NASA TECHNICAL NOTE

NASA TN D-6878
e l

CRITICALITY EXPERIMENTS AND ANALYSIS OF MOLYBDENUM-REFLECTED CYLINDRICAL URANYL-FLUORIDE - WATER SOLUTION REACTORS

by Daniel Fieno, Thomas Fox, and Robert Mueller

Lewis Research Center
Cleveland, Ohio 44135
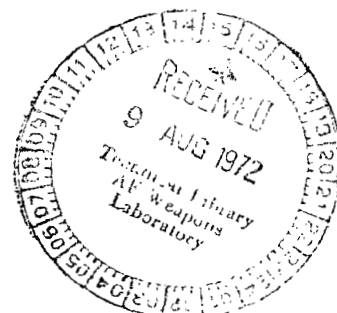
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • JULY 1972

1. Report No. NASA TN D-6878	2. Government Accession No.	3. Recipient's Catalog No.
4. Title and Subtitle CRITICALITY EXPERIMENTS AND ANALYSIS OF MOLYBDENUM-REFLECTED CYLINDRICAL URANYL-FLUORIDE - WATER SOLUTION REACTORS		5. Report Date July 1972
		6. Performing Organization Code
7. Author(s) Daniel Fieno, Thomas Fox, and Robert Mueller		8. Performing Organization Report No. E-6812
		10. Work Unit No. 112-02
9. Performing Organization Name and Address Lewis Research Center National Aeronautics and Space Administration Cleveland, Ohio 44135		11. Contract or Grant No.
		13. Type of Report and Period Covered Technical Note
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D. C. 20546		
		14. Sponsoring Agency Code

15. Supplementary Notes

16. Abstract

Clean criticality data were obtained from molybdenum-reflected cylindrical uranyl-fluoride - water solution reactors. Using ENDF/B molybdenum cross sections, a nine energy group two-dimensional transport calculation of a reflected reactor configuration predicted criticality to within 7 cents of the experimental value. For these reactors, it was necessary to compute the reflector resonance integral by a detailed transport calculation at the core-reflector interface volume in the energy region of the two dominant resonances of natural molybdenum.

17. Key Words (Suggested by Author(s)) Criticality experiment Molybdenum reflector Solution reactor	18. Distribution Statement Unclassified - unlimited		
19. Security Classif. (of this report) Unclassified	20. Security Classif. (of this page) Unclassified	21. No. of Pages 27	22. Price* $3.00

CRITICALITY EXPERIMENTS AND ANALYSIS OF MOLYBDENUM-REFLECTED

CYLINDRICAL URANYL-FLUORIDE - WATER SOLUTION REACTORS

by Daniel Fieno, Thomas Fox, and Robert Mueller

Lewis Research Center

SUMMARY

Clean criticality data were obtained from molybdenum-reflected cylindrical uranyl-fluoride - water solution reactors. Using ENDF/B molybdenum (Mo) cross sections, a nine energy group two-dimensional cylindrical (r,z) S_n calculation of a reflected reactor configuration having a hydrogen to uranium-235 atom ratio of 319.4 predicted criticality to within 7 cents of the experimental value. One-dimensional cylindrical radial S_n calculations were not adequate for predicting the criticality of these reactors; for the seven reactor configurations studied, the calculations were from 0.45 to 1.84 dollars too reactive.

For these reactors, it was necessary to compute the reflector effective resonance integral by a detailed transport calculation at the core-reflector interface volume in the energy region of the two dominant resonances of natural Mo. For the 44.7-eV resonance of Mo^{95}, the resonance integral was computed to be 7.20 barns while for the 131.3-eV resonance of Mo^{96}, the resonance integral was computed to be 3.55 barns.

The calculated fundamental prompt-mode decay constants at delayed critical were in good agreement with the values measured for these reactors.

Molybdenum cross sections from the ENDF/B number 1111 evaluation are adequate for predicting the multiplication factor of the seven reactors presented in this study.

INTRODUCTION

The conceptual design of a compact fast reactor for space power application and a technology program to support the reactor are in progress at Lewis Research Center (ref. 1). The reactor uses uranium nitride as fuel that is clad and supported by a tantalum (Ta) alloy T-111; lithium-7 (Li^7) is used as coolant and the core is reflected by the molybdenum (Mo) alloy TZM, both radially and axially. The nuclear design and criti-

cality experiments for the reactor are described in reference 2.

It has been our experience that attempts to calculate the neutron multiplication factor of past fast critical assemblies fueled with oralloy and containing or reflected by the refractory materials Ta and Mo, have yielded values significantly larger than the experimental factor of unity (ref. 3). The bulk of these discrepancies are attributed to uncertainties in fast neutron cross sections; smaller portions of the discrepancies have been assigned to the use of fewer neutron groups and lower order two-dimensional transport analysis.

In order to test the validity of available cross section evaluations, a series of integral experiments have been performed employing spheres of refractory materials with central neutron sources (ref. 4). Measured and calculated sphere leakage spectra in the energy range 0.5 to 11 MeV were compared as a test of the evaluated total, inelastic scattering, and reaction cross sections. Data for Mo showed general agreement but significant differences were observed for spheres of Ta, tungsten (W), and beryllium (Be). In order to obtain additional data and to widen the neutron energy range of comparison, a series of criticality experiments have been obtained using the NASA enriched uranyl-fluoride - water (UO_2F_2-H_2O) cylindrical solution reactor (ZPR-I) in a configuration having a thick molybdenum radial reflector. Both criticality data and radial neutron leakage spectra for this reactor have been obtained. The present report presents an analysis of the criticality data.

Because of the homogeneity of the cylindrical zones, the critical experiments present relatively clean configurations for analysis by both one- and two-dimensional, multigroup, discrete-ordinate transport theory computer codes. The adequacy of the most recent ENDF/B Mo cross section compilation may be tested by these calculations.

REACTOR AND EXPERIMENT PROCEDURES

The ZPR-I UO_2F_2-H_2O solution reactor used in these tests is a 25.4-centimeter-diameter cylindrical core tank that is reflected radially with a 10.14-centimeter-thick Mo annular reflector. The experimental program involved the measurement of two basic parameters; namely, critical solution height and the fundamental prompt-mode decay constant. Brief descriptions of the critical experiments and the pulsed source experiments are presented. More complete descriptions of the criticality and pulsed source experiment procedures are presented in references 5 and 6, respectively.

ZPR-I with Radial Molybdenum Reflector

ZPR-I is a solution reactor that uses an aqueous solution of highly enriched (93.2

percent uranium-235 (U^{235})) uranyl fluoride (UO_2F_2) as the fuel that is readily varied in concentration. The fuel solution is stored in geometrically safe storage tanks and the critical configuration is achieved by remotely transferring the fuel solution from storage to the stainless-steel reactor tank. In all cases where a critical configuration was to be achieved, the reflector arrangement was assembled first and then the fuel concentration was adjusted so that the core and reflector heights were the same at delayed critical.

The physical arrangement of the core and reflector is shown in figure 1. The radial dimensions are presented in table I. The reactor configuration is located at the approximate center of a large room and is elevated about 20 centimeters above a working grid platform to minimize interactions with nearby structure. The Mo reflector blocks were also stacked on a cylindrical spool-like aluminum platform designed to minimize the structural interactions.

The Mo blocks were fabricated by pressing and sintering and were machine finished. The material is 99.95 percent Mo and has an average density of 9.88 grams per cubic centimeter (96.8 percent of theoretical density). The reflector annulus has been divided into eight equal segments and is constructed from as many as 32 blocks which are 20.32 centimeters in height and eight blocks which are 9.87 centimeters in height. The size of the blocks permitted manual handling and reflector heights could be constructed to a maximum of 91 centimeters in approximately 10-centimeter increments. The radial thickness of the blocks in the radial dimension is 10.14 centimeters and when stacked in a tight array form a cylindrical chimney with an inside diameter of 26.87 centimeters and an outside diameter of 47.15 centimeters.

