BNWL-1522-5
UC-80,
Reactor Technology

TECHNICAL ACTIVITIES REPORT
AEC REACTOR DEVELOPMENT
AND TECHNOLOGY PROGRAMS
OCTOBER, NOVEMBER, DECEMBER 1971
JANUARY, FEBRUARY, MARCH 1972

by

Staff of Battelle-Northwest

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May 1972

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FOREWORD

This report covers all programs except the Waste Studies Program conducted at the Pacific Northwest Laboratory for the U.S. Atomic Energy Commission's Reactor Development and Technology Division. The objective of the Technical Activities Report is to inform the scientific community in a timely manner of the technical progress made on the programs. The report contains brief technical discussions of accomplishments in all areas where significant progress has been made during the reporting period. The results presented should be considered preliminary and do not constitute final publication of the work. A list of publications and papers is given in the report. Anyone wishing to obtain additional information on the work presented is encouraged to contact the author directly.

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1. SUMMARY

HIGH TEMPERATURE LATTICE TEST REACTOR

The High Temperature Lattice Test Reactor has been operated successfully for four years in support of the high-temperature gas-cooled reactors and molten salt breeder reactors. The reactor is currently defueled and in standby status. It was one of the first reactors in this country to use a high speed digital system for process control. Only two interruptions in the schedule resulted from hardware failures during five and a half years of almost continuous service. With the exception of the vertical safety rods, the original materials selected for reactor service have functioned without failure in the high temperature environment.

REACTIVITY EFFECTS IN UO₂-PuO₂ LATTICES DUE TO PuO₂ GRAIN SIZE

The reactivity effect of 25 micron PuO₂ particles relative to homogeneous fuel has been calculated for the Hanford UO₂-PuO₂ lattice experiments. The results indicate a significant trend, with lattice pitch, which is smallest for tight pitches and largest for loose pitches.

NUCLEAR GRAPHITE

An experiment designed to measure the effect of tensile stress on graphite dimensional changes during irradiation was discharged from the ETR. Similar samples under a compressive stress are continuing to be irradiated and will be discharged in February 1972.

Final experimental measurements of the radiolysis of He diluted mixtures of CO and H₂O have been made. A variety of theoretical treatments have been considered and final analysis based on collision theory is in progress.

A capsule for measuring the effect of "cold seeding" on the irradiation behavior of graphite was charged into the ETR in November 1971 and is scheduled for discharge in April 1972.

STEADY STATE AND TRANSIENT SUBCHANNEL CODE DEVELOPMENT AND DATA ANALYSIS

A fuel heat-transfer model was developed for inclusion into the COBRA-IIIB computer program. Only radial conduction within the fuel is considered. Axial conduction is ignored because axial temperature gradients are usually small. Circumferential gradients can be somewhat larger but conduction in this direction is not included because of the necessity to maintain a reasonable limit on core storage for fuel temperatures.

Experimental measurements of local velocity, turbulence intensity and Eulerian time scale were completed in several channels, representing a portion of a multi-rod bundle, using the two-component laser-Doppler flowmeter. Preliminary conclusions from these data indicate that velocity profile, intensity and scale were nearly independent of Reynolds number over the range 50,000 to 200,000. Detailed maps of turbulence intensity and velocity profile showed that a maximum in intensity occurred between the subchannel center and the rod gap in regions away from the channel walls and that secondary flows probably exist in a symmetric pattern which moves fluid into the center of the gap between channels and out near the walls. However, this flow is very small, less than 1% of the axial flow. The addition of a grid spacer increased the intensity by about 100% and decreased the scale. These effects gradually decayed downstream of the grid but were still apparent at $L/D = 20$ downstream of the grid.

CRITICAL EXPERIMENTS WITH HOMOGENEOUS PuO_2 -POLYSTYRENE AT 50 H/Pu

Results and analyses are presented from the latest series of experiments in which the critical parameters of plutonium mixtures is determined over the under-moderated region of the neutron energy spectrum. This latest series of experiments was conducted with PuO_2 -polystyrene fuel having an atomic H/Pu ratio of 49.6 and a ^{240}Pu isotopic content of 18.35 wt%.

In addition to providing basic criticality data, the experiments were designed to: (1) establish the degree of correlation between the PuO₂-polystyrene and the plutonium nitrate solution fueled critical experiments performed at the Hanford Critical Mass Laboratory, (2) provide data for checking previously reported differences obtained by the neutron diffusion theory code HFN and the neutron transport theory code DTF-IV in calculating critical sizes of plutonium-water systems between about 500 and 1000 g Pu/l, (3) substantiate that high exposure, subcritical, fixed volume, Pu-water systems can be made critical by dilution with water.

Based on the data derived from these experiments at 49.6 H/Pu, the HFN code was found to accurately calculate the Pu-water systems at 521 g Pu/l and should be used at this and lower concentrations in preference to the DTF-IV code for calculating criticality parameters. As with the plutonium nitrate solutions at high concentrations, the DTF-IV neutron transport theory code calculations resulted in too small a critical size; however, the data obtained from the PuO₂-polystyrene experiments correlated well with those that have been obtained from plutonium nitrate solutions. The experiments also showed that, for high exposure Pu-water systems, a given size vessel could be just critical ($k_{\text{eff}} = 1.0$) at three different concentrations below about 1000 g Pu/l. Consequently, a critically safe vessel could be made critical by dilution alone if the ²⁴⁰Pu content was high enough.

CRITICALITY OF HIGH BURNUP PLUTONIUM

Correlation of the experimental data for the high burnup plutonium solutions were made using GAMTEC and KENO codes. ENDF/B cross sections were utilized. Concentration of plutonium was varied from 40 to 140 g Pu/l with the plutonium containing 42.9% ²⁴⁰Pu. Effects of the various plutonium isotopes and ²⁴¹Am were calculated. The calculated k_{eff} values average about 1% above unity.

THEORY-EXPERIMENT TESTS USING ENDF/B VERSION II CROSS SECTION DATA

The GAMTEC, MC², and THERMOS codes were used to derive multigroup constants, using data from the ENDF/B-II library, for a number of known critical configurations of fuel chosen for their geometric simplicity and variety of fissile material content. These data were subsequently used in the HFN diffusion theory code to compute effective multiplication factors for a variety of PuO₂-polystyrene, and mixed Pu-U oxide-polystyrene, mixtures. Also, a number of plutonium nitrate, and mixed Pu-U nitrate solutions were examined.

2. HIGH TEMPERATURE LATTICE TEST REACTOR

R. G. Clark

The High Temperature Lattice Test Reactor (HTLTR) is a unique critical facility specifically built for studying the neutronics of reactor lattices at elevated temperatures and is the only reactor that has been operated routinely to 1000°C. Completed in 1967 at a cost of \$3.2 million the facility has been operated by Pacific Northwest Laboratory for the United States Atomic Energy Commission to develop information needed in economics and safety studies for high temperature power reactors.

Briefly the reactor itself is made up of cored, nuclear grade graphite bars which are stacked, keyed, and doweled together to form a solid 10 ft cube weighing over 35 tons, unfueled. A 5-ft-square by 10-ft-long central section is removable to permit loading of different test assemblies. The central test cell of an experimental core can thereby be provided with a flux environment characteristic of its own kind of cells in a critical array. Rated at 2 kW for nuclear operation, it was heated electrically and used nitrogen gas for an inert atmosphere and coolant. A 2 MeV positive ion accelerator provided neutrons for startups.

