X-426A (Revised 1-52)

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ORNL
CENTRAL FILES NUMBER
54-5-170

DATE: May 20, 1954

COPY NO. 28

SUBJECT: Criticality of U-235-H_2O-D_2O Systems In Cylindrical Geometry

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CRITICALITY OF U^{235}-H_2O-D_2O SYSTEMS IN CYLINDRICAL GEOMETRY

Summary

The problem of safely storing large quantities of enriched fuel solution in the HRT dump tanks has motivated criticality studies of long right circular cylinders where the moderator is H_2O, D_2O, or various mixtures of the two. Since the tank may be flooded on the outside during maintenance work, the pipes are considered to have an infinite H_2O reflector. Also, the effect of coating the pipes with a poison for thermal neutrons, say cadmium, has also been considered. The linear concentration in Kg U^{235} per linear foot has been adopted here as a useful unit of concentration. This has the advantage of remaining constant in a horizontal pipe for a given charge of uranium, independent of the quantity of solvent.

The results indicate that for any mixtures of H_2O and D_2O, there is a diameter for which the linear concentration is a minimum. As the mole fraction of D_2O in the moderator is increased, the minimum linear concentration decreases and occurs at larger diameters. These minima are summarized in Table I.

Table I. Minimum Linear Concentration of U^{235}
Infinite H_2O Reflector, 20°C

Mole Fraction H_2O in Solvent	No Cadmium Diameter Inches	No Cadmium Conc. of U^{235} Kg/ft	Cadmium Coated Diameter Inches	Cadmium Coated Conc. of U^{235} Kg/ft
1	9.5	0.65	12	0.82
.5	13.5	.58	17	.75
.25	18	.52	22	.73
.125	21	.47	29	.68
0	24*	.38*	32*	.62*

* This is the value for the largest diameter calculated here; the minimum was not reached.

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Since criticality experiments with H_2O moderator and no cadmium indicated a minimum in linear concentration 10 percent lower, the results obtained here by means of the two-group diffusion model are apparently not conservative. The problem should therefore be treated again by more rigorous methods based on a multi-group model, and it is also recommended that a program of criticality measurements be started as a check on these calculations. The problem will become more urgent in the design of large scale homogeneous reactors and associated chemical plants.

Method of Calculation

Since the systems gain reactivity with decreasing temperature, the calculations were performed for 20°C, the lowest temperature contemplated for the dump tanks. The only method found suitable for calculating the criticality of the two-region configuration over the required range of pipe diameters and mole ratios of H_2O to D_2O is that based on two-group diffusion theory. Although this model implies that the neutron slowing down function is a Yukawa, the actual function and the second moment for mixtures of H_2O and D_2O has never been measured. In fact, it is known that the slowing down in pure D_2O is best represented by a convolution of a Gaussian and a Yukawa.

The second moments of the synthetic Yukawa slowing down kernel for the mixtures were derived from multi-group calculations of the critical buckling of bare spheres.[1,2] To minimize the computational labor, the Feynman-Welton approximation[3,4] to the two-group method was used as shown in Appendix A.

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(1) RM-842, G. Safanov

(2) ORNL 54-1-54, H. C. Claiborne, T. B. Fowler, M. Tobias, "Critical Mass Calculations For a Proposed Rebuilt HRE", January 4, 1954

(3) LA-524, R. Feynman and T. A. Welton

(4) ORNL 53-5-119, M. C. Edlund, "A Two-Group Two-Region Approximation For Blanket Breeder Reactors", May 19, 1953

Two-Group Constants

The constants for the moderator and reflector are summarized in the following table.

Table II. Two-Group Constants for Moderator and Reflector at 20°C

Mole Fraction H_2O	Macroscopic Slow Absorp. Cross-Section cm^{-1} $X10^{-2}$	Slow Diffusion Constant, cm	Square of Diffusion Length cm^2	Fast Diffusion Constant, cm	Yukawa Age cm^2
0	0.00806	0.843	10460	1.25	161
.125	.2513	.550	218.8	1.24	100
.25	.495	.408	82.4	1.22	73
.50	.986	.269	27.3	1.19	49
1.0	1.956	.160	8.18	1.14	33.8

The necessary constants for U^{235} were obtained from BNL-170B.

