REFERENCE 134

R. C. LLOYD, B. M. DURST, AND E. D. CLAYTON, "EFFECT OF SOLUBLE NEUTRON ABSORBERS ON THE CRITICALITY OF LOW-URANIUM-235-ENRICHED UO_2 LATTICES," NUCL. SCI. ENG. 71: 164-169 (1979).

NUCLEAR SCIENCE AND ENGINEERING®

(ISSN: 0029-5639)

THE RESEARCH JOURNAL OF THE AMERICAN NUCLEAR SOCIETY

Indexed in "Engineering Index" and
Abstracted in "INIS Atom Index"

VOLUME 71, NUMBER 2, AUGUST 1979
NSENAO 71 (2) 87-236 (1979) ISSN: 0029-5639

NUCLEAR SCIENCE AND ENGINEERING (ISSN: 0029-5639) is published monthly by the American Nuclear Society, Inc., with business offices at 555 N. Kensington Ave., La Grange Park, Illinois 60525; telephone 312/352-6611. The subscription rate is $63 per volume or $226 for four volumes per calendar year; overseas subscribers add $36 per year for postage and handling. Single copy price is $23 for current and back issues. Microfiche subscriptions are also available at the same rates. Inquiries about the distribution and delivery of NUCLEAR SCIENCE AND ENGINEERING and requests for changes of address should be directed to the American Nuclear Society. Allow 6 weeks for a change to become effective. NUCLEAR SCIENCE AND ENGINEERING is printed in Danville, Illinois 61832, by Interstate Printers and Publishers, Inc. Second-class postage is paid at La Grange Park, Illinois, and at additional mailing offices. Address all manuscript and editorial communications to the editorial offices, NUCLEAR SCIENCE AND ENGINEERING, Building 9204-1, P. O. Box Y, Oak Ridge, Tennessee 37830. European authors may send communications to Karl Wirtz, Karlsruhe, Germany.

NUCLEAR SCIENCE AND ENGINEERING: 71, 164-169 (1979)

Effect of Soluble Neutron Absorbers on the Criticality of Low-Uranium-235-Enriched UO_2 Lattices

R. C. Lloyd, B. M. Durst, and E. D. Clayton

Battelle-Pacific Northwest Laboratories
P.O. Box 999, Richland, Washington 99352

Received January 22, 1979
Accepted March 28, 1979

A series of experiments was performed to determine the effect of soluble neutron absorbers on the criticality of water-moderated latticed assemblies. The soluble absorbers used were boron, gadolinium, and cadmium. These materials were mixed with the moderator and reflector of lattices of 4.3-wt%-^{235}U-enriched UO_2 fuel pins with stainless-steel cladding. Lattice spacings of 22.9, 27.9, and 33.0 mm, center-to-center, were measured to determine the critical number of rods. The value of k_{eff} was computed for each assembly on which criticality was determined. The computed k_{eff} value were up to ~2% above unity in some cases, with the average value for the entire series being 1.007.

INTRODUCTION

Experimental criticality data are needed on soluble neutron absorbers (poisons) to accurately determine their effectiveness for criticality prevention and control of lattice assemblies.[1-3] This paper summarizes the results of a series of experiments performed at the Battelle-Pacific Northwest Laboratories' Critical Mass Laboratory on water-flooded uranium rod lattices poisoned with soluble neutron absorbers. The poisons utilized—boron, cadmium, and gadolinium—were chosen because of their large neutron absorption cross sections. The experiments were oriented toward providing data on the effectiveness of these materials as soluble neutron absorbers for use in nuclear criticality prevention to serve as benchmarks for validating calculational techniques.

[1]R. C. LLOYD, E. D. CLAYTON, and L. E. HANSEN, *Nucl. Sci. Eng.*, **48**, 300 (1972).

[2]R. C. LLOYD and E. D. CLAYTON, *Nucl. Sci. Eng.*, **59**, 21 (1976).

[3]R. C. LLOYD and E. D. CLAYTON, *Nucl. Sci. Eng.*, **62**, 726 (1977).

