REFERENCE 63

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Homogeneous Critical Assemblies of 2 and 3% Uranium-235-Enriched Uranium in Paraffin

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A series of clean critical experiments has been performed with homogeneous mixtures of finely divided $U(2)F_4$ or $U(3)F_4$ dispersed in paraffin with $H/^{235}U$ atomic ratios varying from 133 to 972. The assemblies were constructed in rectangular geometry, and minimum critical masses and volumes in cylindrical and spherical geometries were obtained from buckling conversions. The minimum critical spherical volumes of a $U(2)F_4$-paraffin mixture when unreflected and when reflected with an effectively infinite solid hydrogenous material are 199 and 138 liters, respectively, and the minimum critical ^{235}U masses are 7.5 and 5.4 kg, respectively. The experiments with the $U(3)F_4$-paraffin mixtures were not extensive enough to determine experimentally the values at optimum moderation but transport theory calculations indicate that the unreflected and reflected minimum critical volumes are about 93 and 57 liters, respectively, and that the minimum critical masses are about 4.1 and 2.7 kg of ^{235}U, respectively.

Measurements of the ^{238}U-to-^{235}U fission ratio indicated that in some assemblies as much as 6.5% of the fissions occurred in ^{238}U. Prompt-neutron decay constants, $\alpha = \beta_{eff}/l$, were measured in reflected and unreflected delayed-critical assemblies of several fuel mixtures by the pulsed-neutron technique; the infinite-medium neutron multiplication factor, k_∞, for the various fuel mixtures was inferred from height perturbation measurements.

Transport theory calculations of k_∞ and k_{eff} have been made using various cross-section sets.

INTRODUCTION

Over the years there has been a continuing need for a variety of experimental criticality data that could be used for testing calculational models and methods and as a guide in establishing criticality safety criteria in the storage, handling, and processing of fissile materials. In contrast to the multiplicity of data that have been obtained on systems of highly ^{235}U-enriched uranium at H/U atomic ratios greater than 20, relatively little data have been collected on homogeneous critical systems of uranium of very low ^{235}U enrichment and of low H/U atomic ratios.

This paper summarizes work accomplished from 1958 to 1968 at the Oak Ridge Critical Experiments Facility (CEF) with critical systems of finely divided UF_4 dispersed in paraffin (nominally CH_2) to produce essentially homogeneous mixtures

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with H/U atomic ratios ranging from 4 to 20. The uranium in the fuel mixtures contained either 2 or 3 wt% ^{235}U. To obtain results that would be easily interpretable and unambiguously comparable to calculations, the experimental assemblies were made as simple as possible. The assemblies were self-supporting rectangular parallelepipeds and, except for a few of the earliest experiments, contained no internal controls or significant quantities of other foreign materials. The experiments included measurements of critical dimensions for each fuel mixture both unreflected and reflected with hydrogenous material, flux distributions from which extrapolation distances were determined, cadmium ratios, fission ratios, and of prompt-neutron decay constants. Also, the infinite-medium neutron multiplication factor, k_∞, of each material was obtained from height perturbation measurements.

During the decade that experiments were in progress, many of the data were published in progress reports or as more formal descriptions of particular segments of the program.[1]

MATERIALS

The fuel in these experiments was produced in batches by blending powdered UF_4 in molten paraffin ($C_{25}H_{52}$) and then pouring the mixture into a thin layer on a plastic sheet moving across a chilling table. After the mixture of UF_4 and paraffin had solidified it was broken into small pieces, cooled with dry ice, and ground into a fine powder. Each batch of powder was then homogenized in a rolling drum, sampled, and analyzed for uranium content, impurities, and homogenity. If necessary, the material was remixed or cross blended or both until the uranium content differed from the desired value by less than 1%. The material was then pressed into rectangular parallelepipeds with dimensions varying from 0.63 × 2.54 × 2.54 cm to 10.16 cm cubes and covered with 0.002-cm-thick aluminum foil. Some of the physical properties of each of the mixtures are summarized in Table I and in detail in Ref. 2. The maximum dimension of the UF_4 particles in the mixtures was <297 μm in the 2% enriched fuel and <40 μm in the 3% enriched fuel. The fuel contained insignificant neutron absorbing impurities.

Three reflector materials were used in these experiments. Plexiglas, a methyl methacrylate plastic ($C_5O_2H_8$) with a density of 1.18 g/cm^3, was employed for the bottom reflector. Paraffin ($C_{25}H_{52}$) with a density of 0.93 g/cm^3 was the side and top reflector material in early experiments. In later experiments, polyethylene (CH_2, ρ = 0.92 g/cm^3) was the top and side reflector material.

EXPERIMENTAL RESULTS

Delayed-critical systems of UF_4-paraffin mixtures, both unreflected and reflected with effectively infinite hydrogenous material, were arranged on an apparatus comprised of two tables in the manner shown in Fig. 1 and described by Rohrer et al.[3] Closure of the sections was by remotely controlled horizontal motion. The Critical Experiments Facility and equipment are further described in Ref. 4.

The assemblies were located so that the minimum distance between the surface of any assembly and the shielding walls was 2.7 m and the distance between the lower surface of any assembly and the floor was 1.0 m. Experiments have shown that the effect of neutrons reflected from the floor and walls was negligible. To reduce the effect on the normally unreflected assemblies of neutrons reflected by the apparatus, a five-layer honeycomb of 7.62- × 7.62-cm-square, 91.4-cm-long tubes, made of 0.119-cm-thick Type 1100 aluminum, was placed between the assemblies and the top of the steel table. The assemblies were stacked directly on top of the honeycomb structure except those with mixture U(2)F_4-6. This mixture was assembled on a 0.318-cm-thick sheet of aluminum which was on top of the honeycomb and provided a flatter surface on which to stack the fuel blocks. The effect of neutron reflection by this support structure including the table top was determined by measuring the reactivity worth of a duplicate placed on top of near-critical assemblies. The dimensions of a critical unreflected assembly were then obtained by adding an increment of fuel height, equivalent in reactivity to the support

[1] J. T. MIHALCZO et al., "Preliminary Report on 2% U^{235}-Enriched UF_4-C_{25}-H_{52} Critical Assemblies," ORNL-CF-59-4-120, Oak Ridge National Laboratory (1959); J. T. MIHALCZO, *Trans. Am. Nucl. Soc.*, **3**, 74 (1960); J. T. MIHALCZO, *Trans. Am. Nucl. Soc.*, **5**, 387 (1962); J. T. MIHALCZO and S. J. RAFFETY, *Trans. Am. Nucl. Soc.*, **11**, 676 (1968); also see the following Annual Progress Reports of the Oak Ridge National Laboratory Neutron Physics Division: ORNL-3193, p. 131 (1961); ORNL-3360, p. 37 (1962); ORNL-3499, p. 62 (1963); ORNL-4234, p. 28 (1967); ORNL-4280, p. 51 (1968).

