Y-DR-107

CALCULATED CRITICALITY OF WATER MODERATED OXIDES OF URANIUM-233, THORIUM-232, AND CARBON MIXTURES

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ABSTRACT

The criticality of mixtures of ^{233}U and ^{232}Th oxides with various amounts of carbon and water are explored by calculations performed with the KENO and ANISN codes utilizing the Hansen-Roach neutron cross-section sets. The influence of varying the mixture ratios of thorium and/or carbon to uranium is evidenced. The advantages of detailed analysis of particular process operations and a suggested margin of subcriticality in the application of the calculated results in nuclear safety considerations are given.

INTRODUCTION

One of the fissile materials being considered for use in gas cooled reactors is the oxide of uranium-233 utilized as small particles coated with carbon and fabricated into fuel elements. The material may be mixed at different weight ratios with the oxide of thorium-232 for consideration as a breeder fuel. The preparation and fabrication of the elements will require an examination of the potential for criticality during the fuel process cycle. While examination of specific operations would be more direct, it is believed that the calculation of fundamental criticality data would be of more general use to programs using this fissile material. The criticality data is necessarily calculated; there being virtually no experimental data for the materials in the compositions proposed. The Hansen-Roach(1) neutron cross-section sets were used in this study in the KENO(2) Monte Carlo code and in the ANISN(3) code.

MATERIAL COMPOSITION

The nominal oxide density is assumed not to exceed 10.65 g/cm^3. Depending upon the thorium content in the mixture, usually expressed as the weight ratio of Th:U, the corresponding atomic ratio satisfies the relations:

$$\frac{N_{Th}}{N_U} = \frac{A_U}{A_{Th}} \left(\frac{1}{w} - 1 \right) \equiv R$$

where

$$N = N_U + N_{Th}$$

and w is the weight percent uranium in the mixture. Also,

$$N_U = \frac{N}{R + 1}, \quad N_{Th} = \frac{NR}{R + 1}$$

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1. Gordon E. Hansen and William H. Roach, "Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies," LAMS-2543, Los Alamos Scientific Laboratory (1961).
2. G. E. Whitesides and N. F. Cross, "KENO - A Multigroup Monte Carlo Criticality Program," CTC-5, Union Carbide Corporation Computing Technology Center (1969).
3. Ward W. Engle, Jr., "A Users Manual for ANISN," K-1693, Oak Ridge Gaseous Diffusion Plant (1967).

and

$$A = \frac{A_U}{w(R + 1)} ,$$

the atomic mass of the thorium and uranium in the mixture.

Assuming the application of carbon as a coating to the oxide particles results in an averaged particulate density, ρ, less than $\rho_O = 10.65$ g/cm^3, then

$$\frac{N_C}{N} = \frac{\rho_O(1 - \rho/\rho_O)}{\rho} \frac{A}{A_C}$$

where the subscript C denotes the element carbon.

The resulting oxide density in a uniform mixture may be expressed in terms of a defined N_C/N_U ratio as

$$\rho = \rho_O \left\{1 + \frac{\rho_O}{\rho_C} \frac{A_C}{A} \frac{N_U}{N} \frac{N_C}{N_U}\right\}^{-1} .$$

There may be situations in the fuel process cycle where water may become a constituent of the mixture. This condition is conservatively simulated by assuring the mixture is uniformly distributed and the water content defined by simple volume displacement. Let ρ' denote the resultant density in a unit volume of the mixture when water is present, then

$$N_H = (1 - \rho'/\rho) \frac{N_O}{9} ,$$

represents the hydrogen number density, and defines the fractional density change as

$$\frac{\rho'}{\rho} = \left\{1 + 9 \frac{N_U}{N_O} \frac{N_H}{N_U}\right\}^{-1} ,$$

in terms of the N_H/N_U atomic ratio.

These simple relations define the full compositions for specific values of N_C/N_U and N_H/N_U of interest.

CALCULATED CRITICALITY DATA

The critical radii for spheres and infinite cylinders closely reflected by a 20-cm thickness of water were determined with the ANISN code. The results are presented in Table 1 for various thorium-, carbon-, and hydrogen-to-uranium atomic ratios. The results are also represented in graphic form. The carbon density was taken as 1.8 g/cm^3.

The reflected spherical critical mass of ^{233}U as a function of ^{233}U density is given in Fig. 1 for various carbon- and hydrogen-to-uranium ratios in the absence of thorium. Similar representation of the ^{233}U mass is displayed in Fig. 2 for a Th:U weight ratio of unity and in Fig. 3 for a weight ratio of 4:1. Figures 4-6 present the data for the reflected-infinite cylinder radius as a function of uranium concentration for the three Th:U weight ratios.

Apparent in Figs. 1 and 4 is the increase in the critical mass and cylinder radius, respectively, upon the addition of carbon as a diluent to the uranium. The addition of thorium produces a similar result as shown in Fig. 7 for the critical mass and in Fig. 8 for the cylinder radius where carbon is absent from the mixtures.

Table 1. Calculated Critical Dimensions of Water Reflected Spheres and Infinite Cylinders for Various Mixtures of $^{232}UO_2$, Carbon, and Water.

$H/^{233}U$	$\rho(^{233}U)$ (g/cm^3)	Sphere Radius (cm)	Mass kg ^{233}U	Infinite Cylinder Radius (cm)
Th:U = 0:1		$N_C:N_U$ = 0		
0	9.364-0	7.12	14.16	4.09
1	6.877-0	7.79	13.58	4.53
2	5.433-0	8.25	12.77	4.83
3	4.491-0	8.55	11.74	5.03
10	2.028-0	9.16	6.53	5.48
30	7.90 -1	9.39	2.74	5.71
100	2.52 -1	10.05	1.10	6.29
300	8.55 -2	11.95	0.61	7.81
500	5.15 -2	13.90	0.58	9.32
600	4.30 -2	14.93	0.60	10.11
Th:U = 0:1		$N_C:N_U$ = 11.4		
0	2.310-0	15.5	36.03	9.65
1	2.120-0	14.9	29.38	9.27
2	1.960-0	14.4	24.52	8.95
3	1.822-0	14.0	20.90	8.66
10	1.221-0	12.1	9.06	7.43
30	6.28 -1	10.7	3.22	6.57
100	2.33 -1	10.5	1.13	6.61
300	8.32 -2	12.1	0.62	7.90
500	5.06 -2	14.0	0.59	9.42
600	4.24 -2	15.1	0.61	10.20
Th:U = 0:1		$N_C:N_U$ = 23.9		
0	1.265-0	20.60	46.32	13.2
1	1.206-0	19.60	38.04	12.5
2	1.152-0	18.80	32.06	11.9
3	1.103-0	18.00	26.95	11.4
10	8.50 -1	14.70	11.31	9.2
30	5.13 -1	11.96	3.68	7.4
100	2.15 -1	11.03	1.21	7.0
300	8.08 -2	12.40	0.65	8.1
500	4.97 -2	14.20	0.60	9.5
600	4.17 -2	15.20	0.61	10.3

