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D. W. Magnuson

UNION CARBIDE

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OAK RIDGE, TENNESSEE

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CRITICAL CONCENTRATION OF $U(37)O_2F_2$ AQUEOUS SOLUTION IN AN UNREFLECTED 69.2-cm-DIAMETER ALUMINUM SPHERE

D. W. Magnuson

OAK RIDGE Y-12 PLANT
P. O. Box Y, Oak Ridge, Tennessee 37830

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The experiments with $U(37)O_2F_2$ aqueous solution[1] followed the series of experiments with $^{233}UO_2(NO_3)_2$ and $U(93)O_2(NO_3)_2$ solutions in the 69.2-cm-diam sphere.[2] The critical concentrations of ^{233}U and ^{235}U were used to evaluate the ratio of $\overline{\eta\sigma_a}(233)/\overline{\eta\sigma_a}(235)$ some years ago when the accepted value of $\bar{\eta}(233)$ was questioned. The purpose of the experiment reported here was to measure the increase in ^{235}U critical concentration and, hence, the increase in critical mass due to the increase in the ^{238}U content in the 69.2-cm-diam sphere.

The $U(37)O_2F_2$ concentration in an aqueous solution was adjusted so that when an aluminum spherical vessel was completely filled the multiplication factor was greater than unity and the excess reactivity was measured by means of a positive reactor period. The critical conditions are summarized in Table 1. The critical conditions for $U(93)O_2(NO_3)_2$ solution in the same sphere are also given for comparison and there is only a small difference in the critical ^{235}U density or mass. In these well moderated solutions there is only a small amount of neutron absorption in ^{238}U.

A comparison of the calculated multiplication factors using the DSN[3] and ANISN[4] transport codes with different Hansen-Roach 16-group cross-section sets is presented in Table 2. The calculated value of 1.0005 is to be compared to the experimental value of 1.0011.

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1. The notation U(37) is used to signify uranium enriched to 37 wt% ^{235}U.
2. R. Gwin and D. W. Magnuson, Nucl. Sci. Eng. 12, 364 (1962); and R. Gwin and D. W. Magnuson, "Critical Experiments for Reactor Physics Studies," ORNL-CF-60-4-12, Oak Ridge National Laboratory (1960).
3. B. G. Carlson, C. Lee, and J. Worlton, "The DSN and TDC Neutron Transport Codes," LAMS-2346 and LAMS-2346 Appendix I, Los Alamos Scientific Laboratory (1959).
4. Ward W. Engle, "A User's Manual for **ANISN**, A One-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering," K-1693, Oak Ridge Gaseous Diffusion Plant (1967).

Table 1. Critical Conditions for $U(37)O_2F_2$ and $U(93)O_2(NO_3)_2$ Aqueous Solutions in a 69.2-cm-diam Sphere.

Aluminum Vessel Description		
Measured Volume:	173.6 liter	
Inside Diameter[a]	69.20 cm	
Outside Diameter[b]	69.84 cm	
Axial Port Diameters[c]	5.08 cm	
Solution	$U(37)O_2F_2$	$U(93)O_2(NO_3)_2$
Density, g/cm^3	1.057	1.0288
Uranium, mg of U/g	49.13	19.56
^{235}U, wt%	36.96	93.18
^{235}U, Density, mg/cm^3	19.19	18.75
^{235}U Mass, g	3331	3255
$H:^{235}U$ Atomic Ratio	1343	1378
Multiplication Factor,[c] k_{eff}	1.0011	1.0012

a. Average diameter calculated from the volume.
b. The average wall thickness was 0.32 cm and was measured to have a reactivity value of $\rho = 6.9 \times 10^{-4}$.
c. The reactivity of top axial or polar port when filled with solution was measured to be 0.72×10^{-4} and the multiplication factors have been corrected by 0.72×10^{-4} for both top and bottom ports.

Table 2. Calculated Multiplication Factors of a 69.2-cm-diam Sphere of $U(37)O_2F_2$ Aqueous Solution.

Code	Quadrature	Hansen-Roach ^{238}U Cross-Section Set[a]	k_{eff}
DSN	S4	U238Y	1.0097
ANISN	S4	U238Y	1.0060
ANISN	S4	U23812	1.0115
ANISN	S8	U23812	1.0005

a. The infinite dilution ($\sigma_p = \infty$) cross-section set, U238Y, was used in the DSN calculations in 1962 but the set U23812 ($\sigma_p = 10,000$) is the set appropriate for this solution and the present transport code, ANISN, was used to evaluate the change in cross-section set.

A measurement related to the critical experiment and the description of the neutron energy flux was the magnitude of the epithermal flux parameter, λ, which is defined by the equation

$$\varphi = \int_0^{0.2\ \mathrm{eV}} \varphi(\mathrm{MB})\, dE + \lambda \int_{0.2\ \mathrm{eV}}^{\infty} \frac{dE}{E} \quad .$$

In this representation, the neutron energy flux consists of a Maxwell-Boltzman distribution, φ(MB), plus a high energy component inversely proportional to the neutron energy, λ/E. The values of λ were 0.03 from cadmium ratio measurements with gold and U(93) foils. The estimated error is approximately 10% due to uncertainties in the resonance integrals and foil self-shielding corrections. The detailed description of this measurement method is given in Refs. 2 and 5 where it was found that $\lambda = 0.7195\ \Sigma_a$ where Σ_a is the total macroscopic absorption thermal neutron cross section for the solution. This equation predicts a value of 0.0395 for λ which should be compared to the measured value above.