[2] S. J. RAFFETY and J. T. MIHALCZO, "Homogeneous Critical Assemblies of 2 and 3% Enriched Uranium in Paraffin," Y-DR-14, Union Carbide Corporation, Oak Ridge Y-12 Plant (1969).

[3] E. R. ROHRER et al., *Neutron Phys. Div. Ann. Prog. Rept. Sept. 1, 1961*, ORNL-3193, p. 168, Oak Ridge National Laboratory (1961).

[4] "A Safety Analysis of the Oak Ridge Critical Experiments Facility," ORNL-TM-349, Rev. 1, Oak Ridge National Laboratory (1967).

TABLE I

Physical Properties, Critical Buckling, Extrapolation Distances, Reflector Savings, and Prompt-Neutron Decay Constants for UF_4-Paraffin Mixtures

Mixture No. and Enrichment[a]	Density (g/cm^3)		Weight Fraction of UF_4 (%)	$H/^{235}U$ Atomic Ratio	Ratio of Average Assembly Density to UF_4-CH_2 Density[b]	Critical Buckling (10^{-3} cm^{-2})	Extrapolation Distance (cm)	Reflector Savings[c] (cm)	Prompt-Neutron Decay Constant at Delayed Criticality (sec^{-1})	
	UF_4-CH_2	Uranium							Unreflected	Reflected
U(2)F_4-1	4.490	3.135	92.13	195.2	0.9835	4.393 ± 0.059	2.49 ± 0.21	5.48 ± 0.06	273 ± 3	---
U(2)F_4-2	3.930	2.639	88.61	293.9	0.9835	5.892 ± 0.118	2.43 ± 0.23[d]	4.77 ± 0.11	---	---
U(2)F_4-3	3.450	2.220	84.91	406.3	0.9835	6.568 ± 0.122	2.38 ± 0.24	4.28 ± 0.10	---	---
U(2)F_4-4	3.160	1.968	82.17	495.9	0.9835	6.651 ± 0.116	2.16 ± 0.22	4.02 ± 0.07	---	---
U(2)F_4-5	2.883	1.723	78.84	613.6	0.9817	6.205 ± 0.073	2.21 ± 0.11	3.81 ± 0.16	139 ± 2	137 ± 2
U(2)F_4-6	2.302	1.223	70.17	971.7	0.9930	3.616 ± 0.030	2.59 ± 0.11	3.45 ± 0.08	105 ± 2	107 ± 2
U(3)F_4-1	4.462	3.110	91.95	133.4	0.9826	6.516 ± 0.051	2.15 ± 0.07	5.85 ± 0.07	343 ± 3	258 ± 2
U(3)F_4-2	3.443	2.208	84.63	276.9	0.9843	10.207 ± 0.122	2.31 ± 0.07	4.44 ± 0.16	242 ± 2	208 ± 2

[a]The ^{235}U enrichment of the uranium, expressed in weight percent, as indicated by the number in parentheses is 2.00 ± 0.02 for the U(2)F_4 material and 3.00 ± 0.02 for the U(3)F_4 material.

[b]The density listed in Column 2 is of the fuel material as pressed in the individual blocks. Because of the aluminum foil covering the fuel, irregularities in fuel block dimensions, and small voids introduced in stacking the fuel blocks, the tabulated densities must be reduced by these factors to obtain average densities of the experimental assemblies. All results given in this report correspond to these reduced densities.

[c]The reflector was 15.2-cm-thick methacrylate plastic (Plexiglas) on the bottom of all assemblies. The top and sides of assemblies of mixtures U(2)F_4-1, -2, -3, and -4, and U(3)F_4-1 were reflected with 15.2-cm-thick paraffin; the top and sides of assemblies of mixtures U(2)F_4-5 and -6 and U(3)F_4-2 with 15.2-cm-thick polyethylene.

[d]Since the extrapolation distance was not measured for Mixture U(2)F_4-2, this value is estimated from the values for Mixtures U(2)F_4-1 and -3.

