
Criticality Experiments with Subcritical Clusters of 2.35 Wt% and 4.31 Wt% ^{235}U Enriched UO_2 Rods in Water with Uranium or Lead Reflecting Walls

Undermoderated Water-to-Fuel Volume Ratio of 1.6

Prepared by S. R. Bierman, B. M. Durst, E. D. Clayton

Pacific Northwest Laboratory
Operated by
Battelle Memorial Institute

Prepared for
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Commission

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ABSTRACT

A series of criticality experiments with undermoderated (1.6 water-to-fuel volume ratio) 2.35 wt% and 4.31 wt% ^{235}U enriched UO_2 rods in water were performed to provide data on the reactivity effects of lead and depleted uranium reflecting walls. This data furnishes well defined benchmarks for use in validating calculational techniques employed in analyzing fuel shipping and storage systems having lead or uranium biological shields. For each fuel enrichment, the critical separation between three subcritical fuel clusters was observed to increase as either 77mm thick depleted uranium or 102mm thick lead reflecting walls were moved towards the fuel. A maximum critical separation was observed for both the lead and the depleted uranium reflecting walls with a water gap between the fuel clusters and the reflecting walls. For both fuel enrichments, this optimum water gap was about 25mm for the depleted uranium walls and about 10mm for the lead walls.

SUMMARY

The results from the sixth in a series of criticality experiments funded by the United States Nuclear Regulatory Commission are presented in this paper. The purpose of these experiments is to provide a data base for benchmarking computational methods used in the criticality analysis of fuel transportation and storage systems.

The initial five series of experiments in this program were concerned with determining the critical separation between clusters of either 2.35 wt% or 4.31 wt% ^{235}U enriched UO_2 fuel rods immersed in water at near optimum neutron moderation and at an undermoderated water-to-fuel volume ratio of 1.6. The effect that various neutron absorber plates, positioned between the fuel clusters, had on the critical separation was investigated in these previous experiments. Also the effect that simulated biological shields of lead, steel and depleted uranium had on the critical separation was investigated, but not for both neutron moderation conditions. In these previous experiments, data were obtained for the lead and depleted uranium only at the near optimum neutron moderation condition. This sixth series of experiments are concerned with the lead and depleted uranium walls and fuel clusters at the undermoderated water-to-fuel volume ratios of 1.6.

Measurements similar to those performed previously with lead and depleted uranium were repeated with the same materials used previously, but with the fuel lattices at a water-to-fuel volume ratio of 1.6 (16.84mm square lattice pitch for the 2.35 wt% enriched fuel and 18.92mm square lattice pitch for the 4.31 wt% enriched fuel). Either lead or depleted uranium walls were positioned parallel to, on either side of, and at various distances from three subcritical fuel clusters aligned in a row. The results obtained with these undermoderated fuel clusters are similar to those obtained with fuel clusters having near optimum neutron moderation. For either degree of moderation, the critical separation between fuel clusters increased to a maximum as the reflecting walls were moved towards the fuel clusters. This point of maximum effect was observed to occur at an optimum water gap spacing between the fuel and reflecting walls that appeared to be dependent on the degree of neutron moderation in the fuel clusters. The optimum water gap between the depleted uranium reflecting walls and the fuel clusters at the 1.6 water-to-fuel volume ratio covered by this report is near 25mm. In the previous measurements, with fuel clusters near optimum neutron moderation, this optimum water gap was observed to occur at about 20mm. Similarly, the optimum water gap between lead reflecting walls and the undermoderated 1.6 fuel clusters is about 10mm whereas it was less than 5mm for the near optimum moderated fuel clusters. These experimental results for the optimum water gap between the reflecting walls and the fuel clusters are in general agreement with previously reported calculations on heavy metal reflectors.

The experimental data indicate that lead, backed by water and in close proximity to the fuel, is a better neutron reflector than depleted uranium under the same conditions. Although the lead and uranium are of different thickness, the undermoderated experiments covered by this report and the previous experiments with fuel clusters near optimum neutron moderation indicate that with less than 15mm of water between the reflecting walls and the fuel, lead has the better neutron reflecting properties. Similarly, lead and depleted uranium appear to be better reflectors than steel under either condition of moderation and any distance from the fuel.

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PREFACE

The American National Standard - ANSI N16.1-1969 (Nuclear Criticality Safety in Operations With Fissionable Materials Outside Reactors) makes provisions for criticality limits "derived from calculations made by a method shown to be valid by comparison with experimental data, provided sufficient allowances are made for uncertainties in the data and the calculations." This philosophy is widely applied in governmental and industrial operations and accepted by governmental regulatory agencies. Consequently, a principal concern in criticality analysis work is the validation of the codes and nuclear data used in criticality evaluations. To obtain such confirmatory data in the area of transportation and storage of light water reactor (LWR) type fuel, the United States Nuclear Regulatory Commission (NRC) began funding an experimental program in 1976 at the Pacific Northwest Laboratory Critical Mass Laboratory. The primary objective of these NRC experiments was, and still is, to provide clean, well defined, integral data that could be described in calculations exactly as-run without corrections or approximations having to be made. Nearly all of the world's experimental data directly applicable to the criticality assessments of LWR transportation and storage systems have been obtained from the experiments funded by this NRC program.

The report presented herein covers data from the sixth set of experiments in this program. The data were obtained from experimental assemblies that simulated transportation conditions involving either lead or depleted uranium as biological shielding materials. Since one of the earlier reports covered experiments of a similar nature, this current report has been identified as part II for continuity and to avoid confusion.

The same UO_2 fuel, lattice grid plates, neutron absorber plates, and biological materials have been used throughout these experiments. However, during this period of time, some of these parameters have become better defined as a result of repeated analysis. For example, the 4.31 wt% ^{235}U enriched UO_2 rods were originally identified as having a ^{235}U enrichment of 4.29 wt%. Multiple analysis of the rods during the course of these six sets of experiments have resulted in the more correct average of 4.31 wt% quoted in this and some of the more recent reports. Although there are no redefinitions of significance, the values quoted in this report should be considered the latest and best values to use.

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CRITICALITY EXPERIMENTS WITH
SUBCRITICAL CLUSTERS OF 2.35 WT% AND 4.31 WT% ^{235}U
ENRICHED UO_2 RODS IN WATER WITH
URANIUM OR LEAD REFLECTING WALLS - VOLUME 2

INTRODUCTION

A research program, funded by the United States Nuclear Regulatory Commission, to provide experimental criticality data on conditions simulating light water reactor (LWR) fuel shipping and storage configurations was begun in 1976 at the Critical Mass Laboratory of the Pacific Northwest Laboratory. The primary objective of this program is to provide data suitable for use in confirming calculational techniques and nuclear data used in the criticality assessment of LWR type fuel element shipping packages. It is anticipated that the data would be applicable to criticality assessments of similar conditions encountered in such areas as fuel element handling and storage facilities.

