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CRITICAL MASS STUDIES - PART XIII

BOROSILICATE GLASS RASCHIG RINGS IN AQUEOUS URANYL NITRATE SOLUTIONS

J. T. Thomas, J. K. Fox* and E. B. Johnson

ABSTRACT

The effect of commercially available borosilicate glass Raschig rings on the criticality of aqueous uranyl nitrate solutions enriched in U^{235} has been experimentally investigated. The natural-boron content of the glass varied from 0.5 to 5.7 weight per cent, and the volume of the vessel occupied by the glass ranged from 20.9 to 30 per cent. Results from exponential experiments, using a critical layer of solution above the column of solution-ring mixture as a neutron source, have provided estimates of the material buckling of the mixture as a function of solution concentration, boron content of the glass, and the glass volume present. It has been shown, for example, that the buckling is negative (i.e., $k_\infty < 1$) if glass containing 4 wt% boron occupies more than 22% of the mixture volume, whereas the same concentration of glass containing only 0.5 wt% of boron results in positive values of the buckling except for solutions more dilute than about 72 g of U per liter ($H:U^{235} \cong 380$).

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INTRODUCTION

The capacity, or safety, of processes with fissile materials can be increased significantly by introducing neutron-absorbing elements into the process stream provided, of course, the absorber is compatible with process materials. Borosilicate glass, long a material of chemical engineering usage, has recently been studied for this application by Bidinger et al.[1] and, in fact, its use is becoming extensive in large capacity storage vessels and in components of process trains. A program of experiments intended to better establish bases for use of these glasses in nuclear safety problems was initiated at the Oak Ridge Critical Experiments Facility several years ago[2] when a few measurements were made with Pyrex glass and uranium solutions at several concentrations in a single container. A more extensive study has now been completed in which Raschig rings, varying both in dimensions and in boron content, were immersed in cylinders of different diameters, containing enriched uranyl nitrate solutions of several concentrations.

It was impossible to make most of the solution-ring mixtures critical and, in fact, the source neutron multiplication was often too low to allow sensible extrapolation. Addition of only a few inches of solution above a very subcritical mixture established a critical system which served as a source of neutrons distributed over the mixture and had spectral characteristics allowing a measure of the neutron relaxation length in the mixture. The material buckling estimated from these exponential-experiment data is somewhat uncertain owing to the unavailability of values of the radial extrapolation distance. It has been possible to show, however, that the material buckling of some of the mixtures studied is negative.

EQUIPMENT AND MATERIALS

The largest vessel employed was a stainless steel cylinder, 48 in. in diameter and 60 in. high, having a 1/8-in.-thick lateral wall and a very shallow, 1/4-in.-thick, dished bottom. A 1/16-in.-thick sheet of stainless steel covered the top of the tank to minimize evaporation during the experiments. Two other vessels used in the experiments were

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1. G. H. Bidinger, C. L. Schuske and D. F. Smith, "Nuclear Safety Experiments on Plutonium and Enriched Uranium Hydrogen Moderated Assemblies Containing Boron," RFP-201 (Oct. 13, 1960).
2. J. K. Fox and L. W. Gilley, Neutron Physics Division Annual Progress Report, ORNL-2842, p. 78 (Nov. 9, 1959).

an aluminum cylinder 30 in. in diameter and 60 in. in height, having a lateral wall thickness of 1/8 in. and a 1/2-in.-thick bottom, and a 20-in.-dia, 60-in.-high stainless steel cylinder having a 1/16-in.-thick lateral wall and a 1/2-in.-thick bottom. An effectively infinite water reflector could be provided on the bottom and lateral surface of each cylinder and in some experiments cadmium sheet, 0.032 in. thick, covered the lateral surfaces of the cylinder.

The fissile material was aqueous uranyl nitrate solution enriched* to 92.6% in U^{235}. The principle impurities present were 600 and 100 ppm of aluminum and iron, respectively. The solution concentration was varied between 415 and 63.3 g of U per liter.

The borosilicate glass Raschig rings were commercial items having properties of interest listed in Table I.

