CRITICALITY OF ARRAYS OF ^{233}U SOLUTION

R. C. LLOYD and E. D. CLAYTON
Battelle Memorial Institute, Pacific Northwest Institute Laboratory, Richland, Washington 99352
and
J. H. CHALMERS
Authority Health and Safety Branch, United Kingdom Atomic Energy Authority, Risley, Warrington, Lancashire, England

KEYWORDS: *critical arrays nuclear safety, Monte Carl calculations, ^{233}U solution fissionable material*

Received September 29, 1967
Revised November 27, 1967

The results of neutron multiplication measurements performed with arrays of ^{233}U solution apply to criticality safety considerations in handling solutions at a concentration of ≈ 330 g ^{233}U/liter and are useful in checking computational methods. The measurements were made with ≤ 17.3 kg ^{233}U in both reflected and unreflected arrays. Critical numbers of bottles were determined as a function of spacing, and the effect of adding moderating material between the bottles comprising an array was also examined. Monte Carlo calculations were found to reproduce the experimental data reasonably well, with k_{eff} being computed to within about 0.03 of unity for those cases compared.

INTRODUCTION

The criticality of interacting arrays of fissionable materials is important in fuel processing, shipment, and storage. The difficulty in predicting criticality of complex arrays is further increased by the lack of experimental data that may be used to confirm calculations. Some previous experiments have been performed with single or isolated units of ^{233}U solutions in various geometries,[1,2] but no data have been reported on the criticality of interacting arrays of this material. To obtain such data, multiplication measurements were performed with unreflected and Lucite-reflected arrays of bottles of ^{233}U solution.[3] The effect on criticality of adding Lucite moderator between the bottles was also studied.

Various empirical and semitheoretical methods have been proposed for predicting criticality of arrays, but they are subject to limitations on the available experimental data.[4-6] We have made Monte Carlo calculations to compute the criticality of some of the arrays for which we have also measured criticality empirically. Such comparisons were also made on complex arrays having both external reflection and internal Lucite moderation. The results of the calculations further demonstrate the reliability of the Monte Carlo technique in handling these kinds of criticality problems.

MEASUREMENTS

The ^{233}U was in the form of uranyl nitrate hexahydrate, $UO_2(NO_3)_2 + 6H_2O$, at a concentration of ≈ 330 g ^{233}U/liter; the uranium solution, ≈ 0.53 $\underline{M}$ in excess nitrate acid concentration, was contained in three-liter polyethylene bottles. The isotopic content of the solution is given in Table I. The bottles were 17.75-in. high and 4.7-in. o.d., with a wall thickness of 0.100 in. The average solution height was ≈ 11.5 in., corresponding to 960 g uranium per bottle. Table II shows that the bottles varied in content and given averages of solution contents for each experiment. This variation amounted to less than ± 20 g ^{233}U/liter except for Experiment 21 where it was ± 40 g

TABLE I

Isotopic Content of the Uranium Solution

^{232}U	≈ 4 ppm
^{233}U	98.2%
^{234}U	0.8%
^{238}U	1.0%

TABLE II

Averages for Actual Bottles Used in Experiments

Experiment Number	Solution Height in.	U g	Volume liter	^{233}U Concentration g/liter	Specific Gravity
1	11.78	946	2.98	318.3	1.437
2	11.67	951	2.94	323.1	1.444
3	11.59	947	2.93	323.0	1.444
4	11.40	961	2.89	332.5	1.455
5	11.77	947	2.95	321.1	1.445
6	11.72	962	2.95	326.2	1.450
7,8,9,10,11	11.71	934	2.94	318.1	1.438
12	11.64	947	2.93	323.2	1.444
13A	11.67	951	2.94	323.1	1.444
13B,14,15,16,17	11.59	947	2.94	323.0	1.444
18,19,20	11.40	961	2.89	332.5	1.455
21	11.25	895	2.87	312.6	1.429

U/liter. Use of the average concentration should introduce minimum error since criticality is quite insensitive to concentration over this range.

The bottles were placed in subcritical arrays on a remotely operated split-table device. Because the reactivity of an array can change rapidly with spacing, separate neutron multiplication measurements were made to determine that the number of bottles for each spacing could be safely loaded. The neutron multiplication for the fixed number of bottles at each spacing was then, in turn, used to predict the critical spacing for the array. Thus, the experiments required a series of multiplication measurements—one for each spacing.

