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**CALCULATED NEUTRON MULTIPLICATION FACTORS OF UNIFORM
AQUEOUS SOLUTIONS OF ^{233}U AND ^{235}U**

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CALCULATED NEUTRON MULTIPLICATION FACTORS OF UNIFORM AQUEOUS SOLUTIONS OF ^{233}U AND ^{235}U

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TABLE OF CONTENTS

ABSTRACT	1
I. INTRODUCTION	3
II. COMPUTATIONS AND RESULTS	4
III. ACCURACY OF THE RESULTS	6
IV. CONCLUSIONS	8
V. ACKNOWLEDGEMENTS	8

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ABSTRACT

Computations of the effective neutron multiplication factor of single units of aqueous solutions of $^{233}\text{UO}_2\text{F}_2$ and $^{235}\text{UO}_2\text{F}_2$ are reported for guidance in the specification of limits applicable to processes, such as storage and transport, for these fissile isotopes. Graphs are presented of k_{eff} as a function of such parameters as the mass of fissile material, the chemical concentration, the dimensions of spheres and infinitely long cylinders, and the thickness and areal density of infinite slabs. Transport theory (DTF) codes in the S_n approximation with Hansen-Roach cross sections were utilized and the results agree with relevant experiments to within 0.01 in k_{eff} .

I. INTRODUCTION

In 1958 a Standard, entitled "Safety Standard for Operations with Fissionable Materials Outside Reactors," was drafted by a subcommittee of the Standards Committee of the American Nuclear Society (ANS) and Sectional Committee No. 6 of the American Standards Association. In 1964 this standard was designated an American Standard, ASA No. 1-1964^a.

Increased basic knowledge and operating experience have made desirable a revision of ASA No. 1-1964 which would extend it in various ways and which would include, in particular, the specification of limits on single process variables for guidance of nuclear criticality safety. Such a revision is now being prepared by Subcommittee 8 of the Standards Committee of the ANS.

The computations reported here were made to provide background information as an aid to the specification of the limits for uniform aqueous solutions of ^{233}U and ^{235}U . Limits are established for the spherical mass and volume, the concentration of the solution, the diameter of an infinitely long cylinder, and the thickness as well as areal density of a slab infinite in two dimensions.

In each case the value of k_{eff} was calculated for various assumed values of one parameter with all others optimized. In all cases the uranium is isotopically pure ^{233}U or ^{235}U in an aqueous solution of UO_2F_2 surrounded by an infinitely thick water reflector. The shape is taken as spherical in the calculations for masses and volumes. In all cases the chemical concentration is taken as that giving greatest reactivity except of course where k_{eff} vs concentration for an infinite volume of solution is being investigated.

a. This standard is now designated as USA Standard No. 1-1964.

II. COMPUTATIONS AND RESULTS

All computations were made by the S_n method, as applied by the DTF code,¹ using the Hansen-Roach 16-group cross sections.²

For spherical and slab shapes the angular segmentation (n) was taken as six; for cylindrical shapes, n was taken as eight. A few test cases indicated that a finer quadrature (greater value of n) would not change the results significantly.

A spatial mesh of 0.2 cm was used for the thin slabs and 0.5 to 1.0 cm for the regions and geometries of larger dimension. A smaller mesh was shown to have no significant effect on the results.

With respect to the energy mesh of the Hansen-Roach 16-group scheme, a finer mesh was investigated by replacing the first five groups (energy $> 10^5$ eV) with 17 groups and examining the fast leakage from the core. As with the angle and space variables, the discrete treatment of energy corresponding to 16 groups was shown to cause no significant error.

It was concluded that the method, the angular quadrature, and the spatial mesh as described gives essentially an "exact" solution to the transport equation for the problems considered here. In other words, whatever errors there are in the values of k_{eff} are due to uncertainties in the cross sections and not to the method. The magnitude of these errors is discussed in Section III.

The procedure leading to the establishment of minimum values of the various parameters at a selected value of k_{eff} is illustrated in the following discussion. In the determination of a recommended mass, k_{eff} of spherical volumes of solutions at three chemical concentrations were calculated as a function of sphere radius. The results for ^{235}U are shown in Fig. 1a(235). The accompanying curves, Fig. 1b(235), show the same results plotted as k_{eff} as a function of the mass of ^{235}U in the various spheres. The latter results were cross plotted in Fig. 1c(235)

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1. B. G. Carlson et al., "DTF Users Manual," UNC Phys/Math-3321, Vol. I (Nov. 1963), Vol. II (May 1964).
 2. G. E. Hansen and W. H. Roach, "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos Scientific Laboratory (1961).

as k_{eff} vs ^{235}U concentration at constant mass for several masses. The maxima of these curves were plotted, in turn, in Fig. I(235) as maximum values of k_{eff} as a function of mass. The resulting relation which turns out to be roughly linear gives the largest value of k_{eff} attainable with a given mass of ^{235}U in aqueous solution under optimum conditions, that is, in a fully water-reflected sphere at the most nuclearly reactive chemical concentration. This relation shows that about 820 g of ^{235}U in $\text{U}(100)\text{O}_2\text{F}_2$ solution^b will be critical and that about 710 g of ^{235}U will have a k_{eff} of 0.97.

In a like manner the maximum value of k_{eff} attainable with various spherical volumes of solution of $\text{U}(100)\text{O}_2\text{F}_2$, in a narrow range around the smallest possible critical volume, is obtained from the three curves of k_{eff} as a function of concentration ($\text{H}/^{235}\text{U}$ ratio) in Fig. 2(235) and the plot of the maxima of these curves in Fig. II(235). These results give 6.1 liters as the minimum critical volume.

The variation of k_{eff} with the concentration of solution in volumes infinite in all dimensions, given in Fig. III(235), shows a ^{235}U concentration of 0.0118 g/cc as that of the most dilute solution which can be made critical.

Similarly the finite dimensions of cylindrical and slab-shaped geometries with infinite transverse dimensions and the areal density (mass of ^{235}U per unit area) of an infinite slab corresponding to various maximum values of k_{eff} were obtained, respectively, from Figs. 4(235), and IV(235), from Figs. 5(235) and V(235), and from Figs. 6a, b, and c(235) and VI(235).

An identical series of operations, reported in a similarly numbered series of curves, has established corresponding limiting dimensions for units of ^{233}U solution.

The results are summarized in Table 1 where the critical dimensions and the dimensions corresponding to an arbitrarily selected subcritical condition ($k_{\text{eff}} = 0.97$) are recorded.

b. $\text{U}(100)$ signifies uranium enriched to 100% in the ^{235}U isotope.

Table 1. Dimensions of Single Units of Aqueous Solutions
of ^{235}U and ^{233}U

All parameters optimized.

Parameter	k_{eff}	^{235}U		^{233}U	
		1.00	0.97	1.00	0.97
Mass (g)		820	710	570	510
Volume (liters)		6.1	5.4	3.7	3.3
Isotopic Solution Concentration (g/l)		11.8	11.1	11.2	10.6
Diameter of Infinite Cylinder (cm)		14.3	13.6	11.9	11.3
Thickness of Infinite Slab (cm)		4.9	4.4	3.2	2.8
Areal Isotopic Concentration (g/cm ²)		0.42	0.39	0.37	0.34

III. ACCURACY OF THE RESULTS

To evaluate the accuracy of the computational procedure, a number of critical experiments was analyzed in order to compare the computed values of k_{eff} with the experimental value (unity). Since the configurations of interest and the version of the S_n code used were both one-dimensional, comparison was made with critical experiments of a one-dimensional or close to one-dimensional shape. It was possible to select among the many experiments that have been done over the years at the Oak Ridge Critical Experiments Facility a group for both ^{233}U and ^{235}U solutions which cover a broad range of concentrations.

Table 2 presents the results of this comparison of theory and experiment. It is concluded that, overall, the computed values of k_{eff} are accurate to about $\pm 1\%$; there is a bias of about $+ 0.5\%$ in those for the ^{233}U solutions.

Table 2. Comparison of Computed Values of the Effective Neutron Multiplication Factor for Aqueous Solution of UO_2F_2 with Experiment.

All units were surrounded by an effectively infinite water reflector except as noted.

Shape	Dimensions ^a (cm)	Hydrogen Atoms Fissile Atoms	Computed k_{eff}	Experiment Reference
²³³ U				
Cylinder	Diam = 12.7 Height = 56.5	.74.1	1.005	3 (p. 15)
Sphere	26.4	378.1	1.007	4
Sphere	32.0	663.1	1.009	4
Sphere ^c	69.2	1533.0	1.000	5
²³⁵ U				
Thin slab ^e	1.995 in. x 47.20 in. x 58 in.	44.7	1.005	5
Sphere	23.0	76.1	0.995	7 (p. 42)
Sphere	23.6	126.5	0.988	7 (p. 42)
Sphere	26.4	239.3	0.994	4
Sphere	32.0	515.1	1.006	4
Sphere	55.8	1270.0	1.008	7 (p. 42)

- a. Diameter unless otherwise specified.
 b. The uranium contained about 98% ²³³U.
 c. This was an unreflected solution of $\text{UO}_2(\text{NO}_3)_2$.
 d. The uranium contained about 93.2% ²³⁵U.
 e. The container was 0.8775 in. thick methacrylate plastic surrounded by effectively infinitely thick water.

