

## REFERENCE 84

**E. B. JOHNSON AND R. K. REEDY, JR., "CRITICAL EXPERIMENTS WITH SPERT-D FUEL ELEMENTS," OAK RIDGE NATIONAL LABORATORY REPORT ORNL - TM- 1207 (JULY 1965).**



**OAK RIDGE NATIONAL LABORATORY**  
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**UNION CARBIDE CORPORATION**  
for the  
**U.S. ATOMIC ENERGY COMMISSION**



ORNL-TM-1207

DATE - July 14, 1965

CRITICAL EXPERIMENTS WITH SPERT-D FUEL ELEMENTS

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ABSTRACT

The critical dimensions of lattices of SPERT-D fuel elements in several non-reactor environments were determined to establish specifications for use in storage, transportation, and chemical processing operations. Each fuel element contained about 300 g of  $^{235}\text{U}$  in 22 aluminum-clad flat plates. In addition to lattices with water moderator and reflector, a dilute aqueous solution of uranyl nitrate was used in some experiments to simulate a dissolver. In still other experiments, varying amounts of boron were added to the uranyl nitrate solution to determine its effect as a soluble neutron absorber in chemical process equipment.

It was shown, for example, that a minimum of about 3.5 kg of  $^{235}\text{U}$  is required in a critical lattice moderated and reflected by water. This mass is reduced to 2.8 kg (contained in the elements) when U(92,6) solution having a  $^{235}\text{U}$  concentration of 3.99 g/liter was substituted for water. It was increased to 13.6 kg when 1.118 g of natural boron was added to each liter of the uranyl nitrate solution.

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## CRITICAL EXPERIMENTS WITH SPERT-D FUEL ELEMENTS

## INTRODUCTION

A part of the continuing program of the Critical Experiments Facility at ORNL is the generation of data, primarily for the  $^{235}\text{U}$  isotope, which are required in various areas of the nuclear industry. An attempt is made to obtain data under conditions, commensurate with availability of materials, schedules, and recognized importance, such that they are applicable to a variety of problems and extend or supplement existing information. The data so generated are used for the safe and economic solution of problems associated with the handling of fissile materials outside conventional nuclear reactors.

One problem of recurrent concern to plant design and operating personnel, as well as to the economist, is the batch size, in terms of quantity of fissile materials, which can be safely charged into a dissolver of specified dimensions, and the attendant precautions which may be necessary to assure criticality safety.

A case in point is a dissolver at the Idaho Chemical Processing Plant, operated by Phillips Petroleum Company at NRTS, in which it is intended that the fuel elements from the first core of the Pressurized Water Reactor (PWR) be put into solution. Since each of these fuel elements contains more  $^{235}\text{U}$  (about 300 g in zircalloy-clad plates) than elements for which "clean" critical data have been determined, there was a question of the criticality safety of the dissolver under foreseeable conditions. Consequently, a series of experiments was performed at ORNL to determine the critical dimensions of lattices of these elements in a variety of environments. SPERT-D fuel elements, quite similar in nuclear properties to the first core of the PWR were used because of availability. The resulting critical data will serve as bases for establishing the safety of arrays of these elements both in the dissolver environment, in water storage and in transportation under potential accident conditions. These measurements extend those made previously by Fox and Gilley<sup>1</sup> using ORR elements each containing about 168 g of  $^{235}\text{U}(93)$ .

## DESCRIPTION OF THE ELEMENTS

The SPERT-D "300 g" fuel element is shown in Fig. 1. The outside container was 3-in.-square, type 6060-T6 aluminum tubing, 27.625 in. long. Into two opposite sides of this tube fitted grooved side plates, of the same alloy, which positioned and supported the fuel plates. There were 22 grooves in each side plate. A fuel plate was flat and

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1. J. K. Fox and L. W. Gilley, "Critical Experiments with Arrays of ORR and BSR Fuel Elements," ORNL-CF-58-9-40 (1958).

