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CRITICALITY EXPERIMENTS WITH SUBCRITICAL CLUSTERS OF LOW ENRICHED UO_2 RODS IN WATER WITH URANIUM OR LEAD REFLECTING WALLS

REACTORS

KEYWORDS: uranium dioxide, criticality, water, spent fuel storage, moderators, spent fuel casks, neutron reflectors, lead, depleted uranium

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A series of criticality experiments with 2.35 and 4.31 wt% ^{235}U enriched UO_2 rods in water has provided well-defined benchmark-type data showing that both depleted uranium and lead reflecting walls, submerged in the water reflector, are better neutron reflectors than water alone. For each fuel enrichment, the critical separation between three subcritical, near optimally moderated fuel clusters was observed to increase as either 77-mm-thick depleted uranium or 102-mm-thick lead reflecting walls were moved toward the fuel. The maximum reactivity effect was observed for the depleted uranium with ~20 mm of water between the reflecting walls and the fuel region, whereas for the lead, a maximum effect was obtained with essentially no water between the reflecting walls and the fuel region. This maximum reactivity effect was observed to occur at the same spatial separation between the fuel and reflecting walls for both fuel enrichments. However, the measurements indicated that the magnitude of this phenomenon is dependent on the ^{235}U enrichment of the fuel. The lead reflecting walls increased the critical separation between fuel clusters a maximum of 67% for the 2.35 wt% ^{235}U enriched fuel and at least 152% for the 4.31 wt% enriched fuel. Similar results were observed with the depleted uranium reflecting walls.

INTRODUCTION

A research program, funded by the U.S. Nuclear Regulatory Commission (NRC), to provide experimental criticality data on conditions simulating light water reactor fuel shipping and storage configurations was begun in 1976 at the Battelle-operated Critical

Mass Laboratory at Hanford. The initial two series¹⁻³ of experiments in this program were concerned with determining the critical separation between clusters of either 2.35 or 4.31 wt% ^{235}U enriched UO_2 fuel rods immersed in water with various absorbing materials in the water region between the fuel clusters. The third series of experiments in this program is covered in this paper and involves the same fuel immersed in water as before; however, this third set of experiments is concerned with determining the effect that depleted uranium or lead reflecting walls adjacent to the fuel clusters have on the critical separation between the fuel clusters. They are intended to simulate shipping and storage conditions in which biological shielding materials are present. The objective of these experiments, as in the previous experiments, is to provide clean, definable, integral data that can be described in calculations exactly as run without corrections or approximations having to be made. No particular attempt is made to obtain parametric correlations between different fuels or biological shielding material.

EXPERIMENTS

The experiments consisted of determining the critical separation between three subcritical clusters of rods aligned in a row with either depleted uranium or lead walls parallel on either side of, and at various distances from, the row of fuel clusters. Similar measurements⁴ in 1951 at the Oak Ridge National Laboratory (ORNL) had shown that natural uranium slugs in the water adjacent to a water-flooded 93 wt% ^{235}U enriched fuel array would increase the reactivity of the array, and that this increased reactivity would be a maximum with ~25 mm of water between the fuel and the natural uranium slugs. Also, similar experiments^{4,5} have shown that lead reflecting walls should also increase the reactivity of the water