3. J. K. Fox et al., "Critical Mass Studies, Part VIII, Aqueous Solutions of ²³³U," ORNL-2143, Oak Ridge National Laboratory (Sept. 1959).
 4. J. T. Thomas et al., "A Direct Comparison of Some Nuclear Properties of U-233 and U-235," ORNL-1992, Oak Ridge National Laboratory (Nov. 1955); Nucl. Sci. Eng. 1, 20 (1956).
 5. R. Gwin and D. W. Magnuson, Nucl. Sci. Eng. 12, 364 (1962).
 6. J. K. Fox et al., Nucl. Sci. Eng. 3, 694 (1958).
 7. J. K. Fox et al., "Neutron Physics Division Annual Progress Report for the Period Ending Sept. 1, 1958," ORNL-2609, p. 42, Oak Ridge National Laboratory (1958).

IV. CONCLUSIONS

The calculated values of the dimensions of single units of aqueous solutions of ^{235}U and ^{233}U under optimum reactivity conditions afford establishment of specifications for chemical and other processes whereby nuclear criticality may be avoided. The uncertainty in the calculation of k_{eff} , shown by comparison with experiment to be no more than 0.01, should be considered when specifying the dimension. In the example shown in Table 1, a safety factor of 0.02 was first stipulated as an arbitrary condition; to it was added 0.01 to cover the uncertainty in the calculation, hence the selection of the dimensions corresponding to $k_{\text{eff}} = 0.97$. The span of values of k_{eff} given in the graphs, however, permit selection of dimensions corresponding to other degrees of subcriticality to conform to particular operating conditions. They may be easily obtained from the graphs designated by Roman numerals. The bias apparent in the ^{233}U calculations adds a bit of additional conservatism.

V. ACKNOWLEDGEMENTS

The author gratefully acknowledges the consulting help of John Knight and Elliot Whitesides of the Computer Sciences Center with respect to the computations; and of J. T. Thomas, Dale Magnuson, and E. B. Johnson at the Oak Ridge Critical Experiments Facility with respect to individual experiments.

²³⁵U Results

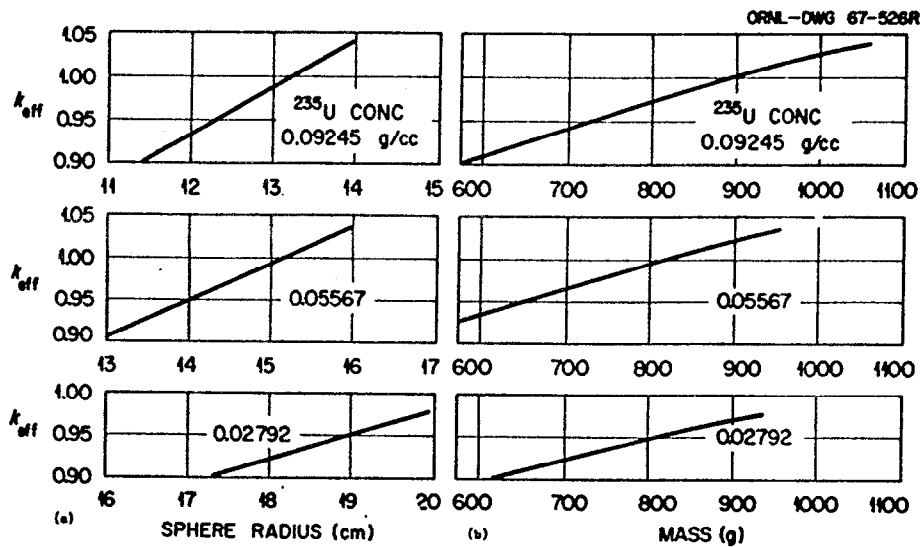


Fig. 1a(235). k_{eff} vs Radius of Sphere for Solutions of Three ^{235}U Concentrations.

Fig. 1b(235). k_{eff} vs Mass of ^{235}U in Spheres for Solutions of Three Concentrations.

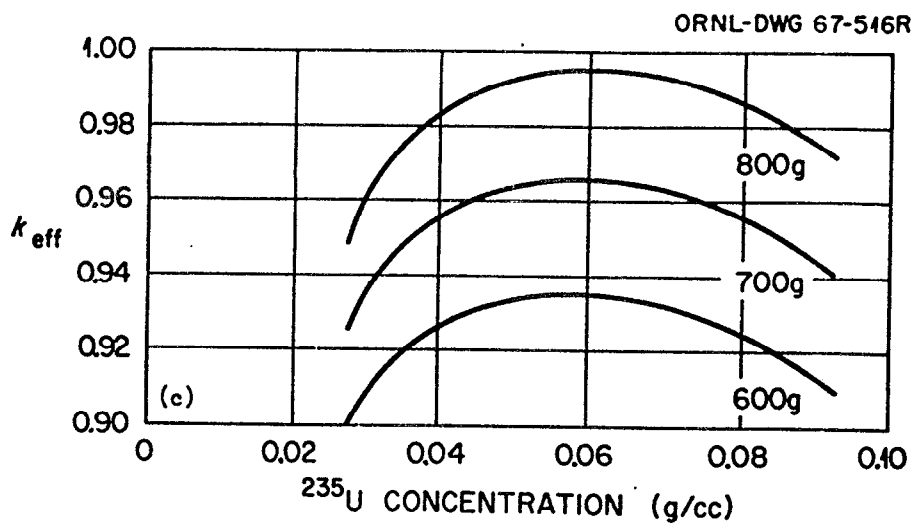


Fig. 1c(235). k_{eff} vs Concentration for Various Masses of ^{235}U .

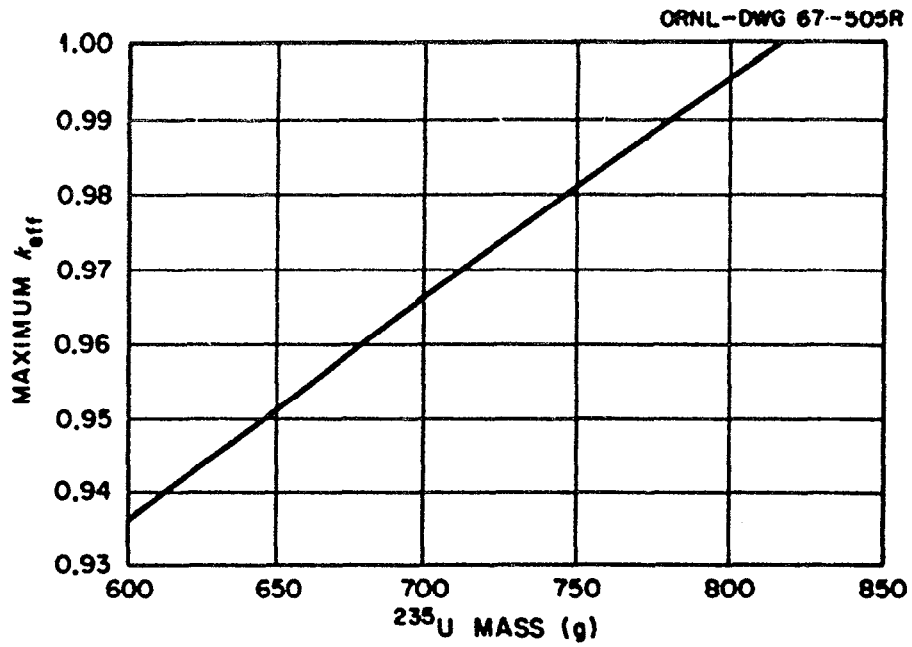


Fig. I(235). Maximum k_{eff} vs Mass of ^{235}U .

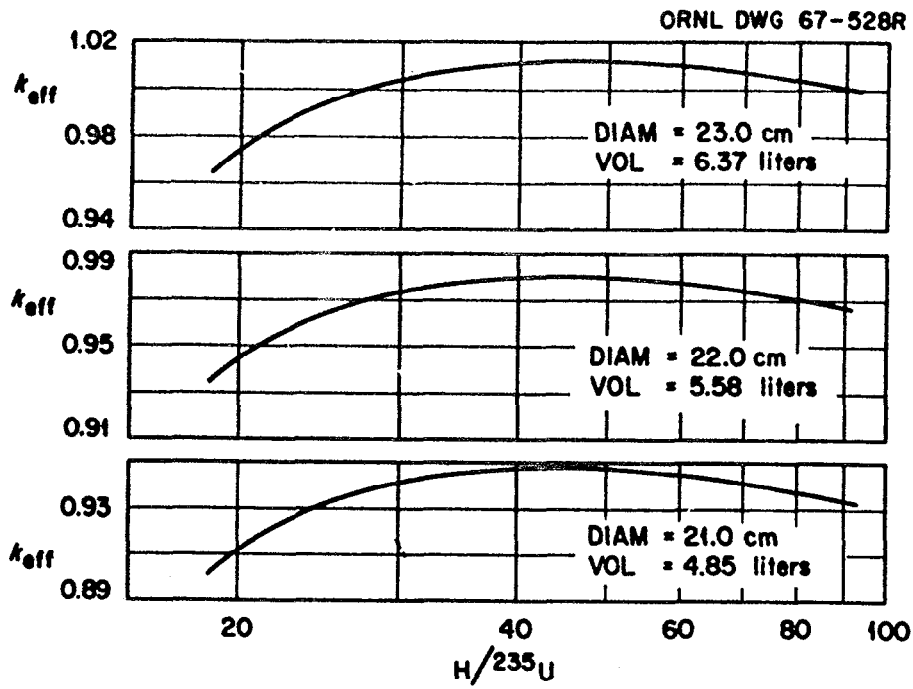


Fig. 2(235). k_{eff} vs $\text{H}/^{235}\text{U}$ for Three Spherical Volumes.