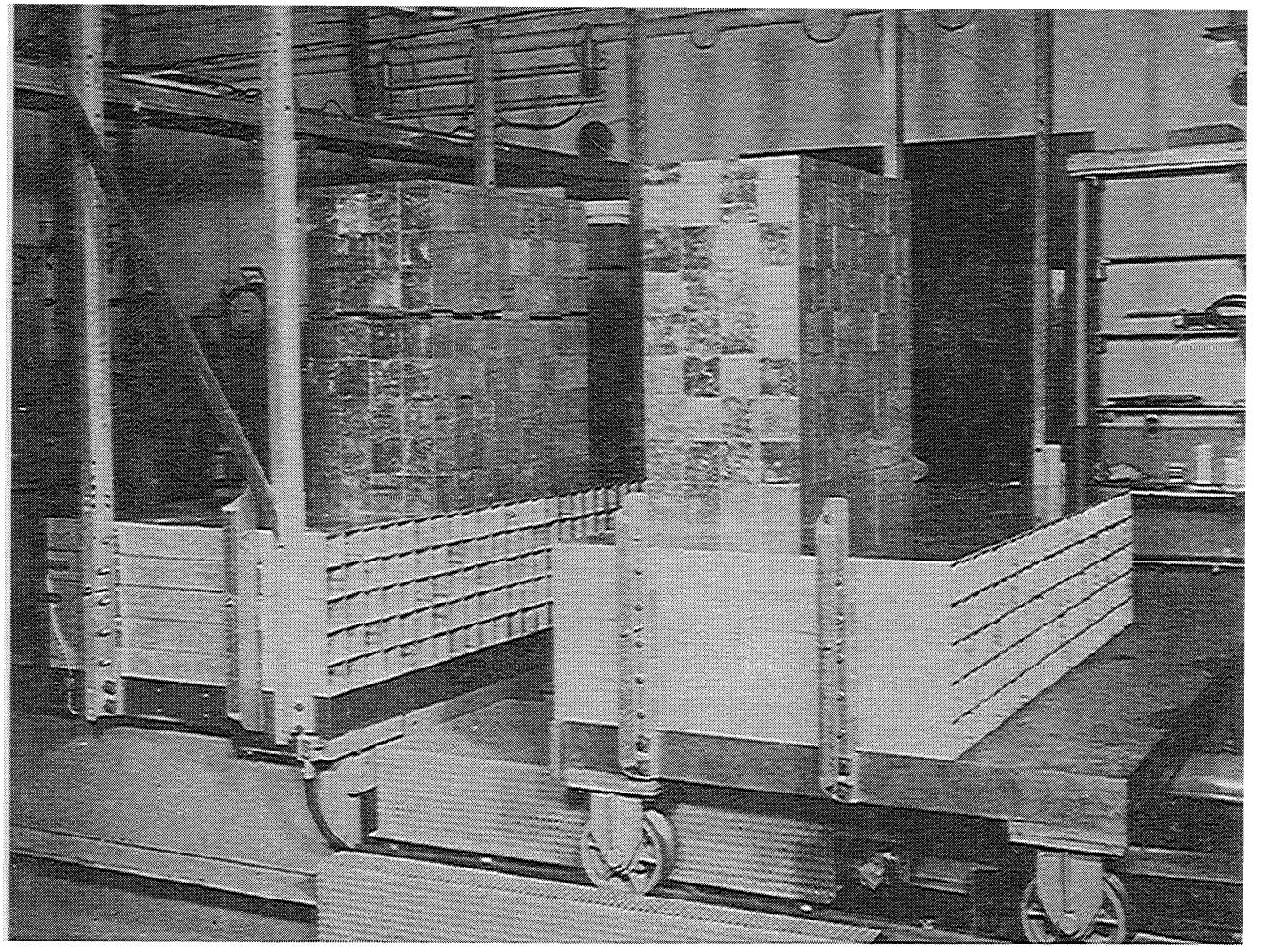

Fig. 1. An experimental assembly on the remotely operated criticality testing equipment.

structure, to the dimensions of the critical assembly mounted on the table. The critical dimensions of the assemblies are given in Table II.

In all experiments with reflected assemblies, the bottom reflector consisted of Plexiglas pieces inserted into the top two layers of the aluminum tubes in the support structure resulting in a 15.2-cm-thick reflector with a Plexiglas volume fraction of 0.918 and an aluminum volume fraction of 0.062. The sides and top of assemblies of fuel mixtures $U(2)F_4$-1 through -4 and $U(3)F_4$-1 were reflected with a 15.2-cm-thick layer of paraffin; those of mixtures $U(2)F_4$-5 and -6 and $U(3)F_4$-2 were reflected with a 15.2-cm-thick layer of polyethylene. An experiment with mixture $U(2)F_4$-5 indicated that the difference in reactivity worth between 15.2 cm of paraffin and 15.2 cm of polyethylene on top of a near cubic assembly was only 0.7 cents, and an experiment with mixture $U(2)F_4$-1 indicated that 15.2 cm of Plexiglas on top of a near cubic assembly was worth only about 5 cents more than 15.2 cm of paraffin. It was therefore assumed that, within experimental errors, the three reflector materials were equivalent. Other measurements with mixture $U(2)F_4$-3 have shown that these materials, for thicknesses >15 cm, have the same reflection effects as water.

Extrapolation distances were determined for the various fuel mixtures by flux mapping. The relative neutron flux at points on a horizontal line across the center of critical assemblies was measured either by foil activations or by fission chamber traverses. The detectors included foils of $U(2)F_4$ homogeneously mixed in paraffin, gold and cadmium-covered gold, and ^{235}U and ^{238}U fission chambers. Several irradiations with each type of detector were made in a given assembly and the data were then fitted to a cosine function by least-squares analysis. A three-parameter fit determined the buckling, the amplitude, and the

TABLE II

Critical Dimensions of Reflected* and Unreflected Rectangular Parallelepipedal Assemblies of UF_4-Paraffin Mixtures and the Effect of Support Structure on Unreflected Experimental Assemblies

Critical Dimensions[a]		Worth of Support Structures of Unreflected Assemblies (cm of fuel height)
Reflected (cm)	Unreflected (cm)	
Mixture $U(2)F_4$-1		
56.22 × 56.22 × 112.88	71.47 × 71.47 × 94.14	0.89
61.33 × 61.33 × 79.46	76.65 × 76.65 × 78.08	0.77
71.55 × 71.55 × 58.03	81.75 × 86.75 × 66.71	0.63
74.11 × 74.11 × 54.71		
Mixture $U(2)F_4$-2		
51.11 × 51.11 × 73.87	56.22 × 56.22 × 122.47	
61.33 × 61.33 × 49.12	61.33 × 61.33 × 79.60	b
71.55 × 71.55 × 40.70	71.55 × 71.55 × 57.92	
	92.00 × 92.00 × 46.18	
Mixture $U(2)F_4$-3		
56.22 × 56.22 × 49.54	45.97 × 61.21 × 144.5	2.64
53.67 × 53.67 × 54.29	51.07 × 56.13 × 110.5	0.85
	51.07 × 61.21 × 88.27	0.60
	61.21 × 61.21 × 64.85	0.46
	61.21 × 71.57 × 56.68	0.61
Mixture $U(2)F_4$-4		
46.00 × 46.00 × 96.57	45.99 × 61.3 × 144.83	2.37
51.11 × 51.11 × 62.97	51.1 × 56.23 × 112.09	1.11
61.33 × 61.33 × 44.53	51.1 × 61.3 × 88.74	0.59
	56.23 × 61.3 × 72.38	0.63
	61.3 × 61.3 × 64.64	0.42
	61.3 × 66.5 × 59.68	0.50
Mixture $U(2)F_4$-5		
56.32 × 61.29 × 54.08	53.80 × 56.30 × 126.98	0.42
	61.30 × 66.54 × 66.52	0.43
	102.28 × 112.64 × 41.59	0.59
Mixture $U(2)F_4$-6		
76.51 × 76.44 × 82.42	76.50 × 76.45 × 119.45	0.58
76.43 × 81.53 × 77.66	81.45 × 86.70 × 88.22	0.52
81.52 × 81.60 × 73.02	101.97 × 101.85 × 66.64	0.65
Mixture $U(3)F_4$-1		
43.47 × 43.47 × 86.39	56.47 × 56.47 × 86.64	0.83
46.02 × 46.02 × 67.57	56.25 × 61.36 × 74.38	0.76
51.14 × 51.14 × 51.27	61.4 × 61.4 × 66.00	0.65
56.25 × 56.25 × 43.41		
61.36 × 61.36 × 38.67		
Mixture $U(3)F_4$-2		
40.81 × 40.80 × 39.49	40.90 × 40.93 × 116.80	0.37
	48.59 × 51.14 × 48.53	0.42
	81.71 × 81.66 × 31.34	0.53

*The bottom reflector for all mixtures was 15.2 cm of methacrylate plastic (Plexiglas) while the top and sides of mixtures $U(2)F_4$-1, -2, -3, and -4 and $U(3)F_4$-1 were reflected with 15.2 cm of paraffin, and mixtures $U(2)F_4$-5 and -6 and $U(3)F_4$-2 with 15.2 cm of polyethylene.