The reactor tank is constructed of type 304 stainless steel, 0.16 centimeter thick, and has an outside diameter of 25.72 centimeters. Allowing for the corrosion protective coating on the tank wall, there is a gap of 0.57 centimeter between reactor tank and reflector when only the blocks are used as a reflector. This gap was designed to accommodate the insertion of a cadmium (Cd) sleeve between the core and reflector as shown in figure 1. The Cd has been constructed of 0.089-centimeter-thick Cd sheet canned in stainless steel. The inserts are in the form of half cylinders constructed to eliminate gaps in the Cd layer that would permit radial leakage of subcadmium neutrons. The Cd sleeve was adjusted in length to match the height of the reflector used. Molybdenum inserts about 0.48 centimeter thick were also fabricated to fill the radial gap. Owing to dimensional tolerances, this is about the maximum amount of material that can be readily fitted into the gap and fills about 85 percent of the gap. These pieces were also fabricated in half cylinders of enough different sizes to permit a match of all of the reflector heights used during this study. The density of this material was measured and found to be within 1 percent of the theoretical density of 10.2 grams per cubic centimeter.
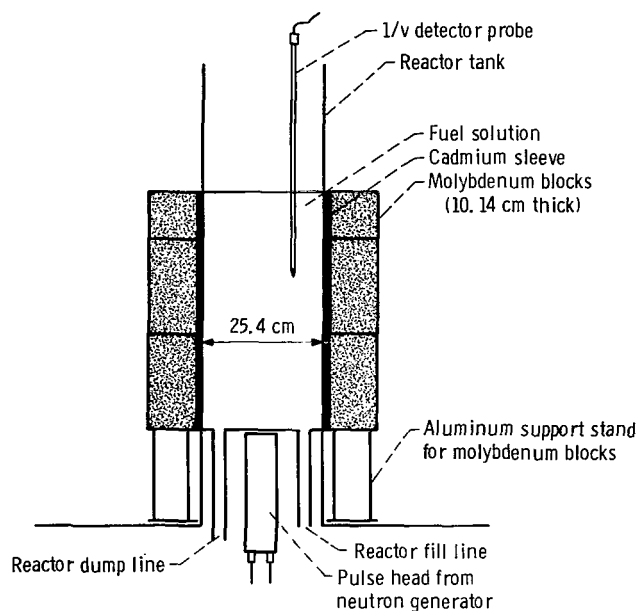
3

Figure 1. - Schematic diagram of NASA ZPR-I solution reactor with annular molybdenum reflector. (Cadmium insert sleeve is shown between stainless-steel tank wall and molybdenum reflector.)

TABLE I. - RADIAL DIMENSIONS OF THE NASA

ZPR-I SOLUTION REACTOR HAVING AN

ANNULAR MOLYBDENUM

REFLECTOR

	Inside radius, cm	Outside radius, cm
Fuel solution[a]	0	12.70
Type 304 stainless-steel tank	12.70	12.86
Insert region[b]	12.86	13.43
Mo reflector	13.43	23.57

[a] UO_2F_2 salt in H_2O solution with concentration varied so that fuel solution height was same as Mo reflector height for given experiment.

[b] Insert region either was left void or contained 0.089-cm-thick Cd sleeve clad in stainless steel, or 0.48-cm Mo sleeve.

4

Pulsed Neutron Source Experiments

The experimental arrangement used to conduct the pulsed neutron source experiments is the same as described in reference 6. The pulsing head from the neutron generator is located on the axis of the reactor core directly beneath the tank and the neutron detector is located in the core at a position found to be optimum for studying the fundamental prompt mode (see fig. 1). The neutron pulse generator is a small positive-ion sealed tube accelerator which produces 14.3-MeV neutrons by means of the ^{3}H(d, n)^{4}He reaction. The neutron pulse intensity is about 10^6 neutrons per pulse with the pulse width being 3.5 microseconds and the rate about 5 pulses per second. The neutron detector probe is a small $1/v$ gas proportional counter filled with helium-3 (^{3}He) gas. This probe is less than 1 centimeter in diameter and has a sensitive volume of 1 cubic centimeter which approximates a point detector. The pulses from the detector were fed through a preamplifier and amplifier to a 400-channel analyzer which was operated in a multiscaler mode using 100- and 200-microsecond channel widths. System resolution time has been measured as 1.12 microseconds with this arrangement.

The output data from the 400-channel analyzer is reduced using the digital computer program GRIPE II developed by Kaufman (ref. 7). This program calculates the fundamental prompt-mode decay constant and can be used to calculate the shutdown reactivity of a subcritical solution system. The data reduction methods are described in detail in reference 6.

Experimental Program

A series of experiments were made using the Mo reflected solution reactor. Several measurements were made of the reactivity effect associated with the voided gap between the core and the reflector, with the Cd sleeve inserted, and with the Mo sleeve inserted. Fuel concentration was varied over a range of hydrogen to uranium-235 atom ratios H/X of 319.4 to 615 during the tests. Delayed critical configurations were achieved with nominal reflector heights of 30.2, 40.6, 50.5, and 70.8 centimeters. The maximum height was limited by the height of the core tank to 75 centimeters.

Two sets of delayed critical experiments were run at each reflector height. The first set of experiments had the 0.57-centimeter gap between the core and reflector voided; for the second set the gap was filled with Mo inserts. To achieve the same critical height for the two sets required an adjustment in fuel concentration. The worth of the Mo sleeve can thus be measured in terms of a change in fuel concentration. If the reactivity is known as a function of fuel concentration, it is thus possible to get a reactivity worth for the Mo sleeve by this technique.

Another measurement of the reactivity worth of the Mo insert as well as the worth of a Cd insert was made using the pulsed neutron source technique. The most reactive configuration, which was the arrangement with the Mo insert, was used as the reference case. By using the fuel concentrations for the clean core cases previously run, the delayed critical condition was established with the pulsing equipment in place for fuel concentrations with H/X values of 330, 540, and 615. At each fuel concentration there was a small increase in critical height due to the presence of the detector probe and the proximity of the pulsing head. The new critical height was used throughout the pulsed experiments at each fuel concentration. The Cd insert and void gap arrangements were both subcritical at the reference fuel heights for all fuel concentrations used.

The reactivity worth measured by this pulsed source technique is considered to be a better value than the worth that could be obtained as a function of fuel concentration change (see ref. 6). Most important in this application, however, is the fact that it is not necessary to measure the fuel concentration to obtain the reactivity worth. It is only necessary to assure that the fuel concentration has not changed between the critical case and the shutdown case being compared. Although fuel concentration is a difficult variable to measure with precision, it is relatively easy to assure that no significant change in concentration has occurred in repeating a reference case. The pulsed source technique used in this way is most advantageous.

TRANSPORT CALCULATIONS

Calculation of the Absolute Criticality of the

Molybdenum Reflected Solution Reactors

The reactor configurations considered are such that a two-dimensional cylindrical (r, z) calculation accurately describes the geometry. This means that the absolute multiplication factor of each reactor configuration could be obtained subject only to errors in the neutron cross section data if parameters such as the number of neutron energy groups, which are controllable, are adequate. Thus, a two-dimensional calculation for a given reactor configuration could serve as a standard for assessing the error in the critical height obtained from a corresponding one-dimensional radial calculation.

Since the delayed critical reactors were achieved with essentially the same fuel solution heights and Mo reflector heights, two-dimensional effects are minimized. This implies that one-dimensional cylindrical radial calculations with an appropriate axial buckling could be done for the large number of cases required and that the absolute multiplication factor of a given reactor could be obtained by applying a correction factor based on the standard two-dimensional result. However, reactor parameters, for ex-

ample, the effective delayed neutron fraction which are needed in the analysis of the pulsed source experiments are not sensitive to small errors in the absolute reactivity of a given reactor.

A two-dimensional cylindrical (r,z) S_n calculation using the DOT code (ref. 8) was made to obtain the absolute multiplication factor for one of the delayed critical reactors. This reactor had a hydrogen to uranium-235 atom ratio of 319.4. The experimental reactor (and reflector) height was 30.74 centimeters. This reactor was critical with the gap between the Mo reflector and the stainless-steel core vessel left void.

One-Dimensional S_n Calculations

The bulk of the calculations for this work were performed using a one-dimensional S_n transport code developed at the Lewis Research Center. Radial calculations were performed for the cylindrical critical systems with the fuel solution, stainless-steel containment vessel, gap (voided or filled with Mo or Cd) between the containment vessel and the reflector, and the Mo reflector explicitly included in the calculations. The leakage through the ends of the critical cylinders was accounted for through a buckling term. The eigenvalue for the system was taken to be the height of these cylindrical reactors for a given value of the fundamental prompt-mode decay constant α_0^P.

