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RFP-1489

August 10, 1971

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MIXTURES OF PLUTONIUM AND ENRICHED
URANIUM IN SLAB GEOMETRY

C. Lee Schuske

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U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

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Printed in the United States of America
Available from the
National Technical Information Service
U. S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

August 10, 1971
Publication Date

RFP-1489
UC-46 CRITICALITY
STUDIES
TID-4500 - 56th Ed.

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Prepared under Contract AT(29-1)-1106
for the
Albuquerque Operations Office
U. S. Atomic Energy Commission

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ACKNOWLEDGMENTS

The helpful suggestions of Deanne Dickinson and Douglas C. Hunt in the preparation of this paper are gratefully acknowledged. L. E. Jackson is acknowledged for preparing the Computer Code DTF input data.

MIXTURES OF PLUTONIUM AND ENRICHED URANIUM IN SLAB GEOMETRY

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Abstract. The multiplication factors have been calculated for several systems composed of alternate slab regions of plutonium and enriched uranium. The slab systems are unreflected and consist of either pure metal or metal-water solutions at a hydrogen-to-fissile element atomic ratio of 50. The calculations were done with the Computer Code DTF in the S_4 approximation.

Slab geometry is used as a simplifying model to simulate possible complex array-storage combinations of plutonium and enriched uranium. The analysis indicates that certain combinations of pure metal are much more reactive than others. For the pure metal case with plutonium surrounded by uranium, the reactivity is 12 percent greater than when the plutonium surrounds the uranium. It is concluded that criticality safety limits for metal storage should include safety margins to account for the effect. For similar hydrogenous systems of plutonium and enriched uranium, only a slight effect was noted.

INTRODUCTION

In the development of nuclear criticality safety criteria for storage of fissile units, the question arises as to whether it is permissible to randomly mix units of plutonium (Pu) and uranium (U). For example, in a storage array of fissile units, should plutonium and uranium units be alternated or would another arrangement be less reactive.

In this paper, multiplication factor (k_{eff}) calculations are performed on unreflected systems composed of alternate slab regions of plutonium and uranium, in order to provide an answer to the question posed.

The approach avoids the costly Monte Carlo multiplication factor calculations necessary for finite arrays containing discrete fissile units. In addition, the reported approach permits a unique determination of system-composition effects by eliminating solid-angle effects of array calculations.

CALCULATIONAL PROCEDURES AND RESULTS

The Computer Code DTF-II^{1,2} with the Hansen and Roach³ 16-group neutron cross sections is used in all critical parameter calculations. All calculations are done in the S_4 approximation using a uniform 75-interval spatial mesh and a 10^{-3} convergence criterion. Table I contains the material and nuclear densities used in all calculations.

The first step in the calculational procedure is to obtain the individual critical thicknesses of plutonium and enriched uranium bare slabs. These are 3.68 and 7.16 centimeters (cm), respectively for the metal slabs, and 12.39 and 12.87 cm for the moderated slabs.

Next, a composite plutonium-uranium slab system is formed by placing a half-critical thickness plutonium slab next to a half-critical thickness uranium slab. Further composite systems are obtained by successively dividing the plutonium and uranium regions and alternating the plutonium and uranium. Figure 1 is a plot of k_{eff} as a function of the step homogenization for the bare metal slabs. Figure 2 depicts the systems reviewed. The limit of this process is a homogenous plutonium-uranium mixture. As shown in Figure 2, the total thickness of the plutonium and uranium regions are individually held constant. The calculations indicated that regardless of the degree of homogenization, the multiplication factor for composite systems always remains less than unity.

A second series of calculations was performed to determine if preferential locations of fissile material would lead to a more reactive configuration. Two configurations were chosen. The first consisted of plutonium in the center and uranium on the outside (Configuration I), and the second of uranium in the

¹ B. G. Carlson, W. J. Worlton, W. Gruber, and M. Shapiro. *DTF Users Manual*. UNC Physics - Mathematics 3321. United Nuclear Corporation, White Plains, New York. Volume I, November 1963. Volume II, May 1964.

² B. G. Carlson. *Methods in Computational Physics, Statistical Physics*. Volume I. Editors: Bernie Alder *et al.* Academic Press, New York. 1963. Pages 1-42.

³ Gordon E. Hansen and William H. Roach. *Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies*. LAMS-2543. University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico. 1960.

FIGURE 1. Plot of k_{eff} as a Function of Degree of Homogenization for Bare Metal Slabs. Refer to Systems A, B, and C in Figure 2.

TABLE I. Densities Used in Computations.