EXPERIMENTS AND DATA

The experiments on lattices of fuel rods immersed in water include measurements at each of three different lattices spacings with soluble neutron absorbers. Figure 1 shows a typical loading of the lattice assembly. The center-to-center fuel rod spacings were 22.9, 27.9, and 33.0 mm in triangular arrangement. The UO_2 pellets of 4.3-wt% ^{235}U enrichment were housed in stainless-steel tubes with an outer diameter of 14.4 mm (cladding thickness of 0.813 mm). A detailed description of the fuel rods is given in Fig. 2.

The fuel rods were positioned in the lattice template assembly, as shown in Fig. 3. The top lattice plate was made of a 6.35-mm-thick polycarbonate (Lexan) plate, having a density of 1.2 g/cm^3. The bottom lattice plate was composed of a 15.9-mm-thick Type 304L stainless-steel plate drilled completely through with a 6.35-mm-thick backing plate made of aluminum (Type 6061). The thickness of the bottom lattice plate was selected so that the bottom of the fuel region would nearly coincide with the top of the steel lattice plate. The top and bottom

0029-5639/79/0008-0164$02.00/0

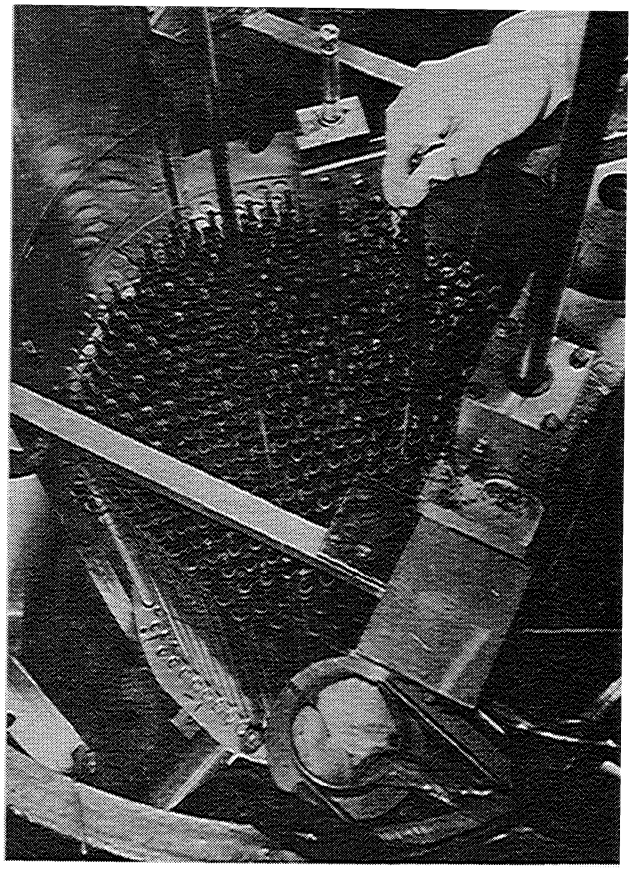


Fig. 1. Lattice assembly.

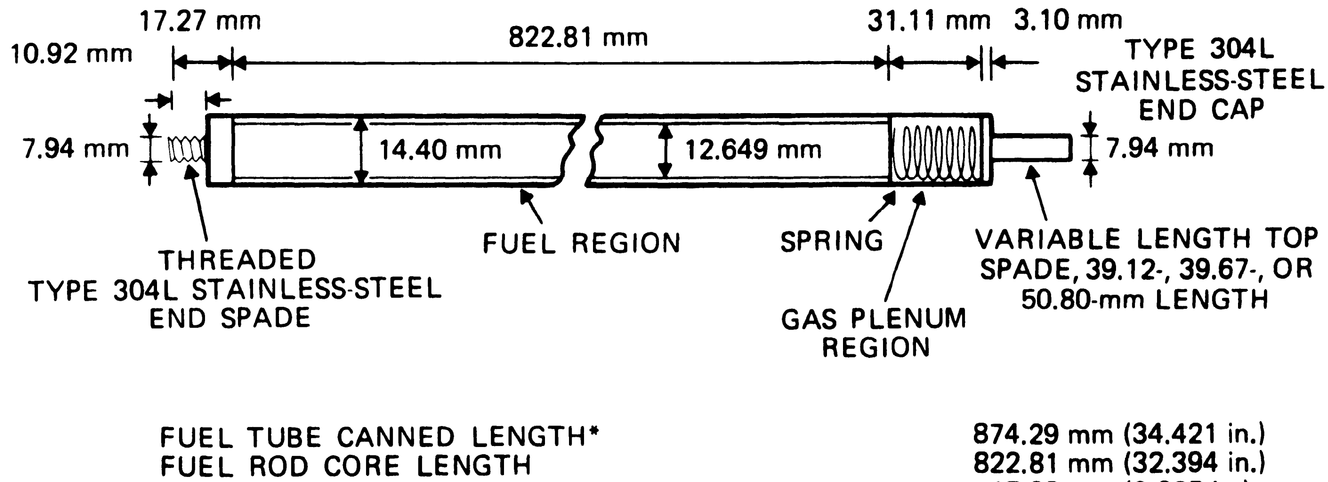
lattice plates were spaced 838 mm by three threaded spacer rods composed of Type 6061 aluminum, 25.4 mm in diameter, positioned outside the fuel loading. The fuel rod assemblies were reflected on all sides by at least 150 mm of water.