[a]The dimensions given have been adjusted to account for control rod guides if present and for effect of support structure. The first two numbers are the horizontal dimensions of the core with an estimated accuracy of ±0.10 cm. The third number is the height with an estimated accuracy of ±0.40 cm for the unreflected assemblies of the second listed mixture and ±0.20 cm for all other cases.

best center of the relative flux distribution in the assembly. To define a distribution free from higher spatial modes, data near the boundaries of the assembly were not used. From the bucklings thus determined and the actual measured dimensions of the assemblies, the extrapolation distances were obtained. The extrapolation distances for the different fuel mixtures are given in Table I and are plotted as a function of the $H/^{235}U$ ratio in Fig. 2. Since the error in the measurements was between 3 and 14% and since no definite correlation was found between the magnitude of the extrapolation distance and the dimension of the assembly or the type of detector, the extrapolation distance for a given mixture is the average of the values measured in that mixture. In systems of the size considered here, the buckling was not very dependent on the extrapolation distance, and critical mass calculations by buckling conversions from one geometry to another were affected even

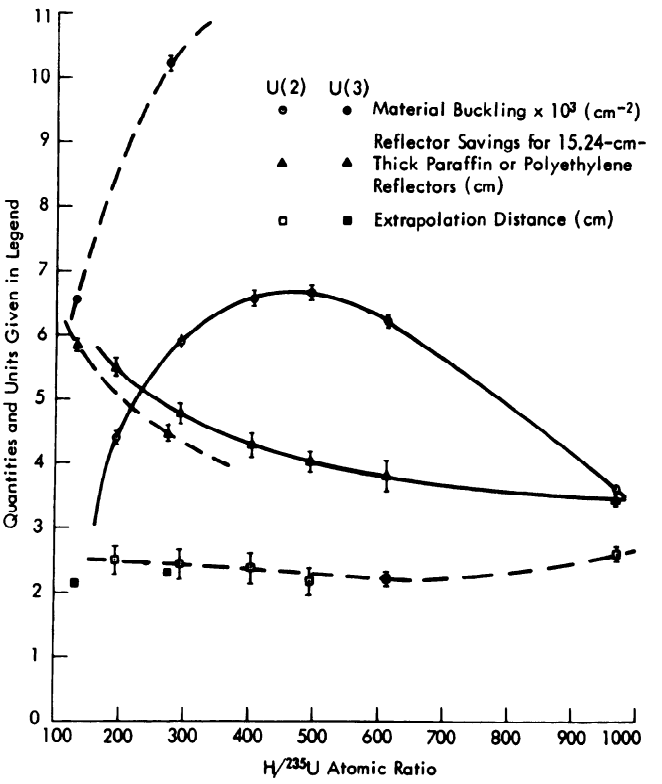

Fig. 2. Material buckling, reflector savings, and extrapolation distance for 2 and 3% ^{235}U-enriched UF_4-paraffin mixtures as a function of the $H/^{235}U$ atomic ratio.

[b]The effect of the support structure was not measured for the second mixture. However, experiments with mixtures $U(2)F_4$-1 and -3 indicated that the effect of the support structure was approximately canceled by the effect of the rod guides so neither correction was made.

less since errors due to extrapolation distance uncertainties tend to cancel out.

These average values of the extrapolation distance were used in the usual equation[5] to calculate the geometric buckling of a number of unreflected critical assemblies. The material buckling of each fuel mixture, which was assumed to be the average of the geometric bucklings calculated for the experimental critical assemblies of that mixture, is given in Table I with error limits produced from the uncertainties in the extrapolation distances and in the critical dimensions. The dependence of buckling on the $H/^{235}U$ atomic ratio of the fuel mixtures is shown in Fig. 2. If it is assumed that the buckling and the extrapolation distance of critical assemblies of a given material are independent of geometry, the buckling relations may be used to convert experimental critical data to the minimum critical volumes and masses which are given in Figs. 3 and 4 for unreflected

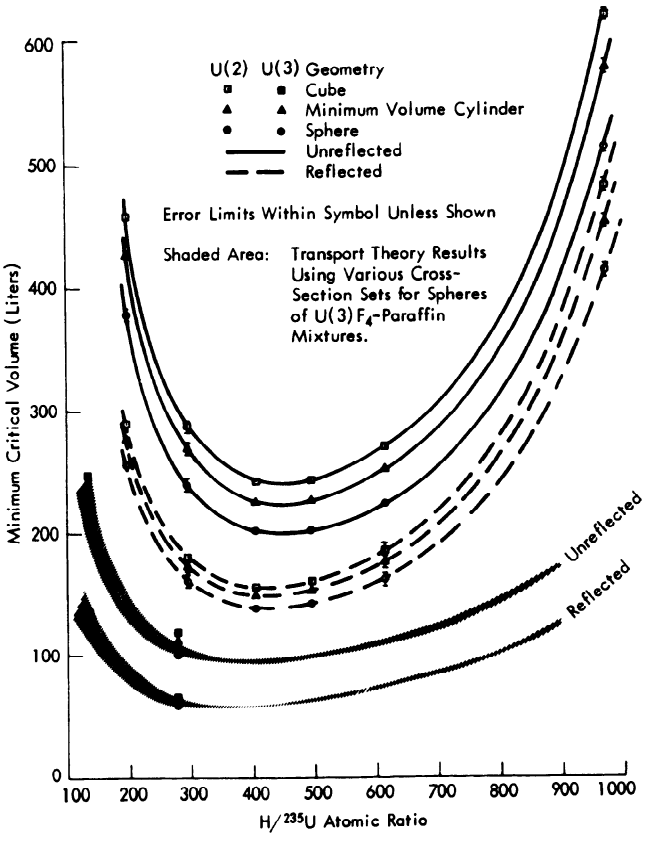

Fig. 3. Critical volume of 2 and 3% ^{235}U-enriched UF_4-paraffin mixtures as a function of the $H/^{235}U$ atomic ratio. The values plotted were obtained from experimental data by buckling conversions.

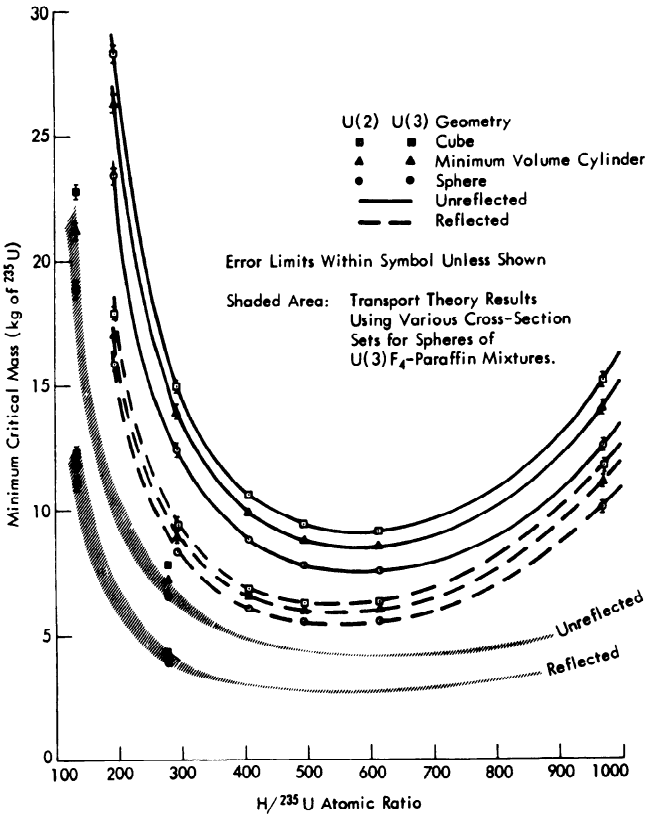

Fig. 4. Critical mass of 2 and 3% ^{235}U-enriched UF_4-paraffin mixtures as a function of the $H/^{235}U$ atomic ratio.

rectangular, cylindrical, and spherical geometries.

