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TECHNICAL ACTIVITIES QUARTERLY REPORT
AEC REACTOR DEVELOPMENT
AND TECHNOLOGY PROGRAMS
JULY, AUGUST, SEPTEMBER 1970

October 1970

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1522-1

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By

Staff of Battelle-Northwest

Program Coordinator:

F. G. Dawson

October 1970

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FOREWORD

This report covers all programs except the Waste Studies Program conducted by the Pacific Northwest Laboratory for the U.S. Atomic Energy Commission's Reactor Development and Technology Division. The objective of the Technical Activities Quarterly 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 quarter. 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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1.0 SUMMARY

PLUTONIUM UTILIZATION PROGRAM

Examination of hot-press and cold-press-sinter fuel irradiated in PRTR shows significantly more restructuring in hot-press pellet fuel and fuel-cladding in both types.

Transient tests conducted in SPERT show that although the cladding failure threshold energy is reduced by the presence of large PuO_2 particles in mixed-oxide fuels, the energy required for prompt fuel dispersal, which is the primary safety consideration, does not appear to be affected.

Electron microprobe investigations show that plutonium migrates toward the center of mixed-oxide fuels during irradiation and that uranium possibly moves outward toward the periphery of the rod.

Neutron yields and neutron and gamma dose rates of fuel assemblies scheduled for irradiation in San Onofre Reactor were measured at the reactor site. Agreement between measured and calculated neutron yields is excellent. Gamma dose rates are higher than those measured for a typical BWR fuel assembly, but the differences are expected. Neutron dose rates are undergoing additional refinement.

Discontinuities occurred in axial gamma scans of non-defected and defected UO_2 - PuO_2 cold-press-sinter rods. Pre- and postirradiation diameter profile measurements show that the cladding has plastically deformed and become oval shaped.

Profilometer data on mixed-oxide fuel rods irradiated in Saxton Core II show that maximum ovality of 0.0035 inch formed during irradiation.

A model for thermal neutron scattering in water was developed for use in the BMC-1 Monte Carlo code. Initial results show promise but there are several problem areas in the model which need correcting. The model is based on the data from ENDF/B file 7 for water.

The isotopic data obtained by Westinghouse from the destructive analysis of UO_2 -6.6 wt% PuO_2 rods irradiated in SAXTON CORE-II have been used to derive ratios of effective cross sections. Cross section ratios for samples from two regions of the core with different neutron energy spectrums are presented.

A series of experiments with a 1.0-inch square light water moderated lattice containing UO_2 - PuO_2 fuel rods have been conducted in the Physical Constants Testing Reactor (PCTR). These experiments provide reactivity data as a function of PuO_2 particle size distributed throughout the natural UO_2 diluent in 1/2-inch diameter, zircalloy clad, fuel rods. The concentration of natural boron, dissolved in the water moderator, corresponding to k_∞ of unity was measured as a function of PuO_2 particle size.

A special version of the computer code THERMOS, called GRANIT, has been completed and is in routine use. Position dependent thermal neutron spectra can be calculated for doubly heterogeneous systems using the GRANIT code.

HIGH TEMPERATURE REACTOR PHYSICS PROGRAM

The analysis of the first High Temperature Gas-Cooled Reactor (HTGR) experiment performed in the High Temperature Lattice Test Reactor (HTLTR) has been completed and a report of the work is in final form.

The analysis of the second HTGR experiment performed in the HTLTR has been completed and the report is in draft form.

Suitable fuel is being obtained for an HTLTR experiment using ThO_2 and PuO_2 in an HTGR lattice.

A series of reactor lattice physics experiments are to be conducted in the HTLTR in support of the Molten Salt Breeder Reactor Program.

The experiments in multizone critical assemblies (such as the HTLTR and PCTR) require a determination of neutron life-time for interpretation of reactivity measurements for the composite loading. An experimental method has been devised for this determination which uses a series of constant reactivity differences obtained from various pairs of rising reactor periods.

A procedure has been established to inspect routinely the vertical rods of the HTLTR to detect abnormal deformations during experimental programs at high temperatures.

A project to determine the feasibility of using the Fast Neutron Cavity in the Physical Constants Test Reactor for cross-section evaluation has been completed. The results indicate that the FNC can be used successfully for this purpose

STEADY STATE AND TRANSIENT SUBCHANNEL CODE DEVELOPMENT AND DATA ANALYSIS

COBRA-III has all the analysis capability of COBRA-II and can consider transients up to, but not including, sonic velocity propagation effects. The numerical scheme performs a boundary value solution to more properly include the effects of crossflow resistance, and the numerical solution is stable for all time steps.

Design of the experimental apparatus that will be used to investigate the turbulent flow structure of single-phase flow in rod-bundles is in progress. A common flow channel will be used to accommodate a variety of subchannel sizes and shapes.

PLUTONIUM CRITICALITY STUDIES

Critical Experiments with Unmoderated PuO₂

The results and analyses are presented from the latest series of experiments in a continuing program for determining the critical parameters of plutonium mixtures having concentrations that are typical of wet powders, precipitates, slurries and polymers. Previous measurements in this program were made on 15 H/Pu fuel having ²⁴⁰Pu isotopic concentrations of 2.2 and 8.08 wt% and on 5 H/Pu fuel having a ²⁴⁰Pu isotopic concentration of 11.46 wt%. This latest series of experiments were conducted with fuel in which the ²⁴⁰Pu isotopic content has been increased to 18.35 wt% and the H/Pu atomic ratio decreased to essentially zero.