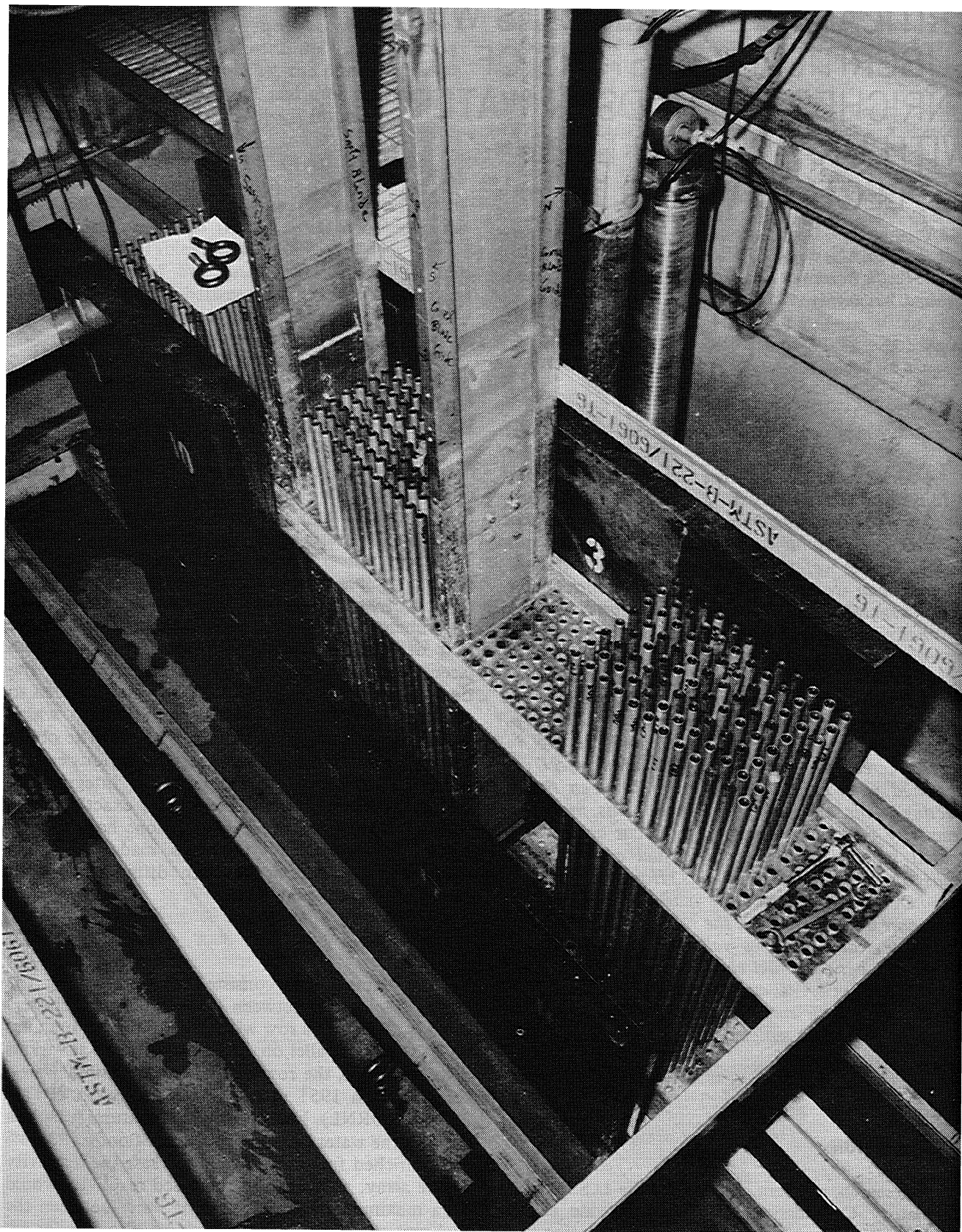


Fig. 1. Typical experiment assembly with depleted uranium walls partially removed.

flooded array. The measurements covered in Ref. 4 indicated that the critical mass of a completely inundated system is insensitive to separation of core and lead reflector up to a separation of ~50 mm and that at an ~100-mm separation, the lead was completely isolated from the core. Considering this earlier information, the experiments presented in this paper were designed to particularly provide data over the first 50 mm of separation between fuel and reflecting walls.

A photograph of a typical assembly, with the uranium walls partially constructed, is shown in Fig. 1. The system is provided with a safety blade and a control blade. Both of these are shown inserted on

either side of the center fuel cluster in Fig. 1. They would be fully withdrawn whenever data are being obtained. Also, of course, the walls would be completed and the entire system flooded with water to a depth of at least 150 mm above the top of the fuel before any measurements were made. The grid plates used for latticing the fuel rods into clusters were an acrylic ($C_5H_8O_2$)_n material (Plexiglas) having about the same density (1.18 g/cm³) and neutron moderating characteristics as water. A detailed graphic layout of the experimental system is given in Fig. 2.

The fuel clusters consisted of UO_2 rods equally spaced on a square pitch to provide water-to-fuel-volume ratios near optimum neutron moderation.