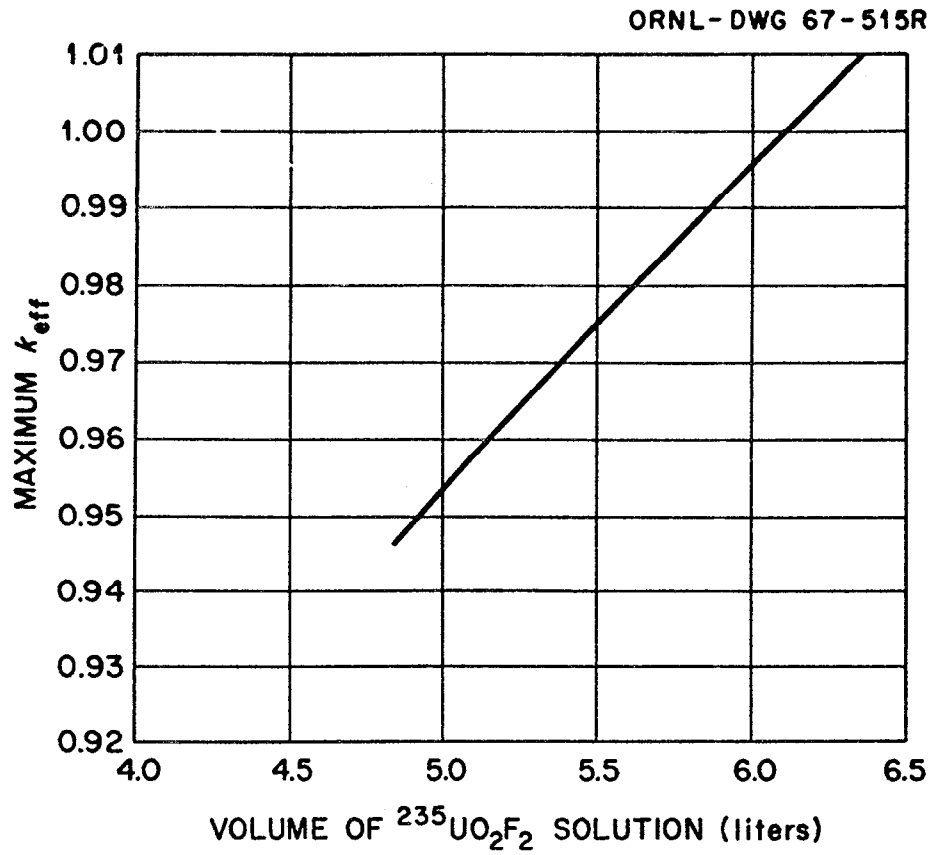


Fig. II(235). Maximum k_{eff} vs Spherical Volume of ^{235}U .

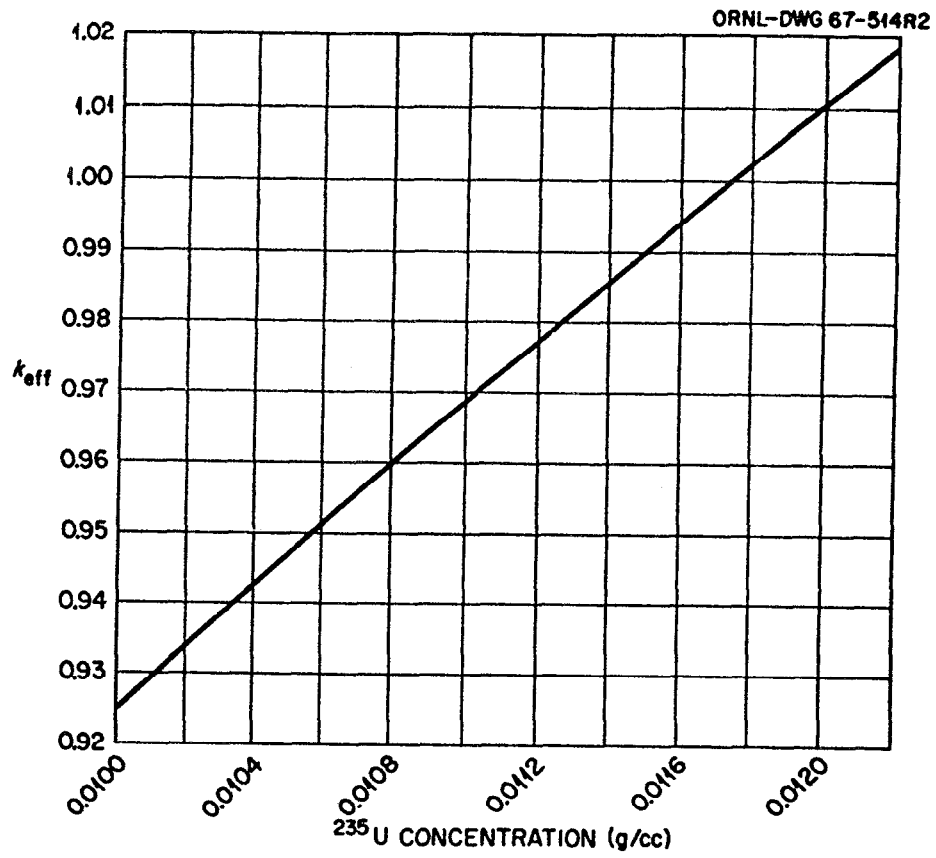


Fig. III(235). k_{eff} vs Concentration of ^{235}U .

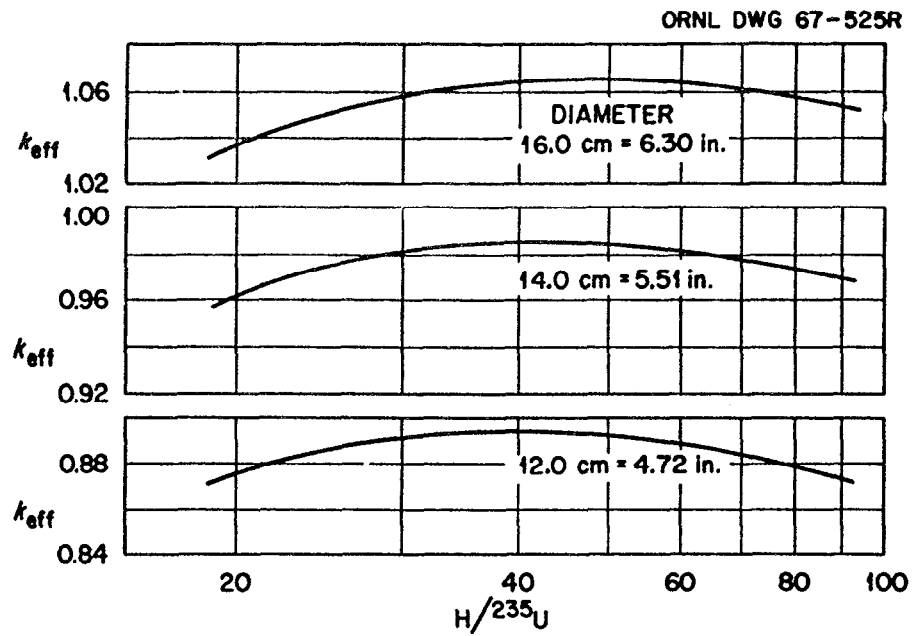


Fig. 4(235). k_{eff} vs $H/^{235}\text{U}$ for Cylinders of Three Diameters.