The remote split-table machine (RSTM), used in these experiments, has a table top of 0.03-in.-thick steel plate supported by an aluminum honeycomb material that provides good support strength but low neutron reflection. A low-density aluminum honeycomb material was also used to provide accurate spacing between bottles and to ensure that the bottles remained upright.

Criticality of the bottles of solution was determined for both bare and Lucite-reflected systems. In the unreflected assemblies, stability of the outer bottles was maintained by aluminum frames attached to small magnets fixed to the thin-steel base plate. Unreflected assemblies of 9 and 16 bottles and a double-tier array of 18 bottles were measured.

In the reflected arrays, the Lucite was placed touching the outside surface of the bottles, i.e., boxing in the array. The thickness of the top and bottom Lucite reflector was 4.5 in. and the side reflectors were 6 in. Experiments were performed with Lucite moderator positioned between the bottles of an array in both unreflected and r flected assemblies. Figure 1 shows a 16-bot unreflected assembly, with this internal moderat at the same height as the solution.

RESULTS

Results obtained from plotting the recipro count rate (inverse neutron multiplication curv as the arrays were built up, and extrapolati these curves to predict criticality, are present in Table III. During the experiments it was not that an improvement in linearity of the neutr multiplication curves could be obtained by plotti

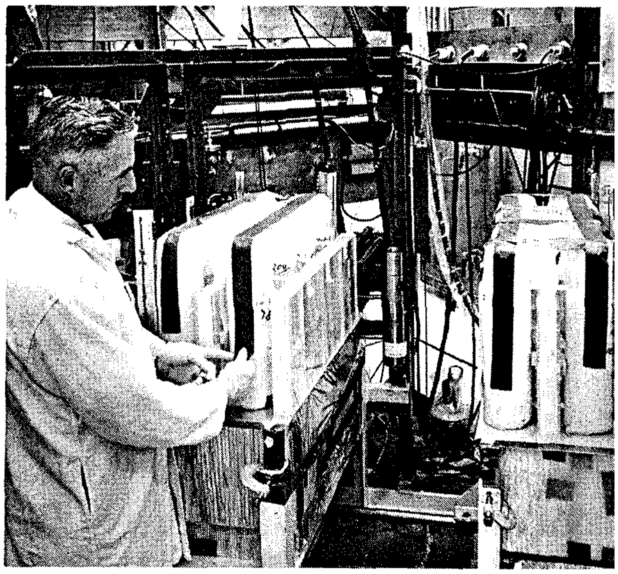

Fig. 1. 16-Bottle moderated unreflected array of ^{233} solution.

the spacing count-rate ratio vs spacing, rather than reciprocal count rate vs spacing. This permitted an earlier, better estimate of criticality; however, either method of plotting the data provides the same estimate in the limit as criticality is approached. The usefulness of the first type plot is that a better estimate of criticality is obtained during the initial portion of the experiment, although we have not found a theoretical explanation for this empirical observation. A single row of nine bottles unreflected was observed to be subcritical, and extrapolation of the inverse neutron multiplication curves indicated an infinitely long, single line would probably be subcritical as well. The data for the reflected row of bottles indicated that criticality would be achieved with more than two but less than three bottles. Three bottles in line with surface-to-surface (S-S) spacing[a] of 0.6 in. would be critical when reflected. S-S spacings for criticality were determined for unreflected arrays of 2 × 3, 3 × 3, and 4 × 4 bottles in single-tier geometry and for reflected arrays of 2 × 2 and 3 × 3 bottles.

A S-S critical spacing of 0.75 in. was also measured for a double-tier 3 × 3 bottle unreflected array, which compares with a 0.60-in. spacing for the 3 × 3 bottle single-tier array. The double-tier array had a spacing of 7.0 in. between the fuel of the upper and lower tiers.

Figure 2 shows the number of bottles required for criticality plotted vs S-S separation. The critical number of bottles is much more sensitive to

---

[a] S-S spacing refers to the outer bottle surface-to-outer bottle surface spacing.