All the sensors, the transducers and controllers for closed loop control, nuclear flux measurements and data handling were interfaced to a general purpose high-speed mini-computer, a Digital Equipment Corporation PDP 7/9, which is an 18-bit machine with a 16,000 word core memory and is equipped with three low speed magnetic tape units and two industrial type teletypewriters.

Five major experiments were conducted in HTLTR at temperatures up to 1000°C. Four of the experiments (1,2,3,4) were in support of the High Temperature Gas-cooled Reactor (HTGR) Program and one experiment⁽⁵⁾ was in support of the Molten Salt Breeder Reactor (MSBR) program. In these experiments the temperature dependence of the infinite medium neutron multiplication factor, k_{∞} , was determined for different lattice compositions and

configurations. In addition, information on the thorium doppler coefficient and on the absorption cross sections of selected neutron absorbers (poisons) was obtained as a function of temperature.

The operating history of HTLTR is shown in Figure 2.1, where the reactor temperature is plotted as a function of calendar time. Initial fuel loading to critical occurred in November of 1967, shown as NUCLEAR START-UP. The first HTGR experiment using ^{235}U C_2 test fuel was completed in 1969, with the reactor being heated to 1000°C for the first time in August 1969. In the following 25 months, the remaining four experiments were completed. During these experiments, over 1880 hours were logged with the reactor assembly at 1000°C , and in the last experiment, a test lattice containing substantial amounts of plutonium oxide was successfully studied at temperatures up to 1000°C .

HTLTR was one of the first nuclear reactors in this country to use a high speed digital system for process control and its use contributed significantly to stable control and precision results. For example, it became possible as experience was gained to routinely maintain thermal equilibrium in the entire reactor for periods of three hours or more, such that the temperature of the central section (test volume) remained within $\pm 0.05^\circ\text{C}$ and the reactor pressure was held constant to within ± 0.03 torr. The excess reactivity of the reactor could be reproduced to $\pm 0.01\%$ during these intervals.

Only two interruptions in the schedule resulted from hardware failures in the computer during the five and a half years of its service and during which time it was almost incontinuous operation. Potential operating or safety problems at these times were precluded by audit features in the software package.

With one exception, materials selected for reactor service have functioned without failure in the high temperature environment. This includes 80 thermocouples with inconel sheaths, hangars of Hastelloy-B to support the insulating brick, and TD nickel for the structural portion of the horizontal control rods and the vertical safety rods. The exception has

FIGURE 2.1. HTLTR Experimental History

been the vertical safety rods, which were routinely cycled out of and into the high temperature region and which had to be redesigned to accept the thermal shock without deforming. All fuel and graphite appear to be in their original condition.

The reactor is now defueled and on standby status.

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3. REACTIVITY EFFECTS IN UO_2 - PuO_2 LATTICES DUE TO PuO_2 GRAIN SIZE

V. O. Uotinen

The development and evaluation of calculational methods for use in the physics design of plutonium recycle cores have been based largely on the results of lattice experiments^(1,2,3,4) performed with UO_2 - PuO_2 rods. Most of these fuel rods contained mixtures of UO_2 and PuO_2 powders. A known source of error in most past analyses of these experiments has been the assumption that the fuel is homogeneous. While this assumption has been recognized as a source of error, the magnitude of this error has only been estimated until recently.

The PuO_2 powders used in the Hanford rods (UO_2 -1.5 wt% PuO_2 ; UO_2 -2 wt% PuO_2 ; and UO_2 -4 wt% PuO_2)⁽¹⁾ contained particles ranging in size from essentially zero microns to about 44 microns. It has been determined experimentally that the "average particle diameter" is ~25 microns.⁽²⁾

It has been demonstrated experimentally⁽⁵⁾ that having the PuO_2 in the form of particles of this size can result in significant changes in reactivity relative to homogeneous fuel. This effect is the result of self-shielding in the PuO_2 particles. Self-shielding of ^{239}Pu is important in the thermal energy range and results in a negative reactivity effect. Self-shielding of ^{240}Pu is important in the energy range around the 1.056 eV resonance of ^{240}Pu and results in a positive reactivity effect. The net effect is the combination of these competing effects and depends on such things as the size of the particles, the Pu enrichment, the relative amounts of the ^{239}Pu and ^{240}Pu isotopes, and the water-to-fuel volume ratio.

The reactivity effect of 25 micron PuO_2 particles relative to homogeneous fuel has been evaluated for the approximately 30 Hanford UO_2 - PuO_2 lattice experiments using the computer codes GRANIT⁽⁶⁾ and EGGNIT.⁽⁷⁾

These codes permit one to calculate the self-shielding within finite absorber grains embedded in a matrix of diluent material. The codes GRANIT and EGGNIT were used to calculate unit cell four-group constants for selected lattices assuming PuO_2 grain sizes of zero and 25 microns. These four-group constants were used in a one-dimensional diffusion theory calculation to obtain values of k_{eff} . The change in calculated k_{eff} between cases having zero and 25 micron PuO_2 particles is shown in Figure 3.1 for each fuel type.

The reactivity effect is in the range 0.1-0.7% $\Delta k/k$ and is negative for all cases studied. However, it can be seen that at tighter pitches, especially for lattices containing fuel with a large amount of ^{240}Pu , the effect would be expected to be positive. The results indicate a significant trend with lattice pitch, the net effect being smallest for tight pitches and largest for loose pitches. This trend is in agreement with the calculations reported recently by Hellens⁽⁸⁾ for two mixed oxide lattices. The results of this study show that it is important to consider the reactivity effect due to the finite grain size of the PuO_2 in these fuels when using these experiments as the basis of methods evaluation and development.

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FIGURE 3.1. Reactivity Effect of Finite PuO_2 Grains Relative to Homogeneous Fuel

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4. NUCLEAR GRAPHITE

G. L. Tingey

IN-REACTOR CREEP

W. J. Gray

The effect of stress on the dimensional changes which occur during irradiation of graphite is being evaluated in our constant-stress creep studies. A description of the irradiation schedule for this experiment was contained in a recent quarterly report.⁽¹⁾ This irradiation schedule has recently been revised to comply with AEC directives to bring the program to an orderly termination.

The tensile creep capsule was discharged as planned from the ETR in November after a single cycle of irradiation. At present, the capsule is being stored at the National Reactor Test Site pending determination of its usefulness to future programs. The two compressive creep capsules, under irradiation at 550 and 800°C, respectively, will be discharged in February 1972 at which time the fluence achieved will be about 1.0×10^{22} n/cm² ($E > 0.18$ MeV). Although this is less than one-half of the goal exposure, it is nevertheless the highest exposure creep data ever generated. Analysis of this data should, therefore, expand our understanding of creep in graphite significantly, although no data will be available beyond "turnaround."

RADIOLYTIC REACTIONS IN GRAPHITE MODERATED REACTORS

R. P. Turcotte and G. L. Tingey

During the last quarter, supplementary data on the radiolytic reaction of carbon monoxide and water vapor were obtained at 465 vpm and 940 vpm reactant concentrations in one atmosphere helium. Agreement with earlier results was satisfactory and a temperature dependence for the reaction was confirmed which suggests an activation energy which is progressively negative with increasing dilution.

Treatment of the kinetic data has been considered as affected by collision theory and by the theory of absolute reaction rates. It was concluded that in the absence of a definite assignment of molecularity for the main reaction, absolute reaction rate theory cannot be applied. Collision theory, which suggests inclusion of a $T^{1/2}$ term in the rate expression, remains the best route to interpretation of the data. The final analyses and writeup of the $\text{CO} + \text{H}_2\text{O}$ radiolysis experiments are underway.