In this fast neutron spectrum the percent change in spherical critical mass per percent change in ²⁴⁰Pu content was determined to be 2.07 for the reflected case and 1.88 for the bare case. In general, the results from these experiments indicate that the values for the critical sizes and masses of plutonium given in such references as TID-7028 should be increased for the highly concentrated systems. Although this completes the data needed for establishing the criticality curves in the fast region of the neutron spectrum, data are still needed in the intermediate region corresponding to plutonium concentrations between about 500 and 1500 g/liter.

NUCLEAR GRAPHITE

In-Reactor Creep

A series of graphite samples have been irradiated under a compressive stress of 1000 to 3000 psi. Dimensional changes are reported for these samples after an exposure of about 1.5×10^{21} n/cm². No meaningful creep constants can be calculated until completion of the second irradiation period.

Radiolytic Reactions in Graphite Moderated Reactors

The temperature coefficient was determined for the radiolytic reaction of carbon dioxide with hydrogen in a helium carrier gas. Those mixtures diluted with helium showed a much lower temperature coefficient than the mixtures without any helium present. These data permit an accurate extrapolation of radiolytic reactions to HTGR temperatures.

Effect of Sample Size on Dimensional Stability

A series of graphite samples which had previously been irradiated at low temperatures were irradiated at high temperature to about 1×10^{21} n/cm² ($E > 0.18$ MeV). This low exposure data suggests that cold seeded samples contract at a greater rate than samples irradiated totally at the high temperature. However, further exposure is required before definite conclusions can be drawn.

High-Temperature Irradiations

The capsule for irradiation of graphite samples at temperatures from 900 to 1600 °C has been recharged into the ETR. This capsule contains both previously irradiated samples and "new" samples.

Analysis of Coolant-Graphite Reactions

The code for calculating graphite oxidation in a high temperature gas cooled reactor (GOP) has been improved to include the greater flexibility required for evaluation of the 1100 MW_e HTGR.

Mechanism of Irradiation Damage

A study is underway to investigate the mechanism of radiation damage to graphite. Particular emphasis will be placed on coefficient of thermal expansion measurements to correlate with radiation damage.

CORROSION PRODUCT TRANSPORT STUDY

A ten-day test of an in-line, electrically activated crud generator device indicated that crud release rates from mild steel surfaces were not increased significantly by variations in electrical potential. A colorimetric procedure that uses Ferrozine as the indicator appears adequate for measurement of iron concentration in the 1 to 2 ppm range without preconcentration of the samples.

2. PLUTONIUM UTILIZATION PROGRAM

PRTR FUEL EVALUATIONS

D. G. Carter and M. D. Freshley

With the shutdown of PRTR, the irradiation performance of selected mixed-oxide rods irradiated in the reactor is being evaluated by means of destructive postirradiation examination. Recent adjustments in the scope of the program resulted in reducing the extent of planned fuel evaluations as described in previous quarterly reports. The PRTR fuel evaluations are divided into two groups as follows.

- High Power Density Fuel Evaluation
- Extended Burnup Fuel Evaluation

High Power Density Fuel Evaluation

Postirradiation examination of selected PRTR High Power Density fuel rods which were irradiated as part of the Batch Core Experiment (BCE) is continuing and the current status of the examination is summarized in Table 2.1. Most of the 66 High Power Density 19-rod cluster elements irradiated in the BCE were vibrationally compacted particle fuel, although pellet fuel fabricated by both the hot-press and cold-press-sinter processes was included.

Revised fission gas release values for five pellet-containing fuel rods based upon analytically determined burnup values using the ^{148}Nd isotopic dilution technique are summarized in Table 2.2. Hot-press pellet fuel releases significantly more fission gas than cold-press-sinter pellet fuel that operated under comparable conditions. The higher gas release is attributed to higher fuel temperatures which exist in hot-press pellet fuel because of its high stoichiometry ($\text{O/M} = \sim 2.15$) and resulting lower thermal conductivity.

TABLE 2.1. Summary of PRTR High Power Density Fuel Rod Postirradiation Examination Status

Element Number	Rod No.	Fuel Type	¹⁴⁸ Nd Peak Burnup, MWd/MTM	Gamma Scan	Profilometer Scan	Fission Gas Analysis	Burnup Analysis	Ceramography
6065	FE-74	UO ₂ -2 wt% PuO ₂ (VP)	4,863	X	Z	X	X	Deleted
	FO-17	(VP)	5,837	X	Z	X	X	Y
	FS-07	(VP)	9,176	X	Y	X	X	Deleted
6519	FR-72	(VP)	5,240	X	Y	X	X	Y
	FN-86	(VP)	3,297	X	Z	X	X	X
	FE-69	(VP)	2,884	X	Z	X	X	X
6520	FR-78	(VP)	9,630	X	Y	X	X	Y
6700	A-14	(HP)	8,150	X	Y	X	X	X
	A-103	(CPS)	3,290	Z	Y	X	X	X
	A-102	(CPS)	3,220	X	Y	X	X	X
6701	A-23	(HP)	3,430	X	Y	X	X	X
	A-12	(HP)	3,800*	Z	Y	X	Deleted	X