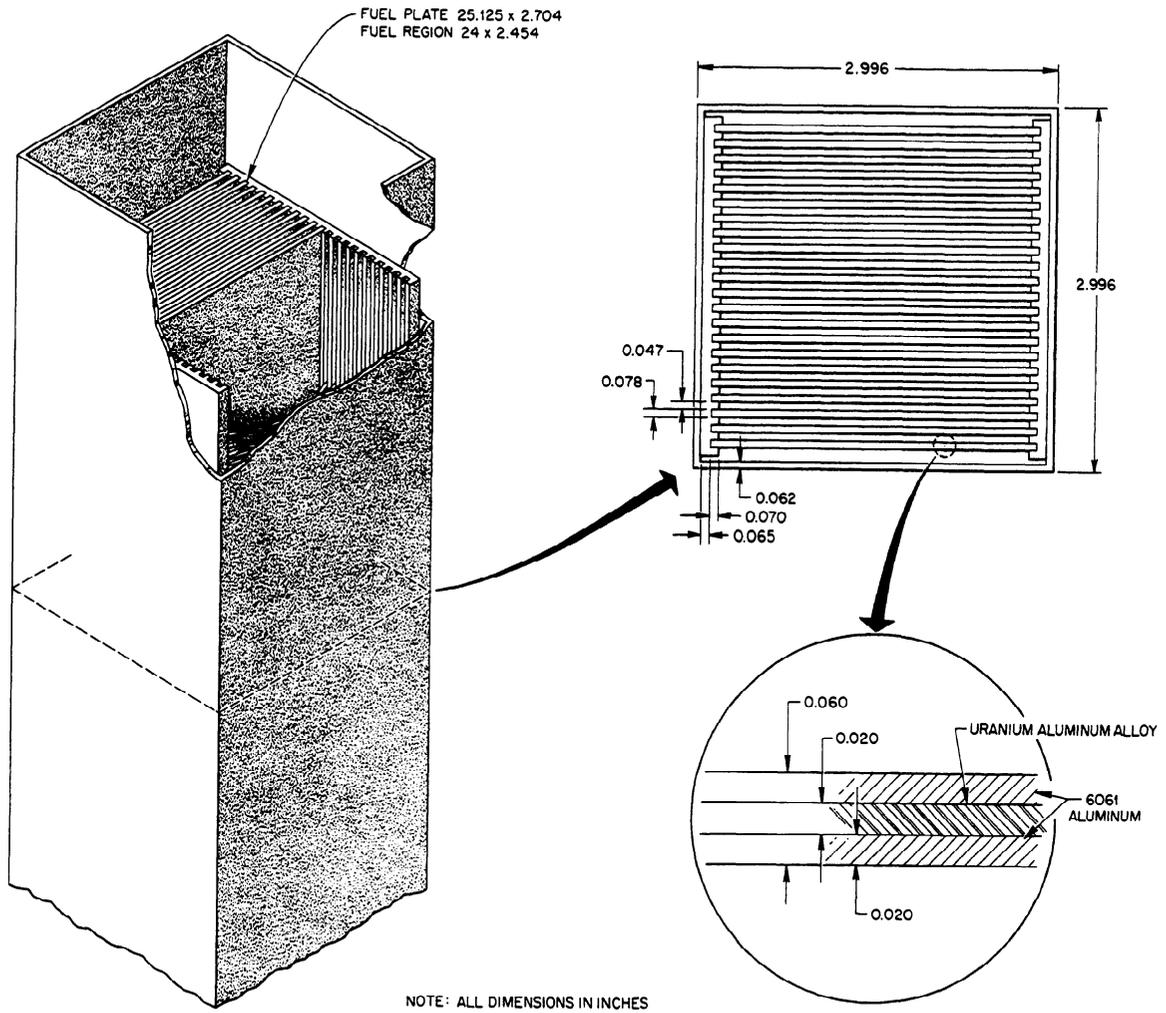


Fig. 1. Sketch of SPERT-D Fuel Element.

consisted of a uranium-aluminum alloy core [U(93.17)-Al], containing 23.8 wt% uranium, completely clad with 0.020-in.-thick type 6061 aluminum; the average  $^{235}\text{U}$  content of each plate was 13.93 g, with individual plates varying between 14.7 and 13.2 g. Each fuel element contained on the average, 306.46 g of  $^{235}\text{U}$  in 22 fuel plates. Within the tolerances of the structure, the distance between the outside of the box and the edge of the fuel, in a direction parallel to the plane of the fuel plates, was  $0.267 \pm 0.042$  in. and, in a direction perpendicular to the plane of the fuel, was  $0.180 \pm 0.025$  in.

Each element was normally equipped with an end box and a handle, each attached with screws, which located the side plates and the fuel plates longitudinally within the outer box. Removal of appropriate end boxes and handles permitted placing two boxes end-to-end. Fuel plates of one element could then be displaced so that their ends were in contact with those of another element, thereby making it possible to simulate elements whose fuel length was a multiple of that of the standard 2-ft element.

#### DESCRIPTION OF MATERIALS

Demineralized water was the reflector and moderator for many of the experiments.

In order to simulate dissolver environment, a dilute (3.99 g of  $^{235}\text{U}$  per liter) aqueous solution of  $\text{U}(92.6)\text{O}_2(\text{NO}_3)_2$  was used as the reflector and moderator in some experiments; the presence of this solution as internal moderator within an element added 6.6 g of  $^{235}\text{U}$  to the fuel content of the element. In still other experiments natural boron, as  $\text{H}_3\text{BO}_3$  was added to this dilute uranyl nitrate solution to obtain boron concentrations varying between 0.389 and 1.118 g per liter. The isotopic analysis of the boron showed 19.72 atom per cent  $^{10}\text{B}$  and 80.26 atom per cent  $^{11}\text{B}$ .

#### EXPERIMENTS

##### Water-Moderated and -Reflected Lattices

Most of the experiments were performed with elements having a nominal 24 in. fuel length. In a few cases, however, longer (6-foot) "elements" were prepared in the manner described previously. In all cases, except as noted, the data describe lattices of 2-ft-long elements.

The experiments by which the optimum moderation and, therefore, the minimum critical mass, of spaced fuel elements was established used lattices which were rectangular in cross section; the structure of the fuel elements allowed adjustment to that number of fuel plates necessary for criticality and, therefore, a much more definitive measure of the critical mass than is possible with fixed-plate elements.

In order to preserve rectangular geometry, each partial element in an outer row contained the same number of fuel plates. The desired spacing between the elements was established and maintained by milled strips of Plexiglas placed between the outer surfaces of the fuel element boxes. One experiment in this series used a "rounded" lattice, in which the cross section approximated a circle, in order to determine the minimum critical mass at the optimum spacing between elements. (No investigation was made of varying the spacing between plates in an element; all plates were spaced in adjacent grooves in the side plates.) A limited number of measurements was made with elements nominally 6 ft long to determine whether effective fuel length influenced the location of the minimum. Table 1 and Fig. 2 show all of these data. Also shown in Fig. 2 are the corresponding results<sup>1</sup> from lattices of ORR fuel elements, each of which contained about 168 g of  $^{235}\text{U}$  in 19 curved plates.

A series of submerged slab lattices was investigated in order to study possible storage configurations. The experience with ORR elements showed that a slab of 16 fuel elements was infinitely long since additional elements did not increase the reactivity. Some of the slabs contained 0.025-in.-thick cadmium sheet between rows; since increasing the length of the cadmium beyond the boundaries of a finite lattice decreased the reactivity, the critical dimensions were determined with cadmium not extending beyond the lattice. Table 2 gives the data for many of the slab geometries which were studied, and for all which were critical; the quoted uncertainties indicate the quantity which was varied, i.e., mass or spacing. An effectively infinitely long slab two elements thick, spaced 0.5 in. between outside surfaces, was subcritical; such a slab at any spacing between elements will be subcritical. Figure 3 is a photograph of two individually subcritical slabs with the elements spaced 0.50 in. between outside of boxes, which were critical when the slabs were separated  $6.37 \pm 0.010$  in. and submerged.