The calculational procedure followed is described in reference 6 and is summarized briefly here. One-dimensional, radial S_n calculations with an assigned value of α_0^P were made for a given critical experiment to obtain both the fundamental prompt-mode flux and the static-mode adjoint flux. These calculations yield the reactor height for the assigned value of α_0^P and the calculated reactivity $\rho_s^c(\$)$ by using either of the following two equivalent expressions:

$$\rho_s^c(\$) = \frac{K_{eff} - 1}{K_{eff} \ \bar{\beta}_0}$$

or

$$\rho_s^c(\$) = \frac{\alpha_0^P}{\dfrac{\bar{\beta}_0}{\Lambda_0}} + 1$$

where $\rho_s^c(\$)$ is the calculated value of the static reactivity in dollar units, K_{eff} is the

static-mode multiplication factor, $\overline{\beta}_0$ is the fundamental prompt-mode effective delayed neutron fraction, Λ_0 is the fundamental prompt-mode generation time in seconds, and α_0^P is the fundamental prompt-mode decay constant in inverse seconds. The value of α_0^P is varied (and correspondingly, the reactor height) until the reactivity $\rho_s^c(\$)$ is zero. This calculated height for $\rho_s^c(\$)$ equal to zero gives the calculated delayed critical configuration which is compared to an experimental value.

Nine neutron energy groups were used, eight fast and one thermal group which included an up-scattering transfer component. The fast group neutron cross sections for the UO_2F_2-H_2O fuel solutions, the type 304 stainless-steel reactor tank, and Cd were obtained using the GAM II code (ref. 9). The neutron cross sections for the Mo reflector were obtained from the ENDF/B data files and averaged over the fundamental mode fluxes and currents generated by GAM II for the various fuel solutions. The thermal group cross sections for all materials were obtained using the GATHER II code (ref. 10). The energy boundaries for each neutron energy group are given in table II. The delayed neutron data for U^{235} which are needed for the calculations are given in table III (ref. 11). All of the delayed neutrons are assumed to appear in energy group four where the energy varies from 0.22 to 0.82 MeV.

The one-dimensional radial S_n transport calculations were all performed using an S_4 angular quadrature order with elastic scattering treated through the P_1 order. The work of reference 12, although performed for an unreflected solution reactor, indicates that increasing the angular quadrature order, the elastic scattering order, or the number of thermal groups had a negligible effect on the eigenvalue determined for the delayed critical data.

TABLE II. - NEUTRON ENERGY GROUP

BOUNDARIES

Group	Energy	Lethargy
1	14.92 to 4.966 MeV	-0.4 to 0.7
2	4.966 to 2.231 MeV	0.7 to 1.5
3	2.231 to 0.821 MeV	1.5 to 2.5
4	0.821 to 0.224 MeV	2.5 to 3.8
5	0.224 MeV to 9.12 keV	3.8 to 7.0
6	9.12 keV to 454 eV	7.0 to 10.0
7	454 to 8.32 eV	10.0 to 14.0
8	8.32 to 0.414 eV	14.0 to 17.0
9	0.414 to 0 eV	17.0 to ∞

TABLE III. - DELAYED NEUTRON DATA FOR

URANIUM-235

[From ref. 11, p. 90; $\beta = 0.0065$.]

Delayed group	Decay constant, λ, sec^{-1}	Relative abundance, B_i/β	Delayed neutron fraction, B_i
1	0.0124	0.033	2.145×10^{-4}
2	.0305	.219	14.235×10^{-4}
3	.111	.196	12.740×10^{-4}
4	.301	.395	25.675×10^{-4}
5	1.14	.115	7.475×10^{-4}
6	3.01	.042	2.730×10^{-4}

Calculated kinetic parameters are presented in table IV for four fuel solutions having H/X values of 319.4, 447, 528, and 596. These fuel concentrations, whose atom densities are given in table V, were chosen so that fuel solution height and the Mo side reflector height was as nearly the same as possible for four critical experiments. The nominal experimental heights for these cylindrical systems were 30.2, 40.6, 50.5, and 70.8 centimeters. These calculations were performed for the gap between the core tank and Mo annular side reflector left void.

Figure 2 shows the variation in the fundamental prompt-mode effective delayed neutron fraction $\bar{\beta}_0$ with respect to H/X for the fuel solution. The value of β, the delayed neutron fraction, was taken to be 0.0065 for U^{235}. Figure 3 shows the fundamental prompt-mode generation time Λ_0 as a function of H/X. Figure 4 shows the ratio of the parameter $\bar{\beta}_0/\Lambda_0$ as a function of H/X. Since $\alpha_0^P = -\bar{\beta}_0/\Lambda_0$ at delayed critical, the negative of the data shown in figure 4 is also the fundamental prompt-mode decay constant α_0^P at delayed critical. Figure 5 shows the reactivity coefficient $\Delta\rho_s^c(\$)/\Delta h$ as a function of H/X. This coefficient gives the worth in dollars per centimeter change in reactor height at delayed critical. Figure 6 shows the reactivity coefficient $\Delta\rho_s^c(\$)/\Delta\alpha_0^P$ as a function of H/X. This coefficient gives the change in reactivity due to a change in the fundamental prompt-mode decay constant.

TABLE IV. - TABULATION OF CALCULATED DELAYED CRITICAL DATA

[Calculated values are for void gap between stainless-steel core tank and molybdenum annular reflector.]

Hydrogen to uranium-235 atom ratio, H/X	319.4	447	528	596
Effective delayed neutron fraction, $\bar{\beta}_0$	7.907×10^{-3}	7.752×10^{-3}	7.652×10^{-3}	7.564×10^{-3}
Neutron generation time, Λ_0, μsec	26.84	35.27	40.15	44.00
Ratio of effective delayed neutron fraction to neutron generation time, $\bar{\beta}_0/\Lambda_0$, sec^{-1}	294.6	219.8	190.6	171.9
Reactivity coefficient, $\Delta K_{eff}/\Delta h$, cm^{-1}	9.896×10^{-3}	5.207×10^{-3}	2.923×10^{-3}	1.408×10^{-3}
Reactivity coefficient, $\Delta\rho_s^c(\$)/\Delta h$, $\$\text{-cm}^{-1}$	1.252	0.672	0.382	0.186
Reactivity coefficient, $\Delta\rho_s^c(\$)/\Delta\alpha_0^P$, $\$\text{-sec}$	3.394×10^{-3}	4.550×10^{-3}	5.247×10^{-3}	5.817×10^{-3}

TABLE V. - ATOM DENSITIES OF FUEL SOLUTIONS REQUIRED TO

YIELD NOMINAL EXPERIMENTAL DELAYED CRITICAL HEIGHTS

OF 30.2, 40.6, 50.5, AND 70.8 CENTIMETERS

[Void gap between stainless-steel tank and molybdenum reflector.]