The fission rate distribution along a diameter of the sphere was calculated by the DSN transport code and was measured with a ^{235}U fission counter which was 0.64-cm-o.d. by 2.5-cm long. The results normalized to unity at the center of the sphere are compared in Fig. 1. The flux distributions between points located 25 cm on opposite sides of the center were fitted to the equation $\phi = (A \sin B \bar{R})/B \bar{R}$ to evaluate the buckling, B^2, and the extrapolated radius $\bar{R}$. The calculated and measured distributions resulted in values of $\bar{R}$ of 37.47 and 36.66 cm, respectively, corresponding to extrapolation distances of 2.87 and 2.06 cm. An extrapolation distance of 2.16 cm was measured for a $U(93)O_2(NO_3)_2$ critical solution in this same sphere.[2] These extrapolation distances include the reflector savings resulting from neutrons reflected by the aluminum vessel.

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5. D. W. Magnuson, Nucl. Sci. Eng. 44, 266 (1971).

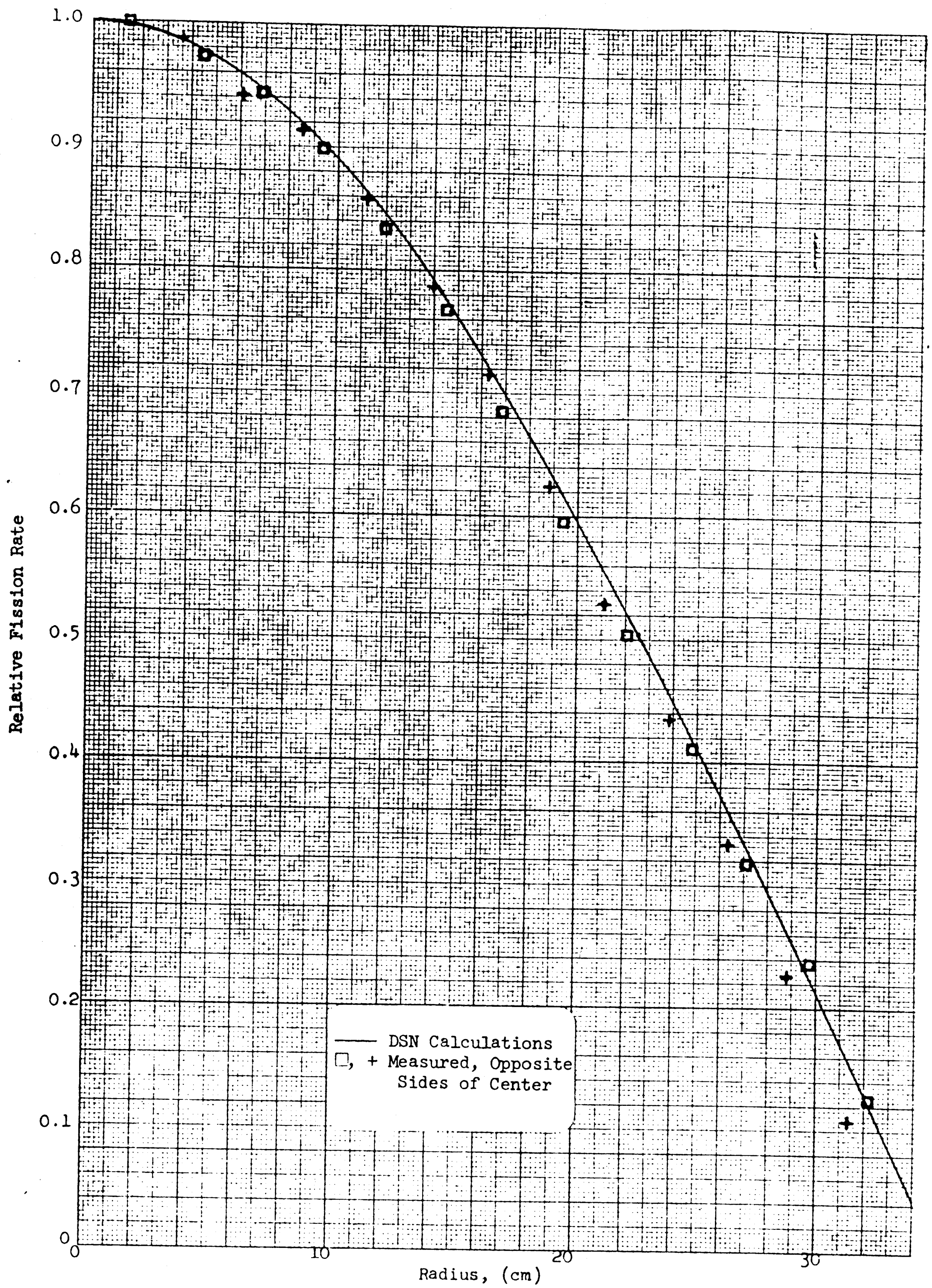

Fig. 1. Calculated and Measured Radial Fission Rate Distributions in a 69.2-cm-diam Sphere of $U(36.96)O_2F_2$ Aqueous Solution.