(VP) = *Vibrationally compacted, pneumatically impacted fuel rod*

(CPS) = *Cold-press-sinter pellet fuel rod*

(HP) = *Hot-press pellet fuel rod*

X = *Examination completed*

Y = *Examination in progress*

Z = *Examination not scheduled*

* = *Estimated burnup*

TABLE 2.2. Summary of Postirradiation Examination Results for Pellet-Containing UO₂-2 wt% PuO₂ Fuel Rods Irradiated in the PRTR

Rod No.	Rod Type	Estimated Peak Power, kW/ft	Estimated Peak Burnup, MWd/MTM	¹⁴⁸ Nd Peak Burnup, MWd/MTM	Fission Gas Releases, %
A-102	CPS ^(a)	16-18	3,650	3,220	19.3
A-103	CPS	16-18	3,650	3,290	17.9
A-14	HP ^(b)	16-18	9,600	8,150	60.9
A-12	HP	10-14	3,800	---	36.7
A-23	HP	10-14	3,800	3,430	37.9

a. CPS refers to cold-press-sinter pellet fuel

b. HP refers to hot-press pellet fuel

Ceramographic examination of cold-press-sinter pellet fuel rods (A-102 and A-103) irradiated in PRTR shows fission product inclusions and lenticular shaped pores in the columnar grain growth region near the central void (Figure 2.1). A nonuniform oxide layer formed on the inner surface of the Zircaloy-4 cladding (Figure 2.2) and what appears to be a reaction between the contiguous oxide layer and the fuel occurred in some regions of the fuel-cladding interface (Figure 2.3).

Extensive restructuring occurred in hot-press pellet fuel during irradiation in PRTR (Figure 2.4). In addition to the normal fission product migration, which occurs in oxide fuels during irradiation, the beta-gamma autoradiographs indicate the presence of high fission product concentrations associated with cracks in the fuel at the fuel-cladding interface. Again, a uniform reaction occurred between the contiguous oxide layer on the inner surface of the cladding and the fuel (Figure 2.5).

Neg F4060

250X

FIGURE 2.1. Transverse Section of a UO_2 -2 wt% PuO_2 Cold-Press-Sinter Pellet Fuel Rod (A-103) Irradiated in PRTR Showing Fission Product Inclusions and Lenticular Shaped Pores Near the Central Void

ZIRCALOY
CLADDING

ZrO_2 LAYER

UO_2 - PuO_2 FUEL

Neg F3806

750X

FIGURE 2.2. Fuel-Cladding Interface from a UO_2 -2 wt% PuO_2 Cold-Press-Sinter Pellet Fuel Rod (A-103) Showing Intermittent Fuel-Cladding Reaction

Neg F3807

750X

FIGURE 2.3. Fuel-Cladding Interface from a UO_2 -2 wt% PuO_2 Cold-Press-Sinter Pellet Fuel Rod (A-103) Showing Contiguous Fuel-Clad Interaction

In addition, what appears to be severe localized oxidation occurred in some regions of the inner surface of the cladding in these hot-press pellet fuel rods (Figures 2.6 and 2.7). The severe reaction is probably the result of the excess oxygen ($\text{O/M} \approx 2.15$) in this type of fuel.

Extended Burnup Fuel Evaluation

Postirradiation evaluation of selected fuel rods irradiated to extended burnups in PRTR fringe positions continued. The status of the examination of these rods is summarized in Table 2.3.

Neg F3854

Photomicrographs

Neg 1824

 β - γ Autoradiographs

Neg 1826

 α Autoradiographs

Neg F3822

Neg 1817

Neg 1818

FIGURE 2.4. Transverse Sections from the Peak Power Position of $\text{UO}_2\text{-PuO}_2$ Hot-Press Pellet Fuel Rods Irradiated in PRTR. The estimated peak heat rating in Rod A-12 (top) ranged from 10 to 14 kW/ft and from 16 to 18 kW/ft in Rod A-14 (bottom).

Neg F4062

250X

FIGURE 2.5. Fuel-Cladding Interface from a UO_2 -2 wt% PuO_2 Hot-Press Pellet Fuel Rod (A-14) Showing Contiguous Fuel-Clad Reaction

The fuel structure in a vibrationally compacted UO_2 -2 wt% PuO_2 rod shows no evidence of fuel melting although irradiation at a peak linear heat rating greater than 20 kW/ft during the first part of the irradiation should have produced a melting condition (Figure 2.8). Vapor phase transport phenomena eradicated evidence of the once-molten structure during subsequent operation at an estimated heat rating of 10 kW/ft to a burnup of about 13,000 MWd/MTM. This phenomenon has been observed during the examination of other mixed-oxide fuel rods. The beta-gamma autoradiographs show an unexpected heterogeneous fission product distribution in the homogenized region of this mechanically-mixed high burnup fuel.