Results and Discussion

The critical linear concentration of U^{235} for right circular cylinders of infinite length reflected by H_2O is plotted in Figure 1 as a function of diameter. In Figure 2 are similar plots for the case of the pipe coated with a black poison for thermal neutrons.

The reliability of the calculations can only be assessed for the pure H_2O moderator with no cadmium by comparison with experimental results and with calculations by the variational method using four slowing down groups.(5) This comparison is shown in Figure 3 where the experimental points represent extrapolations(6) to infinite length of data for finite cylinders(7) with a

(5) LA-399, Eugene Greuling, "Theory of Water-Tamped Water Boiler", September 27, 1945

(6) K-905, R. L. Macklin, "Cylindrical Reactor Dimensions", May 9, 1952

(7) K-343, C. K. Beck, A. D. Callihan, J. W. Morfitt, R. L. Murray, "Critical Mass Studies, Part III", April 19, 1949

wall of 1/16" stainless steel. For the smaller pipe diameters, the method of calculation used here tends to be non-conservative; the minimum in the linear concentration being 10 percent lower than calculated. This discrepancy is probably due to two causes: first, as the diameter decreases, the calculated neutron leakage from the pipe becomes more sensitive to the assumed slowing down characteristics of the medium; second, the approximations made in the calculation of the probability of absorption in the core of a neutron which is thermalized in the reflector also become more important with decreasing diameter.

One would expect that the reliability would improve for the pipes with cadmium coating since there is no return to the core of neutrons thermalized in the reflector; and furthermore, criticality is reached in large diameter pipes, minimizing the importance of the slowing down model. Thus, there is a considerable advantage in the use of the coating on the pipe arising not only from the higher fuel storage permitted, but the increased confidence in the calculations as well.

The envelopes to the curves in Figures 1 and 2 define the minimum linear concentration for criticality for each pipe size. If the presence of H_2O and D_2O in any ratio is possible, then the envelopes dictate the safe fuel concentration. Furthermore, since the minimum represented by the envelope increases as the pipe diameter decreases, a horizontal pipe will not become critical if the moderator alone is removed, say by evaporation, if one conservatively considers the remaining solution constrained in a circular cylinder of decreasing diameter. This argument obviously leads to

conservatism since the actual configuration in a partially filled horizontal cylinder has a greater leakage than a circular cylinder of the same cross-sectional area.

Recommendations

In view of the uncertainty of the magnitude of the error in these results, they should be used with considerable caution and with as large a safety factor as can be tolerated. This is especially true for the pipes with no cadmium covering containing D_2O or D_2O-H_2O mixtures as moderator. It is strongly recommended that multi-group calculations should be programmed for the computing machines as an improved approach to the problem. Furthermore, this should be supported by criticality experiments at least with pure D_2O moderator to check the reliability of the calculations. The problem of safe containers for the conditions discussed here will most probably arise even more frequently as larger scale homogeneous reactors and associated chemical plants are designed.

The writers wish to acknowledge the very helpful discussions with M. C. Edlund.

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FEYNMAN-WELTON APPROXIMATION

Appendix A

For the concentrations considered here, the resonance escape probability, p, can be taken as unity. Denoting the fast group by the subscript 1 and the slow group by 2, the uncoupled diffusion equations are:

(1) $D_{1c}\nabla^2\Phi_{1c} + (k_1-1)\Sigma_{1c}\Phi_{1c} = 0$ fast, core

(2) $D_{1B}\nabla^2\Phi_{1B} - \Sigma_{1B}\Phi_{1B} = 0$ fast, reflector

(3) $D_{2c}\nabla^2\Phi_{2c} + (k_2-1)\Sigma_{2c}\Phi_{2c} = 0$ slow, core

(4) $D_{2B}\nabla^2\Phi_{2B} - \Sigma_{2B}\Phi_{2B} = 0$ slow, blanket

In cylindrical coordinates, these become

(1') $$\frac{d^2\Phi_{1c}}{dr^2} + \frac{1}{r}\frac{d\Phi_{1c}}{dr} + (k_1-1)\frac{\Sigma_{1c}}{D_{1c}}\Phi_{1c} = 0$$