EFFECT OF SAMPLE SIZE ON DIMENSIONAL STABILITY OF GRAPHITE

W. J. Gray

The study of the effect of sample size on irradiation induced dimensional changes in graphite has been expanded to include an investigation of a phenomenon called "cold-seeding" wherein graphite samples are briefly irradiated at low temperatures prior to the normal high-temperature irradiation. A large number of samples which have been previously "cold-seeded" are currently being irradiated to high fluences at high temperatures in the ETR.

The "cold-seeded" samples were charged into the ETR in November for the third period of high-temperature irradiation. Some difficulty was experienced with temperature control in one of the three cells but appropriate corrections have enabled the capsule to operate normally. Discharge is scheduled in April 1972 following three cycles of irradiation which will bring total fluences for some samples up to about $5 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$). At this time, the data will be analyzed and a final report prepared to be issued in June.

HIGH-TEMPERATURE IRRADIATIONS

W. J. Gray and W. C. Morgan

Capsule GEH-13-422, which was inserted in the ETR at the beginning of Cycle 113, operated satisfactorily and was discharged during the shutdown at the middle of Cycle 114 (January 1972). The capsule discharge

was moved ahead to enable the normal dimensional and physical property measurements as well as an additional series of destructive measurements to be completed prior to the end of FY-1972. Length-change measurements have been completed for all 196 samples; measurement of thermal expansion and sonic modulus are proceeding.

MECHANISM OF IRRADIATION DAMAGE

W. C. Morgan and W. J. Gray

Some reordering of priorities has been done as a result of the planned close-out of the Nuclear Graphite program. Work has been curtailed on long-term subtasks where significant advances were not expected prior to the end of FY-1972; on the other hand, availability of the high-temperature irradiated samples will allow acceleration of the destructive testing and analysis subtask.

Samples have been selected from GEH-13-422 for measurement of high temperature thermal expansion and annealing up to 2800°C following the normal series of measurements made on irradiated samples.

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5. STEADY STATE AND TRANSIENT SUBCHANNEL CODE DEVELOPMENT AND DATA ANALYSIS

D. S. Rowe and B. M. Johnson

The objective of this study is to develop improved methods for analyzing heat transfer and fluid flow in rod bundle nuclear elements during both steady state and transient conditions. The study includes the continued development of the COBRA computer program and experiments to help verify and implement this computer program.

COBRA-III COMPUTER PROGRAM

The objective of this portion of the study is to develop improved mathematical models and computer program for predicting the steady-state and transient thermal-hydraulic performance of rod bundle nuclear fuel elements.

The major effort for this reporting period was directed toward the development of a fuel heat transfer model for the COBRA-III computer program. This important improvement was needed to allow thermal capacity effects to be included during transient analyses. In the previously published version of COBRA-III (BNWL-B-82) the fuel heat flux was assumed to be known from input data. This was rather unsatisfactory since it required a prior knowledge of the fuel heat transfer. A more satisfactory approach is to let the heat flux occur naturally by changing the heat generation, fuel surface temperature, heat transfer coefficient or fluid temperature.

The fuel heat transfer model now included in COBRA-IIIB considers only radial conduction within the fuel. Axial and circumferential heat conduction are presently ignored. Axial conduction can be ignored because axial temperature gradients are usually small. Circumferential conduction is ignored to maintain a reasonable limit on core storage for fuel temperatures. Storing temperatures at one time for each radial node in each rod and at every axial location uses a large amount of core storage. Further

subdivision circumferentially increases the core storage by at least 4 or 6 depending upon the amount of detail required.

Figure 5.1 shows the node layout used for the conduction model. The fuel is divided into equal radial nodes plus a node for the cladding. For N nodes this gives $N + 1$ temperatures where the temperature at $i = N + 1$ is at the outer surface of the cladding. A gap conductance coefficient is assumed to exist at the fuel-clad interface. The surface heat transfer coefficient can be arbitrarily specified through heat transfer correlations. An average heat transfer coefficient is determined from a circumferentially weighted average of the heat transfer coefficients in the subchannels surrounding a fuel rod. A similar calculation is performed to obtain an average fluid temperature.

FIGURE 5.1. Fuel Model Node Designation.

The numerical solution uses a stable implicit finite difference scheme. An implicit scheme was chosen since it is stable for all time steps as is the flow solution in COBRA-IIIB. The equations used for the numerical solution are as follows:

$i = 1$, temperature at fuel center

$$\left(\frac{\rho c}{\Delta t} + \frac{4k}{\Delta r^2} \right) u_1' - \frac{4k}{\Delta r^2} u_2' = q_1''' + \frac{\rho c u_1}{\Delta t}$$

$1 < i \leq N-1$, temperature in fuel region

$$\begin{aligned} \left(-\frac{k}{\Delta r^2} + \frac{k}{2(i-1)\Delta r^2} \right) u_{i-1}' + \left(\frac{\rho c}{\Delta t} + \frac{2k}{\Delta r^2} \right) u_i' + \left(-\frac{k}{\Delta r^2} - \frac{k}{2(i-1)\Delta r^2} \right) u_{i+1}' \\ = q_i''' + \frac{\rho c u_i}{\Delta t} \end{aligned}$$

$i = N$, fuel temperature at fuel-clad interface

$$\begin{aligned} \left(-\frac{2k}{\Delta r^2} \right) u_{N-1}' + \left(\frac{\rho c}{\Delta t} + \frac{2k}{\Delta r^2} + \frac{2h_G}{\Delta r} + \frac{h_G}{(i-1)\Delta r} \right) u_N' + \left(-\frac{2h_G}{\Delta r} - \frac{h_G}{(i-1)\Delta r} \right) u_{N+1}' \\ = q_N''' + \frac{\rho c u_N}{\Delta t} \end{aligned}$$

$i = N+1$, cladding temperature

$$-\left(\frac{h_G}{t_c} \frac{r_N}{r_{N+1}} \right) u_N' + \left(\frac{\rho c}{\Delta t} + \frac{h_G}{t_c} \frac{r_N}{r_{N+1}} \right) u_{N+1}' + \frac{h_s}{t_c} u_{N+L}' = q_{N+1}''' + \frac{\rho c u_{N+1}}{\Delta t} + \frac{h_s}{t_c} u_c$$

These equations can be arranged as a set of simultaneous equations of the form

$$[A] \{u\} = \{b\}$$

where $[A]$ is a tridiagonal matrix. This system of equations is solved by using a compact Gaussian elimination routine. Only the three diagonals of matrix $[A]$ are stored to reduce core storage.

For steady state solutions, the temperatures are obtained through iteration with a large value of Δt . This is the same procedure used for the flow solution. For transients the temperature solution is done only once per time step using data obtained during the first iteration on the flow solution. This is a valid procedure when the fuel time constant is large compared to the transit time of the flow through the bundle. This also permits storage of fuel temperatures at one time only and, therefore, conserves core storage.

To accommodate the fuel heat transfer calculations, three new subroutines are now included in COBRA-IIIB. Subroutine HEAT calculates the heat addition to each subchannel from the fuel rod heat flux. This heat flux can be specified directly from input as done previously or it can be determined from the fuel temperatures. Subroutine TEMP calculates the radial temperatures in each fuel rod using the previously described model. It sets up the matrix $[A]$ and $\{b\}$ and solves for $\{u\}$ by calling Subroutine GAUSS. GAUSS is a very compact routine that can quickly solve a tridiagonal system of equations.