Table I - Description of Borosilicate Glass Raschig Rings

Manufacturer	Type	Size (in.) O. D. x I. D. x Length	Natural Boron Content wt%
Kimble Glass Co.	R-6	1.50 x 1.25 x 1.70	0.5
" " "	KG-33	1.50 x 1.25 x 1.70	4.1
" " "	KG-33	0.625 x 0.43 x 0.625	3.9
" " "	EN-1	1.50 x 1.25 x 1.70	5.7
Corning Glass Co.	Pyrex	1.80 x 1.50 x 1.90	4.0
" " "	Pyrex	1.50 x 1.25 x 1.70	4.0

A BF_3 proportional counter, 0.25 in. O. D. by 1.0 in. long, was used to determine the neutron flux distribution in the exponential experiments.

DESCRIPTION OF EXPERIMENTS

It was apparent from early source neutron multiplication tests that many of the mixtures of solution and glass rings would be subcritical even though the tests were made in volumes as large as 48 in. in both diameter and height and the chemical concentration was near that required for minimum critical volume, at least when unpoisoned. In fact there was strong evidence that an infinite quantity of some of the mixtures could not be made critical. A critical system could be established, however, by raising the level of the solution a few inches above the glass bed, thereby providing the conditions necessary

* This salt of enriched uranium is designated by $U(93)O_2(NO_3)_2$

for exponential experiments. The critical slab was operated at a power sufficient to provide a source of neutrons whose distribution was measured in the adjacent exponential column. This unique coupling of the two systems provided a radially symmetric neutron source with almost matched spectra.

RESULTS

The conditions necessary to sustain criticality during the flux measurements in the cylinders are given in Table II. Random irregularities in the top surface of the ring bed introduced an uncertainty of ±0.3 in. in the glass-solution mixture height and must be considered in any attempt to assign a thickness to the critical slab of solution when reflected on one surface.

Figure 1 shows the flux distribution obtained in the reflected and unreflected 48-in.-dia cylinder arbitrarily normalized to unity at a point 28 in. above the bottom of the column. The deviations of the points from linearity are probably due to the random orientation of the glass rings in the vicinity of the reentrant tube, i.e., the formation of "pockets" of solution. The traverse is representative of those obtained in the other experiments.

The material buckling for cylindrical geometry can be expressed as

$$B_m^2 = \frac{J_o}{(R + \lambda)^2} - \gamma^2$$

where R is the cylinder radius, λ is an appropriate radial extrapolation distance, and γ is the reciprocal neutron relaxation length. Retaining only the fundamental mode leads to the following z-dependent relation for the flux

$$\phi = \phi_o e^{-\gamma z}.$$

Portions of each flux distribution, selected to avoid boundary perturbations, fitted by least squares to the latter relation yields the values of γ listed in Table II.

Setting $\lambda = 0$ gives maximum values of B_m^2 so that conservatism is introduced upon application of these results to safety problems. Imposing this condition on the data for the mixtures of solution at a concentration of 415 g of U per liter and glass with an approximate boron content of 4 wt%, both bare and water reflected, yields the upper two curves of Fig. 2 where B_m^2 is shown as a function of the volume per cent of the mixture occupied by the glass in a 20-in.-dia cylinder. A similar pair of points from a 30-in.-dia cylinder with

Table II. Results from Experiments with Mixtures of Uranyl Nitrate Solution and Borosilicate Glass
U^{235} Enrichment = 92.6 wt%