The reflector savings, λ_r, was determined for several reflected experimental critical systems from a buckling equation of the form $B^2 = \pi^2[T + 2(\lambda_0 + \lambda_r)]^{-2}$, where the buckling, B^2, and the extrapolation distance, λ_0, had the same values as for the unreflected assemblies. The side lengths, T, were the dimensions of reflected experimental assemblies. The average value of reflector savings found for each fuel mixture is reported in Table I and is plotted as a function of $H/^{235}U$ atomic ratio in Fig. 2. These values of reflector savings were then used to calculate the minimum critical volumes and masses for rectangular, cylindrical, and spherical systems reflected by 15.2 cm of paraffin or polyethylene. These results are also shown in Figs. 3 and 4.

The change in the neutron spectrum in critical assemblies as the $H/^{235}U$ atomic ratio changes was investigated by measuring cadmium ratios and ^{238}U-to-^{235}U fission ratios in the center of the core. Cadmium ratios, defined as the ratio of the activity produced in a bare foil to that produced in an identical cadmium-covered foil in the same

[5]SAMUEL GLASSTONE and MILTON C. EDLUND, *The Elements of Nuclear Reactor Theory*, D. Van Nostrand Company, Inc., Princeton, New Jersey (1952).

neutron flux, were measured with 0.0025-, 0.0051-, and 0.0127-cm-thick gold foils, with 0.0254-cm-thick 9.3 wt% In-Al alloy foils, with 0.0127-cm-thick 10 wt% U(93)-Al alloy foils, and with 0.0102-cm-thick U(93) metal foils. For some assemblies it was verified experimentally that these ratios did not depend on position except near the core boundaries. The results of these measurements are plotted in Fig. 5 as a function of $H/^{235}U$ atomic ratio.

The ratio of the ^{238}U-to-^{235}U fission rates was measured in the critical assemblies by fission chambers or foil activations or both. In the foil measurements the detectors consisted of 98 wt% UF_4 and 2 wt% paraffin in 1.27-cm-diam by 0.155-cm-thick foils. The uranium of some foils, the "enriched foils," contained 2 wt% ^{235}U and that in the "depleted foils" contained 0.184 wt% ^{235}U with the remaining uranium considered to be ^{238}U. The enriched and depleted foils were irradiated in pairs in symmetrical positions about the center of a critical assembly and the activation due to fission product formation subsequently determined.

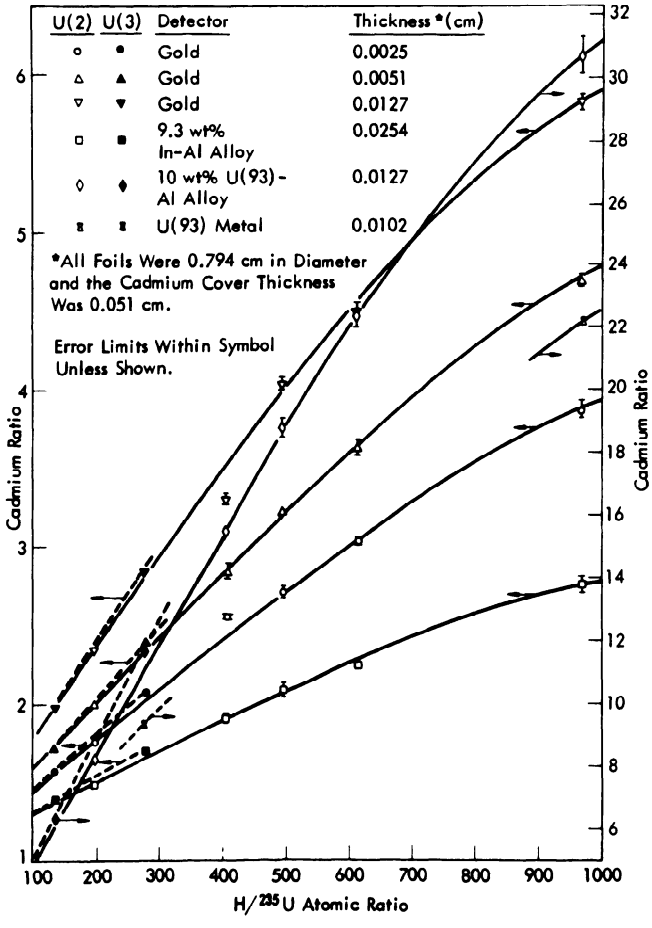

Fig. 5. Cadmium ratios in critical assemblies of UF_4-paraffin mixtures.

The chambers for the fission ratio measurements were 2.54 cm long and 0.635 cm in diameter. One of the chambers contained 0.3471 mg (101 $\mu g/cm^2$) of uranium enriched to 97.34% ^{235}U and the other contained 0.3989 mg (120 $\mu g/cm^2$) of uranium with about 7 parts of ^{235}U per million parts of ^{238}U. The two chambers were placed in symmetrical flux positions in the assemblies and data were obtained from them simultaneously. Since the chambers were identical except for the ^{235}U loading, it was assumed that their pulse-height distributions would be the same and therefore the efficiency for fission detection would be the same if the voltages and the amplifiers for the two channels were identical. To check on the amplifier balance and to correct for small differences in the amplifiers and possible small asymmetries in the neutron flux, data were taken with the detectors in each of the four possible combinations of position and amplifier arrangement and the average of the four counting ratios was used to calculate the fission ratio.