Material	Nuclear Density (atoms per barn centimeter)
Plutonium Metal	Plutonium-239 = 0.04941
Uranium Metal (93.0 percent Uranium-235 and 7.0 percent Uranium-238)	Uranium-235 = 0.04311 Uranium-238 = 0.00324
Plutonium Metal and Water Mixture ($\frac{\text{Hydrogen}}{\text{Plutonium}} = 50$)	Plutonium-239 = 0.00129 Hydrogen = 0.06517 Oxygen = 0.03258
Uranium Metal (93.0 percent Uranium-235 and 7.0 percent Uranium-238) and Water Mixture ($\frac{\text{Hydrogen}}{\text{Uranium}} = 50$)	Uranium-235 = 0.00120 Uranium-238 = 0.00009 Hydrogen = 0.06506 Oxygen = 0.03253

NOTE: Density of Plutonium = 19.6 grams per cubic centimeter (g/cm^3)

Density of Uranium = 18.1 g/cm^3

Density of Water = 1.00 g/cm^3

center with plutonium on the outside (Configuration II). The configurations are depicted in Figure 3 with the associated results given in Table II. Of the two configurations, the one with the plutonium in the center was much more reactive having a k_{eff} of 1.054, while the second configuration had a k_{eff} of only 0.948.

A final series of calculations was performed for bare moderated-metal systems. In the calculations, the fissile metal is homogenized in water at a hydrogen-to-fissile atomic ratio of 50. The configurations and the calculational results are shown in Figure 4 and Table II, respectively. The multiplication-factor values change in the same sense as in the previous bare pure metal case. The resultant reactivity changes are, however, much smaller.

In Figure 5, Configurations I and II, the total uranium region thickness is maintained at 3.582 cm (that is the uranium critical half-thickness). Only the plutonium thickness is varied to obtain critical systems. These calculations show an approximately 12 percent greater total slab thickness with the plutonium on the outside of the slab than when the plutonium is placed in the center.

Legend

Total thickness of each slab = 5.42 centimeters.

Total uranium thickness of each slab = 3.58 centimeters.

Total plutonium thickness of each slab = 1.84 centimeters.

FIGURE 2. Bare Metal Systems A, B, and C.

FIGURE 3. Bare Metal Slab Systems.

FIGURE 4. Bare Homogeneous Slabs at a Hydrogen-to-Fissile Element Atomic Ratio of 50.

DISCUSSION

As noted from the results, the most reactive position for the plutonium in a composite plutonium-enriched uranium system is in the center. This can be shown qualitatively as follows.

The reason simply stated is that plutonium has higher values of eta (η) (the average number of fission neutrons produced per absorbed incident neutron), than does uranium. When the plutonium is placed at the center, where the neutron flux is maximum, its effectiveness becomes enhanced.

The following expression can be written to describe the above statement:

$$k_{\text{eff}} = \frac{\sum_{ij} P_{ij} \eta_{ij} \phi_{ij}}{\sum_{ij} \phi_{ij}}$$

Where: k_{eff} is the system multiplication factor

P_{ij} is the system nonabsorption probability for energy group i of neutrons in the spatial interval j

ϕ_{ij} is the flux of neutrons in the i th energy group in the interval j

η_{ij} is the average number of fission neutrons produced per absorbed incident neutron

In the pure metal systems, this effect is more pronounced than in the moderated systems for the following reasons. In the pure metal systems, only the neutrons in the top six energy groups [which range down to 17 kiloelectron volts (keV)] contributed significantly to the total reactivity and, in these fast energy groups, the average ratio $\eta_{\text{Pu}}/\eta_{\text{U}}$ is about 1.23, while in the lower ten groups, the average ratio $\eta_{\text{Pu}}/\eta_{\text{U}}$ is about 1.12. The last 10 groups contribute significantly to the reactivity of the moderated case.

FIGURE 5. Critical ($k_{eff} = 1$) Bare Metal Slab Thickness as a Function of Fissile Locations.

TABLE II. The k_{eff} as a Function of Fissile Location (see Figures 3 and 4 for fissile arrangements).

Type of System	Configuration I (k_{eff})	Configuration II (k_{eff})	Figure No.
Bare Metal Plutonium and Uranium Slabs	1.054	0.948	3
Bare Moderated Plutonium and Uranium Slabs	1.002	0.996	4

CONCLUSIONS

As noted, any degree of homogenization of plutonium and enriched uranium slabs results in a multiplication factor less than that for corresponding systems of uranium or plutonium alone.

When plutonium is placed in the center of a composite slab system and the uranium is placed on the outside, the mixture results in the most reactive configuration. In the pure metal case, the effect is significant, but for the moderated case only a small effect occurs. The effect noted for bare metal should be considered in the development of criticality rules for mixed storage of plutonium and enriched uranium metal.