Diameter of Cylinder (in.)	Reflector[a]	Type of Glass	Boron Content of Glass (wt%)	Glass Content of Mixture (vol %)	Glass Height (in.)	Solution Height (in.)	γ (Relaxation Length)$^{-1}$ ($in.^{-1}$)	Error in γ (95% Confidence Level)
Solution Concentration: 415 g of U per liter ($H:U^{235}$ = 59)								
48	Air	EN-1	5.7	24.1	46.5	44.68[b]	-	-
	H_2O	EN-1	5.7	24.1	46.5	44.68[b]	-	-
	Air	EN-1	5.7	24.1	30.0	34.31	0.189	± 0.022
	H_2O	EN-1	5.7	24.1	30.0	34.25	0.179	± 0.027
30	H_2O	Pyrex	4.0	20.9	50.0	50.0[b]	-	-
	H_2O	Pyrex	4.0	20.9	35.9	39.49	0.092	± 0.016
	Air	Pyrex	4.0	20.9	35.9	39.75	0.118	± 0.015
20	H_2O	EN-1	5.7	24.1	34.6	40.64	0.257	± 0.017
	Air	EN-1	5.7	24.1	34.6	41.03	-	± -
	Air	KG-33	4.1	24.1	35.6	41.32	0.219	± 0.007
	H_2O	KG-33	4.1	24.1	35.6	40.84	0.198	± 0.018
	H_2O	Pyrex	4.0	20.9	33.8	38.53	0.145	± 0.011
	Air	Pyrex	4.0	21.0	33.6	38.67	0.171	± 0.016
	H_2O	Pyrex	4.0	21.0	33.6	38.29	0.142	± 0.017
	H_2O	KG-33	3.9	30.0	27.2	32.37	0.307	± 0.013
	Air	KG-33	3.9	30.0	27.2	32.69	0.322	± 0.012
	Air	R-6	0.5	24.0	21.4	10.86	Note that this mixture was critical	
	H_2O	R-6	0.5	24.0	21.4	8.34		
Solution Concentration: 279 g of U per liter ($H:U^{235}$ = 92)								
20	Air	KG-33	4.1	24.1	35.6	41.48	0.257	± 0.016
	H_2O	KG-33	4.1	24.1	35.6	41.12	0.239	± 0.029
Solution Concentration: 141 g of U per liter ($H:U^{235}$ = 191)								
20	Air	KG-33	4.1	24.1	35.6	41.47	0.312	± 0.022
	H_2O	KG-33	4.1	24.1	35.6	41.05	0.321	± 0.026
	Air	R-6	0.5	24.0	21.0	25.68	0.112	± 0.007
	H_2O	R-6	0.5	24.0	21.0	24.95	0.074	± 0.013
20[c]	Air	R-6	0.5	24.0	21.0	25.63	0.108	± 0.006
	H_2O	R-6	0.5	24.0	21.0	25.29	0.088	± 0.005

Table II. Contd.

Diameter of Cylinder (in.)	Reflector[a]	Type of Glass	Boron Content of Glass (wt%)	Glass Content of Mixture (vol %)	Glass Height (in.)	Solution Height (in.)	γ (Relaxation Length)$^{-1}$ ($in.^{-1}$)	Error in γ (95% Confidence Level)
Solution Concentration 94.4 g of U per liter ($H:U^{235}$ = 290)								
20[c]	Air	R-6	0.5	24.0	21.0	26.86	0.185	± 0.011
	H_2O	R-6	0.5	24.0	21.0	26.51	0.176	± 0.011
Solution Concentration: 63.3 g of U per liter ($H:U^{235}$ = ~~406~~)								
20[c]	Air	R-6	0.5	24.0	21.0	28.23	0.264	± 0.016
	H_2O	R-6	0.5	24.0	21.0	27.78	0.265	± 0.014

a. In instances of water reflection the bottom and lateral surfaces, to approximately the height of the solution, were surrounded by an effectively infinite thickness of water.
b. This assembly was subcritical.
c. 32-mil-thick cadmium sheet around cylinder.