Conditions that must be considered, and thus simulated by the experiments, involve subcritical clusters of rods (fuel elements) separated in space with and without neutron absorbing and biological shielding materials. The results from 5 sets of earlier experiments in this program have been reported on previously(1,2,3,4,5) and have provided data on a wide assortment of conditions which are commonly encountered, or could be expected to occur, in shipping and storage systems. These conditions and the experiments are briefly summarized as follows:

- Fuel clusters of 2.35 wt% ^{235}U enriched UO_2 rods immersed in water at near optimum neutron moderation with neutron absorber plates between the fuel clusters.
- Fuel clusters of 4.31 wt% ^{235}U enriched UO_2 rods immersed in water at near optimum neutron moderation with neutron absorber plates between the fuel clusters.
- Fuel clusters of 2.35 wt% and 4.31 wt% ^{235}U enriched UO_2 rods immersed in water at near optimum neutron moderation with lead and depleted uranium reflecting walls on either side of the fuel clusters.
- Fuel clusters of 2.35 wt% and 4.31 wt% ^{235}U enriched UO_2 rods immersed in water at a 1.6 water-to-fuel volume ratio with neutron absorber plates and flux traps between the fuel clusters. Measurements were also made with some fuel clusters containing water holes.
- Fuel clusters of 2.35 wt% and 4.31 wt% ^{235}U enriched UO_2 rods immersed in water with steel reflecting walls on either side of the fuel clusters. Measurements were made with the fuel clusters at an undermoderated water-to-fuel volume ratio of 1.6 and at a neutron moderation near optimum for each of the fuels.

The conditions simulated by the experiments covered in this report involved undermoderated fuel clusters of either 2.35 wt% or 4.31 wt% ^{235}U enriched UO_2 rods in the presence of lead and depleted uranium biological shielding materials. The previous experiments with these materials were performed, as indicated above, with the fuel clusters at near optimum neutron moderation. In this series of experiments the effect that depleted uranium and lead reflecting walls have on the critical separation between fuel clusters is investigated at the undermoderated water-to-fuel volume ratio of 1.6 typical of Pressurized Water Reactor (PWR) and Boiling Water Reactor (BWR) type fuel elements.

EXPERIMENTS

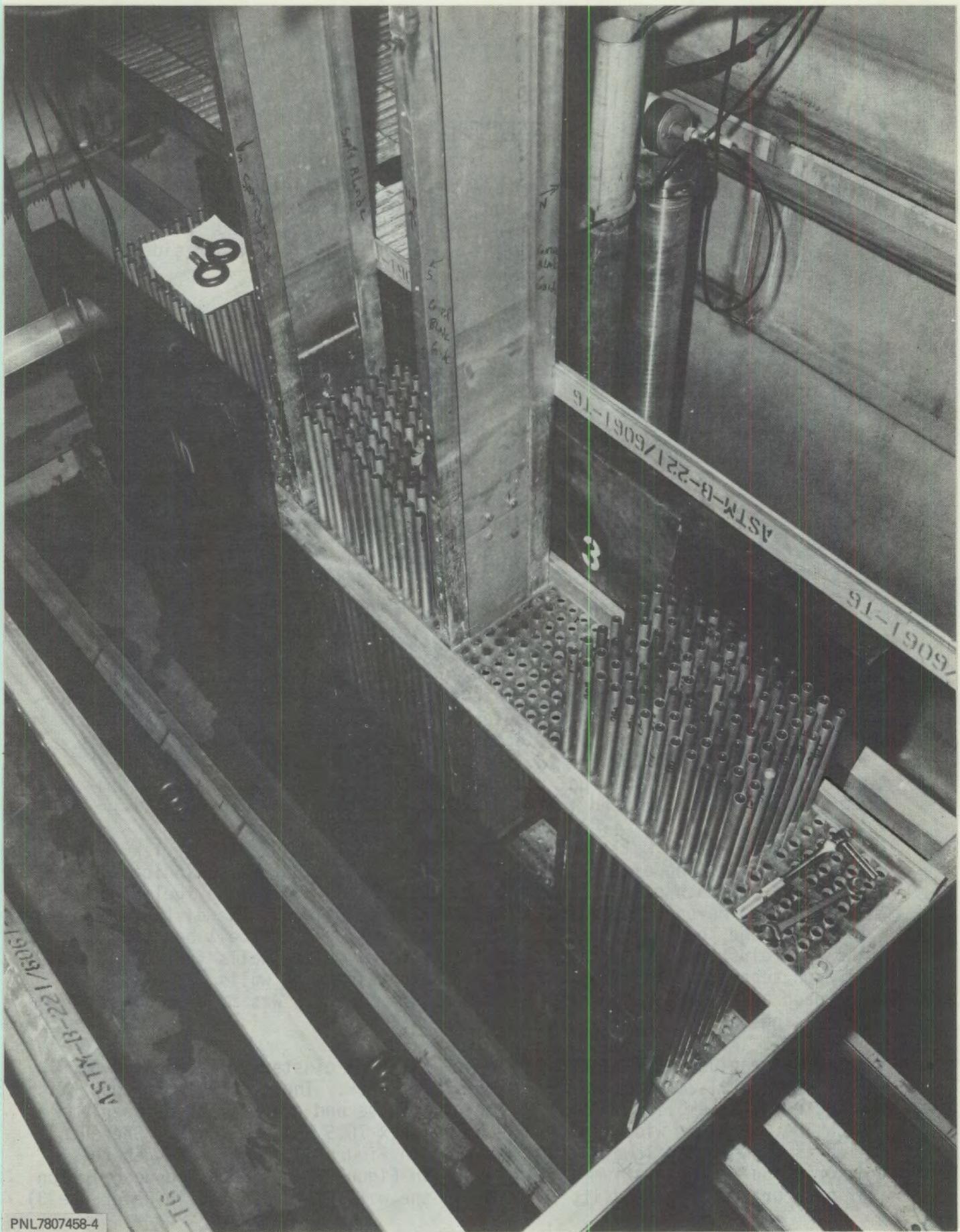
As in the previous experiments with lead and depleted uranium reflecting walls, these current experiments consisted of determining the critical separation between three sub-critical clusters of fuel rods aligned in a row with reflecting walls of lead or depleted uranium parallel on either side of, and at various distances from, the row of fuel clusters.