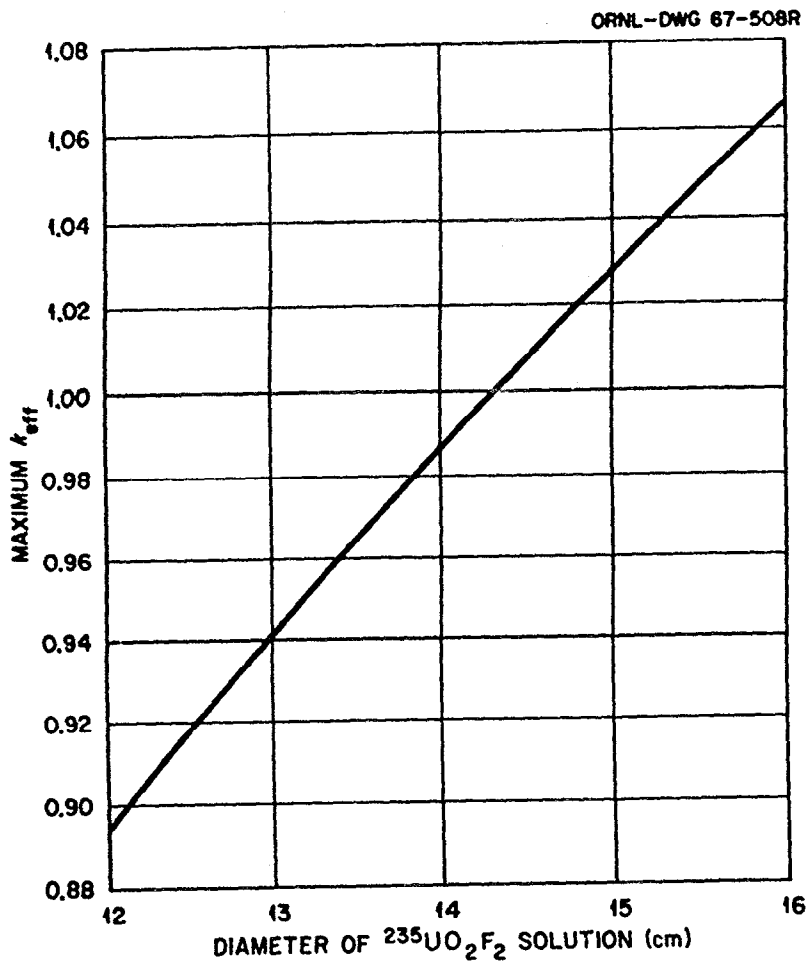


Fig. IV(235). Maximum k_{eff} vs ^{235}U Cylinder Diameter.

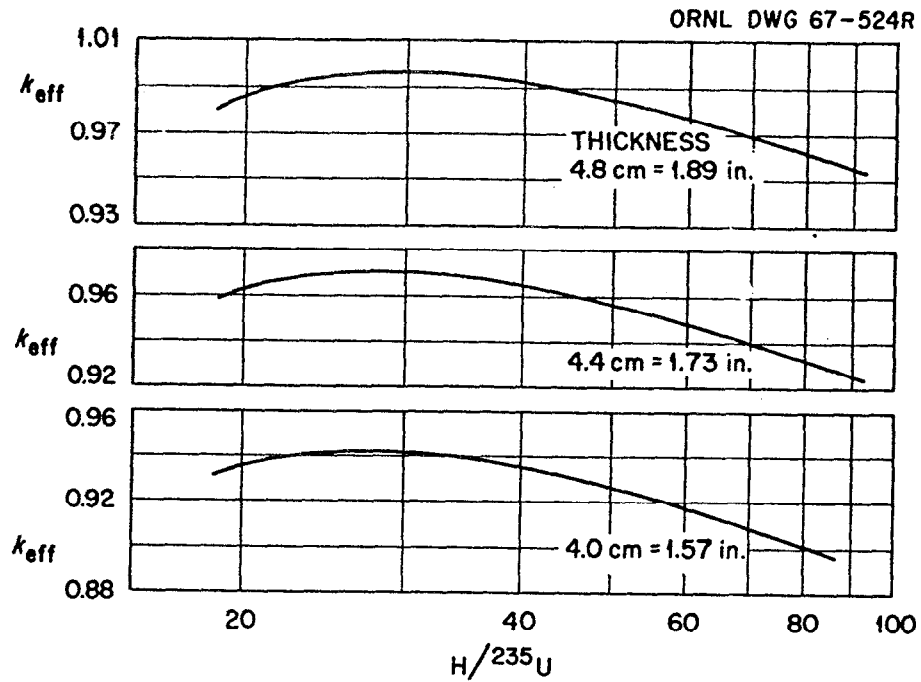


Fig. 5(235). k_{eff} vs $H/^{235}\text{U}$ for Infinite Slabs of Three Thicknesses.

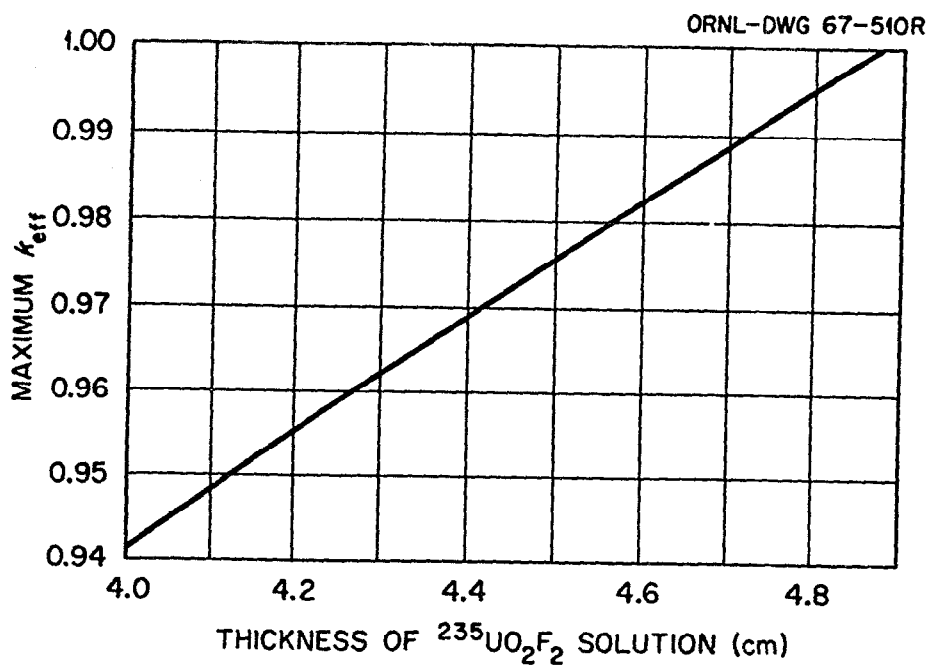


Fig. V(235). Maximum k_{eff} vs Infinite ^{235}U Slab Thickness.

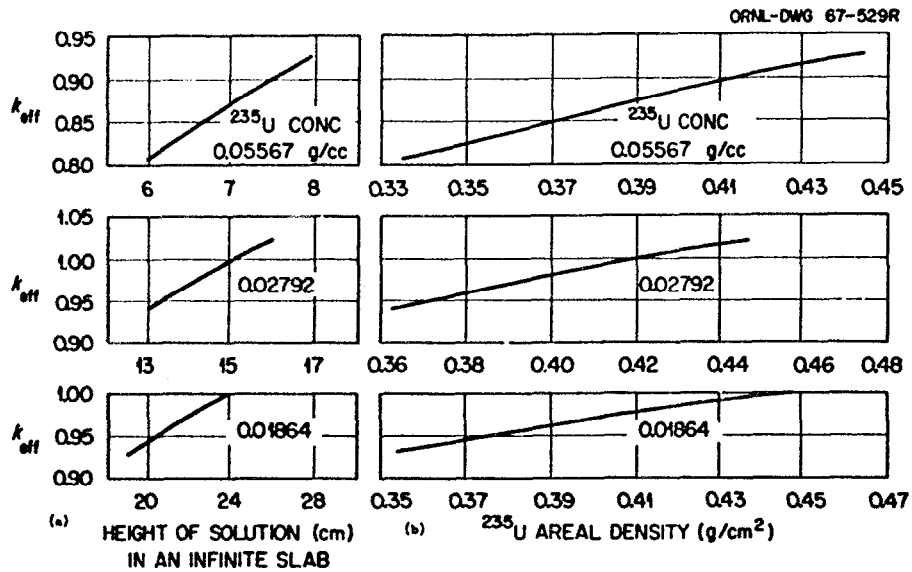


Fig. 6a(235). k_{eff} vs Height of ^{235}U Solution in Infinite Slabs for Solutions of Three Concentrations.

Fig. 6b(235). k_{eff} vs Density of ^{235}U per Unit Area of Infinite Slabs for Three Concentrations.

