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USE OF BOROSILICATE GLASS RASCHIG RINGS AS A NEUTRON ABSORBER IN SOLUTIONS OF FISSILE MATERIAL

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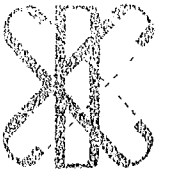

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.95

UC-46, CRITICALITY STUDIES

Y-CDC-8
July 14, 1971

UNION CARBIDE CORPORATION
Nuclear Division

OAK RIDGE Y-12 PLANT

Contract W-7405-eng-26
With the U.S. Atomic Energy Commission

USE OF BOROSILICATE GLASS RASCHIG RINGS AS A NEUTRON ABSORBER IN SOLUTIONS OF FISSILE MATERIAL

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FOREWORD

The Criticality Data Center was established under the auspices of the U.S. Atomic Energy Commission for the development of methods allowing extension and application of data derived from experiments and from analyses to problems in nuclear criticality safety, as well as for the review and evaluation of the data themselves. A necessary part of this program is a medium whereby information germane to the intent of the Center is made available. This report series has been inaugurated for that purpose.

The first five reports were published by and identified with the Oak Ridge National Laboratory. Subsequent reports, however, issued from the Y-12 Plant, are identified by a number sequence including the prefix Y-CDC.

Inquiries should be directed to E. B. Johnson, P. O. Box Y, Oak Ridge, Tennessee 37830.

Reports in This Series:

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Y-CDC-9 Criticality of Liquid Mixtures of Highly ^{235}U-Enriched Uranium Hexafluoride and Hydrofluoric Acid by Robert Caizergues, Edouard Deilgat, Pierre Lécorché, Louis Maubert, and Henri Revol (1971).

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USE OF BOROSILICATE GLASS RASCHIG RINGS AS A NEUTRON ABSORBER IN SOLUTIONS OF FISSILE MATERIAL

J. P. Nichols, C. L. Schuske, and D. W. Magnuson

ABSTRACT

Borosilicate glass raschig rings have had limited use as a neutron absorber in solutions of fissile material for primary criticality control. This report describes the properties of raschig rings in physical, chemical, and radiation environments, the specifications for individual rings and for packed beds of rings, and the calculations and experiments which have resulted in the establishment of recommended limits on the concentration of solutions contained in large vessels packed with the rings. The studies reported here have also resulted in the proposed standard, "Use of Borosilicate Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material."

INTRODUCTION

This report presents background material, references, and supporting data for a proposed standard, "Use of Borosilicate Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material," prepared by Subcommittee 8 of the Standards Committee of the American Nuclear Society. The proposed standard specifies conditions whereby borosilicate glass raschig rings provide primary nuclear criticality control in processes with solutions of fissile materials. The practice of packing vessels with rings for this purpose has been successful for several years within facilities operated under contract with the U. S. Atomic Energy Commission. Dependence on the presence and effectiveness of the rings to prevent criticality in such vessels has been relied upon for fissile uranium and/or plutonium solution storage at the Rocky Flats Plant (RFP) of the Dow Chemical Company[1-5] since 1959 and at the Oak Ridge National Laboratory (ORNL)[6,7] since 1961. This practice was extended at the RFP Critical Mass Laboratory,[8] the ORNL ^{233}U Liquid Storage Facility,[9] and the Nuclear Fuels Services Fuel Processing Plant.[10] Process applications include evaporators having diameters as large as 40 in., storage vessels of 250-gal and greater capacity, and 55-gal transportable liquid carriers. Operating experiences with continuous storage of large quantities of plutonium, ^{233}U, and ^{235}U-enriched uranium solutions extend over significant times.

The concensus of responsible individuals having interest in the development of this operational practice was that specific guidance should be provided only for the use of borosilicate glass raschig rings rather than address the more general subject of all types of solid neutron absorbers. It is believed that the resulting specific proposed standard will be widely utilized in nuclear fuel cycle industries. It will also aid the nuclear safety specialist by calling attention to certain pertinent factors in planning systems of neutron absorbers that are beyond the scope of the proposed standard.

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1. References are listed beginning on page 59.

The requirements of the proposed standard, which are identified by the verb "shall," pertain to those applications in which the glass raschig rings are used for primary criticality control. In these applications, principal or sole dependence is placed on the presence and effectiveness of the rings to prevent a nuclear chain reaction. Much of the information in the proposed standard will be useful, however, in evaluating the adequacy of the presence of raschig rings to prevent a nuclear chain reaction should the improbable failure of another type of primary control occur.

1. EFFECTS OF PROCESS ENVIRONMENT ON GLASS

The borosilicate glass specified in the proposed standard is the low-expansion, corrosion-resistant type that is conventionally used for chemical laboratory glassware. This type of glass contains ~80 wt % SiO_2, 4 wt % Na_2O, 2 wt % Al_2O_3, and 11.8 to 13.8 wt % B_2O_3, and is sold by Corning Glass Works under the trade name of "Pyrex," type 7740, and by Owens-Illinois, Kimble Glass Company, as "Kimax," type KG-33. Other types of borosilicate glasses are marketed. In general, the glasses with more than 14 wt % B_2O_3 are less corrosion resistant.

1.1 Chemical Environment

Low-expansion, corrosion-resistant borosilicate glass has good resistance to corrosion by all acids except HF and hot concentrated H_3PO_4. It is resistant to all concentrations of HCl to 150°C, to all concentrations of H_2SO_4 to about 120°C, and to all concentrations of HNO_3 to the boiling point. It is resistant to acidic salt solutions that do not contain fluoride ions to temperatures of 100°C and greater and to chlorinated hydrocarbons.[11] Typically, the low-expansion, corrosion-resistant glass is attacked to a depth of less than 10^{-6} in. in the first day following exposure to acidic solutions at temperatures less than 100°C.[12]

The good corrosion resistance of the glass depends on its relatively high content of SiO_2 and relatively low content of Na_2O and B_2O_3; good corrosion resistance results from comparatively rapid cooling of the glass after forming to minimize phase separation. The mechanism of chemical attack is a leaching of certain constituents, primarily Na_2O, from the glass. The amount of extract appears to vary with the square root of the time exposure, consistent with a diffusion-controlled process. The amount of extract varies approximately logarithmically with the reciprocal of the absolute temperature.[13]

Low-expansion, corrosion-resistant borosilicate glass is not recommended for exposure to solutions that are basic or contain free fluoride ions. At a temperature of 95°C the depth of attack in one year varies

from 0.01 in. at a pH of 10 to 0.3 in. at a pH of 14. The depth of attack in one year when exposed to 2% NaOH solution varies from 0.01 in. at 60°C to 0.3 in. at 100°C. The rate of attack by 3% (1 M) HF solution varies from about 0.01 in./day at 15°C to about 0.1 in./day at 85°C.[14] The rate of attack by HF solutions does not appreciably decrease with time, as with most other leachants, because the fluoride attacks the SiO_2 base as well as the other phases. In general, the glass is attacked rapidly by concentrated phosphoric acid only if the temperature is greater than 200°C.

Tests of the corrosion resistance of annealed and tempered 1.5-in.-diam borosilicate glass raschig rings in acid and basic solutions have been performed at RFP[15] and ORNL.[16] Table 1 summarizes the weight losses of 10-ring samples that were exposed to 7 N HNO_3 at 95°C for 48 hours. In these tests there was no appreciable difference due to the heat treatment. The weight loss was 0.001 to 0.003% for the two types of low-expansion, corrosion-resistant glass and 0.01 to 0.02% for the borosilicate glass that contained about 19 wt % B_2O_3. This type of nitric acid test is one of the most effective for distinguishing low-expansion, corrosion-resistant borosilicate glass from other types of glass and has been adopted as the chemical acceptance test specified in Section 4.2 of the proposed standard. Of the common commercial glasses, essentially only those which are relatively pure silica have better resistance to corrosion by acids.[12]

Table 2 presents measured weight losses for samples of borosilicate glass raschig rings that were exposed to the vapors from boiling HNO_3 for 48 hours. The weight loss was negligible for the low-expansion, corrosion-resistant glass but was about 0.03% for the glass containing 19 wt % B_2O_3.

The measured weight losses of borosilicate glass rings following exposure to 1.0 N NaOH solution at 95°C for six hours are reported in Table 3. The weight loss was large compared to the acidic exposures and relatively independent of the type of borosilicate glass.

Table 1. Measured Weight Losses of Ten-Ring Samples of Borosilicate Glass Raschig Rings Exposed to 7 N HNO_3 at 95°C for 48 Hours.

Type of Glass[a]	State	Weight of 10 Rings (g) Initial	Weight of 10 Rings (g) Final	Weight Loss (mg)	Weight Loss (%)
A	Annealed	576.232	576.221	11	0.002
	Annealed	576.841	576.829	12	0.002
	Tempered	576.896	576.889	6	0.001
	Tempered	575.550	575.537	13	0.002
B	Annealed	551.362	551.346	16	0.003
	Annealed	549.616	549.605	11	0.002
	Tempered	553.091	553.084	7	0.001
	Tempered	549.961	549.955	6	0.001
C	Annealed	430.740	430.691	49	0.011
	Annealed	438.389	438.316	73	0.017
	Annealed	429.972	429.917	55	0.013
	Annealed	431.085	430.918	67	0.016
	Annealed	442.691	442.617	74	0.017
	Annealed	438.486	438.420	66	0.015
	Tempered	429.245	429.192	53	0.012
	Tempered	430.017	429.936	81	0.019

a. Glass types A and B were commercial low-expansion, corrosion-resistant borosilicate glasses containing ~13 wt % B_2O_3. Glass type C was a commercial borosilicate glass containing ~19 wt % B_2O_3.

Table 2. Measured Weight Losses of Ten-Ring Samples of Borosilicate Glass Raschig Rings Exposed to the Vapors from Boiling 7 N HNO_3 for 48 Hours.

Type of Glass[a]	State	Weight of 10 Rings (g) Initial	Weight of 10 Rings (g) Final	Weight Loss (mg)	Weight Loss (%)
A	Annealed	575.398	575.391	7	0.001
	Annealed	573.664	573.655	9	0.002
B	Annealed	551.624	551.623	1	0.000
	Annealed	549.322	549.322	0	0.000
	Tempered	548.986	548.988	0	0.000
	Tempered	551.466	551.465	1	0.000
C	Annealed	431.916	431.774	142	0.033
	Annealed	429.837	429.709	128	0.030
	Annealed	445.116	444.955	161	0.036
	Annealed	440.422	440.283	139	0.032
	Tempered	427.616	427.597	119	0.028
	Tempered	430.482	430.325	157	0.036

a. Glass types A and B were commercial low-expansion, corrosion-resistant borosilicate glasses containing ~13 wt % B_2O_3. Glass type C was a commercial borosilicate glass containing ~19 wt % B_2O_3.

Table 3. Measured Weight Losses of Ten-Ring Samples of Borosilicate Glass Raschig Rings Exposed to 1.0 N NaOH at 95°C for Six Hours.

Type of Glass[a]	Stage	Weight of 10 Rings (g)		Weight Loss	
		Initial	Final	(mg)	(1%)
A	Annealed	576.688	576.167	521	0.09
	Annealed	576.612	576.094	518	0.09
	Tempered	580.767	579.887	869	0.15
	Tempered	571.132	569.996	1136	0.20
B	Annealed	550.510	549.965	545	0.10
	Annealed	543.305	542.764	641	0.12
	Tempered	538.916	538.597	309	0.06
	Tempered	548.612	548.258	354	0.06
C	Annealed	434.513	433.772	741	0.17
	Annealed	438.998	438.196	802	0.18
	Annealed	442.616	441.875	741	0.17
	Annealed	438.471	437.772	699	0.16
	Tempered	430.562	429.570	592	0.14
	Tempered	430.411	429.738	673	0.16

a. Glass types A and B were commercial low-expansion, corrosion-resistant borosilicate glasses containing ~13 wt % B_2O_3. Glass type C was a commercial borosilicate glass containing ~19 wt % B_2O_3.

Tests made at ORNL[17] compared the corrosion resistance of borosilicate glasses containing 13 and 19 wt % B_2O_3. Samples of annealed rings were exposed to 2 and 6 N HNO_3 at 25 and 65°C for 650 hours. There was no discernible attack or weight change of the low-expansion, corrosion-resistant, 13 wt % B_2O_3 glass in any of the experiments. The rings with the higher B_2O_3 content were noticeably etched and suffered 0.01 to 0.02% weight loss for both acid concentrations at 25°C; at 65°C the etching was severe, the weight loss was about 0.1%, edges were chipped, and internal cracks developed.

1.2 Physical Environment

1.2.1 Mechanical Environment. Highly tempered rings are recommended for those applications in which rings may be agitated. Annealed rings have appreciable structural integrity, however, as is indicated by the following data that were obtained from observations of solution-filled vessels packed with annealed rings that occupied 24% of the volume. The dimensions of the rings were 1.25 in. i.d., 1.50 in. o.d., and 1.70 in. long.

a. Neither breakage nor significant settling of rings was observed in tanks at ORNL that were periodically subjected to air sparge at a rate of 1 scfm per square foot of horizontal projected tank area.

b. At RFP the rings in 55-gal drums used regularly for transport of solution settled 2 to 4 in. in a year because of jarring of the drums.

c. Dropping a simulated ORNL shipping container a distance of 3 ft onto an unyielding surface resulted in breakage of ~20% of the rings and a 5% decrease in the height of the packed rings. Five successive drops from a height of 5 ft resulted in breakage of ~50% of the rings and a 20% decrease in the packed height.

Although the tempered rings are significantly more resistant to breakage, when they do fail they often disintegrate into small particles. At RFP[18] drop tests were performed on 55-gal drums filled with randomly packed tempered rings 1.07 in. i.d., 1.50 in. o.d., and 1.75 in. long. Two successive drops from a height of 6 ft onto a concrete pad broke ~9% of the rings and diminished the packed height by 5%.

A further comparison of the breakage resistance of annealed and tempered rings will be made in Section 2.5 of this report (p. 14).

1.2.2 Radiation Environment. Borosilicate glass is ideally suited for use as a fixed neutron absorber in solutions of ^{235}U, ^{233}U, and ^{239}Pu that are relatively free of beta and gamma radiation. Although there is only limited experience as a basis for an evaluation of the radiation damage incurred when borosilicate glass is exposed to the intense alpha, beta, gamma, and neutron radiation encountered in contaminated solutions of these isotopes, some guidelines can be given.

Beta-Gamma Radiation: Pyrex glass is slightly discolored by exposure to $\sim 10^5$ rads of beta-gamma radiation and is essentially opaque after exposures to 10^9 rad. However, no deleterious mechanical changes, such as loss in strength or leach resistance, are expected[19] below beta-gamma doses of 10^9 to 10^{10} rad.

Mike[20] irradiated Pyrex-type glass with 2 MeV electrons (range 0.2 in.)[a] to doses as great as 3×10^8 rad. The glass was colored an olive brown but there was no measurable change in the flexural strength, resistance to corrosion by water or dilute acid, density, electrical resistivity, or heat of solution.

In an application at ORNL, ^{233}U solution at a concentration of 100 g/liter and contaminated with the daughters of ^{232}U was stored for about four years in a tank packed with Pyrex raschig rings. There was no measurable degradation or leaching of the rings. The approximate exposure of the rings to beta-gamma radiation was 3×10^6 rad at a rate of 100 rad/hr, 0.03 W of alpha radiation per liter, and about 10 rad of neutron radiation (a fluence of 10^{10} n/cm^2).

Borosilicate glass raschig rings are not recommended for use in the "head-end" of a reactor-fuel reprocessing plant where the beta-gamma radiation from typical solutions of fission products would be the order of 10^6 rad/hr, thus reducing the useful life of the rings to between one month and one year. Other suitable methods of criticality control, including use of boron-stainless-steel neutron absorbers, soluble neutron absorbers, favorable geometry, or concentration control, should be considered.

Neutron Radiation: Permissible neutron doses to borosilicate glass are not as well known as the permissible beta-gamma radiation doses. In Rockwell's[21] compilation it is stated that Pyrex is darkened but not appreciably damaged by a thermal neutron fluence less than 3×10^{19} n/cm^2. Bowman[22] reports that Pyrex is not appreciably damaged by a fluence of 5×10^{19} n/cm^2, although $\sim 4\%$ of the boron would be transmuted. Truell[23] has found that a thermal neutron fluence of $\sim 5 \times 10^{15}$ n/cm^2 invariably cracked borosilicate glass with 8 wt % boron presumably because of pressure buildup from the $^{10}B(n,\alpha)^7Li$ reaction.

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[a] The range in glass of 0.05 MeV electrons is about 1 mil.

While the permissible neutron doses are not known precisely, it appears that they are much higher than those normally encountered in a radiochemical plant. There is no appreciable neutron production in solutions containing ^{235}U. Glass will be exposed to neutron dose rates less than 10^{13} n/cm^2-yr ($\sim 10^4$ rad/yr) when immersed in a solution containing ^{233}U, contaminated by 1000 ppm of ^{232}U, at a concentration of 200 g/liter; or in a solution containing ^{239}Pu, contaminated by 2% ^{238}Pu and 12% ^{240}Pu, at a plutonium concentration of 200 g/liter.