Table 1 (Cont'd)

$H/^{233}U$	$\rho(^{233}U)$ (g/cm^3)	Sphere Radius (cm)	Mass kg ^{233}U	Infinite Cylinder Radius (cm)
Th:U = 0:1		$N_C:N_U$ = 46		
0	7.03-1	25.88	51.04	16.96
1	6.84-1	24.63	42.81	16.07
2	6.67-1	23.48	36.17	15.27
3	6.50-1	22.46	30.85	14.56
10	5.53-1	17.98	13.46	11.48
30	3.87-1	13.91	4.36	8.77
100	1.89-1	11.87	1.32	7.54
300	7.69-2	12.75	0.67	8.37
500	4.82-2	14.47	0.61	9.73
600	4.07-2	15.44	0.63	10.48
Th:U = 0:1		$N_C:N_U$ = 100		
0	3.37-1	32.2	47.13	21.5
1	3.33-1	30.7	40.36	20.5
2	3.28-1	29.5	35.27	19.6
3	3.24-1	28.4	31.09	18.8
10	2.98-1	23.1	15.39	15.2
30	2.42-1	17.5	5.43	11.3
100	1.46-1	13.7	1.57	8.8
300	6.87-2	13.6	0.72	9.0
500	4.49-2	15.1	0.65	10.2
600	3.83-2	16.0	0.66	10.9
Th:U = 1:1		$N_C:N_U$ = 0		
0	4.681-0	10.79	24.63	6.50
1	3.964-0	10.92	21.62	6.64
2	3.438-0	11.03	19.33	6.71
3	3.035-0	11.05	17.15	6.73
10	1.667-0	10.71	8.58	6.54
30	7.29 -1	10.28	3.32	6.32
100	2.45 -1	10.55	1.21	6.64
300	8.47 -2	12.31	0.66	8.07
500	5.12 -2	14.26	0.62	9.58
600	4.28 -2	15.3	0.64	10.38

Table 1 (Cont'd)

$H/^{233}U$	$\rho(^{233}U)$ (g/cm^3)	Sphere Radius (cm)	Mass kg ^{233}U	Infinite Cylinder Radius (cm)
Th:U = 1:1		$N_C:N_U$ = 11.9		
0	1.805-0	18.07	44.59	11.49
1	1.687-0	17.43	37.45	11.06
2	1.584-0	16.91	32.07	10.70
3	1.493-0	16.34	27.29	10.31
10	1.063-0	13.84	11.80	8.64
30	5.84 -1	11.71	3.93	7.29
100	2.26 -1	11.06	1.28	7.00
300	8.24 -2	12.49	0.67	8.20
500	5.03 -2	14.37	0.62	9.66
600	4.21 -2	15.39	0.64	10.45
Th:U = 1:1		$N_C:N_U$ = 75.9		
0	4.19-1	33.21	64.27	22.30
1	4.13-1	31.69	55.05	21.21
2	4.06-1	30.26	47.13	20.20
3	4.00-1	28.96	40.71	19.28
10	3.61-1	33.16	18.79	15.20
30	2.82-1	17.20	6.01	11.10
100	1.60-1	13.48	1.64	8.67
300	7.16-2	13.59	0.75	8.98
500	4.61-2	15.16	0.67	10.24
600	3.91-2	16.11	0.69	10.97
Th:U = 4:1		$N_C:N_U$ = 0		
0	1.872-0	19.56	58.68	12.76
1	1.746-0	19.30	52.58	12.50
2	1.635-0	18.95	46.61	12.31
3	1.538-0	18.45	40.46	11.95
10	1.086-0	15.04	15.48	9.81
30	5.91 -1	12.66	5.02	8.01
100	2.27 -1	11.84	1.58	7.57
300	8.25 -2	13.33	0.82	8.82
500	5.04 -2	15.23	0.75	10.31
800	4.22 -2	16.31	0.77	11.14

Table 1 (Cont'd)

$H/^{233}U$	$\rho(^{233}U)$ (g/cm^3)	Sphere Radius (cm)	Mass kg ^{233}U	Infinite Cylinder Radius (cm)
Th:U = 4:1		$N_C:N_U$ = 21.16		
0	8.78-1	29.23	91.85	19.65
1	8.49-1	28.54	82.65	19.12
2	8.21-1	27.47	71.30	18.34
3	7.97-1	26.26	60.41	17.48
10	6.55-1	19.92	21.71	12.99
30	4.35-1	14.99	6.14	9.62
100	2.00-1	12.76	1.74	8.21
300	7.86-2	13.73	0.85	9.10
500	4.89-2	15.52	0.77	10.51
600	4.11-2	16.56	0.78	11.32
Th:U = 4:1		$N_C:N_U$ = 122.13		
0	2.48-1	47.91	114.24	33.20
1	2.46-1	45.04	94.18	31.12
2	2.44-1	42.55	78.73	29.32
3	2.41-1	40.38	66.48	27.75
10	2.27-1	31.76	30.47	21.55
30	1.93-1	22.59	9.32	15.03
100	1.27-1	16.42	2.36	10.80
300	6.40-2	15.51	1.00	10.37
500	4.28-2	16.81	0.85	11.45
600	3.68-2	17.74	0.86	12.18

