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UO₂F₂ AQUEOUS SOLUTIONS
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PARAMETERS FOR TWO GROUP ANALYSIS OF CRITICAL EXPERIMENTS
WITH WATER REFLECTED SPHERES OF UO_2F_2 AQUEOUS SOLUTIONS

Introduction

A great many critical experiments have been performed with water reflected UO_2F_2 aqueous solutions over an extensive range of concentrations. Those in spherical geometry are particularly attractive for calculations because of their simplicity. Using the data from spheres it is possible to obtain a system of parameters that will permit two-group two-region calculations to be made with reasonable accuracy. It is not surprising that one can create a consistent set of constants over an extended concentration range when consideration is given to the number of parameters that are adjustable. A disappointing fact is the inability to extract acceptable values for the dimensions of infinite slabs and cylinders using the constants derived for the finite spheres. The parameters, however, do suit the experimental data and help pin point the vague concentrations at which the minimum mass and volume occur as well as establish these values.

I - Experimental Data

Three spheres, spun of 2S aluminum, having nominal diameters of 11.52, 13.21, and 15.96 cm, have been used. Their critical conditions are listed in Table 1. The oxyfluoride solution was enriched to ~90% in the U-235 isotope.

Table 1. Critical Conditions for Water Reflected Spheres

Diameter (cm)	<u>U-235 Concentration</u>		Volume (Liters)	Mass U-235 (kg)
	g/L	H/U-235		
11.52	649.05	35.8	6.400	4.154
11.52	483.13*	49.9	6.400	3.092
13.21	95.11	269.8	9.660	0.918
15.96	49.41	524.0	17.020	0.841

*This critical concentration was obtained from the extrapolation of a series of measurements made with the sphere about 99% filled.

The variation of critical mass and volume in the 11.52 cm diameter sphere with concentration was such that the mass and volume decreased with decreasing concentration, the volume passing through a minimum and increasing again, as shown by the dashed curve in Fig. 1. The extrapolated

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Fig. 1 - The critical mass vs critical volume in spherical geometry.

point appearing in Table 1 was obtained by extending the mass vs volume curve until the void present was again zero. Also shown in Fig. 1 are the critical points for the other spheres.

Measurements were made of the fuel cadmium fraction at the higher concentrations and at the most dilute concentration. These fission rate measurements are made by exposing, in the solution, capsules containing U-235. Each capsule consists of a 3/8-in. O. D. by 1/4-in. cylindrical Lucite tube filled with 25 to 50 mg of 90% enriched UO_2F_2 . The capsule (with and without cadmium covers) are closely fitted in a thin walled Lucite tube which, in turn, is inserted into another Lucite tube (1/2-in. O. D.) and immersed in the solution. After exposure, the gamma activity of the U-235 fission products in each capsule is counted. Several of the measurements were made in partially filled spheres.* These are shown in Fig. 2 as triangles. The circles are measurements made in full spheres. It is to be noted that the difference between the filled and partially filled spheres is slight. The measured indium cadmium fraction is also shown in Fig. 2 where the isolated square point is a measurement made in a 15 cm diameter cylinder. The indium foils (bare and cadmium covered) were sealed in Teflon tape, mounted on a stringer, and immersed in the solution. After exposure the foils are removed from the tape and counted.

II - Theory and Method

The nuclear constants required in the two-group two-region calculation, as a function of concentration, are the fast and slow diffusion coefficients, D , the thermal diffusion lengths, L , the neutron ages, γ , for both the core and reflector, and the infinite multiplication factors, k_∞ . In the following let the subscript notation c and r denote the core and reflector regions respectively and the numbers 1 and 2 denote the fast and slow neutron groups. The critical determinant for spherical geometry, in which shell absorption has been included in the boundary condition of equating the thermal neutron currents, may be written as

$$\begin{vmatrix} \alpha_1 + \frac{D_{1r}}{D_{1c}} K_F & \alpha_2 + \frac{D_{1r}}{D_{1c}} K_F \\ S_1 \gamma_1 + S_3 \alpha_3 (K_F - K_S) & S_2 \gamma_2 + S_3 \alpha_3 (K_F - K_S) \end{vmatrix} = 0 \quad (1)$$

* A portion of this data appears in "Semiannual Progress Report, Physics Division, for Period Ending March 10, 1955", ORNL-1926 p 6 ff.