The assumptions made concerning the similarity of the two fission chambers and their efficiency were verified experimentally in experiments with the $U(2)F_4$-6 fuel mixture. A 256-channel pulse-height analyzer recorded the pulse-height spectrum from each chamber and a linear extrapolation to zero pulse height was made to obtain a correction for the small fission pulses that were not recorded. This correction was <12% of the recorded counts in all cases and the percentage correction was determined to an accuracy of about 5%. The correction for fission counts lost because fission fragments do not escape from the uranium deposit was <0.7% for the thicknesses in these chambers.[6] It was negligible in the fission ratio because the platings are nearly the same in the two chambers. The ^{238}U-to-^{235}U fission ratio in the counters thus obtained was $(4.74 \pm 0.10) \times 10^{-4}$. Since this value was only 3.5% less than the value obtained $[(4.91 \pm 0.27) \times 10^{-4}]$, assuming the detectors and counting systems were identical and within the error limits quoted, the validity of the assumption was verified. These measurements with foils and counters were interpreted to obtain the ratio of ^{238}U fission-to-^{235}U fission and the fast fission factor for these assemblies. The results are given in Table III and are shown in Fig. 6.

The prompt-neutron decay constant, $\alpha = \beta_{eff}/l$, was measured in reflected and unreflected delayed-critical assemblies of several fuel

[6] FREDERICK S. KIRN, "Neutron Detection with an Absolute Fission Counter," IAEA Symposium on Neutron Detection, Dosimetry and Standardization, Harwell, England (1962).

TABLE III

Parameters for and Results of Calculations of Neutron Ages and Infinite-Medium Neutron Multiplication Factors for UF_4-Paraffin Mixtures with 2 and 3% ^{235}U Enrichment

		Mixture No.							
		U(2)F_4-1	U(2)F_4-2	U(2)F_4-3	U(2)F_4-4	U(2)F_4-5	U(2)F_4-6	U(3)F_4-1	U(3)F_4-2
S,	ratio of ^{238}U fissions to ^{235}U fissions in fuel (10^{-2})	6.48 ± 0.34	5.20 ± 0.28	4.35 ± 0.23	3.87 ± 0.20	3.41 ± 0.18	2.42 ± 0.13	6.91 ± 0.36	4.29 ± 0.22
$\bar{\alpha}^{28}$	average neutron capture-to-fission ratio in ^{238}U above the fission threshold energy	0.147 ± 0.040	0.143 ± 0.040	0.141 ± 0.040	0.140 ± 0.040	0.139 ± 0.040	0.137 ± 0.040	0.146 ± 0.040	0.141 ± 0.040
$\bar{\beta}$,	fraction of fission neutrons that are delayed (10^{-3})	7.06 ± 0.31	6.96 ± 0.29	6.89 ± 0.28	6.85 ± 0.27	6.81 ± 0.26	6.72 ± 0.24	7.10 ± 0.31	6.88 ± 0.28
ϵ,	fast fission factor[a]	1.042 ± 0.007	1.034 ± 0.006	1.029 ± 0.005	1.026 ± 0.004	1.022 ± 0.004	1.016 ± 0.002	1.045 ± 0.008	1.028 ± 0.005
L^2,	thermal-neutron diffusion area (cm^2)	1.69 ± 0.08	1.67 ± 0.08	1.72 ± 0.09	1.79 ± 0.09	1.86 ± 0.09	2.11 ± 0.11	1.15 ± 0.06	1.22 ± 0.06
$\frac{\Delta\rho}{\Delta B^2}$,	change in reactivity per unit change in buckling (cm^2)	-36.80 ± 0.52	[b]	-26.69 ± 0.37	-24.85 ± 0.35	-23.65 ± 0.33	-24.05 ± 0.34	-32.89 ± 0.46	-23.07 ± 0.32
τ_d,	average delayed-neutron age-to-thermal energy[c] (cm^2)	27.0 (27.5)	21.5 (21.9)	18.6 (19.1)	17.3 (17.8)	16.0 (16.8)	14.5 (15.1)	25.7 (26.2)	17.5 (18.1)
τ_p,	prompt-fission-neutron age-to-thermal energy (cm^2)								
	inferred from height perturbation measurements[d]	42.6 ± 3.2	[b]	31.8 ± 2.5	28.8 ± 2.0	26.4 ± 1.8	24.4 ± 1.6	42.8 ± 4.0	32.0 ± 3.1
	from transport calculation[c]	45.5 (46.1)	37.9 (38.6)	33.9 (34.6)	32.0 (32.7)	30.1 (30.9)	27.5 (28.3)	44.1 (44.7)	32.6 (33.2)
k_∞,	infinite-medium neutron multiplication factor								
	inferred from height perturbation measurements[d]	1.196 ± 0.014[e]	[b]	1.222 ± 0.018	1.205 ± 0.014	1.177 ± 0.011	1.096 ± 0.006	1.290 ± 0.027	1.343 ± 0.032
	from transport calculation[f]	1.204 (1.147)	1.229 (1.189)	1.217 (1.190)	1.202 (1.188)	1.170 (1.169)	1.086 (1.091)	1.302 (1.243)	1.341 (1.312)

[a]The value of $\bar{\nu}^{25}$ is 2.43 ± 0.02. The value of ν^{28} for mixtures U(2)F_4-1 is 2.73 ± 0.12; for the other mixtures it is 2.74 ± 0.12.

[b]The parameters necessary for determining these quantities were not measured.

[c]These values are one-sixth of the second moment of the flux distribution of the 16th (thermal) group of an S_{32} transport theory calculation. The first number was obtained by reducing the original Hansen-Roach 16-group cross sections for ^{238}U in the resonance region according to a scheme developed by Knight.[9] The reduction factors were 0.74, 0.78, 0.83, 0.87, 0.93, and 1.00 for mixtures U(2)F_4-1 to -6, respectively, and 0.74 and 0.83 for mixtures U(3)F_4-1 and -2, respectively. The values in parenthesis were obtained by using the revised Hansen-Roach cross-section set.[10]

[d]The τ_d's derived from the calculations employing the cross-section modification by Knight were used to obtain these values.

[e]Since the value measured in the PCTR by Neeley[8] was 1.216 ± 0.013, the average measured value is 1.206 ± 0.010.

[f]The first value was obtained by using the Knight cross-section modification and the value in parenthesis was obtained from the revised Hansen-Roach cross sections.

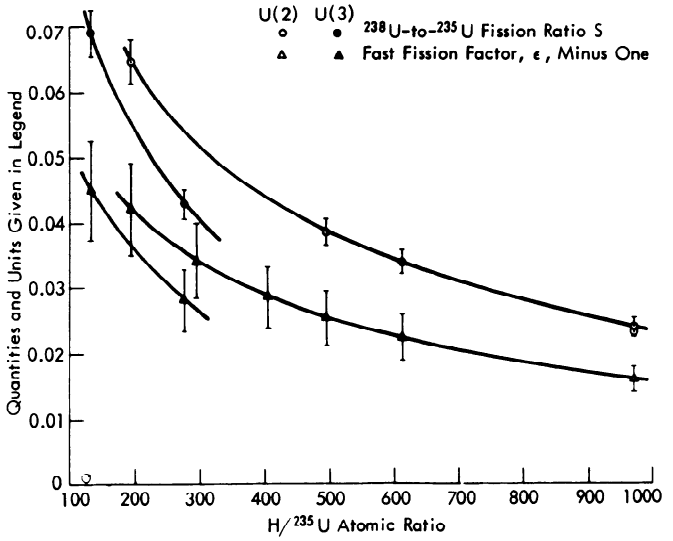

Fig. 6. ^{238}U-to-^{235}U fission ratio and fast fission factor in 2 and 3% ^{235}U-enriched UF_4-paraffin mixtures as a function of the $H/^{235}U$ atomic ratio.