#### Solution-Moderated and -Reflected Lattices

In order to minimize the volume of solution required without severely limiting the number of elements which could be used, the experiments intended to simulate dissolver environment were performed in a stainless steel tank 37.75 in. ID and 35.625 in. high. These dimensions permitted a 4-in.-thick bottom reflector and a 5-in.-thick top reflector on the fuel region. It was not practical to determine whether these dimensions were effectively infinite for the dilute aqueous  $\text{U}(92.6)\text{O}_2(\text{NO}_3)_2$  solution or for the solution with boron added. The elements were spaced within the tank with 0.5 in. between the outside surfaces of the fuel boxes. Although not investigated, this spacing was assumed to give maximum reactivity for the simulated dissolver solutions. The broad minima of Fig. 2 imply that little error is introduced by this assumption.

Table 3 and Fig. 4 summarize the results of these experiments. The series was terminated at a boron concentration of 1.118 g per liter because as many latticed elements were necessary to achieve criticality at this concentration as the tank could contain and still maintain the desired 4-in.-thick, minimum, side reflector. Figure 5 shows the lattice

Table 1. Effect on Critical Mass of Spacing Between SPERT-D Fuel Elements in Rectangular Arrays Moderated and Reflected by Water

Spacing			Dimensions of Lattice (Number of Elements)	Critical Number of Elements	Critical Mass (kg of $^{235}\text{U}$ )
$s^a$ (in.)	$d^b$ (in.)	$d^c$ (in.)			
2-Foot-Long Elements					
0.00	$0.53 \pm 0.08$	$0.36 \pm 0.05$	4 x 3.77	$15.09 \pm 0.10$	$4.63 \pm 0.03$
0.25	$0.78 \pm 0.08$	$0.61 \pm 0.05$	4 x 3.16	$12.64 \pm 0.10$	$3.87 \pm 0.03$
0.50	$1.03 \pm 0.08$	$0.86 \pm 0.05$	4 x 3.09	$12.36 \pm 0.17$	$3.79 \pm 0.05$
0.50 <sup>d</sup>	$1.03 \pm 0.08$	$0.86 \pm 0.05$		$11.35 \pm 0.06$	$3.48 \pm 0.02$
0.75	$1.28 \pm 0.08$	$1.11 \pm 0.05$	4 x 3.16	$12.64 \pm 0.10$	$3.87 \pm 0.03$
1.00	$1.53 \pm 0.08$	$1.36 \pm 0.05$	4 x 3.70	$14.82 \pm 0.10$	$4.54 \pm 0.03$
1.25	$1.78 \pm 0.08$	$1.61 \pm 0.05$	5 x 4.03	$20.14 \pm 0.11$	$6.16 \pm 0.04$
1.50	$2.03 \pm 0.08$	$1.86 \pm 0.05$	6 x 5.34	$32.04 \pm 0.13$	$9.82 \pm 0.04$
1.60	$2.13 \pm 0.08$	$1.96 \pm 0.05$	7 x 6.68	$46.77 \pm 0.31$	$14.33 \pm 0.10$
6-Foot-Long Elements					
0.00	$0.53 \pm 0.08$	$0.36 \pm 0.05$	4 x 3.20	$12.81 \pm 0.11$	$11.78 \pm 0.10$
0.50	$1.03 \pm 0.08$	$0.86 \pm 0.05$	3 x 3.36	$10.09 \pm 0.11$	$9.28 \pm 0.10$
1.25	$1.78 \pm 0.08$	$1.61 \pm 0.05$	4 x 4	$16.00 \pm 0.11$	$14.71 \pm 0.10$

- a. Spacing between outside of fuel boxes.  
 b. Distance between fuel in adjacent elements in a direction parallel to the plane of the fuel plates.  
 c. Distance between fuel in adjacent elements in a direction perpendicular to the plane of the fuel plates.  
 d. This array was approximately circular in cross section in order to minimize the critical mass.