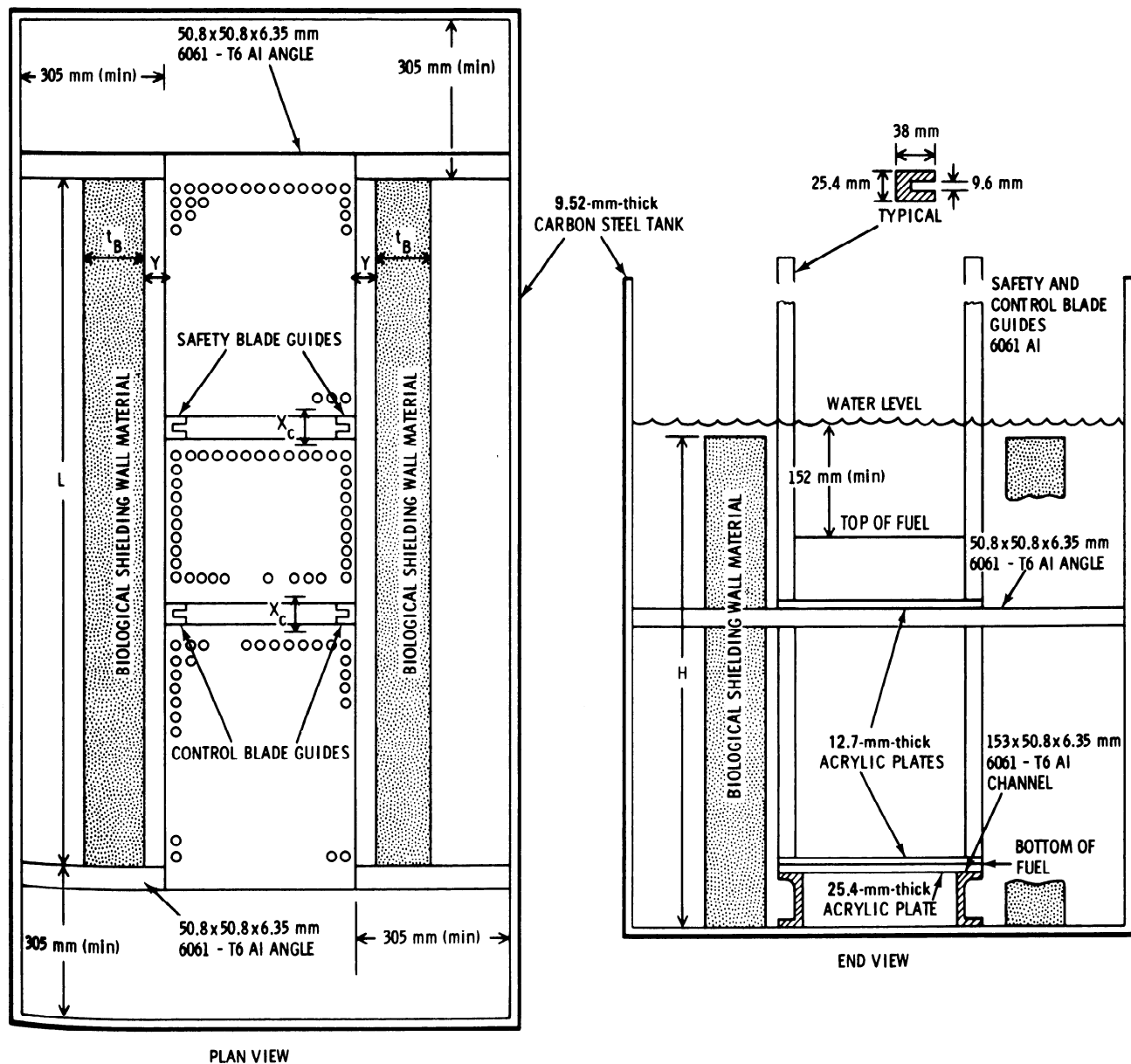
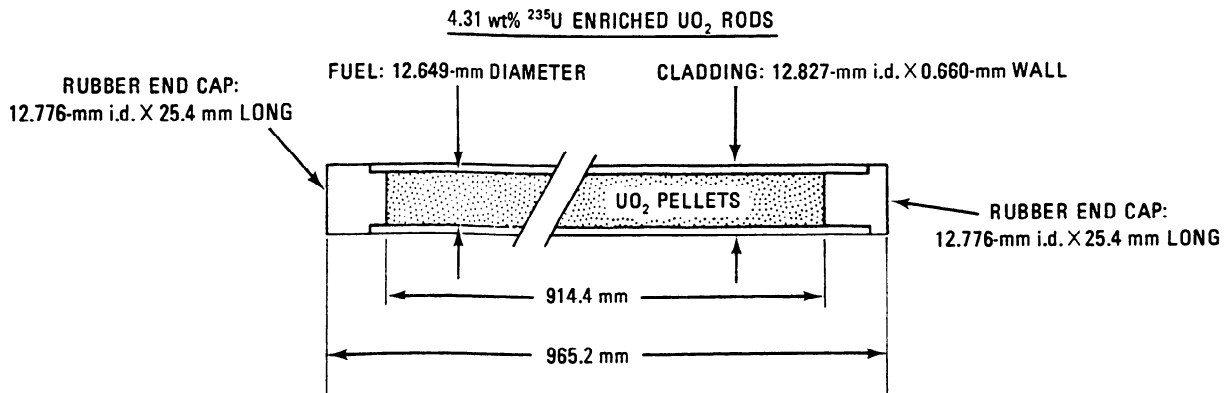