Alpha Radiation: Investigators at Lawrence Radiation Laboratory[24] and ORNL[25] found that HNO_3 or LiCl-HCl solutions containing ^{242}Cm at a concentration of about 20 W/liter erode Pyrex vessels at a rate of about 1 mil/day. Tuck[26] found that visible quantities of a siliceous solid are produced in 6 months by 25 ml of radium chloride solution at a concentration of 0.003 W/liter contained in a soda-glass vessel. Subsequent studies showed that Pyrex is attacked at a rate one-tenth less than is soda glass. Baybarz[27] found that solutions of 2 W/liter of ^{241}Am in mixtures of HCl-LiCl eroded Pyrex glass at rates of less than 1 mil/yr.

The effect of alpha radiation is to cause surface damage of the glass since the range of 5 to 6 MeV alpha particles is about 1 mil. The mechanism of erosion is not well understood but it is presumed that the atomic dislocations tend to cause less local chemical durability and that pressure buildup causes exfoliation.[28] Such processes, which are influenced by heat and mass transport, tend to explain the large difference in the erosion rates caused by solutions in which 2 and 20 W/liter, respectively, are produced. The rate of attack probably varies as the square of the alpha particle energy; therefore, a lower rate is expected for the common uranium and plutonium isotopes than for ^{241}Am and ^{242}Cm which emit alpha particles of higher energy.

Surface damage is not expected to cause disastrous failure, even of tempered borosilicate glass rings. Sand blasting tempered rings[15] caused no detectable change in the breaking strength. The primary effect of surface erosion on heavily tempered rings is to shift the location of the neutral tension-compression axis.

Since the energy deposition from alpha-particle absorption in solutions of ^{235}U, ^{233}U, and Pu at the maximum concentrations allowed by the proposed standard is less than 10^{-4}, 0.1, and 2 W/liter, respectively, it appears that alpha-radiation damage can be tolerated provided the rings are inspected periodically. This surmise is corroborated

qualitatively by continuing experiments with rings in concentrated plutonium solutions at RFP. It is known that glasses exhibit the phenomenon of energy storage during irradiation with photons, neutrons, and charged particles. The amount of energy stored is generally less than 1% of the absorbed radiant energy (i.e., less than 2 cal/gram for exposures of 10^8 rads). This phenomenon should be considered as a possible mechanism for sudden release of thermal energy in systems operating near the recommended upper range of radiation exposure.

2. SPECIFICATIONS FOR RASCHIG RINGS

Borosilicate glass raschig rings appropriate to the applications described in the proposed standard will meet the following specifications.

2.1 Composition

2.1.1 ^{10}B Content. The proposed standard specifies that "the boron in the glass shall contain a concentration of ^{10}B isotope such that the ^{10}B-to-^{11}B atomic ratio is not less than 0.240." Recent data indicate that most, if not all, of naturally occurring boron will fall within this specification. Finley et al.[29] determined that the ^{10}B-to-^{11}B ratio in seven mineral samples from various parts of the world varied from 0.2415 to 0.2550. Alt[30] determined that this ratio in a number of U.S. samples was in the range 0.247 ± 0.006. The ^{10}B-to-^{11}B ratios in borosilicate glass rings from different vendors which have been used at ORNL have been in the range of 0.245 ± 0.001.

A method for ^{10}B-to-^{11}B analysis is not specified in the proposed standard because of the differences in analytical capabilities available to various laboratories.

2.1.2 Boron Content. The distribution of the ionic states of boron in borosilicate glass is complex but it is traditional to report the boron content of glass in terms of B_2O_3. The boron content of the glass may be determined by the method of chemical analysis required by the proposed standard or by any other method of comparable accuracy and precision.

2.1.3 Density. The density of low-expansion, corrosion-resistant borosilicate glass is 2.23 ± 0.02 g/cm^3 at 25°C.[31]

2.2 Chemical Acceptance Test

The nitric acid chemical acceptance test, based on experiments of the type described in Table 1, and the above determinations of chemical composition provide reasonable assurance that the rings will have acceptable corrosion resistance.

2.3 Ring Dimensions

Typical dimensions and tolerances of rings that will occupy at least 24 and 32 vol % of the packed volume are as follows:

Dimension	24 to 27 vol %	32 to 35 vol %
Outside diameter (in.)	1.500 + 0.000 - 0.060	1.500 + 0.000 - 0.060
Wall thickness (in.)	0.150 + 0.060 - 0.000	0.210 + 0.060 - 0.000
Length (in.)	1.750 + 0.035 - 0.125	1.750 + 0.035 - 0.125

Both sizes of rings randomly pack to a density of 350 to 400 pieces per cubic foot.

2.4 Surface Finish

It is good practice to require that the rings be fire polished. This helps to prevent personnel injuries in handling, minimizes the generation of fine particles caused by attrition, and heals small cracks.

2.5 Mechanical Shock-Resistance Test

The tumbler illustrated in Fig. 1 has been used to determine the survival of highly tempered and untempered rings as a function of the number of rotations of the tumbler. Test results shown in Fig. 2 indicate that more than 75% of the highly tempered rings survived 10 revolutions without breakage and about 60% would apparently remain unbroken after a very large number of revolutions. On the average, only about 60% of the annealed but untempered rings survived 10 revolutions and about 30% would survive a large number of revolutions.

Mechanical tests of rings in equipment of this type are specified in the proposed standard.

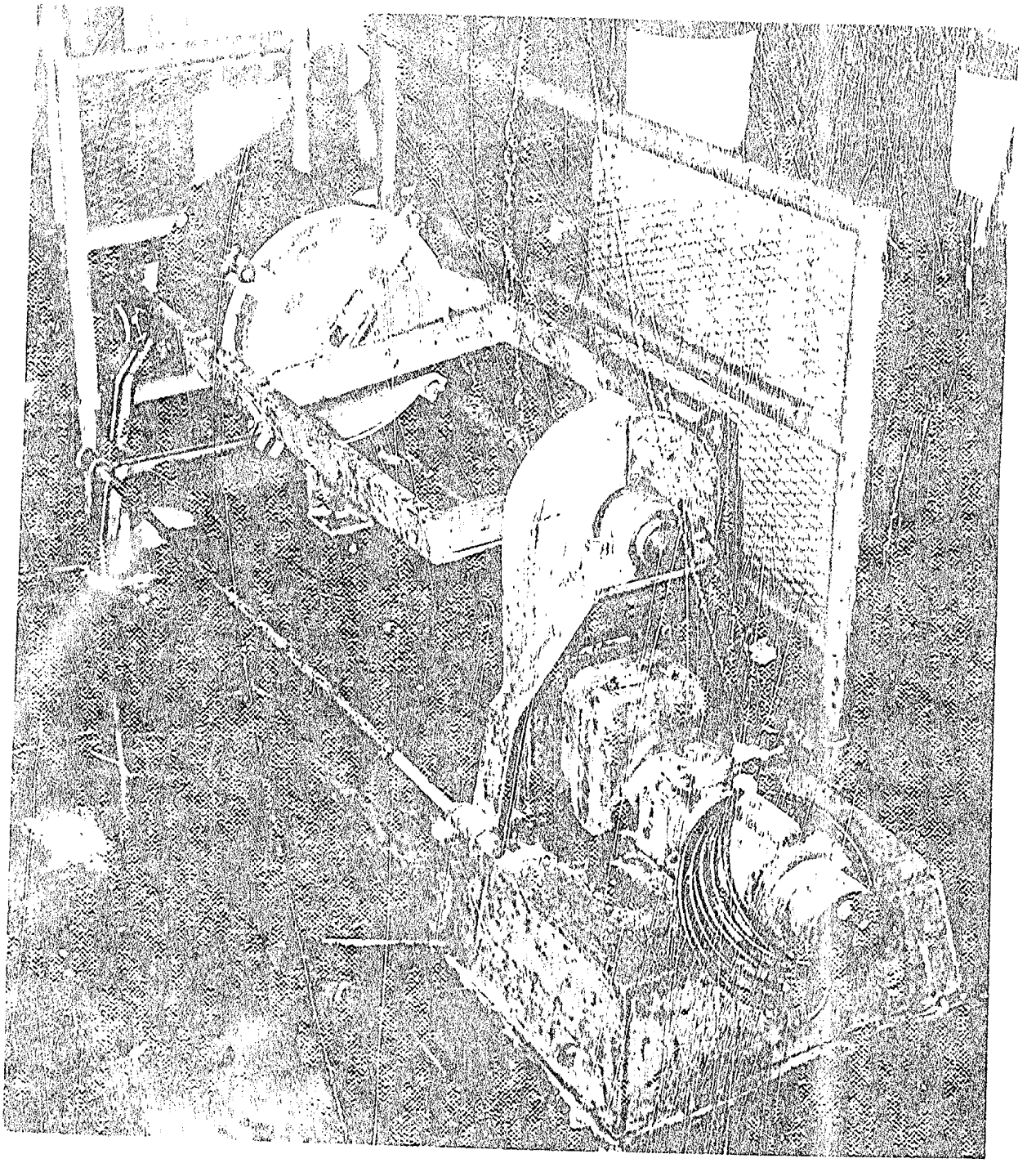

Fig. 1. Rocky Flats Tumbler Drum for Mechanical Shock Resistance Test.

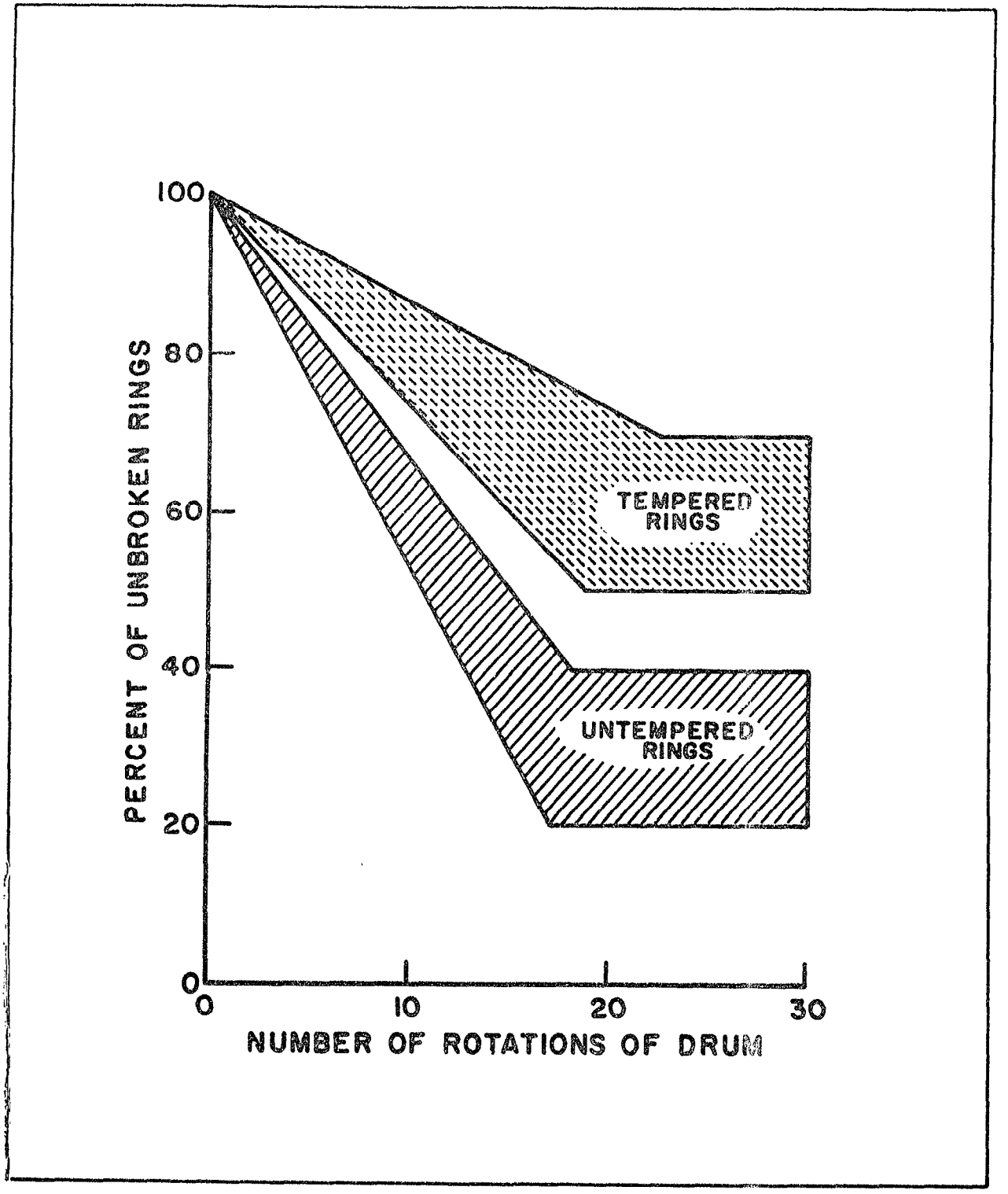

Fig. 2. The Effect of Number of Rotations on the Percentage Survival of Tempered and Untempered 1.5-in.-o.d. Raschig Rings Subjected to the Standard Mechanical Shock Resistance Test.

3. SPECIFICATIONS FOR PACKED VESSELS

Early application of the raschig rings included random packing in unmodified standard vessels of large dimensions. Over the past 10 yr various modifications of vessels to facilitate the use of rings have evolved. Several of these special design features listed below are shown in Figs. 3 and 4.

1. A glass port on top of the vessel facilitates visual inspection of the level of the packed rings.

2. Capacitance probes preclude overfilling the vessel by stopping pumps. (Overfilling would not result in a nuclear incident because the rings completely fill the vessel.)

3. An external torus, connected to the pump system, allows recirculation and mixing of the fissile solution.

4. A drop-out port on the bottom of the vessel permits easy removal of rings for cleaning.

5. Rings may be contained in a perforated steel basket.

Tubes for sampling glass rings are desirable when the liquid in the vessel is corrosive.

In the ORNL ^{233}U Liquid Storage Facility,[9] which has a capacity of 500 kg of ^{233}U as a solution at a concentration of 250 g of ^{233}U/liter, the five 240-gal tanks are instrumented for remote determination of liquid level, liquid specific gravity, temperature, and ring level. The ring level is determined periodically by a vertical traverse of a central "dry" reentrant pipe with an ultrasonic transducer and sensor.[b] The solution is mixed by air sparging.[32,33] Radiolytic hydrogen is continuously purged from the tanks by air flow from the pneumatic instruments. A full-length stringer of rings may be withdrawn for sampling through a perforated reentrant pipe that passes through the 4-in.-thick lead shielding wall. These capabilities are illustrated in Fig. 5.

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[b] In subsequent installations the ultrasonic method has been abandoned in favor of simpler procedures which employ radiography or periodic recalibration of the free volume of the vessels as a function of height.

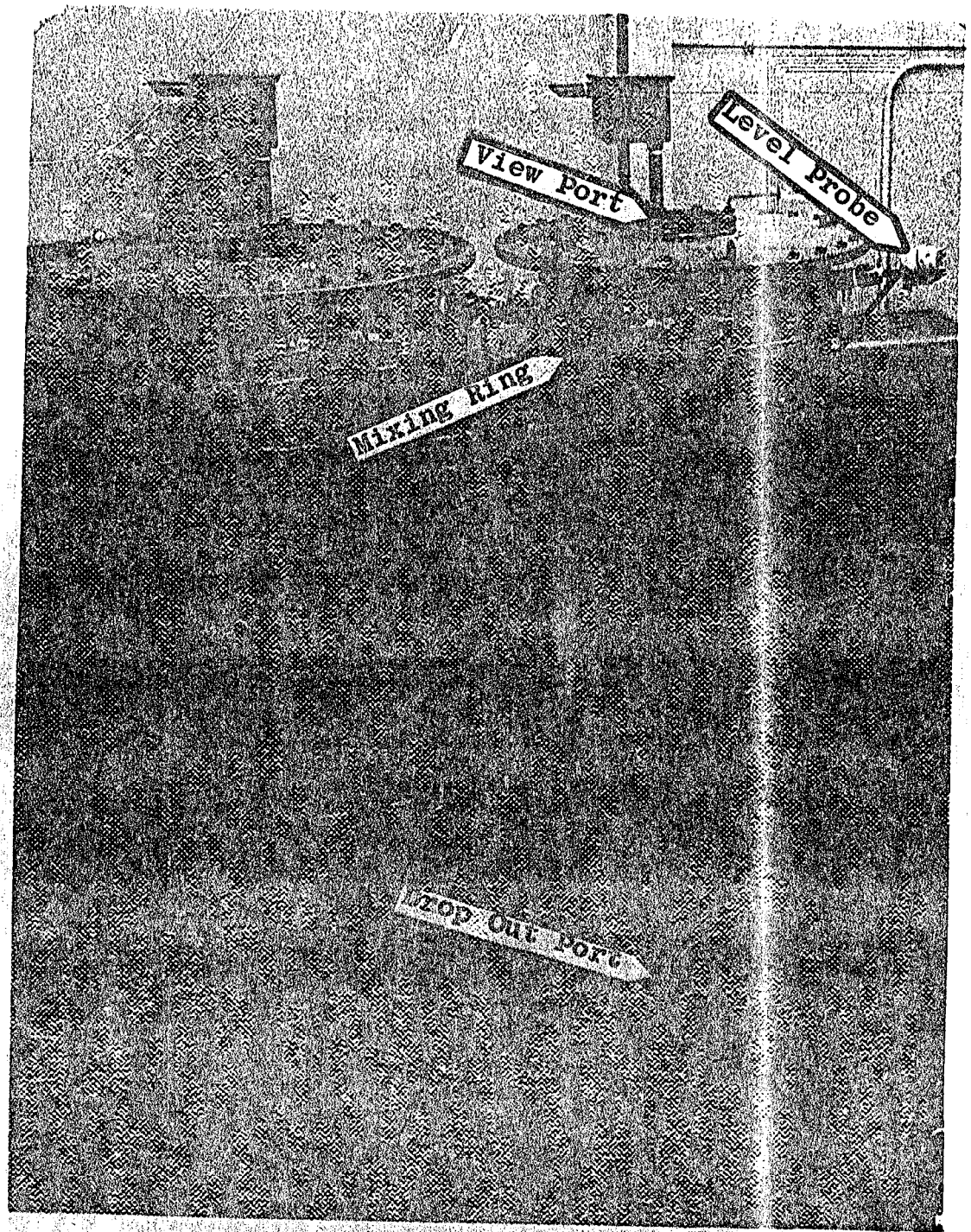

Fig. 3. Uranium Solution Storage Tanks in the Rocky Flats Plant Critical Mass Laboratory.