EXPERIMENTAL STUDY OF FLOW STRUCTURE IN ROD BUNDLES

Experimental measurements of local velocity, turbulence intensity and Eulerian time scale were completed in several types of rod bundle channels. These experiments were performed to obtain a more complete description of turbulent flow structure of single-phase flow in rod bundles. This information is needed to develop better mixing correlations for use in subchannel computer programs such as COBRA.

The measurements were performed with a two-component laser-Doppler velocimeter (LDV). This device can measure fluid velocities at a point in a flow stream without inserting a probe into the flow. It operates from the principle that light scattered from very small particles moving with the fluid experiences a frequency shift that is related to the fluid velocity. By using a system of optics and electronics, this Doppler shift can be accurately measured to obtain the instantaneous fluid velocity at the

measurement point. The fluctuating component of the velocity signal can also be processed with a correlation computer to obtain autocorrelation functions which provides an estimate of turbulence scale.

A common flow channel was used to accommodate the variety of subchannels shown in Figure 5.2. This was accomplished by using a flow channel that consisted of two common side plates containing glass windows for entrance and exit of the laser beam. Inserts of two different thicknesses were placed between the side plates to create rectangular flow channels. Subchannels were formed within these channels by inserting rods, or half-rods, to form the desired subchannel shapes. Rods with glass windows were used where appropriate to enable passage of the laser beam. A flow straightening and calming section was included to control the level of upstream turbulence and provide a uniform velocity profile at the channel inlet.

FIGURE 5.2. Subchannel Flow Models Used for Turbulent Flow Studies

Over 2000 velocity and turbulence intensity data points were obtained during the study. These data are presently being reduced and analyzed. Preliminary conclusions from the data are as follows.

- Velocity, intensity, and scale were found to be nearly independent of Reynolds number over the range from 50,000 to 200,000. Turbulence intensity showed a slight decrease with increasing Reynolds number. Axial component turbulence intensity values typically ranged from 0.03 to 0.05 in regions away from the channel walls.
- Subchannel shape did not significantly affect the axial and transverse components of turbulence intensity in the gap of flow channels D and E. The transverse component was found to be about 30% less than the axial component.
- The added degrees of lateral freedom in channels A and B caused only a slight increase in the axial component of intensity in the rod gaps. There was no evidence of a macroscopic transverse pulsation through the rod gaps.
- Detailed maps of turbulence intensity showed that maximum turbulence intensity occurred between the subchannel center and the rod gap in regions away from the channel walls. The minimum intensity occurred at the subchannel center.
- Detailed velocity maps and the turbulence data suggest the possibility of secondary flows that move in a symmetric pattern as shown in Figure 5.3. Measurement of secondary flows was attempted but they were below the measurement accuracy of the LDV. If secondary flows did exist they were less than about 1% of the axial velocity.
- The addition of a grid spacer to Channel A significantly modified the turbulent flow structure. It increased the intensity by about 100% which then decayed gradually downstream of the grid. The grid also decreased the scale which then gradually increased downstream of the grid. At $L/D = 20$ the influence of the grid on both intensity and scale was still apparent.

FIGURE 5.3. Secondary Flow Pattern Suggested by Maps of Axial Component Velocity and Turbulence Intensity

All data to date has been obtained at a pitch-to-diameter (p/d) ratio of 1.25. To evaluate the effect of gap spacing, measurements are to be made in Channel A using 1 1/8 inch rods giving a p/d = 1.125. These measurements are scheduled for the last quarter of FY-1972.

EXPERIMENTAL STUDY OF FLOW BLOCKAGE

An experimental study was started to investigate the flow in rod bundle subchannels containing blockages. These data are needed to help validate and to provide input to the COBRA program for flow blockage analysis.

The blockage measurements are being performed in Section B shown in Figure 5.2. Area reductions of 90% are being considered for both "orifice" and "swelling" type blockages. The extent of flow blockage is also being considered by progressively blocking subchannels starting at one side of the flow channel.

Both velocity and pressure information are being obtained in this study. The velocity is being measured with the two component LDV. Axial and transverse pressure gradients are being measured to within 0.001 psi using a precision servomanometer. Both velocity and pressure data are being taken at several locations upstream and downstream of the blockages.

NOMENCLATURE

C	-	Specific heat
Δr	-	Fuel node size
Δt	-	Time step
h_g	-	Fuel-clad gap conductance
h_s	-	Surface heat transfer coefficient including clad resistance
k	-	Thermal conductivity
N	-	Number of nodes
q'''	-	Power density
r	-	Radius
ρ	-	Density

t_c - Clad thickness
 u - Temperature
 u_c - Coolant temperature

6. CRITICAL EXPERIMENTS WITH HOMOGENEOUS

PuO₂-POLYSTYRENE AT 50 H/Pu

S. R. Bierman and E. D. Clayton

INTRODUCTION

Part of the research effort at the Pacific Northwest Laboratory operated Critical Mass Laboratory is concerned with determining the critical parameters of plutonium mixtures over the entire range of neutron moderation. To the extent possible, plutonium nitrate solutions have been used as the experimental fuel, since it offers the advantages of theoretical densities, easily varied fuel compositions, and easily varied geometries. However, the practical concentration limit for which plutonium nitrate solutions are stable is less than 500 g Pu/l. Consequently, to obtain experimental data in that portion of the neutron energy spectrum characteristic of concentrations above 500 g Pu/l, homogeneous mixtures of PuO₂-polystyrene have had to be used.

Experimental results have been reported for plutonium nitrate solutions^(1,2) having concentrations between about 25 and 435 g Pu/l and for PuO₂-polystyrene fueled assemblies^(3,4,5) ranging in H/Pu atomic ratios corresponding to essentially unmoderated PuO₂ down to 1620 g Pu/l. To obtain a closer tie-in of data between these solution- and solid-type experiments, a series of critical experiments was performed with a homogeneous mixture of PuO₂-polystyrene at an H/Pu atomic ratio corresponding to a theoretical density, Pu-water concentration of 521 g Pu/l (49.6 H/Pu). The results of these latest series of experiments are presented in this paper.

Data in the concentration range between 500 and 1000 g Pu/l are also needed to resolve the differences⁽⁶⁾ that have been observed between diffusion theory and transport theory computations using the computer codes HFN⁽⁷⁾ and DTF-IV.⁽⁸⁾ The diffusion theory code, HFN, has been shown to be quite accurate for concentrations below 300 g Pu/l, and indications

are it is accurate up to about 500 g Pu/l; however, it calculates too large a critical size above, at least, 1600 g Pu/l. Conversely, the transport theory code, DTF-IV, has been shown to be quite accurate for concentrations above 1600 g Pu/l but calculates a smaller critical size than HFN for concentrations between about 300 and 1600 g Pu/l.

Calculations have also indicated that, for plutonium systems having a ^{240}Pu isotopic content greater than 5 wt%, there are three different plutonium concentrations below about 1000 g Pu/l for which k_{eff} is unity⁽⁶⁾ in the same sized vessel. That is, for Pu-water systems having a ^{240}Pu isotopic content greater than 5 wt%, a subcritical Pu concentration exists which can be made critical by dilution alone. Critical experiments with plutonium nitrate solutions⁽⁹⁾ having a ^{240}Pu isotopic content of 23.2 wt% have tended to substantiate this phenomenon in that a smaller critical slab thickness was required for 202 g Pu/l solution than for 284 g Pu/l solution. However, in this concentration range the critical size is not very sensitive to concentration changes. To provide additional data on this aspect of the effects of ^{240}Pu on criticality, as well as to provide data on the neutron poisoning effects of ^{240}Pu , the experimental fuel was fabricated using PuO_2 having a relatively high ^{240}Pu content.

