Calculation of Critical Experiments involving $U(37)O_2F_2$ Solution

K. L. Goluoglu and A. W. Krass

Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6271, USA goluoglukl@ornl.gov

INTRODUCTION

Critical experiments were conducted at the Oak Ridge Critical Experiment Facility (ORCEF) to determine the critical concentration for an unreflected 69.2-cm-diameter sphere of UO_2F_2, at an enrichment of ~37 percent U^{235}, by weight. These experiments were a continuation of previous efforts to determine critical dimensions for fissile materials in simple geometry. Some of the earlier experiments in this vessel have been published as part of the OECD handbook.[1]

The reports concerning these experiments have only recently become available. Until August 2005, Refs. 2 and 3 were still classified. [2,3] These documents, along with experimental logbooks[4] and unclassified papers available on the experimental campaign and facility are being used to generate a computer model for this critical experiment.

BACKGROUND

A program to determine the critical conditions of aqueous solutions of uranium salts enriched in U^{235} was initiated in 1947. The experiments were part of a long range program of ORNL personnel at the ORCEF, and numerous reports document the results of these experiments. The particular series of experiments dealt with in this paper were performed between 1958 and 1960.

The experiments being analyzed involve a $U(37)O_2F_2$ solution in a sphere. The uranium was enriched to ~37 percent U^{235} by weight. The vessel was constructed of aluminum, with an inside diameter of 69.20 cm and a wall thickness of 0.64 cm. The sphere was unreflected, and was the same sphere used for the experiments originally reported in ORNL-2609[5] and later analyzed in the OECD report HEU-SOL-THERM-013.[6] The measured volume of the tank was 173.6 L.

The $U(37)O_2F_2$ concentration in the solution was adjusted such that when the aluminum vessel was completely filled the multiplication factor was greater than unity and the excess reactivity was measured by means of a positive reactor period. The final solution used to achieve criticality had a density of 1.057 g/cm^3. The solution contained 49.19 mg of U/g of solution. The U^{235} content reported was 36.96 wt%. This value has some uncertainty associated with it, which is noted in the Y-12 reports. The U^{235} density is reported as 19.19 mg/cm^3, with a total mass of 3331 g. The reported H:U^{235} ratio is 1343.

The experimental k_{eff} for the critical sphere was measured to be 1.0011. The experimenters also published an adjusted k_{eff} to represent the critical mass only, to account for small differences between an idealized system and the physical reality of the experiment. The adjustment accounts for things such as the small amount solution in the polar port of the sphere and the aluminum walls of the sphere. The experimenters compared their calculated values for a solution in an idealized aluminum shell to the 1.0011 measurement.

CURRENT RESULTS AND FUTURE WORK

Preliminary calculations for this system have been performed with the CSAS25 control module of the SCALE 5.0 code system.[7] Calculations were performed using either NITAWL or CENTRM for cross-section processing. 16-energy-group Hansen-Roach and 238-energy-group ENDFB-VI cross sections were used. The results are shown in Table 1.

The results shown in Table 1 are for a simple case, and do not model the structure or room of the experiment other than the aluminum tank. The calculated results by the experimenters, ca. 1973, using Hansen-Roach cross-sections and ANISN, show a similar under-prediction of k_{eff} when calculating the solution sphere in the aluminum shell (k_{eff}=1.0005).

Table 1. Preliminary Results for a $U(37)O_2F_2$ Solution Sphere in Aluminum Shell

Case	Cross-section set	Cross-section code	Calculated k_{eff}
1	Hansen-Roach	NITAWL	1.0095±0.002
2	238group	CENTRM	0.9971±0.0013
3	238group	NITAWL	0.9996±0.0017

Work is currently underway to determine more precisely the possible sources of the excess reactivity and to quantify the uncertainties associated with the experiment. The problem is also being examined in MCNP5.

REFERENCES

1. *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, NEA/NSC/DOC(95)03, September 2005 Edition, Organization for Economic Co-Operation and Development.
2. D. W. Magnuson, *Critical Concentration of Uranium Solution: Critical Concentration of UO_2F_2 Aqueous Solution in an Unreflected 69.2-Centimeter-Diameter Aluminum Sphere*, Y-DR-118, Oak Ridge Y-12 Plant, Oak Ridge, TN, November 3, 1973.
3. D. Callihan, *Critical Dimensions of Aqueous Solutions of $U(37)O_2F_2$*, Y-DR-135, Oak Ridge Y-12 Plant, Oak Ridge, TN, February 7, 1975.
4. The ORNL Critical Experiments Logbooks, available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory at: http://www-rsicc.ornl.gov/rsiccnew/criticallist.htm
5. J. K. Fox, L. W. Gilley, R. Gwin, and J. T. Thomas, *Critical Parameters of Uranium Solutions in Simple Geometry*, Neutron Physics Annual Progress Report, ORNL-2609, Oak Ridge National Laboratory, 1958.
6. M. Pitts and F. Rahnema, *Unreflected 174-Liter Spheres of Enriched Uranium Nitrate Solutions*, HEU-SOL-THERM-13, *International Handbook of Evaluated Criticality Safety Benchmark Experiments*, September 2005 Edition, Organization for Economic Co-Operation and Development.
7. *SCALE: A Modular Code System for Performing Standardized Computer Analysis for Licensing Evaluations*, ORNL/TM-2005/39, Version 5, Vols. I-III, April 2005. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-725.