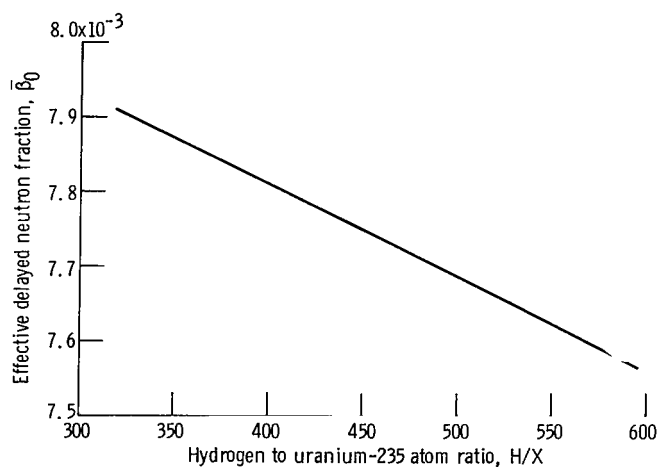| Nuclide	Hydrogen to uranium-235 atom ratio, H/X			
	319.4	447	528	596
	Delayed critical height, h, cm			
	30.2	40.6	50.5	70.8
	Atom densities, atoms/b-cm			
Hydrogen	0.066019	0.066233	0.066315	0.066367
Oxygen	.033453	.033435	.033427	.033422
Fluorine	4.436×10^{-4}	3.179×10^{-4}	2.695×10^{-4}	2.389×10^{-4}
Uranium-235	2.067×10^{-4}	1.482×10^{-4}	1.256×10^{-4}	1.114×10^{-4}
Uranium-238	1.506×10^{-5}	1.079×10^{-5}	9.15×10^{-6}	8.11×10^{-6}

Figure 2. - Effective delayed neutron fraction at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.
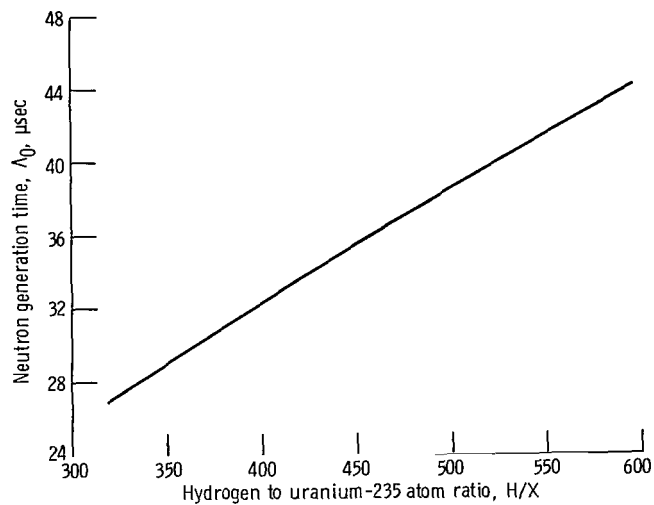
10

Figure 3. - Neutron generation time at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.
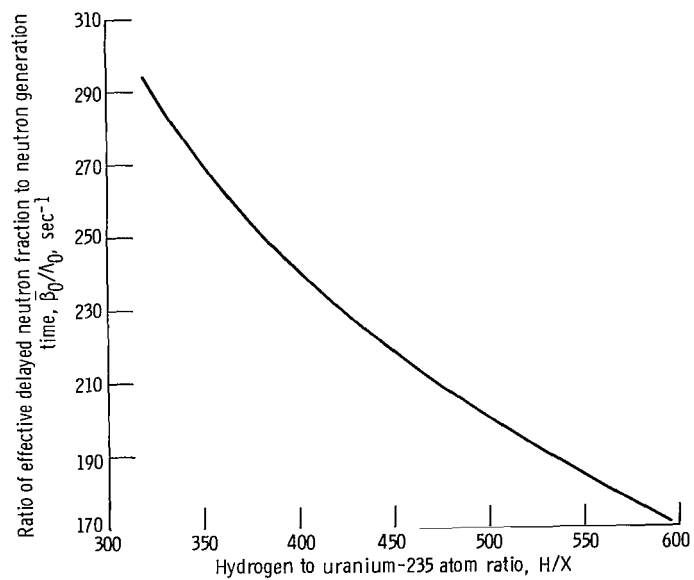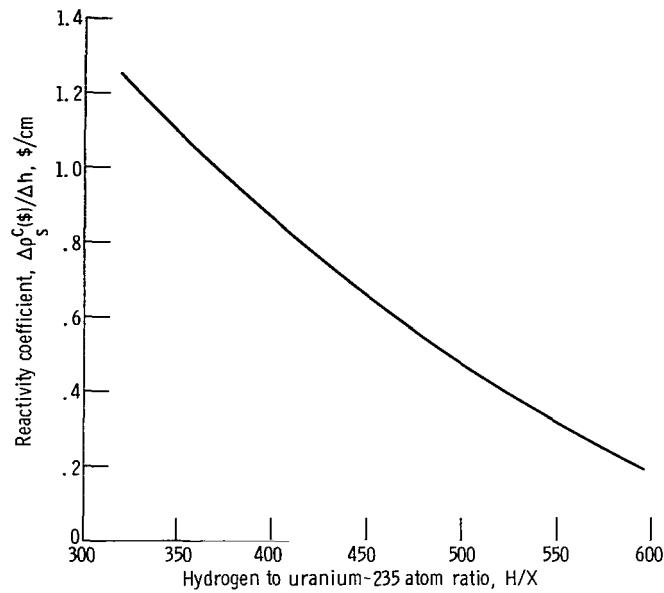

Figure 4. - Ratio of effective delayed neutron fraction to neutron generation time at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.

11

Figure 5. – Reactivity coefficient in dollars per centimeter of reactor height at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.
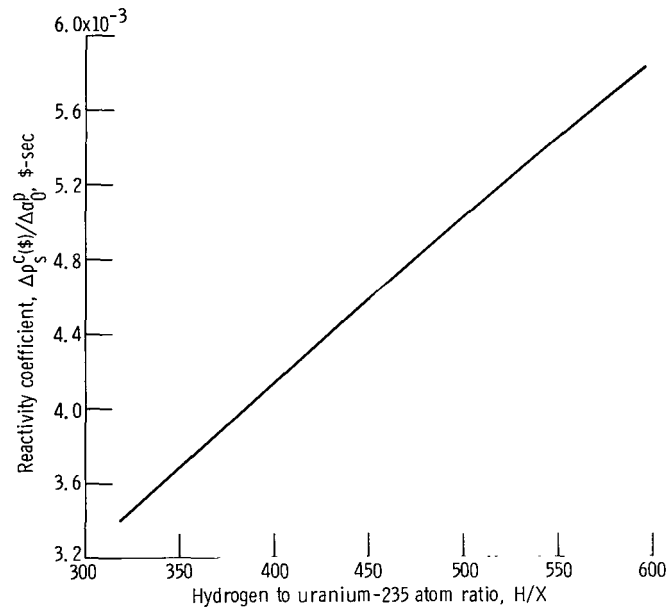

Figure 6. – Reactivity coefficient in dollar-seconds at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.

MOLYBDENUM CROSS SECTIONS

ENDF/B Molybdenum Cross Section Evaluation

The ENDF/B Mo data was compiled in 1966 by Pennington and Gajniak (ref. 13) and issued as ENDF/B material number 1025. The primary source of information for this evaluation by Pennington and Gajniak is the evaluation prepared by Schmidt (ref. 14). Pennington and Gajniak made the following modifications to the data compiled by Schmidt:

(1) Resonance parameters were taken from reference 15, plus some smooth capture cross sections were used from 4 eV to 1 keV.

(2) Inelastic scattering cross sections from 200 keV to 1.5 MeV, for four resolved levels, were obtained from reference 16.

(3) The (n,2n) and (n,3n) cross sections were obtained from reference 17.

(4) The data range was extended from 10 to 15 MeV: (1) the total cross section by using the data in references 18 and 19, (2) the (n,γ) cross section by assuming a 1/E behavior, and (3) the nonelastic cross section was assumed to be constant.

(5) Above the (n,2n) threshold, the inelastic cross section was assumed to be the difference between the nonelastic cross section and the sum of the cross sections for various other reactions.

(6) Nuclear temperatures were estimated for continuum inelastic scattering as in reference 20.

(7) Legendre coefficients for elastic scattering were obtained up to 6 MeV from reference 21 and above 6 MeV, the coefficients were obtained from optical model calculations.

The Mo cross sections have been further revised and issued as ENDF/B material number 1111. A number of changes in the ENDF/B #1025 evaluation were made. The most important change for the purpose of this study was in the high energy (n,γ) cross sections. This (n,γ) cross section data was reduced in value so that computed central reactivity worths in fast assemblies correspond to experimental reactivity determinations.

Result of an Integral Experiment to Test Inelastic Scattering
Cross Sections of Molybdenum

In order to test the validity of the ENDF/B #1111 Mo cross sections, an integral experiment has been performed employing a Mo sphere with a centrally located neutron source (ref. 4). Here, measured and calculated sphere leakage spectra in the energy range 0.5 to 11 MeV are compared as a test of the evaluated inelastic scattering cross

sections. A 54 Ci Am-Be source was used as a source of fast neutrons at the center of a 22.96-centimeter-diameter Mo sphere whose shell thickness was 8.38 centimeters. The Mo was in the form of a powder having an average density of 3.73 grams per cubic centimeter. This gave a shell thickness of 0.74 total mean free paths at 4 MeV. The leakage spectrum was measured at a distance of 200 centimeters using an NE213 liquid scintillator neutron spectrometer. Room scattered neutron backgrounds were accounted for by using a shadow cone. The leakage spectrum was calculated with 51 energy groups and an S_n discrete ordinate transport code.