Fig. 2. Graphical arrangement of simulated shipping container critical experiments.

Each fuel cluster was rectangular in shape, contained the same number of fuel rods, and had the same outside dimensions. As in the previously reported measurements,³ complete sets of data were obtained at two different ^{235}U enrichments, 2.35 and 4.31 wt%. A detailed description of each type fuel rod is given in Fig. 3. The chemical impurities of the water used in these experiments are given in Table I.

The reflecting walls consisted of either depleted

uranium or lead and were constructed on either side of the row of fuel clusters, as indicated in Fig. 2. In each case, the walls were an equal distance from the fuel clusters and extended beyond the fuel clusters in all directions. The distance between the walls and the fuel clusters was varied from zero (the cell boundary of the fuel clusters) to infinity (complete removal of the walls from the system). At each separation between fuel clusters and reflecting walls (dimension Y



CLADDING: 6061 ALUMINUM TUBING

LOADING:

ENRICHMENT = 4.31 ± 0.01 wt% ^{235}U

FUEL DENSITY = 10.40 ± 0.06 g/cm³

URANIUM ASSAY = 88.055 ± 0.261 wt% OF TOTAL FUEL COMPOSITION

UO_2 = 1203.38 ± 4.12 g/ROD

END CAP:

C = 58 ± 1 wt%

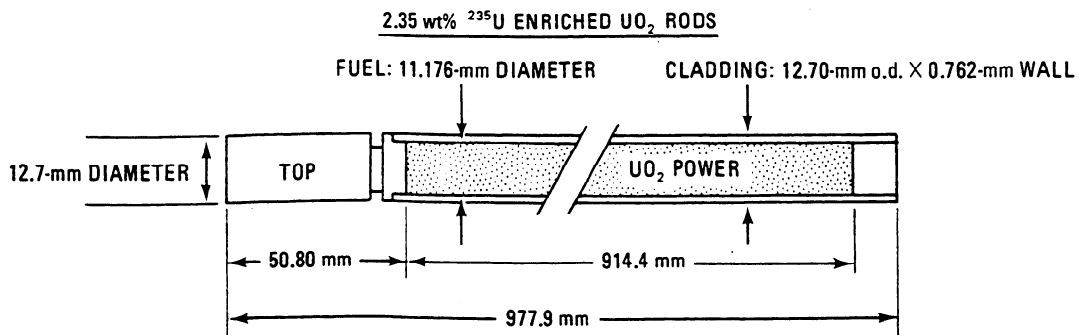
S = 1.7 ± 0.2 wt%

H = 6.5 ± 0.3 wt%

O = 22.1 wt% (BALANCE)

Ca = 11.4 ± 1.8 wt%

Si = 0.3 ± 0.1 wt%



CLADDING: 6061 ALUMINUM TUBING SEAL WELDED WITH A LOWER END PLUG OF 5052-H32 ALUMINUM AND A TOP PLUG OF 1100 ALUMINUM

LOADING:

ENRICHMENT = 2.35 ± 0.05 wt% ^{235}U

FUEL DENSITY = 9.20 mg/mm³ (84% TD)

URANIUM ASSAY = 88.0 wt%

UO_2 = 825 g/ROD (AVERAGE)

Fig. 3. Description of fuel rods.