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Fig. 2 - Measured fuel and indium cadmium fraction on full (○) and partially filled (△) spheres. The square point is a measurement in a 15 cm diameter cylinder.

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where the symbols have the definitions

$$\begin{aligned}\alpha_1 &= \mu R_0 \cot \mu R_0 - 1 & K_F &= 1 + \chi_{1r} R_0 \\ \alpha_2 &= \nu R_0 \coth \nu R_0 - 1 & K_S &= 1 + \chi_{2r} R_0 \\ \alpha_3 &= \frac{D_{2r}}{D_{2c}} & \gamma_1 &= (\alpha_1 - \alpha_4 + \alpha_3 K_S) \\ \alpha_4 &= \frac{\Sigma_a(\text{shell}) t_{\text{shell}}}{D_{2c}} & \gamma_2 &= (\alpha_2 - \alpha_4 + \alpha_3 K_S).\end{aligned}$$

From the characteristic equation for the core of a reflected reactor, the quadratic in B^2 , the material buckling has the two values μ^2 and $-\nu^2$ defined by

$$\mu^2 = \frac{1}{2} \left\{ -\left(\frac{1}{L_{2c}^2} + \frac{1}{\tau_c}\right) \pm \sqrt{\left(\frac{1}{L_{2c}^2} + \frac{1}{\tau_c}\right)^2 + \frac{4(k_\infty - 1)}{\tau_c L_{2c}^2}} \right\} \quad (2)$$

It is seen that the material buckling is a function of τ_c , L_{2c}^2 , and k_∞ . The infinite multiplication factor is taken as

$$k_\infty = \eta f p \epsilon \quad (3)$$

where the fast fission factor, ϵ , has been assumed equal to unity, the resonance escape probability* as given Fig. 3, η is constant and equal to 2.08, and f , the thermal utilization, is calculated for the core materials using absorption cross sections for 2200 m/s neutrons. The variation of L_{2c}^2 with concentration is obtained from the expression

$$L_{2c}^2 = \frac{D_{2c}}{\Sigma_a(c)} \quad (4)$$

where D_{2c} is defined as

$$D_{2c} = \Sigma_a(H) L_0^2 \left(\frac{\rho_0}{\rho} \right)^2, \quad (5)$$

the subscript zero referring to water and the density ratio is that of hydrogen in the core. L_{2c}^2 and D_{2c} as a function of concentration are shown in Fig. 4.

The age, however, is not taken as the age to thermal energy but rather as the age to some mean energy of fission production, E_m . Since the mean energy at which fissions occur increases as the concentration increases, the age is expected to decrease with increasing concentration. The simple expression which corrects the age for density variation, namely,

* The figure is from "HRP Quarterly Progress Report, Period Ending March 31, 1953," ORNL-1554.

It is equally convenient to assume $p\epsilon = 1$ which would entail only moderate changes in the derived parameters.

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Fig. 3 - Resonance escape probability as a function of the atomic ratio $^2\text{H}/\text{U-238}$.