The results of this integral experiment are shown in figure 7. The agreement between experiment and calculation is satisfactory and similar to the results obtained for the bare source. From this integral experiment, one can conclude that the inelastic scattering and total cross sections for Mo in the energy range from 0.5 to 11 MeV are
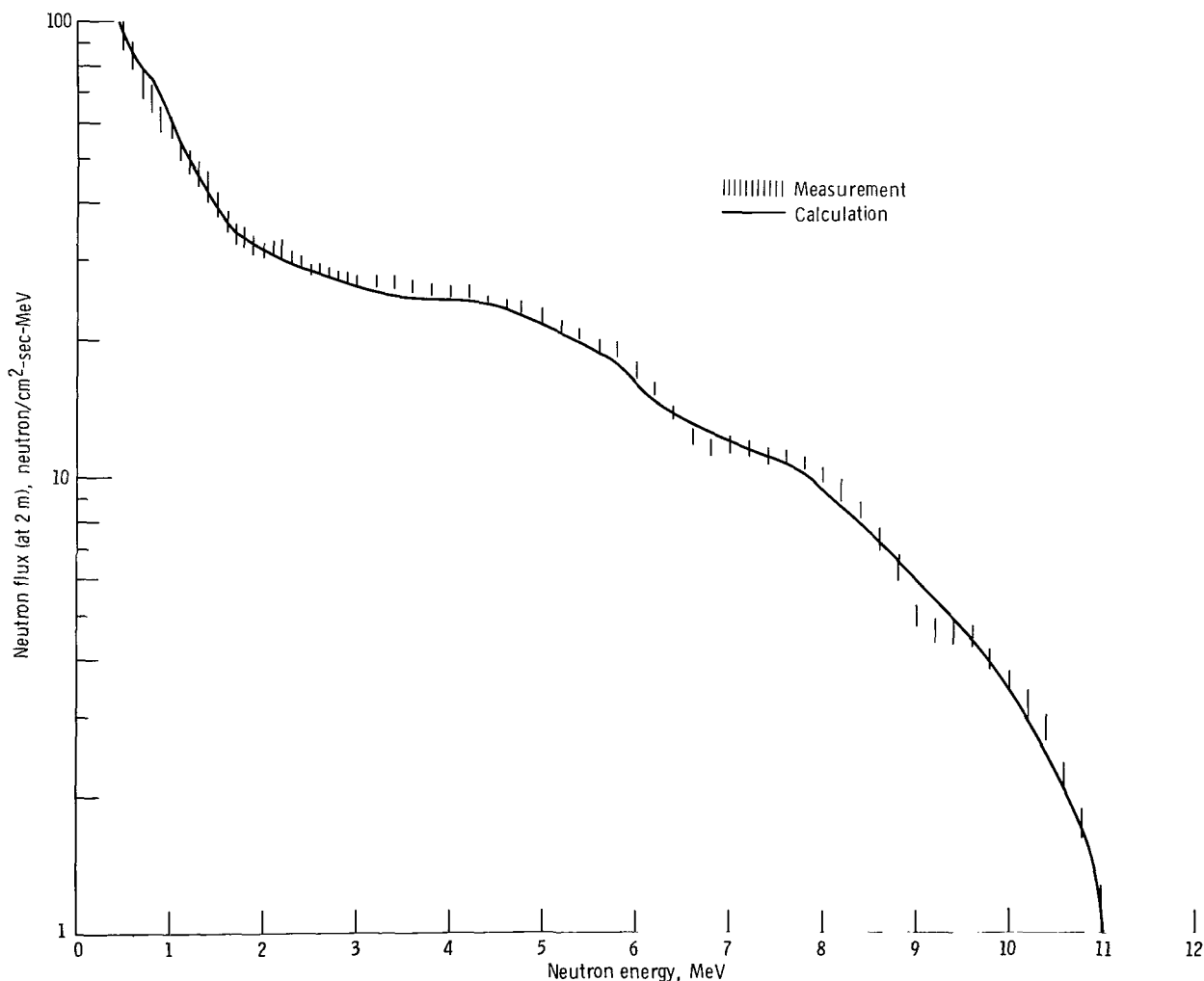

Figure 7. - Leakage spectrum for source enclosed by molybdenum shell (54 Ci Am-Be neutron source with intensity of 1.3×10^8 neutrons/sec).

14

satisfactory. However, the other Mo ENDF/B #1111 neutron cross sections need to be further tested by experiments such as the criticality experiments presented in this work.

Resonance Cross Sections

Molybdenum has a number of absorption resonances which yield an infinitely dilute resonance integral of 29.3 barns for the ENDF/B #1111 evaluation. Table VI lists the resonances which are the largest contributors to the resonance integral. The 44.7-eV resonance of Mo^{95} and the 131.3-eV resonance of Mo^{96} contribute 22.8 barns to the infinitely dilute resonance integral. The remaining resonances of Mo give small resonance integrals and it is expected that the energy and spatial self-shielding factors for these resonances will not differ appreciably from unity.

Since criticality calculations using Mo absorption cross sections computed using the GAM II approximate resonance calculation gave reactivity differences of $3.76 in the critical heights as compared to absorption cross sections based on the infinitely dilute resonance integral, an accurate evaluation of the resonance integral of the two main resonances of Mo was needed. Therefore, effective resonance integrals for the two most important resonances of Mo were computed using the following procedure: For each resonance considered, Doppler broadened absorption and scattering cross sections

TABLE VI. - RESONANCES WHICH ARE THE MOST

IMPORTANT CONTRIBUTORS TO THE INFINITELY

DILUTE MOLYBDENUM RESONANCE INTEGRAL[a]

Isotope	Resonance energy, eV	Resonance integral contribution for natural molybdenum, b
Mo^{95}	44.7	17.3
	358.5	.4
Mo^{96}	131.3	5.5
Mo^{97}	70.9	0.5
Mo^{98}	12.1 (p wave)	0.8
	467.4	.7
Mo^{100}	363.7	0.6

[a]Infinitely dilute resonance integral for ENDF/B
1111 Mo data is 29.3 b.

were computed for a temperature of 293 K. Then down-scattering transfer cross sections P_0 and P_1 were computed for Mo and water for the group structure chosen to represent each resonance. Finally, P_1S_4 transport calculations were performed for the actual reactor geometry with water representing the fuel solution. The results of these transport calculations for the effective resonance integral are presented in table VII.

The data in table VII indicate that the calculation of the effective resonance integral was not sensitive to the geometry used or to the height of the configurations. Calculations were also made in periodic slab geometry for the same thickness of water but varying the Mo thickness. The effective resonance integral for both the 44.7- and 131.3-eV resonances approached the infinitely dilute values as the Mo thickness became progressively smaller.

For the Mo reflected solution reactors considered in this report, the effective resonance integral of the 44.7-eV resonance of Mo^{95} was taken as 7.20 barns, while the effective integral of the 131.3-eV resonance of Mo^{96} was taken as 3.55 barns. Both resonances occur in group 7 of the energy group structure presented in table II. The ENDF/B #1111 Mo absorption cross sections in group 7 were adjusted so that the effective resonance integral of the two resonances considered was 10.75 barns.

TABLE VII. - EFFECTIVE RESONANCE INTEGRALS FOR THE 44.7-eV

RESONANCE OF MOLYBDENUM-95 AND THE 131.3-eV

RESONANCE OF MOLYBDENUM-96

[The calculations were P_1S_4 using a water region 12.7 cm thick and a molybdenum thickness of 10.14 cm.]