(2') $$\frac{d^2\Phi_{1B}}{dr^2} + \frac{1}{r}\frac{d\Phi_{1B}}{dr} - \frac{\Sigma_{1B}}{D_{1B}}\Phi_{1B} = 0$$

(3') $$\frac{d^2\Phi_{2c}}{dr^2} + \frac{1}{r}\frac{d\Phi_{2c}}{dr} + (k_2-1)\frac{\Sigma_{2c}}{D_{2B}}\Phi_{2c} = 0$$

(4') $$\frac{d^2\Phi_{2B}}{dr^2} + \frac{1}{r}\frac{d\Phi_{2B}}{dr} - \frac{\Sigma_{2B}}{D_{2B}} = 0$$

The usual boundary conditions hold.

(5) $\Phi_{1c}(a) = \Phi_{1B}(a)$

(6) $$-D_{1c}\left.\frac{d\Phi_{1c}}{dr}\right)_{r=a} = -D_{1B}\left.\frac{d\Phi_{1B}}{dr}\right)_{r=a}$$

[redacted]

(7) $\Phi_{2c}(a) = \Phi_{2B}(a)$

(8) $-D_{2c} \left.\frac{d\Phi_{2c}}{dr}\right)_{r=a} = -D_{2B} \left.\frac{d\Phi_{2B}}{dr}\right)_{r=a}$

(9) $\Phi_{1B}(\infty) = 0 \quad ; \quad \Phi_{2B}(\infty) = 0.$

The solutions are given in terms of Bessel functions:

(10) $\Phi_{1c} = A_1 J_o(B_1 r) \quad ; \quad \Phi_{1B} = E_1 K_o(\mathcal{H}_1 r)$

$\Phi_{2c} = A_2 J_o(B_2 r) \quad ; \quad \Phi_{2B} = E_2 K_o(\mathcal{H}_2 r)$

where the following substitutions have been made;

$$B_1^2 = (k_1-1)\frac{\Sigma_{1c}}{D_{1c}} \quad ; \mathcal{H}_1^2 = \frac{1}{\tau_B} = \frac{\Sigma_{1B}}{D_{1B}}$$

$$B_2^2 = (k_2-1)\frac{\Sigma_{2c}}{D_{2c}} \quad ; \mathcal{H}_2^2 = \frac{1}{L_o^2} = \frac{\Sigma_{2B}}{D_{2B}}$$

L_o is the diffusion length in H_2O.

From the boundary conditions (5), (6), (7), and (8),

(12) $B_1 a \frac{J_1(B_1 a)}{J_o(B_1 a)} = \frac{D_{1B}}{D_{2c}} \mathcal{H}_1 a \frac{K_1(\mathcal{H}_1 a)}{K_o(\mathcal{H}_1 a)}$

(13) $B_2 a \frac{J_1(B_2 a)}{J_o(B_2 a)} = \frac{D_{2B}}{D_{2c}} \mathcal{H}_2 a \frac{K_1(\mathcal{H}_2 a)}{K_o(\mathcal{H}_1 a)}$

which are solved for B_1 and B_2.

From considerations of neutron balance, there is obtained the criticality equation:

$$(14) \quad k_1 k_2 = \eta f \left[1 + (k_1 - 1) p_{12} \right]$$

where p_{12} is the probability that a neutron which is thermalized in the blanket is absorbed in the core. In terms of z, the ratio in the core of uranium to moderator macroscopic cross-section, (14) becomes

$$(15) \quad z = \frac{1 + L_o^2 B_2^2}{\dfrac{\eta (1 + c)}{1 + \gamma B_1^2} - 1}$$

where

$$C = \frac{D_{2c} L_o^2 (\text{core})}{D_{2B} L_o^2 (\text{reflector})} A_1^2 A_2^2 \int_{\text{core}} r dr \; J_o(B_1 r) \, J_o(B_2 r) \int_{\text{reflector}} r dr \; J_o(B_1 r) \, J_o(B_2 r$$

where A_1 and A_2 are normalization constants (over the core).