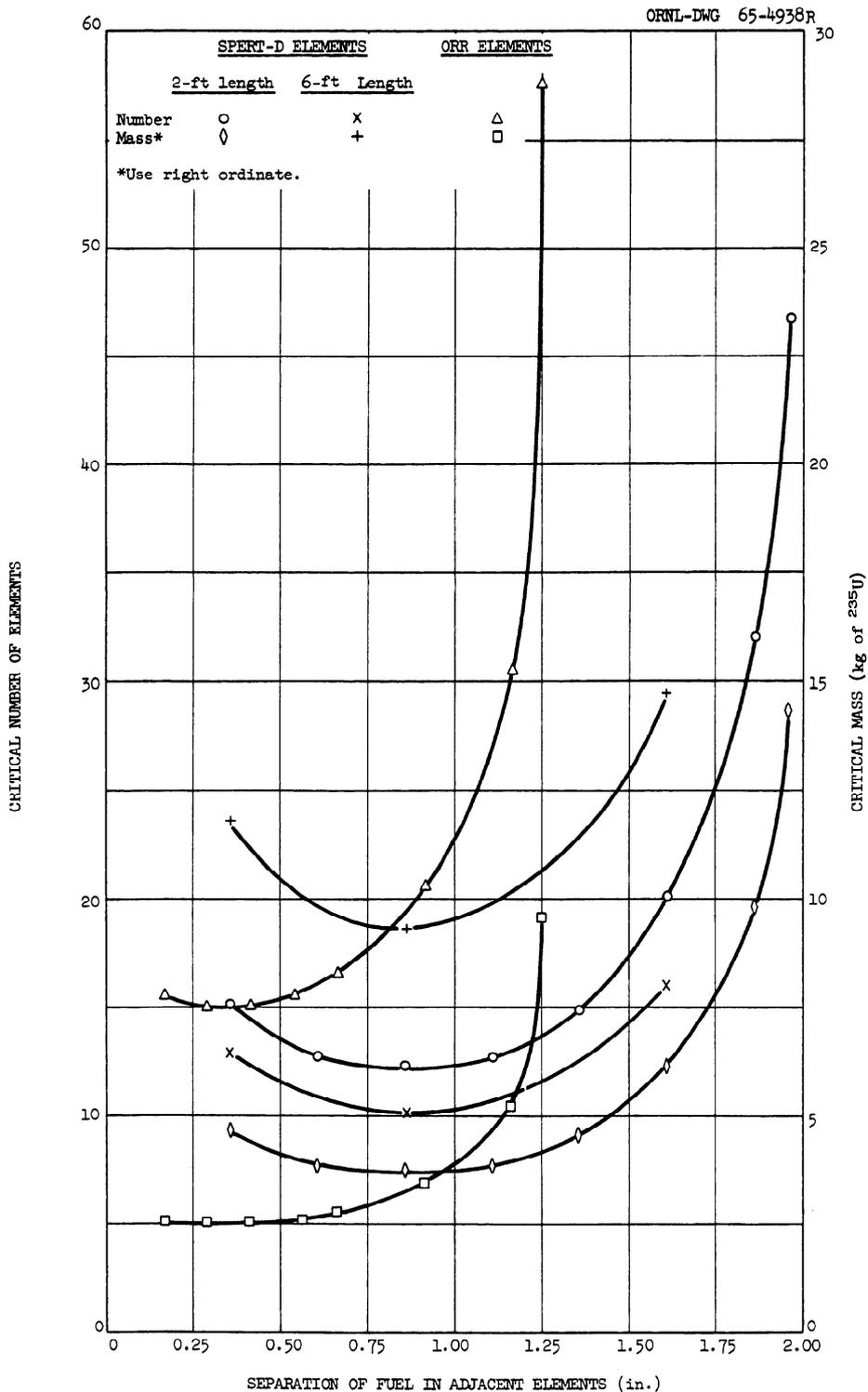


Fig. 2. Critical Number and  $^{235}\text{U}$  Mass of SPERT-D and ORR Fuel Elements as a Function of Fuel Separation Between Adjacent Elements. The full SPERT-D elements each contained 306.5 g of  $^{235}\text{U}$  in 22 plates and the ORR elements each contained about 168 g in 19 plates. The spacing is that between fuel in adjacent elements in a direction perpendicular to the plane of the fuel plates.

Table 2. Critical Slab Lattices of SPERT-D Fuel Elements Moderated and Reflected by Water

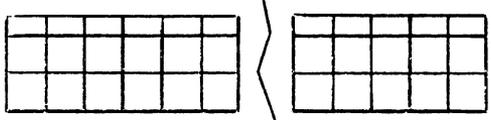
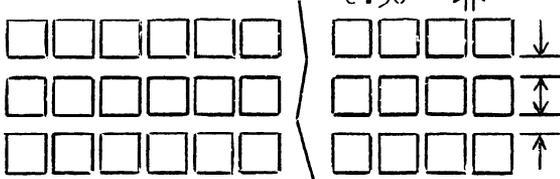
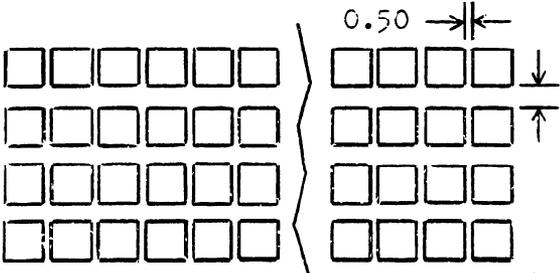
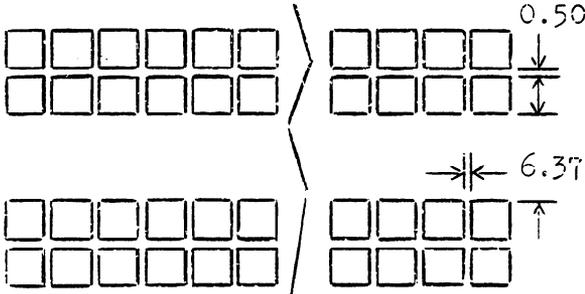
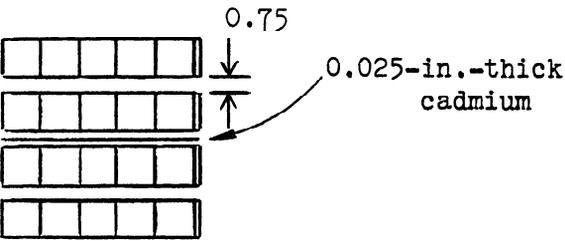
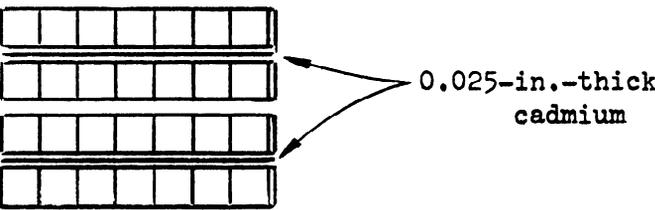
Lattice (All dimensions in in.)	Spacing <sup>a</sup> (in.)		Critical Mass (kg of <sup>235</sup> U)
	Between Elements	Between Rows	
 16 x 2.32 elements	0.00	0.00	11.37 ± 0.20
 16 x 3 elements	0.50	2.19 ± 0.03	14.71
 16 x 4 elements	0.50	2.56 ± 0.02	19.62
 2 slabs, each 16 x 2 elements	0.50	0.50; 6.37 ± 0.01 between slabs	9.81 each 19.62 total
8 x 8 elements; 0.025-in.-thick cadmium between rows	0.50	0.50	19.62; sub-critical

Table 2. (cont.)

Lattice (All dimensions in in.)	Spacing <sup>a</sup> (in.)		Critical Mass (kg of <sup>235</sup> U)
	Between Elements	Between Rows	
 <p>4 x 5.04 elements</p>	0.00	0.75	6.19 ± 0.06
 <p>4 x 7.04 elements</p>	0.00	0.75	8.64 ± 0.05

- a. The spacing is that between the outside of fuel element boxes; the horizontal separation between fuel in adjacent elements, in a direction perpendicular to the plane of the fuel plates, was this spacing plus 0.36 in.; the vertical separation, in a direction parallel to the plane of the fuel plates, was this spacing plus 0.53 in.