Resonance energy, eV	44.7 (Mo^{95})	131.3 (Mo^{96})
Inifinitely dilute resonance integral, b	17.3	5.5
Number of energy groups in S_n calculation	26	21
Computed resonance integral, b Cylindrical geometry, 30.45-cm height, no return current boundary condition	7.20	3.55
Slab geometry, 30.45-cm height, no return current boundary condition	7.15	3.54
Periodic slab geometry, 30.45-cm height	7.14	3.54
Periodic slab geometry, infinite height	7.16	----------

COMPARISON OF EXPERIMENTS AND CALCULATIONS

The comparison of the calculations with experiment will be made in terms of two quantities: (1) the delayed critical height, and (2) the fundamental prompt-mode decay constant. These two quantities were measured for each reactor configuration and are sufficient to establish the absolute criticality.

Delayed Critical Heights

Void gap between stainless-steel core tank and <u>molybdenum reflector</u>. The delayed critical height was measured for four fuel solution concentrations. These fuel solutions had hydrogen to uranium 235 atom ratios H/X of 319.4, 447, 528, and 596 which gave critical reactor heights of 30.74, 40.54, 50.55, and 70.42 centimeters, respectively. The experimental heights were corrected to a fuel solution temperature of 20° C and for loss of water by evaporation. Both of these corrections were small in comparison with the reactor heights.

The reactor with the smallest delayed critical height, that is, the reactor with a fuel solution having an H/X of 319.4, was computed using the two-dimensional transport program DOT. This calculation served as a standard for assessing the error in the absolute criticality determination obtained from a corresponding one-dimensional S_n transport calculation. This DOT calculation was made using cylindrical (r, z) geometry and used nine energy groups, a P_1 scattering order, and an S_4 angular quadrature. The experimentally determined height of 30.74 centimeters was used in this calculation. The computed effective static multiplication factor was 1.00054 which indicates, upon using the value of β_{eff} for this H/X from table IV, that the calculation is within 6.8 cents of the experimental multiplication factor of unity.

A one-dimensional cylindrical radial calculation for this same reactor yielded a delayed critical height of 29.27 centimeters. Using the reactivity coefficient $\Delta \rho_s^c (\$)/\Delta h$ for this H/X from table IV indicates that the one-dimensional calculation is too reactive by 1.84 dollars. Similar results can be expected for the other three critical reactors.

A comparison of experimental and one-dimensional calculated values of the delayed critical height, h, as a function of H/X for the four fuel solutions considered has been made in figure 8 and table VIII. All of these calculations predicted critical heights which were less than the experimental delayed critical heights. As shown in table VIII, these one-dimensional calculations were over-reactive by amounts of 184 to 115.4 cents for solution concentrations which varied from an H/X of 319.4 to 596.
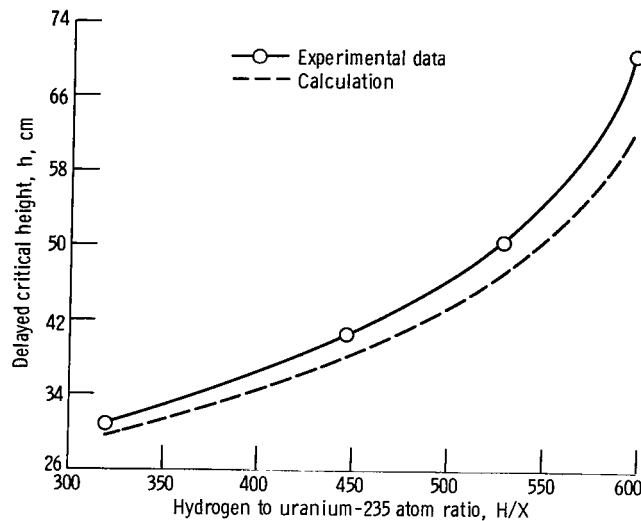
Figure 8. - Delayed critical height as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector. (Void gap between stainless-steel tank and molybdenum reflector.)

TABLE VIII. - TABULATION OF DELAYED CRITICAL HEIGHTS AND

FUNDAMENTAL PROMPT-MODE DECAY CONSTANTS

[Computed values are based on one-dimensional cylindrical radial calculations.]

Hydrogen to uranium-235 atom ratio, H/X	Delayed critical height, h, cm		Fundamental prompt-mode decay constant, α_0^P, sec^{-1}	
	Experimental[a]	Calculated	Experimental	Calculated
Void gap between stainless-steel tank				
319.4	30.7±0.1	29.27 ([b]184.0)	-281±2	-293
447	40.5±0.1	38.02 ([b]169.3)	-214±2	-218
528	50.6±0.1	47.53 ([b]115.4)	-187±2	-191
596	70.4±0.1	61.97 ([b]157.2)	-166±2	-173
Gap filled with molybdenum				
330	30.2±0.1	29.6 ([b]66.9)	-274±2	-285
540	49.9±0.1	47.8 ([b]71.8)	-185±2	-186
615	68.9±0.1	65.6 ([b]45.8)	-165±2	-166

[a]Experimental heights corrected to a fuel solution temperature of 20° C and for evaporation.

[b]Reactivity worth in cents of height difference between experimental and calculated values.

18

Molybdenum sleeve between stainless-steel tank and molybdenum reflector. - Three critical reactor experiments were performed for a Mo sleeve filling the 0.57-centimeter gap between the stainless-steel reactor tank and the Mo reflector. A comparison of experimental and one-dimensional calculated values of the delayed critical height, h, as a function of H/X is made in figure 9. These data are also presented in table VIII. These critical experiments were performed for slightly leaner fuel concentrations than those with the voided gap. The leaner fuel concentrations were necessary to maintain the fuel solution height the same as the Mo reflector height for these more reactive experiments.
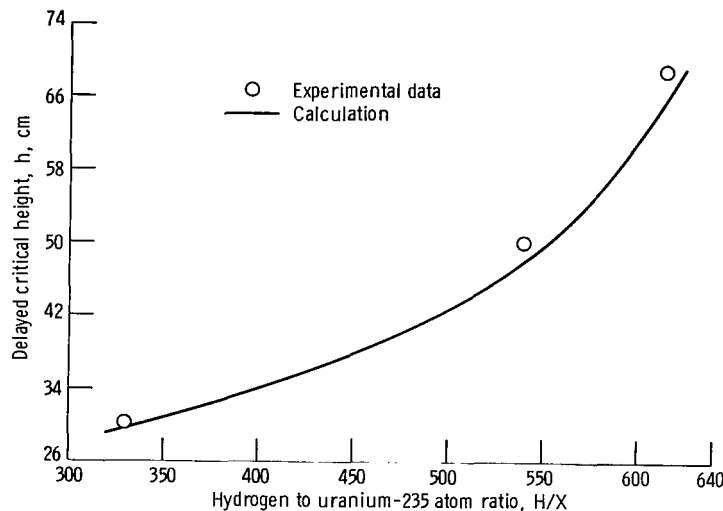

Figure 9. - Delayed critical height as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector. (Gap between stainless-steel tank and molybdenum reflector filled with molybdenum.)

At an H/X of 330, the calculated height is 0.56 centimeter less than the experimental height. At H/X of 540 and 615, the calculated heights are 2.08 and 3.27 centimeters less than the measured height. By using the calculated reactivity coefficient $\Delta\rho_S^c(\$)/\Delta h$ given in table IV, the difference of the calculated and experimental delayed critical heights in terms of reactivity is about 67, 72, and 46 cents for the reactors with H/X of 330, 540, and 615, respectively.

Fundamental Prompt-Mode Decay Constants

The fundamental prompt-mode decay constant α_0^P has been measured for the various critical systems considered by using the pulsed source technique. The measured

data are shown in figure 10 as well as being presented in table VIII. The decay constant α_0^P is primarily, for these reactors, a function of the fuel concentration. This can be seen to some extent in figure 10 since the experimental data all fell on one curve regardless of whether or not the gap between the core tank and reflector is left void or filled with a Mo sleeve.