After each series of experiments with a neutron absorber, the rods were rinsed with water, wiped dry, washed in detergent, rinsed, and wiped dry to reduce any residual neutron absorber remaining on the rods. This effect was double-checked in one case after the use of boron. Criticality of the rods in water was remeasured and found to agree exactly with the earlier measurements.

The critical number of fuel rods, N_c, was determined at various concentrations of gadolinium, cadmium, and boron in the water moderator with the 22.9-mm lattice and with gadolinium and boron in the 27.9- and 33.0-mm lattices. Soluble neutron absorbers were not intermixed in any of the experiments. The data from these experiments are summarized in Figs. 4, 5, and 6.

Analysis of the data for the 22.9-mm lattice spacing shows that the critical number of fuel pins increased from ~223 in water without a neutron absorber to ~422 in water containing 0.21 g Gd/ℓ. The critical number was 392 rods for 1.1 g Cd/ℓ in water and 433 rods for ~0.6 g B/ℓ in water. Figure 4 is a graph of the critical number plotted against the neutron absorber concentration for the 22.9-mm lattice.

For the 27.9-mm lattice, the critical number of



FUEL TUBE CANNED LENGTH*	874.29 mm (34.421 in.)
FUEL ROD CORE LENGTH	822.81 mm (32.394 in.)
FUEL PELLET LENGTH	15.88 mm (0.625 in.)
CLADDING o.d.	14.40 mm (0.567 in.)
CLADDING THICKNESS (TYPE 304L STAINLESS STEEL)	0.813 mm (0.032 in.)
FUEL COLUMN DIAMETER	12.649 mm (0.498 in.)
UO_2 DENSITY (% THEORETICAL)	94.9 ± 0.6
ENRICHMENT	4.29 wt% ^{235}U

*MEASURED FROM BOTTOM OF END SPADE TO BOTTOM OF TOP SPADE.

Fig. 2. Description of 4.3-wt%-^{235}U-enriched fuel rods.

