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SUMMARY

Several experiments have been performed at ORNL with light water solutions of uranyl nitrate (highly enriched in either U^{233} or U^{235}) in an essentially bare sphere 27 inches in diameter. This report presents the results of several calculations with elementary bare reactor theory and a discussion of the observed discrepancies between the calculated and experimental results. If the observed critical concentration is used in the calculations, the calculated effective multiplication constant is less than unity; thus a higher critical concentration would be predicted than is actually observed.

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A COMPARISON OF ELEMENTARY CRITICALITY CALCULATIONS WITH EXPERIMENTAL RESULTS

C. W. Nestor, Jr.

Introduction

The purpose of this report is to compare the predictions of elementary bare reactor theory to experimental results for a set of specific "clean" critical experiments, and to investigate some of the possible sources of the observed discrepancies. There are two areas of major importance, namely the uncertainties in the microscopic cross section data and in the slowing down properties of light water, and the uncertainties in the chemical and isotopic analyses of the reactor fuel solutions.

Method and Sources of Nuclear Data

The effective multiplication constant k_e of a bare reactor was taken to be of the form

$$k_e = \frac{\eta \epsilon p f K(B^2)}{1 + L^2 B^2} \quad (1)$$

(Symbols are defined in Table 1)

The uncertainties in the chemical and isotopic analyses affect k_e primarily through the thermal utilization f ; the value of L^2 is of course changed by changes in the cross sections, but since $L^2 B^2 \approx 0.02$ for this reactor, the resultant changes in k_e are negligible. The quantity ϵp has been calculated¹ for cases of interest by numerical integration of cross section data (but neglecting the increase of ν with neutron energy). The effect of changes in the factors of (1) on the calculated values of k_e may be estimated by the use of the formula

Table 1. Definition of Symbols

k_e	effective multiplication constant
η	number of fission neutrons produced per thermal neutron absorbed in fuel
f	thermal utilization
ϵp	product of fast effect (total fissions per thermal fission) and resonance escape probability
$K(B^2)$	epithermal nonleakage probability
L^2	square of diffusion length, cm^2
B^2	geometric buckling, cm^{-2}
R	reactor radius, cm
d	extrapolation distance, cm
σ_s	scattering cross section of U^{233}
$\bar{\sigma}_s^H$	average microscopic scattering cross section of hydrogen, barns
N_H	hydrogen concentration, atoms per barn centimeter
$\bar{D}$	average diffusion coefficient, cm
Σ_{tr}	average macroscopic transport cross section, cm^{-1}
σ_a	microscopic absorption cross section
Σ_a	macroscopic absorption cross section
(superscripts: F fissile material	
H hydrogen	
B boron	
N nitrogen	

$$\frac{\delta k_e}{k_e} \approx \frac{\delta(\eta)}{\eta} + \frac{\delta(f)}{f} + \frac{\delta(\epsilon p)}{\epsilon p} + \frac{\delta(K)}{K} + \frac{\delta(P)}{P} \quad (2)$$

where $P = (1 + L^2 B^2)^{-1}$.

The thermal utilization was assumed to be of the form

$$f = \frac{\Sigma_a^F}{\Sigma_a^F + \Sigma_a^H + \Sigma_a^B + \Sigma_a^N} ; \quad (3)$$

and as an approximation to $\frac{\delta f}{f}$ we have

$$\frac{\delta f}{f} \approx (1-f) \frac{\delta \Sigma_a^F}{\Sigma_a^F} - \frac{\Sigma_a^H \left(\frac{\delta \Sigma_a^H}{\Sigma_a^H} \right) + \Sigma_a^B \left(\frac{\delta \Sigma_a^B}{\Sigma_a^B} \right) + \Sigma_a^N \left(\frac{\delta \Sigma_a^N}{\Sigma_a^N} \right)}{\Sigma_a^F + \Sigma_a^H + \Sigma_a^B + \Sigma_a^N} \quad (4)$$

The use of (4) leads to an approximate limit of error to be expected from the reported uncertainties in cross sections and material analyses.