A calculated curve is also shown in figure 10. The calculated values of α_0^P are in good agreement with the measured values. This can be best judged by using the reactivity coefficient $\Delta\rho_s^c(\$)/\Delta\alpha_0^P$ as given in table IV, which indicates that differences in
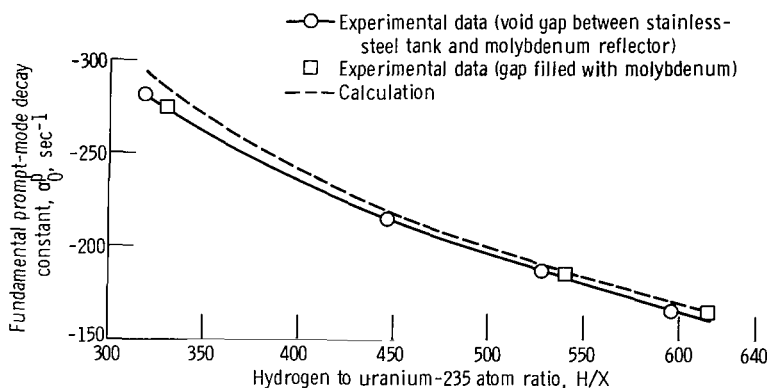

Figure 10. – Fundamental prompt-mode decay constant at delayed critical as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.

α_0^P correspond to differences in reactivity of less than 5 cents over the range of fuel concentrations used. Physically, this means that the nuclear cross section data used provide a good representation of the transport, moderating, and thermalization properties for the fuel solution.

Reactivity Worth of Molybdenum Sleeve and Cadmium Insert

Pulsed source subcritical measurements provide an excellent means for determining the reactivity worths caused by removal of the Mo liner or its replacement by a stainless-steel clad Cd sleeve for the three experiments for which the reactors were critical with the Mo filled gap. If the experiments are done in this manner, the fuel solution height is the same as the Mo reflector height and, thus, two-dimensional chimney effects are avoided. Experimental measurements of α_0^P are presented in table IX for various configurations.

20

TABLE IX. - EXPERIMENTAL FUNDAMENTAL PROMPT-MODE DECAY

CONSTANTS FOR MOLYBDENUM REFLECTED SOLUTION REACTORS

Experimental configuration	Hydrogen to uranium-235 atom ratio, H/X		
	330	540	615
	Fundamental prompt-mode decay constant, α_0^P, sec^{-1}		
Gap filled with molybdenum (delayed critical)	-274±2	-185±2	-165±2
Voided gap (subcritical)	-604±4 [a](-1.20)	-380±4 [a](-1.05)	-320±2 [a](-0.94)
Stainless-steel clad cadmium sleeve (subcritical)	-587±4 [a](-1.14)	-425±4 [a](-1.30)	-380±2 [a](-1.30)

[a]Subcritical reactivity worth in dollars relative to the reactor with gap
filled with Mo.

The method of Simmons and King (ref. 22) can be used to determine the experimental subcritical reactivities for the configurations involved since the departure from the delayed critical configuration is not too great. By this method, the subcritical reactivity is given by

$$\rho_s^c(\$) = - \frac{\alpha_0^P}{\left(\alpha_0^P\right)_{DC}} + 1$$

where α_0^P is the decay constant for the subcritical configuration and $\left(\alpha_0^P\right)_{DC}$ is the decay constant for the corresponding delayed critical configuration. Reference 5 gives a discussion of this method for determining subcritical reactivities.

Removal of the Mo sleeve from the delayed critical reactors with fuel solution H/X of 330, 540, and 615 gives an experimental reactivity worth for the Mo sleeve of -1.20, -1.05, and -0.94 dollars, respectively. Calculated reactivity worth of the Mo sleeve was -39, -49, and -53 cents for the systems with an H/X of 330, 540, and 615, respectively. The calculated reactivities are lower than the experimental values by factors of one-third to one-half. These data are shown in figure 11 along with the experimental data.

Replacement of the Mo sleeve by a stainless-steel clad Cd liner gave experimental reactivity worths of -1.14, -1.30, and -1.30 dollars for the reactors with an H/X of
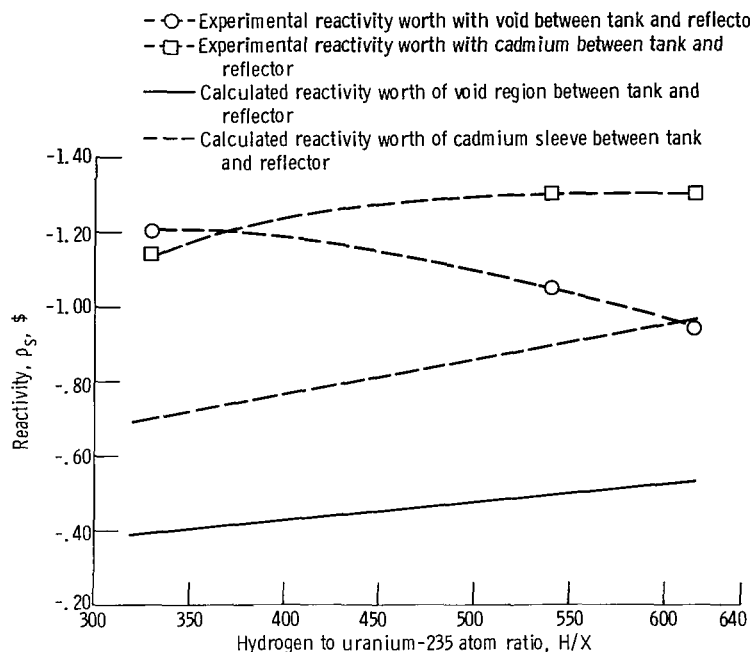
Figure 11. - Subcritical reactivity caused by removal of molybdenum sleeve as a function of hydrogen to uranium-235 atom ratio for NASA solution reactor with molybdenum side reflector.

330, 540, and 615, respectively. Calculated reactivity worths as given in figure 11 were -70, -89, and -97 cents for the corresponding values of H/X, respectively. The calculated reactivities are lower than the experimental reactivities by 39 to 25 percent.

There are a number of reasons why the calculated reactivity is much lower than the experimental reactivity. The most important factor is the inability of the one-dimensional calculations to properly treat the neutron streaming in the voided gap. Here, the two-dimensional calculations would be more appropriate. Another factor is that the measured thickness of the various inserts and claddings varies within fabrication tolerances and limitations.

Thermal Flux Distributions

The thermal neutron flux distribution in the radial direction at the reactor midplane was measured using dysprosium-aluminum foils for the reactor system having a fuel solution with an H/X of 319.4. The dysprosium activity was measured for the insert region between the stainless-steel core tank and the Mo reflector voided and for an insert consisting of a Cd sleeve clad with stainless steel. These measured distributions are shown in figure 12 and are normalized to the activity measured at the center of the reactor.
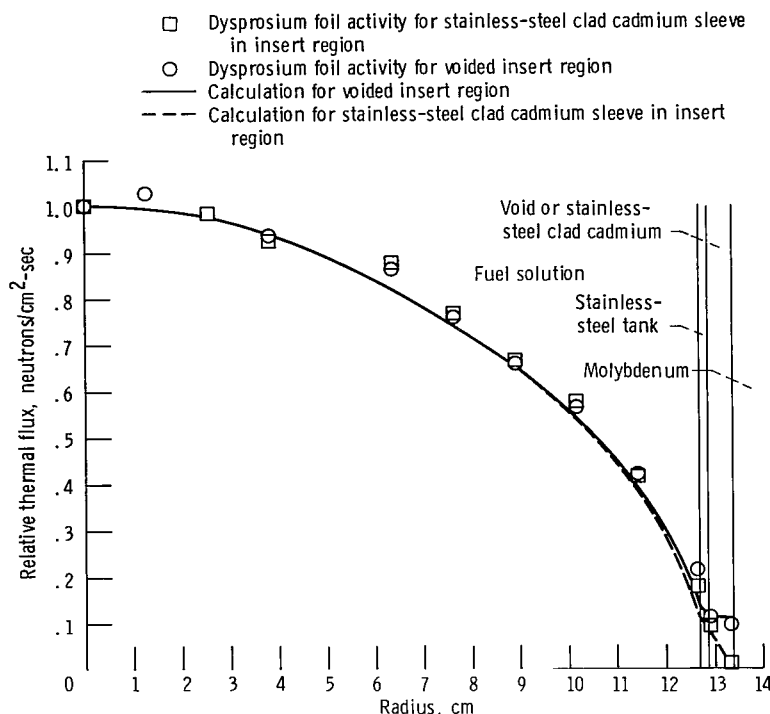
22

Figure 12. - Relative thermal flux distribution for reactor with hydrogen to uranium-235 atom ratio of 319.4.

