ORNL-4280

Contract No. W-7405-eng-26

NEUTRON PHYSICS DIVISION

ANNUAL PROGRESS REPORT

For Period Ending May 31, 1968

F. C. Maienschein, Director
A. D. Callihan, Associate Director

Group Leaders

C. E. Clifford, Reactor and Weapons Radiation Shielding
R. G. Alsmiller, Theoretical Space and High-Energy Accelerator Shielding
R. W. Peelle, Experimental Space and High-Energy Accelerator Shielding

OCTOBER 1968

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

2.16 CRITICAL EXPERIMENTS WITH AQUEOUS SOLUTIONS OF $^{233}UO_2(NO_3)_2$

J. T. Thomas

The criticality of ^{233}U under conditions of interest to nuclear criticality safety as well as in basic geometries suitable for theoretical analysis has been studied in a series of experiments conducted over the past 18 months. In one series aqueous uranyl nitrate solutions of ^{233}U were examined in a large capacity vessel containing borosilicate glass in order to define systems having k_∞ of unity or less. In a second series as many as 27 nearly identical subcritical cylindrical volumes of the solution were assembled in air-spaced arrays both unreflected and reflected by polyethylene to establish parameters useful in storage and transport specifications. A final series of experiments determined the criticality of water-reflected and unreflected spherical and cylindrical volumes.

The ^{233}U isotopic content of the uranium of the uranyl nitrate solution was 97.59%. There were no impurities present in significant quantities.

Borosilicate Glass Experiments

Exponential experiments with uranyl nitrate solution having uranium concentrations of 333 and 204 g/liter were performed in an unreflected aluminum cylinder 50.8 cm in diameter and 18.3 cm high having a lateral wall thickness of 1.5 mm and a 1.27-cm-thick bottom. Flux traverses were made with a 6.4-mm-diam BF_3 counter along the axis of the cylinder through a mixture of solution and randomly oriented glass raschig rings. A neutron source was provided by adding sufficient solution above the glass rings to produce a critical slab.

A description of the raschig rings is given in Table 2.16.1. The conditions described in this table define mixtures having a negative material buckling and hence values of k_∞ less than unity. Thus, ^{233}U as an aqueous uranyl nitrate solution at a uranium concentration of 333 g/liter may be stored in any quantity, either mass or volume, provided that the borosilicate glass uniformly occupies 38% of the volume of the container.

Spaced Subcritical Components

Cylindrical containers, of 4.63 liters capacity and fabricated of 0.25-mm-thick stainless steel, had an outside diameter and a height of 18.28 and 17.67 cm, respectively. They were identically filled to within ±0.5 g of solution. Critical assemblies of reflected and unreflected arrays at uranium concentrations of 333 and 204 g/liter were constructed. The reflector material was 15.2-cm-thick polyethylene (ρ = 0.93 g/cm^3) located from the peripheral cylinders of the array by a distance equal to one-half the surface separation between cylinders. The critical conditions for the arrays of cylinders at the two uranium concentrations are summarized in Table 2.16.2.

Simple Geometries

Presented in Table 2.16.3 are the critical conditions for water-reflected and unreflected spherical and cylindrical volumes of the solution which had a value of k_{eff} of 1.0000 ± 0.0005. A concentration at which a sphere was critical was first established, and then several critical cylindrical volumes were measured. The results for unreflected cylinders have been corrected for the 1.27-cm-thick aluminum base of the container, so the results describe cylindrical volumes having aluminum on the lateral surface only.