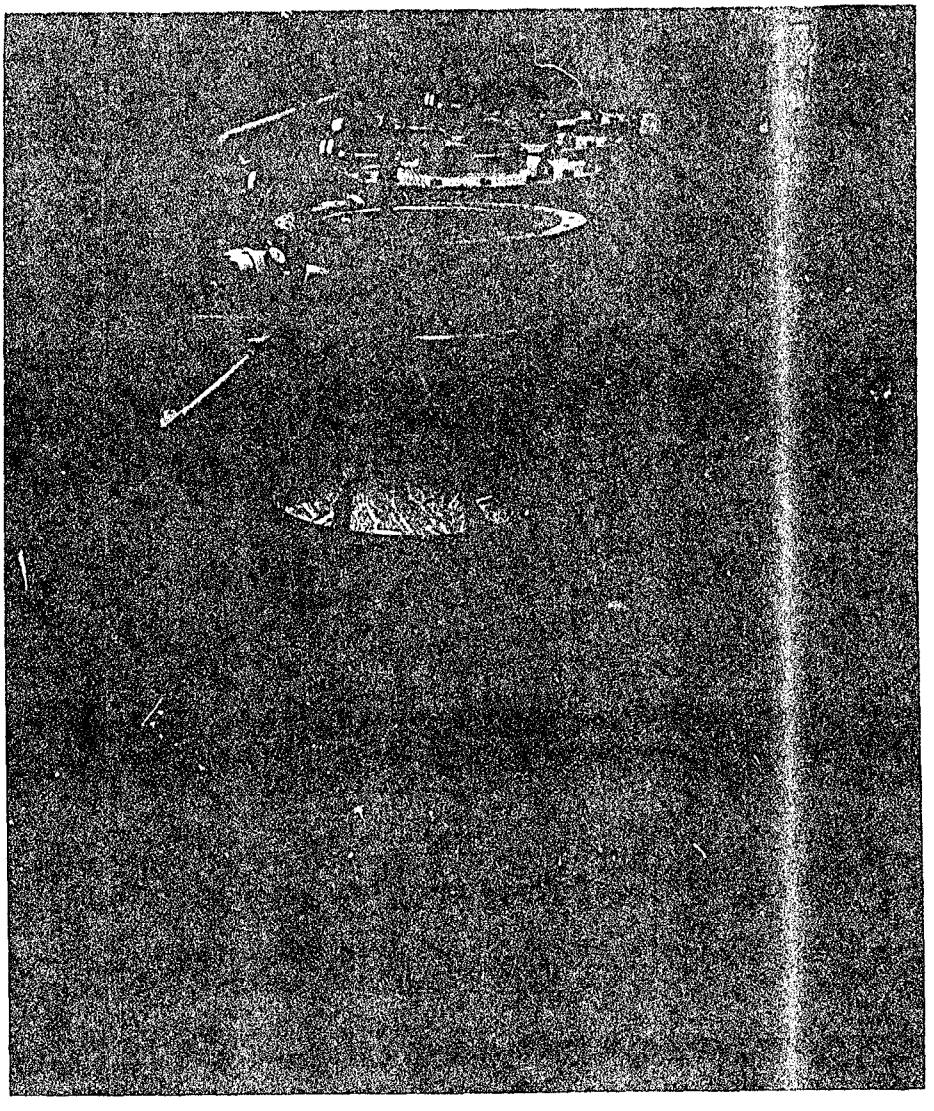

Fig. 4. Plutonium Solution Storage Tanks in the Rocky Flats Plant Critical Mass Laboratory.

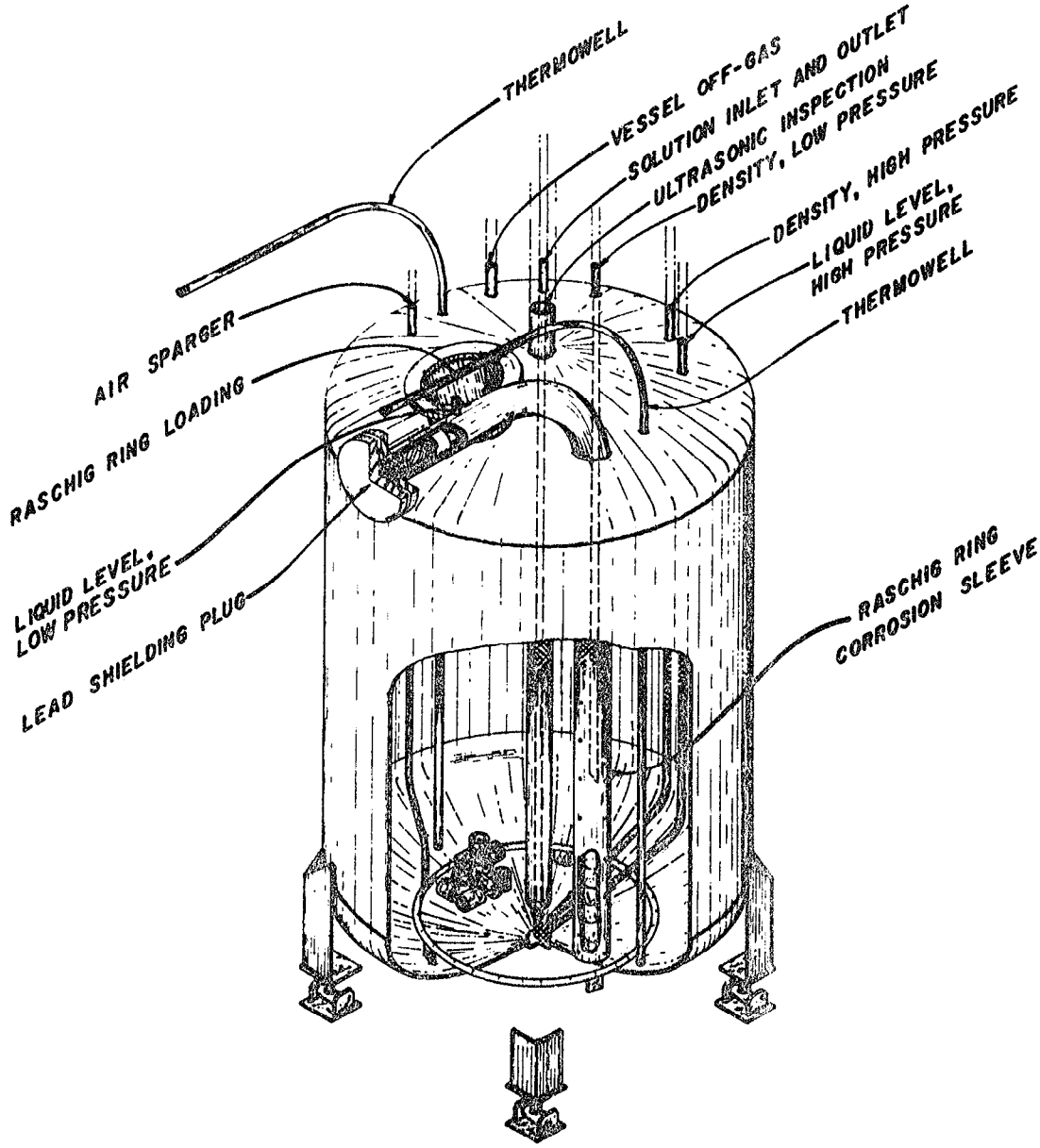

Fig. 5. Oak Ridge National Laboratory Storage Tank for ^{233}U Solution.

The stainless-steel floor pan beneath the tanks is filled with raschig rings to a depth of 11 in. as protection should a tank leak and the solution be diluted. Figure 6 is a photograph of a storage facility.

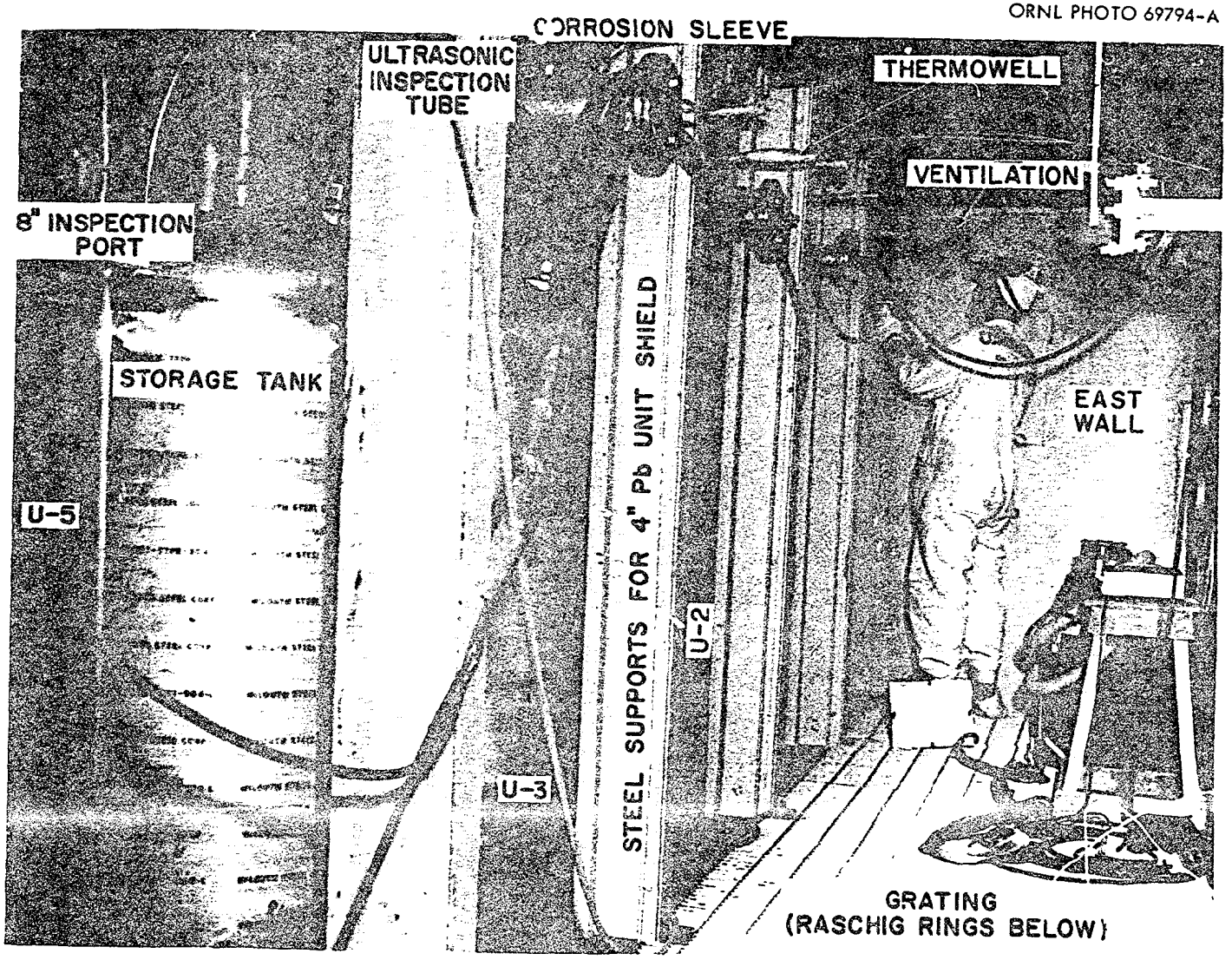

Fig. 6. Oak Ridge National Laboratory ^{233}U Liquid Storage Facility During Construction.

4. MAINTENANCE INSPECTION

Section 6 of the proposed standard specifies the inspection procedures which insure that the packed rings continue to provide the necessary protection against criticality. The vessels shall be inspected periodically to determine if the rings have settled. If the rate of settling (determined by the decrease in ring level between inspection intervals of less than 13 months) indicates that there will be no rings in the upper 5% of the vessel before the next inspection, rings should be added to restore the specified packing condition. The inventory of fissile material in the vessel shall be redetermined from the measured volume of solution and a reanalysis of the fissile material content. If any discrepancy in mass from the previous inventory, including possible analytical errors, exceeds 1000 g of fissile material, rings shall be removed for analysis of deposits. If the rings do not contain the fissile material, the vessel and rings shall be cleaned. The rings shall also be inspected for changes in their chemical and plysical properties. A limit of 50 g of U or Pu per liter of glass for the fissile-solids accumulation has been allowed instead of specifying the requirements necessary to maintain the fissile material in solution. For example, in a packed vessel having a glass volume fraction of 0.32 and a solution volume fraction of 0.68, this limit would increase the solution concentration 23.5 g per liter if the solids were dissolved and is conservatively smaller than the amount of fissile solids accumulation assumed for the criticality calculations (see Section 5).

Inspection procedures are necessary to insure that undue amounts of fissile material have not precipitated from the solution. The material balance inspection for solids may impose inconvenience in some types of operations. At this time, the authors are not confident of alternative procedures and, because of the importance of fissile solids, have decided not to provide a relaxation clause. It is expected that the 1000 g limit, for the "mass unaccounted for" possibly as precipitates and not as solids accumulated on the rings, will be reviewed periodically for possible revision if data substantiating other methods become available. The use of glass raschig rings for primary criticality control is not recommended if samples of rings cannot be withdrawn for inspection from regions suspected of harsh chemical or physical attack.

5. CRITICALITY OF FISSILE SOLUTIONS CONTAINING BOROSILICATE GLASS

Neutron absorbers in fissile solutions increase greatly the concentration of fissile isotopes that can be safely stored in large vessels. Borosilicate glass raschig rings have been in use for this purpose in several laboratories and plants, and nuclear safety has been assured by direct comparison to neutron multiplication measurements or other experiments. There has been a need for a calculational model for which safe concentrations could be predicted for many different solution-glass combinations. The analytic study, reported in this section, uses a simple model for comparing calculations to experimental results. This model, called the Cylindrical Tube Model, consists of an infinite tube of glass in a larger cylinder of solution. A mirror boundary condition at the outer surface of the model returns all neutrons; there is no leakage. The ANISN transport code[34] in the S_4 approximation with Hansen-Roach 16-group cross sections[35,36] was used. Results are also compared to other calculational models to verify that the simplifications do not seriously affect the results. The geometry in these other models was amenable to Monte Carlo codes and not to transport codes; hence, these calculations were done using the KENO Monte Carlo code[37] with the same cross sections. The geometry of some of the critical experiments was also not suited for transport calculations and the KENO Monte Carlo code was used for this comparison. The results of ANISN transport and KENO codes, when used with the same cross sections, have been shown to be consistent.[38]

The Hansen-Roach 16-group cross sections have been extensively used in one- and two-dimensional transport codes. Their accuracy for calculations of simple geometry has been proved through many years of use and therefore justifies their use in this study.

The initial part of the calculational program was designed to determine the dependence of k_∞ on various parameters such as boron content, glass volume fraction, radii changes, and solution volume fraction outside the glass using the Cylindrical Tube Model. These preliminary

calculations and the evaluation of the limited experimental data have led to a better understanding of the important variables. It is therefore with more confidence that the Cylindrical Tube Model can be used to make survey calculations for the range of glass properties specified in the proposed standard for the use of borosilicate glass raschig rings for criticality control.

5.1 Preliminary Calculations of the Dependence of k_∞ on Variables in the Cylindrical Tube Model

As a result of an experimental program with uranyl nitrate solution at an enrichment of 92.6 wt % ^{235}U and glass raschig rings, Thomas et al.[39] have reported that k_∞ should be unity in a system containing fissile solution and glass if the solution contained 415 g U per liter and the glass volume fraction was 0.222, the glass having 4 wt % boron, or in a system of the same solution having the glass volume fraction of 0.24 with 3.3 wt % boron. Because the experiments included raschig ring diameters from 0.61 to 1.85 in., it was uncertain what would be the appropriate dimensions of the glass and solution in the Cylindrical Tube Model for the calculations of the experimental results. Therefore, a number of preliminary calculations[40] were made using the ANISN transport code and a solution of 415 g of U per liter to establish the dependence of the calculated k_∞ on boron content and dimensional changes which altered glass volume fractions and solution volume fractions outside the glass. These exploratory calculations, later expanded to include larger glass fractions, illustrated some of the variables and considerations that may affect k_∞ for a fissile solution containing glass raschig rings.

Raschig ring dimensions[c] of 1.25 in. i.d. ($R_1 = 1.5875$ cm) and 1.50 in. o.d. ($R_2 = 1.905$ cm) and the solution o.d. of 1.6929 in. ($R_3 = 2.1500$ cm). which was estimated from an assumed glass volume fraction[d] of 0.24, were chosen to be the initial dimensions of the Cylindrical Tube Model. These

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[c] Although raschig ring dimensions are usually measured in inches, the radii for the model are given in centimeters because these were used in the calculations.