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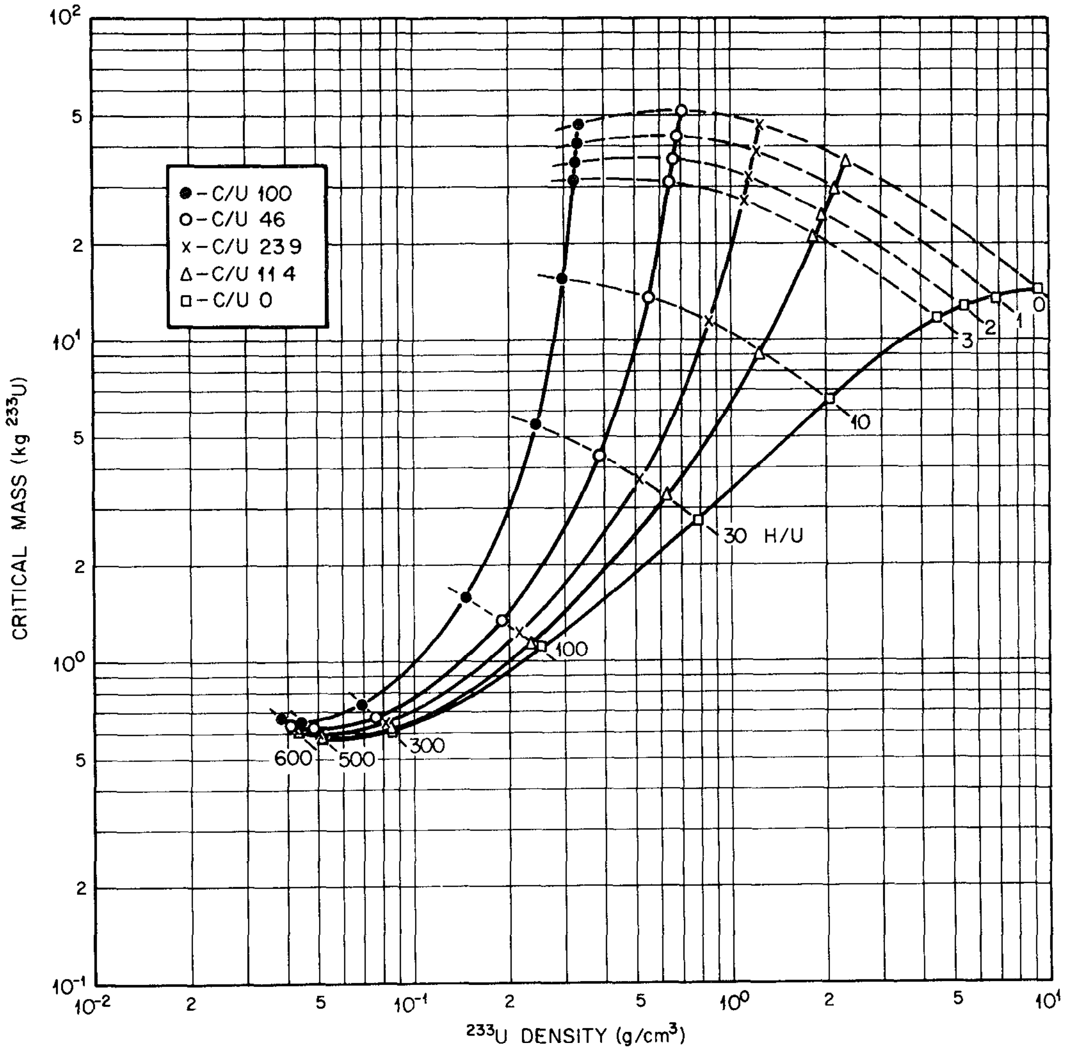

Fig. 1. Critical Masses of $^{233}UO_2$ in Spherical Geometry (water reflected) with Various H/U and C/U.

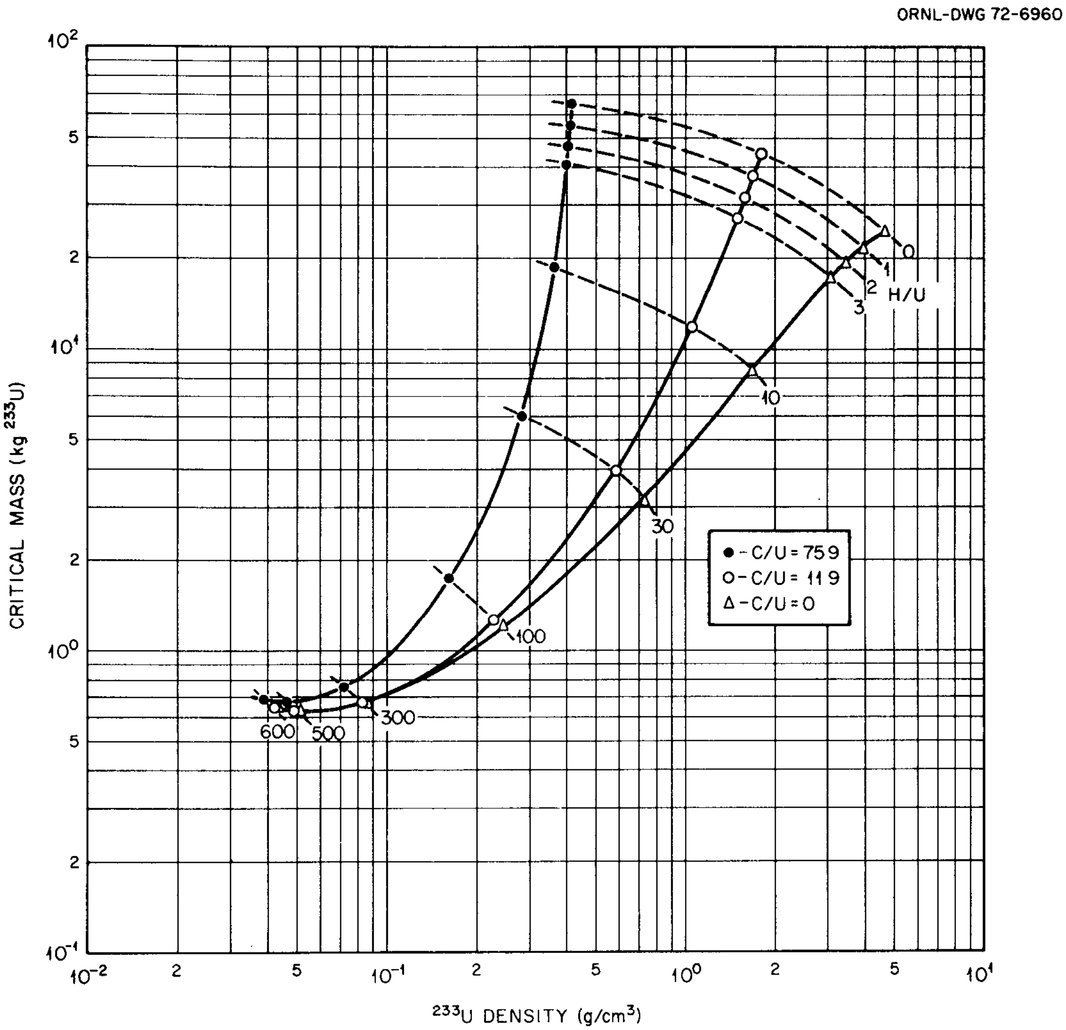

Fig. 2. Critical Masses of (1 Th:1 ^{233}U)O_2 in Spherical Geometry (water reflected) with Various H/U and C/U.