Neg F4064

250X

FIGURE 2.6. Fuel-Cladding Interface from A UO₂-2 wt% PuO₂ Hot-Press Pellet Fuel Rod (A-14) Showing Severe Localized Attack

Neg F4065

250X

FIGURE 2.7. Fuel-Cladding Interface from a UO₂-2 wt% PuO₂ Hot-Press Pellet Fuel Rod (A-14) Showing Severe Localized Attack

TABLE 2.3. Summary of the Postirradiation Examination Status for Fuel Rods Irradiated to Extended Burnup in PRTR Fringe Positions

Element Number	Rod No.	Fuel Type	Estimated Peak Burnup, MWd/MTM	Gamma Scan	Profilometer Scan	Fission Gas Analysis	Burnup Analysis	Ceramography
1037	A-1330	UO ₂ (SW)	12,075*	X	Z	X	X	X
	A-1377	↓	12,033*	X	Z	X	X	X
	A-1398	↓	15,368*	X	Z	X	X	X
	A-1325	↓	---	Z	Z	X	Z	Deleted
5118	DB-63	UO ₂ -0.5 wt% PuO ₂ (VP) (MM)	18,500	X	Z	X	Z	Y
	DA-48	↓	18,500	Z	Z	X	Z	Deleted
5186	ZZ-17	↓	---	X	Z	X	X	Deleted
	ZZ-7	↓	---	X	Z	X	X	Deleted
	ZZ-12	↓	12,500	X	Z	X	X	Y
	CN-5	↓	12,500	X	Z	Lost	Z	X
	CN-7	↓	12,500	Z	Z	X	Z	Z
	CM-85	↓	12,500	Z	Z	X	Z	Z
5224	CS-27	UO ₂ -1 wt% PuO ₂ (SW) (PI)	13,500	X	Z	X	Deleted	X
5226	DF-80	↓	11,500	X	Z	X	Deleted	Y
6003	5C-27	UO ₂ -2 wt% PuO ₂ (VP) (PI)	13,000	X	Z	X	Deleted	X
6005	5F-36	↓	8,000	X	Z	X	X	Y
6521	No. 6	ThO ₂ -5 wt% PuO ₂ (VP) (PI)	Low	X	Z	Lost	Z	X

(VP) = Vibrationally compacted fuel rod

(SW) = Swage compacted fuel rod

(CPS) = Cold-press-sinter pellet fuel rod

(MM) = Heterogeneously enriched or incrementally loaded fuel material

(PI) = High-energy-rate pneumatically impacted fuel

X = Examination completed

Y = Examination in progress

Z = Examination not scheduled

* = Burnup values determined by ¹⁴⁸Nd analysis

Neg F3798

Photomicrograph

Neg 1821

 β - γ Autoradiograph

Neg 1822

 α Autoradiograph

FIGURE 2.8. Transverse Section of a Vipac UO_2 -2 wt% PuO_2 Fuel Rod Irradiated in PRTR to an Estimated Peak Burnup of 13,000 MWd/MTM

TRANSIENT TESTING

M. D. Freshley, E. A. Aitken,* and R. L. Johnson**

A knowledge of the behavior of reactor fuels when subjected to high energy, short duration power excursions is essential to permit a complete evaluation of the effect of fuel performance under accident conditions on reactor safety. A joint program involving Pacific Northwest Laboratory, General Electric Company, and Idaho Nuclear Corporation was conducted in SPERT to determine whether special considerations associated with adding PuO_2 enrichment to UO_2 fuels affect transient behavior. Emphasis was directed toward investigating the possible effects of large PuO_2 particles or agglomerates which may be inadvertently introduced in mixed-oxide fuels when utilizing standard cold-press-sinter pellet fabrication techniques. Transient behavior of the fuel is an important consideration when evaluating the design basis accident for a given reactor system.

Four fuel pins containing 0.272-inch diameter enriched (7% ^{235}U) UO_2 pellets with a single PuO_2 microsphere embedded in alternate pellets in the 5.1-inch long fuel column were assembled in 0.020-inch thick Zircaloy-2 cladding with a nominal 0.004-inch diametral gap. The PuO_2 microspheres were strategically located within the pellets ranging from the center to the surface to investigate the possible influence of different spatial positions relative to the cladding. A nominal PuO_2 particle size of 500 μm was selected for the initial tests because it is a size that can be readily detected by neutron radiography and because it is considered to be near the upper size limit for particles that could be inadvertently introduced in the fabrication process. A standard pin design was used to permit a comparison of the results from these tests with the results of other tests involving different fuel types.

* General Electric Company

** Idaho Nuclear Corporation

Test capsule instrumentation in SPERT permitted measurement of the following dynamic responses during irradiation: fuel column axial displacement, fuel rod cladding axial displacement, capsule pressure, water column velocity, and cladding surface temperature rise. Postirradiation measurements included hydrogen gas evolution and flux wire measurements.

Each of the four fuel pins was subjected to a single power burst that resulted in adiabatic energy depositions of 189, 200, 213, and 237 cal/g of UO_2 . The test conditions and results of the transient tests are summarized in Table 2.4.