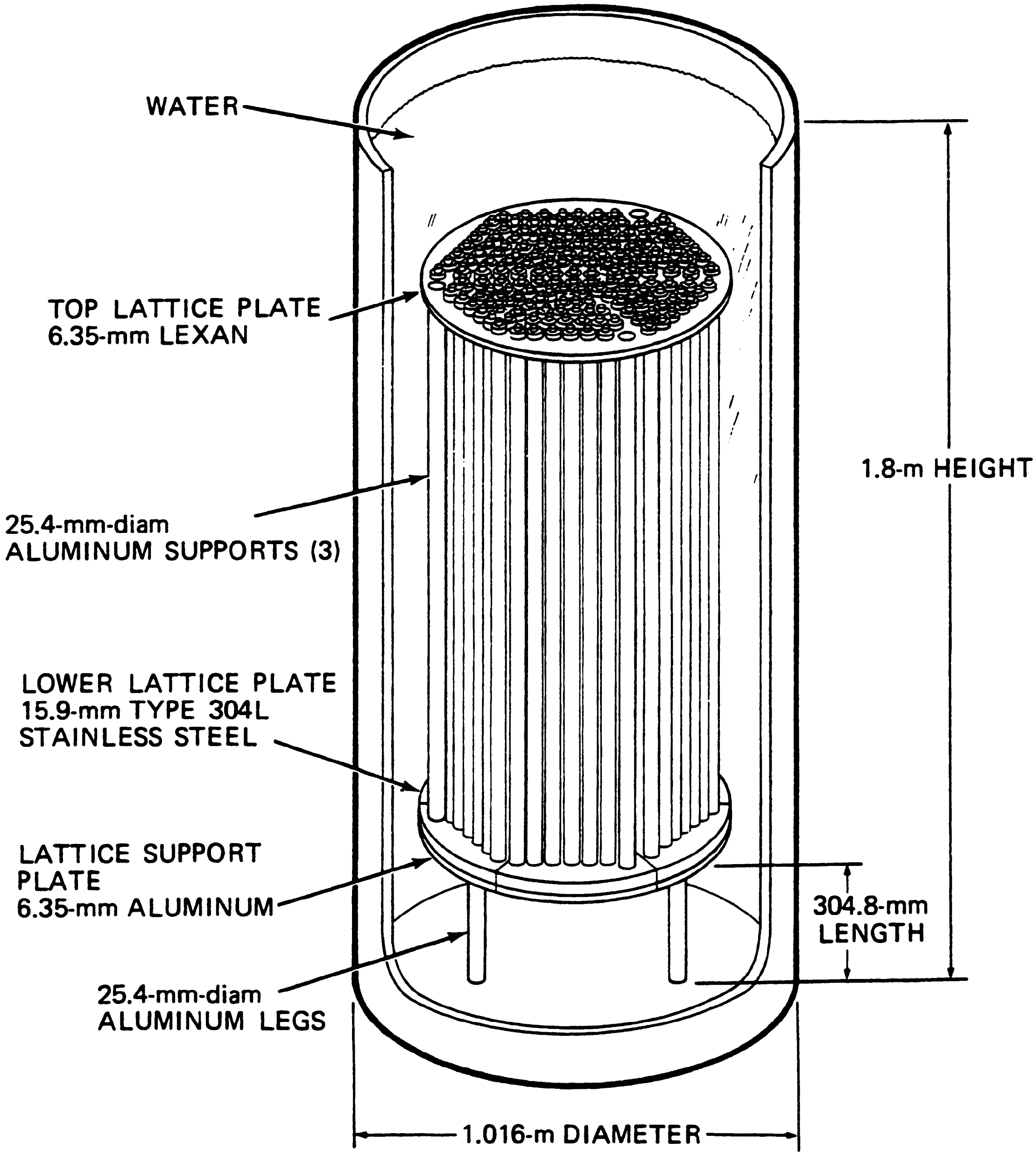


Fig. 3. Schematic diagram of the lattice assembly.

fuel pins varied from ~167 in water without a neutron absorber to ~342 in water containing 0.12 g Gd/ℓ and 350 in water containing 0.38 g B/ℓ. Figure 5 is a graph of the critical number of rods plotted against neutron absorber concentration for the 27.9-mm lattice.

The critical number for the 33.0-mm lattice varied from 187 in water to ~317 in water containing 0.044 g Gd/ℓ, and 434 in water containing 0.22 g B/ℓ. Figure 6 gives the critical number of rods plotted against the neutron absorber concentration for the 33.0-mm lattice.

It is apparent from these data that gadolinium is the most effective neutron absorber per unit weight of poison in solution, with boron and cadmium following in order of decreasing effectiveness. This was expected as the thermal-neutron absorption cross section of absorber follows similar suit.

The critical number of rods has been plotted versus lattice spacing in Figs. 7 and 8 for various concentrations of soluble neutron absorbers in the moderator and reflector of the lattice assemblies. The soluble neutron absorbers are more effective at the larger lattice spacings, which have better moderation

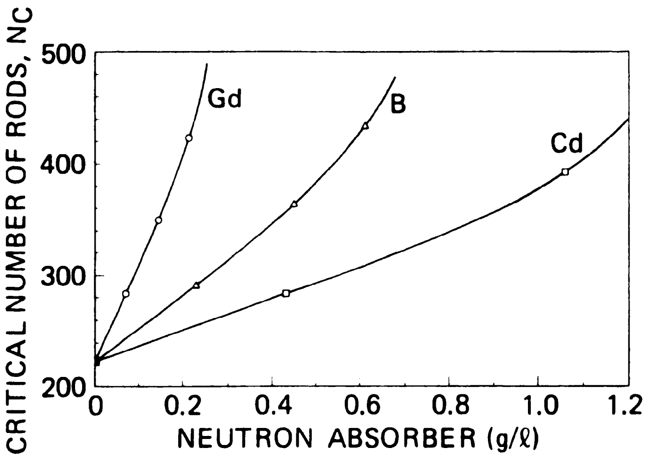


Fig. 4. Effect of neutron absorber on 22.9-mm (0.9-in.) lattice.

