sheaths used in the critical core cuboid are given in Table III.

TABLE 11

Cathoid Dimensions

Category	OUTER REFLECTOR CATHOID						INNER REFLECTOR CATHOID						CORE CATHOID					
	Dimensions (cm)			*Gap (cm)	N-S Direction		Transverse Thickness (cm)	Dimensions (cm)			Dimensions (cm)			Dimensions (cm)			Last Complete Layer (cm)	
	North Table	South Table	Vertical		E-W	C		D	F	G	H	I	J	K	N-S			E-W
				Table			Table								Table	Table	Table	Table
1A	60.9 ± 0.2	74.8 ± 0.2	128.3 ± 0.2	134.4 ± 0.1	0.8 ± 0.2	—	1.27	35.0 ± 0.3	35.4 ± 0.1	70.1 ± 0.2	70.4 ± 0.2	52.4 ± 0.3	34.7 ± 0.2	34.7 ± 0.1	68.8 ± 0.2	50.8 ± 0.1	32.9 ± 0.1	
1B	60.9 ± 0.2	74.8 ± 0.2	128.3 ± 0.2	134.4 ± 0.1	0.8 ± 0.2	—	1.27	34.9 ± 0.1	35.4 ± 0.2	61.9 ± 0.2	70.1 ± 0.2	52.4 ± 0.2	34.5 ± 0.1	34.7 ± 0.1	68.8 ± 0.1	50.8 ± 0.1	32.9 ± 0.1	
2A	61.8 ± 0.1	74.5 ± 0.1	128.3 ± 0.2	134.4 ± 0.1	0.8 ± 0.2	—	1.91	36.1 ± 0.1	35.0 ± 0.1	69.8 ± 0.1	69.8 ± 0.1	83.0 ± 0.1	35.0 ± 0.1	35.0 ± 0.1	69.8 ± 0.1	69.4 ± 0.1	51.6 ± 0.1	
2B	61.8 ± 0.1	74.5 ± 0.1	128.3 ± 0.2	134.4 ± 0.1	0.8 ± 0.2	—	1.91	36.1 ± 0.1	35.0 ± 0.1	69.8 ± 0.1	69.8 ± 0.1	83.0 ± 0.1	35.0 ± 0.1	35.0 ± 0.1	69.8 ± 0.1	69.4 ± 0.1	51.6 ± 0.1	
4A	60.8 ± 0.2	74.7 ± 0.3	128.3 ± 0.2	134.4 ± 0.1	0.7 ± 0.1	—	1.27	35.1 ± 0.1	35.7 ± 0.1	70.4 ± 0.1	70.3 ± 0.2	70.3 ± 0.2	34.9 ± 0.1	34.9 ± 0.1	69.3 ± 0.1	68.8 ± 0.1	51.3 ± 0.1	
4B	60.8 ± 0.2	74.7 ± 0.3	128.3 ± 0.2	134.4 ± 0.1	0.7 ± 0.1	—	1.27	35.0 ± 0.1	35.6 ± 0.2	70.0 ± 0.2	70.1 ± 0.1	70.3 ± 0.2	34.8 ± 0.1	35.0 ± 0.1	69.4 ± 0.1	68.8 ± 0.1	51.0 ± 0.1	
6A	58.4 ± 0.2	74.9 ± 0.4	128.3 ± 0.2	134.4 ± 0.1	—	1.5 ± 0.6	—	32.6 ± 0.2	49.3 ± 0.1	64.4 ± 0.1	64.4 ± 0.1	83.0 ± 0.1	31.8 ± 0.1	48.3 ± 0.1	64.3 ± 0.1	60.2 ± 0.1	63.9 ± 0.2	
6B	58.4 ± 0.2	74.9 ± 0.4	128.3 ± 0.2	134.4 ± 0.1	—	1.3 ± 0.1	—	32.6 ± 0.2	48.3 ± 0.1	64.4 ± 0.1	64.4 ± 0.1	83.0 ± 0.1	31.8 ± 0.1	48.3 ± 0.1	64.3 ± 0.1	60.2 ± 0.1	63.9 ± 0.2	
7	59.6 ± 0.2	74.5 ± 0.1	128.3 ± 0.2	134.4 ± 0.1	0.6 ± 0.1	0.6 ± 0.1	—	74.0 ± 3.2	49.0 ± 0.1	64.1 ± 0.1	61.0 ± 0.1	83.0 ± 0.1	32.2 ± 0.1	47.6 ± 0.1	64.0 ± 0.1	81.2 ± 0.1	63.4 ± 0.1	
Repeat Driver	58.3 ± 0.1	73.4 ± 0.1	128.3 ± 0.2	134.4 ± 0.1	0.9 ± 0.1	0.8 ± 0.1	—	32.7 ± 0.1	47.7 ± 0.1	77.5 ± 0.1	76.1 ± 0.1	83.1 ± 0.1	30.9 ± 0.1	46.3 ± 0.1	76.3 ± 0.1	76.9 ± 0.1	76.9 ± 0.1	