TABLE I
Major Chemical Impurities of Water in the Simulated Shipping Container Experiments

Component	Concentration ^a (ppm)
Cl	30.2 ± 5.8
NO_3^-	0.42 ± 0.16
Cr^{+6}	<0.01
Zn	0.26 ± 0.07
Mn	<0.01
Pb	<0.005
Fl	0.15 ± 0.04
Fe	<0.03
Cu	<0.01
Cd	0.006 ± 0.001
SO_3	6.6 ± 0.04
Dissolved solids	137 ± 5

^aError limits are standard deviations observed in three samples.

in Fig. 2), a critical approach on the water separation, X_c , between fuel clusters was made by incrementally decreasing the spacing separating the fuel clusters. In each measurement, the fuel clusters were centered between the reflecting walls in the X - Y plane.

The uranium walls on either side of the fuel clusters were ~1.5 m long \times ~1.2 m high and were ~76 mm thick. These walls were constructed by assembling 5 long \times 2 high tongue and groove slabs of uranium, each 304.6 ± 0.9 mm wide \times 609.5 ± 2.5 mm high \times 76.5 ± 0.4 mm thick. One such slab is shown being lowered into position in Fig. 4. A complete general description of the uranium wall is given in Fig. 5. Each individual uranium slab was radiographed along its entire length to ensure uniform density in each slab and that each slab was free of internal voids >1.5 mm in diameter. All except four slabs met this criterion. In each of these four slabs, three voids of up to ~6 mm in diameter and of undefined thicknesses were observed within 125 mm of either the top or bottom edges of the slabs. These four slabs were positioned in the walls such that these slightly nonuniform areas were located on the edges of the walls.

The lead walls on either side of the fuel clusters were ~1.6 m long \times ~1.2 m high and were ~0.1 m thick. These walls were constructed by stacking, 8 long \times 24 high, lead bricks, each ~205 \times 102 \times 51 mm, one on top of the other. A generalized diagram of the lead walls is given in Fig. 6 as an aid to computer input.

EXPERIMENTAL DATA

Each of the experiments is physically described in Table II. The measurement data obtained are sum-

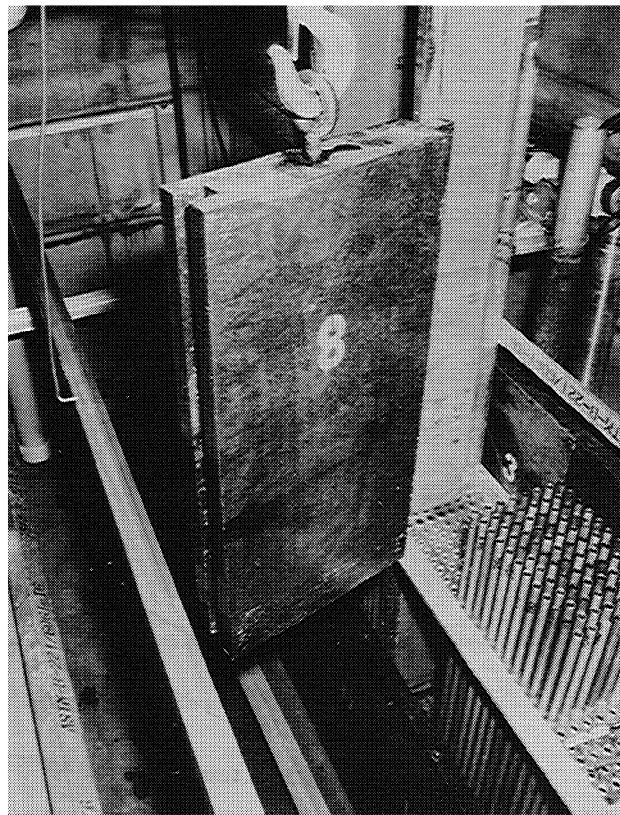


Fig. 4. Depleted uranium slab being lowered into position.

marized in Table II and Fig. 7. Both the lead and the uranium reflecting walls caused an increase in the critical separation between the fuel clusters. As can be seen in Fig. 7, the data are very similar for both the 2.35 and the 4.31 wt% ^{235}U enriched fuels, except the effect of the reflecting walls was observed to be much greater for the 4.31 wt% enriched fuel.