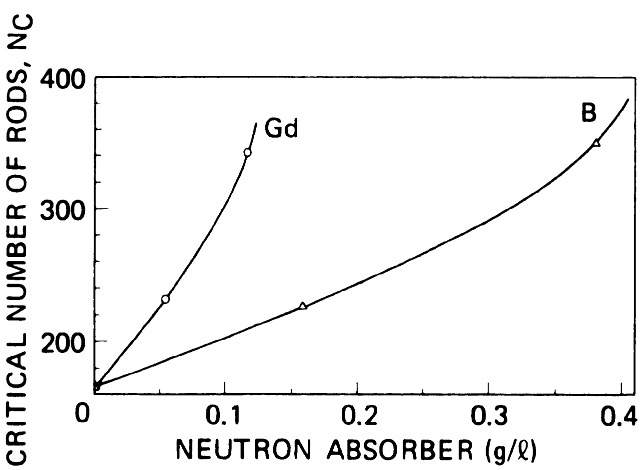


Fig. 5. Effect of neutron absorber on 27.9-mm (1.1-in.) lattice.

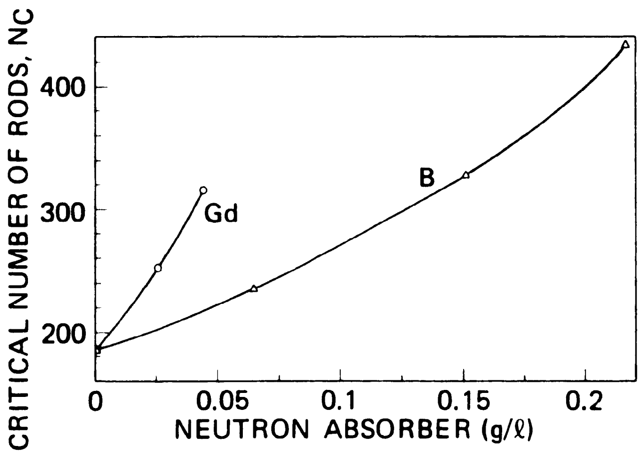


Fig. 6. Effect of neutron absorber on 33-mm (1.3-in.) lattice.

(larger water-to-uranium volume ratios). Also, for a fixed concentration of neutron absorber, the lattices with larger water-to-uranium volume ratios will simultaneously contain more neutron absorber per uranium rod, i.e., the neutron absorber-to-uranium ratio will be higher since the total amount of neutron absorber in the lattice is the product of water volume X

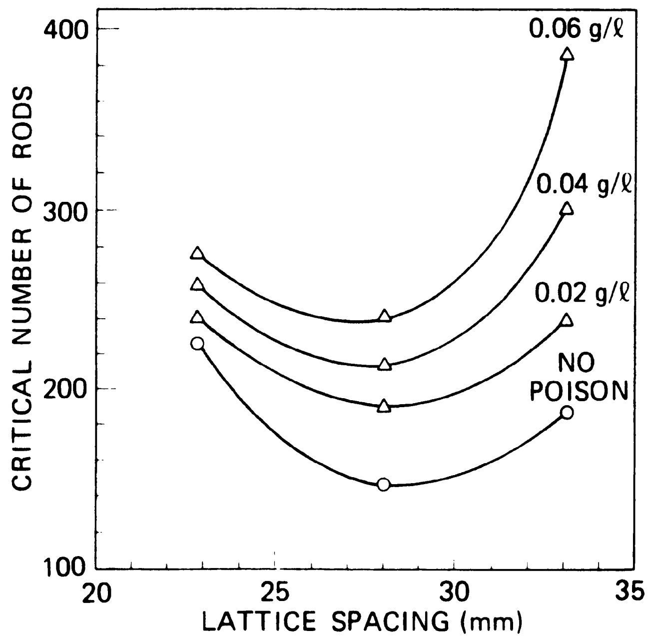


Fig. 7. Effect of gadolinium on the critical number of rods.