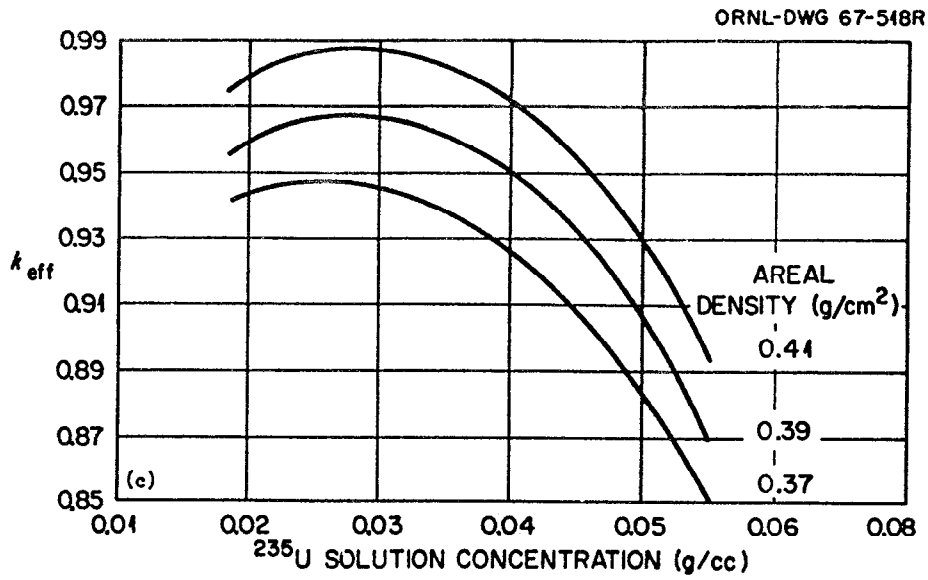


Fig. 6c(235). k_{eff} vs Concentration of ^{235}U for Various Densities per Unit Area.

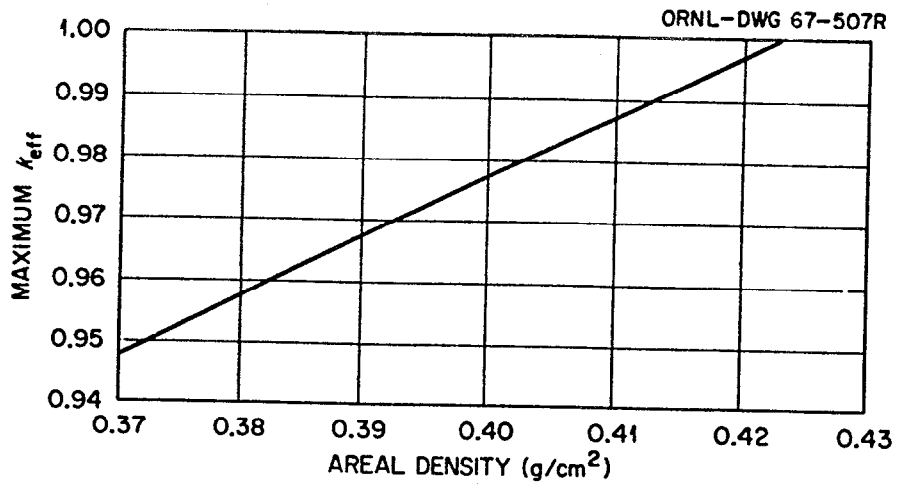


Fig. VI(235). Maximum k_{eff} vs ^{235}U Mass per Unit Area.

²³³U Results

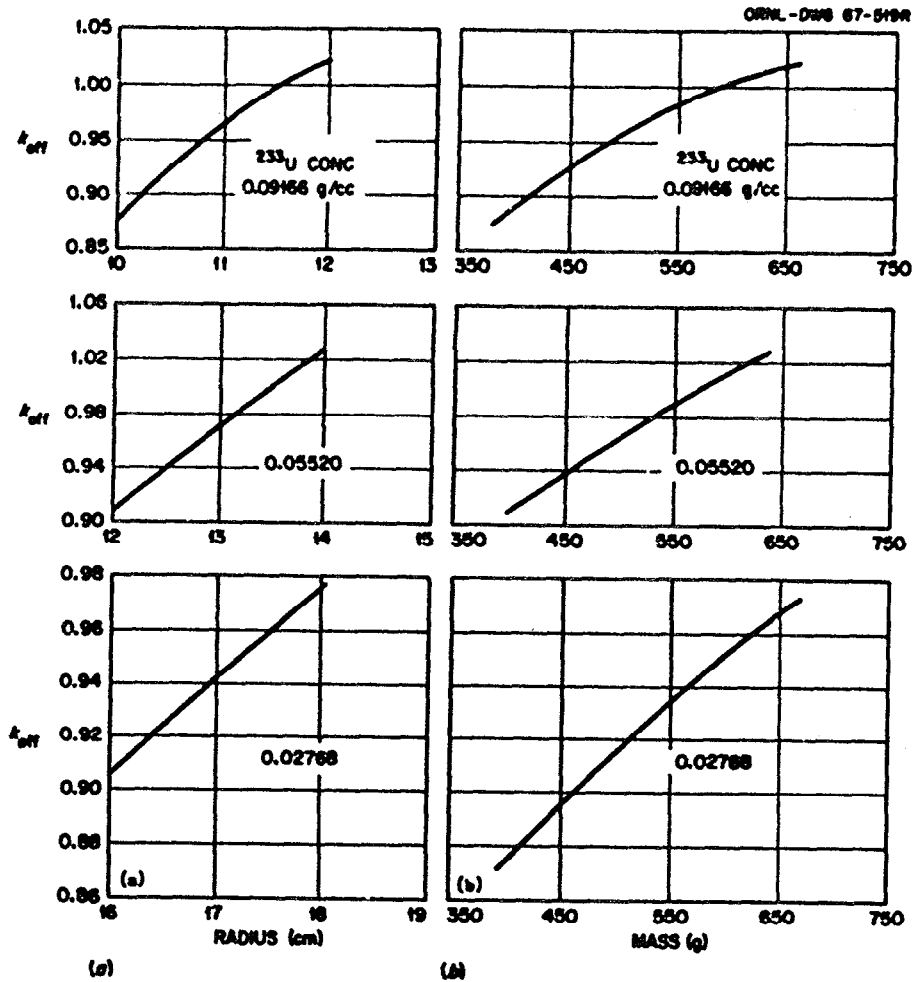


Fig. 1a(233). k_{eff} vs Radius of Spheres for Solutions of Three ^{233}U Concentrations.

Fig. 1b(233). k_{eff} vs Mass of ^{233}U in Sphere for Solutions of Three Concentrations.

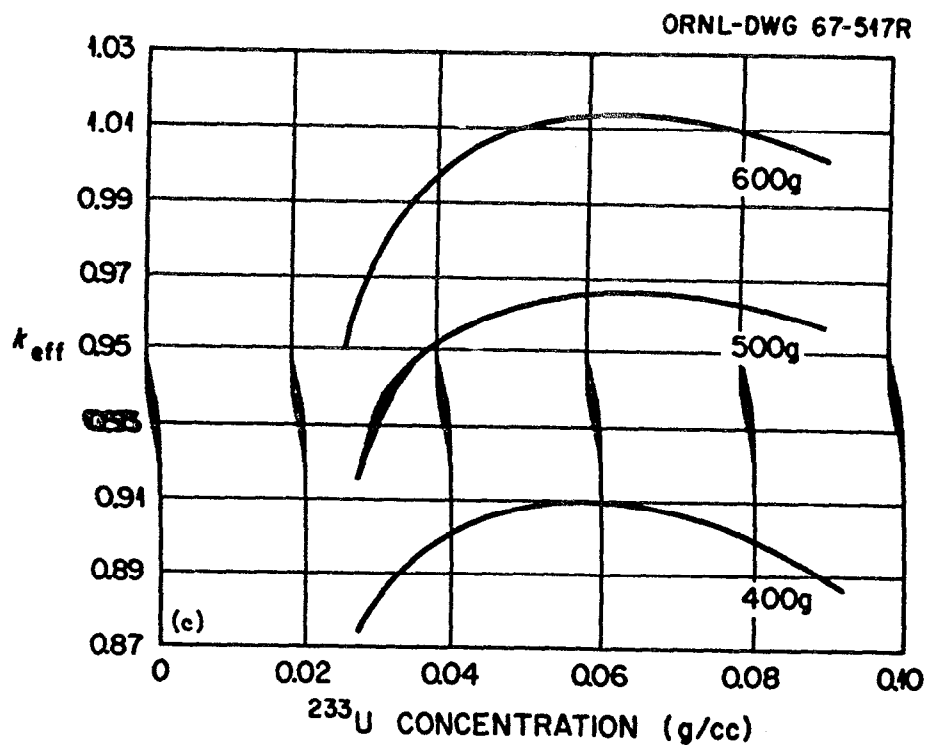


Fig. 1c(233). k_{eff} vs Concentration for Various Masses of ^{233}U .