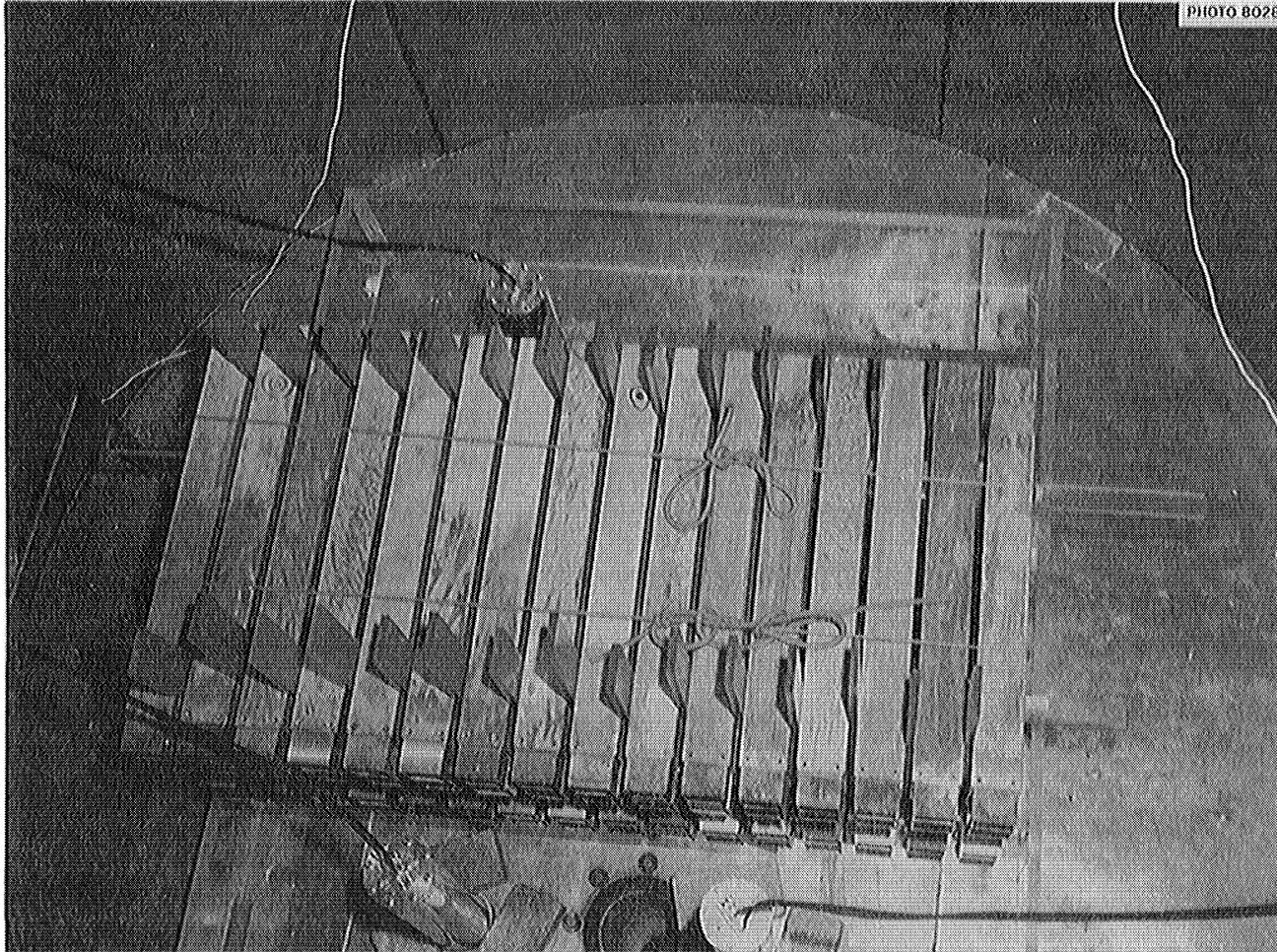


Fig. 3. Two Individually Subcritical 32-Element Slabs of SPERT-D Fuel Elements Forming a Critical System When Submerged. The separation between the elements within each slab was 0.5 in. in both directions; the separation between slabs was 6.37 in.

Table 3. Effect on Critical Mass of Dilute  $U(92.6)O_2(NO_3)_2$  and Boron of Several Concentrations in the Reflector and Moderator Water of Lattices of SPERT-D Fuel Elements Spaced 0.5 in. Between Outside Surfaces

$^{235}U$ Concentration (g/liter)	Boron Concentration (g/liter)	Critical Mass <sup>a</sup> (kg of $^{235}U$ )	Critical Number of Elements	Dimensions of Lattice (No. of Elements)
0	0	$3.48 \pm 0.02^b$	$11.35 \pm 0.06^b$	-
0	0	$3.79 \pm 0.05$	$12.36 \pm 0.17$	4 x 3.09
3.99	0	$2.76 \pm 0.02^b$	$9.01 \pm 0.06^b$	-
3.99	0	$2.86 \pm 0.02$	$9.33 \pm 0.06$	3 x 3.09
3.99	0.389	$5.15 \pm 0.03$	$16.80 \pm 0.10$	4 x 4.20
3.99	0.579	$6.76 \pm 0.03$	$22.06 \pm 0.10$	5 x 4.41
3.99	0.773	$8.90 \pm 0.03$	$29.04 \pm 0.10$	6 x 4.96
3.99	0.871	$10.15 \pm 0.03$	$33.12 \pm 0.10$	6 x 5.55
3.99	1.118	$13.6 \pm 0.05^b$	$44.38 \pm 0.16^b$	-

a. The values do not include 6.6 g of  $^{235}U$  introduced into each element by the aqueous  $U(92.6)O_2(NO_3)_2$  moderator solution at a concentration of 3.99 g of  $^{235}U$ /liter.

b. These lattices were approximately circular in cross section; all others were rectangular.