A photograph of a typical assembly, with depleted uranium walls partially constructed, is shown in Figure 1. The system is provided with a safety and a control blade. Both of these are shown, fully inserted, on either side of the center fuel cluster in Figure 1. Both blades would be fully withdrawn whenever data were being obtained. Also, of course, the entire system would be flooded with water to a depth of at least 150mm above the top of the fuel before any measurements were made. A detailed graphic layout of the experimental system is given in Figure 2. All structural materials are on the periphery of the experimental assembly except for four 51mm wide type 6061 aluminum spacer bars between the fuel and reflecting walls, the lattice grid plates and the aluminum guides for the safety and control blades. The lattice grid plates were fabricated from 12.7mm thick polypropylene(0.904 g/cm³) sheets.

Measurement data were obtained for both the 2.35 wt% and the 4.31 wt% ²³⁵U enriched UO₂ rods in square pitched, water flooded, lattices. Data were obtained for each enrichment at neutron moderation in the fuel clusters approximating that found typically in Boiling Water Reactor and Pressurized Water Reactor fuel elements (1.6 water-to-fuel volume ratio). In the measurements, the distance between the outer cell boundaries of the fuel clusters and the near surface of the reflecting walls was varied from zero to that approaching infinity (removal of the walls from the system). A detailed description of each type fuel rod is given in Figure 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 positioned on either side of the row of fuel clusters as indicated in Figure 2. In each case the walls were 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, a critical approach on the water separation between fuel clusters was made by incrementally decreasing the spacing separating the fuel clusters.

The uranium walls on either side of the fuel clusters were about 1.5m long by about 1.2m high and were about 76mm thick. These walls were constructed by assembling, 5 long by 2 high, tongue and grove slabs of uranium each 304.6 ± 0.9 mm wide $\times 609.5 \pm 2.5$ mm high $\times 76.5 \pm 0.4$ mm thick. One such slab is shown being lowered into position in Figure 4. A complete general description of the uranium wall is given in Figure 5. (A detailed description of each uranium slab and its location in the walls is presented in Reference 3).



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FIGURE1. PHOTOGRAPH OF A TYPICAL EXPERIMENTAL ASSEMBLY

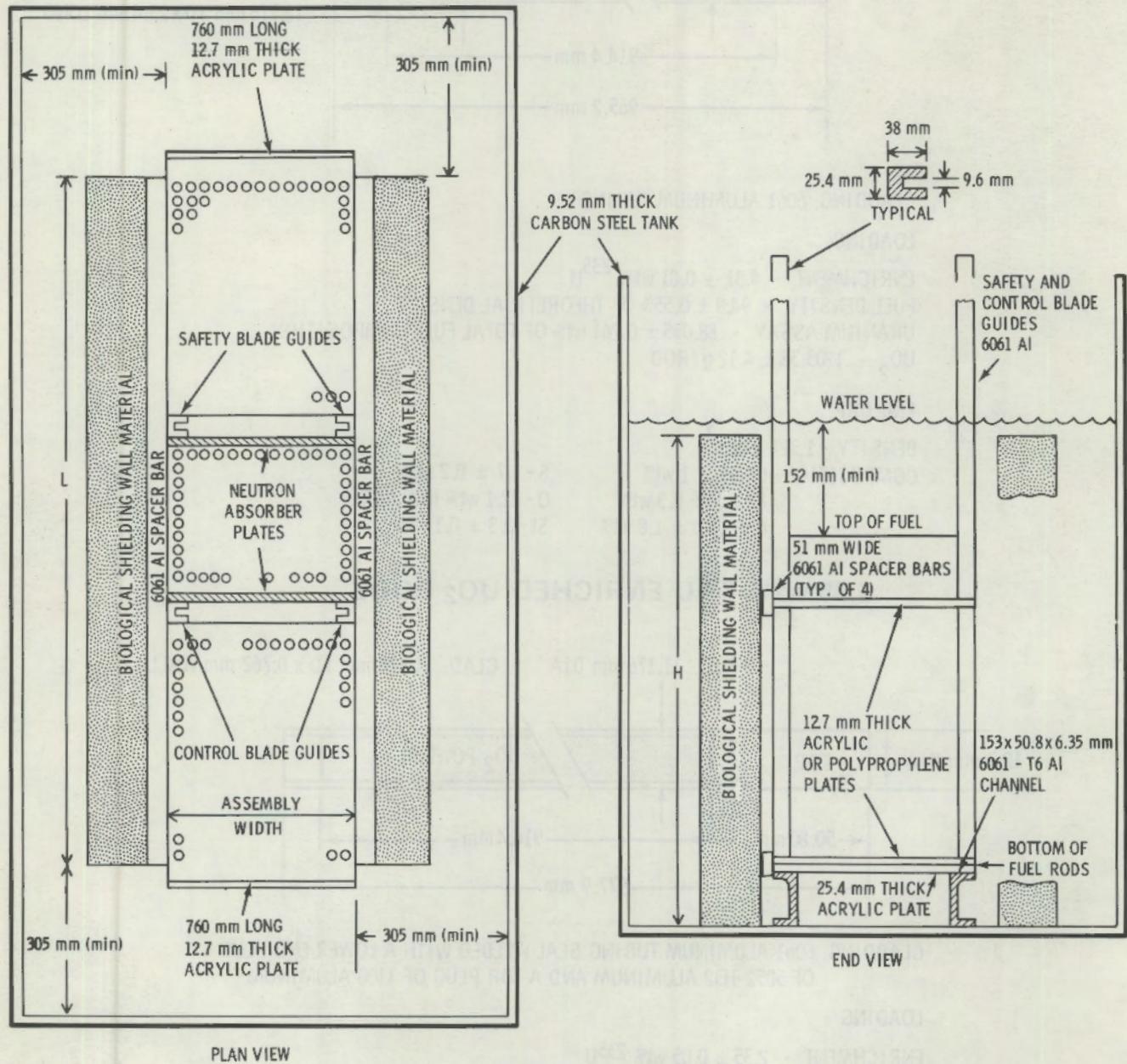
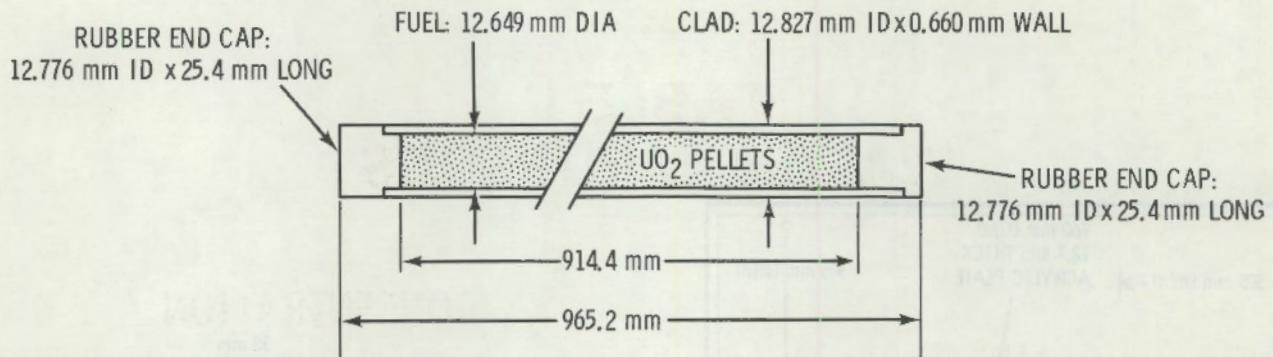