Fig. 4 - The square of the thermal diffusion length and the thermal diffusion coefficient for UO_2F_2 solutions as a function of the atomic ratio $H/U-235$.

$$\tau_c \propto \rho^{-2} \quad (6)$$

is not satisfactory because, at high concentrations, τ_c will not cause the determinant in equation (1) to vanish. This can be shown by solving equation (1) for the fast diffusion coefficient in the core and examining the resulting expression. Recalling the definitions of S_1 , S_2 and S_3 , there obtains

$$-D_{1c} = D_{1r} \left\{ \frac{\tau_{cR_0}(\alpha_2 - \alpha_1) \alpha_5 \alpha_6 + K_F \tau_r L_{2c}^2 (\chi_{1r} + \chi_{2r}) (\alpha_5 \gamma_2 - \alpha_6 \gamma_1)}{\tau_r L_{2c}^2 (\chi_{1r} + \chi_{2r}) (\alpha_1 \alpha_5 \gamma_2 - \alpha_2 \alpha_6 \gamma_1)} \right\} \quad (7)$$

where $\alpha_5 \equiv 1 + L_{2c}^2 \mu^2$ and $\alpha_6 \equiv 1 - L_{2c}^2 \gamma^2$. Consideration of the quantities in this expression reveals that unless α_1 is less than α_2 , D_{1c} will have a negative value. Hence the following inequality subsists

$$\cot \mu R_0 < \frac{\gamma}{\mu},$$

since $\coth \gamma R_0 = 1$. The relation given in (6) is inadequate because the resulting τ_c fails to satisfy this inequality. Therefore, to approximate τ_c as a function of concentration E_m is assumed proportional to the fuel cadmium fraction (C. F.) measured near the center of reactivity and shown in Fig. 2, thus*

$$\frac{1}{\tau_c} \propto E_m \propto \text{C. F.} \quad (8)$$

The resulting variation of τ_c with concentration is shown in Fig. 5 where τ_c has been normalized to 26.00 cm² at an H/U-235 of 524.

The reflector water constants are assumed independent of the core concentration and equal to the values listed in Table 2.

Table 2. Reflector Water Constants

$D_{1r} =$	1.135 cm
$D_{2r} =$	0.160 cm
$L_{2r}^2 =$	8.123 cm ²
$\tau_r =$	27.00 cm ²

The remaining constant to be characterized as a function of concentration is the fast diffusion coefficient in the core. This is accomplished by substituting in equation (7) the constants determined in the manner described above and the experimental data of Table 1.

* Preliminary unpublished measurements and analysis by R. Gwin at the ORNL Critical Experiments Laboratory give ages in fair agreement with those used here.

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Fig. 5 - The age to fission in UO_2F_2 solutions as a function of the atomic ratio H/U^{235} .

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D_{1c} as a function of the core concentration is shown in Fig. 6 along with the calculated values of μ from equation (2). Figure 7 presents the values of ν from equation (2), shown as a function of μ , for completeness.

III - Results

The calculated critical spherical radii, over the range of concentrations defined by $35 < H/U-235 < 525$, are presented in Fig. 8. The circled points are the experimental data of Table 1. The results from the determinations of minimum critical mass and volume are given in Table 3.

Table 3. Minimum Critical Mass and Volume

	U-235 Concentration		Volume (liters)	Mass U-235 (kg)
	g/L	H/U-235		
Minimum Volume*	537.86	44.	6.291	3.383
Minimum Mass	58.81	440.	13.434	0.790

* U-235 Enrichment ~90%.

It is significant to point out that these constants fail to give reasonable results when applied to the geometries of the infinite slab and cylinder. This is probably due to the dependence of the measured cadmium fraction on geometry as indicated by the point in Fig. 2 determined from measurements in a cylinder. The implication is, therefore, that the constants have a geometric dependence, hence, a consistent set of constants in one geometry may not satisfy another.

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Fig. 6 - The square root of the core material buckling, μ , and the fast diffusion coefficient for UO_2F_2 solutions as a function of H/U-235.

Fig. 7 - The core material buckling values, k vs. V , for UO_2F_2 solutions.

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Fig. 8 - Critical spherical radii for water reflected UO_2F_2 aqueous solutions as a function of the atomic ratio H/U-235.

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P-Uranium and Uranium Compounds

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- ## 1. Uranyl Fluorides -- Criticality Studies

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