TABLE III

Interaction Data for Bottles of ^{233}U Solution

Experiment Number	Reflector Condition	Configuration	Number of Bottles for Criticality[a]	Estimated Critical Surface-to-Surface Spacing (in)	Remarks
U-1	Unreflected	1 × 9	> 9	0	Single Row
U-2	Unreflected	2 × 3	6.1	0	Double Row
U-3	Unreflected	3 × 3	9	0.60	
U-4	Unreflected	4 × 4	16	1.16	
U-5	Reflected	1 × 2	> 2	0	≈ 2.8 Bottles, Single Row
		1 × 3	< 3	0	Table stopped at 0.9 in.
U-6	Reflected	1 × 3	3	0.6	Single Row
U-7	Reflected	2 × 2	4	2.18	
U-8	Reflected	2 × 2	4	2.48	$\frac{1}{2}$-in. Lucite Moderator Between Bottles
U-9	Reflected	2 × 2	4	2.58	$\frac{3}{4}$-in. Lucite Moderator Between Bottles
U-10	Reflected	2 × 2	4	2.66	1-in. Lucite Moderator Between Bottles
U-11	Reflected	2 × 2	4	2.50	$1\frac{1}{2}$-in. Lucite Moderator Between Bottles
U-12	Reflected	3 × 3	9	3.98	
U-13A	Unreflected	2 × 3	6.3	1.00	1-in. Lucite Moderator Between Bottles
U-13B	Unreflected	3 × 3	9	1.60	1-in. Lucite Moderator Between Bottles
U-14	Unreflected	3 × 3	9	1.17	$\frac{1}{2}$-in. Lucite Moderators Between Bottles
U-15	Unreflected	3 × 3	9	1.78	$1\frac{1}{2}$-in. Lucite Moderator Between Bottles
U-16	Unreflected	3 × 3	9	1.87	$1\frac{3}{4}$-in. Lucite Moderator Between Bottles
U-17	Unreflected	3 × 3	9	1.90	2-in. Lucite Moderator Between Bottles
U-18	Unreflected	4 × 4	16	2.50	2-in. Lucite Moderator Between Bottles
U-19	Unreflected	4 × 4	16	2.47	$2\frac{1}{2}$-in. Lucite Moderator Between Bottles
U-20	Unreflected	4 × 4	16	2.42	$1\frac{1}{2}$-in. Lucite Moderator Between Bottles
U-21	Unreflected	3 × 3 × 2	18	0.75	Double Tier

[a] Fractional number of bottles indicates extrapolation to the critical number with the spacing fixed.

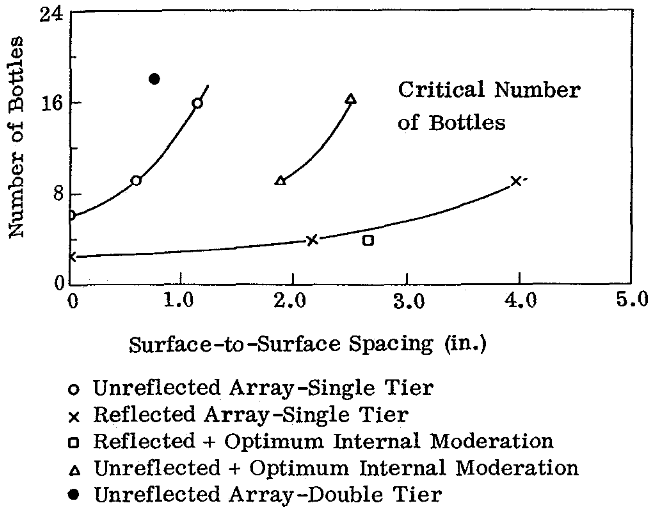

Fig. 2. Criticality of ^{233}U solution in polyethylene bottles.

spacing for the unreflected array than for the reflected array. Points of optimum internal moderation for maximum critical spacing of the array are also shown for comparison as determined from plotting spacing vs moderator thickness (Figs. 3 through 5). Figure 3 gives data on critical S-S spacing vs thickness of added Lucite moderator for a four-bottle reflected array. The most effective thickness, as here defined, is that thickness of moderator that results in the smallest critical number of bottles, or conversely, the largest critical spacing for a given number of bottles. The most effective thickness of added moderator was about 1 in. in the reflected array. Figures 4 and 5 give results of critical S-S spacing vs added Lucite moderator thickness for unreflected arrays comprising 9 and 16 bottles. These results indicate the most reactive unreflected loading to be obtained with a moderator thickness of about two inches between the bottles.