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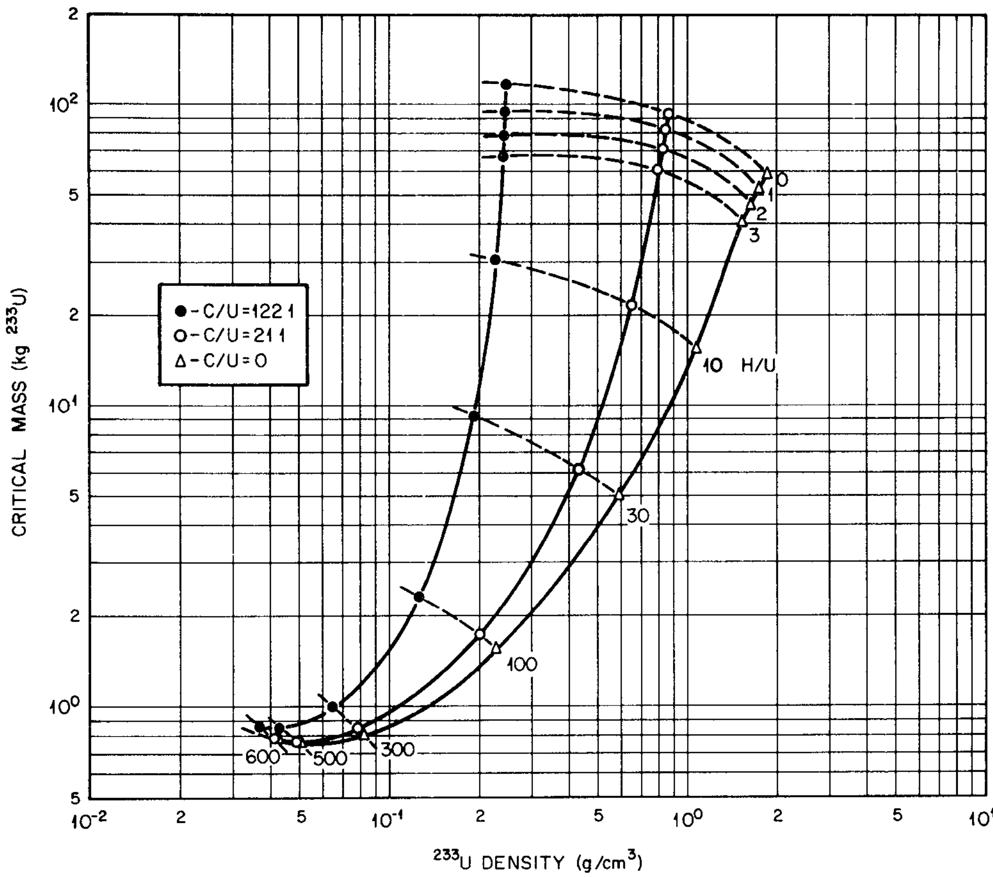

Fig. 3. Critical Masses of (4 Th:1 ^{233}U)O_2 in Spherical Geometry (water reflected) with Various H/U and C/U.

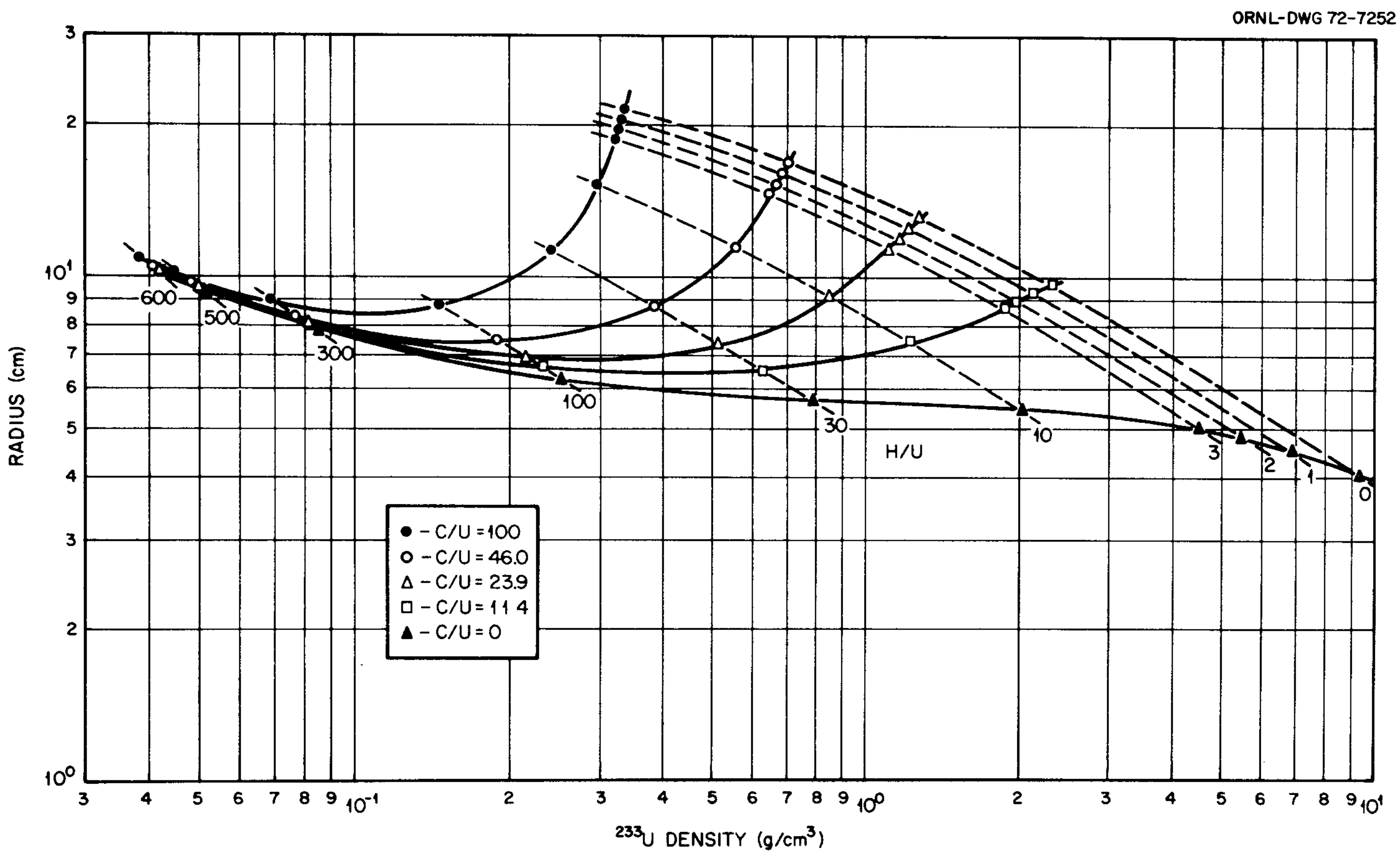

Fig. 4. Estimated Critical Radius of Infinite Cylinders of Water-Moderated $(Th,U)O_2$ (water reflected) with Various C/U-Th:U = 0:1.