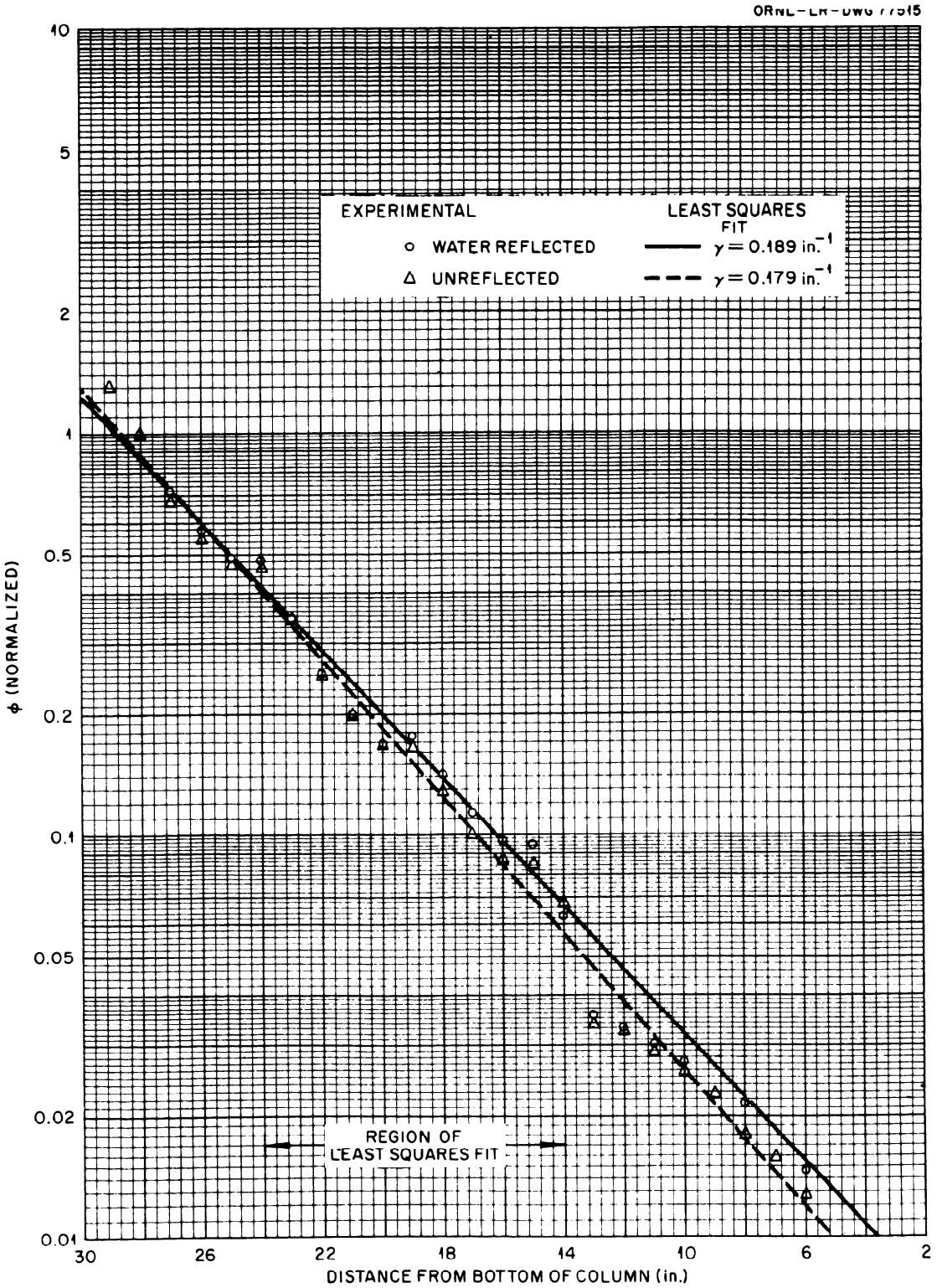

Fig. 1. Neutron Flux Distribution Along Axis of 48-in.-dia Cylinder.