EXPERIMENTAL ERRORS

The error in critical spacing is due primarily to the uncertainty in extrapolation of the inverse neutron multiplication curves and the uncertainty in positioning the bottles within the array. Three counters were used simultaneously for the neutron multiplication measurements in which the inverse multiplication curves were plotted vs spacing. The arrays were subcritical in each case, but the uncertainty in critical spacing, as a result of extrapolation, is estimated to be about 0.03 in. This uncertainty comes from the difference in values for criticality predicted by the separate curves, while another uncertainty of about 0.02 in. can

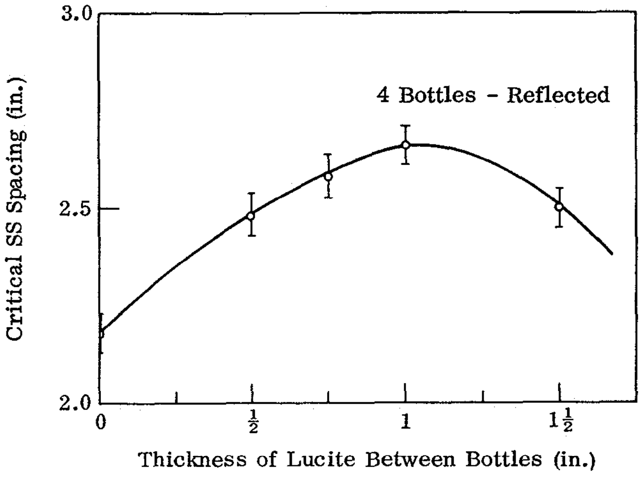

Fig. 3. Effectiveness of moderation between bottles of ^{233}U.

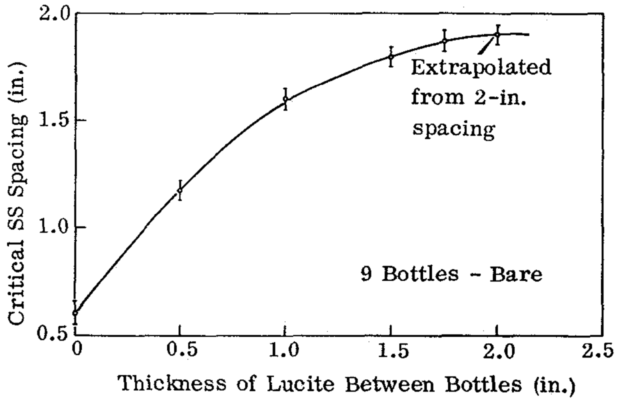

Fig. 4. Effectiveness of moderation between bottles of ^{233}U solution (9-bottle unreflected array).

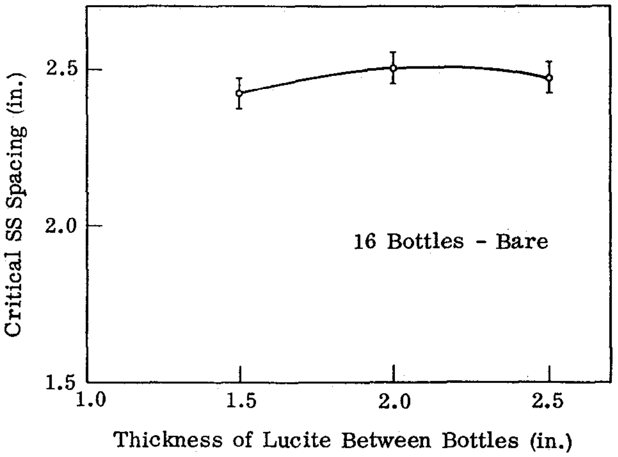

Fig. 5. Effectiveness of moderation between bottles of ^{233}U solution (16-bottle unreflected array).

result from positioning error in loading the arrays. Therefore, the uncertainty in the quoted critical spacings is ≈0.05 in.