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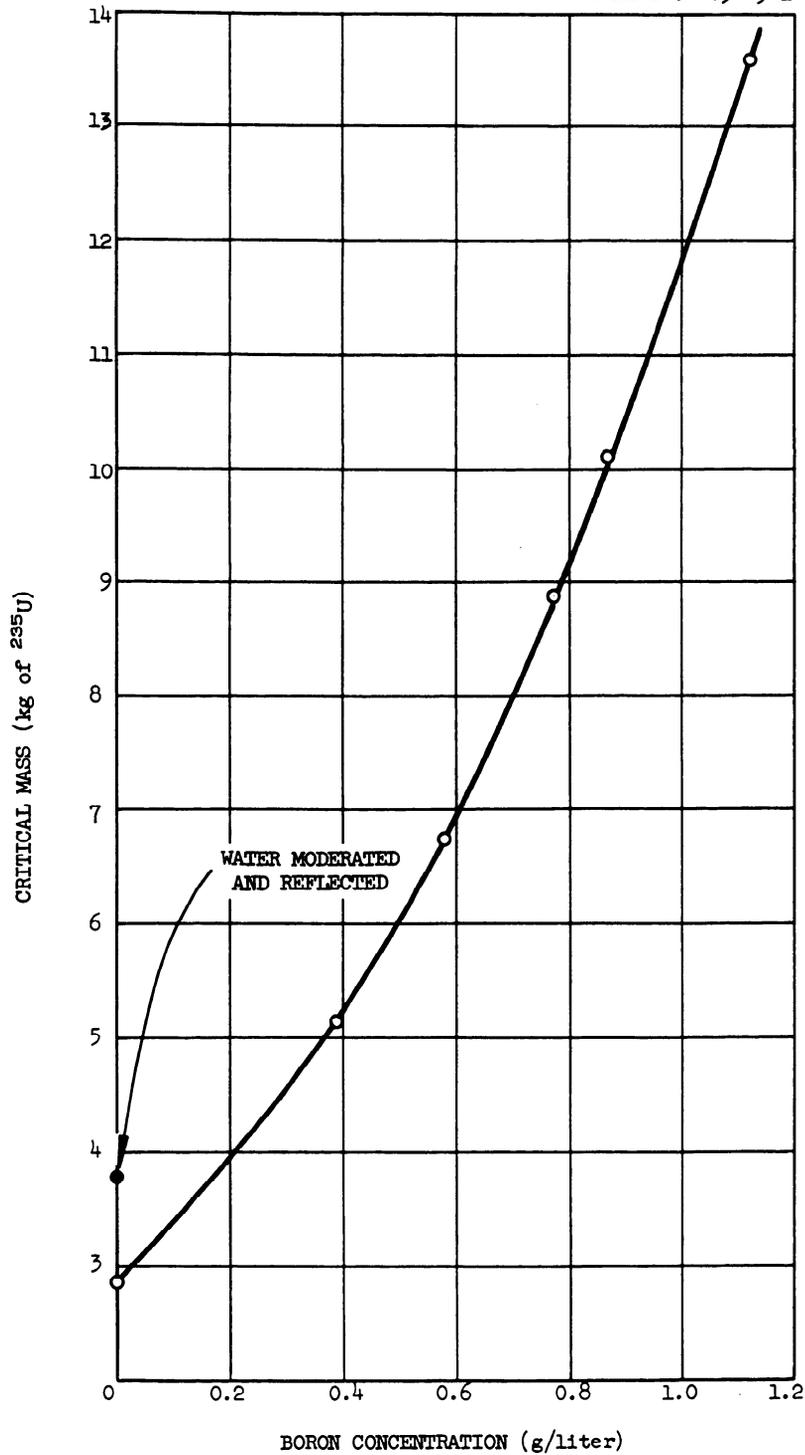


Fig. 4. Critical Mass of SPERT-D Fuel Elements Moderated and Reflected by Dilute  $\text{U}(92.6)\text{O}_2(\text{NO}_3)_2$  Aqueous Solution Containing 3.99 g of  $^{235}\text{U}$  Per Liter and Various Boron Concentrations. Spacing between fuel element boxes = 0.5 in.