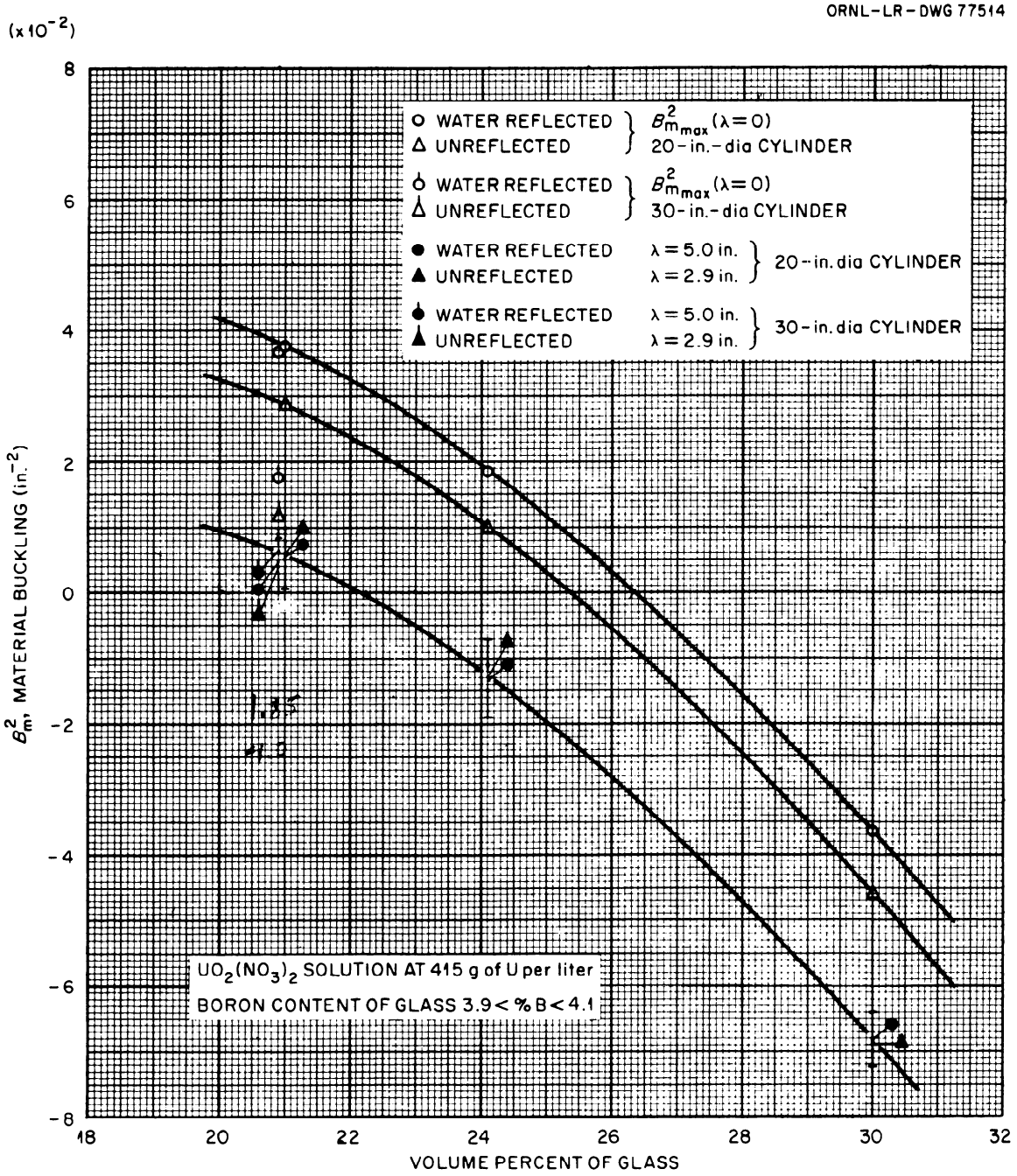

Fig. 2. Material Buckling of Mixtures of $U(93)O_2(NO_3)_2$ Solution and Borosilicate Glass Raschig Rings Containing 4 Weight Percent Boron as a Function of Percent Volume of Glass in the Mixture.

glass occupying 20.9 volume per cent are also shown.

These two curves can be superposed, as they should be since B_m is a property of the material and not of the reflector, by assuming 5.0 and 2.9 in. as the radial extrapolation distance of reflected and unreflected cylinders, respectively. Application of these same extrapolation distances to the data from the 30-in.-dia cylinder gives values of B_m^2 consistent with those from the smaller cylinder. This superposition is shown in the lower curve of Fig. 2.

These values of the extrapolation distance were assumed correct for all mixtures studied containing 24.1 volume per cent glass in the solution of the above concentration although the boron content of the glass varied. The results are plotted in Fig. 3 where the material buckling is shown as a function of the boron content. The buckling of this cylinder without glass, based on an estimated critical height of 6.3 ± 0.1 in., is shown for comparison by the point plotted at 19.9×10^{-2} on the ordinate. Since $B_m^2 = 0$ for an infinite critical system ($k_\infty = 1$), it may be concluded, from Fig. 3, that $k_\infty \leq 1$ for every volume of a mixture of aqueous $U(93)O_2(NO_3)_2$, at a concentration of 415 g of U per liter, and borosilicate glass provided the glass occupies at least 24.1% of the volume, contains a minimum of 3.3 wt% of boron and is uniformly distributed. Similarly, from Fig. 2, $k_\infty \leq 1$ if the glass contains 4 wt% of boron and occupies 22.1% of the volume of the mixture.

The assembly containing type R-6 Raschig rings (0.5 wt% boron) in concentrated solution was the only one which was critical with the solution height equal to or less than the height of the ring bed (see Table II). These experiments and others with subcritical mixtures of these rings and solutions of lower concentration are reported in Fig. 4 where the maximum value of B_m^2 ($\lambda = 0$) is plotted against the solution concentration. The results indicate that any system containing Raschig rings of 0.5 wt% boron and occupying at least 24 volume per cent of the mixture will have a value of $k_\infty < 1$ if the solution concentration is $\leq$ 72 g of U per liter ($H:U^{235} \geq 380$).