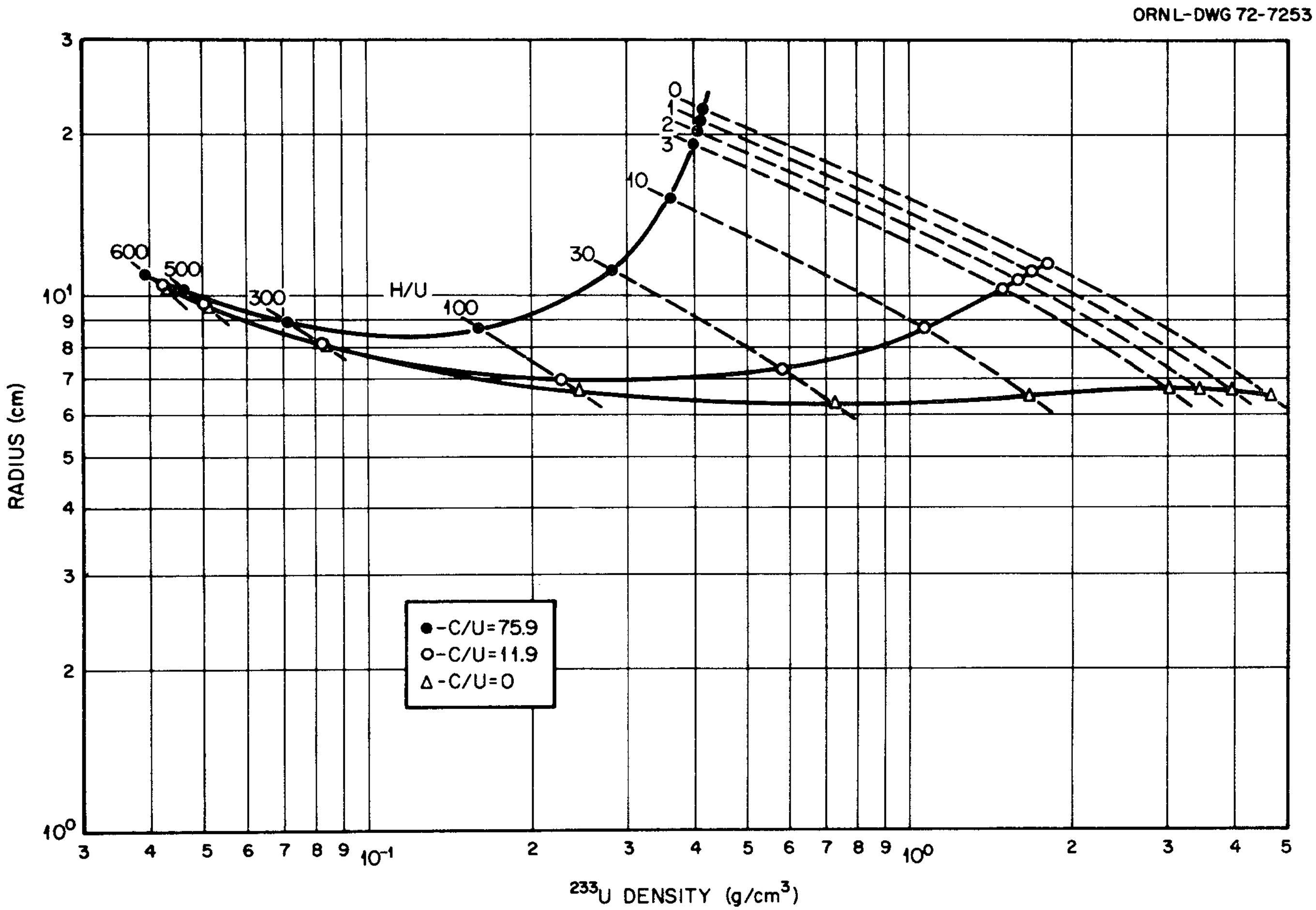

Fig. 5. Estimated Critical Radius of Infinite Cylinders of Water-Moderated $(Th,U)O_2$ (water reflected) with Various C/U-Th:U = 1:1.

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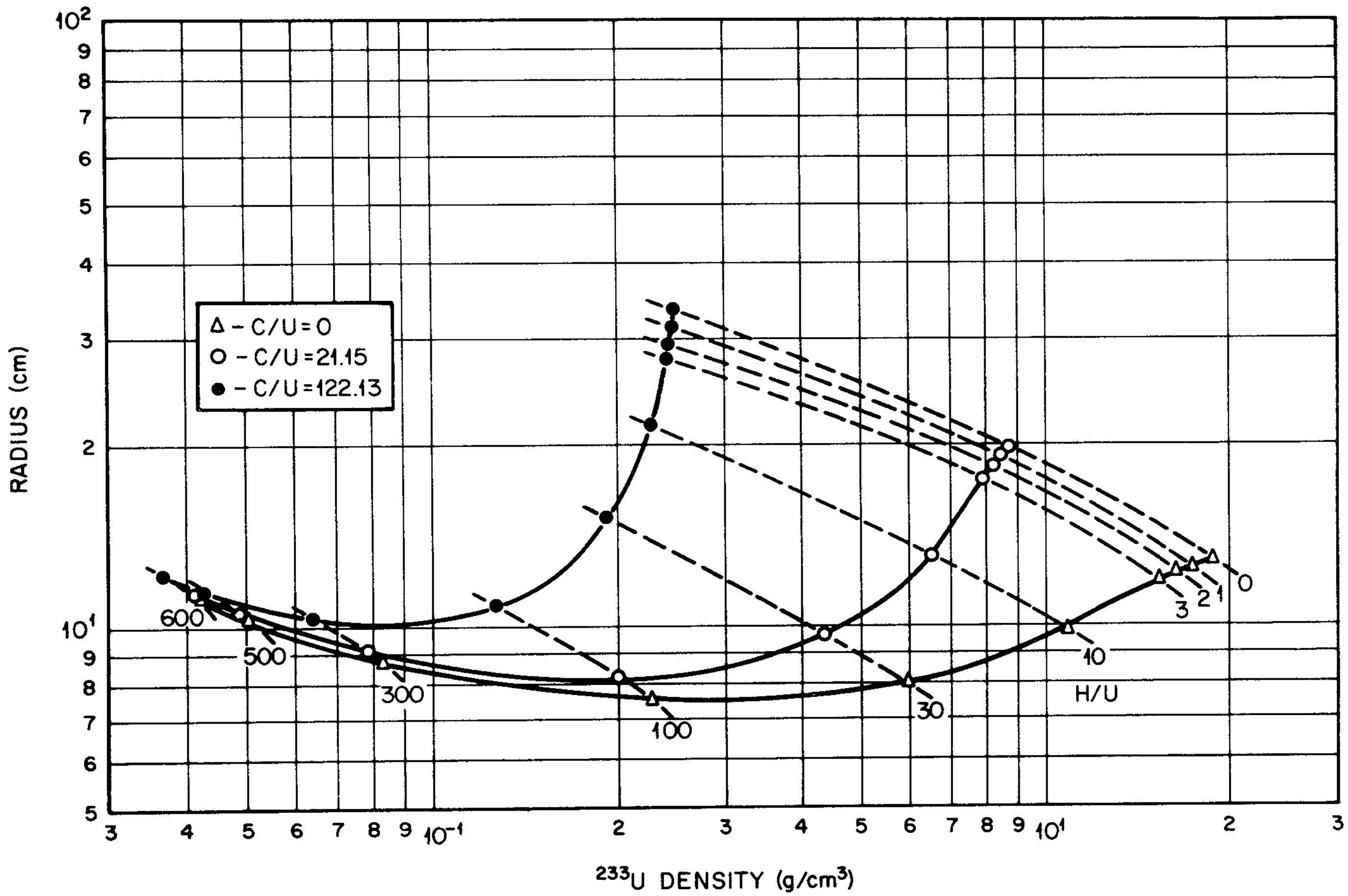