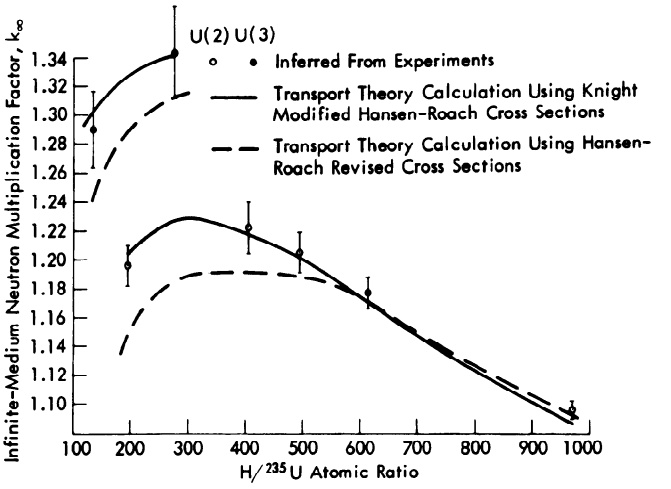

Fig. 7. The infinite-medium neutron multiplication factor of 2 and 3% ^{235}U-enriched UF_4-paraffin mixtures as a function of the $H/^{235}U$ atomic ratios.

mixtures by the pulsed-neutron technique. Pulses of neutrons for these experiments were produced by a 150-kV Cockcroft-Walton accelerator utilizing the (d, T) reaction. The detectors were BF_3 neutron counters in most cases, although some measurements were made with proton-recoil-type scintillators and NaI(Tl) scintillators.[7] Within experimental errors the results were independent of the detector. The data were recorded by a 256-channel time analyzer and the decay constant α was obtained from a nonlinear least-squares fit of these data to an equation of the form $y = B_1 + B_2 \exp(-\alpha T)$. The data were analyzed to determine that the decay was purely exponential and the decay constants are given in Table I.

The infinite-medium neutron multiplication factor for the various fuel mixtures was inferred from height perturbation measurements by a method previously described by Mihalczo[8] which agreed with the results of measurements in the Physical Constants Testing Reactor for the mixture $U(2)F_4$-1. The parameters used in the calculation of the infinite-medium multiplication factors and the results are given in Table III and the infinite-medium multiplication factor is plotted as a function of $H/^{235}U$ atomic ratio in Fig. 7.

TRANSPORT AND MONTE CARLO CALCULATIONS

The ANISN[11] transport theory code has been used to obtain calculated values of k_{eff} for the delayed-critical spheres of each fuel mixture. Calculations were performed with three different sets of cross sections; the revised Hansen-Roach[10] 16-group set, the Knight-modified[9] Hansen-Roach 16-group set, and the 18-group set of Bell et al.[10] All the calculations were performed using a quadrature order of four and a convergence criterion of 10^{-4}. The ^{238}U cross sections used in the resonance groups were selected by linear interpolation between tabulated values for various total scattering cross sections per ^{238}U atom. The ^{235}U cross sections for an infinitely dilute ^{235}U system were used in all cases. The 0.002-cm-thick aluminum foil on the fuel blocks was treated as a void in these calculations since a calculation with mixture $U(2)F_4$-1 indicated it was worth <0.01% in k_{eff}. Voids introduced in stacking the blocks were accounted for by reducing the homogeneous density of the materials by the density reduction factors listed in Table I. As can be seen from the results listed in Table IV, the calculations that utilized the

[7] J. T. MIHALCZO, *Trans. Am. Nucl. Soc.*, **5**, 387 (1962).

[8] J. T. MIHALCZO, "Comparison of k_∞ Measurements in a Critical Assembly with k_∞ Measurements in the Physical Constants Testing Reactor," ORNL-CF-60-4-24, Oak Ridge National Laboratory (1960); J. T. MIHALCZO and VICTOR I. NEELEY, *Nucl. Sci. Eng.*, **13**, 6 (1962).

[9] J. R. KNIGHT in Appendix I of Report K-1663, "Hydrogen Moderation—A Primary Nuclear Safety Control for Handling and Transporting Low-Enrichment UF_6," by C. E. NEWLON and A. J. MALLETT, Oak Ridge Gaseous Diffusion Plant (1966).

[10] G. I. BELL et al., "Los Alamos Group-Averaged Cross Sections," LAMS-2941, Los Alamos Scientific Laboratory (1963).

[11] WARD W. ENGLE, Jr., "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 IV

Calculated Multiplication Factors and Prompt-Neutron Decay Constants for Delayed-Critical Assemblies of UF_4-Paraffin Mixtures

Fuel Mixture	$H/^{235}U$ Atomic Ratio	ANISN Transport Code[a]			KENO Monte Carlo Code[b]	
		Revised Hansen-Roach 16-Group Cross Sections	Knight-Modified Hansen-Roach 16-Group Cross Sections	Bell 18-Group Cross Sections	Near Cubic Experimental Critical Assemblies	Sphere from Buckling Conversions
$U(2)F_4$-1	195.2	0.9596 (228)[d]	1.007 (236)	0.9866 (202)	1.007 ± 0.004[c]	0.998 ± 0.003
$U(2)F_4$-2	293.9	0.9755	1.0076	0.9909	1.002 ± 0.004	
$U(2)F_4$-3	406.3	0.9766	0.9985	0.9896	0.992 ± 0.005	1.001 ± 0.004
$U(2)F_4$-4	495.9	0.9837	0.9952	0.9863	0.992 ± 0.005	
$U(2)F_4$-5	613.6	0.9874 (142)	0.9885 (142)	0.9870 (112)	0.993 ± 0.005	0.990 ± 0.004
$U(2)F_4$-6	971.7	0.9920 (108)	0.9886 (109)	0.9890 (81)	0.994 ± 0.005	0.996 ± 0.005
$U(3)F_4$-1	133.4	0.9728 (332)	1.0179 (333)	0.9968 (314)	1.015 ± 0.004	1.023 ± 0.005
$U(3)F_4$-2	276.9	0.9869 (241)	1.0082 (240)	1.0017 (217)	1.003 ± 0.005	1.007 ± 0.004

[a]The results shown are for delayed-critical spheres inferred from experimental results by buckling conversions.