FIGURE 2. GRAPHICAL ARRANGEMENT OF SIMULATED SHIPPING CONTAINER CRITICAL EXPERIMENTS

8106234-1

4.31 wt% ^{235}U ENRICHED UO_2 RODS



CLADDING: 6061 ALUMINUM TUBING

LOADING:

ENRICHMENT - 4.31 ± 0.01 wt% ^{235}U

FUEL DENSITY - $94.9 \pm 0.55\%$ OF THEORETICAL DENSITY

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

UO_2 - 1203.38 ± 4.12 g / ROD

END CAP:

DENSITY - 1.321 g/cm^3

COMPOSITION- C - 58 ± 1 wt%

H - 6.5 ± 0.3 wt%

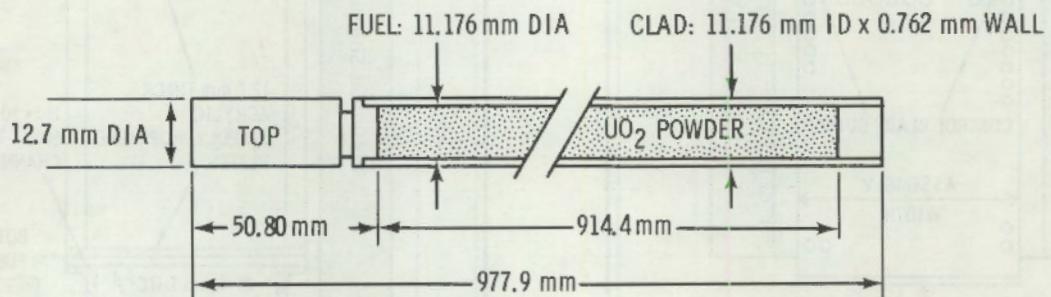
Ca - 11.4 ± 1.8 wt%

S - 1.7 ± 0.2 wt%

O - 22.1 wt% (BALANCE)

Si - 0.3 ± 0.1 wt%

2.35 wt% ^{235}U ENRICHED UO_2 RODS



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/mm^3 (84% THEORETICAL DENSITY)

URANIUM ASSAY - 88.0 wt%

UO_2 - 825 g / ROD (AVERAGE)

FIGURE 3. DESCRIPTION OF FUEL RODS

TABLE I
WATER IMPURITIES

COMPONENT	CONCENTRATION g / m ³ (ppm)
Cl	≤5
NO ₃	0.02
Cr ⁺⁶	<0.01
Zn	16
Mn	<0.01
Pb	<0.005
F	0.18
Fe	24
Cu	<0.01
Cd	0.001
SO ₃	14.5
DISSOLVED SOLIDS	61 ±3