TABLE 2.4. Summary of Test Conditions and Results of Transient Tests with Pins Containing Discrete PuO_2 Particles in Pellet Fuel

Test No.	Energy Deposition, cal/g UO_2	Reactor Period, msec	Max. Meas. Cladding Temp, $^{\circ}\text{C}$	Residual Rod Growth, in.	Results
1	189	6.78	1450	0.026	No external damage
2	200	6.22	1600	0.013	No external damage
3	213	5.79	1725	0.007	Two clad penetrations by PuO_2 particles
4	237	5.32	1825	---	Four clad penetrations by PuO_2 particles, clad severely wrinkled

No cladding penetrations occurred in the two fuel pins irradiated at 189 and 200 cal/g UO_2 . The most noticeable effects of irradiation were a slight bowing of the pins and the formation of a dark zirconium oxide layer over the active fuel length. Small discolored areas may be evidence of localized overheating associated with PuO_2 particles near the surface of the pellets.

Cladding penetrations approximately 600 to 1200 μm in diameter developed in the two pins irradiated at energies above 213 cal/g UO_2 at some of the locations where PuO_2 microspheres were positioned within 100 μm of the cladding. Although the general condition of the pin irradiated at an energy input of 213 cal/g UO_2 was not adversely affected by the irradiation, the cladding was penetrated by two small holes associated with PuO_2 particles near the surface. Severe cladding distortion and wrinkling indicative of melting occurred in the pin irradiated at an energy input of 237 cal/g UO_2 and four small penetrations associated with PuO_2 particles occurred (Figure 2.9). In addition, there were numerous other small cracks and penetrations in the cladding that were not directly associated with PuO_2 particles.

The detailed postirradiation examination of these pins is in progress. Preliminary results show that even in the pins subjected to the lower energy inputs, microspheres further removed from the cladding expanded by a factor of approximately 2 within the surrounding UO_2 matrix and developed a cavity in the center of the microsphere (Figure 2.10). Molten PuO_2 extruded for distances greater than 1000 μm into pre-existing cracks in the UO_2 .

It is postulated that cladding penetrations were caused by localized melting as the molten PuO_2 particles were ejected from the fuel surface by the force created by their vapor pressure at very high temperatures. The amount of fuel released was limited primarily to the PuO_2 microspheres.

The prompt cladding failure threshold for fresh homogeneous mixed-oxide fuel is above 225 cal/g UO_2 . Based upon the results of these experiments, the cladding failure threshold for thermal recycle fuel containing 500 μm diameter PuO_2 agglomerates is

FIGURE 2.9. Posttransient Appearance of a Zircaloy-Clad Pellet Containing Pin Irradiated at an Energy Input of 237 cal/g UO_2 . (Small cladding penetrations were caused by the expulsion of discrete PuO_2 particles placed near the surface of the pellets.)

Neg 61153

|←500 μm→|

FIGURE 2.10. A 500 μm PuO_2 Particle in a UO_2 Matrix After Transient Irradiation in SPERT. (The diameter of the particle increased to 800 μm and a central cavity formed.)

only slightly lower at 213 cal/g UO_2 although no significant fuel dispersal was associated with cladding penetrations. The threshold energy for prompt fuel dispersal, which is accompanied by a detectable conversion of thermal to mechanical energy, commences at about 325 cal/g UO_2 . A primary safety consideration, which is prompt fuel dispersal accompanied by a significant conversion of thermal to mechanical energy, occurs at approximately 425 cal/g UO_2 and does not appear to be significantly affected by the presence of a 500 μm diameter PuO_2 agglomerate.

PLUTONIUM AND FISSION PRODUCT MIGRATION IN MIXED-OXIDE FUELS
DURING IRRADIATION

M. D. Freshley and J. K. Bahl*

Plutonium and fission product migration in mixed-oxide fuels during irradiation are important considerations when assessing fuel performance. Quantitative studies utilizing the shielded electron-microprobe show that significant plutonium redistribution occurs in high performance mixed-oxide fuels during irradiation. Qualitative evaluations of fission product migration utilizing autoradiography indicate that this technique is a sensitive indicator of the fission product distribution in irradiated fuels and that migration of some species is coincident with the fuel restructuring phenomenon and occurs rapidly during irradiation.

Studies were performed on vibrationally compacted UO_2 -2 wt% PuO_2 fuel pin irradiated for approximately 230 hours in PRTR at a maximum linear heat rating of 20 kW/ft which produced fuel melting at the time of shutdown. The 86% of theoretical density fuel with an O/M ratio of 2.015 was clad in Zircaloy-4 with an 0.565-inch outside diameter and a nominal 0.030-inch wall thickness.

Plutonium, uranium and fission product concentrations were evaluated by scanning along different radii on the specimen with a $1\text{ }\mu\text{m} \times 10\text{ }\mu\text{m}$ beam in 5 or 10 μm intervals. The 10 μm dimension was perpendicular to the radial direction of travel.