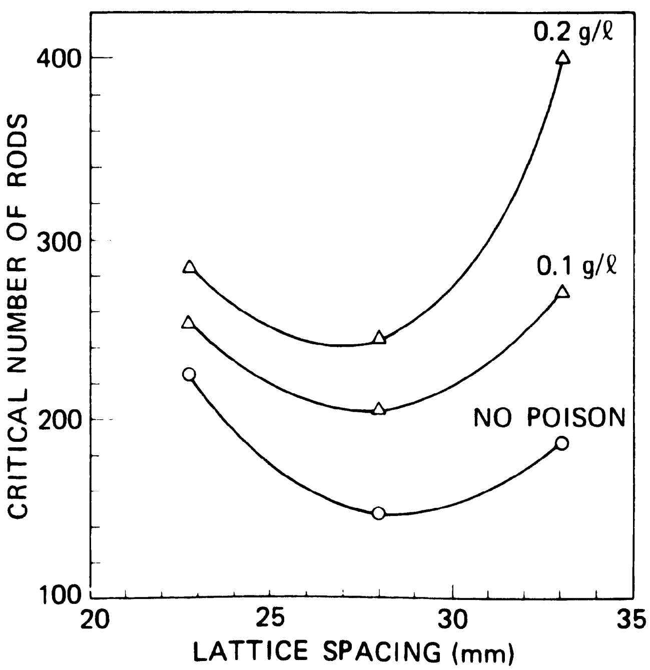


Fig. 8. Effect of boron on the critical number of rods.

TABLE I

Effect of Neutron Absorber on Criticality of Heterogeneous Lattice Assembly

Experiment Number	Triangular Lattice Pitch (mm)	Number of Fuel Rods Required for Criticality[a]	Neutron Absorber[b] Concentration (g/ℓ)	Total NO_3 (g/ℓ)	Excess Acid Molarity M	k_{eff}[c,d]
001	22.86	223.3	0	---	---	1.010 ± 0.008
002	22.86	289.8	0.2307 B	---	---	1.003 ± 0.007
003	22.86	363.0	0.4514 B	---	---	0.992 ± 0.006
004	22.86	433.2	0.6053 B	---	---	1.003 ± 0.006
006	22.86	282.6	0.4293 Cd	0.722	0.004	1.013 ± 0.006[e]
007	22.86	391.7	1.060 Cd	1.418	0.004	1.021 ± 0.005[e]
009	22.86	283.4	0.0722 Gd	0.230	0.0023	1.005 ± 0.007
010	22.86	349.5	0.145 Gd	0.398	0.0036	0.995 ± 0.008
011	22.86	422.4	0.213 Gd	0.527	0.0044	1.016 ± 0.006
012	27.94	166.6	0	---	---	0.997 ± 0.005
013	27.94	225.6	0.158 B	---	---	1.006 ± 0.006
014	27.94	349.9	0.380 B	---	---	1.005 ± 0.006
016	27.94	232.3	0.0547 Gd	0.1927	0.0021	0.994 ± 0.008
017	27.94	341.8	0.1169 Gd	0.2707	0.0021	1.006 ± 0.006
018	33.02	187.4	0	---	---	1.024 ± 0.005
019	33.02	234.0	0.0643 B	---	---	1.012 ± 0.005
020	33.02	434.0	0.2154 B	---	---	0.992 ± 0.005
021	33.02	327.5	0.1507 B	---	---	1.013 ± 0.006
023	33.02	252.7	0.0257 Gd	0.1571	0.00204	1.009 ± 0.007
024	33.02	316.8	0.0440 Gd	0.1805	0.00207	1.016 ± 0.007

[a]Experiment error in number of rods is ≤0.3%.

[b]Boron in the form H_3BO_3, cadmium in the form $Cd(NO_3)_2 \cdot 4H_2O$, and gadolinium in the form $Gd(NO_3)_3$.

[c]Calculated using EGGNIT/ENDF/B-IV and THERMOS/ENDF/B-III.

[d]The bottom fuel rod end spade was modeled as part of the Type 304L stainless-steel support plate, and top spades were modeled as homogenized Type 304L stainless steel in the water region above the fuel. The top lattice plate, made of Lexan, was considered to be water equivalent.