Fig. 6. Estimated Critical Radius of Infinite Cylinders of Water-Moderated $(Th,U)O_2$ (water reflected) with Various C/U-Th:U = 4:1.

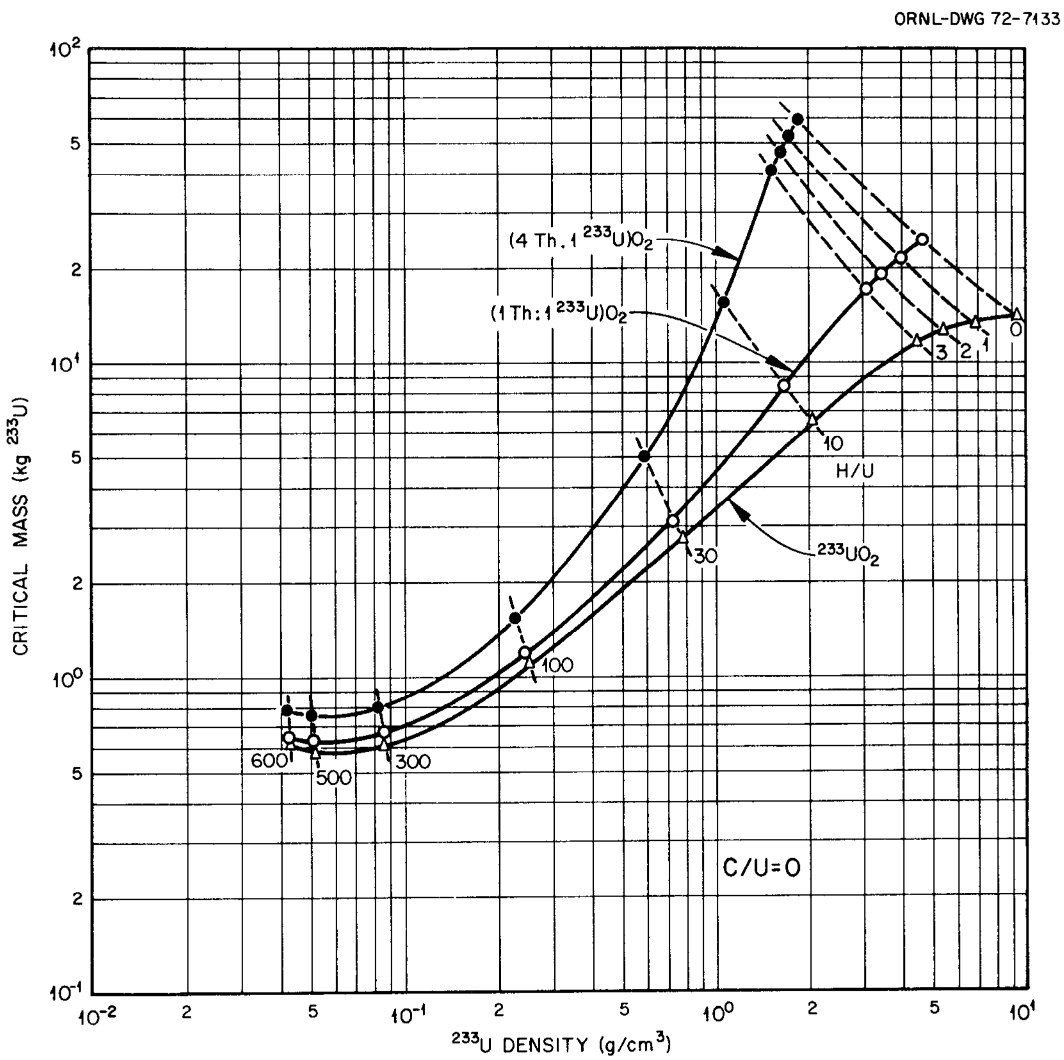

Fig. 7. Critical Masses of $^{233}UO_2$ in Spherical Geometry with Various Amounts of ThO_2 (water reflected).