This error does not apply to Experiments 1 and 5 which involved unreflected single rows of bottles. Due to the nature of the arrays and the observed neutron multiplication in these two cases, it was only possible to define upper or lower limits, i.e., critical number of bottles >9 in one case and between 2 and 3 in the other.

In the unreflected unmoderated array, an experimental uncertainty of 0.05 in. would correspond to a variation in k_{eff} of about ±0.006. This is concluded from examination of the values of k_{eff} calculated over a range of spacings from 0 to 0.6 in. (Fig. 6).

CALCULATIONS

A series of Monte Carlo-type calculations was made by using the GEM Code,[7] where the geometric complexities of the experimental arrays can be considered in as much detail as required. GEM is a Monte Carlo code written originally to study criticality problems, but it was later developed into a code versatile enough to perform calculations for a complete reactor. For the calculations, the system is divided into an inner "core" surrounded by a "reflector." The tracking cycle directly yields a surface multiplication M and a reflection R at the chosen boundary. The product MR gives a measure of the criticality of the system; when $1/M$ is equal to R, the system is critical, with k_{eff} being unity. The basic method of tracking is the assumption that within each regional boundary only one material is present. To avoid approximations in cases where a region contains more than one material or where the boundaries do not conform to the standard geometries, a technique of HOLE routines[7] has been developed. The HOLE routine technique enables one to specify a system directly, taking into account voids and the various materials that may exist in any region. This technique determines which material is present at a neutron collision point; it decides the conditions of collision and whether the neutron should continue and at what energy and in which direction. The results of calculating k_{eff} for 11 of the experimentally measured arrays are presented in Table IV. The standard deviation in the computed k_{eff} comes from the statistical uncertainty in the number of neutron histories traced in the Monte Carlo calculations. (To be in agreement with experiment, k_{eff} should have been unity, allowing for experimental uncertainties.) As seen from the table, even for the least accurately computed cases, k_{eff} is computed from the GEM Monte Carlo code to within about 0.03 of experimental unity.

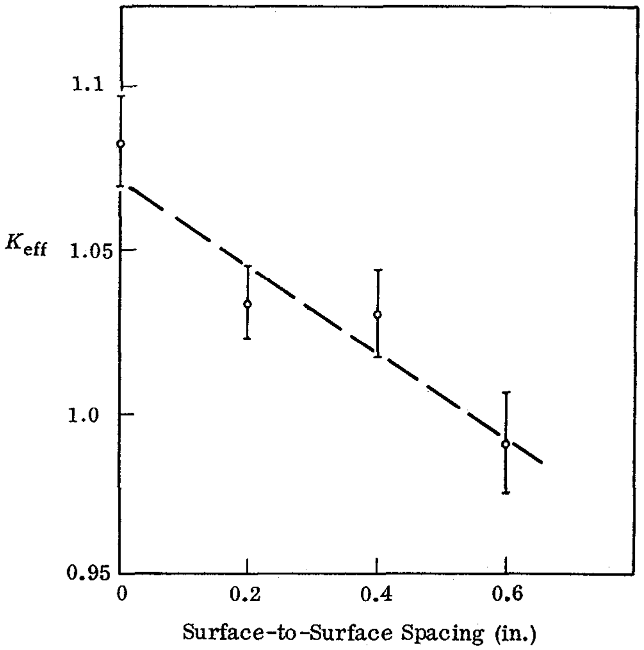

Fig. 6. Sensitivity of k_{eff} on S-S spacing.

A number of comparisons have been made between computed and measured values for ^{235}U systems that demonstrate the reliability of the Monte Carlo technique.[8-10] The error range of ±0.03 on the calculated k_{eff} for most of the critical experiments with ^{235}U systems is similar to that found in our experiments with ^{233}U systems. The sensitivity of k_{eff} to spacing was also examined (Fig. 6). When arrays are being examined by computational methods, the sensitivity of the particular system to spacing should always be investigated before any conclusions are made on the "safe" working spacing. If an array is found to be extremely unsensitive to changes in spacing, then an assumed factor of safety (on spacing) may consequently be nonexistent.