[d] The glass volume fraction = $(R_2^2 - R_1^2)/R_3^2$.

ring dimensions and glass volume fraction are typical values in the experiments of Thomas et al.

The variation of the calculated k_∞ as a function of the boron content in the glass for three different dimensions in the model is shown in Fig. 7. The first is described above. The second resulted from an increase in R_1 to 1.6329 cm, decreasing the glass volume fraction to 0.208; R_3 was then decreased to maintain the original glass volume fraction of 0.24. (See Items 1-9, Table 4, for the complete description of each calculational model.) It is seen that the dependence of k_∞ on boron content appears to be the same for the three models; however, the k_∞ values for the several dimensions in the model differ by as much as 3.3%.

If all dimensions are changed by the same scale factor, the volume fractions for solution and glass remain unchanged. The variation of k_∞ as a function of the scale factor normalized to unity at 1.50 in. o.d. for the glass tube is given in Fig. 8. (See Items 1 and 10-15, Table 4.) For a 0.1-in. decrease in glass o.d. from 1.5 in. o.d., $\Delta k_\infty = -0.0148$ (Items 1 and 12, Table 4). It is therefore conservative to have glass which is smaller than the diameter specified. The change in k_∞ for the range of glass outside diameters used in the experiments of Thomas et al., i.e., from 0.61 to 1.85 in., is estimated from Fig. 8 to be +0.18. It was this large change in k_∞ with ring size which emphasizes the difficulty in choosing appropriate ring dimensions for the estimated or interpolated experimental conditions for which $k_\infty = 1$.

In Fig. 9, the variation of k_∞ is shown as a function of the volume fraction outside the glass.[e] (See Items 7 and 36-43, Table 4.) The glass fraction and R_3 were held constant. The glass geometry changes from an outer annulus to a centrally located cylinder as the volume fraction outside the glass ranges from 0 to 0.76. The effects of the absorber and fuel rearrangement are quite pronounced, k_∞ changing from 1.07 to 1.20 with a minimum of 0.95.

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[e] The volume fraction outside the glass = $(R_3^2 - R_2^2)/R_3^2$.

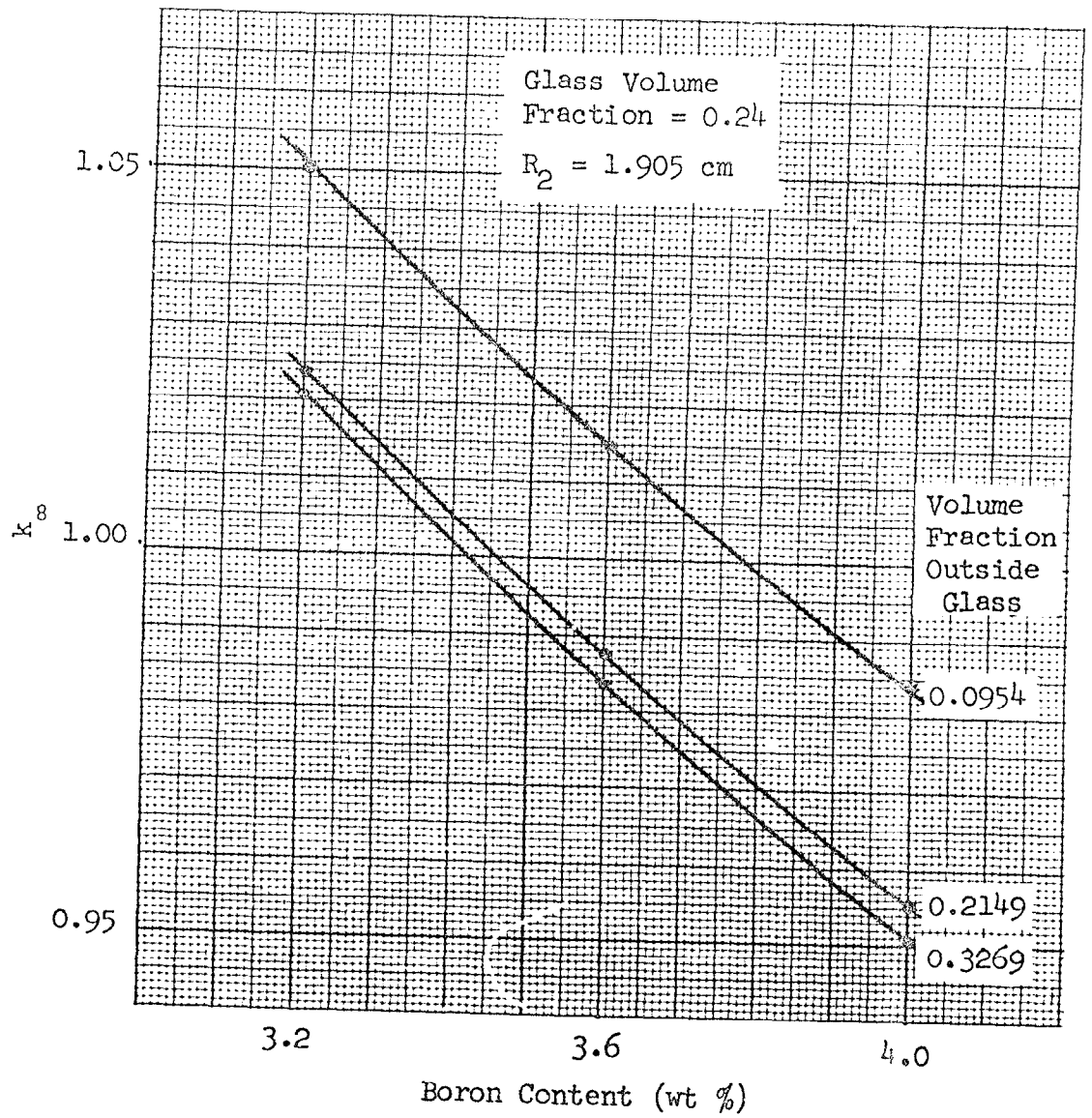

Fig. 7. Dependence of Calculated k_∞ upon Boron Content of Glass for Three Variations of the Dimensions of the Cylindrical Tube Model.

Table 4. Values of k_∞ from Preliminary ANISN Calculations by the Cylindrical Tube Model for 415 g/liter Uranyl Nitrate Solutions Using Various Dimensions and Boron Content.[a]

No.	Cylindrical Tube Model Radii (cm) Glass Inside R_1	Glass Outside R_2	Solution Outside R_3	Boron Content (wt %)	Volume Fractions Glass[b]	Solution Outside Glass[c]	k_∞
1	1.5875	1.9050	2.1500	4.0	0.24	0.2149	0.9556
2	1.5875	1.9050	2.1500	3.6	0.24	0.2149	0.9878
3	1.5875	1.9050	2.1500	3.2	0.24	0.2149	1.0243
4	1.6329	1.9050	2.0029	4.0	0.24	0.0954	0.9841
5	1.6329	1.9050	2.0029	3.6	0.24	0.0954	1.0153
6	1.6329	1.9050	2.0029	3.2	0.24	0.0954	1.0504
7	1.[illegible]36	1.9050	2.3220	4.0	0.24	0.3269	0.9510
8	1.5280	1.9050	2.3220	3.6	0.24	0.3269	0.9830
9	1.5280	1.9050	2.3220	3.2	0.24	0.3269	1.0211
10	1.2700	1.5240	1.7200	4.0	0.24	0.2149	0.9107
11	1.3758	1.6510	1.8633	4.0	0.24	0.2149	0.9258
12	1.4817	1.7780	2.0067	4.0	0.24	0.2149	0.9408
13	1.6933	2.0320	2.2933	4.0	0.24	0.2149	0.9696
14	1.7992	2.1590	2.4367	4.0	0.24	0.2149	0.9839
15	1.9050	2.2860	2.5800	4.0	0.24	0.2149	0.9964
16	1.5875	1.9050	2.2663	4.0	0.216	0.2934	0.9962
17	1.5875	1.9050	2.4038	4.0	0.192	0.3720	1.0572
18	1.6220	1.9050	2.1500	4.0	0.216	0.2149	1.0028
19	1.6559	1.9050	2.1500	4.0	0.192	0.2149	1.0538
20	1.6329	1.9050	2.1113	4.0	0.216	0.1859	1.0075
21	1.6329	1.9050	2.2393	4.0	0.192	0.2763	1.0497
22	1.5698	1.9050	2.3220	4.0	0.216	0.3269	0.9994
23	1.6105	1.9050	2.3220	4.0	0.192	0.3269	1.0514
24	1.4558	1.9050	2.3220	4.0	0.28	0.3269	0.8766
25	1.3797	1.9050	2.3220	4.0	0.32	0.3269	0.8077
26	1.4796	1.9050	2.4593	4.0	0.240	0.40	0.9643
27	1.5240	1.9050	2.4593	4.0	0.216	0.40	1.0119
28	1.5709	1.9050	2.4593	4.0	0.192	0.40	1.0629
29	1.3912	1.9050	2.4593	4.0	0.28	0.40	0.8912
30	1.3014	1.9050	2.4593	4.0	0.32	0.40	0.8250
31	1.6607	1.9050	1.9050	4.0	0.24	0.00	1.0193
32	1.9000	1.9050	2.3971	4.0	0.24	0.3684	0.9568
33	1.4500	1.9050	2.5220	4.0	0.24	0.4298	0.9737
34	1.5280	1.9050	2.4480	4.0	0.216	0.3944	1.0106
35	1.5280	1.9050	2.5960	4.0	0.192	0.4615	1.0821
36	2.0243	2.3220	2.3220	4.0	0.24	0.0000	1.0755
37	1.7552	2.1000	2.3220	4.0	0.24	0.3821	0.9831
38	1.6450	2.0000	2.3220	4.0	0.24	0.3581	0.9611
39	1.3950	1.8000	2.3220	4.0	0.24	0.3991	0.9514
40	1.1252	1.6000	2.3220	4.0	0.24	0.3252	0.9813
41	0.8161	1.4000	2.3220	4.0	0.24	0.6365	1.0468
42	0.5182	1.2500	2.3220	4.0	0.24	0.7102	1.1212
43	0.0000	1.1375	2.3220	4.0	0.24	0.7600	1.2020
44	1.4213	1.9050	2.3971	4.0	0.28	0.3684	0.8827
45	1.3380	1.9050	2.3971	4.0	0.32	0.3684	0.8149

a. The changes in the model and the results of these calculations are discussed in Section 5.1.

b. The glass volume fraction = $(R_3^2 - R_1^2)/R_3^2$.

c. The volume fraction outside the glass = $(R_3^2 - R_2^2)/R_3^2$.

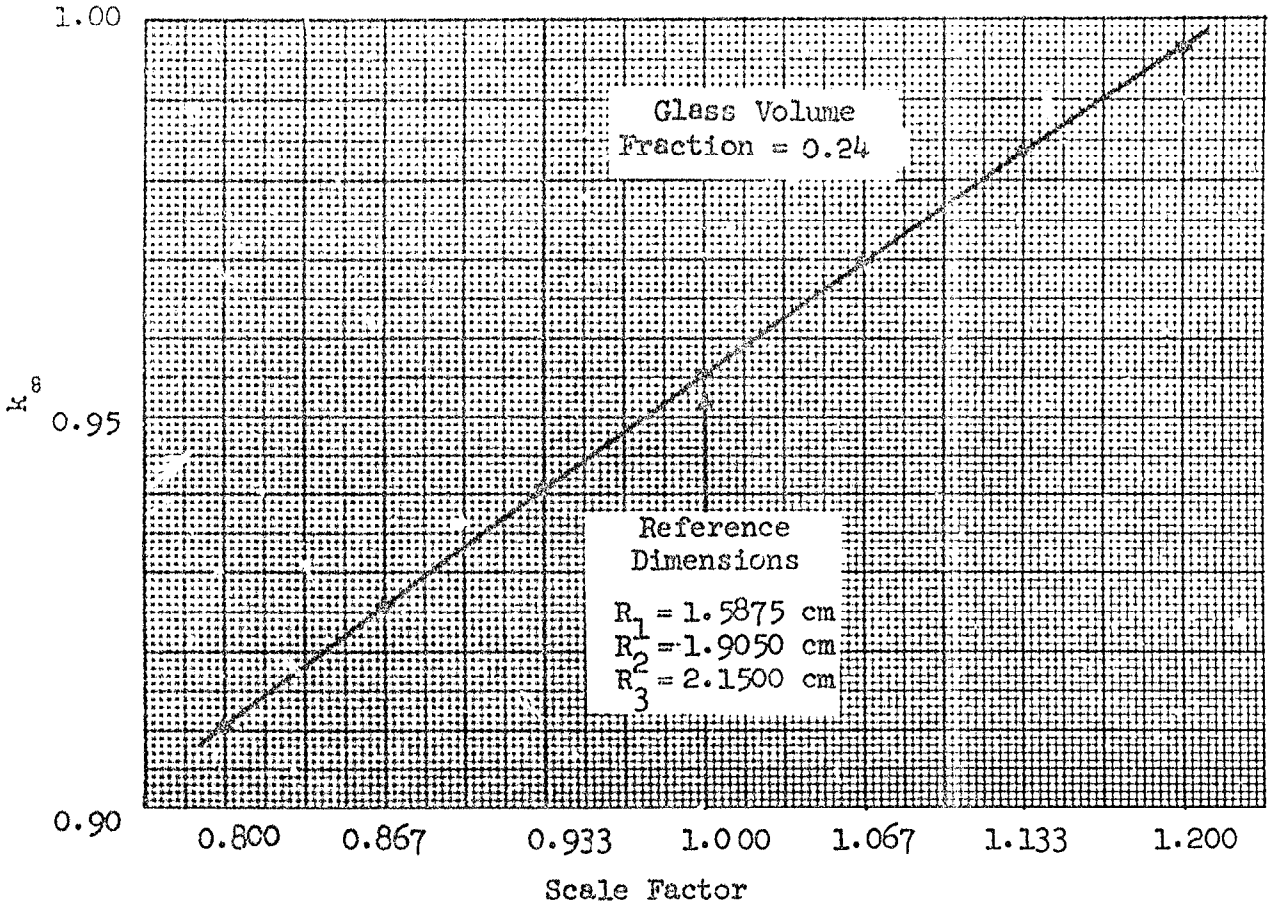

Fig. 8. Dependence of k_∞ upon the Dimensions in the Cylindrical Tube Model, All Diameters Scaled by the Same Factor. The glass tube outside diameter varied from 1.2 to 1.8 in.

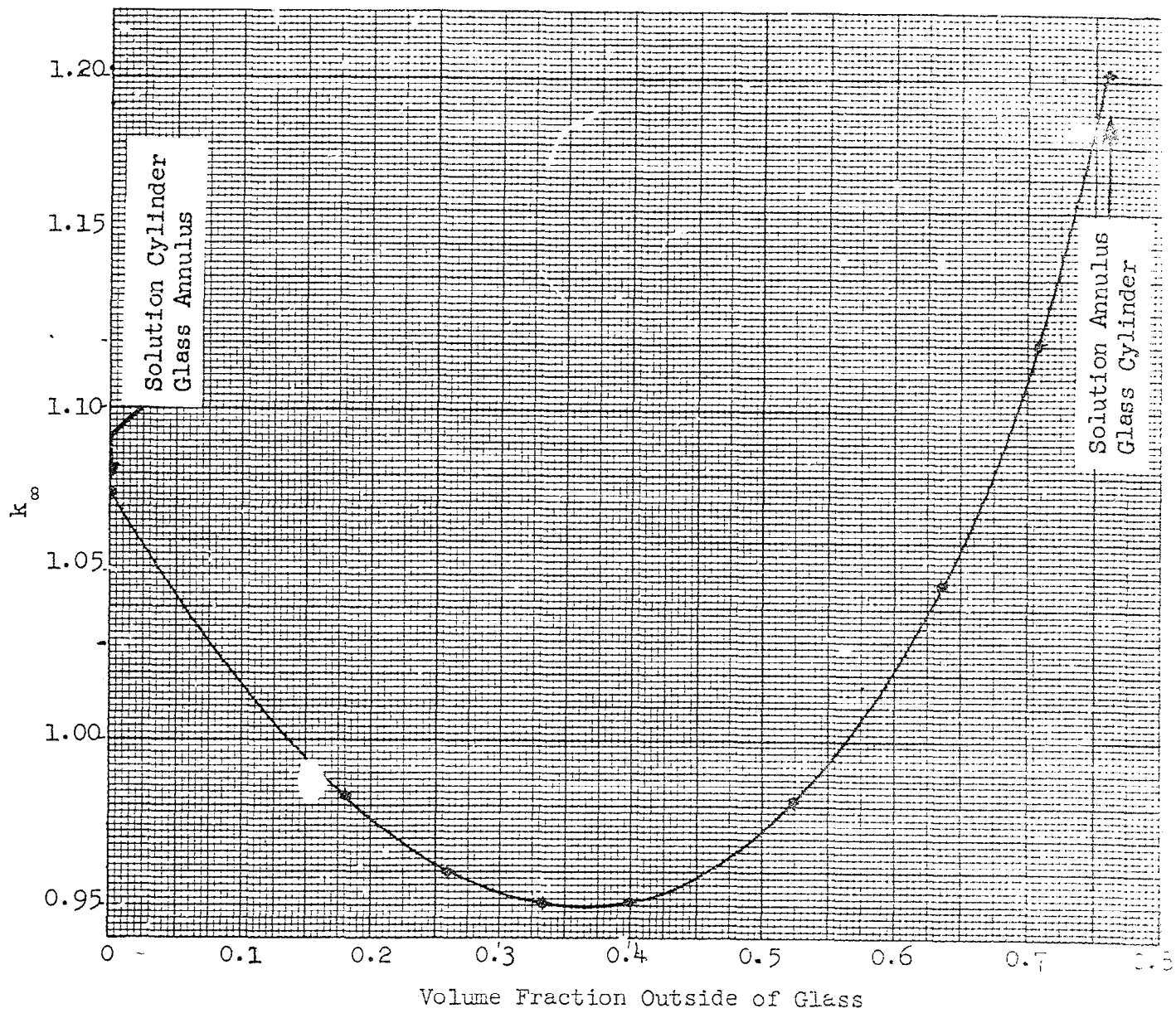

Fig. 9. Dependence of Calculated k_∞ on Volume Fraction Outside Glass.