EXPERIMENTS AND DATA REDUCTION

The experiments were carried out using our remote split-table machine⁽³⁾ and PuO_2 -polystyrene in the form of 2- x 2- x 2-in. and 2- x 2- x 1/2-in. blocks, each compressed to a density of 367 ± 4 g Pu/l and encased, for contamination control, in 6 mil thick tape (MM&M #471). The fuel blocks were fabricated with PuO_2 having a ^{240}Pu isotopic content of 18.35 ± 0.22 wt%. A detailed description of the fuel is presented in Table 6.1. The reflected assemblies were fully reflected with 15 cm of Plexiglas and its composition is also given in Table 6.1.

TABLE 6.1. Description of Fuel

Dimension of 2 in. Cubes without Cladding, LxWxH	5.09 x 5.09 x 5.09 ± 0.01 cm
Average Dimension of Stacked 2-in. Cubes with Cladding, LxWxH	5.19 x 5.19 x 5.16 ± 0.04 cm
Average Thickness of Cladding, cm	0.018
Composition of Cladding, Atoms per Barn - cm	
H	4.489 x 10 ⁻²
C	3.113 x 10 ⁻²
Cl	0.724 x 10 ⁻²
Composition of Plexiglas, Atoms per Barn - cm	
H	5.712 x 10 ⁻²
C	3.570 x 10 ⁻²
O	1.428 x 10 ⁻²
Composition of Compacts, Atoms per Barn - Cm(a)	
²³⁸ Pu	2.125 x 10 ⁻⁶
²³⁹ Pu	6.952 x 10 ⁻⁴
²⁴⁰ Pu	1.689 x 10 ⁻⁴
²⁴¹ Pu	3.952 x 10 ⁻⁵
²⁴² Pu	1.045 x 10 ⁻⁵
²⁴¹ Am	7.345 x 10 ⁻⁶
O	2.082 x 10 ⁻³
H	4.575 x 10 ⁻²
C	4.505 x 10 ⁻²
PuO ₂ Particle Size, mm	
Maximum	0.0070
Mean	0.0012
Minimum	0.0003
Plutonium Density, g/cm ³	0.367 ± 0.004

a. ²⁴¹Am content determined May 27, 1970. Reflected experimental data obtained by June 6, 1970. Bare experimental data obtained September 9, 1970.

Experimental data were obtained at room temperature from both bare and Plexiglas-reflected parallelepipeds. The critical sizes of each of these assemblies are presented in Table 6.2. Replacement type experiments were performed to experimentally determine the corrections required to account for the effects of stacking voids and the cladding material used on each fuel block for contamination control. Thus, each critical assembly shown in Table 6.2 represents a solid mass of 49.6 ± 0.7 H/Pu fuel at a density of 367 ± 4 g Pu/l.

TABLE 6.2. Experimental Data from PuO₂-Polystyrene Compacts
Plutonium at 18.35 wt% ²⁴⁰Pu, 49.6 ± 0.7 H/Pu
Atomic, 367 g Pu/l

REFLECTOR	CRITICAL DIMENSIONS, cm			CRITICAL MASS
	LENGTH	WIDTH	HEIGHT ^(a)	Kg OF Pu
PLEXIGLAS	61.08	61.08	16.35 ± 0.06	22.4 ± 0.3
PLEXIGLAS	50.90	61.08	17.48 ± 0.05	19.9 ± 0.2
PLEXIGLAS	50.90	50.90	18.68 ± 0.08	17.7 ± 0.2
PLEXIGLAS	50.90	45.81	19.69 ± 0.07	16.8 ± 0.2
PLEXIGLAS	40.72	45.81	22.06 ± 0.06	15.1 ± 0.2
PLEXIGLAS	40.72	40.72	23.58 ± 0.10	14.3 ± 0.2
PLEXIGLAS	40.72	30.54	29.64 ± 0.12	13.5 ± 0.2
BARE	46.08 ± 0.50	40.96 ± 0.44	49.12 ± 0.50	34.0 ± 0.3

^(a) CRITICAL HEIGHT CORRECTED FOR VOIDS AND CLADDING.
BARE ASSEMBLIES CORRECTED ALSO FOR EFFECTS OF
STRUCTURAL SUPPORTS.

Although there was an insufficient amount of fuel to achieve criticality unreflected, a bare critical assembly was built using a driver region of PuO_2 -polystyrene having an H/Pu atomic ratio of 5. (Experiments with this 5 H/Pu fuel have been previously reported.)⁽⁴⁾ The worth of the 5 H/Pu driver region was calculated in terms of an equivalent height of the 49.6 H/Pu fuel region. Based on this corrected measurement, a bare assembly $46.08 \times 40.96 \times 49.12 \pm 0.050$ cm containing 34.0 ± 0.3 kg of plutonium, would be critical. By using a calculated extrapolation distance of 2.72 ± 0.2 cm for the bare assembly and the bare critical dimensions given above, the critical buckling for this 49.6 H/Pu, 367 g Pu/l, PuO_2 -polystyrene material was determined to be $116.2 \pm 1.0 \text{ m}^{-2}$.

The reflected critical assemblies ranged over a series of slabs from about 30 cm to 16 cm in thickness to permit determining the critical thickness of a slab of this fuel, infinite in two dimensions. The critical dimensions of each of these reflected assemblies are shown in Table 6.2. The critical height of each of these assemblies is plotted in Figure 6.1, as an inverse function of its core cross sectional area. A least squares fit extrapolation of this data to an inverse cross sectional area of zero yields a critical thickness of 10.87 ± 0.14 cm for a slab of this PuO_2 -polystyrene material infinite in two dimensions. A corresponding extrapolation distance of 9.04 ± 0.10 cm was obtained for the reflected infinite slab by equating the critical dimensions of each reflected assembly to the critical buckling of $116.2 \pm 1.0 \text{ m}^{-2}$ determined for this material for the bare assembly. The extrapolation distances thus obtained for each reflected assembly are also shown in Figure 6.1, as an inverse function of the respective assembly's core cross sectional area.

DATA ANALYSIS

For establishing criticality limits and for checking nuclear constants and calculational techniques, these experimental slab data have been expressed in equivalent spherical, cylindrical, and rectangular geometries.

FIGURE 6.1. Extrapolation Distances and Measured Critical Heights

The experimental data were converted at the critical buckling of 116.2 m^{-2} to obtain critical sizes for these geometries, both bare and fully reflected. These derived critical sizes are shown in Table 6.3. The standard deviations on the calculated extrapolation distances are estimated to be $\pm 0.2 \text{ cm}$. The remaining standard deviations shown were obtained by propagation of errors. Also shown in Table 6.3 are critical sizes for Pu-water and PuO_2 -water at theoretical densities corresponding to a 49.6 H/Pu atomic ratio. These were obtained by making transport theory corrections to the 367 g Pu/l PuO_2 -polystyrene data for density and the presence of carbon.