For either enrichment, the maximum effect of the uranium walls was observed with the walls ~20 mm from the fuel clusters. The fuel clusters were 19 rods long \times 16 rods wide for the 2.35 wt% enriched fuel and 13 rods long \times 8 rods wide for the 4.31 wt% enriched fuel to obtain a nearly common critical separation (83 mm) for both enrichments at full water reflection. This critical separation was observed to increase to its maximum of ~139 mm for the 2.35 wt% enriched fuel clusters; however, in this region of maximum effect, the 13- \times 8-rod 4.31 wt% enriched fuel clusters would have been supercritical at any separation. (A single cluster, 8 rods wide, was experimentally determined to require only 101.5 ± 0.5 rods for criticality.) Consequently, to obtain benchmark-type data for the 4.31 wt% enriched fuel at this point of maximum effect, the fuel clusters were reduced in size to 12 rods long \times 8 rods wide.

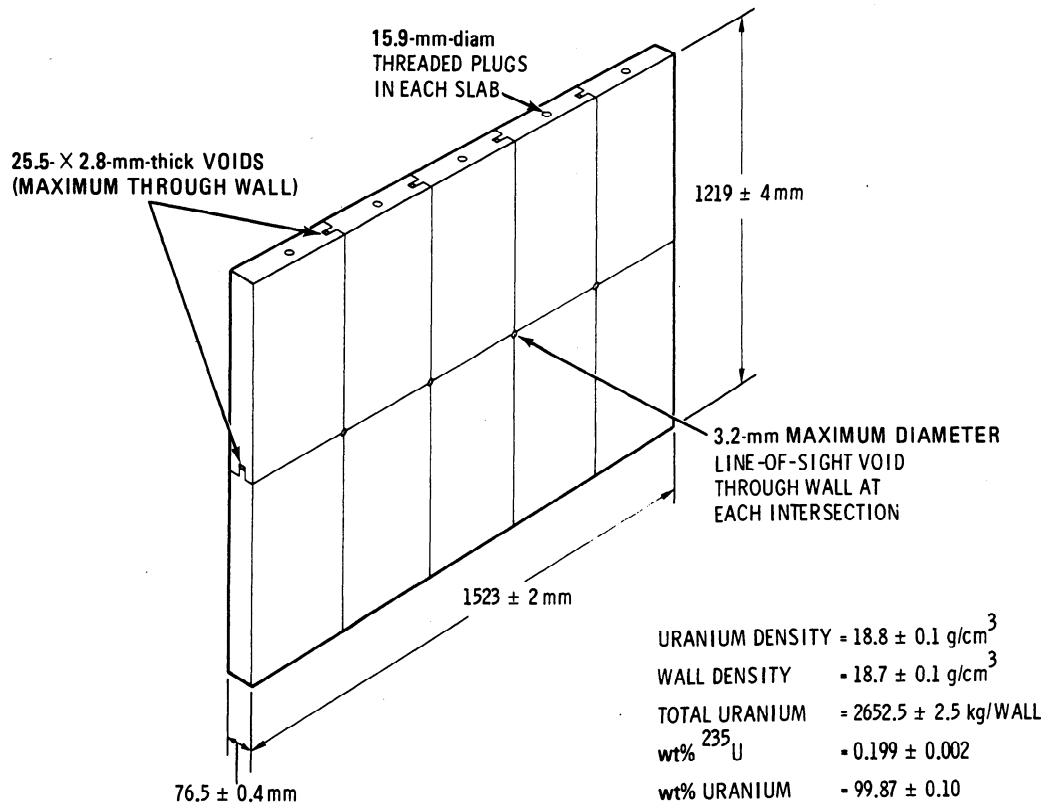


Fig. 5. Assembled uranium wall.