The variation in k_∞ with the volume fraction outside the glass for various glass volume fractions from 0.192 to 0.32 (see Items 1, 4, 7, 16, 35, 44, and 45 in Table 4) is given in Fig. 10. For these calculations a constant glass tube o.d. of 1.50 in. was assumed ($R_2 = 1.905$ cm) and the variations in the glass volume fraction and the volume fraction outside the glass were achieved by changing R_1 and R_3. The glass contained 4 wt % boron. Also shown in the figure by connecting dashed lines are the calculations which have equal glass inside radius (R_1). These lines connect data points for which the glass volume fraction is no longer constant. These lines illustrate the variation in calculated k_∞ as the volume of solution outside the glass was changed. It is well known that the number raschig rings that can be put into a fixed volume varies with the manner in which they are dumped and stirred, thereby varying the glass volume fraction and the volume fraction outside the glass. Packed beds of rings have been observed to have volume fractions outside the rings from 0.33 to 0.41 (see Table 7, Section 5.3) and k_∞ increases by 0.08 over this range for $R_1 = 1.5280$ cm and $R_2 = 1.9050$ cm.

It is noted that the initial calculational model (Item 1, Table 4), which was based on nominal ring dimensions and a measured glass fraction, did not predict a value of the volume fraction outside the glass, consistent with the volume fractions observed in experimental packed vessels. This discrepancy emphasized the fact that accurate glass dimensions would be necessary in the calculations of k_∞ for experiments and that it would not be fruitful to estimate appropriate radii for the $k_\infty = 1$ conditions interpolated from exponential experiments using many sizes of rings. Therefore, each critical and exponential experiment for which there were available accurate measurements of the ring diameters was evaluated independently. It was expected that this comparison of calculation and experiment would then provide the proper validation of a calculational method which would then be used to make survey calculations for the various fissile solutions-raschig ring mixtures.

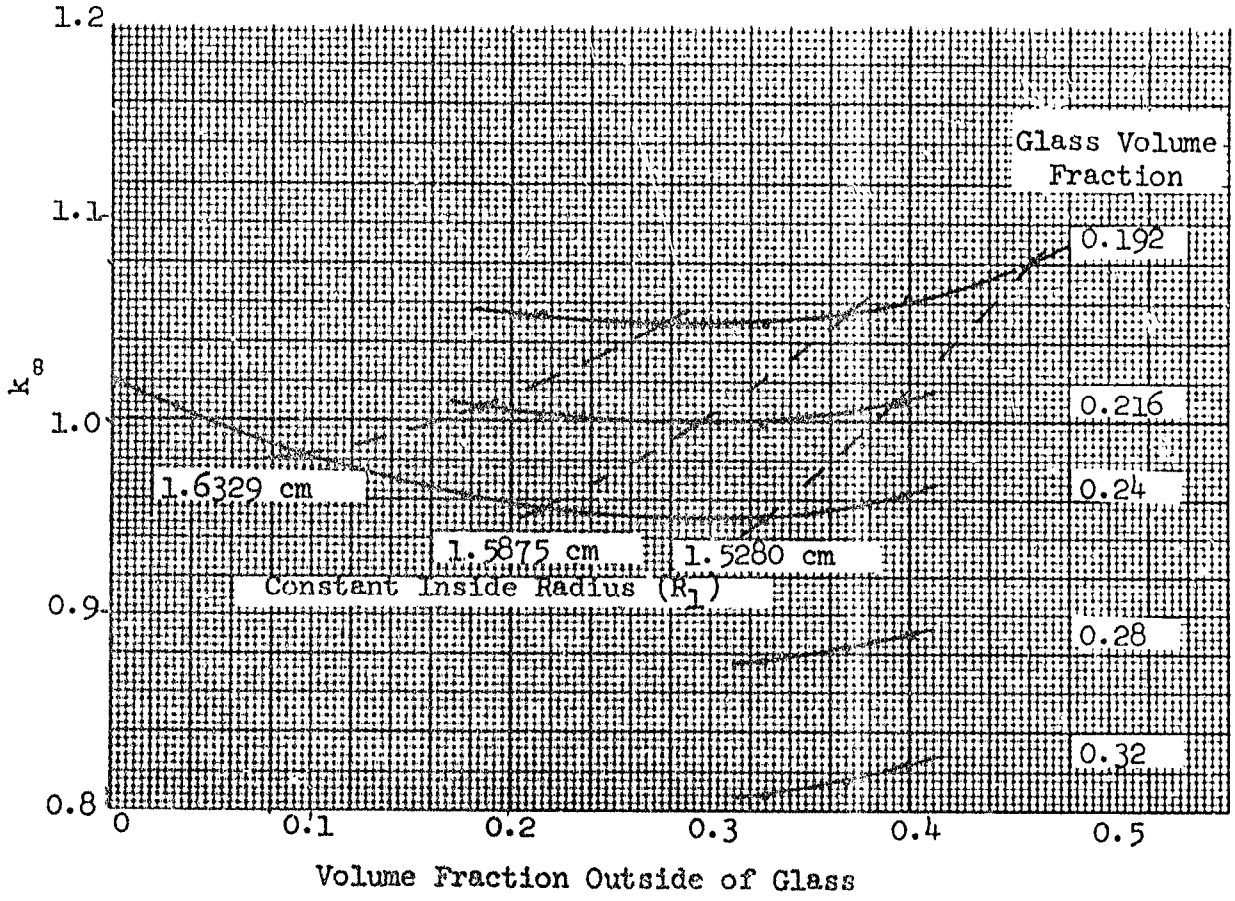

Fig. 10. Calculated k_∞ for Various Glass Volume Fractions as a Function of Volume Fraction Outside of Glass for Constant Glass Tube o.d. of 1.50 in. (R_2 = 1.905 cm).

5.2 Critical Experiments with ^{235}U Solutions and Glass Pipes

The critical experiments of Fox and Gilley[41] provide some reference experimental data for validation of computation methods and cross sections. In these experiments various sizes of Pyrex glass pipe were placed in hexagonal arrays in cylindrical vessels. The critical height of fissile solution was determined. Six experiments, described in Table 5, were selected for this validation. The KENO Monte Carlo code with a geometry routine[42] which describes the hexagonal array of the glass pipes and with Hansen-Roach 16-group cross sections was used to calculate k_{eff} for the critical experiments [$k_{eff}(exp) = 1$].

The calculated k_{eff} of the two experiments having no glass pipes were about 2.5% less than 1. Previous experience with the KENO code and Hansen-Roach cross sections has resulted in better agreement with experiment (± 0.01) for hydrogen-moderated and -reflected systems, and the present larger disagreement is not understood. The calculated k_{eff} for the glass pipe-solution systems, with one exception, were also less than unity by about the same amount and it is believed that the calculational method is satisfactory for calculating glass pipe-fissile solution systems. These calculations are also listed in Table 5.

5.3 Raschig Ring Exponential Experiments

A calculational scheme was used to evaluate some of the parameters in a simple model for raschig ring exponential experiments in cylindrical geometry so that an experimental value for $k_\infty(exp)$ is obtained from measured reciprocal relaxation lengths for comparison to k_∞ calculated from the Cylindrical Tube Model. The equation for obtaining k_∞ from the experimental data is

$$k_\infty(exp) = 1 + M^2B_r^2 - M^2\gamma^2$$

where

M^2 = $L^2 + \tau$ = the migration area

τ = the neutron age to thermal energy

B_r^2 = the radial buckling

γ = the reciprocal relaxation length

L = the thermal diffusion length.

Table 5. Calculated Multiplication Factors Using the KENO Monte Carlo Code for $^{235}UO_2(NO_3)_2$ Solution Critical Experiments with Glass Pipes.

Glass Pipes			Critical Height of Solution[a] (in.)		Calculated[b] Multiplication Factor k_{eff}	
No.	Diameter (in.)	Hexagonal Pitch (in.)	Unreflected	Water[c] Reflected	Unreflected With Reflector Water Tank[d]	Water Reflected[e]
0			5.97	4.98	0.974 ± 0.005	0.969 ± 0.006
19	2.0	3.00	16.7	9.75	0.975 ± 0.004	0.959 ± 0.007
31	1.5	2.75	26.6	12.7	0.975 ± 0.004	1.000 ± 0.008

a. 87.4 wt % ^{235}U at a concentration of 331.2 g of U per liter, $H/^{235}U = 80.26$, $N/U = 2.15$. These revised values are slightly different from those reported in Ref. 41.
b. No correction is included for the sections of glass pipes and 20-in.-diam steel cylinder above the solution nor for the 3-in.-diam solution port at the bottom of the cylinder. Including these the increase in k_{eff} was 0.004 for the unreflected experiment with 19 glass pipes and 0.002 for the unreflected experiment with 31 pipes.
c. Reflector water height was equal to solution height, no top reflector.
d. The reflector water tank was 9 ft in diameter with a 7/16-in.-thick steel wall. The 1 1/16-in.-thick steel bottom was 9 in. below the critical experiment cylinder.
e. The 3-in.-diam solution port was assumed to be water for these calculations.

The reciprocal relaxation length is determined from the exponential dependence of the neutron flux in the z direction of the cylinder filled with raschig ring-solution mixture in the exponential experiment. $M^2 (= \tau + L^2)$ and $M^2B_r^2$ are calculated parameters. The neutron age, τ, which is equal to one-sixth the second moment of the slowing down distribution from a point fission source, was calculated using the ANISN-S_{32} method of Raffety and Mihalczo.[43] The thermal diffusion length, L, is inversely proportional to $\sqrt{\Sigma_{at}}$. The radial nonleakage probability, $1/(1 + M^2B_r^2)$, was calculated from the equation

$$k_{eff} = \frac{k_\infty}{1 + M^2B_r^2}$$

where k_∞ was calculated for the fissile solution-raschig ring mixture using the Cylindrical Tube Model. Cell-averaged, homogenized cross sections, generated from the k_∞ calculation, were used in a subsequent ANISN reactor calculation of k_{eff} for an infinitely long cylinder of the fissile solution-raschig ring mixture. Thus, $M^2B_r^2$ was derived from the two calculations. The calculated values of M^2, $M^2B_r^2$ and the experimental value of γ were then used to obtain $k_\infty(exp)$ for comparison to the calculated value of k_∞.

Table 6 summarizes these calculations for the ^{235}U and ^{233}U exponential experiments[39,44] where there were available measurements of the raschig ring inside and outside diameters. k_∞ was also calculated for the raschig rings and the plutonium solution concentration which was determined experimentally[45] to have $k_\infty(exp) = 1$ and is included in the table. The raschig rings used in the experiments are described in Table 7.

The differences between the experimental values and the calculated values of k_∞ for these raschig ring-fissile solution systems are used to determine the value of the calculated k_∞ for which subcriticality is assured, see Section 5.6.

Table 6. Comparison of Calculated k_∞ with Experimentally Determined k_∞ Values.

U or Pu Concentration (g/liter)	Raschig Ring Type[a]	Cylinder Diameter (in.)	Reflector	Reciprocal (Relaxation Length) γ^2 (cm^{-2})	ANISN Calculations k_∞	ANISN Calculations k_{eff}	ANISN Calculations τ (cm^2)	L^2 (cm^2)	M^2 (cm^2)	$M^2B_r^2 = \frac{k_\infty}{k_{eff}} - 1$	k_∞(exp)
Uranyl Nitrate Solution Containing 92.6 wt % ^{235}U[b]											
415	EN-1	48	Air	0.00554	0.8180	0.7765	39.03	0.19	39.22	0.0534	0.836 ± 0.048
415	EN-1	48	H_2O	0.00497	0.8180	0.7816	39.03	0.19	39.22	0.0466	0.852 ± 0.054
415	Pyrex	30	H_2O	0.00131	1.0674	0.9766	38.25	0.20	38.45	0.0930	1.043 ± 0.015
415	Pyrex	30	Air	0.00216	1.0674	0.9520	38.25	0.20	38.45	0.1212	1.038 ± 0.020
415	EN-1	20	H_2O	0.01024	0.8180	0.6957	39.03	0.19	39.22	0.1758	0.774 ± 0.051
415	Pyrex	20	H_2O	0.00326	1.0674	0.9047	38.25	0.20	38.45	0.1798	1.062 ± 0.011
415	Pyrex	20	Air	0.00453	1.0674	0.8522	38.25	0.20	38.45	0.2525	1.078 ± 0.021
415	Pyrex	20	H_2O	0.00313	1.0674	0.9047	38.25	0.20	38.45	0.1798	1.060 ± 0.027
415	KG-33	20	H_2O	0.01461	0.7115	0.5968	42.91	0.15	43.06	0.1922	0.563 ± 0.053
415	KG-33	20	Air	0.01607	0.7115	0.5596	42.91	0.15	43.06	0.2714	0.579 ± 0.051
415	R6	20	Air	c	1.5467	c	41.11	0.28	41.39		
415	R6	20	H_2O	d	1.5467	d	41.11	0.28	41.39		
141	R6	20	Air	0.00194	1.1990	0.9417	39.81	0.65	40.46	0.2732	1.195 ± 0.009
141	R6	20	H_2O	0.00085	1.1990	0.9919	39.81	0.65	40.46	0.2088	1.174 ± 0.011
141	R6	20 + 0.032 Cd	Air	0.00181	1.1990	0.9417	39.81	0.65	40.46	0.2732	1.200 ± 0.008
141	R6	20 + 0.032 Cd	H_2O	0.00120	1.1990	0.9613	39.81	0.65	40.46	0.2473	1.199 ± 0.005
94.4	R6	20 + 0.032 Cd	Air	0.00530	1.0178	0.7986	39.75	0.77	40.52	0.2745	1.060 ± 0.024
94.4	R6	20 + 0.032 Cd	H_2O	0.00480	1.0178	0.8135	39.75	0.77	40.52	0.2745	1.057 ± 0.023
63.3	R6	20 + 0.032 Cd	Air	0.01080	0.8278	0.6495	39.86	0.93	40.79	0.2745	0.834 ± 0.052
63.3	R6	20 + 0.032 Cd	H_2O	0.01088	0.8278	0.6614	39.86	0.93	40.79	0.2516	0.808 ± 0.045
Uranyl Nitrate Solution Containing 97.6 at. % ^{233}U[e]											
332.6	A	20	Air	0.00863	0.9362	0.7079	49.58	0.20	49.78	0.3225	0.893 ± 0.007
345.9	B	20	Air	0.00723	0.8927[f]	0.6988	47.50	0.15	47.65	0.2960	0.951 ± 0.006
204.1	B	20	Air	0.01432	0.6329	0.4921	45.27	0.18	45.45	0.2861	0.632 ± 0.014
Plutonium Nitrate Solution Containing 95.8 at. % ^{239}Pu[g]											
182	C	--	--	0.00000	1.0274	--	--	--	--	--	1.000

a. See Table 8 for raschig ring descriptions.
b. See Ref. 39 for complete description.
c. Critical height = 10.86 in. k_{eff} was calculated by KENO to be 0.956 ± 0.006 using cell-averaged cross sections from the ANISN k_∞ calculation.
d. Critical height = 8.34 in. k_{eff} was calculated by KENO to be 0.925 ± 0.006 using cell-averaged cross sections from the ANISN k_∞ calculation.
e. See Ref. 44 for complete description.
f. S_8 calculation was 0.9042 which should be compared to 0.951.
g. See Ref. 45 for complete description.

Table 7. Descriptions of Raschig Rings Used in Experiments.

Type	Boron Content wt %	Measured Dimensions o.d. (in.)	Measured Dimensions i.d. (in.)	Measured Dimensions Length (in.)	Measured Glass Volume Fraction	Radii of Cylindrical Tube Model R_1 (cm)	Radii of Cylindrical Tube Model R_2 (cm)	Radii of Cylindrical Tube Model R_3 (cm)	Solution Frection Outside Glass
EM-1	5.70	1.56	1.25	1.73	0.241[a]	1.5875	1.9812	2.4145	0.3267
Pyrex	3.99	1.853	1.515	1.893	0.209[a]	1.9245	2.3533	2.9638	0.3696
KG-33	3.88	0.61	0.43	0.64	0.300[a]	0.5461	0.7747	1.0032	0.4037
R-6	0.5%	1.60	1.28	1.69	0.240[a]	1.6256	2.0320	2.4887	0.3333
A	3.97	1.458	0.939	1.719	0.380	1.1925	1.8517	2.2979	0.3506
B	3.93	0.617	0.426	0.652	0.345	0.5410	0.7836	0.9651	0.3408
C	4.0	1.181	1.024	1.181	0.161	1.3000	1.5000	1.8650	0.3531

a. These measured values may be uncertain by ± 0.02.