Values are also shown in Table 6.3 which have been corrected to zero ^{240}Pu content to show the large effect that ^{240}Pu has on the critical size at this near optimum concentration for neutron poisoning with ^{240}Pu . In this relatively thermalized neutron energy system, the percent change in the spherical critical mass of total plutonium per percent change in ^{240}Pu content is 12.1 for the reflected case and 10.2 for the bare case, as compared to values of about 2 for an unmoderated Pu-water system containing up to 20 wt% ^{240}Pu . A GAMTEC-II⁽¹⁰⁾ calculated neutron energy spectrum, using the original GAMTEC-II cross sections library, is shown in Table 6.4 for the PuO_2 -polystyrene fuel used in the experiments.

For consistency with previously reported experimental results and data presentation on plutonium systems, the cross section data for the original GAMTEC-II code developed⁽¹⁰⁾ at the Hanford Critical Mass Laboratory were used in analyzing and presenting these current data. Currently, a thorough testing and evaluation of the ENDF/B cross section data is in progress⁽¹²⁾ with these and other previously reported critical experiments. Preliminary k_{eff} values, however, based on ENDF/B-II cross section data for the bare and the Plexiglas-reflected critical infinite slabs of PuO_2 -polystyrene at 49.6 H/Pu are shown in Table 6.5 as an initial comparison between the original GAMTEC-II cross sections (CML) and the ENDF/B-II cross sections. In each case, 18-group GAMTEC-II averaged cross sections were used with the diffusion theory code, HFN,⁽⁷⁾ and the transport theory code, DTF-IV.⁽⁸⁾ The ENDF/B-II cross sections were obtained from the ENDF/B-II point energy

TABLE 6.3. Critical Dimensions in Spherical, Cylindrical, Cubic and Slab Geometries at 49.6 H/Pu, Atomic Ratio

GEOMETRY	PuO ₂ -POLYSTYRENE 0.367 g Pu/cc 18.35 WT% ²⁴⁰ Pu(a)		²³⁹ Pu - WATER 0.521 g Pu/cc 0.0 WT% ²⁴⁰ Pu	²³⁹ PuO ₂ - WATER 0.508 g Pu/cc 0.0 WT% ²⁴⁰ Pu	Pu - WATER 0.521 g Pu/cc 18.35 WT% ²⁴⁰ Pu	PuO ₂ - WATER 0.508 g Pu/cc 18.35 WT% ²⁴⁰ Pu
	λ	χ(b)	χ(b)	χ(b)	χ(b)	χ(b)
	cm	cm	cm	cm	cm	cm
BARE ASSEMBLIES						
INFINITE SLAB	2.65 ± 0.2	23.85 ± 0.29	13.40 ± 0.16	13.60 ± 0.16	19.55 ± 0.24	19.85 ± 0.24
SPHERE	2.65 ± 0.2	26.50 ± 0.21	15.49 ± 0.12	15.69 ± 0.12	22.02 ± 0.17	22.30 ± 0.17
INFINITE CYLINDER	2.64 ± 0.2	19.67 ± 0.21	11.32 ± 0.12	11.48 ± 0.12	16.26 ± 0.17	16.50 ± 0.18
CUBE	2.72 ± 0.2	45.04 ± 0.30	-----	-----	-----	-----
REFLECTED ASSEMBLIES(c)						
INFINITE SLAB	9.04 ± 0.10	10.87 ± 0.14	5.22 ± 0.07	5.34 ± 0.07	9.43 ± 0.12	9.64 ± 0.12
SPHERE	9.12 ± 0.10	20.03 ± 0.12	11.65 ± 0.07	11.81 ± 0.07	17.19 ± 0.10	17.45 ± 0.10
INFINITE CYLINDER	9.08 ± 0.10	13.23 ± 0.12	7.39 ± 0.07	7.51 ± 0.07	11.39 ± 0.10	11.58 ± 0.10
CUBE	9.20 ± 0.10	32.08 ± 0.17	-----	-----	-----	-----

(a) ISOTOPIC CONCENTRATION

(b) CRITICAL THICKNESS OF SLAB OR CUBE, CRITICAL RADIUS OF SPHERE OR CYLINDER

(c) PuO₂-POLYSTYRENE REFLECTED WITH PLEXIGLAS, OTHER SYSTEMS REFLECTED WITH WATER

TABLE 6.4. Calculated 18-Group Neutron Flux for PuO₂-Polystyrene

367 g Pu/l, 49.6 H/Pu Atomic Ratio

18.35 wt% ²⁴⁰Pu Isotopic Content

<u>Group</u>	<u>Lower Energy</u>	<u>Relative Flux</u>
1	7.79 MeV	0.0028
2	6.07	0.0083
3	4.72	0.0184
4	3.68	0.0293
5	2.87	0.0401
6	1.74	0.1094
7	1.35	0.0545
8	183.00 keV	0.2738
9	24.80	0.1152
10	3.36	0.0799
11	454.00 eV	0.0731
12	101.00	0.0512
13	37.30	0.0292
14	13.70	0.0280
15	5.04	0.0245
16	1.86	0.0242
17	0.683	0.0152
18	0	0.0229

TABLE 6.5. Comparison Between HFN and DTF-IV Calculations
at 49.6 H/Pu Atomic Ratio

Critical Assembly				k_{eff}			
				DTF-IV		HFN	
Core	Reflector	Geometry	Slab Thickness	CML (a)	ENDF/B (b)	CML (a)	ENDF/B (b)
PuO ₂ -Polystyrene at 367 g Pu/l; 18.35 wt% ²⁴⁰ Pu in Pu	None	Inf. Slab	23.85 ± 0.29	1.028	1.049	1.014	1.041
PuO ₂ -Polystyrene at 367 g Pu/l; 18.35 wt% ²⁴⁰ Pu in Pu	Full Plexiglas	Inf. Slab	10.87 ± 0.14	0.998	0.009	0.998	1.004

- a. 18-group GAMTEC-II averaged cross sections using the original GAMTEC-II library.⁽¹⁰⁾
b. FLANGE-ETOG-ENDF/B-II 18 group GAMTEC-II averaged cross sections.

library data by using the FLANGE code to process the thermal data and the ETOG code to process the epithermal data for broad group averaging in GAMTEC-II. As can be seen in the table, calculated k_{eff} values using these cross section data were consistently higher than comparable values obtained from the calculations with the CML cross sections. Both sets of calculations indicate, also, that the bare infinite slab thickness of 23.85 cm may be slightly conservative with respect to criticality safety.

As with previously published data^(3,4,5) from undermoderated PuO₂-polystyrene experiments, these current data are shown graphically in Figures 6.2, 6.3, 6.4, and 6.5 along with the previous data. In all three geometries the critical sizes for ²³⁹Pu-water derived from the current experiments are slightly larger than those shown in TID-7028⁽¹³⁾ and are in general agreement with the previously reported data derived from undermoderated PuO₂-polystyrene experiments.

FIGURE 6.2. Data Derived from Critical Slabs of PuO_2 - Polystyrene

FIGURE 6.3. Data Derived from Critical Slabs of PuO₂-Polystyrene

FIGURE 6.4. Data Derived from Critical Slabs of PuO_2 -Polystyrene

FIGURE 6.5. Water Reflected Spherical Critical Radii of Pu (metal) - Water Mixtures

In Figure 6.5, previously reported spherical critical radii obtained from plutonium nitrate solutions are also included. Also shown in Figure 6.5 are calculated spherical critical radii obtained by using GAMTEC-II⁽¹⁰⁾ 18-group CML cross sections with the transport theory code, DTF-IV⁽⁸⁾ and the diffusion theory code, HFN.⁽⁷⁾ Previous studies⁽¹¹⁾ have indicated that little improvement can be gained in the transport theory calculated values for these type systems by using higher than first order anisotropic scattering or fourth order of quadrature. Consequently, the S_4 approximation with P_1 scattering was used in the DTF-IV calculations. As can be seen in Figure 6.5, the spherical critical radius for 521 g Pu/l ^{239}Pu -water, derived from the 49.6 H/Pu PuO_2 -polystyrene experiments, is in good agreement with that previously obtained⁽²⁾ from a plutonium nitrate solution experiment at 435 g Pu/l. (The water-reflected spherical critical radius for ^{239}Pu -water at 435 g Pu/l is 12.00 cm and at 521 g Pu/l it is 11.65 cm.) Also, it can be seen in Figure 6.5 that, at 521 g Pu/l the diffusion theory code, HFN, yields a more correct calculated value of the critical size than does the transport theory code, DTF-IV.