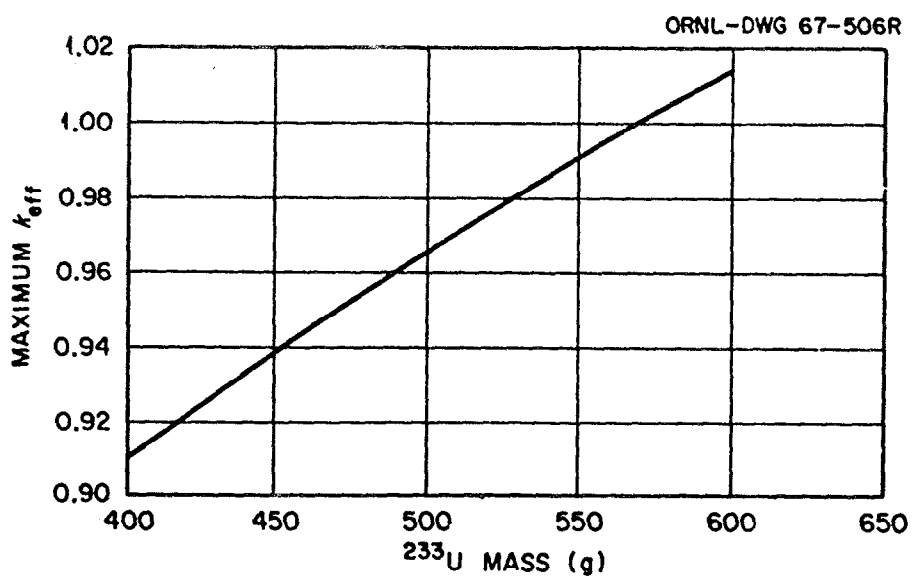


Fig. I(233). Maximum k_{eff} vs Mass of ^{233}U .

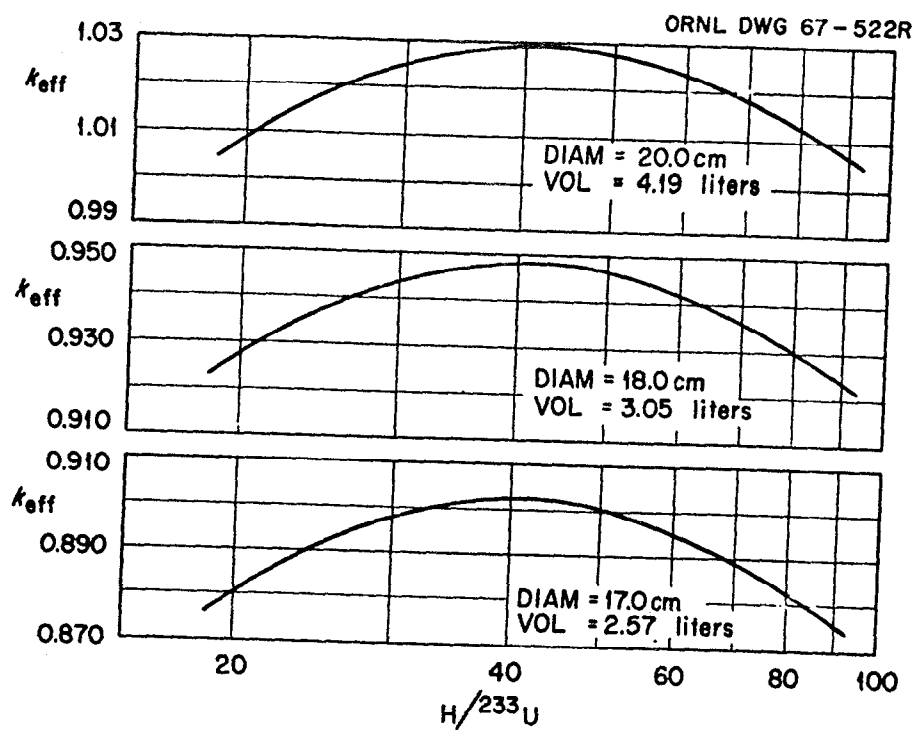


Fig. 2(233). k_{eff} vs $H/^{233}\text{U}$ for Three Spherical Volumes.

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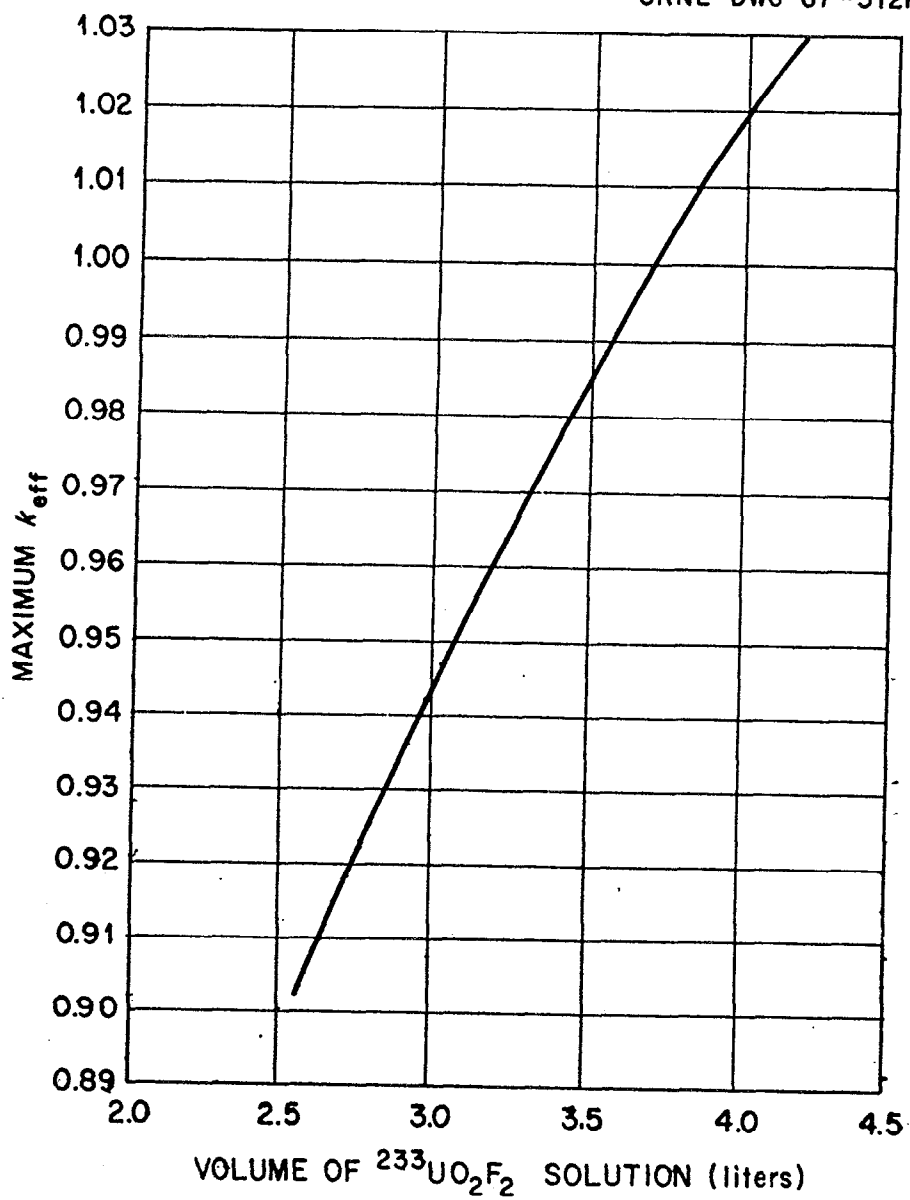


Fig. II(233). Maximum k_{eff} vs Spherical Volume of ^{233}U .

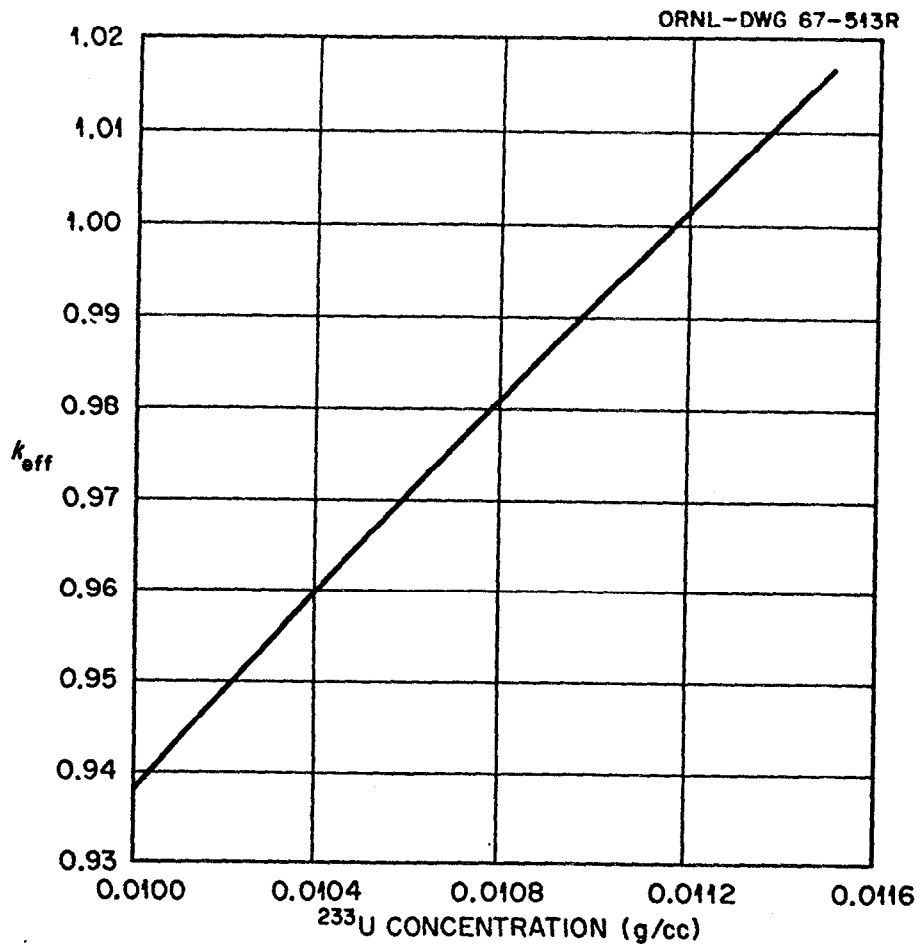


Fig. III(233). k_{eff} vs Concentration of ^{233}U .