5.4 Comparison of the Calculated k_∞ from the ANISN-Cylindrical Tube Model with Those from Other Calculational Models

The Cylindrical Tube Model may be in error for two reasons: first, the unknown and complex geometry of the solution outside the rings in a packed bed was approximated by an annulus of solution having a perfect mirror reflector; second, neutrons from the solution can enter the surface at the ends of a ring which has a surface-to-volume ratio different from that for a glass tube.

Estimates of the latter effect were made by changing the glass and solution radii in the Cylindrical Tube Model to maintain both the same glass volume fraction and the surface-to-volume ratio for the rings. The measured ring height was used and the ends were assumed to be flat for the ring surface calculation. In one method, the inside radius of the glass (R_1) was increased to maintain a constant surface-to-volume ratio and the solution radius (R_3) was decreased to maintain the correct glass volume fraction. In the second method, the glass outside radius (R_2) was decreased and the solution radius (R_3) was decreased. It is observed from the results of these calculations given in Table 8 that the Cylindrical Tube Model using the measured ring radii predicts a higher k_∞ value, a conservative value for nuclear criticality safety.

Table 8. Variation of k_∞ with Changes in Radii to keep the Surface-to-Volume Ratio Equal for Tubes and Rings.

		k_∞		
			Tube Surface-to-Volume Ratio Equal to That of Ring	
	Raschig Ring[a]	Tube Model	Inside Radius, R_1, Increased	Outside Radius, R_2, Decreased
^{235}U	Pyrex	1.0674	1.0596	1.0572
^{239}Pu	C	1.0274	1.0250	1.0241

a. See Table 7 for ring description.

It is also observed that the arbitrary increase in the inside radius of the glass represented by Items 4, 5, and 6 in Table 4 when compared to Items 1, 2, and 3 show a 3% increase in k_∞. For these cases the volume fraction outside the glass changes in a very unrealistic manner and the indicated reverse trend is ignored.

Two other models were evaluated using the KENO Monte Carlo code and Hansen-Roach 16-group cross sections. In one model, the geometry of the solution outside the glass was an infinitely long square tube of solution in which was placed the cylindrical glass tube. In the actual calculation, a cube of solution having a mirror boundary on all six faces contained one raschig ring whose length was the cube dimension. The second model resulted from some calculations by Webster[46] who had suggested that raschig rings with axes oriented mutually perpendicularly should be a better approximation to a random array than a square array of tubes. Using the 05R Monte Carlo code,[47] he calculated k_{eff} for $7 \times 7 \times 9$ arrays[f] of rings having various glass sizes and compared the results to k_{eff} for the same array in which the rings were oriented to make parallel glass tubes. The k_{eff} value for arrays having rings oriented mutually perpendicularly were lower by 1 to 6% depending upon glass size; however, none of the glass sizes was comparable to the ring sizes of the experiments or the proposed standard.

A calculational method was devised to calculate k_∞ for raschig rings oriented mutually perpendicularly from the first method by the use of a special boundary condition at the surfaces of the cube; i.e., all neutrons which arrive at one surface of the cube are not reflected but returned to another surface and a similar permutation is used for neutrons at the other surfaces.[48] The k_∞ values calculated from the Cylindrical Tube, the Square Array, and the Mutually Perpendicular Raschig Ring Models are given in Table 9. It is observed that the Square Array Model predicts higher k_∞ than the other two models in which the values are similar.

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[f] Larger arrays could not be described because of limitations of the computer memory.

Table 9. Comparison of Cylindrical Tube, Square Array Tube, and Mutually Perpendicular Raschig Ring Models.

Nitrate Solution of	U or Pu Concentration (g/liter)	Raschig Ring[a]	k_∞ Cylindrical Tube Model	k_∞ Square Array Tube Model	k_∞ Mutually Perpendicular Raschig Ring Model
^{235}U	415	Pyrex	1.0674	1.084 ± 0.005	1.066 ± 0.004
^{235}U	415	EN-1	0.8180	0.852 ± 0.003	0.818 ± 0.004
^{233}U	332.6	A	0.9362	0.971 ± 0.003	0.939 ± 0.003
^{233}U	345.9	B	0.8927	0.918 ± 0.003	0.906 ± 0.003
^{239}Pu	182	C	1.0274	1.043 ± 0.005	1.037 ± 0.005

a. See Table 7 for ring description.

5.5 Survey Calculations for Raschig Rings in ^{233}U, ^{235}U, and ^{239}Pu Nitrate Solutions

The survey calculations of the variation of k_∞ with fissile solution concentration for four glass volume fractions were done with the ANISN transport code in the S_4 approximation using Hansen-Roach 16-group cross sections. The methods used to obtain the atomic densities in the fissile solutions and the dimensions of the Cylindrical Tube Model for four glass volume fractions are now described. A number of perturbations from these standard conditions and the comparisons of calculated k_∞ values are also included in this section.

5.5.1 Molar Volumes and Water Displacement by Fissile Compounds. Aqueous solutions of uranyl nitrate, $UO_2(NO_3)_2$, are the most common chemical form used in chemical processing plants. Davis and Mrocheck[49] report values for the molar volume of $UO_2(NO_3)_2$ averaging 92.55 cm^3 which was used to obtain the amount of water displaced for the various concentrations of uranium.

Aqueous solutions of plutonium nitrate usually have various amounts of excess nitric acid for solution stability, but the excess nitric acid was assumed to be zero because it is conservative to neglect the effect of the acid on k_∞, i.e., k_∞ will be lower for solutions containing excess acid. The molar volume of $Pu(NO_3)_4$ was estimated to be 108.6 cm^3 from the density equation used for criticality calculations in the United Kingdom.[50] The value of 86.5 cm^3 was obtained from the density equation given by Richey.[51] The value of 108.6 cm^3 appeared to fit the analytical data for the composition of the solutions used in critical experiments[51] and was therefore used for this work.

5.5.2 Glass Composition. The minimum value of 3.66 wt % boron, the minimum $^{10}B/^{11}B$ isotopic ratio of 0.24, and the minimum glass density of 2.22 g/cm^3 result in an effective 10.3% decrease in the nominal boron density in 4 wt % B glass. The normal boron atom density was reduced from 4.966×10^{21} to 4.455×10^{21} atoms/cm^3 and is equivalent to 3.57 wt % boron in glass. This is consistent with the specifications for raschig rings; see Section 2.1.

5.5.3 Dimensions of the Cylindrical Tube Model for Various Glass Fractions. Two sets of specifications (see Section 2.3) for glass raschig rings were examined to estimate in the Cylindrical Tube Model the dimensions of the outside solution diameter and glass volume fractions based on the range of glass diameters included by the tolerances and assumed values for the volume fraction of solution outside the glass. This latter assumption represents the range of values observed in experiments (see Table 7).[g] It is noted in Table 10 that the calculated glass fraction ranges from 0.30 to 0.42 for the first, and from 0.21 to 0.34 for the second set of raschig ring specifications if volume

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[g] Variations in the volume fraction outside the glass are dependent on the manner in which a raschig ring is oriented with respect to its nearest neighbors. It is well known that stirring while rings are added to a vessel decreases the volume occupied by the rings by decreasing the volume fraction outside the glass.

Table 10. Raschig Ring Dimensions, Tolerances, and Variation of Glass Fraction from Which the Dimensions of Cylindrical Tube Model for Various Glass Fractions were Selected for Survey Calculations.

Raschig Ring Dimensions and Tolerances[a] (in.)			Raschig Ring Maximum and Minimum Dimensions		Calculated Glass Fractions for Volume Fractions Outside Glass of			Calculated Outside Solution Diameter (in.) for Volume Fractions Outside Glass of		
Diameter	Wall Thickness	Length	Outside Diameter (in.)	Inside Diameter (in.)	0.4038	0.3615	0.3268	0.4038	0.3615	0.3268
1.500	0.219	1.750	1.500	0.942	0.3611	0.3867	0.4077	1.9426	1.8772	1.8282
+0.000	+0.060	+0.035	1.500	1.062	0.2973	0.3185	0.3357	1.9426	1.8772	1.8282
-0.060	-0.000	-0.125	1.440	0.882	0.3725	0.3990	0.4206	1.8650	1.8020	1.7551
			1.440	1.002	0.3075	0.3293	0.3472	1.8650	1.8020	1.7551
1.500	0.150	1.750	1.500	1.080	0.2871	0.3075	0.3241	1.9426	1.8772	1.8282
+0.000	+0.060	+0.035	1.500	1.200	0.2146	0.2298	0.2423	1.9426	1.8772	1.8282
-0.060	-0.000	-0.125	1.440	1.020	0.2970	0.3181	0.3353	1.8650	1.8020	1.7551
			1.440	1.140	0.2225	0.2383	0.2512	1.8650	1.8020	1.7551

Dimensions of Cylindrical Tube Models for Survey Calculations

Glass Volume Fraction	Volume Fraction Outside Glass	Glass Tube Inside Radius R_1 (cm)	Glass Tube Outside Radius R_2 (cm)	Outside Solution Radius R_3 (cm)
0.20	0.3354	1.5926	1.9050	2.3368
0.24	0.3354	1.5227	1.9050	2.3368
0.28	0.3354	1.4491	1.9050	2.3368
0.32	0.3354	1.3716	1.9050	2.3368
0.24	0.4000	1.4756	1.9050	2.4593
0.32	0.4000	1.3014	1.9050	2.4593

a. See Section 2.3.

fractions outside the glass range from 0.3268 to 0.4038. An average value of the solution diameter was 1.84 in. which then resulted in a volume fraction outside the glass of 0.3354 for a glass o.d. of 1.50 in.

Four glass fractions, 0.20, 0.24, 0.28, and 0.32, were selected for survey calculations using the Cylindrical Tube Model. It was very important to choose the glass and solution radii so that the calculations would have meaning with respect to each other. The ring outside dimension was specified to the the largest permitted by the tolerances on the rings, 1.50 in. o.d. ($R_2 = 1.905$ cm). Since a solution diameter of 1.84 in. ($R_3 = 2.3368$ cm) was arbitrarily selected, the volume fraction outside the glass was 0.3354 for the survey calculations. After the dependence of k_∞ on the volume fraction outside the glass was found (see Figs. 9 and 10), additional calculations of k_∞ were made with this fraction increased to 0.40 by increasing the outside solution diameter to 1.9365 in. ($R_3 = 2.4593$ cm). The inside glass diameter was then calculated from the above dimensions and the desired glass volume fraction. Table 10 is a summary of the two raschig ring specifications, dimensions of the Cylindrical Tube Model for variations of volume fraction outside glass, and glass volume fraction based on maximum and minimum dimensions of the raschig rings. The last part of the table gives the dimensions of the Cylindrical Tube Model selected for the survey calculations.

5.5.4 Survey Calculations. The survey calculations of k_∞ for various concentrations of fissile solutions of $^{233}UO_2(NO_3)_2$, $^{235}UO_2(NO_3)_2$, and $Pu(NO_3)_4$, the latter having several different isotopic compositions, and four glass volume fractions were performed using the ANISN transport code in the S_4 approximation with the Hansen-Roach 16-group cross sections.[40] These results are given in Table 11 and presented graphically in Figs. 11 through 16 for the various fissile solutions. The volume fraction outside the glass was constant, 0.3354, for all of these calculations.

Table 11. Values of k_∞ from Survey Calculations for Raschig Rings in Fissile Solutions at Various Concentrations and Glass Volume Fractions.[a]

U or Pu Concentration (g/liter)	k_∞					
			^{239}Pu			
	^{233}U	^{235}U	0% ^{240}Pu	5% ^{240}Pu	10% ^{240}Pu	12% ^{240}Pu-12% ^{241}Pu
			Glass Volume Fraction = 0.20			
100	0.7058		0.8749	0.8245	0.7824	0.7989
150	0.9047		1.0176	0.9541	0.9064	0.9311
200	1.0567	0.8214		1.0394	0.9891	1.0237
250		0.9077		1.1009	1.0403	1.0814
300		0.9793		1.1447	1.0754	1.1219
350		1.0385			1.1203	1.1517
400					1.1429	1.1928
450					1.1845	1.2333
500						1.2531
			Glass Volume Fraction = 0.24			
100	0.6372		0.7925			
150	0.8266		0.9318	0.8742	0.8324	0.8545
200	0.9747	0.7397	1.0341	0.9618	0.9151	0.9467
250	1.0863	0.8235		1.0231	0.9696	1.0047
300		0.8946		1.0699	1.0084	1.0518
350		0.9544			1.0526	1.0855
400		1.0055			1.0781	1.1259
450					1.1204	
500					1.1284	1.1900
600						1.2208
			Glass Volume Fraction = 0.28			
150	0.7571		0.8528			
200	0.9007		0.9545	0.8891		0.8748
250	1.0104		1.0166	0.9498	0.9029	0.9380
300	1.1062		1.0751	0.9991	0.9447	0.9851
350		0.8780	1.1225	1.0374	0.9875	1.0215
400		0.9296		1.0687	1.0157	1.0612
450		0.9714		1.1014	1.0583	1.1037
500		1.0103			1.0679	1.1289
600					1.1051	1.1626
			Glass Volume Fraction = 0.32			
200	0.8326		0.8797			
250	0.9396		0.9406	0.8800		0.8713
300	1.0345		0.9999	0.9307	0.8830	0.9205
350	1.1148		1.0484	0.9710	0.9245	0.9593
400	1.1839		1.0897	1.0046	0.9551	0.9985
450		0.9008	1.1137	1.0363	0.9979	1.0413
500		0.9414	1.1452	1.0623	1.0084	1.0684
600		1.0119				

a. See Table 10 for the dimensions selected for the Cylindrical Tube Model.

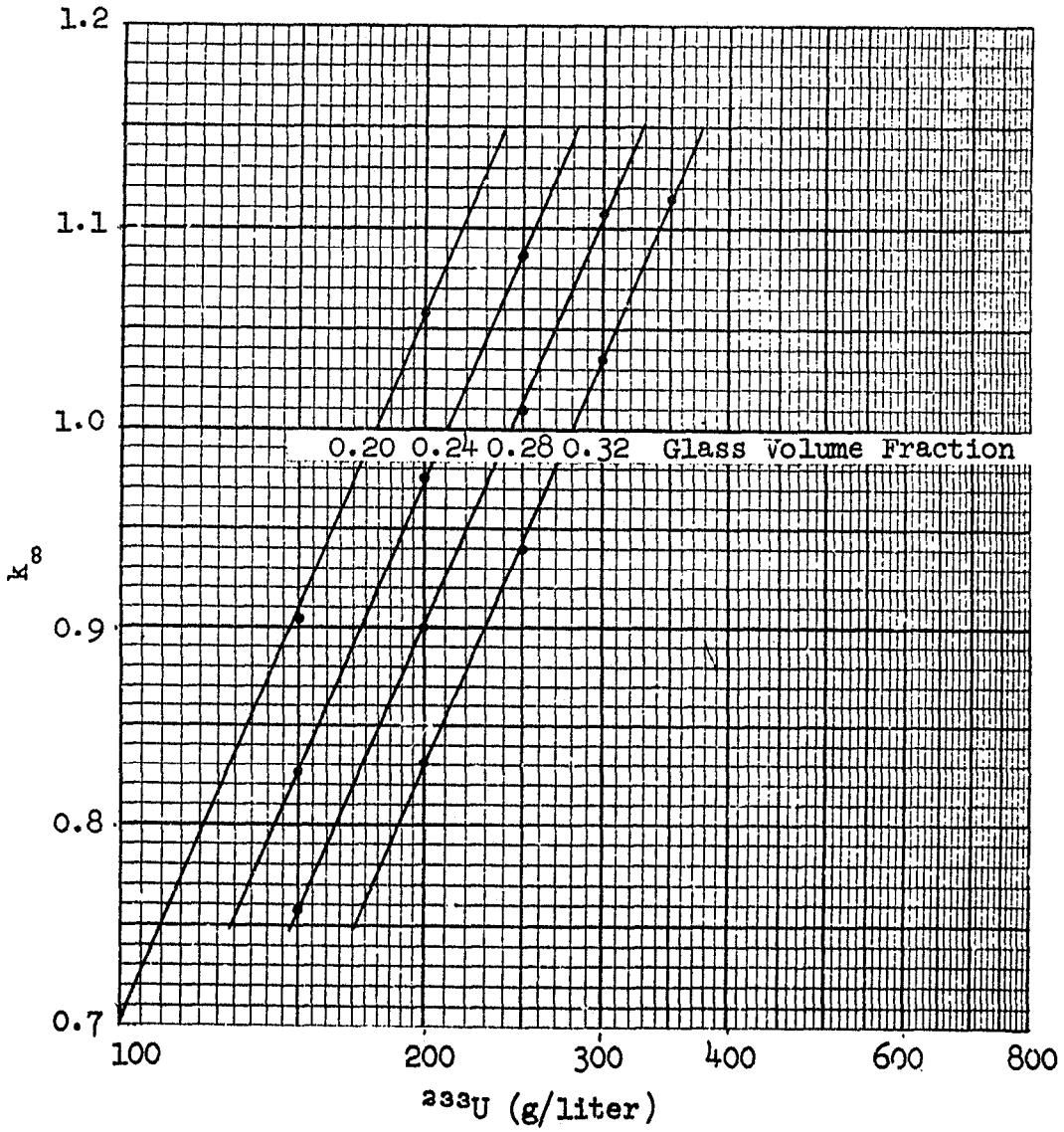

Fig. 11. k_∞ for Raschig Rings in $^{233}UO_2(NO_3)_2$ Solutions for Various Glass Volume Fractions.