Although the neutron poisoning effects of 18.35 wt% ^{240}Pu are given in Table 6.3, its effect on ^{239}Pu -water at 521 g Pu/l, and the indication that three concentrations exist for which a given size vessel is just critical ($k_{\text{eff}} = 1$), is better illustrated in Figure 6.5. As can be seen in Figure 6.5., the critical radius for this system is slightly less than that calculated by the diffusion theory code HFN for this same system containing 20 wt% ^{240}Pu . However, the critical radius, at 521 g Pu/l, is considerably larger than some of the HFN-calculated critical radii at lower concentrations. This substantiates the observations made by Lloyd⁽⁹⁾ with plutonium nitrate solutions at concentrations between 200 and 300 g Pu/l. It also confirms that the critical size of a Pu-water system can experience a minimum at a concentration other than that of the pure metal. Consequently, in setting criticality limits the credibility of diluting, as well as concentrating, in a given vessel must be established whenever the neutron poisoning effect of ^{240}Pu is considered, and if, as is generally the case, the Pu concentration is below about 1000 g/l.

To further evaluate the phenomenon in which a subcritical vessel of fixed volume could become critical by dilution with water, further data at 1000 g Pu/l would be useful with plutonium containing about 20 wt% ^{240}Pu . Experiments reported by Lloyd⁽⁹⁾ provide data for the slightly overmoderated case and the experiments reported on in this paper provide data at the concentration for which ^{240}Pu effects are essentially a maximum.

CONCLUSIONS

The results from the critical experiments reported on in this paper provide a correlating point between the plutonium critical experiments performed with plutonium nitrate solution and those performed with PuO_2 -polystyrene compacts. A good correlation was found to exist between the two types of experiments. It was also observed that, due, apparently, to compensating errors, the diffusion theory code HFN calculates the critical condition better than the transport theory code DTF-IV at concentrations of 521 g Pu/l and less.

In contrast to data previously obtained with unmoderated PuO_2 , the density effect of having the plutonium in the oxide form had essentially no effect on the reactivity at 49.6 H/Pu, whereas the neutron poisoning effect of the 18.35 wt% ^{240}Pu was at least five times that observed for the unmoderated PuO_2 . It appears, also, from the experiments that, for high exposure Pu-water systems, a given size vessel could be just critical ($k_{\text{eff}} = 1.0$) at three different concentrations below about 1000 g Pu/l. That is, a critically safe vessel could be made critical by dilution alone if the ^{240}Pu content were high enough. The experiments also imply that the ENDF/B-II cross section data result in conservative values with respect to nuclear criticality safety; however, considerably more testing against experimental data is needed to establish conclusions of this kind.

ACKNOWLEDGEMENTS

The authors wish to express their appreciation to Mrs. Donna Andersen for her assistance in the compilation of this manuscript, and to L. E. Hansen for his efforts in obtaining the ENDF/B-II cross section data.

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7. THE CRITICALITY OF HIGH BURNUP PLUTONIUM

R. C. Lloyd and E. D. Clayton

A series of experiments have been completed with plutonium obtained from extreme high burnup fuel to establish the combined effects of the various isotopes of plutonium on criticality. The isotopic composition of the plutonium, as measured on August 11, 1971, was: ^{238}Pu -0.2 wt%, ^{239}Pu -41.4 wt%, ^{240}Pu -42.9 wt%, ^{241}Pu -10.8 wt%, and ^{242}Pu -4.7 wt%. A non-negligible amount of Americium-241 also was present to the extent of 1.08 wt% of the total Pu. The experiments were performed in a 24-inch diameter, water-reflected, cylindrical vessel. Ten data points were obtained with the critical heights and masses determined for various plutonium concentrations and nitric acid molarities in the plutonium nitrate solutions. Plutonium concentrations covered the range of about 40 to 140 g Pu/l with nitric acid molarities ranging from about 1.5 to 5.

The data from the criticality experiments (conducted during August 9-19, 1971) for the plutonium nitrate solutions are included, for completeness, with the correlated calculational results. The experimental results were analyzed by means of the KENO Monte Carlo Code, utilizing ENDF/B-II cross section data. In this case, the epithermal groups were obtained through application of the ETOG and GAM processing codes, and the thermal group data via the FLANGE code followed by the TEMPEST portion of the GAMTEC code. The k_{eff} values are given in Table 7.1. The checks against the integral critical experiments display a somewhat positive bias with the average computed k_{eff} being about 1% above unity.

The effects of the individual isotopes have been separately examined to determine their contribution to the reactivity. The computed effect for the 1.08 wt% ^{241}Am is to lower the value of k_{eff} from 0.8 to 1.2%.

<u>Pu Conc.</u>	<u>δk of ^{241}Am</u>
40.6 g/l	- 0.008
85.5 g/l	- 0.010
140.0 g/l	- 0.012

TABLE 7.1. Criticality of High Burnup Plutonium Solutions

<u>Pu Conc. (g/l)</u>	<u>Nitric Acid Molarity</u>	<u>Critical Height in 24-in. Dia. Cylinder</u>	<u>Critical Volume</u>	<u>Computed k_{eff} With ENDF/B-II Data and KENO Code</u>
140.0	5.02	54.70 cm.	158.3 l	0.993 ± 0.007
116.0	4.14	50.55	147.9	1.020 ± 0.006
99.3	3.67	48.26	141.2	1.012 ± 0.007
85.5	3.12	47.00	137.5	1.020 ± 0.005
75.6	2.74	47.29	138.3	1.006 ± 0.007
65.1	2.41	49.12	143.7	1.014 ± 0.007
56.3	2.05	52.83	154.5	1.022 ± 0.007
46.8	1.70	63.47	185.7	1.014 ± 0.005
40.6	1.46	80.92	236.7	1.007 ± 0.005

The effects of the other isotopes have been examined for the solution at 85.5 g Pu/l. The results are:

	<u>δk</u>
^{238}Pu (0.2 wt%)	- 0.001
^{242}Pu (4.7 wt%)	- 0.009
^{240}Pu (42.9 wt%)	- 0.371
^{241}Pu (10.8 wt%)	+ 0.103
^{241}Pu replaced by ^{239}Pu	- 0.036

In the case of ^{241}Pu , the change of k_{eff} is that which would result from adding 10.8 wt% to the solution originally containing no ^{241}Pu , so as to result in the final concentration of 85.5 g Pu/l. It is more appropriate to ask how much more reactive the ^{241}Pu would be relative to a like addition of ^{239}Pu . This is shown in the last entry. If the 10.8 wt% ^{241}Pu were replaced with a like amount of ^{239}Pu , k_{eff} would be less by about 3.6%, i.e. treating the ^{241}Pu as equivalent to ^{239}Pu would cause a significant error of nearly 4% in k_{eff} , which would be nonconservative (k_{eff} calculated about 4% too small). Thus, these results show the importance of considering the effects of all isotopes that may be present in criticality evaluations pertaining to high burnup fuel.