In the case of the cadmium covered pipes, C is zero and $J_o(B_2 a)$ is made to vanish at an extrapolation distance, ε, beyond the pipe radius. Thus, $B_2 = 2.405/(a + \varepsilon)$.

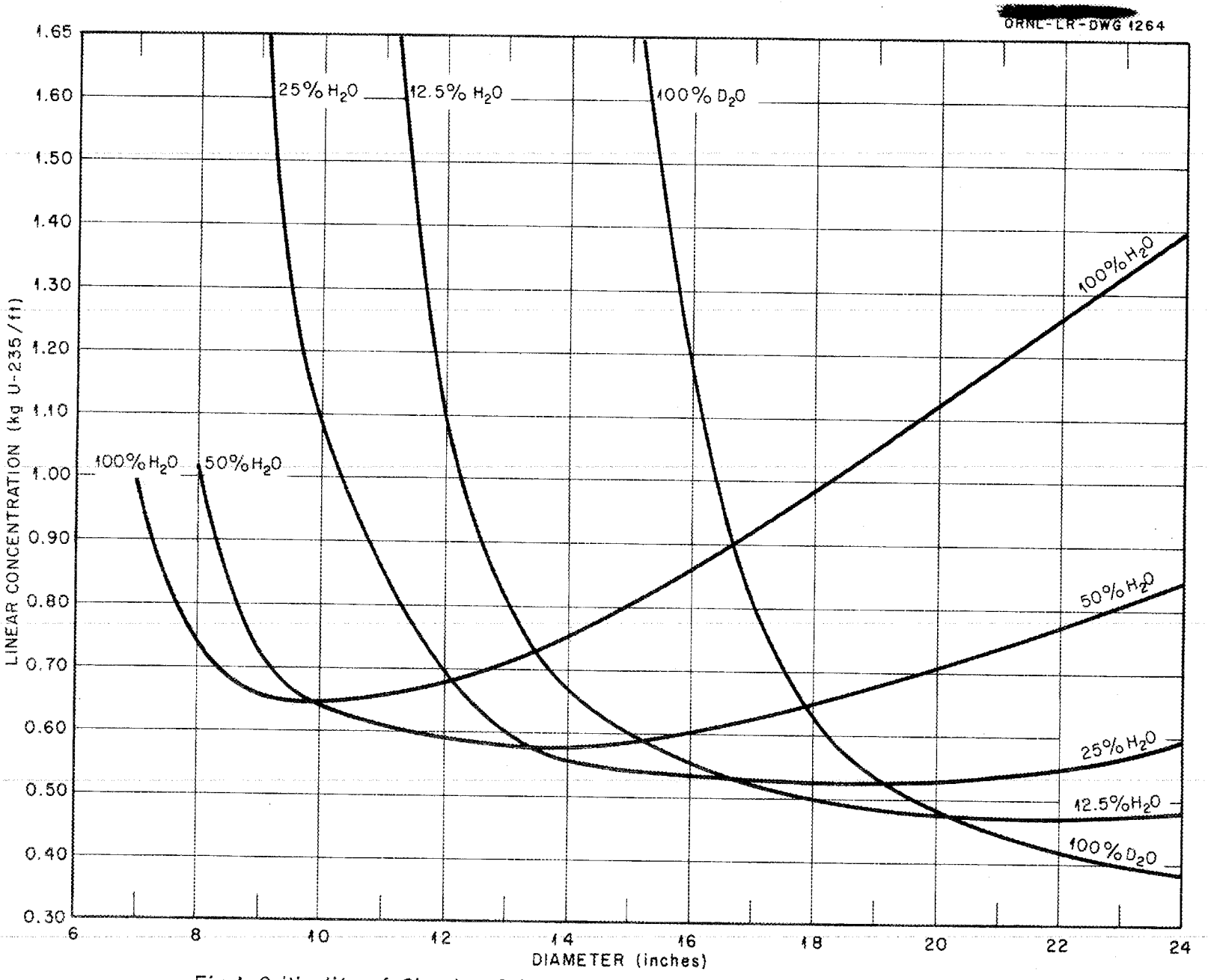

Fig. 1. Criticality of Circular Cylinders of Infinite Length, H_2O Reflected, 20°C