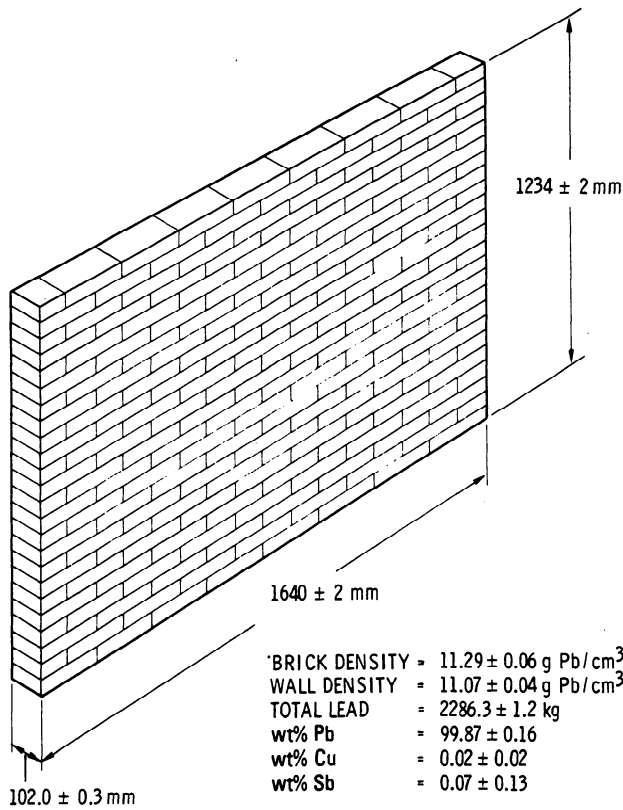


Fig. 6. Assembled lead wall.

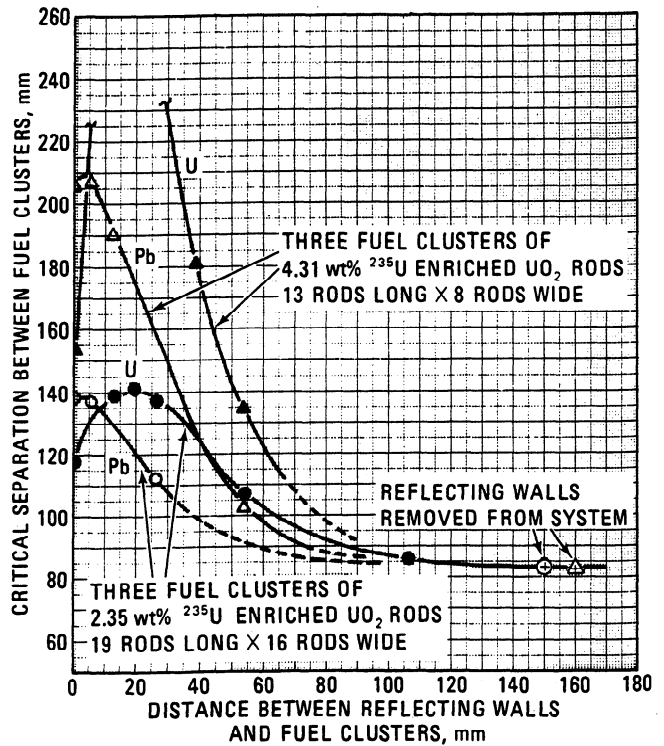


Fig. 7. Critical separation between fuel clusters of 2.35 and 4.31 wt% ²³⁵U enriched UO₂ rods in water with depleted uranium or lead walls.

TABLE II
Experimental Data on Clusters of 2.35 and 4.31 wt% ^{235}U Enriched UO_2 Rods in Water with Depleted Uranium or Lead Reflecting Walls*

Distance Between Reflecting Walls and Fuel Clusters ^a (mm)	2.35 wt% Enriched Fuel			4.31 wt% Enriched Fuel		
	Fuel Clusters 20.32-mm Square Pitch ^b	Critical Separation Between Fuel Clusters ^c		Fuel Clusters 25.40-mm Square Pitch ^b	Critical Separation Between Fuel Clusters ^c	
		Uranium Walls ^d (mm)	Lead Walls ^e (mm)		Uranium Walls ^d (mm)	Lead Walls ^e (mm)
0	3-19 × 16	118.3 ± 0.2	138.4 ± 0.1	3-13 × 8	153.8 ± 0.1	206.2 ± 0.1
6.60 ± 1.02	3-19 × 16	---	137.2 ± 0.1	---	---	207.8 ± 0.2
13.21 ± 0.76	3-19 × 16	139.3 ± 0.1	---	---	---	190.4 ± 0.2
19.56 ± 1.02	3-19 × 16	141.1 ± 0.1	---	3-12 × 8	153.2 ± 0.1 ^f	---
26.16 ± 0.76	3-19 × 16	137.0 ± 0.2	112.5 ± 0.8	---	---	---
39.12 ± 0.76	---	---	---	3-13 × 8	180.5 ± 0.5	---
54.05 ± 1.02	3-19 × 16	106.9 ± 0.2	---	3-13 × 8	134.9 ± 0.2	103.0 ± 0.2
106.76 ± 1.52	3-19 × 16	85.6 ± 0.2	---	---	---	---
∞	3-19 × 16	83.1 ± 0.4	83.1 ± 0.4	3-13 × 8	82.4 ± 0.3	82.4 ± 0.3
∞	3-20 × 16 ^g	91.3 ± 0.2	91.3 ± 0.2	---	---	---

*Error limits shown are one standard deviation.