*See Figure 7.

**[20] gaps where neutron leakage might occur are reported.

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FIGURE 7

Dashed lines indicate core boundaries.
and solid lines represent reflector boundaries.

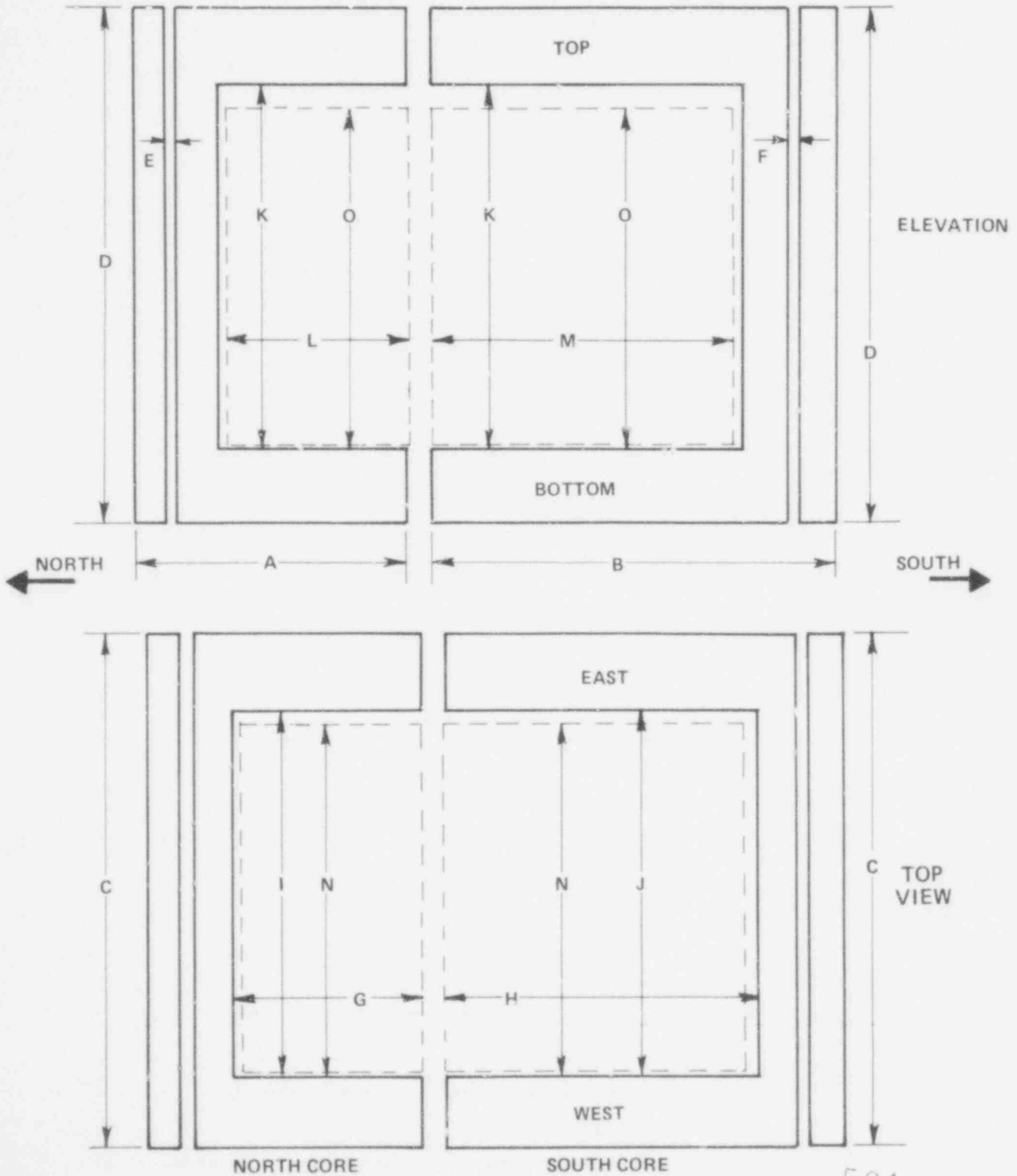


TABLE III

Critical Core Cuboid
Plastic and Absorber Sheath
Material Dimensions

Category	Plastic		Absorber Sheath		
	Total Weight (g)	Thickness (cm)	Total Weight (g)	Thickness (cm)	Material
1A	62345	2.43	--	--	--
1B	62345	2.43	--	--	--
2A	99110	2.43	75616	0.117	steel
2B	96678	2.43	74346	0.117	steel
4A	86845	2.43	5196	0.054	PVC
4B	85347	2.43	4968	0.05	PVC
6A	62681	0.93	--	--	--
6B	60857	0.93	--	--	--
7	55956	2.45	--	--	--

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MATERIAL ANALYSIS

Uranium Oxide

Impurity analysis of the wet ($H/U = 0.77$) compacted samples of the uranium oxide was performed using atomic absorption and emission spectrograph methods. Average values of the impurities and the method of analysis are given in Table IV.

Two dry, uncompact uranium oxide samples were analyzed for particle size. The analysis indicates 93% of the particles are between one and ten microns in size. Table V gives the particle size analysis by averaging the results of the two samples analyzed.

Non-Fissile Materials