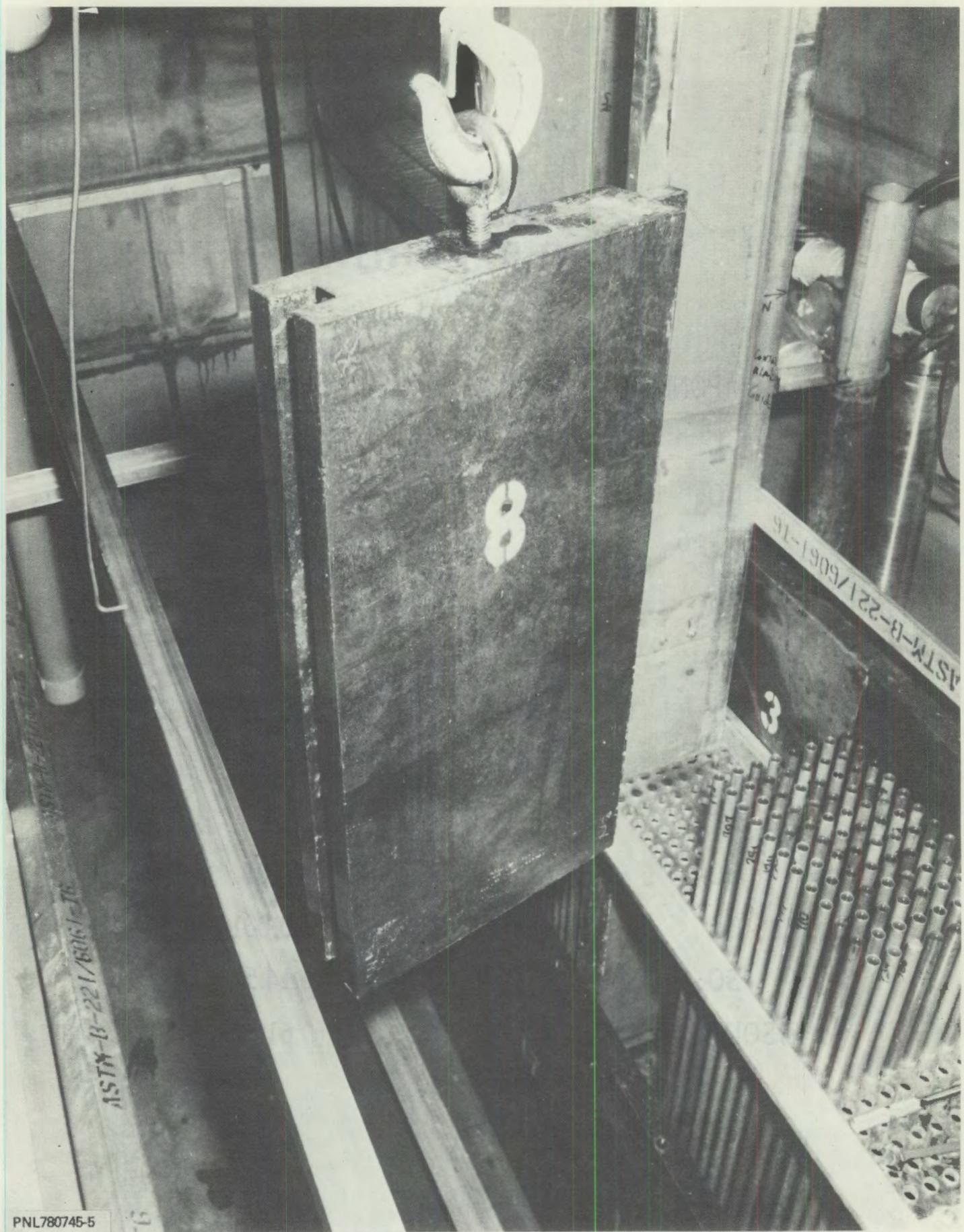
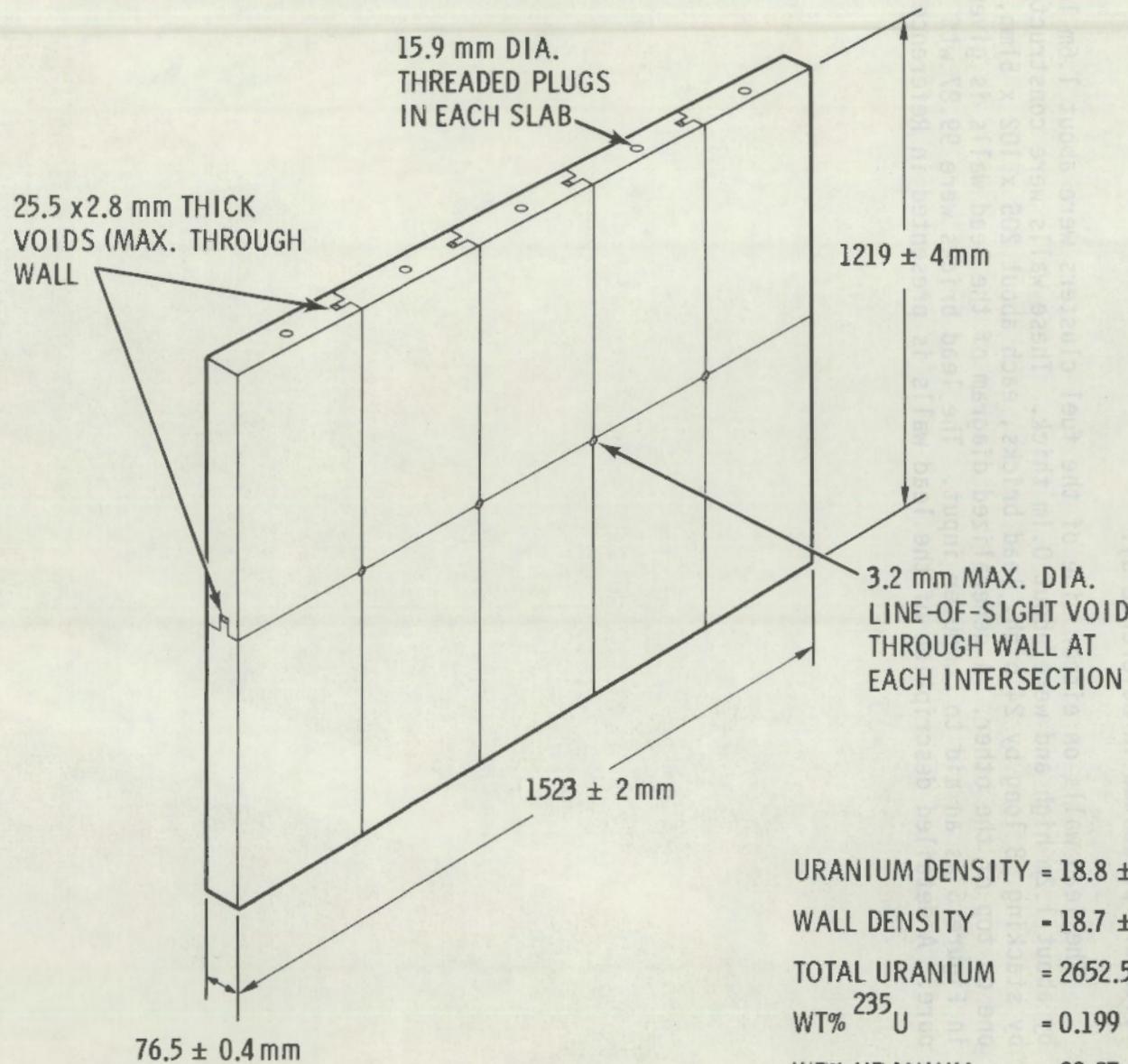