The nonleakage probability $K(B^2)$ used in these calculations was the form derived by Weinberg, Trubey and Moran from the slowing down data of Hill, Roberts and Fitch.² The Fourier transform of the slowing down kernel was computed by numerical integration for various values of B^2 ; these results were found to be well represented by the empirical function

$$K(B^2) = \left[(1 + B^2 \tau_1)(1 + B^2 \tau_2)(1 + B^2 \tau_3) \right]^{-1} \quad (5)$$

with

$$\tau_1 = 26.27 \text{ cm}^2$$

$$\tau_2 = \tau_3 = 2.26 \text{ cm}^2$$

$$\tau = \tau_1 + \tau_2 + \tau_3 = 30.8 \text{ cm}^2$$

It should be noted that this value is in essential disagreement with the currently accepted theoretical value,³ but in excellent agreement with the result of J. W. Wade,⁴ namely

$$\tau = 31 \pm 1 \text{ cm}^2$$

Further, the form (5) gives $K(B^2)$ at indium resonance, 1.44 ev, and not at "thermal." In view of some controversy connected with the size of the correction term, it was neglected, but the effect of various corrections was estimated from (2). There is a possibility that the fission spectrum of U^{233} may differ sufficiently from that of U^{235} to produce a change in the nonleakage probability; there is however no quantitative information available at present.

The geometric buckling B^2 is of the form

$$B^2 = \pi^2 / (R + d)^2 \quad (4)$$

where d is the so-called extrapolation distance. An estimate of this quantity was made in the following way. Since the migration area $L^2 + \tau$ is made up chiefly of the age τ , the majority of the leakage is nonthermal and the extrapolation distance d should be estimated on the basis of an average fast transport mean free path. If it is assumed that the major contribution is due to hydrogen scattering and take

$$\sigma_s^H = 20 \text{ barns}$$

$$N_H = 0.0667 \text{ atoms/barn centimeter.}$$

the following is obtained:

$$\frac{1}{\bar{D}} = 3 \bar{\Sigma}_{tr} \approx 1.33 \text{ cm}^{-1}$$

$$d \approx 2.13 \bar{D} = 1.6 \text{ cm}$$

There is also the effect associated with the aluminum wall. Taking a "reflector savings" equal to the wall thickness (0.32 cm) gives

$$d \approx 1.9 \text{ cm}$$

Due to the "back of an old envelope" nature of this estimate, the calculations were performed for three values of d , 1.5, 2.0 cm, and 2.5 cm.

The cross sections and eta values for U^{233} and U^{235} (from the "World Consistent Set" in the second edition of BNL-325), thermal cross sections from the same compilation and the ϵp values reported by Terasawa are listed in Table 2.

Experimental Results

The experimental results and a discussion thereof, prepared by D. W. Magnuson, are given in the Appendix. In addition, the following information has been obtained:⁵ each stated uranium concentration is the average of four determinations; each nitrate and boron concentration is the average of two determinations. The standard error associated with the uranium analyses is 0.2% while those associated with both the boron and nitrate are 5%. The chemical analyses were determined as grams of constituent per gram of solution. The densities of the solutions were also determined; errors in this measurement were neglected here.

Discussion of the Results of Calculations

The results of calculations with the model discussed above are presented in Table 3. The values of k_e in parentheses are those calculated with the higher values of ϵp reported by Terasawa.¹ Every calculated value is less than unity. There is an apparent trend in cases 5 through 9 (all fueled with U^{233}) for the calculated k_e to decrease with increasing boron concentration; however, this effect, as well as the apparent discrepancy between case 3 and

Table 2. Nuclear Data

1. ϵp

U^{235} :	1.000	(S. Terasawa)
	1.006	(Weinberg, Trubey and Lessig)
U^{233} :	1.012	(S. Terasawa, $\sigma_s = 10$ bn)
	1.016	(S. Terasawa, $\sigma_s = 15.3$ bn)

2. Microscopic data

Element	η	σ_a	(2200 m/sec values, BNL-325, Ed. II)
U^{235}	$2.07 \pm .02$	694 ± 8	
U^{233}	$2.28 \pm .02$	582 ± 6	
H	0	$0.332 \pm .002$	
O	0	0	
N	0	$1.88 \pm .05$	
B	0	755 ± 2	

3. L^2

$$L^2 = \frac{\sum_a^{H_2O} (kT)}{\sum_a (kT)} L^2(H_2O), \quad L^2(H_2O) = 7.339 \text{ cm}^2 \text{ at } 20^\circ\text{C}$$

Table 2. Nuclear Data - contd

4. Concentrations in atoms/bn cm at 20°C

hydrogen $N_H = 6.674 \times 10^{-2}$

oxygen $N_O = 3.337 \times 10^{-2}$

Case	Fissile uranium N_F	Nitrogen N_N	Boron N_B	
1	4.807×10^{-5}	1.569×10^{-4}	0.0	} U^{235}
2	5.621	2.128	0.521×10^{-5}	
3	6.394	2.392	1.041	
4	6.796	2.548	1.281	
5	4.339	1.178	0.0	} U^{233}
6	4.514	1.224	0.133	
7	4.682	1.274	0.259	
8	4.848	1.319	0.383	
9	5.009	1.363	0.508	

cases 1, 2, and 4, can be accounted for by the uncertainties in the cross sections. For example, in case 3, application of formula (4) gives a limit of error estimate for $\frac{\delta f}{f}$ of 2.5% which is much larger than the observed discrepancy in the calculated value of f .