The metal analysis of the north table scram guide (denoted as S5 in Figures 8, 9, 10, and 17 of Reference 3) and the steel lower channel framework portion of the table (denoted as S6 in Figures 8, 9, 10, and 17 of Reference 3) has been completed and is given in Table VI.

Transite used in the north reflector during some of the concrete-reflected critical experiments was analyzed. Transite composition is given in Table VII.

TABLE IV
Oxide Impurity Analysis

Impurity	Weight ppm	Method of Analysis*
Al	37	E
B	0.3	E
Be	< 0.4	E
Bi	< 5	E
Ca	18	E
Cd	< 5	A
Cr	128	A
Cu	185	A
Fe	312	A
K	< 25	E
Mg	13	E
Mn	< 3	E
Mo	< 4	E
Ni	16	E
P	< 70	E
Pb	< 10	E
Si	128	A
Sn	< 5	E
V	< 5	E
Zn	< 17	E

*A = atomic absorption method.
E = emission spectrographic method.

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TABLE V
Uranium Oxide
Particle Size Analysis

Particle Size (microns)	Percent of Particles in Range
< 1	1.05
1 - 10	93.00
10 - 25	4.45
25 - 40	0.75
40 - 60	0.35
60 - 80	0.10
80 - 100	0.15
> 100	0.15

TABLE VI
Elemental Composition of Steel

Location of Steel	Weight-Percent by Element														All Others ²
	C	Sn	Si	Cr	Mn	Fe ¹	Ta	Cu	Mo	Co	Na	Ti	W	Zn	
north table scram guide bars	0.63	0.1	0.1	0.01	1.1	97.679	< 0.1	0.1	0.01	0.02	0.01	0.01	0.05	0.01	0.071
lower portion of table	0.25	0.1	0.1	0.1	0.7	98.231	< 0.1	0.1	0.01	0.02	0.01	0.01	< 0.05	0.01	0.209

¹ Obtained by subtracting sum of other fourteen columns from 100%.