[e]Calculated using EGGNIT/ENDF/B-IV and TEMPEST/ENDF/B-IV.

neutron absorber concentration. The effectiveness of these chosen neutron absorbers in the more thermal systems is not surprising since their absorption cross section is highest in the thermal or near-thermal region.

When added in the form of H_3BO_3, $Cd(NO_3)_2 \cdot 4H_2O$, and $Gd(NO_3)_3$, boron, cadmium, and gadolinium are all very soluble in either water or slightly acid solutions. However, it was observed that in a slightly basic solution (i.e., water having a pH > 7), cadmium precipitated, probably in the form $Cd(OH)_2$. Upon the addition of sufficient acid to correct the pH, the cadmium went into solution.

A similar behavior was not observed with gadolinium, as the water solutions were always slightly acidic. The gadolinium, in the oxide form Gd_2O_3, was dissolved in sufficient nitric acid before being added to the water solutions. As can be seen from these figures, the concentrations of neutron absorbers in water solution never exceeded a few grams per litre and hence were well within solubility limits for these compounds.

CORRELATION OF CALCULATIONS WITH EXPERIMENTS

For each of these critical experiment determinations, k_{eff} values were computed to check how well standard calculational techniques agree with experimental results. The k_{eff} values were calculated using the KENO IV (Ref. 4) computer code. The calculations were made using 17 epithermal broad-group cross sections. These data were generated by the EGGNIT code[5] using ENDF/B-IV data processed by

[4]G. E. WHITESIDES and N. F. CROSS, "KENO–A Multigroup Monte Carlo Criticality Program," CTC-5, Oak Ridge Computing Technology Center (1969).

[5]C. R. RICHEY, "EGGNIT: A Multigroup Cross Section Code," BNWL-1203, Battelle-Pacific Northwest Laboratories (1969).

the FLANGE (Ref. 6) and ETOG (Ref. 7) codes; a single thermal group was generated using the THERMOS code[8] with the ENDF/B-III library. All rod systems were modeled in a "smeared rod" geometry, that is, cell-averaged cross sections were generated for fuel rods in water, and these cross sections in turn were used in modeling the fuel region as a homogeneous cylindrical zone. The 6.35-mm Lexan top lattice plate that supported fuel during the course of the experiments was assumed to be water equivalent, while the bottom 15.9-mm stainless-steel lattice plate was modeled exactly. The results of these calculations, as well as all pertinent experiment data, are summarized in Table I. The computed criticality factors (k_{eff}) were up to ~2% above unity in some cases, with the average value for the entire series being 1.007.

[6]H. C. HONECK and D. R. FINCH, "FLANGE-II (VERSION 71-1): A Code to Process Thermal Neutron Data from an ENDF/B Tape," DP-1278 (ENDF-152), Savannah River Laboratory (1971).

[7]K. E. KUSNER, R. A. DANNELS, and S. KELLMAN, "ETOG-1: A FORTRAN-IV Program to Process Data from the ENDF/B File to the MUFT, GAM, and ANISN Formats," WCAP-3845-1 (ENDF-114), Westinghouse Electric Corporation (1969).

[8]C. L. BENNETT and W. L. PURCELL, "BRT-1: Battelle-Revised-THERMOS," BNWL-1434, Battelle-Pacific Northwest Laboratories (1970).

CONCLUSIONS

Data have been presented herein that can be used in criticality control and in validation of calculational schemes for low-^{235}U-enriched uranium rods in poisoned water solutions. The effectiveness of three such neutron absorbers—boron, cadmium, and gadolinium—has been compared at different lattice pitches, and it was observed on a unit weight basis that gadolinium was the most effective absorber.

The neutron absorbers can be readily added to water systems in the form of H_3BO_3, $Cd(NO_3)_2 \cdot 4H_2O$, and $Gd(NO_3)_3$. However, some care must be taken to ensure that the solution is slightly acidic to prevent precipitation of poison from solutions containing cadmium or gadolinium nitrate.

Calculations were performed to check the agreement of standard calculational techniques with experimental data. The EGGNIT and THERMOS computer codes were used to generate 18 multigroup cross sections for input into the computer code KENO IV. Calculations of these experiments were slightly conservative, being ~1% high in terms of k_{eff}.

ACKNOWLEDGMENT

This work was prepared for the U.S. Department of Energy (formerly the U.S. Energy Research and Development Administration).