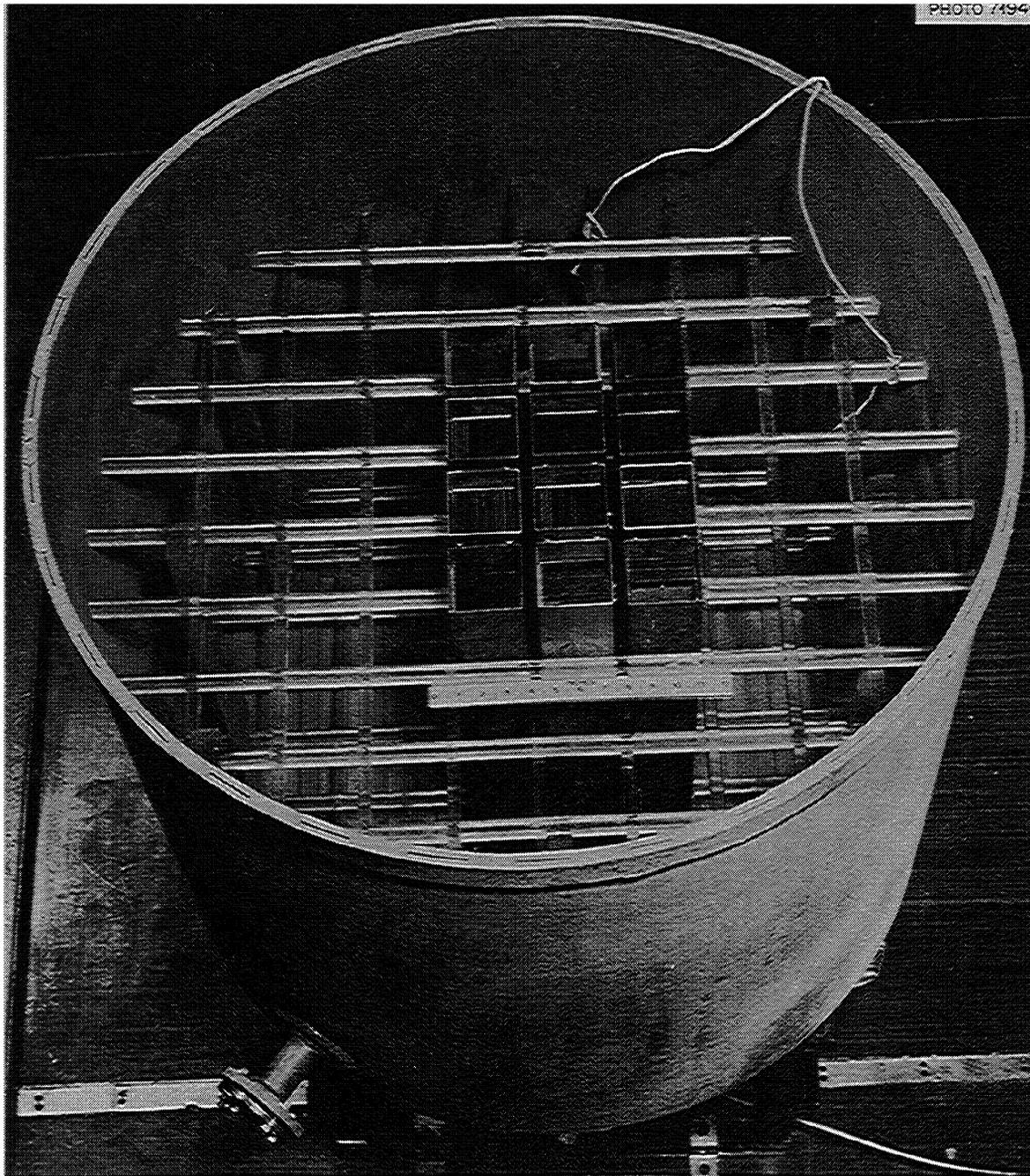


Fig. 5. Critical Lattice of SPERT-D Fuel Elements When Moderated and Reflected by Dilute Uranyl Nitrate Solution. The aqueous  $U(92.6)O_2(NO_3)_2$  solution contained 3.99 g of  $^{235}U$  per liter.

which was critical with a moderator and reflector of dilute  $U(92.6)O_2-(NO_3)_2$  solution containing 3.99 g of  $^{235}U$  per liter, and Fig. 6 shows the corresponding lattice when 1.118 g of boron had been added to each liter of the dilute uranyl nitrate solution.

Because neutron-sensitive counters and a neutron source may be located on the periphery of the actual dissolver to monitor possible inadvertent approaches to criticality, an attempt was made to determine the source neutron multiplication curve for the lattice shown in Fig. 6. Figure 7 shows the reciprocal source neutron multiplication determined by each of three counters, located as shown in the inset, as a function of the amount of  $^{235}U$  in the lattice. It is not surprising that the data from all of the counters were not truly representative of the neutron multiplication by the lattice because of averse geometric effects, nor that the counter located adjacent to the neutron source was the most insensitive to lattice multiplication.

#### SUMMARY AND CONCLUSIONS

Since only a limited number of experiments utilized elements with a nominal 6-foot fuel length, the following comments apply only to lattices of elements having the normal 2-foot fuel length and, therefore, only to lattices no more than 2 feet in the direction of the length of fuel in the elements.

The maximum reactivity of a lattice of 300-g SPERT-D fuel elements occurs at a spacing between the outside surfaces of the boxes equal to about 0.5 in. while the maximum reactivity of 168-g ORR fuel elements occurs at a spacing of 0.25 in. At a spacing greater than 2 in. between outside surfaces more than 50 SPERT-D elements are required for criticality. Any slab two elements thick will be subcritical. An 8 x 8-element lattice spaced 0.5 in. in both directions is subcritical with cadmium sheet between adjacent rows; the presence of the cadmium more than doubles the critical lattice dimensions at this spacing.

The use of aqueous uranyl nitrate solution at a concentration of 3.99 g of  $^{235}U$  per liter as the moderator and the reflector decreased the critical mass of elements by more than the quantity of uranium added to each element by the solution. The addition of 1.118 g of boron to each liter of the dilute aqueous uranyl nitrate solution increased the critical number of elements by a factor of about 3.5 over that with water moderator and reflector and by a factor of about 5 over that with no boron in the uranyl nitrate solution.

The source neutron multiplication curves, observed with the source and the detectors located on the periphery of the cylindrical container of the submerged elements, extrapolate to apparent critical masses far in excess of the true ones so long as the loading is less than about 80% of the critical loading.