² Sum of twenty other metallic elements.

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TABLE VII
Transite Composition

Element	Weight-Percent
Ca	35
Al	1.7
Si	9.5
Mg	4.6
Fe	1.9
Na	0.3
K	0.2
C	0.64
S	< 0.001
O	46.16

Density 2.44 g/cc

THE SECOND WETNESS: H/U = 1.25

One hundred twenty nine (129) oxide cans were injected with sufficient distilled water to raise the H/U atomic ratio from 0.77 to 1.25. All but four of these were "regularly packed" cans consisting of twenty-eight blocks of compacted oxide weighing \sim 540 g each. The remaining four were "special" cans having a U-shaped notch designed to clear the metal driver support rod and other experimental equipment. Because of this notch, the oxide in those cans weighed only 82% of that in the "regular" cans and received correspondingly less water. Seven cans, including one "special" can, were left at the first wetness since they are not needed for projected experiments and doing so retains the capability for future laboratory analyses at the lower H/U ratio.

The weight of water to be injected was calculated recognizing that several materials contribute hydrogen to an experimental can:

[a] The water initially injected to create the first wetness (Nov-Dec, 1977).

[b] Moisture absorbed into the relatively hygroscopic uranium oxide after it had been calcined at high temperatures by National Lead of Ohio in 1976.

[c] The plastic sandwich bag which contained the loose oxide powder at the time of compaction.

[d] A second plastic bag wrapped around each block as it was placed into a can (to reduce water migration between blocks within a can).

[e] The vinyl tape used to seal the aluminum lid to the aluminum can.

[f] The thin mylar tape placed over the water injection holes to prevent moisture loss or gain during the experimental program.

[g] The water to be injected to yield the second wetness.

Table VIII shows the hydrogen and uranium data for a typical can resulting from the above considerations. The two total element weights from Table VIII may be used to calculate the expected resulting atomic ratio at the second wetness:

$$H/U = \frac{67.8 \text{ g H/can} + 1.008}{12750 \text{ g U/can} + 237.95} = 1.255$$

The atomic weights used are the textbook value for hydrogen and the product of measured isotopic enrichment weight-fractions (see Table IX) and textbook isotopic weights for each isotope of uranium:

$$237.95 = .95437(238.0508) + .04461(235.0439) \\ + .00088(236.0457) + .00030(234.0409)$$

The second watering was accomplished by injecting 224 g of water (item g of Table VIII) equally into the 56 holes

TABLE VIII

Hydrogen Contribution for a Typical "Regularly Packed"
Oxide Can Yielding H/U \approx 1.25

Description	Weight (g)	Hydrogen Weight- Fraction	Hydrogen Weight (g)
a. first water injection	273.2	.1119	30.6
b. initial moisture	37.8	.1119	4.2
c., d. plastic bags	53.0	.1401	7.4
e. vinyl tape	3.0	.0592	0.2
f. mylar tape	4.0	.0683	0.3
g. this water injection	224.0	.1119	25.1
Total hydrogen weight per can			67.8
Uranium oxide weight including initial moisture			15129
Uranium oxide weight excluding initial moisture			15091
Uranium assay			0.8449
Total uranium weight per can			12750

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TABLE IX
 Uranium Isotopic Enrichment

Date Measured	Weight-Percent of Uranium Isotope				Remarks (analysis by)
	U-234	U-235	U-236	U-238	
4/26/76		4.479			1st shipment, powder (National Lead)
8/10/76		4.476			2nd shipment, powder (National Lead)
11/10/78	.030	4.465	.086	95.419	H/U = .77 (Rocky Flats)
3/12/79	.030	4.425	.090	95.455	last at H/U = .77 (Rocky Flats)
Average	.030	4.461 ±.025	.088	95.437	total = 100.016 weight-percent

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drilled through the aluminum sides of a can. Thus, 4 g of water were injected per hole, and Figure 8 shows one experimenter making these injections. The top 2 or 3 layers of oxide blocks would accept the water readily. The bottom several layers took longer because the oxide appeared to be less permeable to water penetration. Eventually, every oxide block received its total share of water, 4 g in each of two holes, and the plastic wrappers around each block reduces the possibility that the water will migrate between blocks.