^aPerpendicular distance between the cell boundary of the fuel clusters and the reflecting walls.

^bNumber of fuel clusters, rods long X rods wide, aligned in a row.

^cPerpendicular distance between the cell boundaries of the fuel clusters.

^dWalls 76.5 ± 0.4 mm thick.

^eWalls 102.0 ± 0.3 mm thick.

^fA single fuel cluster, 8 rods wide, was determined from experiments to require 101.5 ± 0.5 rods for criticality.

^gCritical separations of 91.3, 91.6, 91.5, and 91.8 mm obtained in previous experiments at this lattice pitch. No previous measurements with three 19 × 16 rod clusters.

The maximum effect of the lead walls occurred with the walls at or very near the cell boundary of the fuel clusters. The data indicated that a maximum effect might exist with ~3 mm of water between the fuel clusters and the lead walls. Based on the data obtained with the uranium walls, this unobserved maximum effect could be quite large for the 4.31 wt% ^{235}U enriched fuel but is probably quite small for the 2.35 wt% ^{235}U enriched fuel when compared to the effect with no water between the fuel clusters and the lead walls. To investigate this possibility, calculations were performed with the KENO-IV computer code.⁶ (It was virtually impossible, with any degree of accuracy, to separate the lead walls and fuel clusters less than ~6 mm in the experiments.) The calculations were made using 17 epithermal broad-group cross sections, generated using the EGGNIT code⁷ and FLANGE-ETOG-processed ENDF/B-IV data,^{8,9} and a single thermal group, generated using THERMOS (Ref. 10) with ENDF/B-III data. At the experimentally determined critical condition with 6.6 mm separating the lead walls from the fuel clusters, a k_{eff} of 1.002 ± 0.005 was calculated for both enrichments. Reducing this separation to 3.3 mm and using the critical separations between fuel clusters indicated

by the curve in Fig. 7 (139 mm for the 2.35 wt% ^{235}U enriched fuel and 209 mm for the 4.31 wt% ^{235}U enriched fuel) resulted in calculated k_{eff} values of 0.996 ± 0.005 and 1.003 ± 0.005, respectively, for the 2.35 and 4.31 wt% ^{235}U enriched fuels. It would appear, therefore, that the maximum effect of the lead walls occurs essentially with the walls at the fuel zone boundary for both fuels.

The critical separations of 83.1 ± 0.4 mm and 82.4 ± 0.3 mm obtained with an infinite amount of water between the fuel and the reflecting walls are arbitrarily shown at 150 and 160 mm in Fig. 7 for the 2.35 and 4.31 wt% ^{235}U enriched fuel, respectively.

CONCLUSIONS

Both depleted uranium and lead reflecting walls submerged in a water reflector are better neutron reflectors than water alone. This effect is a maximum with ~20 mm of water between the uranium reflecting walls and the fuel region. The effect is a maximum with essentially no water between the lead and the fuel. The data also indicate that this phenomenon is dependent on the ^{235}U enrichment of the fuel. In

the experiments covered in this paper, the lead reflecting walls increased the critical separation between fuel clusters a maximum of 67% for the 2.35 wt% ²³⁵U enriched fuel (83 to 139 mm) and at least 152% for the 4.31 wt% enriched fuel (82 to 208 mm). Similar results were observed with the depleted uranium reflecting walls.

The results obtained with the depleted uranium reflecting walls are in close agreement with results reported on similar measurements⁴ at ORNL in 1951. A reinterpretation of the ORNL data can easily be made to obtain an exact agreement for the condition at which the maximum reactivity effect is observed, i.e., 20 mm of water between the fuel region and the uranium reflecting walls. Consequently, it would appear that this point of maximum reactivity effect is not dependent on the enrichment of the fuel region. (ORNL experiments were performed with 93 wt% ²³⁵U enriched fuel.)

As mentioned earlier, some measurements with lead reflecting walls in water were also performed in 1951 at ORNL. The results reported for lead in this paper do not agree with the conclusions drawn from these earlier experiments. In contrast, the data presented in this paper show that a lead reflecting wall in a completely inundated fuel system has a large effect on the reactivity of the system, and that this effect is very sensitive to the separation between the lead reflecting walls and the fuel region.

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