Calculated radial thermal flux distributions are also shown in figure 12. The measured and calculated thermal flux distributions are in agreement with each other and exhibit the same features. The flux distributions for this reactor with and without the Cd insert are essentially the same within the core region. However, the flux distributions do differ, as expected, within the insert region. Thus the Mo reflector is effective primarily as a reflector of fast neutrons and has little effect on the thermal neutron flux distribution in the core because of its large thermal absorption cross section.

CONCLUSIONS

A number of conclusions result from this criticality study of molybdenum-reflected cylindrical uranyl-fluoride - water solution reactors:

1. A nine group two-dimensional cylindrical (r, z) transport calculation predicted the criticality to within 7 cents.

2. Nine group one-dimensional cylindrical radial transport calculations for the seven reactor configurations studied were from 0.45 to 1.84 dollars too reactive.

3. The effective resonance integral of molybdenum (Mo) must be accurately calculated for these reactors. The effective resonance integral of the 44.7-eV resonance of

Mo^{95} was computed to be 7.20 barns, while the effective resonance integral of the 131.3-eV resonance of Mo^{96} was computed to be 3.55 barns.

4. The calculated fundamental prompt-mode decay constants at delayed critical were in good agreement with the values measured for these reactors.

5. The differential reactivity worths of the Mo sleeve and stainless-steel clad cadmium (Cd) sleeve as obtained from one-dimensional S_n calculations were not in agreement with measured reactivity worths.

In summary, then, Mo cross sections from the ENDF/B #1111 evaluation are adequate for predicting the absolute multiplication factor of the seven reactors presented in this report.

Lewis Research Center,
    National Aeronautics and Space Administration,
        Cleveland, Ohio, May 4, 1972,
            112-02.

REFERENCES

1. Krasner, Morton H.; Davison, Harry W.; and Diaguila, Anthony J.: Conceptual Design of a Compact Fast Reactor for Space Power. NASA TM X-67859, 1971.

2. Mayo, Wendell; Klann, Paul G.; and Whitmarsh, Charles L., Jr.: Nuclear Design and Experiments for a Space Power Reactor. NASA TM X-67857, 1971.

3. Mayo, Wendell; and Lantz, Edward: Analysis of Fuel Loading Requirements and Neutron Energy Spectrum of a Fast Spectrum, Molybdenum-Reflected Critical Assembly. NASA TM X-52762, 1970.

4. Shook, D. F.; Fieno, D.; Ford, C. H.; and Alexander, R. L.: An Integral Test of Inelastic Scattering Cross Sections Using Measured Neutron Spectra from Thick Shells of Ta, W, Mo, and Be. Proceedings of the Third Conference on Neutron Cross Sections and Technology. AEC Rep. CONF-710301, Mar. 1971, p. 98.

5. Fox, Thomas A.; Mueller, Robert A.; and Fieno, Daniel: Criticality Study of NASA Solution Reactors with 25.4-Centimeter-Diameter Cylindrical Stainless-Steel Tanks. NASA TM X-2381, 1971.

6. Fieno, Daniel; Fox, Thomas A.; Mueller, Robert A.; and Ford, C. Hubbard: Analysis and Experiments with a Pulsed Neutron Source for an Unreflected Solution Reactor up to $50 Shutdown. NASA TN D-5487, 1969.

7. Kaufman, N. C.: GRIPE II - A Computer Program for the Analysis of Data from a Pulsed-Neutron Experiment. Rep. IN-1085, Idaho Falls National Reactor Testing Station, Sept. 1967.

8. Mynatt, F. R.: A Users Manual for DOT: A Two-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering. Rep. K-1694, Union Carbide Corp., 1967.

9. Joanou, G. D.; and Dudek, J. S.: GAM-II. A B_3 Code for the Calculation of Fast-Neutron Spectra and Associated Multigroup Constants. Rep. GA-4265, General Dynamics Corp., Sept. 16, 1963.

10. Joanou, G. D.; Smith, C. V.; and Vieweg, H. A.: GATHER-II. An IBM-7090 FORTRAN-II Program for the Computation of Thermal-Neutron Spectra and Associated Multigroup Cross Sections. Rep. GA-4132, General Dynamic Corp., July 8, 1963.

11. Keepin, G. Robert: Physics of Nuclear Kinetics. Addison-Wesley Publ. Co., 1965.

12. Fox, Thomas A.; Mueller, Robert A.; and Fieno, Daniel: Pulsed-Neutron-Source Studies with the NASA Zero Power Reactor II. NASA TN D-5956, 1970.

13. Pennington, E. M.; and Gajniak, J. C.: Compliation of ENDF/B Data for Magnesium, Titanium, Vanadium, Molybdenum, and Gadolinium. Rep. ANL-7387, Argonne National Lab., Mar. 1968.

14. Schmidt, J. J.: Neutron Cross Sections for Fast Reactor Materials. Part I: Evaluation. Rep. KFK-120, Institut fuer Neutronenphysik und Reaktortechnik, Feb. 1966.

15. Pevzner, M. I.; Adamchuk, Yu. V.; Danelyan, L. S.; Efimov, B. V.; Moskalev, S. S.; and Muradyan, G. V.: Investigation of Nuclear Levels by Neutron Spectroscopy. 1. Neutron Cross Sections of Molybdenum Isotopes in the 7-15000 eV Energy Range. Soviet Phys.-JETP, vol. 17, no. 4, Oct. 1963, pp. 803-808.

16. Smith, A. B.; and Hayes, R.: Fast Neutron Scattering from Elemental Mo, Sn, Sb and Te. Nucl. Phys., vol. A93, 1967, pp. 609-630.

17. Pearlstein, S.: Analysis of (n, 2n) Cross Sections for Medium and Heavy Mass Nuclei. Nucl. Sci. Eng., vol. 23, no. 3, Nov. 1965, pp. 238-250.

18. Hughes, Donald J.; and Schwartz, Robert B.: Neutron Cross Sections. Rep. BNL-325, 2nd ed., Brookhaven National Lab., July 1, 1958.

19. Hughes, Donald J.; Magurno, B. A.; and Brussel, M. K.: Neutron Cross Sections. Rep. BNL-325, 2nd ed., Suppl. No. 1, Brookhaven National Lab., Jan. 1, 1960.

20. Yiftah, S.; Okrent, D.; and Moldauer, P. A.: Fast Reactor Cross Sections. Pergamon Press, 1960.

21. Campbell, R. W.; Davis, S. K.; Alter, H.; Dunford, C. L.; and Berland, R. F.: Compilation, Evaluation, and Reduction of Neutron Differential Scattering Data, Vol. IV. Rep. NAA-SR-11980, vol. 4, Atomics International, Apr. 3, 1967.

22. Simmons, B. E.; and King, J. S.: A Pulsed Neutron Technique for Reactivity Determination. Nucl. Sci. Eng., vol. 3, no. 5, May 1958, pp. 595-608.

"The aeronautical and space activities of the United States shall be conducted so as to contribute . . . to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."

— NATIONAL AERONAUTICS AND SPACE ACT OF 1958

NASA SCIENTIFIC AND TECHNICAL PUBLICATIONS

TECHNICAL REPORTS: Scientific and technical information considered important, complete, and a lasting contribution to existing knowledge.

TECHNICAL NOTES: Information less broad in scope but nevertheless of importance as a contribution to existing knowledge.

TECHNICAL MEMORANDUMS: Information receiving limited distribution because of preliminary data, security classification, or other reasons.

CONTRACTOR REPORTS: Scientific and technical information generated under a NASA contract or grant and considered an important contribution to existing knowledge.

TECHNICAL TRANSLATIONS: Information published in a foreign language considered to merit NASA distribution in English.

SPECIAL PUBLICATIONS: Information derived from or of value to NASA activities. Publications include conference proceedings, monographs, data compilations, handbooks, sourcebooks, and special bibliographies.

TECHNOLOGY UTILIZATION PUBLICATIONS: Information on technology used by NASA that may be of particular interest in commercial and other non-aerospace applications. Publications include Tech Briefs, Technology Utilization Reports and Technology Surveys.

Details on the availability of these publications may be obtained from:

SCIENTIFIC AND TECHNICAL INFORMATION OFFICE

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
Washington, D.C. 20546