The results of five radial scans show that the plutonium concentration is uniform in the once-molten fuel region and is 18% higher than the nominal value. The discontinuity in the plutonium concentration occurs at the maximum melt radius which corresponds to the outer boundary of the pore-free band

* *Exchange scientist from Bhabha Atomic Research Center, Bombay, India*

surrounding the central subgrain structure region (Figure 2.11). The uniformity of the plutonium concentration in this region is attributed to a convection mixing phenomenon.⁽¹⁾

Plutonium migration towards the thermal center occurred beyond the molten boundary in the columnar grain growth region. A minimum plutonium concentration equal to the nominal value occurred at 44% of the radius from the thermal center where the fuel temperature was estimated to have been 2500 °C. Plutonium redistribution in the columnar grain region is attributed to preferential evaporation of uranium oxide in the hyperstoichiometric fuel during the continual movement of voids up the thermal gradient by a vapor transport mechanism.^(2,3) A limited amount of plutonium migration up the thermal gradient by a thermal diffusion mechanism is also likely to have occurred in this region.⁽²⁾

Beyond the region where homogeneous solid solution formed during irradiation, the plutonium concentration is erratic because of the nature of the mechanically-mixed mixed-oxide fuel. The average plutonium concentration in the fuel region where relocation phenomena occurred is 12% greater than the nominal concentration in the as-fabricated fuel. This may be the result of preferential evaporation of gross amounts of uranium from the hotter central region of the rod to the cooler peripheral region near the cladding.^(2,3)

Autoradiography indicated the presence of short-lived α -activity associated with the molten region as delineated by the outer boundary of the pore-free band that disappeared within 8 months decay. β - γ autoradiographs show the normal fission product activity distributions associated with once-molten fuel (Figure 2.11) although the fission product concentration in this low burnup sample was not sufficient to be detected by the electron microprobe which has an estimated detectable limit of 500 ppm.

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FIGURE 2.11. Radial Plutonium Concentration in an Irradiated UO_2 -2 wt% PuO_2 Fuel Rod That Operated with Limited Fuel Melting. (Photo shows electron microprobe scan locations and autoradiographs.)

References

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HIGH EXPOSURE PLUTONIUM STUDIES

L. G. Faust and R. L. Gulley

Neutron yields and neutron and gamma dose rates from mixed-oxide fuel assemblies scheduled for irradiation in the San Onofre Reactor were measured at the reactor site. The measurements were made with four assemblies stored in a single row with 12-inch edge-to-edge spacing between assemblies (Figure 2.12).

Each of the assemblies contain 180 plutonium-bearing fuel rods and 16 nonfueled rods in a 14×14 array. Each assembly is 8 inches square by 135 inches long and contain a total of 11.25 kg of plutonium with the following isotopic composition:

<u>Isotope</u>	<u>wt%</u>
^{238}Pu	0.34
^{239}Pu	80.55
^{240}Pu	13.44
^{241}Pu	4.93
^{242}Pu	0.74

FIGURE 2.12. San Onofre Fuel Assembly Arrangement

The PuO_2 concentration in the rods included in each assembly is as follows:

<u>PuO_2 Enrichment</u>	<u>Number of Rods</u>
3.34 wt%	24
3.12 wt%	92
2.93 wt%	64

Gamma dose rates were measured with a thermal luminescence dosimeter (TLD). The results of these measurements are considerably higher than similar measurements made with BWR plutonium-bearing assemblies. However, this is to be expected because the total plutonium contained in these assemblies is nearly twice the amount contained in the most highly plutonium enriched BWR assembly measured to date (i.e. 11.25 kg versus 6 kg plutonium per assembly). In addition, PWR fuel cladding is typically thinner than BWR fuel rod cladding (i.e. 24 mils versus 30 mils), and possible differences in the ages of the

plutonium could significantly affect the measurements. Gamma dose rates measured at different distances from the end assembly (Figure 2.12) are presented below. Dose rate measurements of a BWR assembly⁽¹⁾ containing a total of 6 kg of plutonium are included for comparison.

Distance, in.	Gamma Dose Rates	
	PuO ₂ -UO ₂ Fuel Assemblies	
	Dose Rate, mR/hr	
	PWR	BWR
0.75	36	12
6	13	3.2
24	6.5	1.9

Neutron yields from the four assemblies were measured with a precision long counter located from 4 to 18.75 feet from the end assembly (Figure 2.12). The effect of neutron scatter from surrounding concrete walls in the assembly storage area was considered and experimental correction factors were determined in a low level flux room before the final measurements were made. Using the correction factors, the measured neutron yield (four assemblies) was 1.12×10^7 neutrons per second. A calculated yield of 1.08×10^7 neutrons per second, which is flux-weighted for the geometric configuration, compares favorably with the measured yield. The calculated neutron yield for an individual assembly is 3.56×10^6 neutrons per second.

Neutron dose rate measurements were made with a Tissue Equivalent Proportional Counter (TEPC). Preliminary analysis of this data indicates a neutron dose rate of about 25 mrem/hr at about 2 inches and about 14 mrem/hr at 25 inches from the end assembly (Figure 2.12). These values include a Quality Factor of 10 and 8.4 respectively. Both the dose rate and the Quality Factors are subject to refinement upon completion of the data analysis.

Reference

1. Plutonium Utilization Program Technical Activities Quarterly Report, March, April, and May 1970, BNWL-1442, pp. 3.14-3.15, Battelle-Northwest. June 1970.

DEFECT TESTING

M. D. Freshley and D. G. Carter

The performance of intentionally defected and nondefected mixed-oxide fuels was investigated by performing irradiations in the ETR M-3 Loop facility. This program was terminated after performance of the first defect test.

Mixed-oxide fuel rods incorporating different cold-press-sinter pellet designs were irradiated in a 12-rod cluster assembly as part of the defect testing experiment. The design parameters associated with the different fuel rod designs tested are summarized in Table 2.5.