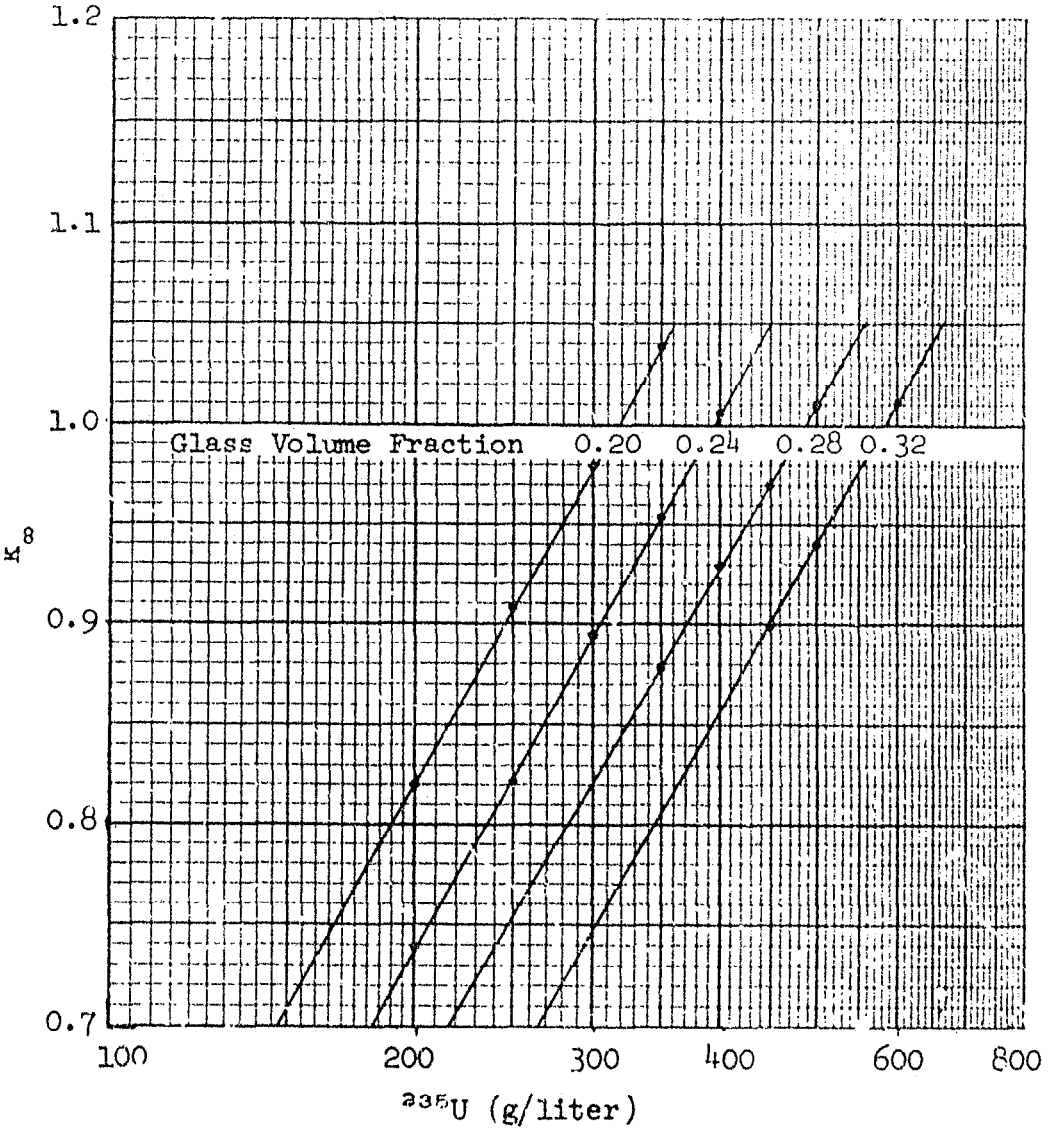

Fig. 12. k_∞ for Raschig Rings in $^{235}UO_2(NO_3)_2$ Solution for Various Glass Volume Fractions.

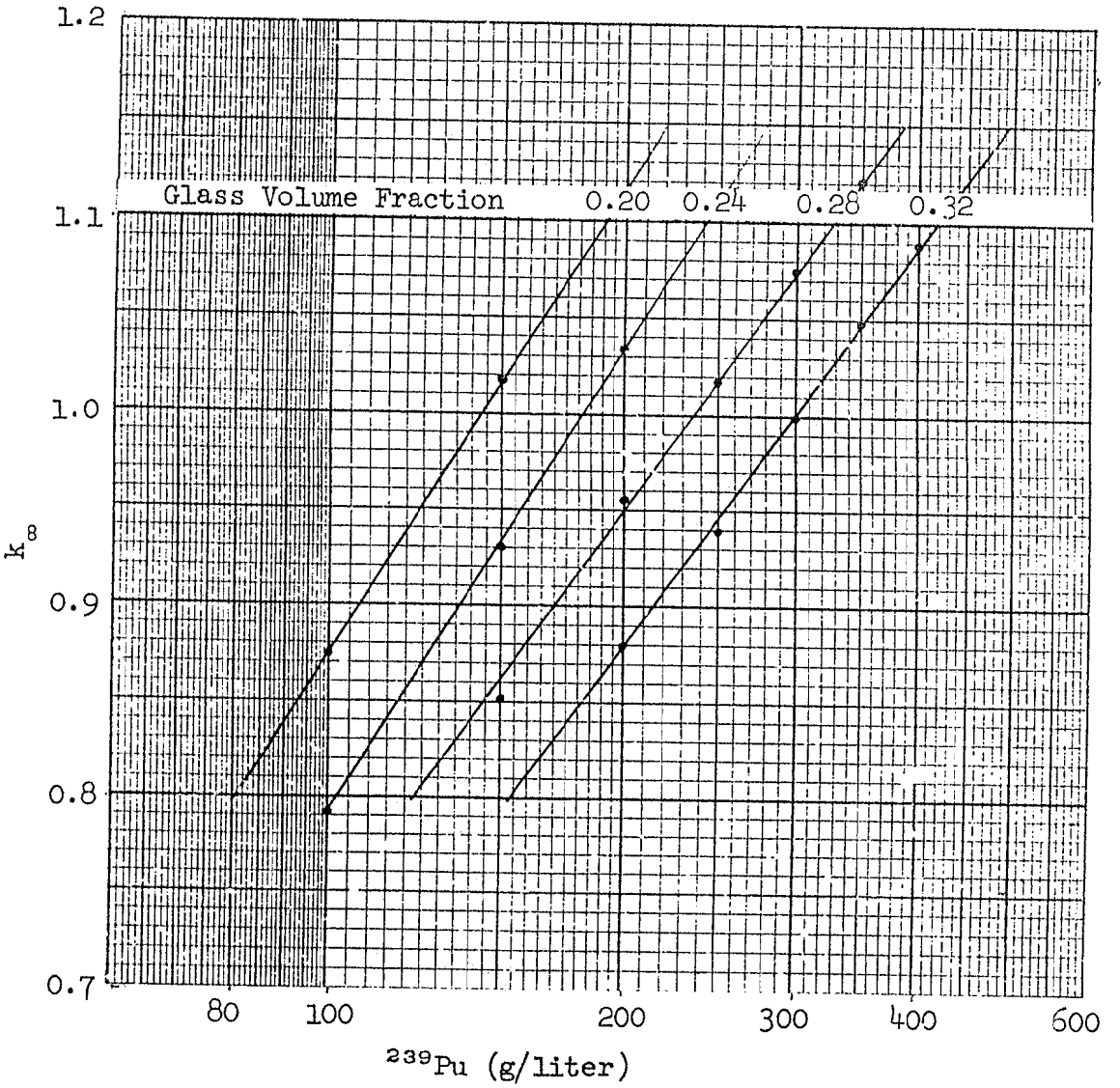

Fig. 13. k_∞ for Raschig Rings in $^{239}Pu(NO_3)_4$ Solutions for Various Glass Volume Fractions.

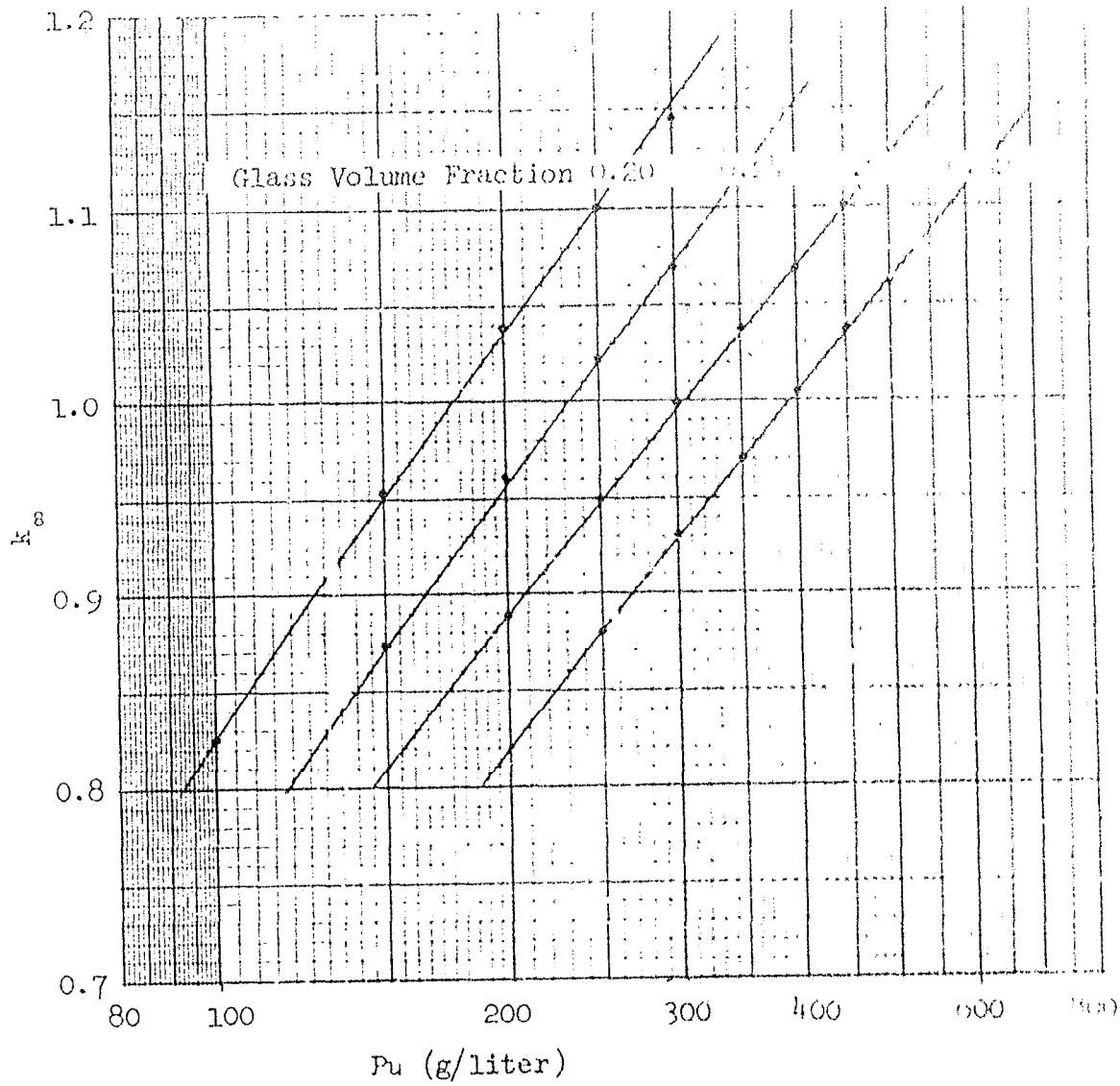

Fig. 14. k_∞ for Raschig Rings in Pu (95% ^{239}Pu, 5% ^{240}Pu) $(NO_3)_4$ Solutions for Various Glass Fractions.

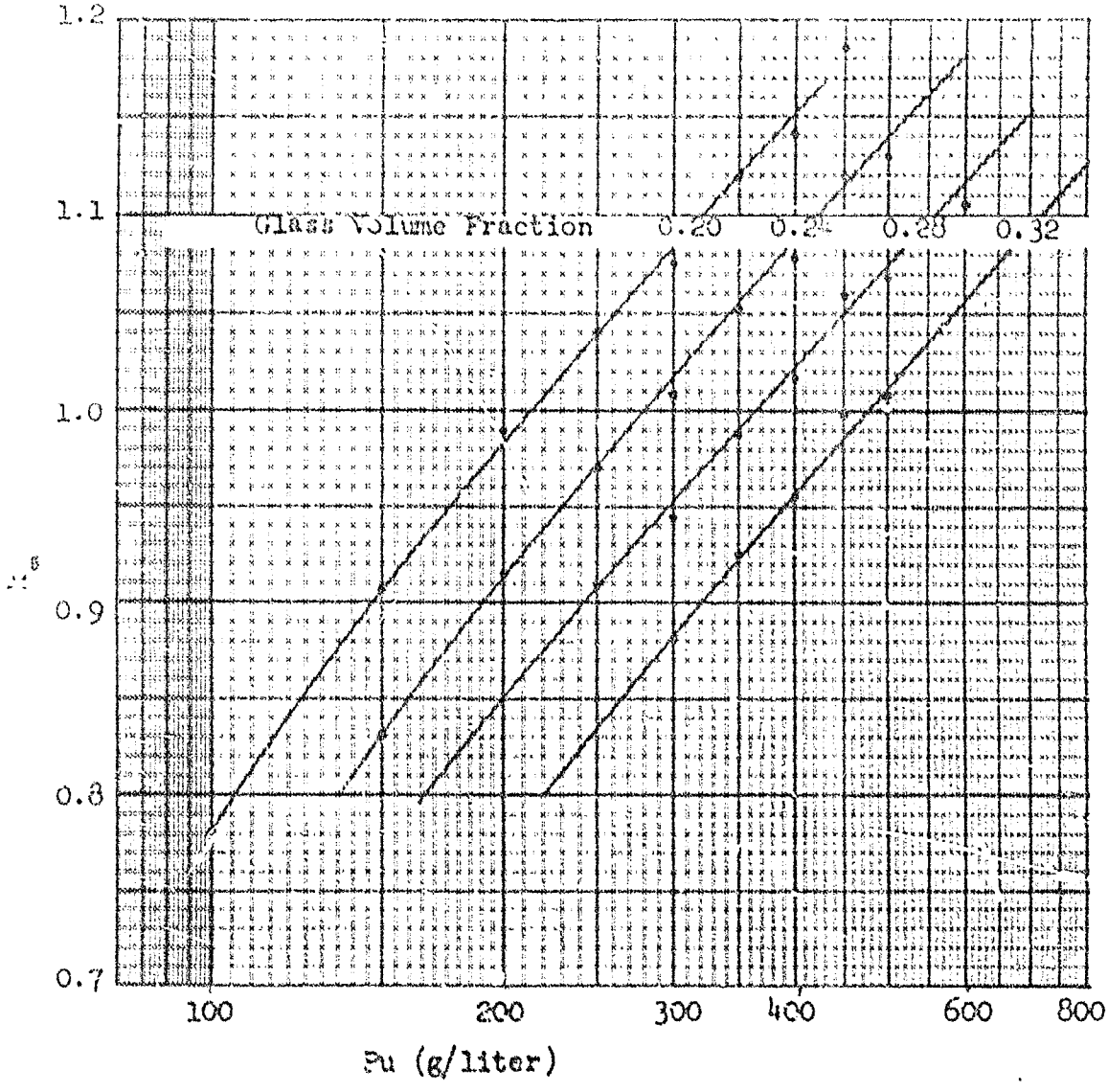

Fig. 15. k_∞ for Raschig Rings in Pu (90% ^{239}Pu, 10% ^{240}Pu) $(NO_3)_4$ Solutions for Various Glass Volume Fractions.