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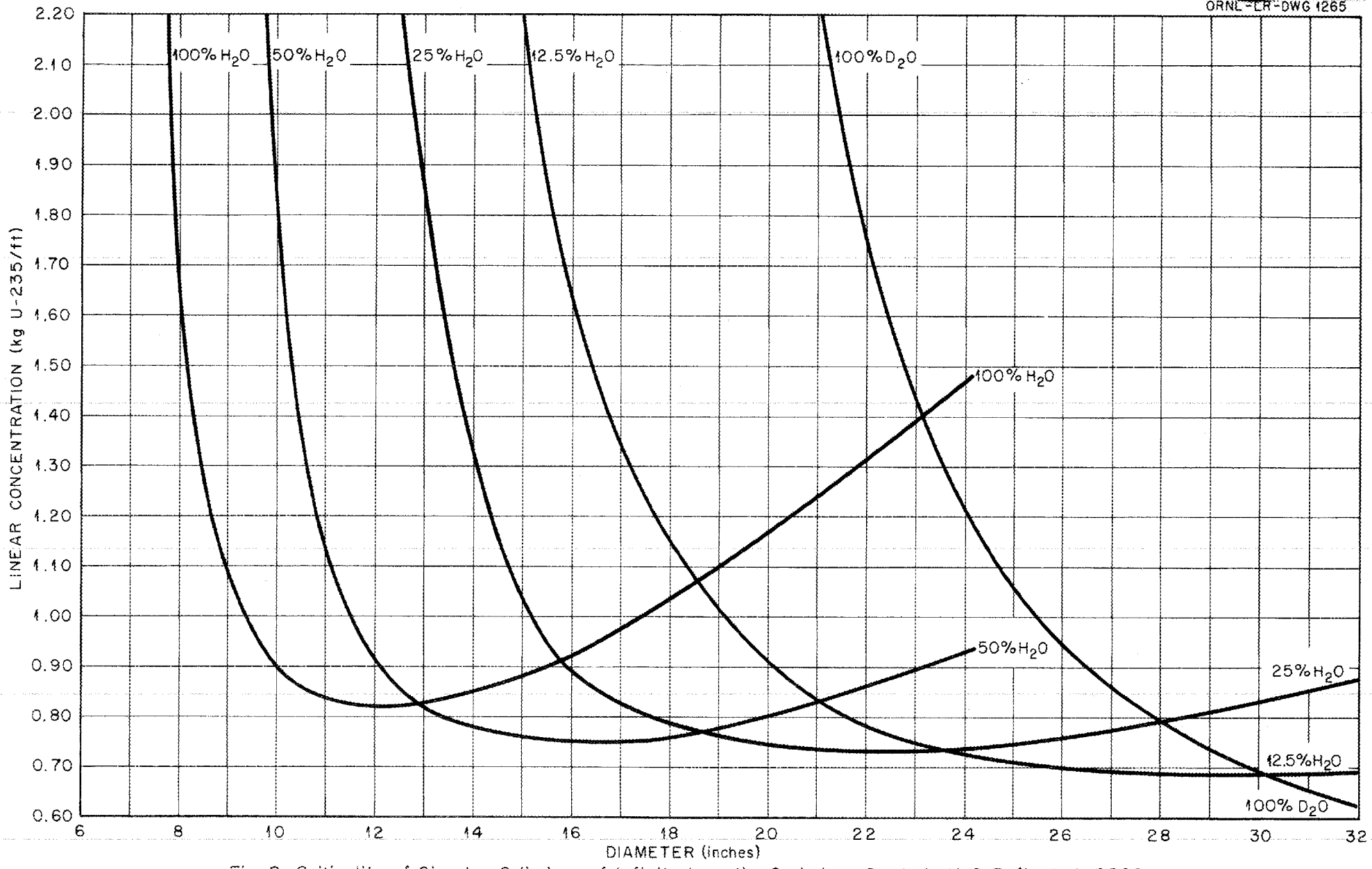

Fig. 2. Criticality of Circular Cylinders of Infinite Length, Cadmium Coated, H_2O Reflected, 20°C