SUMMARY AND CONCLUSIONS

Experiments were performed to determine the criticality of arrays containing up to 18 bottles of ^{233}U solution, yielding data for nuclear criticality safety, and for checking computational methods. The critical number of bottles was found to be much more sensitive to spacing in an unreflected array than when reflected with Lucite. When moderating material in the form of Lucite plates was placed between the bottles, the most effective thickness was about 2 in. in the unreflected array and 1 in. in the reflected array; i.e., these thicknesses produced the maximum increases in reactivity from the unmoderated to the moderated conditions.

TABLE IV

GEM Calculations of k_{eff} for Experimental Arrays of ^{233}U Solution Containers

Experiment Number	Configuration	Lucite Moderator Thickness, in.	Surface-to-Surface Critical Spacing in.	k_{eff}	Standard Deviation
Reflected					
7	2 × 2	0	2.18	0.9844	0.019
9	2 × 2	0.75	2.58	0.9783	0.020
11	2 × 2	1.50	2.50	1.0133	0.033
12	3 × 3	0	3.98	0.9934	0.010
Unreflected					
3	3 × 3	0	0.60	0.9909	0.016
3[a]	3 × 3	0	0.40	1.0303	0.013
3[a]	3 × 3	0	0.20	1.0339	0.011
3[a]	3 × 3	0	0.00	1.0830	0.014
14	3 × 3	0.50	1.17	1.0018	0.012
13B	3 × 3	1.00	1.60	1.0069	0.012
17	3 × 3	1.90	1.90	1.0070	0.009
4	4 × 4	0	1.16	0.9894	0.013
18	4 × 4	2.00	2.50	0.9875	0.014
21	3 × 3 × 2	0	0.75	0.9728	0.013

[a] Computed in these cases (but not measured) for surface-to-surface spacings to investigate the sensitivity of k_{eff} with spacing.

The GEM calculations were found to reproduce the experimental data reasonably well. Even for the least accurately computed cases, k_{eff} would have been computed by the GEM-Monte Carlo code to within about 0.03 of unity.

ACKNOWLEDGMENTS

The authors wish to acknowledge sponsorship of this work by the USAEC under Contract No. AT(45-1)-1830 between the USAEC and Battelle Memorial Institute.

REFERENCES

1. R. GWIN and D. W. MAGNUSON, "The Measurement of Eta and Other Nuclear Properties of ^{233}U and ^{235}U in Critical Aqueous Solutions," *Nucl. Sci. Eng.* 12, 36 (1962).

2. J. K. FOX, L. W. GILLEY, and E. R. ROHER, "Critical Mass Studies Part VIII, Aqueous Solutions of ^{233}U," ORNL-2143, Oak Ridge National Laboratory (1959).

3. R. C. LLOYD and E. D. CLAYTON, "Neutron Interaction Experiments with Bottles of ^{233}U Solution," *Physics Research Quarterly Report July, August, September, 1966,* BNWL-340, Pacific Northwest Laboratory (1966).

4. H. C. PAXTON, "Correlations of Experimental and Theoretical Critical Data," *Criticality Control in Chemical and Metallurgical Plant,* Karlsruhe Symposium, Organisation for Economic Cooperation and Development, European Nuclear Energy Agency (1961).

5. J. T. THOMAS, "Criticality of Large Systems of Subcritical U(93) Components," ORNL-CDC-1, Oak Ridge National Laboratory (1967).

6. H. F. HENRY, C. E. NEWLON, and J. R. KNIGHT, "Extension of Neutron Interaction Criteria," K-1478, Union Carbide Nuclear Company (1961).

7. P. J. HEMMINGS, *The GEM Code,* AHSB (S) R 105, United Kingdom Atomic Energy Authority, Safeguards Division (1967).

8. J. T. THOMAS, "Experimental and Calculated System Criticality," *Criticality Control of Fissile Materials,* Stockholm Symposium, International Atomic Energy Agency (1966).

9. O. C. KOLAR, J. R. MORTON III, and N. L. PRUVOST, "Interaction in Arrays of Fissionable Materials," *Criticality Control of Fissile Materials,* Stockholm Symposium, International Atomic Energy Agency (1966).

10. J. T. THOMAS, "The Effect of Unit Shape on the Criticality of Arrays," ORNL-CDC-4, Oak Ridge National Laboratory (1967).