ACKNOWLEDGEMENTS

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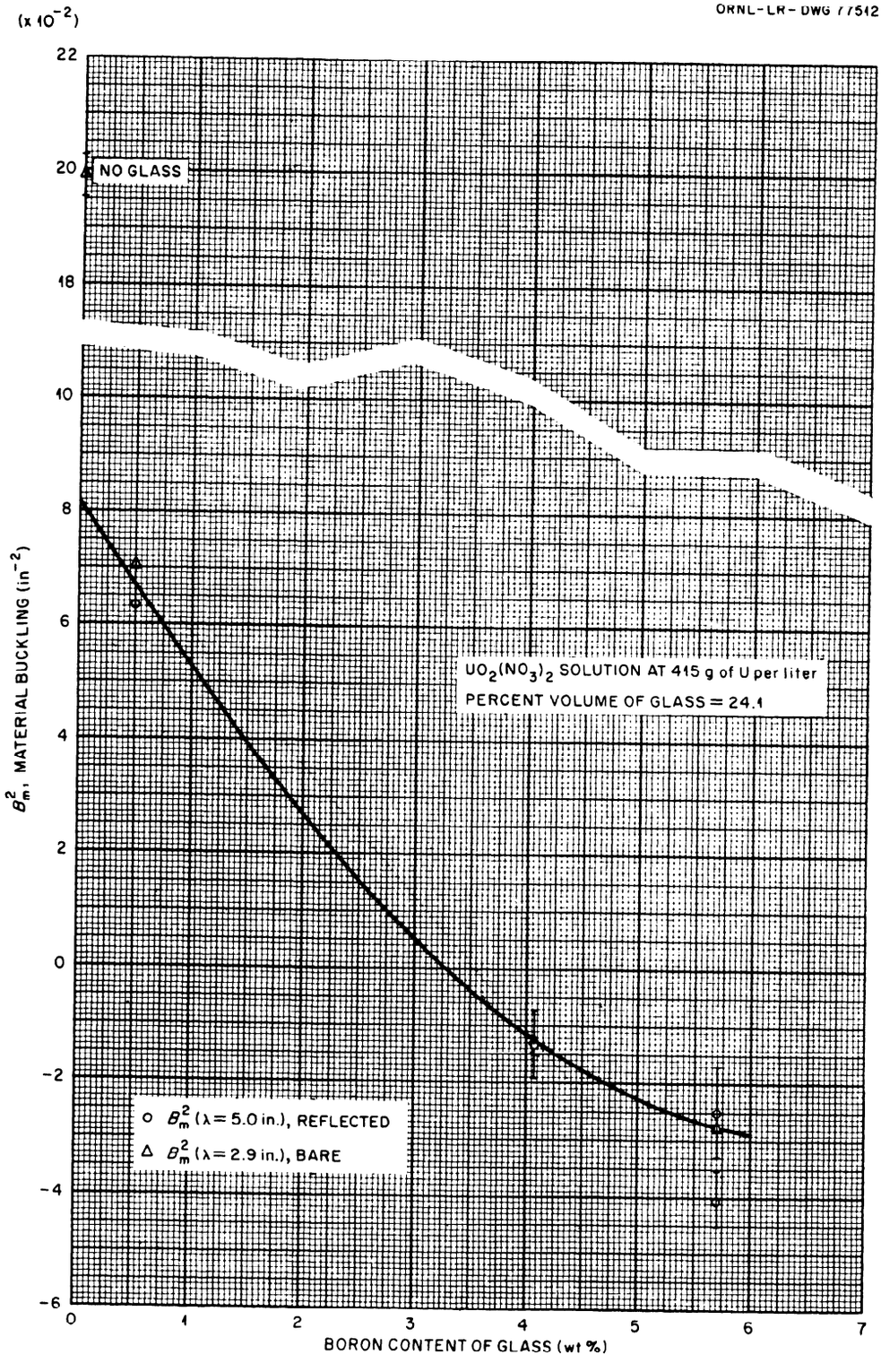

Fig. 3. Material Buckling of Mixtures of $U(93)O_2(NO_3)_2$ Solution and Borosilicate Glass Raschig Rings for a Constant Volume Percent of Glass in the Mixture as a Function of Weight Percent Boron in the Glass.