FIGURE 4. DEPLETED URANIUM SLAB BEING LOWERED INTO POSITION

FIGURE 5. ASSEMBLED URANIUM WALL



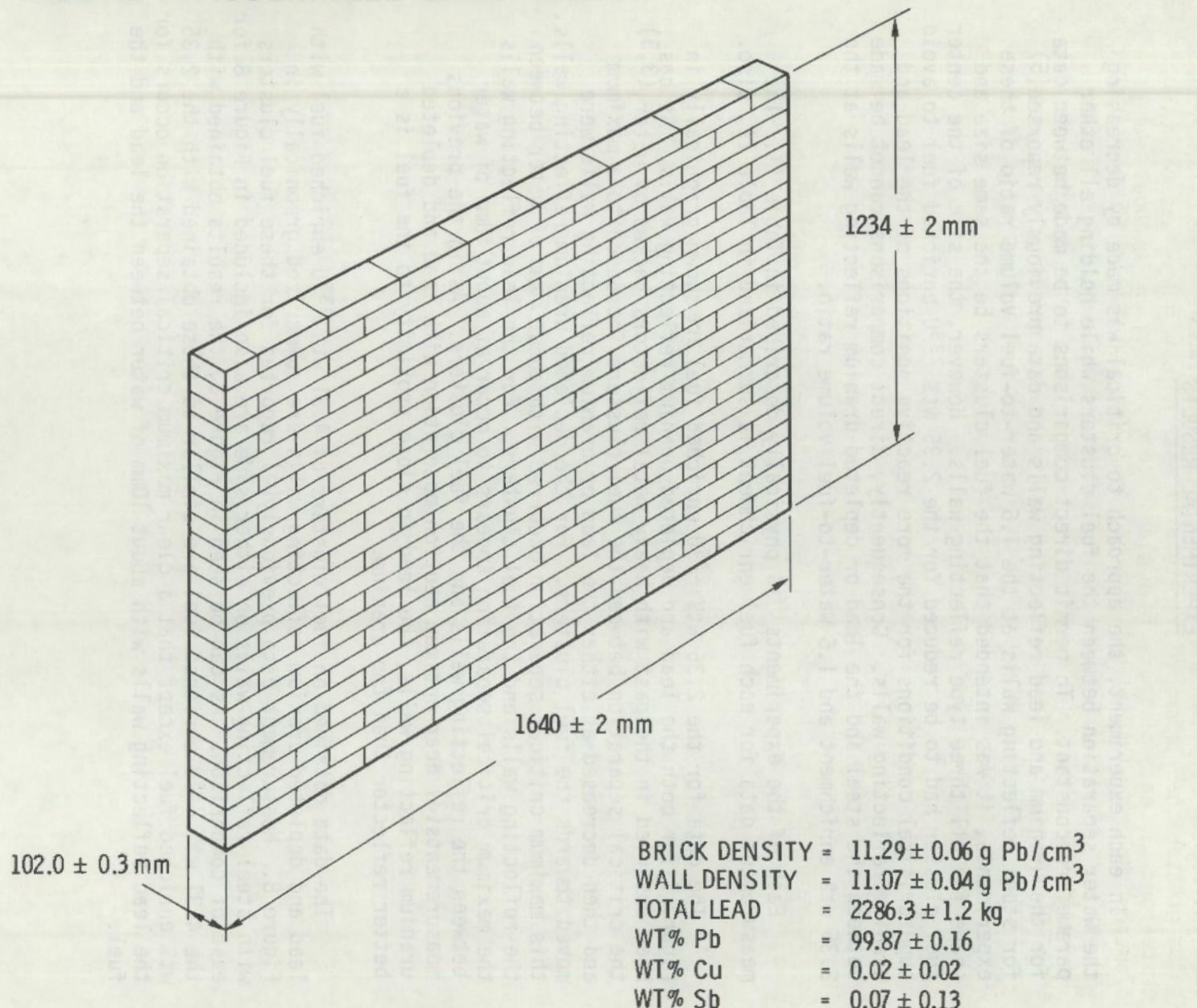
URANIUM DENSITY	$= 18.8 \pm 0.1 \text{ g/cm}^3$
WALL DENSITY	$= 18.7 \pm 0.1 \text{ g/cm}^3$
TOTAL URANIUM	$= 2652.5 \pm 2.5 \text{ kg/WALL}$
WT% ^{235}U	$= 0.199 \pm 0.002$
WT% URANIUM	$= 99.87 \pm 0.10$

Each individual uranium slab was radiographed along its entire length to assure uniform density in each slab and that each slab was free of internal voids greater than 1.5mm in diameter.⁽³⁾ All except four slabs met this criterion. In each of these four slabs, three voids, up to about 6mm in diameter and of undefined thicknesses, were observed within 125mm of either the top or bottom edges of the slabs. These four slabs were positioned in the walls such that these slightly non uniform areas were located on the edges of the walls. The assembled walls were also radiographed to determine the size of line-of-sight voids which occur at the intersection of four adjoining slabs in the walls (see Figure 5). (Radiographs showing the largest voids are presented in Reference 3).

The lead walls on either side of the fuel clusters were about 1.6m long by about 1.2m high and were about 0.1m thick. These walls were constructed by stacking, 8 long by 24 high, lead bricks, each about 205 x 102 x 51mm, one on top of the other. A generalized diagram of the lead walls is given in Figure 6 as an aid to computer input. The lead bricks were 99.87 wt% pure. A detailed description of the lead walls is presented in Reference 3.

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FIGURE 6. ASSEMBLED LEAD WALL



EXPERIMENTAL RESULTS

In each experiment, the approach to critical was made by decreasing the water separation between the fuel clusters while holding all other parameters constant. To permit direct comparisons to be made between data for the uranium and lead reflecting walls and data previously reported(5) for steel reflecting walls at the 1.6 water-to-fuel volume ratio of these experiments, it was intended that the fuel clusters be the same size and shape for all three type reflecting walls. However, the size of the center fuel cluster had to be reduced for the 2.35 wt% ^{235}U enriched fuel to avoid supercritical conditions for the more reactive positions of the lead and uranium reflecting walls. Consequently, direct comparisons cannot be made between the steel and the lead or depleted uranium reflecting walls at the 2.35 wt% enrichment and 1.6 water-to-fuel volume ratio.

Each of the experiments is physically described in Table II. The measurement data for each fuel enrichment is summarized in Table II, also.

The data for the 2.35 wt% ^{235}U enriched fuel are shown graphically in Figure 7 for both the lead and depleted uranium reflecting walls. As has been observed in the past with heavy metal reflectors backed by water,(3,5) the critical separation between the fuel clusters increased to a maximum and then decreased as either the lead or uranium reflecting walls were moved towards the fuel clusters. For the depleted uranium reflecting walls, this maximum critical separation occurred with about 25mm of water between the reflecting walls and the fuel clusters. For the lead reflecting walls the maximum critical separation appears to occur at about 10mm of water between the reflecting walls and the fuel clusters. As in the previous measurements(3) near optimum neutron moderation with lead and depleted uranium reflecting walls, the lead in close proximity to the fuel is a better reflector than the uranium.

The data obtained on fuel clusters of 4.31 wt% ^{235}U enriched fuel with lead and depleted uranium reflecting walls are summarized graphically in Figure 8. Measurement data previously(5) reported for these fuel clusters with steel reflecting walls on either side are also included in Figure 8 for ease of comparison. As can be seen in Figure 8, the results obtained with the 4.31 wt% ^{235}U enriched fuel are similar to those obtained with the 2.35 wt% enriched fuel except that a clear maximum critical separation occurs for the lead reflecting walls with about 10mm of water between the lead and the fuel.