There are certain systematic errors in the calculation arising from the neglect of the nonfissile uranium isotopes and from the neglect of any correction in $K(B^2)$ from 1.44 ev to thermal. Taking account of these errors

would reduce the calculated values of k_e still further. An increase in the age by 3% leads to a 0.6% decrease in k_e . The non- $1/v$ behavior of the absorption cross section of U^{235} leads to about a 0.8% reduction in the k_e values listed in Table 3 for cases 1 through 4.

Table 3. Results of Calculations

Case	k_e $\left(\begin{array}{c} d = 1.5 \\ K(B^2) = 0.8064 \end{array} \right)$	k_e $\left(\begin{array}{c} d = 2.0 \\ K(B^2) = 0.8104 \end{array} \right)$	k_e $\left(\begin{array}{c} d = 2.5 \\ K(B^2) = 0.8150 \end{array} \right)$	f
1	0.975 (0.981)	0.981 (0.987)	0.987 (0.993)	0.5971
2	0.976 (0.982)	0.981 (0.987)	0.987 (0.993)	0.5956
3	0.973 (0.979)	0.979 (0.985)	0.985 (0.991)	0.5929
4	0.975 (0.981)	0.980 (0.986)	0.986 (0.992)	0.5934
5	0.961 (0.965)	0.966 (0.970)	0.972 (0.976)	0.5297
6	0.959 (0.963)	0.964 (0.968)	0.971 (0.975)	0.5286
7	0.958 (0.962)	0.964 (0.968)	0.970 (0.974)	0.5275
8	0.958 (0.962)	0.963 (0.967)	0.969 (0.973)	0.5268
9	0.956 (0.960)	0.961 (0.965)	0.967 (0.971)	0.5256

APPENDIX

SUMMARY OF RECENT ORNL CRITICAL EXPERIMENTS

D. W. Magnuson

A summary of the experimental data for the critical experiments in spherical geometry with U^{235} and U^{233} solutions for the determination of eta has been prepared in the attached table.

The following items are explanatory notes for the table:

Column 1: The excess reactivity, $\zeta = \sum_i \frac{\beta_i}{1 + \lambda_i T}$, was measured as a stable period at several temperatures and these data were extrapolated or interpolated to 20°C. The latest values of the delayed neutron yields and periods (Keepin, Wimett, and Ziegler, Phys. Rev. 107, 1044, (1957)) were used to calculate these reactivities.

Column 2: The density of the solution was measured by weighing a known volume at $25 \pm 1^\circ\text{C}$.

Column 3: The total uranium analyses were made by a potentiometric titration method using ferric sulfate and were checked by a coulometric method on later samples. Standard solutions prepared from specially purified metallic uranium are periodically checked by the analytical chemistry group.

Columns 4 - 8: Isotopic analyses are averages of several samples.

Column 9: Boron analyses were performed using a Mannitol titration method after removal of the cations by ion exchange. The isotopic composition of the boron was not significantly different from "Argonne Standard Boron."

Column 10: Total nitrate analyses were performed by ion exchange and by calculating total anion as NO_3 .

Column 11: Two U^{235} metal foils, 0.002 in. thick and 0.3125 in. in dia were irradiated when located 1 in. from and on opposite sides of the centers of the critical systems. One foil was bare and the other was cadmium covered. The ratio of their fission product gamma-ray activities are given in Column 11. All cadmium covers were 0.026 in. thick.

Additional Notes on Experiments 3 and 4: To each sphere was attached two vertical tubes, 2 in. in dia and, of course, diametrically opposite each other. These tubes served for filling and venting the sphere and the solution contained by the lower one during an experiment contributed to the reactivity. This contribution was evaluated as 0.36×10^{-4} in $\Delta k/k$ in experiment 3 by filling the upper tube to an appropriate height and observing the resulting period.