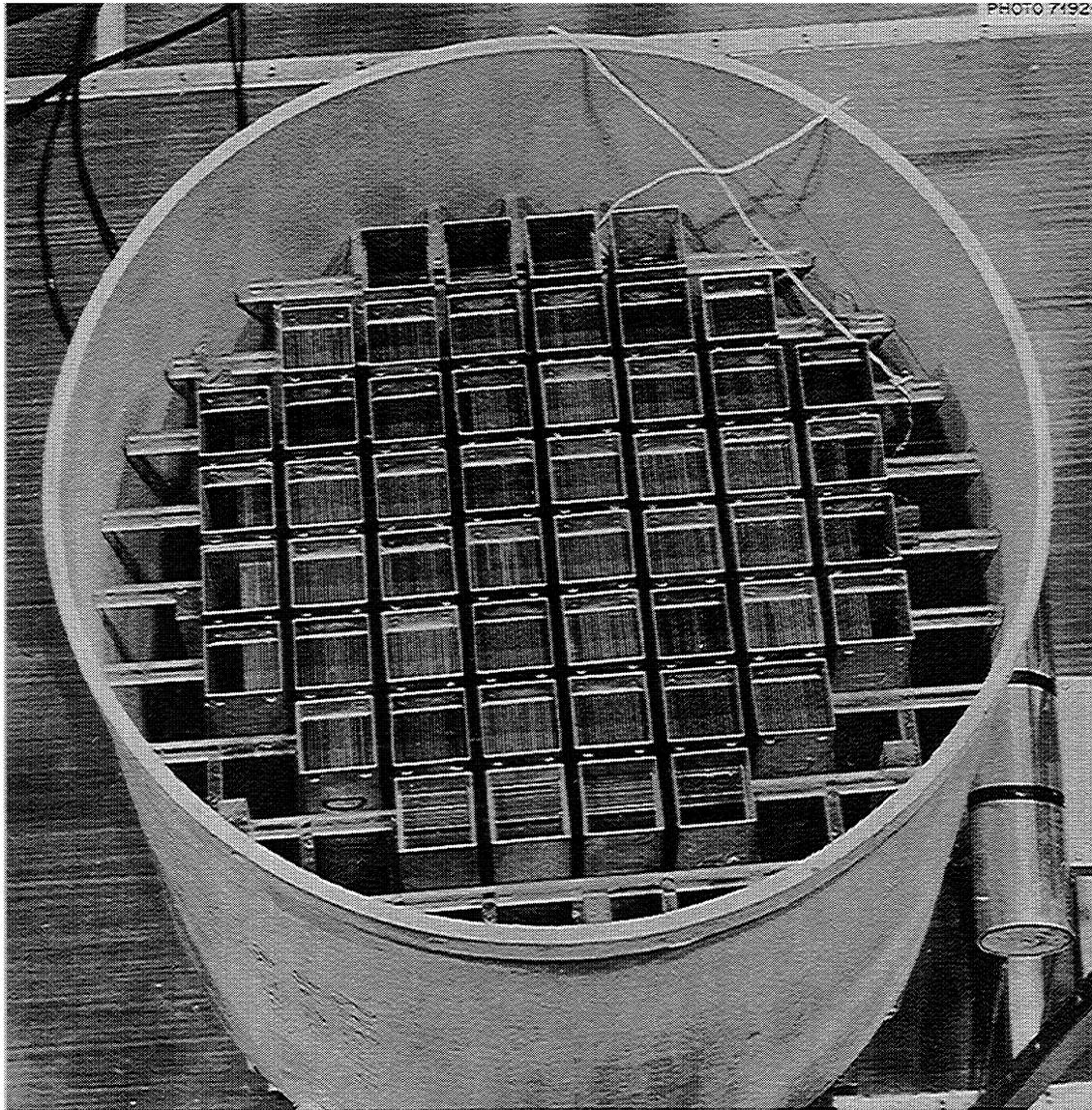


Fig. 6. Critical Lattice of SPERT-D Fuel Elements When Moderated and Reflected by Borated Dilute Uranyl Nitrate Solution. The aqueous borated  $U(92.6)O_2(NO_3)_2$  solution contained 3.99 g of  $^{235}U$  and 1.118 g of boron per liter.

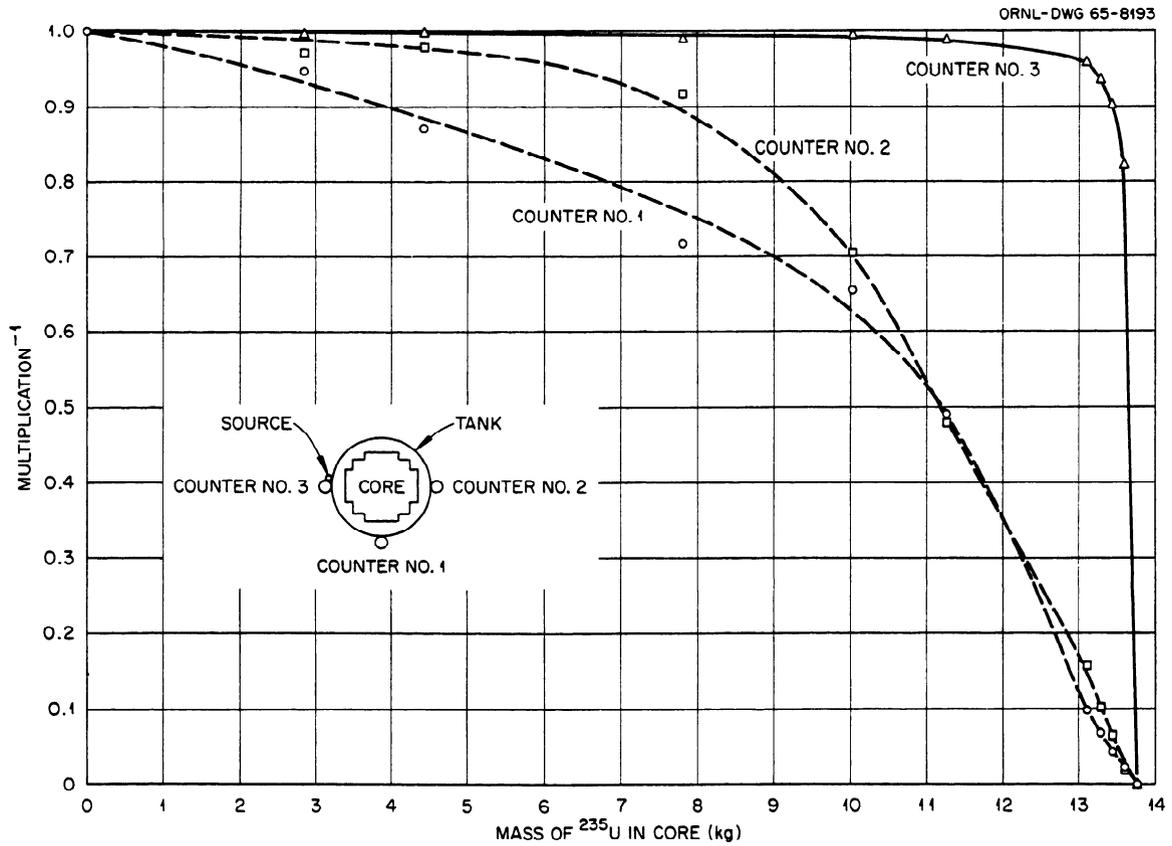


Fig. 7. Source Neutron Multiplication Curves for a Lattice of SPERT-D Fuel Elements in an Aqueous Solution Containing 3.99 g of  $^{235}\text{U}$  per Liter and 1.118 g of Boron per Liter.

## ACKNOWLEDGEMENTS

The authors would like to express appreciation for the assistance of J. K. Fox of Phillips Petroleum Company, Idaho Falls, for guidance in the scope of the experiments, of E. R. Rohrer for instrumentation, and of W. C. Tunnell for special mechanical equipment.