[redacted]

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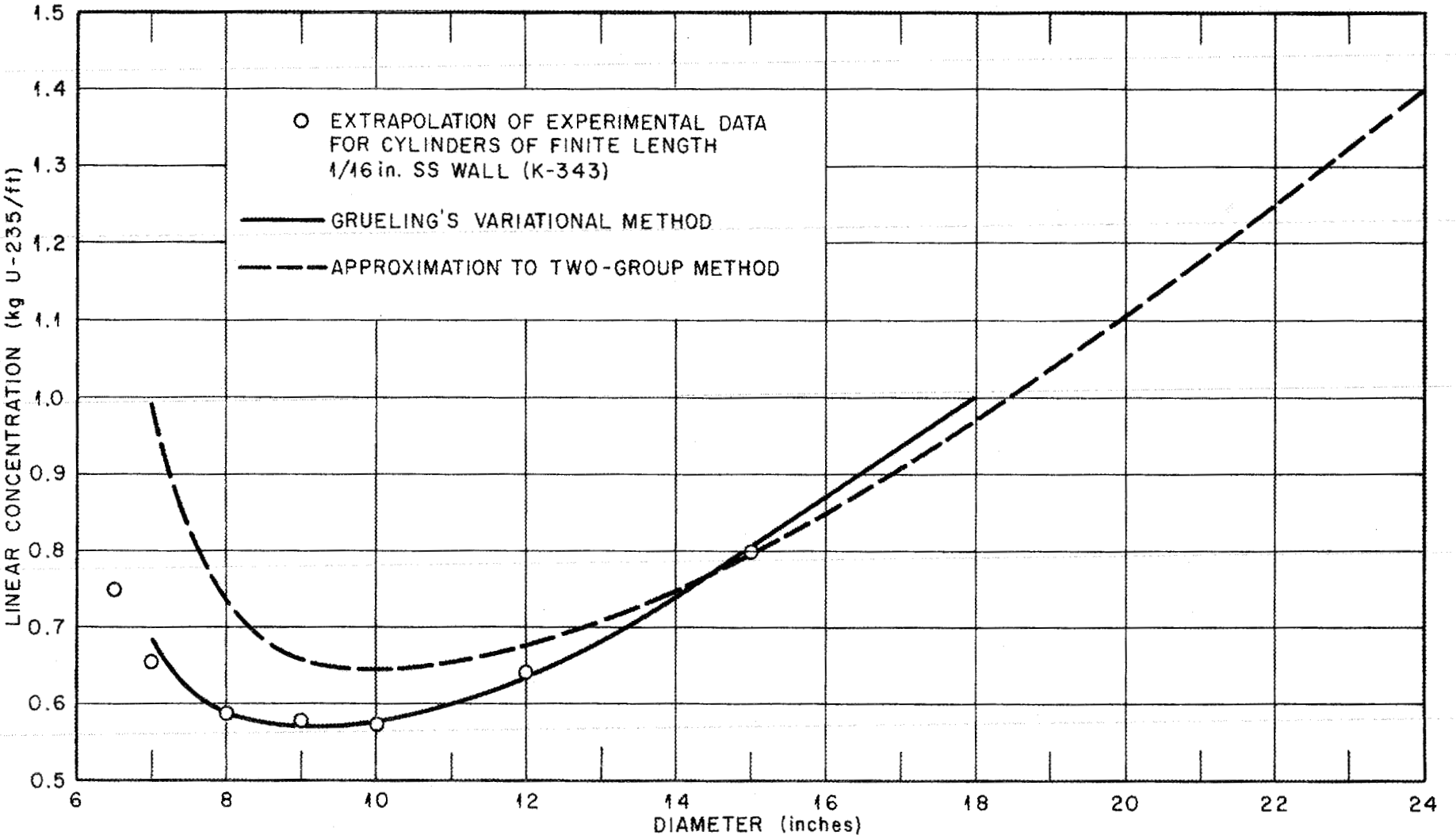

Fig. 3. Comparison of Calculations and Experiment for Criticality of Cylinders of Infinite Length, H_2O Moderated And Reflected, 20°C