A test was performed to estimate the neutron reflection by the aluminum wall of the sphere. A partial-spherical shell of aluminum equal in thickness to the aluminum sphere (0.32 cm) covering 32.7% of the sphere in Experiment 4 contributed a reactivity of 2.3×10^{-4} . In another reflector test in Experiment 4, a cylinder of paraffin 8 in. in dia and 6 in. high encased in a mild steel can with a flat bottom and 0.25 in. wall thickness placed against the outside of the aluminum sphere was worth 2.5×10^{-4} .

A value of the geometric buckling of Experiment 4 was determined to be $7.408 \times 10^{-3} \text{ cm}^{-2}$ from a number of flux traverses measured by miniature fission counters. The corresponding extrapolation distance is 1.9 cm.

Additional Notes on Experiment 11: A similar variety of tests were made in Experiment 11. Fission counter traverses yielded a buckling of $2.567 \times 10^{-3} \text{ cm}^{-2}$ and an extrapolation distance of 1.0 cm. A cylinder of paraffin 20 in. in dia and 8 in. thick, encased in 0.125-in.-thick aluminum was worth 1.7×10^{-4} in $\Delta k/k$ when brought into contact with the sphere. (The paraffin container was made concave to closely fit the spherical surface.) The effect of solution in the fill-tube at the bottom of the sphere was shown to be negligible. Some activation measurements were made in Experiment 11 in addition to those reported in the table. Two bare U^{235} metal foils (0.002 in. thick x 0.3125 in. dia) were first irradiated with faces in contact when located one inch from the center of the sphere. They were also irradiated as single foils each located one inch from the center, that is, the foils were separated 2 in. The average ratio of the activity of the foils when adjacent to the activity when separated was 0.895. Two discs each of U^{233}_{90} and U^{235}_{92} were prepared having a diameter of 0.3125 in. and a metal equivalent thickness of 0.002 in. The cadmium covered to bare activity ratio of these "foils" was 0.0616 for the U^{233} and 0.0241 for U^{235} . This cadmium-covered to bare activity ratio was determined by miniature fission counters lined

with U^{233} and U^{235} . The values obtained were 0.0585 and 0.0203 for U^{233} and U^{235} , respectively. These latter foil and chamber measurements were also made at a point 1 in. from the center of the sphere.

REFERENCES

1. S. Terasawa, The Effect of Epithermal Fission on Aqueous Homogeneous Reactors, ORNL-2553 (1958).
2. Herbert Goldstein, P. F. Zweifel and D. G. Foster, Jr., "The Slowing Down of Neutrons in Hydrogeneous Media - Status of Theory and Experiment," Geneva paper A/CONF. 15/P/2375, (1958).
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TABLE A.1. EXPERIMENTAL DATA FOR CRITICAL SPHERES

Aluminum Sphere Volume = 173.6 liters Wall Thickness = 0.32 cm

Experi- ment Number	(k-1)x10 ⁴	Solution Density (g/ml)	Total Uranium (g/g)	Isotopic Analyses, wt%					Total Boron (mg/g)	Total Nitrate (g/g)	U ²³⁵ Metal Foil Activation-Ratio (Cd covered/Bare)
				U ²³³	U ²³⁴	U ²³⁵	U ²³⁶	U ²³⁸			
1	11.8	1.0288	0.01956	0.00	1.04	93.18	0.27	5.51	0.00	0.0187	0.0274
2	7.3	1.0333	0.02277	0.00	1.04	93.18	0.27	5.51	0.0905	0.0212	
3	9.0	1.0387	0.02577	0.00	1.04	93.18	0.27	5.51	0.18	0.0237	
4	2.8	1.0445	0.02724	0.00	1.04	93.18	0.27	5.51	0.22	0.0251	0.0367
5	5.0	1.0226	0.01676	97.70	1.62	0.04	0.00	0.64	0.00	0.0119	0.0264
6	10.3	1.0253	0.01742	97.70	1.62	0.04	0.00	0.64	0.0233	0.0123	
7	10.9	1.0274	0.01803	97.70	1.62	0.04	0.00	0.64	0.0453	0.0128	
8	3.3	1.0275	0.01867	97.70	1.62	0.04	0.00	0.64	0.0670	0.0132	
9	4.4	1.0286	0.01927	97.70	1.62	0.04	0.00	0.64	0.0887	0.0136	0.0277

Aluminum Sphere Volume = 949.1 liters Wall Thickness = 0.77 cm

10	12.9	1.0216	0.01482	0.01	1.05	93.21	0.54	5.19	0.00	0.0113	0.0235
11	4.6	1.0153	0.01305	97.67	1.54	0.03	0.00	0.76	0.00	0.0076	0.0214

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