[b]The Knight-modified Hansen-Roach 16-group cross-section set was used in all KENO calculations.

[c]The multiplication factor for this fuel mixture calculated with a modified version of the O5R Monte Carlo code and ENDF-B-II cross sections, except for fluorine, was 0.9687 ± 0.0041. The fluorine cross sections were those recommended by Norton.[12] The multiplication factor calculated with ENDF-B-II A cross sections for the uranium isotopes was 0.9669 ± 0.0066.

[d]Numbers in parentheses are the prompt-neutron decay constants in units of sec^{-1} and are to be compared with the measured values of 237, 139, 105, 343, and 242 sec^{-1} for these UF_4-paraffin mixtures.

Knight-modified cross sections are in closest agreement with experimental results at low $H/^{235}U$ ratios where the poorest agreement is obtained with the revised Hansen-Roach set. Calculation of the prompt-neutron decay constant at delayed criticality for the UF_4-paraffin mixtures where measurements have been made are also given in Table IV and close agreement with the experimental results is obtained by use of the Knight-modified Hansen-Roach cross sections.

The critical mass and critical volume of bare and reflected spheres of $U(3)F_4$-paraffin mixtures with $H/^{235}U$ atomic ratios varying from 133 to 867 were calculated using the ANISN code. The results of the calculations, which used the three different sets of cross sections discussed previously, are shown as the shaded area in Figs. 3 and 4. The upper limit of the shaded area at low $H/^{235}U$ atomic ratios was determined by the results obtained from the revised Hansen-Roach cross-section set and the lower limit by the Knight-modified Hansen-Roach cross-section set.

The ANISN transport theory code was also used to obtain values of k_∞ using both the Knight-modified Hansen-Roach 16-group cross sections and the revised Hansen-Roach 16-group set. The results are given in Table II and plotted in Fig. 7. The k_∞ values calculated from the Knight-modified cross sections were in closer agreement with the measured values than those obtained from the revised Hansen-Roach set for fuel mixtures with low $H/^{235}U$ atomic ratios. However, for fuels at higher $H/^{235}U$ ratios, the revised Hansen-Roach set produced k_∞ values in closer agreement with the measured values than did the Knight-modified set. A single calculation using the original Hansen-Roach cross-section set[13] for the fuel mixture at an $H/^{235}U$ ratio of 195 yielded a k_∞ value of 1.114 which was much lower than that obtained from use of either of the other cross-section sets.

Theoretical studies by both Craven[14] and Webster[15] indicated that the effect of UF_4 particle size

[12]D. S. NORTON, "The U.K.A.E.A. Nuclear Data Library," AEEW-M 824, United Kingdom Atomic Energy Authority, Harwell (1968).

[13]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).

[14]C. W. CRAVEN, Reactor Division, ORNL, Personal Communication.

[15]J. WALLACE WEBSTER, "O5R Monte Carlo Calculations of Low-Enriched Uranium Thermal Critical Assemblies, and Evidence of Errors in ^{238}U Cross Sections," ORNL-TM-2187, Oak Ridge National Laboratory (1968).

in the whole assembly was worth <0.5% in k_{eff}. An uncertainty of ±1.2% in the ^{235}U content of the fuel would also be worth only about ±0.5% in k_{eff}. The experimental uncertainties in the critical dimensions and densities of the assemblies produce a negligible effect on k_{eff}. The accuracy of the conversion from experimental data obtained from rectangular parallelepipeds to critical sphere dimensions was also investigated by calculating k_{eff} for several of the experimental assemblies with the KENO Monte Carlo code.[16] Thirty thousand neutron histories were studied in each Monte Carlo calculation, and within statistical accuracy, the results, given in Table IV, are the same for experimental assemblies and corresponding spheres.

The neutron multiplication factors for the three unreflected assemblies of the U(2)F_4-1 fuel mixture, described in Table II, were calculated by Petrie[17] using the Monte Carlo code SUPERKENO and ENDF-B-III cross-section data[18] except for fluorine. The fluorine cross sections were those recommended by Norton.[12] The calculation was performed accounting for the molecular binding in the thermal energy range using the $S_{\alpha\beta}$ scattering models[19] with the data for polyethylene.[20] Elements other than hydrogen were treated as free gas scatterers.[21] This scattering model had previously predicted the measured thermal-neutron spectrum for the U(2)F_4-1 mixture.[22] The resulting neutron multiplication factors for these three assemblies were 0.96, 0.97, and 0.96, respectively, each with a standard deviation of ±0.005. These factors agree with those obtained with the revised Hansen-Roach cross sections.

From these results it is concluded that the discrepancy between experimental values and computed values of k_{eff} is related to the cross sections for ^{238}U in the resonance region. This conclusion is in agreement with those of Stratton[23] and Webster.[15] Perhaps more accurate absorption cross sections for ^{238}U at resonance energies may resolve this problem.

CONCLUSIONS

The minimum critical spherical volumes of a U(2)F_4-paraffin mixture when unreflected and reflected with an effectively infinite hydrogenous reflector are 199 and 138 liters, respectively, and the minimum critical ^{235}U masses are 7.5 and 5.4 kg, respectively. Transport theory calculations of U(3)F_4 mixtures indicate that the unreflected and reflected minimum critical volumes are about 93 and 57 liters, respectively, and that the minimum critical masses are about 4.1 and 2.7 kg of ^{235}U, respectively.

The infinite-medium neutron multiplication factors inferred from the critical experiments agreed with those calculated by transport theory using the Knight-modified Hansen-Roach 16-group cross-section set. However, calculations using the revised Hansen-Roach 16-group cross sections produced much lower values for k_∞ at the lower $H/^{235}U$ atomic ratios indicating that the ^{238}U resonance absorption cross sections used may be in error. Similar results were also obtained from transport theory calculations of k_{eff} where the values determined from the Knight-modified Hansen-Roach 16-group cross-section set agreed quite well with experiment, but when the revised Hansen-Roach 16-group cross-section set was used the k_{eff} value was low by as much as 4% for mixture U(2)F_4-1. This conclusion is in agreement with that of Stratton and Webster and indicates that a more accurate absorption cross section for ^{238}U at resonance energies may resolve this problem. Recent ENDF-B-III cross sections distributed by the National Cross Section Center at Brookhaven National Laboratory and Monte Carlo calculations of the neutron multiplication factor for the fuel mixture with the largest ^{238}U density have not resolved the discrepancy between measurement and calculation.

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[21] R. R. COVEYOU, R. R. BATE, and R. K. OSBORN, *J. Nucl. Energy*, **2**, 153 (1956).

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[23] WILLIAM R. STRATTON, "Correlations of Experiments and Calculations," in *Proc. Nuclear Criticality Safety*, SC-DC-67-1305, Sandia Corporation (1966).