TABLE II
**EXPERIMENTAL DATA ON CLUSTERS OF 2.35 wt% AND 4.31 wt% ^{235}U ENRICHED
 UO_2 RODS IN WATER WITH DEPLETED URANIUM OR LEAD
 REFLECTING WALLS^(a)**

DISTANCE BETWEEN REFLECTING WALLS AND FUEL CLUSTERS (b) (mm)	2.35 wt% ENRICHED FUEL				4.31 wt% ENRICHED FUEL			
	FUEL CLUSTERS (c)	CRITICAL SEPARATION BETWEEN FUEL CLUSTERS (d)		FUEL CLUSTERS (c)	CRITICAL SEPARATION BETWEEN FUEL CLUSTERS (d)		URANIUM WALLS (e) (mm)	LEAD WALLS (f) (mm)
		URANIUM WALLS (e) (mm)	LEAD WALLS (f) (mm)		URANIUM WALLS (e) (mm)	LEAD WALLS (f) (mm)		
0	1-23x18 2-20x18	80.5 \pm 0.2	100.5 \pm 0.3	3-12x16	153.3 \pm 0.3	177.4 \pm 0.3		
5.50 \pm 1.02	1-23x18 2-20x18	-	101.1 \pm 0.2	3-12x16	167.1 \pm 0.2	181.8 \pm 0.5		
13.21 \pm 0.76	1-23x18 2-20x18	95.0 \pm 0.3	-	3-12x16	182.7 \pm 0.5	-		
19.56 \pm 1.02	-	-	-	3-12x16	192.4 \pm 0.4	174.3 \pm 0.2		
25.15 \pm 1.07	1-23x18 2-20x18	98.3 \pm 0.2	-	3-12x16	193.7 \pm 0.4	-		
32.75 \pm 1.48	1-23x18 2-20x18	-	85.0 \pm 0.2	3-12x16	187.8 \pm 0.2	-		
39.12 \pm 1.32	1-23x18 2-20x18	91.9 \pm 0.3	-	-	-	-		
54.05 \pm 1.02	-	-	-	3-12x16	164.0 \pm 0.2	143.5 \pm 0.3 (g)		
∞	1-23x18 2-20x18	-	65.9 \pm 0.2	3-12x16	128.5 \pm 0.2	129.7 \pm 0.3 (h)		
∞	1-25x18 2-20x18	-	72.2 \pm 0.2 (i)	-	-	-		

(a) ERROR LIMITS SHOWN ARE ONE STANDARD DEVIATION

(b) PERPENDICULAR DISTANCE BETWEEN THE CELL BOUNDARY OF THE FUEL CLUSTERS AND THE REFLECTING WALLS

(c) NUMBER OF FUEL CLUSTERS, RODS LONG x RODS WIDE, ALIGNED IN A ROW

(d) PERPENDICULAR DISTANCE BETWEEN FUEL CLUSTERS, ROD SURFACE TO ROD SURFACE

(e) WALLS 76.5 ± 0.4 mm THICK

(f) WALLS 102.0 ± 0.3 mm THICK

(g) DISTANCE BETWEEN REFLECTING WALLS AND FUEL CLUSTERS IS 50.01 ± 0.86 mm

(h) CRITICAL SEPARATION PREVIOUSLY REPORTED IN NUREG/CR-1784

(i) CRITICAL SEPARATIONS OF 72.4 AND 71.9 mm OBTAINED IN PREVIOUS EXPERIMENTS AT THIS LATTICE PITCH. NO PREVIOUS MEASUREMENTS WITH CENTER CLUSTER 23 RODS LONG.

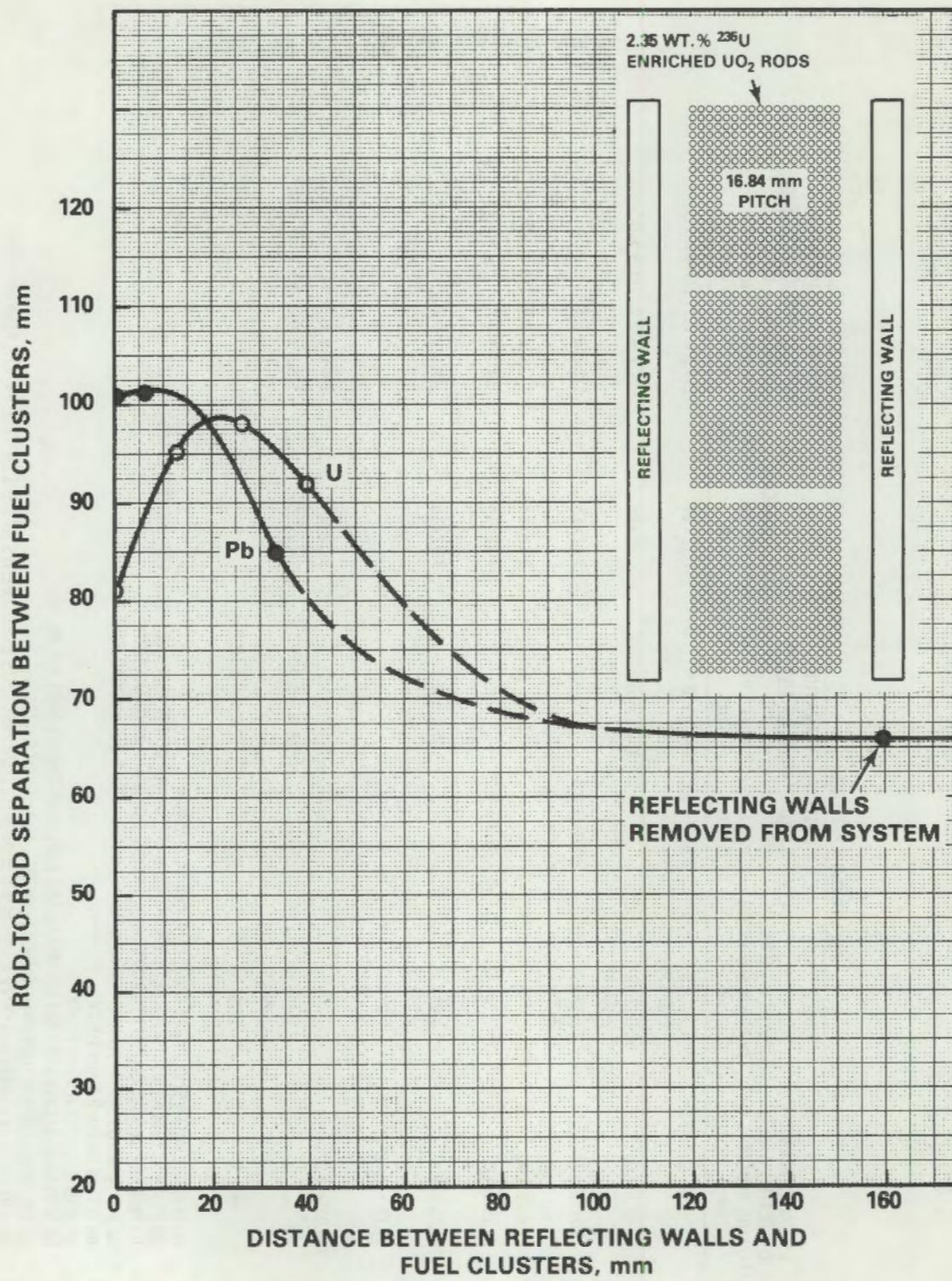


FIGURE 7. CRITICAL SEPARATION BETWEEN UNDER-MODERATED FUEL CLUSTERS OF 2.35 WT% ^{235}U ENRICHED UO_2 RODS IN WATER WITH LEAD OR DEPLETED URANIUM REFLECTING WALLS