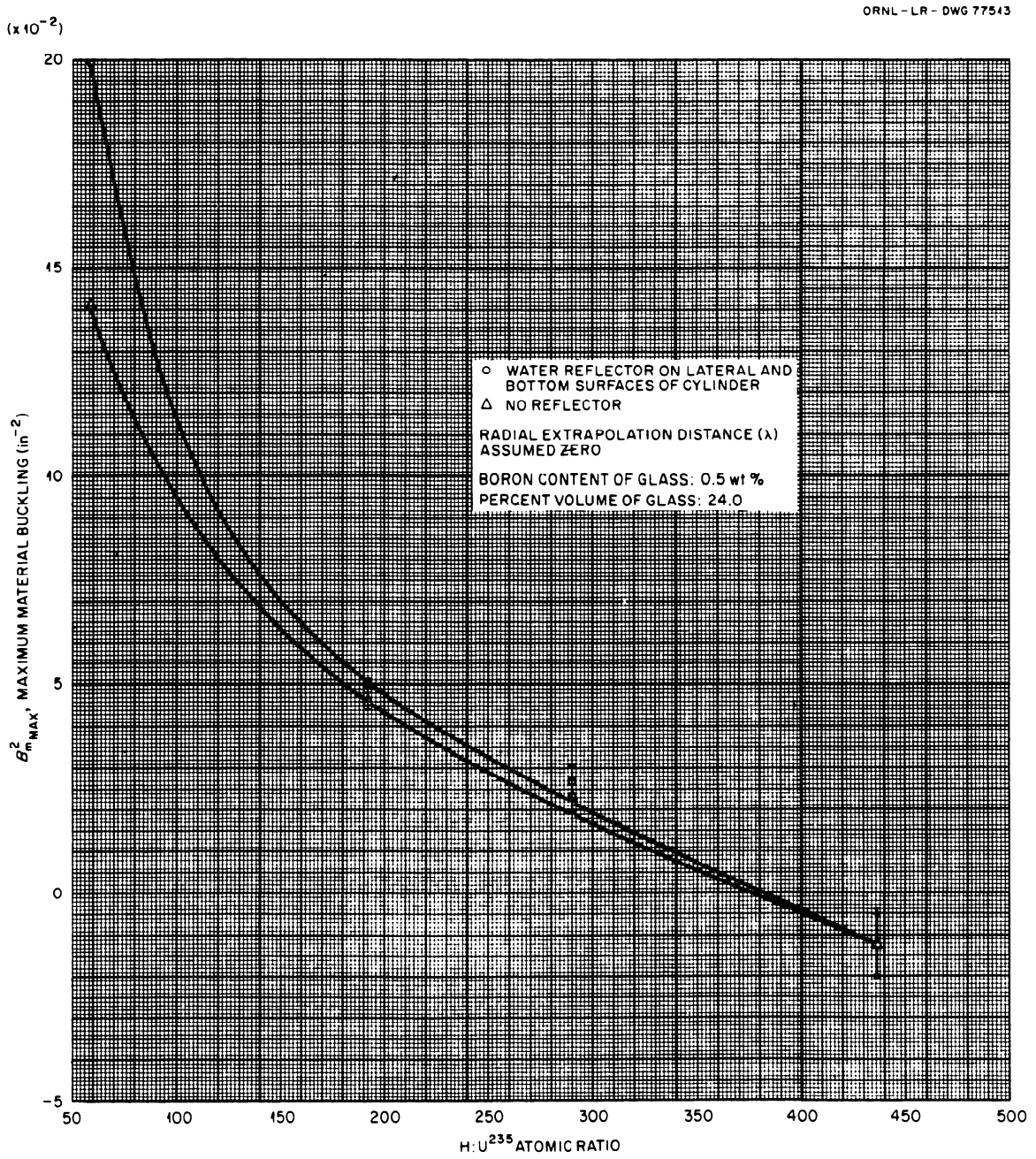

Fig. 4. Maximum Material Buckling ($\lambda = 0$) of Mixtures of $U(93)O_2(NO_3)_2$ Solution and Borosilicate Glass Raschig Rings in a 20-in.-dia Stainless Steel Cylinder.