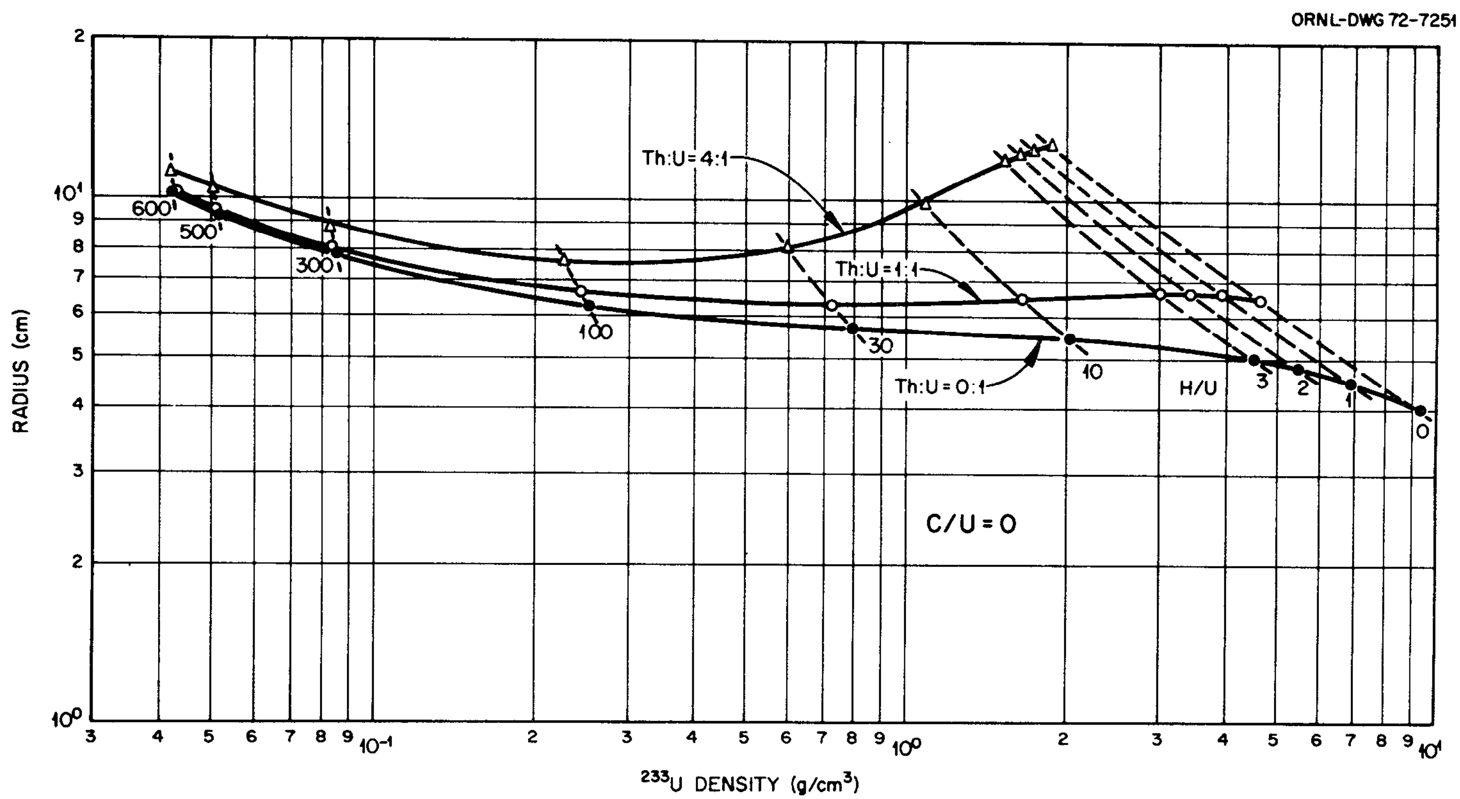

Fig. 8. Calculated Critical Radius of Infinite Cylinders of Water-Moderated $^{233}UO_2$ (water reflected) with Various Amounts of ThO_2.

HTGR COATER

The uranium and thorium oxide particles are coated with carbon in a remotely operated furnace in which methane gas is burned in a fluidized bed region. One of the prototype designs is shown in Fig. 9. This particular operation in the process was examined by calculation to determine loadings necessary to sustain criticality under normal and assumed accident conditions. Comparison of the loadings with the usually employed idealized reflected, spherical critical mass values provides a measure of potential economic gain to be had by this more realistic appraisal of criticality. Further, the dependence of the loadings on the diameter of the hopper section was examined to indicate criticality limitations for possible future designs.

The dimensions and materials of the coater considered in the ORNL Pilot Plant are given in Ref. 4. The configuration was described in the KENO code as shown in Fig. 9 with the exception of the disentrainment chamber and the details of the external structure below the base of the cylindrical section. While these would influence the resultant neutron multiplication factor for the system, their effect is a small fraction of β_{eff} and would be negligible compared to an adopted margin of safety for such an operation.

The inside diameter of the hopper is 12.7 cm and the outside dimensions of the cylindrical section are 30.48-cm-diam by 65.5 cm height. Examination of other furnace sizes consisted of a change in the diameter of the hopper and an increase in other diameters by the dimension of the initial annular regions, while maintaining the height of the system. The capacity of the 12.7-cm-diam hopper was 3.40 liters. Two other diameters used were 22.9 and 27.9 cm having respective capacities of 10.95 and 16.35 liters.

The accident conditions assume the hopper vessel is ruptured and its contents fill the annular v-trough section uniformly to a height consistent with the load and that, concurrently a water rupture floods