Every can received a careful examination coordinated with its watering. The vinyl tape holding the lid to the can was removed and discarded. The lid was flattened if it had become bent through use. One of two layers of mylar tape covering each of eight rows of water injection holes was removed and discarded. Then, the oxide was gouged with an ordinary ice pick to facilitate water injection. The water was injected through the now-pierced remaining layer of mylar tape. After watering one side of a can, it was wiped clean and fresh strips of mylar tape were placed over the four vertical rows of holes to prevent evaporation. The process was then repeated on the opposite side of the can.

Each can was weighed before any of the above steps were accomplished. This constituted the last weighing at the first wetness ($H/U = .77$). Each can was again weighed after watering and retaping. This second weight yielded the first weighing at the second wetness ($H/U \sim 1.25$).

Figure 8

Each syringe contains 4 g of water which is delivered to the oxide through holes drilled in the side of the aluminum can. The entire 112 g of water for one side of a can was measured into a glass jar before beginning the injection process.

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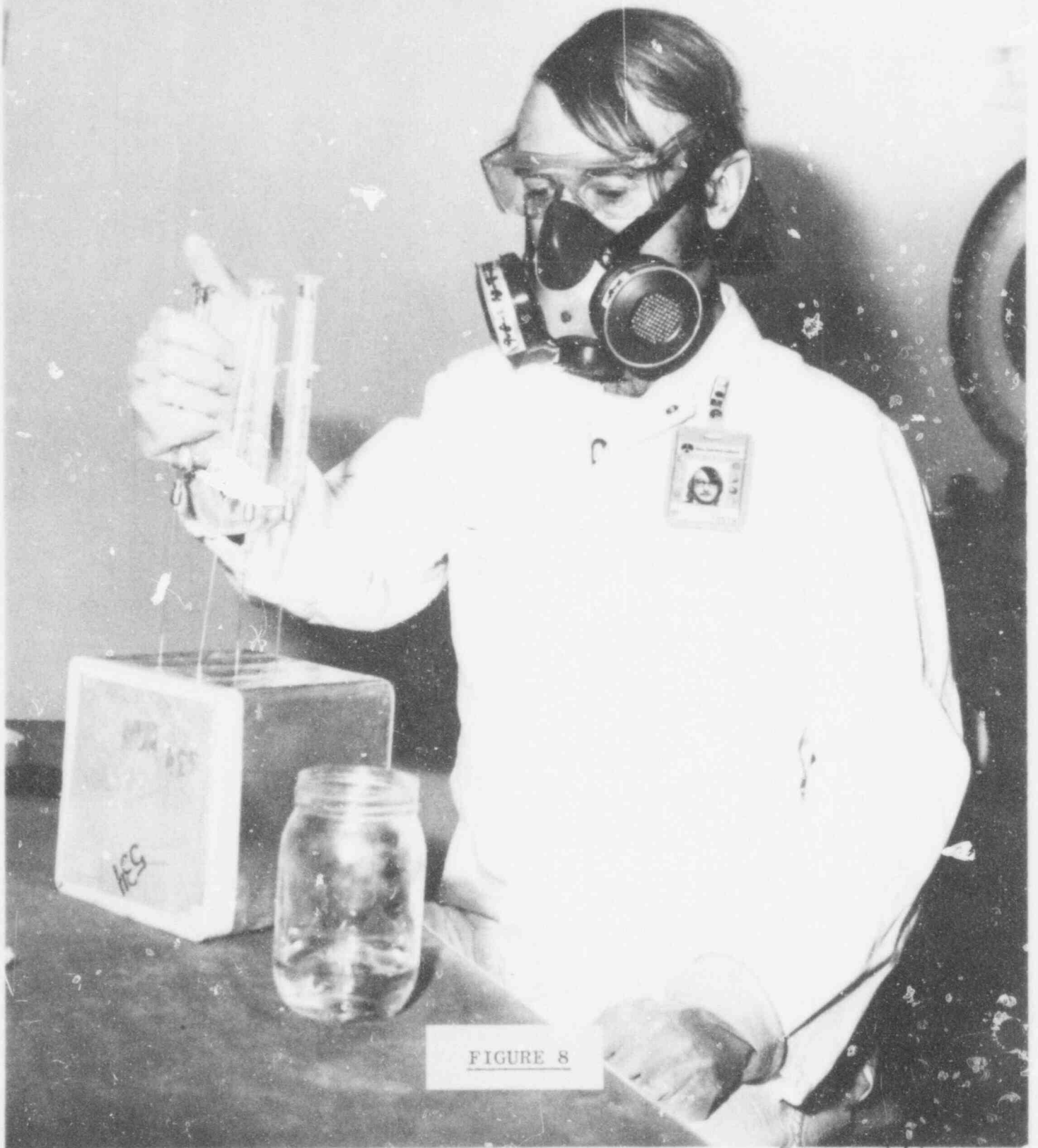