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8. THEORY-EXPERIMENT TESTS USING ENDF/B VERSION II CROSS SECTION DATA

L. E. Hansen and E. D. Clayton

The availability of ENDF/B cross section data, while providing standard cross section data needed for criticality calculations, also creates the necessity of recorrelating calculational results with experimental data. This is required to assess the uncertainties of utilizing these data in criticality safety evaluations.

The ENDF/B data have been checked against integral experiments for a number of fast critical assemblies,⁽¹⁾ but little has been reported concerning the use of these data when applied to the more general cases encountered in nuclear criticality safety (i.e., the conditions encountered external to reactors require consideration of thermal, intermediate, and fast systems). Unfortunately, the checks made on fast critical assemblies provide little, if any, assurance of reliability in regard to the first two systems mentioned.

There are, of course, a multiplicity of methods that can be used to derive broad group averaged cross section data, given the same fundamental cross section data. The schematic diagram in Figure 8.1 shows some of the possible alternative calculational procedures that can be employed to obtain ENDF/B data averaged into broad energy group constants. As can be seen, even the limited number of possible calculational methods shown in Figure 8.1 yields 15 different possible combinations of thermal and epithermal broad group data that can be used in eigenvalue calculations. Consequently, it is necessary to severely limit the number of permutations and combinations of the derived data to be examined so that a greater variety of fuels may be examined.

For our purposes, the primary interest was to determine the accuracy with which reactivities could be computed throughout the entire range of moderation ratios using multipgroup constants generated by the GAMTEC code. GAMTEC is basically a combination of the GAM-I and TEMPEST codes.

FIGURE 8.1. Typical Schematic Diagram for Processing ENDF/B Cross Sections for Use in Multigroup Transport or Diffusion Theory Codes

Consequently, the epithermal ENDF/B-II data were averaged over the 68 epithermal group structure of GAM-I using the ETOG code and the thermal ENDF/B-II data were obtained for the TEMPEST energy mesh using the FLANGE-II code.

As a check on the adequacy of the GAMTEC code, the MC² and THERMOS codes were used to derive multigroup constants for selected appropriate cases. For the MC² and THERMOS codes, fine group cross sections were obtained from the ENDF/B-II library using the ETOE and FLANGE-II codes, respectively.

Eigenvalues computed using the HFN code with multigroup data (18 energy groups) for 21 different experiments selected for their geometric simplicity, are given in Table 8.1. Also shown are calculations that utilized thermal group data from the TEMPEST portion of the GAMTEC code and epithermal data, 17 groups, averaged by the MC² code. Calculations for the two mixed Pu-U nitrate solution spheres were also performed using thermal group data averaged by the THERMOS code. For these homogeneous systems, however, the resulting k_{eff} 's were within 3 mk of the values obtained with thermal data averaged by the TEMPEST code.

It is evident that the effective multiplication factors computed do not reproduce the experimental data with an acceptable degree of accuracy over the entire moderation range.

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TABLE 8.1. Computed Reactivities for Pu and Mixed Pu-U Systems Using ENDF/B-II Cross Sections

GEOMETRICAL CONFIGURATION	REFLECTOR	H/Pu ATOMIC RATIO	PERCENT ²⁴⁰ Pu	CRITICAL CONC. (g Pu/l)	HNO ₃ MOLARITY	TOTAL NO ₃ CONC. (g NO ₃ /l)	WATER CONC. (g H ₂ O/l)	DERIVED* CRITICAL DIMENSION (cm)	HFV COMPUTED MULTIPLICATION FACTORS WITH ENDF/B-II DATA AVERAGED BY	
									(ETOG-GAMTEC FLANGE-TEMPEST)	(ETOE, MC ² FLANGE-TEMPEST)
PuO ₂ -POLYSTYRENE SYSTEMS										
INFINITE SLAB	UNREFLECTED	5.0	11.46	2302	----	---	---	16.83 ⁽²⁾	1.045	1.029
INFINITE SLAB	UNREFLECTED	15.0	2.2	1120	----	---	---	16.09 ⁽³⁾	1.029	1.028
INFINITE SLAB	UNREFLECTED	49.6	18.35	367	---	---	---	23.85 ⁽⁴⁾	1.045	1.078
Pu NITRATE SOLUTION SYSTEMS (THIN STAINLESS-STEEL VESSELS ENCLOSE SPHERICAL SYSTEMS)										
SPHERICAL	WATER	354	4.6	73.0	0.2	86	971	14.568 ⁽⁵⁾	1.008	0.988
SPHERICAL	WATER	344	4.6	74.5	0.4	105	963	14.568 ⁽⁵⁾	1.007	
SPHERICAL	WATER	243	4.6	100.0	1.9	230	898	14.568 ⁽⁵⁾	1.002	
SPHERICAL	WATER	192	4.6	126.0	2.2	262	892	14.568 ⁽⁵⁾	1.013	
SPHERICAL	WATER	87	4.6	269.0	1.1	346	868	14.568 ⁽⁵⁾	1.015	
SPHERICAL	WATER	81	4.6	295.0	0.8	303	893	14.568 ⁽⁵⁾	1.027	
SPHERICAL	UNREFLECTED	668	4.6	39.0	0.4	64	978	19.318 ⁽⁵⁾	1.023	
SPHERICAL	UNREFLECTED	125	4.6	172.0	4.9	486	766	19.318 ⁽⁵⁾	1.030	
INFINITE SLAB	WATER	425	4.6	58.0	2.34	203	909	9.13 ⁽⁶⁾	0.999	
INFINITE SLAB	WATER	383	4.6	58.0	5.0	397	791	9.78 ⁽⁶⁾	0.988	
INFINITE SLAB	WATER	367	18.4	66.5	2.40	219	897	11.38 ⁽⁷⁾	1.004	
INFINITE SLAB	WATER	144	18.4	160.0	3.04	360	839	11.21 ⁽⁷⁾	1.024	
INFINITE SLAB	WATER	89	18.4	241.0	4.16	495	770	11.64 ⁽⁷⁾	1.016	
INFINITE SLAB	WATER	46	18.4	412.0	4.14	664	684	13.01 ⁽⁷⁾	1.043	
Pu-U (DEPLETED) OXIDE POLYSTYRENE SYSTEMS										
INFINITE SLAB	UNREFLECTED	158 (H/X = 47.4)	8.0	111.9 (30 WT% Pu)	---	---	---	21.70 ⁽⁸⁾	1.006	1.022
INFINITE SLAB	UNREFLECTED	209 (H/X = 30.6)	8.0	84.8 (14.62 WT% Pu)	---	---	---	23.22 ⁽⁸⁾	1.025	1.045
Pu-U (DEPLETED) NITRATE SOLUTION SYSTEMS (THIN STAINLESS-STEEL VESSELS ENCLOSE SYSTEMS)										
SPHERICAL	WATER	322 (H/X = 100)	4.6	70.9 (31.11 WT% Pu)	3.12	365	382	17.87**	1.038	
SPHERICAL	WATER	708 (H/X = 224)	4.6	35.1 (31.65 WT% Pu)	1.49	182	920	19.31**	1.034	1.026

* RADIUS OF SPHERES AND THICKNESSES FOR SLABS

(2) (3) (4) (5) (6) (7) (8)

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