4. ORNL DWG 67-2691R1, Oak Ridge National Laboratory.

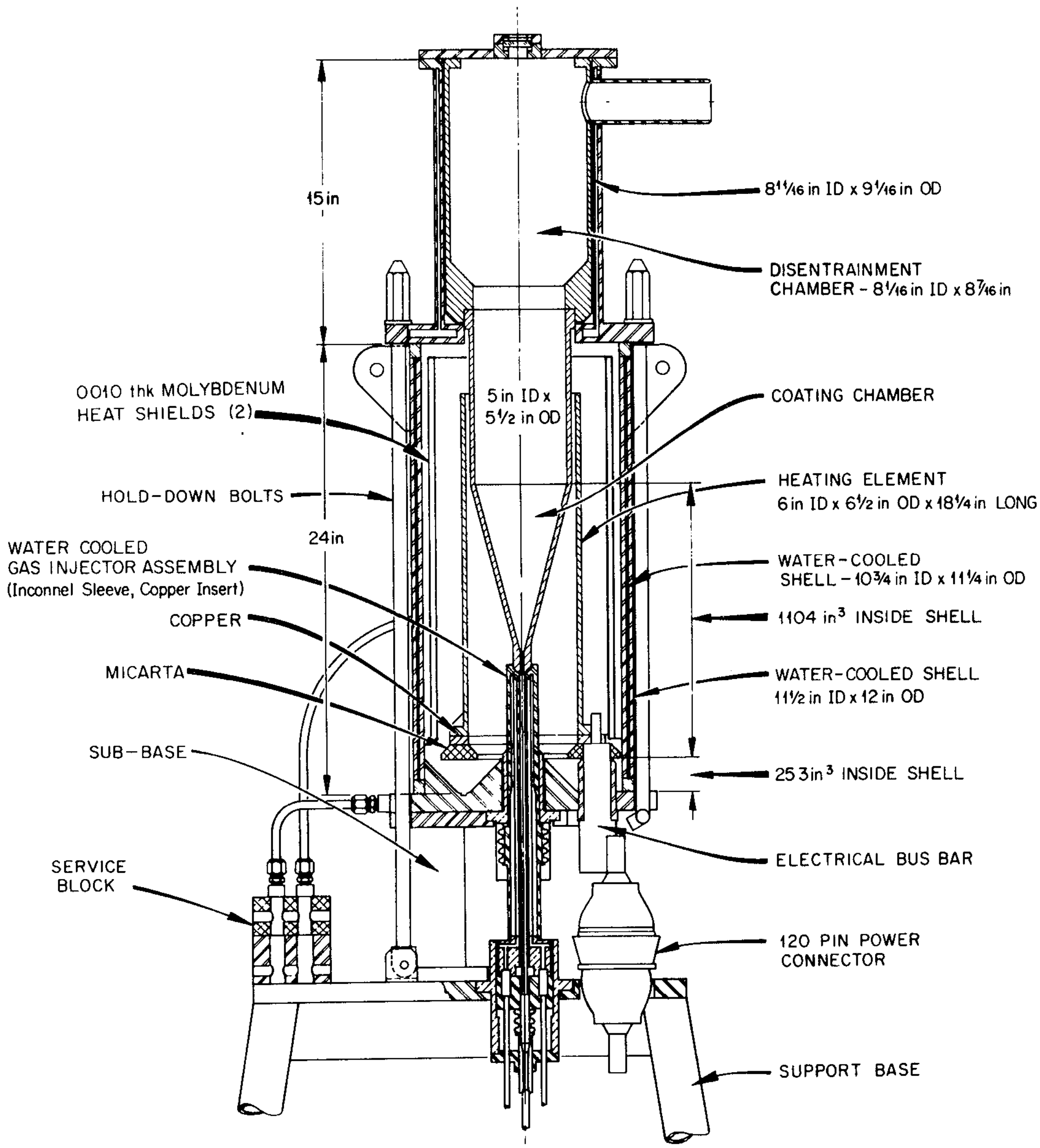

Fig. 9. Coater for HTGR Recycle Fuel

the region and extends above the fuel at least 15 cm. An exception to this top reflector condition was examined in the 12.7-cm-diam hopper. The calculations were performed for three ratios of 0:1, 1:1, and 4:1. The results are summarized in Table 2 where a description of the loading and the corresponding k_{eff} of the system are reported.

It is clear that the most reactive composition is the ^{233}U oxide in the absence of thorium or carbon which act principally as diluents. Confirmation of this effect is evident in the few calculations done for the normal operations of the coater as reported in Table 3 and for another accident condition with partially coated particles in Table 4.

Table 2. Summary of Monte Carlo calculations of Accident Conditions in the Fluidized Bed Coater.

Hopper i.d. (cm)	Atomic Ratios Th:U	H:U	^{233}U Density (g U/cm^3)	Load Volume (liters)	^{233}U Mass (kg)	k_{eff}
12.7	0:1	1.84	5.61	3.40	19.1	0.830
	0:1	1.84	5.61	3.40	19.1	0.692[a]
	0:1	30	0.79	3.40	2.86	0.724
	0:1	50	0.49	3.40	1.7	0.703
	1:1	3.68	2.81	3.40	9.6	0.480[a]
	4:1	9.22	1.12	3.40	3.8	0.315[a]
22.9	0:1	1.84	5.61	10.95	61.4	1.166
	0:1	1.84	5.61	8.89	49.9	1.098
	0:1	1.84	5.61	6.51	36.5	0.969
	0:1	1.84	5.61	6.92	38.8	0.995
	0:1	30	0.79	10.95	8.7	1.064
	0:1	50	0.49	10.95	5.4	1.061
	1:1	3.68	2.81	10.95	30.8	0.965
	4:1	9.22	1.12	10.95	12.3	0.746
27.9	0:1	1.84	5.61	16.35	91.7	1.311
	0:1	1.84	5.61	8.60	48.3	0.980
	0:1	1.84	5.61	8.87	49.8	0.998
	1:1	3.68	2.81	12.30	34.5	0.917
	1:1	3.68	2.81	14.52	40.8	0.999
	4:1	9.22	1.12	16.35	18.4	0.841
	4:1	9.22	1.12	16.35	18.4	0.841

a. Top water reflector absent.

Table 3. Summary of Monte Carlo Calculation of Normal Operation of the Fluidized Bed Coater.

Hopper i.d. (cm)	Atomic Ratios Th:U	H:U	^{233}U Density (g U/cm^3)	Load Volume (liters)	^{233}U Mass (kg)	k_{eff}
12.7	0:1	1.84	5.61	3.40	19.1	0.574
	1:1	3.68	2.81	3.40	9.6	0.340
	4:1	9.22	1.12	3.40	3.8	0.158
22.9	0:1	1.84	5.61	10.95	61.4	0.893
27.9	0:1	1.84	5.61	16.25	91.7	1.015

Table 4. Summary of Monte Carlo Calculations of Accident Conditions in Fluidized Coater with Partially Coated ^{233}U Oxide.

Accident Condition[a]	Hopper i.d. (cm)	Atomic Ratios C:U	H:U	^{233}U Density (g U/cm^3)	Load Volume (liters)	^{233}U Mass (kg)	k_{eff}
A	22.9	23.9	41	0.42	10.95	4.6	0.911
B		23.9	0	0.76	10.95	8.3	0.540
A	27.9	23.9	41	0.42	16.35	6.9	1.019
B		23.9	0	0.76	16.35	12.4	0.571

a. Accident Condition A is same as described for Table 2 in the text. Accident Condition B assumes water floods annular region and hopper is not ruptured.

CONCLUSIONS AND RECOMMENDATIONS

Although the calculations performed in this study have been limited and minimal, they are sufficient to support the nuclear criticality safety evaluations for the operation of the Oak Ridge National Laboratory pilot process plant. The critical parameters of related spherical and cylindrical geometries are basic and have general applicability to all stages of fuel fabrication. Addition of the elements thorium and carbon to the uranium generally cause an increase in the mass necessary to criticality. This behavior is characteristic of diluents[(5)] added to nonhydrogenous systems. Indeed, it is possible to estimate dilution exponents from the data presented and, thus, provide some guidance for criticality in regions not explored by calculations. The addition of water to such systems is to diminish the dilution effect.

The cursory investigation of the Fluidized Bed Coater exemplifies the benefits to be had from detailed calculations of particular process operations that may be restricted because of unknowledgeable criticality considerations. Removal of the geometric restriction in this operation would admit consideration of more favorable designs and, in turn, directly influence the economic analyses of these fuel mixtures by the future industry. A significant feature of allowing larger masses to be processed is the potential improvement in quality control of the fabricated fuel pieces.

Further basic and design studies are recommended. In the application of the data presented to possible criticality safety assessments, a suggested minimal margin of safety in k_{eff} is 5% corresponding to about 85% of the calculated ^{233}U critical masses.

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5. H. C. Paxton et al., "Critical Dimensions of Systems Containing U^{235}, Pu^{239}, and U^{233}," TID-7028, U.S. AEC (1964).

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