FIGURE 8

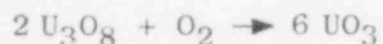
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OXIDE WEIGHT GAIN

A weight gain problem has been discussed in several previous quarterly progress reports (References 1, 2, 3, and 4). The final weighing at the first wetness permits a concluding assessment of the net weight gain over the one year's time that the oxide resided at H/U = 0.77.

The total weight of 125 "regularly packed" oxide cans on March 1, 1978, just prior to the first experiments, was 1,999,033 g, an average of 15,992 g/can. This weight included the can, lid, oxide, water, plastic bags, and two kinds of tape. Between then and March, 1979, the same 125 cans gained a total of 3191 g (0.16%). The average weight per can grew 25.53 g to ~ 16,018 g. Not all cans gained weight at the same rate. The largest gain was 47 g, and the smallest was 4 g. The weight gain for all 125 cans is shown in Figure 9. The standard deviation for this assumed normal distribution is ± 9.64 g.

The weight gain is probably attributable to oxygen absorbed from the atmosphere. There are two possible chemical mechanisms for this process. The first is:



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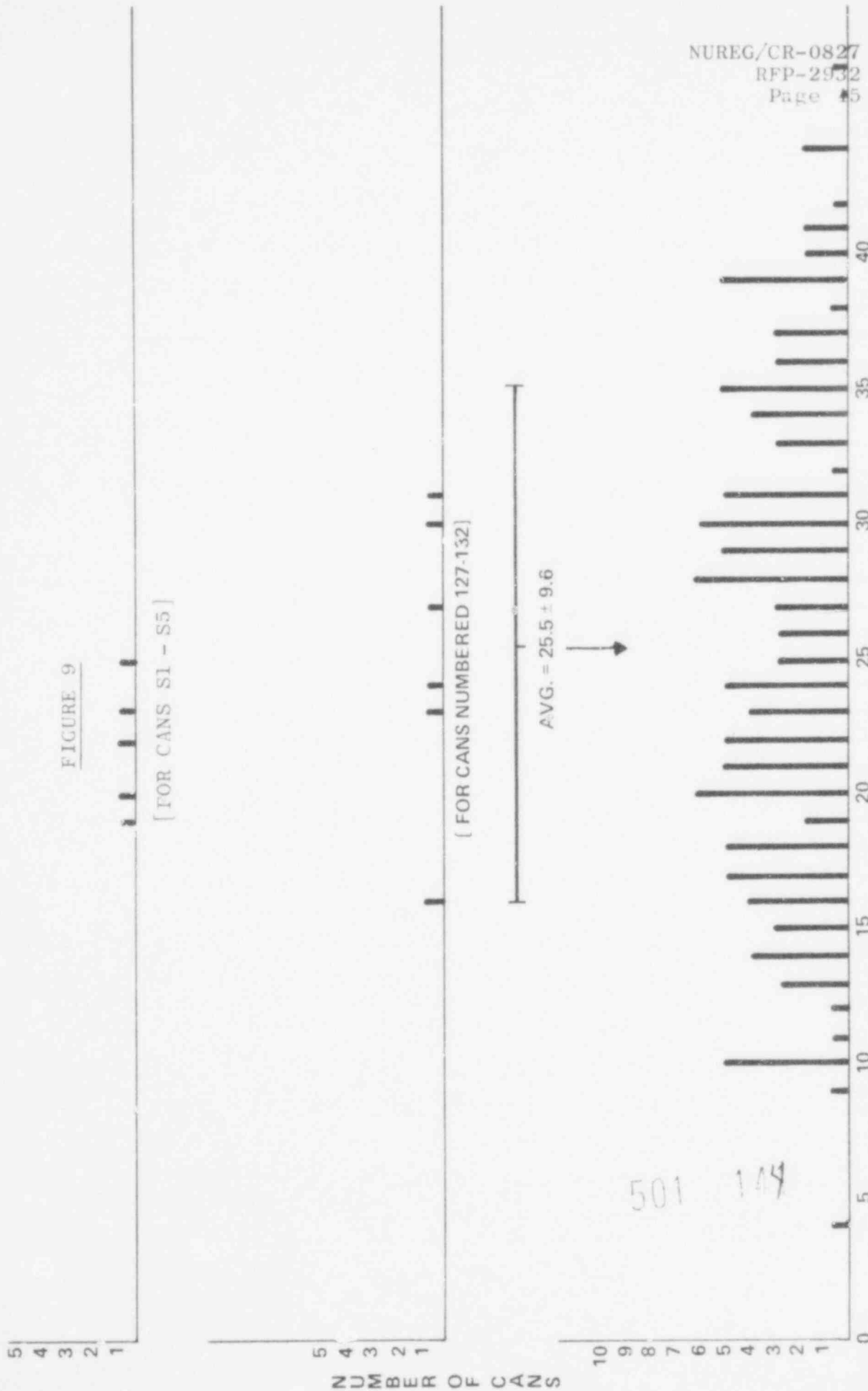
The oxide U_3O_8 is known to be the stoichiometric form at elevated temperatures. It is assumed that this material slowly oxidizes to UO_3 over long periods at room temperature.

Figure 9

Each line represents the number of oxide cans that have gained a certain weight between March 1, 1978, and March, 1979. The 125 cans represented by the lower third of the figure are the "regularly packed" cans referred to in the text. The middle set of cans probably will not be used on the projected program. The five cans along the top are the special ones designed for source and driver support clearance.