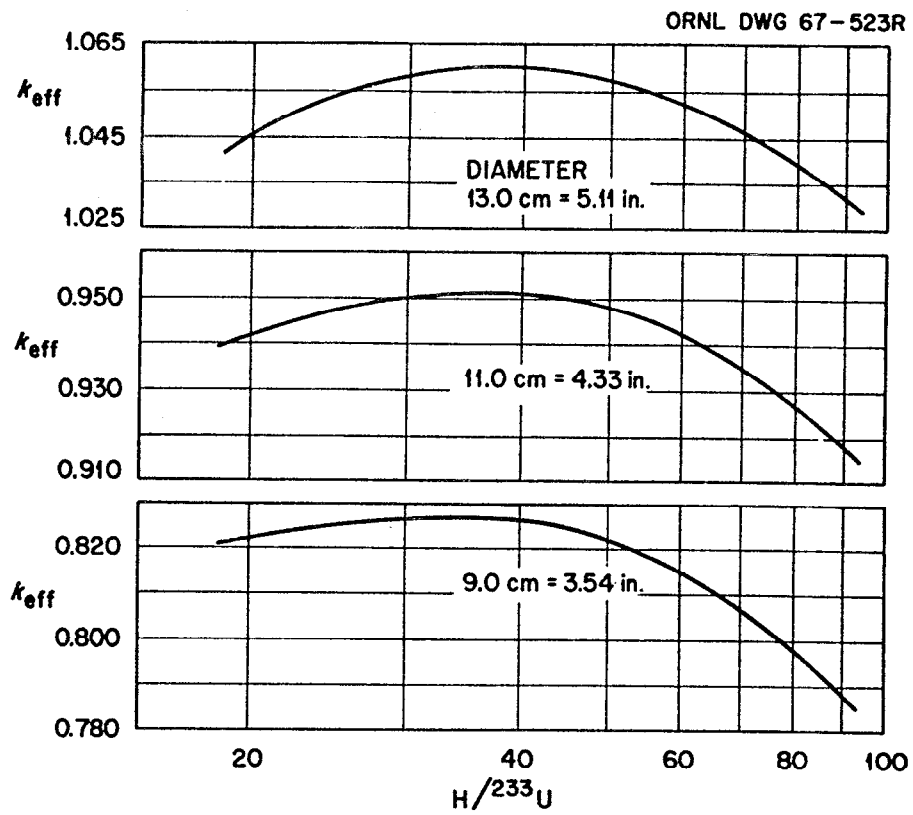


Fig. 4(233). k_{eff} vs $H/^{233}\text{U}$ for Cylinder of Three Diameters.

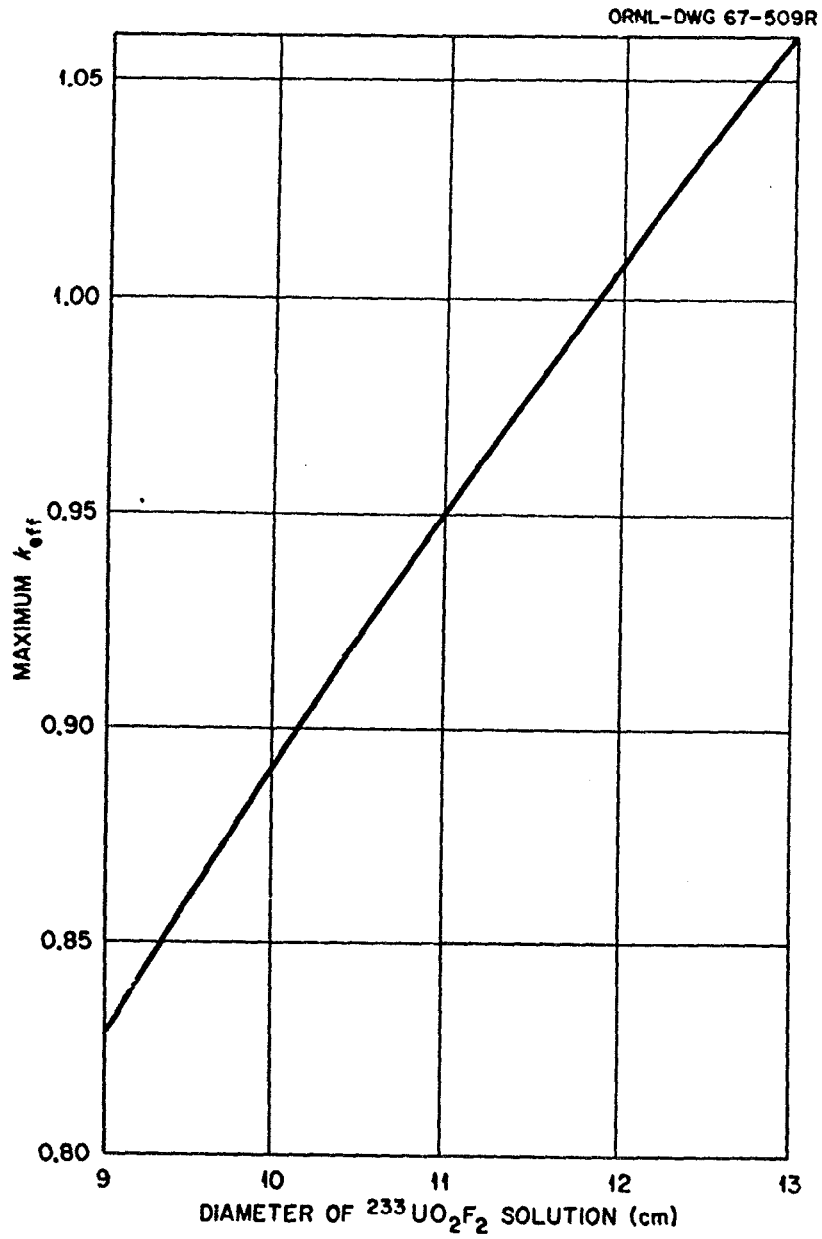


Fig. IV(233). Maximum k_{eff} vs ^{233}U Cylinder Diameter.

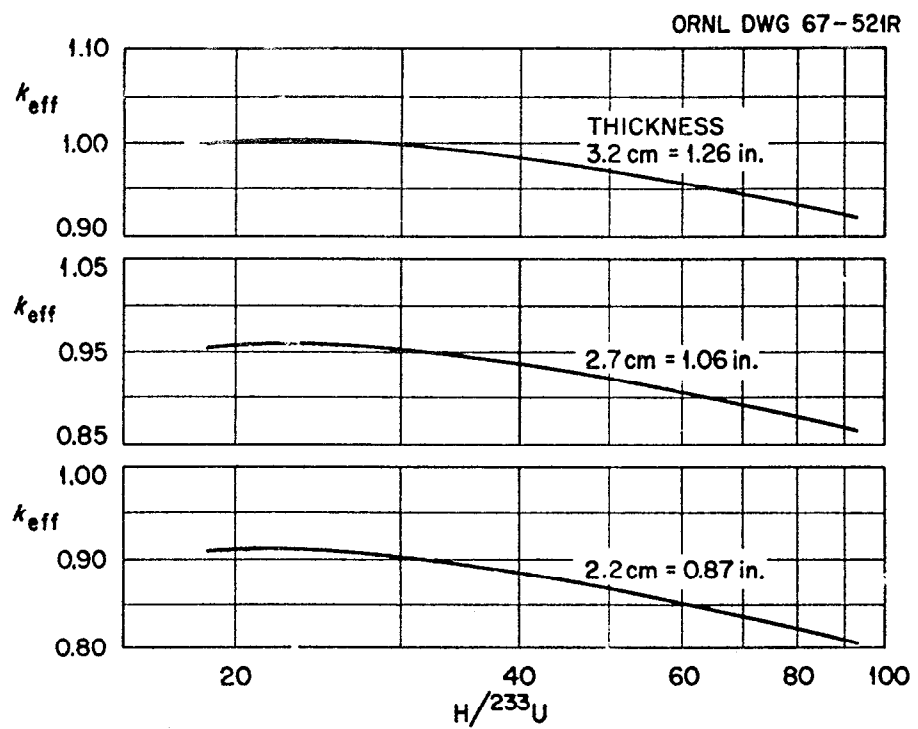


Fig. 5(233). k_{eff} vs H/λ_{233U} for Infinite Slabs of Three Thicknesses.

