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Critical Measurements on UO_3-H_3PO_4 Solutions[1]

During the course of homogeneous reactor fuel studies, critical measurements were carried out with solutions of UO_3 (93.5% U^{235}) dissolved in 4.3 molar H_3PO_4. A brief description of these measurements, their results, and an evaluation of some calculations is presented here.

The basic fuel geometry for critical mass determinations was a right circular cylinder, 15.75 cm in radius. Fuel solution was held in a stainless steel container (0.1 cm wall thickness) surrounded radially and axially by a 9 cm iron reflector. The experimental apparatus was located in the Pajarito Site remotely-controlled critical assembly facility at LASL.

Determinations of critical masses were made as follows. The container was filled to various heights with phosphoric acid. An appropriately canned neutron source was located near the center of the phosphoric acid. Unmultiplied counts were taken with BF_3 proportional counters positioned outside the iron reflector. Multiplied counts were made after replacing the phosphoric acid with fuel solution.

Filling the experimental assembly with fuel solution was done with the top reflector raised, and with the container (together with the bottom reflector) withdrawn below the radial reflector. The solution was transferred into the container from 5-inch diameter 20-liter storage cans. These were pressurized to produce a slow flow of fuel, through Tygon tubing, to the experimental setup. A weighing device holding the storage cans indicated completion of the addition of a preselected amount of fuel. The container and bottom reflector were raised into the radial reflector, and the top reflector was lowered inside the container to the level of the fuel solution, by means of a remotely controlled assembly mechanism, after all personnel had left the experimental area.

[1] Work performed under the auspices of the U. S. Atomic Energy Commission.

Multiplied counting rates from the source neutrons were determined after each addition of fuel. Reciprocal multiplications were plotted against mass of fuel solution and extrapolated to obtain estimated values of the critical mass. The total amount of fuel injected into the assembly was about 98% of the estimated critical loading.

Table I lists properties, compositions, and critical volumes of four different uranium-containing phosphoric acid solutions used in the measurements.

TABLE I

CRITICAL MASS MEASUREMENTS ON CYLINDRICAL VOLUMES (15.75 CM RADIUS) OF UO_3 (93.5% U_{235}) IN 4.3 MOLAR H_3PO_4

Iron reflector thickness was 9 cm

	Solution 1	Solution 2	Solution 3	Solution 4
UO_3 molarity	0.51	0.46	0.41	0.34
H (atoms)/U (atoms)	198	220	248	300
H molarity	101.0	101.4	101.8	102.3
O molarity	62.7	62.7	62.8	62.8
Density (g/cc)	1.326	1.316	1.308	1.293
U concentration (g/liter)	120	108	96	80
U per kg of solution (g)	90.1	82.4	73.2	62.0
Critical height (cm)	16.4 ± 0.2	17.2 ± 0.2	18.0 ± 0.2	19.5 ± 0.3
Critical volume (liters)	12.8	13.4	14.0	15.2
Critical mass of U (kg)	1.53	1.45	1.34	1.22

A critical equation for assemblies containing hydrogeneous materials can be derived in a manner very similar to Fermi age theory (*1*) if it is assumed that slowing-down of neutrons is due only to collisions with hydrogen. The unique feature of scattering by hydrogen is that all final energies from zero to the initial energy E_0 have equal probability $P(E)$: $P(E)\,dE = dE/E_0$. We have calculated bucklings, B^2, for the fuel solutions using a form of slowing-down theory wherein account is taken of the neutrons removed from each energy interval dE by leakage, absorption, and scattering by hydrogen. The fission spectrum was not taken into account (*2*), and the neutron flux was assumed to be separable into a function of energy and a function of position. By fitting the experimental data to calculated values of B^2, the linear extrapolation length, δ, into the reflector was found to be 3.30, 3.28, 3.26, and 3.32 cm, respectively, for the 0.51, 0.46, 0.41, and 0.34 molar UO_3 fuel solutions. The variation is not inconsistent with estimated errors in the calculations and measurements.

We interpret the measurements as showing that the extrapolation length δ is independent of the H/U ratio over the range 200 to 300. This can be explained as follows: since the slowing-down length for fission neutrons is much longer than the diffusion-length of slow neutrons, most of the neutrons which are reflected by the 9 cm iron walls are fast neutrons. While the diffusion length is approximately inversely proportional to uranium concentration, the slowing-down length is nearly independent of uranium concentration. Hence the neutron leakage and the value of δ are also nearly independent of uranium concentration.

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A Contribution to Black Control Rod Worth

In a recent paper, Arnold (*1*) makes a useful suggestion enabling a calculation of control rod worth entirely from a perturbation analysis. An equivalent cross section, $\overline{\Sigma^{\text{rod}}}$, is defined for each group that will lead to the same reactor averaged cross section in the original flux ϕ as the actual property perturbation P in the new flux ϕ'. For example, in the thermal group, it is sufficient that

$$\frac{P_{22}\,\phi_2' + P_{21}\phi_1'}{\int \phi_2'\,dv} = \frac{\overline{\Sigma_2^{\text{rod}}\phi_2}}{\int \phi_2\,dv} \tag{1}$$

where P_{22} represents the perturbation in slow leakage and absorption and P_{21} is the perturbation in the source term from the fast group. The integrals are over the reactor volume.

An equivalent perturbation $\bar{P}_2$, however, can be defined for the terms such as P_{22} and P_{21}, that will lead to the same asymptotic period in the reactor. According to the method of Pigford *et al.* (*2*), it is sufficient that

$$\frac{\phi_2^* P_{22}\,\phi_2' + \phi_2^* P_{21}\,\phi_1'}{\int [(\phi_1^*\phi_1'/V_1) + (\phi_2^*\phi_2'/V_2)]\,dv} = \frac{\phi_2^*\bar{P}_2\phi_2}{\int [(\phi_1^*\phi_1/V_1) + (\phi_2^*\phi_2/V_2)]\,dv} \tag{2}$$

V represents the group speeds. The adjoint flux, ϕ_2^*, cancels in the numerator of Eq. (2) and, on defining

$$F_2 = \frac{\int \phi_2'\,dv \int [(\phi_1^*\phi_1/V_1) + (\phi_2^*\phi_2/V_2)]\,dv}{\int \phi_2\,dv \int [(\phi_1^*\phi_1'/V_1) + (\phi_2^*\phi_2'/V_2)]\,dv} \tag{3}$$

we have

$$\bar{P}_2 = F_2\overline{\Sigma_2^{\text{rod}}}. \tag{4}$$

The F ratios have the following significance: the reactivity is affected by the flux change as well as by the property perturbation. For example, the leakage out of a control rod