Table 2.16.1. Conditions for Mixtures of Uranyl Nitrate Solutions of ^{233}U and Glass Raschig Rings Whereby $k_\infty < 1$

Uranium concentration (g/liter)	333	204
Natural-boron content of glass (wt %)	3.9	3.9
Glass content of mixture (vol %)	38	33
Dimensions (cm) of borosilicate glass raschig rings		
Length	4.45	1.59
Outside diameter	3.81	1.59
Wall thickness	0.56	0.43
Isotopic content of uranium (%)		
^{232}U (ppm)	6.47	
^{233}U	97.54	
^{234}U	1.05	
^{235}U	0.03	
^{236}U	<0.01	
^{238}U	1.39	
sotopic content of boron (%)		
^{10}B	19.74	
^{11}B	80.26	

Table 2.16.2. Critical Parameters for Unreflected and Reflected Arrays of Units of Uranyl Nitrate Solution of Uranium Containing 97.5% ^{233}U

No. of Units in Array[a]	Polyethylene Reflector Thickness[b] (cm)	Center-to-Center Separation of Units (cm)		Average Uranium[c] Density (g/cm^3)
		Horizontal	Vertical	
333 g of U per liter; H:^{233}U = 73; specific gravity, 1.468; 1.432 kg of U per container				
8 (2 × 2 × 2)	0	20.44	19.13	0.179
27 (3 × 3 × 3)	0	25.72	24.57	0.088
8 (2 × 2 × 2)	15.2	31.95	30.36	0.046
27 (3 × 3 × 3)	15.2	41.03	38.57	0.022
204 g of U per liter; H:^{233}U = 119; specific gravity, 1.280; 0.885 kg of U per container				
8 (2 × 2 × 2)	0	20.16	18.81	0.116
27 (3 × 3 × 3)	0	25.01	23.89	0.059
8 (2 × 2 × 2)	15.2	30.23	25.51	0.034
27 (3 × 3 × 3)	15.2	38.05	36.74	0.017

[a]The solution was contained in cylinders of 0.25-mm-thick stainless steel with an outside diameter of 18.28 cm and height of 17.67 cm. The number of units along the edges of the array is given in parentheses.

[b]The polyethylene reflector was located at the cell boundaries.

[c]See Table 2.16.1 for isotopic content.

Table 2.16.3. Critical Conditions of $^{233}UO_2(NO_3)_2$ Aqueous Solution in Water-Reflected and Unreflected Simple Geometries

$UO_2(NO_3)_2$ Solution[a]			Critical Dimensions									
			Spheres		Cylinders[b]							
					50.8-cm diam		38.1-cm diam		25.3-cm diam		20.3-cm diam	
Uranium Concentration (g/liter)	Specific Gravity	H:^{233}U	Radius (cm)	Mass (kg of U)	Height (cm)	Mass (kg of U)	Height (cm)	Mass (kg of U)	Height (cm)	Mass (kg of U)	Height (cm)	Mass (kg of U)
Unreflected Assemblies												
333	1.468	73			13.36	9.02						
204	1.280	122			13.51	5.59	15.14	3.52	24.69	2.53		
131	1.183	195	14.579	1.70	14.07	3.74	16.35	2.44	28.52	1.88		
102	1.144	253	15.078	1.46			17.60	2.05	33.40	1.71		
74.6	1.106	349	15.821	1.24			19.35	1.65	43.69	1.64		
44.6	1.050	581	18.378	1.16			26.37	1.34				
Water-Reflected Assemblies[c]												
132	1.186	194	11.170	0.769			13.42	1.77	17.22	1.14	22.86	0.976
95.0	1.135	273	11.847	0.662			*d*	*d*	19.67	0.939	20.02	0.824
47.9	1.068	548	14.579	0.621			19.6	1.07	31.53	0.757	*d*	*d*

[a]See Table 2.16.1 for isotopic content of the uranium.

[b]Aluminum cylinders had a 1.5-mm-thick wall and 1.27-cm-thick bottom. Spheres were of aluminum with 1.22-mm-thick wall.

[c]There was no reflector on the top of any cylinder. The surface of the reflector water was 24.3 cm above the solution, a distance equal to the bottom reflector thickness.

[d]There was insufficient solution inventory to achieve criticality; the maximum solution height was 12.5 cm in the 38.1-cm-diam cylinder and 68.2 cm in the 20.3-cm-diam cylinder.