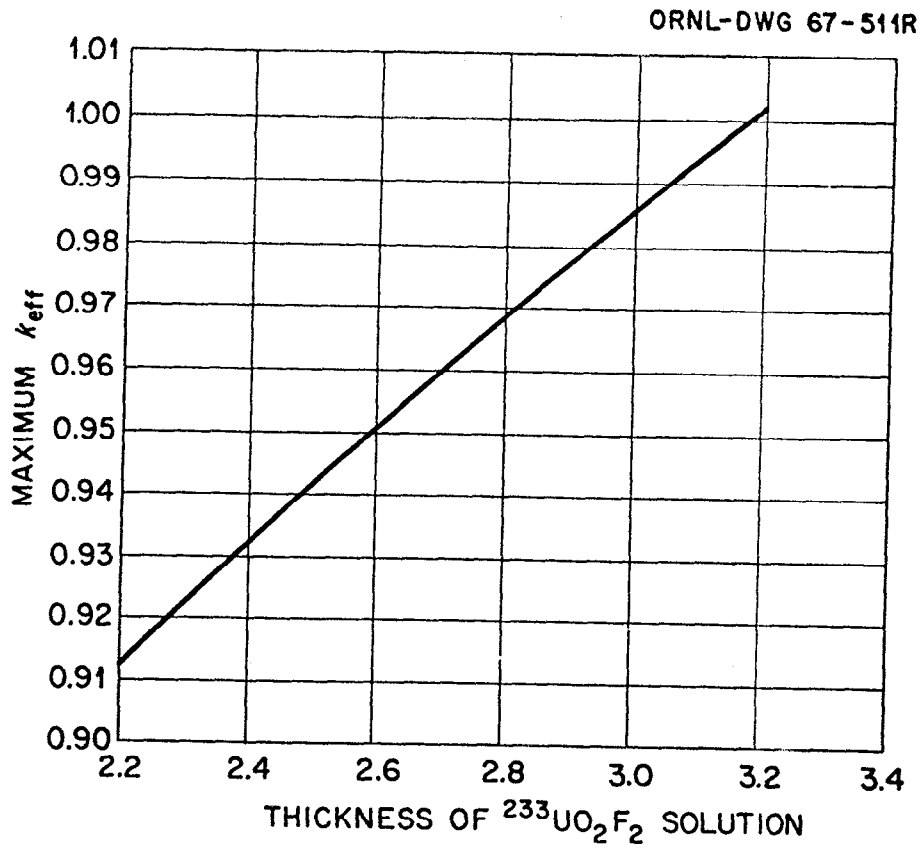


Fig. V(233). Maximum k_{eff} vs Infinite ^{233}U Slab Thickness.

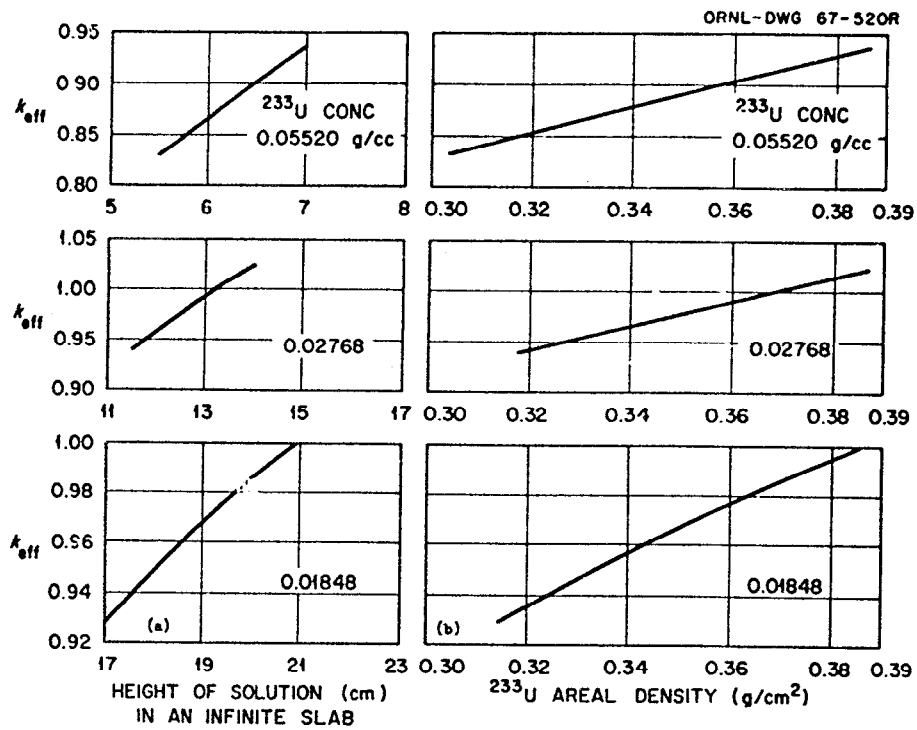


Fig. 6a(233). k_{eff} vs Height of ^{233}U Solution in Infinite Slabs for Solutions of Three Concentrations.

Fig. 6b(233). k_{eff} vs Density of ^{233}U per Unit Area of Infinite Slabs for Solutions of Three Concentrations.

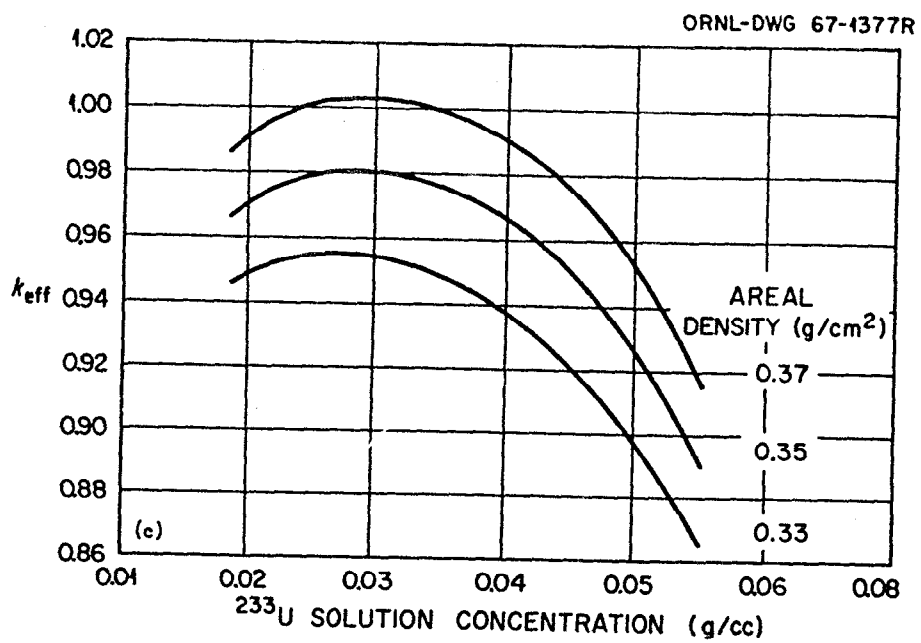


Fig. 6c(233). k_{eff} vs Concentration of ^{233}U for Various Densities per Unit Area.

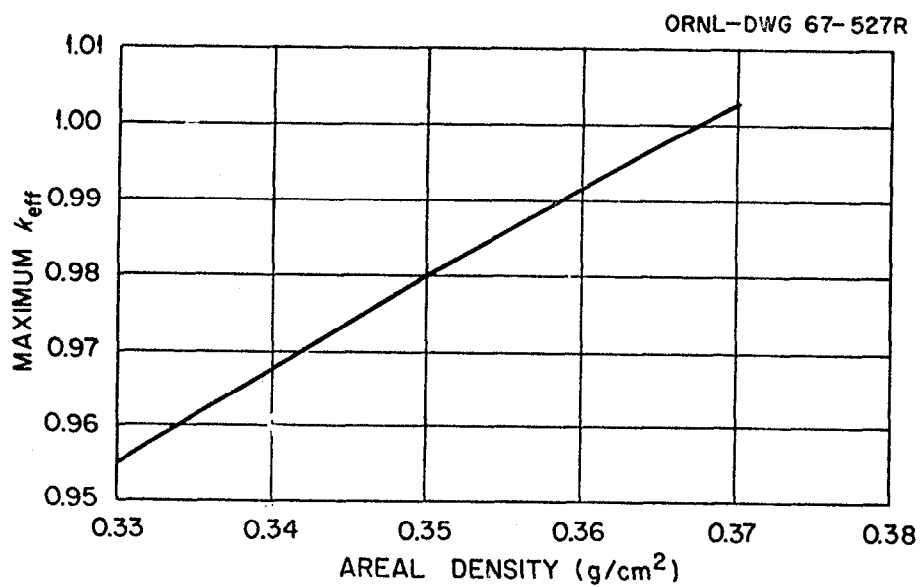


Fig. VI(233). Maximum k_{eff} vs ^{233}U Mass per Unit Area.