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FIGURE 9



However, only small particles of UO_2 (~ 1 micron) are oxidized to UO_3 at room temperatures (Reference 5). Particle size analysis of the uranium oxide indicates 93% of the particles are between one and ten microns (see Material Analysis section of this report). The second possible reaction is surface oxidation of UO_2 [U_3O_8 is $UO_2 \cdot 2(UO_3)$] to $UO_{2.3}$ at room temperature in contact with dry air (Reference 5). X-ray diffraction results of the uranium oxide indicate the presence of $UO_{2.3}$ (Reference 2). Methods to verify the above chemical mechanisms are being discussed.

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16. ABSTRACT (200 words or less) <p>This is the fourteenth (14th) in the series of quarterly progress reports describing reference critical experiments being performed at the Critical Mass Laboratory at Rockwell International's Rocky Flats Plant for the U.S. Nuclear Regulatory Commission (NRC).</p> <p>Eight critical experiments were performed using the low-enriched, damp (H/U ~ .77) uranium oxide (U₃O₈) in the concrete reflector. This concludes the interstitially-moderated experiments. Two critical experiments were repeated. One was repeated ten months after the original experiment to determine the combined effect of oxide weight gain and statistical error of measurements. The second repeat experiment was performed a few hours after the original experiment, so oxide weight gain would not be a factor, to see the effect on critical results of restacking and rearranging the oxide cans.</p> <p>Impurity content and particle size distribution of the oxide were determined by laboratory analysis.</p>				11. CONTRACT NO. Fin No. A1036	
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