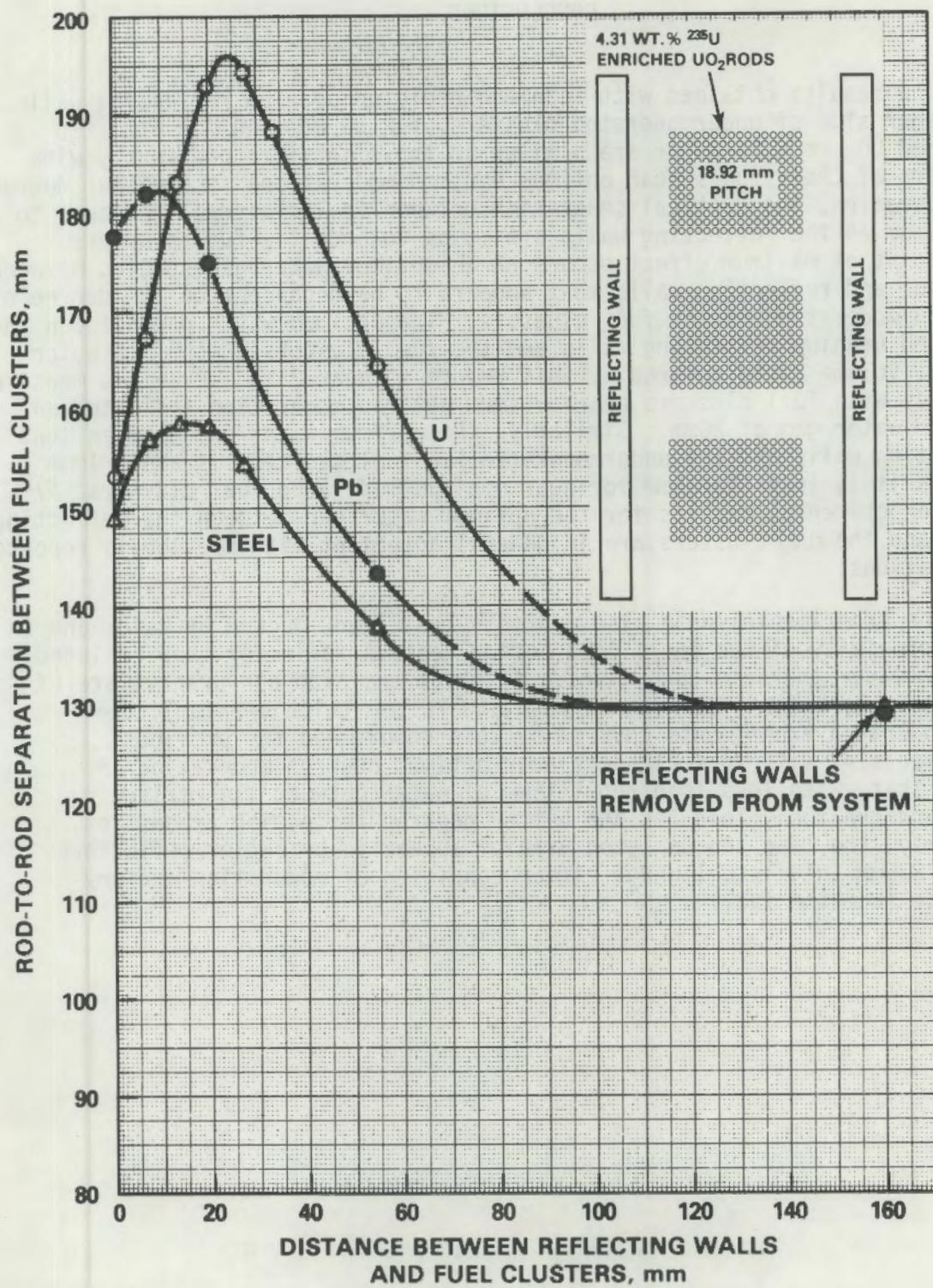


FIGURE 8. CRITICAL SEPARATION BETWEEN UNDER-MODERATED FUEL CLUSTERS OF 4.31 WT% ^{235}U ENRICHED UO_2 RODS IN WATER WITH STEEL, LEAD OR DEPLETED URANIUM REFLECTING WALLS

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CONCLUSIONS

The results obtained with lead and depleted uranium reflecting walls on either side of undermoderated clusters of 2.35 wt% and 4.3 wt% ^{235}U enriched UO_2 rods in water are similar to those obtained previously with clusters of these fuels near optimum neutron moderation. For either degree of moderation, the critical separation between fuel clusters increases to a maximum as the reflecting walls are moved towards the fuel clusters. This point of maximum effect occurs at an optimum water gap spacing between the fuel and reflecting walls that appears to be dependent on the degree of neutron moderation in the fuel clusters. The optimum water gap between the depleted uranium reflecting walls and the fuel clusters at the 1.6 water-to-fuel volume ratio covered by this report is near 25mm. Previous measurements(3) with fuel clusters near optimum neutron moderation indicated an optimum water gap of 20mm. Similarly, the optimum water gap between lead reflecting walls and the undermoderated 1.6 fuel clusters is about 10mm whereas it is less than 5mm for near optimum moderated fuel clusters.(3) These experimental results for the optimum water gap between the reflecting walls and the fuel clusters are in general agreement with previously reported calculations.

The experimental data also indicate that lead, backed by water and in close proximity of the fuel, is a better neutron reflector than depleted uranium under the same conditions. Although the lead and uranium are of different thickness (102mm thick lead and 76.5mm thick uranium), the undermoderated experiments covered by this report and the previous experiments(3) with fuel clusters near optimum neutron moderation indicate that, with less than about 15mm of water between the reflecting walls and the fuel, lead has the better neutron reflecting properties. Similarly, the lead and depleted uranium appear to be better reflectors than 178.5mm thick steel under either condition of moderation and any distance from the fuel.

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The results from the sixth in a series of criticality experiments, are presented in this paper. This sixth set of experiments involve clusters of either 2.35 wt% or 4.31 wt% ^{235}U enriched UO_2 fuel rods immersed in water as in previous experiments. This latest set of measurements are concerned with determining the effect that lead and depleted uranium reflecting walls have on the critical separation between fuel clusters having a 1.6 water-to-fuel volume ratio.

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