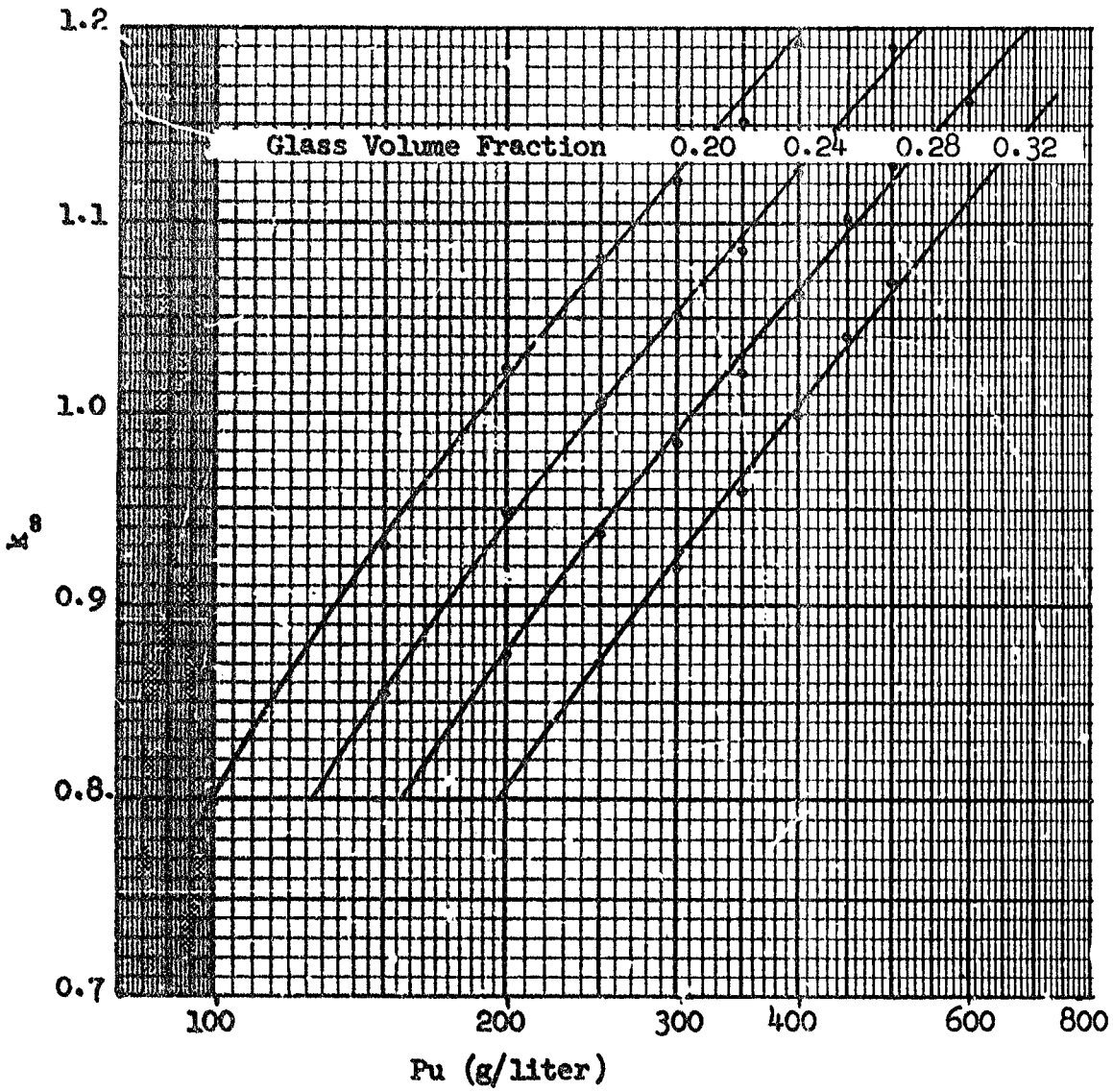

Fig. 16. k_s for Raschig Rings in Pu (76% ^{239}Pu, 12% ^{240}Pu, 12% ^{241}Pu) $(NO_3)_4$ Solution for Various Glass Volume Fractions.

5.5.5 Unpacked Piping in Vessels. It was recommended in Section 4 that raschig rings should not be used for criticality control unless some rings can be removed from the tanks for examination. To to this, rings are placed on stringers inside pipes which contain no rings when the stringer is removed for examination, thereby increasing the reactivity of the storage system. In the proposed standard, pipes up to 2.5 in. in diameter in a square array having 14.5 in. center-to-center spacing (12 in. edge-to-edge spacing) may contain no raschig rings, only solution. A two-region, transport theory calculation in cylindrical geometry was made to evaluate the k_∞ for an infinite array of square cells. A central 2.5-in.-diam cylinder of solution was surrounded by a cylindrical annulus of raschig rings and solution, the latter having a mirror neutron reflector. The area of the entire cylinder was made equal to the square cell area. Cell-averaged homogenized cross sections from the k_∞ calculation for raschig rings and solution were used for the annular region. In Table 12, the k_∞ values for the system with 2.5-in.-diam pipes without rings (Column 4) should be compared to the calculations of k_∞ for solution and rings (Column 3). Reactivity increases from 0.015 to 0.040 were calculated due to the inclusion of unfilled pipes on 14.5-in. centers in the packed infinite vessel.

5.5.6 Neutron Absorption in Nitrogen and Hydrogen Density Variations. It was desirable to know the increase in reactivity if the neutron absorption in the nitrogen were neglected. The nitrogen atom density was set equal to zero for the calculations given in Column 5 in Table 12. By comparison to the entries in Column 3, it is seen that this effect is at most 1% in k_∞. Neutron absorption in excess nitric acid, were its presence included in the calculation, would not be large and it is conservative to neglect it.

Another effect of excess nitric acid in nitrate solutions is the reactivity effect of the reduced hydrogen density. Nitric acid displaces $\sim 30\ cm^3$ of water per mol of acid resulting in a net hydrogen displacement of 0.67 hydrogen atoms per molecule of acid. The hydrogen density in 8.3 M nitric acid solution is 5% less than it is in water.

Table 12. Values of k_∞ from ANISN Calculations of Raschig Rings in Fissile Solutions for Various Conditions.

U or Pu Concentration (g/liter)	Glass Volume Fraction	k_∞ With Volume Fraction Outside Glass = 0.3354[a]	k_∞ With 2.5-in.-diam Pipes Without Rings on 14.5 in. Centers	k_∞ With Nitrogen Density = 0	k_∞ With 0.9 Hydrogen Density	k_∞ With 0.7 Hydrogen Density	k_∞ With Volume Fraction Outside Glass = 0.4000	With 0.001-in.-thick Oxide Layer on Glass: k_∞	With 0.001-in.-thick Oxide Layer on Glass: U or Pu as Oxide (g/liter of rings and solution)
				$^{233}UO_2(NO_3)_2$ Solution					
200	0.24	0.9747	1.0057	0.9768	0.9745	0.9790	0.9849	1.0520	30.0
350	0.32	1.1148	--	1.1179	1.1151	1.1214	1.1285	1.1677	28.7
400	0.32	1.1839	1.2122	--	--	--	--	--	--
				$^{235}UO_2(NO_3)_2$ Solution					
300	0.24	0.8946	0.9238	0.8972	0.8812	0.8556	0.9066	0.9432	30.3
600	0.32	1.0119	1.0386	1.0163	1.0056	1.0000	1.0251	1.0423	29.0
				$^{239}Pu(NO_3)_4$ Solution					
200	0.24	1.0341	1.0583	1.0380	1.0164	0.9760	1.0465	1.1017	32.2
500	0.32	1.1452	1.1692	1.1534	1.1310	1.1038	1.1572	1.1811	30.9
				(95% ^{239}Pu-5% ^{240}Pu)$(NO_3)_4$ Solution					
350	0.24	1.1061	1.1240	1.1124	1.0888	1.0526	1.1156	1.1456	32.2
500	0.32	1.0623	1.0835	1.0698	1.0480	1.0221	1.0732	1.0959	30.9
				(90% ^{239}Pu-10% ^{240}Pu)$(NO_3)_4$ Solution					
500	0.24	1.1284	1.1430	1.1376	1.1147	1.0899	1.1360	1.1562	32.2
600	0.32	1.0493	1.0670	1.0588	1.0396	1.0252	1.0582	1.0768	30.9
				(76% ^{239}Pu-12% ^{240}Pu-12% ^{241}Pu)$(NO_3)_4$ Solution					
600	0.24	1.2208	1.2604	1.2323	1.2125	1.2033	1.2269	1.2468	32.2
600	0.32	1.1051	1.1452	1.1151	1.0973	1.0871	1.1141	1.1357	30.9

a. These data are also given in Table 11 and repeated here for comparison to the other calculations.

Organic solutions may have increased or decreased hydrogen densities. Therefore, the variation of k_∞ with variations in hydrogen density was calculated for hydrogen densities reduced to 90 and 70% of the values used in the reference calculations (Column 3, Table 12) and are given in Columns 6 and 7 of Table 12. Changes in hydrogen density have a small effect on k_∞ and it is noted that Δk is positive for a decrease in hydrogen density for ^{233}U solutions but is negative for the other solutions. The reactivity effect for increases in hydrogen density were extrapolated from these calculations.

5.5.7 Increased Volume Fraction Outside Glass. The survey calculations were made for the volume fraction outside the glass of 0.3354 where k_∞ has a minimum value but within the range observed experimentally (see Table 7). After the k_∞ dependence on the volume fraction outside the glass was established (see Figs. 9 and 10), additional survey calculations were included for a value of this parameter of 0.40, near the maximum observed experimentally. To do this, the Cylindrical Tube Model dimensions were changed. First, with the glass outside radius (R_2) fixed, the solution outside radius (R_3) was increased so that the volume fraction outside the glass was increased to 0.40. This change also changed the glass volume fraction which then was adjusted to its former value by decreasing the glass inside radius (R_1). These dimensions are part of Table 10.

The calculations of k_∞, Column 8, Table 12, using the above dimensions for a volume fraction outside the glass of 0.40, are to be compared to the corresponding k_∞ values for volume fraction outside the glass of 0.3354 (Column 3) and increases in k_∞.

5.5.8 Oxide Layer on Glass. Repeated and continued use of raschig ring-filled vessels may result in a buildup of deposits on the glass surfaces. Therefore, solutions may be added to rings already having some fissile deposits. A 0.001-in.-thick layer of UO_2 or PuO_2, having theoretical density, on the cylindrical glass surfaces in the Cylindrical Tube Model was assumed and 5-region (solution, oxide, glass, oxide,

solution) ANISN calculations were made for selected compositions. This amount of oxide corresponds to ~30 g of uranium or plutonium (see Column 10, Table 12, for the exact amounts) per liter of glass and solution as packed in the vessel. The addition of the oxide increases k_∞ by as much as 0.04 for the limited number of cases which were calculated (compare Column 9 to Column 3 in Table 12).

A comparison of k_∞ for systems with and without oxide deposits was made at constant total U or Pu content. At a glass fraction of 0.24, a solution concentration increase of 1.32 g/liter is equivalent to 1 g of U or Pu as oxide per liter of glass and solution and the oxide volume fraction is 0.00310. At a glass fraction of 0.32, the concentration increase is 1.47 g/liter per gram of U or Pu as oxide and the oxide volume fraction is 0.00305. k_∞ for the cases without oxide having increased solution concentrations (interpolated from Figs. 11 to 16) are larger than k_∞ for cases having the oxide layers for ^{233}U, ^{235}U, and ^{239}Pu. The converse is true for the solutions containing ^{240}Pu. This effect on k_∞ must be related to self shielding in the high density layers of oxide resulting in reduced neutron absorption because all of the ^{240}Pu is no longer homogeneous but some is in a layer on the glass. An allowance of 2% in k_∞ is estimated for this effect; calculations were not made for solution concentrations where k_∞ was less than unity. (See Section 5.6.)

5.6 Recommended Values

The survey calculations were made for 3.57 wt % boron in the glass, equivalent to an atom density of $4.455 \times 10^{21}/cm^3$, the minimum expected from the specifications for the borosilicate glass (Section 2.1). For the safe concentrations, k_∞ less than unity must be selected to include margins for the positive reactivity effects of

a. the maximum value of $[k_\infty(exp) - k_\infty(calc)]$ or bias in the calculations,

b. unpacked piping in vessels,

c. uranium or plutonium deposits on the glass surfaces as UO_2 or PuO_2,

d. increasing the volume fraction outside the glass from 0.33 to 0.40, and

e. a 30% decrease in hydrogen density for ^{233}U solutions or a 10% increase in hydrogen density for ^{235}U or Pu solutions.

These Δk values are given in Table 13 for the various solutions that were used in the survey calculations.

From Figs. 11 through 16, values of the concentrations were obtained for $k_\infty = 0.90$, 0.85, and 0.80 and are listed in Table 14 for various fissile solutions. In Table 15, the values for $k_\infty = 0.85$ are recommended for the standard for glass fractions of 0.24, 0.28, and 0.32 for ^{233}U, ^{235}U, ^{239}Pu with less than 5% ^{240}Pu, and ^{239}Pu with more than 5% ^{240}Pu.

These criticality calculations are also applicable to organic solutions where the hydrogen density in the solvent is neither more than 10% greater nor 30% less than the hydrogen density of the aqueous solution that have been assumed for these calculations. Since the calculated k_∞ values do not vary much with changes in hydrogen density, it is recommended that these limits given in Table 15 be applicable only to systems whose hydrogen content ranges from 75 to 115 g/liter. It is emphasized that the recommended concentration limits for the solutions must be reduced by the amount of the fissile isotope deposits on the glass. Additional calculations have also been made for some assumed conditions of hydrogen density, fissile solution concentration, and fissile deposit. These are given in Table 16 and were chosen to represent some "worst" conditions. It is believed that these calculations confirm the earlier estimates of the reactivity perturbations and that the recommended limits have adequate margins of safety for the conditions described in this report.

Table 13. Reactivity Effects not Included in the Survey Calculations.

			^{239}Pu			
	^{233}U	^{235}U	0% ^{240}Pu	5% ^{240}Pu	10% ^{240}Pu	12% ^{240}Pu and 12% ^{241}Pu
Maximum value of $[k_\infty(exp)-k_\infty(calc)]$	0.047	(0.020)[a]	(0.030)	(0.040)	(0.050)	(0.050)
Calculated Δk for 2.5-in.-diam pipe without rings on 14.5 in. centers	0.031	0.032	0.024	0.021	0.018	0.040
Maximum Δk estimated for 30 g/liter of uranium or plutonium as oxide instead of solution	--	--	--	0.02	0.02	0.02
Maximum value of calculated Δk for 30% decrease in H density	0.007	--	--	--	--	--
Maximum value of calculated Δk for 10% increase in H density	--	0.013	0.018	0.018	0.012	0.008
Calculated Δk for 0.400 volume fraction outside of glass	0.014	0.015	0.012	0.011	0.011	0.009
TOTAL	0.099	0.090	0.084	0.110	0.000	0.127

a. Numbers in parentheses are estimated and may be uncertain by 100%.

Table 14. Fissile Solution Concentrations for Various k_∞ Values and Glass Volume Fractions.

Glass Volume Fraction	Calculated Uranium or Plutonium Solution Concentration (g/liter)					
			^{239}Pu			
	^{233}U	^{235}U	100% ^{239}Pu	5% ^{240}Pu	10% ^{240}Pu	12% ^{240}Pu and 12% ^{241}Pu
			$k_\infty = 0.90$			
0.20	147	245	107	127	146	133
0.24	173	305	136	165	190	172
0.28	200	371	170	209	246	218
0.32	230	449	214	269	320	275
			$k_\infty = 0.85$			
0.20	134	215	93	107	123	114
0.24	156	268	117	139	150	147
0.28	180	323	144	173	200	183
0.32	209	393	181	223	264	231
			$k_\infty = 0.80$			
0.20	121	189	80	92	105	100
0.24	142	235	102	118	135	127
0.28	163	282	122	145	165	156
0.32	188	344	153	185	220	195

a. These values of the concentrations are interpolated values from Figs. 11-16.

Table 15. Recommended Fissile Solution Concentration Limits for 1.5-in.-o.d. Raschig Ring Filled Storage Vessels.

Glass Volume Fraction	Recommended Uranium or Plutonium Concentration Limit[a] (g/liter) for			
			^{239}Pu	
	^{233}U	^{235}U	< 5% ^{240}Pu	> 5% ^{240}Pu
> 0.24	150	270	115	140
> 0.28	180	330	140	170
> 0.32	200	400	180	220

a. Including fissile deposits on glass.

Table 16. Calculated k_∞ Values Using the Cylindrical Tube Model, Five Regions for the Recommended Concentration Limits and Assumed Conditions of Maximum Reactivity.[a]

Glass Volume Fraction	Recommended Concentration Limit (g/liter)	Assumed Concentrations		Calculated k_∞	
		Solution[b] (g/liter)	Oxide (g/liter)	Without Pipes	With 2.5-in.-diam Pipes Without Rings on 14.5 in. Centers
		^{233}U Solutions			
0.24	150	120	30.0	0.8272	0.8527
0.28	180	150	29.3	0.8545	0.8824
0.32	200	170	28.7	0.8448	0.8748
		^{235}U Solutions			
0.24	270	240	30.3	0.9048	0.9349
0.28	330	300	29.6	0.9128	0.9450
0.32	400	370	29.0	0.9216	0.9555
		^{239}Pu Solutions with < 5% ^{240}Pu			
0.24	115	85	32.2	0.8767	0.9037
0.28	140	110	31.5	0.8868	0.9164
0.32	180	150	30.9	0.9167	0.9484
		^{239}Pu Solutions with > 5% ^{240}Pu			
0.24	140	110	32.2	0.9112	0.9359
0.28	170	140	31.5	0.9113	0.9379
0.32	220	190	30.9	0.9311	0.9586

a. The ANISN-S_8 transport code with Hansen-Roach 16-group cross sections was used for these calculations. The volume fraction (of solution) outside the glass was assumed to be 0.40, and Cylindrical Tube Model dimensions for these calculations are taken from Table 10. The oxide layer on the glass tube was assumed to be 0.001 in. thick. The boron density in the glass was 4.455×10^{21} atom/cm^3. The hydrogen density was 120 g/liter except for the ^{233}U cases where a value of 70 g/liter was assumed.

b. All the calculations were made with the solution concentrations reduced by 30 g/liter from the recommended limits. Since the total solution volume fraction (inside and outside the glass) was 76, 72, and 68% for the three glass fractions, the appropriate concentration reduction corresponding to 30 g of fissile material on the glass would be 39.5, 41.7, and 44.1 g/liter, respectively. Hence, the calculations were made for higher average fissile concentrations than the recommended limits and are conservative for nuclear safety by the differences in the concentrations.

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