TABLE 2.5. Design Parameters for Mixed-Oxide Fuel Rods Irradiated During the Defect Testing Experiment

<u>Rod Type</u>	<u>Pellet Design</u>	<u>Diametral Gap, in.</u>	<u>Weight % PuO₂</u>
1	Solid	0.006	4.0
2	Solid	0.010	4.0
3	Dished ^(a)	0.006	4.0
4	Dished	0.010	4.0
5	Cored ^(b)	0.006	4.4

a. Dish volume is 3% of pellet volume.

b. Core volume is 10% of pellet volume.

Each pellet-containing rod is nominally 0.565-inch OD clad in 0.030-inch thick Zircaloy-4 cladding with a 26.8-inch active fuel length. The irradiations were conducted at peak heat ratings ranging from 14 to 23 kW/ft with maximum fuel temperatures near melting in the solid pellet rods and approximately 2100 °C in the cored pellet rods. Peak burnups of approximately 3000 MWd/MTM were achieved before the tests were terminated. The system pressure was 2000 psi at an average coolant temperature of 490 °F.

A Type 2 rod was intentionally defected by drilling a 0.020-inch diameter hole through the cladding in the peak power region after the rod was irradiated to a peak burnup of about 1200 MWd/MTM. The defected rod was subsequently irradiated for 17.3 effective full power days at a linear heat rating of about 13.5 kW/ft which produced maximum fuel temperatures of approximately 1650 °C. Rotation of the element increased the heat rating at the defect to about 20 kW/ft with associated maximum fuel temperatures of 2400 °C for an additional 14.1 effective full power days.

Gross gamma activity levels monitored in the loop piping system increased in proportion with the power but remained well within acceptable limits. There were no indications of particulate fuel release and there were no activity bursts associated with changes in reactor power level, some of which were severe, that characterize the defect behavior of particle fuel.

Axial gamma scanning and diameter profile measurements were made on two nondefected Type 1 rods with a peak burnup of approximately 1200 MWd/MTM and the defected Type 2 rod with a peak burnup of 3000 MWd/MTM. The gross gamma scan of nondefected F-10 shows a smooth fission product activity distribution that is related to the axial power profile (Figure 2.13).

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FIGURE 2.13. Axial Gamma Scan for Nondefected Cold-Press-Sinter Pellet Fuel Rod F-10

BNWL-1522-1

A discontinuity in the gamma scan occurred above the peak power position in rod F-11 (Figure 2.14). The discontinuity is possibly the result of axial movement of the pellet fuel column.

The gamma scan of defected rod F-12 shows two localized variations in the fission product activity curve at and above the peak power position (Figure 2.15). The perturbations, which are possibly the result of axial fuel relocation or slumping, do not appear to be related to the position of the defect.

Pre- and postirradiation diameter profile measurements on the rods show that the cladding has plastically deformed and become oval shaped during irradiation in the pressurized loop (Figures 2.16 through 2.18). The maximum ovality increase in nondefected rods F-10 and F-11, which were assembled with a cold diametral gap of 0.006 inch, is 0.004 inch and 0.009 inch, respectively. The relatively large maximum ovality in rod F-11 is localized. The maximum ovality increase in defected rod F-12, which was assembled with a nominal 0.010-inch diametral gap, is 0.006 inch. Cladding collapse on the defected rod must have occurred during the relatively short predefect irradiation of 35 effective full power days because no collapse type deformation is expected to occur after defecting. There was no evidence of clad ridging at pellet interfaces in any of the rods. These results show that clad deformation, even in cladding that is calculated to be free-standing, commences during the early stages of irradiation under these conditions.

FIGURE 2.14. Axial Gamma Scan for Nondefected Cold-Press-Sinter Pellet Fuel Rod F-11

POINT OF DEFECT

FIGURE 2.15. Axial Gamma Scan for Defected Cold-Press-Sinter Pellet Fuel Rod F-12

FIGURE 2.16. Preirradiation (Top) and Postirradiation (Bottom)
Diameter Profile Scans of Nondefected Fuel Rod F-10

FIGURE 2.17. Preirradiation (Top) and Postirradiation (Bottom)
Diameter Profile Scans of Nondefected Fuel Rod F-11

FIGURE 2.18. Preirradiation (Top) and Postirradiation (Bottom)
Diameter Profile Scans of Defected Fuel Rod F-12

SAXTON FUEL EXAMINATION

D. G. Carter and M. D. Freshley

The dimensional stability of four Zircaloy-4 clad UO_2 -6.6 wt% PuO_2 fuel rods irradiated in the PWR environment of Saxton Core II was evaluated by continuous profilometer scanning the outside diameter of each rod along its full length.

Three of the rods examined contained $94 \pm 2\%$ TD cold-press-sinter pellet fuel and one contained $87 \pm 1\%$ TD vipac particle fuel. The pellet rods were assembled with a nominal 0.007-inch diametral gap in the 0.391-inch OD tubing which had an 0.023-inch wall thickness. The overall length of each rod is 41 inches with an active fuel length of 36.6 inches. The rods were exposed for 6350 effective full power hours to a coolant pressure of 2000 psi at an average inlet temperature of 503 °F and subsequently for 9387 effective full power hours to a coolant pressure of 2200 psi at an average temperature of 483 °F. A summary of the irradiation history and profilometer data for each rod is presented in Table 2.6.

