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# Criticality Studies of Enriched Uranium Metal in $UO_2(NO_3)_2$ Solutions

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#### THE DOW CHEMICAL COMPANY ROCKY FLATS PLANT DENVER, COLORADO

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### CRITICALITY STUDIES OF ENRICHED URANIUM METAL

IN UO2(NO3)2 SOLUTIONS

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#### ABSTRACT

Neutron multiplication measurements were made on 6.5-in. diameter cylindrical assemblies of enriched metal discs immersed in aqueous solutions of enriched  $UO_2(NO_3)_2$ . Diffusion calculations were made on homogeneous mixtures of the enriched metal with varying H:U atomic ratios.

#### ACKNOWLEDGMENTS

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#### 1. INTRODUCTION

Neutron multiplication measurements were made on cylindrical disc assemblies immersed in water and concentrated  $UO_2(NO_3)_2$  solutions. The thickness of the discs was changed to investigate various H:U atomic ratios.

Diffusion calculations were made on homogeneous mixtures of uranium and water to check the experimental data.

#### 2. EXPERIMENTAL MATERIALS

The measuring equipment used in these experiments included scalers, atomic model 1050A, coupled to G. E.  $B^{10}$ lined counters. A LiI(Eu) scintillator was also used.

#### 2.1 Materials

2.1.1 Moderator and Reflector A. Light water.
2.1.2 Fuel (Enriched Uranium) ~90% U<sup>235</sup>
A. Metal discs OD 5-5/8 in. and ID 3/4 in. Thickness ~0.060 in. Weight ~453 g per disc.
B. Fuel (Aqueous solutions of UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> ~90% U<sup>235</sup> enrichment) Concentrations: 102, 308 g of uranium per liter

The experimental arrangement is shown in Figure 1. The experiments were performed by building the metal core in both directions from the source.

Table I summarizes the results of the experiments. The densities were calculated assuming the core to be the smallest cylinder enclosing all of the uranium discs. This volume includes the steel rod down the center.

Experiments were performed with the discs immersed in water and  $UO_2(NO_3)_2$  solutions containing 308 g uranium per liter and 102 g uranium per liter.

Description of Core	Number of Discs in Experiment	Extrapolated Critical No. of discs	Core <u>H:U</u>	Approximate Density of <u>U in Core</u>
8 sheets/ 1/4 in H <sub>2</sub> O	96	~142	0.689	12.20
4 sheets/ $1/4$ in $H_2^0$	100	ω	1.43	8.99
5 sheets/ 1/4 in 308 g/1	96	œ	1. <b>21</b>	9,80
10 sheets/ 1/4 in 308 g/1	96	~150	0.635	12.66
6 sheets/ 1/4 in 102 g/l	96	~176	0 <b>.955</b>	10.87
Solid cylinder - H <sub>2</sub> O	88	97.5	0	17.8
Solid cylinder - 308 g/l	66	88	0	17.8

#### TABLE I

#### 4. CORRELATION OF EXPERIMENT AND CALCULATION

The diffusion calculations were multigroup, fundamental mode calculations done on the Bendix G-15 computer. The cross sections are a sixteen-group set obtained by G. E. Hansen and W. H. Roach of the Los Alamos Scientific Laboratory. The hydrogen cross sections given above were modified in the first five groups to give slightly more slowing down. This resulted in better agreement with experiment.

The calculation yields a critical buckling and an extrapolation length. The extrapolation length used is  $\frac{1}{\overline{\Sigma}_{tr}}$  where  $\overline{\Sigma}_{tr}$  is the flux weighted average of  $\Sigma_{tr}$ . This method gives the best agreement between experiment and calculation. Figure 2 shows the calculated buckling and extrapolation length vs. moderation.

Because the G-15 calculations were made on bare systems, a reflector savings method was used to correct the experimental results so that they could be compared with the calculations.

The reflector savings for uranium metal spheres reflected by water was obtained by taking the reduction in diameters of the metal spheres<sup>(1)</sup> due to specific amounts of water reflector (see Figure 3). It was assumed that the reflector savings for the metal spheres would closely enough opproximate our experiments since the experiments were highly concentrated in uranium. See Table I for the fuel densities.

(1) H. C. Paxton, "Critical Masses of Fissionable Metals as Basic Nuclear Safety Data", USAEC Report LA-1958, 1955.

In correlating the water experiments with the calculations all experimental core dimensions were increased by the appropriate reflector savings from the solid curve in Figure 3. Figure 4 gives a comparison of an experiment and calculations for an H:U = 0.69. The agreement is seen to be adequate considering experimental errors and approximations used in the calculations.

In order to get a reflector savings for the solution cases, the last two experiments in Table I were used. The difference in the length of the solid cylinders is assumed to be the difference in reflector savings between water and 308 g uranium per liter solution. This yields 2.07 in. as the reflector savings due to the solution for these cylinders. In correlating with calculations, the solution around the sides of the cylinders was assumed to be equivalent to water. The solution on the ends was corrected for by using 2.07 in. as the reflector savings. Figure 5 gives a comparison between experiment and calculations for 308 g uranium per liter solution at an H: U = 0.635 the agreement is again seen to be adequate.

There was not enough experimental data to obtain a reflector savings value for the 102 g unchium per liter solution. In this case the reflector savings was assumed to be a linear function of concentration, thus a value between 2.07 in. and the solid curve of Figure 2 was used. Figure 6 shows the correlation for 102 g uranium per liter at an H:U = 0.955.

Figure 7 shows a comparison between critical height vs. moderation for 5-5/8-in. diameter cylinders. The calculated curve was obtained from mixtures of water and  $U^{235}$  only, and appropriate reflector savings were subtracted from the bare values.

Figures 8, 9, and 10 show a comparison of the calculations with curves derived from experiments. (2) The calculated curves are seen to be 10% to 20% higher in mass than the experimental curves in most cases.

The calculations were fundamental-mode diffusion calculations. Some  $S_4$  calculations were done using the same cross as for the diffusion calculations. The diffusion calculations agreed within 5% with the  $S_4$  calculations. The  $S_4$  calculations were infinite cylinders including the steel rod down the center. Somewhat better agreement with the diffusion calculations could be obtained for infinite cylinders if the steel rod were considered void in the diffusion approximation. A reflector savings value obtained by  $S_n$  at an H:U of 1 agreed with the solid curve of Figure 3.

#### 5. CONCLUSION

The G-15 diffusion calculation compares well with the  $S_n$  calculation.

(2) H. C. Paxton, personal communication.

The minimum critical mass of these systems occurred for a single solid mass of uranium metal either in water or  $UO_2(NO_3)_2$  solutions. The calculated critical masses compared reasonably well with the observed extrapolated critical masses.





Figure 1











Figure 7



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Figure 8



H:U Atomic Ratio

Figure 9

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Figure 10