TABLE 2.6. Irradiation History and Profilometer Data for Saxton Core II Rods

<u>Rod Number</u>	<u>Rod Type</u>	<u>Estimated Peak Rod Power, kW/ft</u>	<u>Estimated Peak Burnup, MWd/MTM</u>	<u>Maximum Ovality, in.</u>
F-2(HG)	Pellet	14.0	25,400	0.0035
F-3(AQ)	Pellet	8.5	15,500	0.0034
D-5(KF)	Pellet	9.5	17,400	0.0055
A-3(S3)	Vipac	14.5	26,500	0.0020

Ten unirradiated Saxton rods were profilometer scanned to obtain data on the preirradiation condition of the rods. This data shows a minimum amount of ovality, i.e., less than 0.0005-inch (Figure 2.19). Postirradiation profilometer data on the three pellet-containing rods show maximum ovality of about 0.0035 inches (Figures 2.20 through 2.22). There was no evidence of ridging at pellet interfaces. The smaller ovality measured on the vipac rod (maximum 0.0020 inch) is a result of the improved cladding support provided by this fuel type (Figure 23). These data are in basic agreement with profilometer data obtained by Westinghouse Electric Corporation on similar rods.⁽¹⁾

Reference

1. Saxton Plutonium Program Semi-Annual Progress Report for the Period Ending June 30, 1969, WCAP-3385-20. Westinghouse Electric Corporation, Pittsburgh, Pennsylvania.

NEG. 705000-12

FIGURE 2.19. Diameter Profile Scan of Typical Unirradiated Vibrationally Compacted Saxton Fuel Rod

BNWL-1522-1

2.34

NEG. 705000-20

FIGURE 2.20. Postirradiation Diameter Profile Scan of Pellet-Containing Saxton Fuel Rod F3-8(AQ)

BNWL-1522-1

2.35

NEG. 705000-13

FIGURE 2.21. Postirradiation Diameter Profile Scan of Pellet-Containing Saxton Fuel Rod D5-4(kF)

BNWL-1522-1

NEG. 705000-76

FIGURE 2.22. Postirradiation Diameter Profile Scan of Pellet-Containing Saxton Fuel Rod F2-9 (HQ)

BNWL-1522-1

2.37

NEG. 705000-17

FIGURE 2.23. Postirradiation Diameter Profile Scan of Vibrationally Compacted Saxton Fuel Rod A3-1(S3)

BNWL-1522-1

MONTE CARLO THERMAL NEUTRON SAMPLING MODEL FOR WATER

D. H. Thomsen

An effort to provide a better thermal neutron scattering model for use in the BMC⁽¹⁾ Monte Carlo code has begun. Development work was done on a scattering model for 296 °K water. This model is based on the $S(\alpha, \beta)$ representation of thermal neutron scattering. Presently, ideal gas scattering is used to provide for energy changes. For many calculations this results in a too soft thermal spectrum. The new model should give more correct energy distribution along with a better angular distribution.

The double differential scattering cross section for a chemically bound substance is given by the equation.

$$\frac{\partial \sigma(E_0 \rightarrow E, \mu)}{\partial E \partial \Omega} = \frac{\sigma_\beta}{4\pi kT} \sqrt{\frac{E}{E_0}} e^{-\beta/2} S(\alpha, \beta)$$

where $\beta = \frac{E - E_0}{kT}$ is the dimensionless energy change and

$\alpha = \frac{E_0 + E - 2\mu\sqrt{EE_0}}{akT}$ is the dimensionless momentum change.

It would require a three-dimensional array to select random samples directly from the double differential cross section. This would require too much memory space for many computer applications. However, a space saving method independently suggested by C. W. Lindenmier⁽²⁾ and M. H. Kalos⁽³⁾ and partially developed by K. B. Cady⁽⁴⁾ and Frank G. Bischoff⁽⁵⁾ was used. This method allows sampling using several two-dimensional arrays and hence greatly reduces the required computer memory space.

The idea is to first obtain an energy change and then to use a sampling distribution in the variables α and β to obtain the scattering angle. Memory space is also reduced by treating upscattering and downscattering separately.

The main problem in implementation of this method is processing and fitting the data. Data was obtained from ENDF/B-file 7 for water. The necessary data was processed by using a modified version of the FLANGE-II⁽⁷⁾ code. The data was fit and a FORTRAN subroutine was written to sample from the fitted data.

Very little testing of this routine has been done. However, what has been done indicates that it works fairly well over most of the energy range, but there is a part of the energy range which either has an error in the routine or has an inadequate fit to the data. It is expected that further development and testing of this model must be postponed due to reduced funding and other higher priority work.

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THE DETERMINATION OF RATIOS OF EFFECTIVE CROSS SECTIONS FROM
BURNUP DATA FROM THE SAXTON PLUTONIUM PROGRAM

P. E. Coudron and R. P. Matsen

The isotopic data obtained by Westinghouse from the destructive analysis of UO_2 -6.6 wt% PuO_2 rods irradiated in SAXTON Core II⁽¹⁾ have been used to derive preliminary values of ratios of effective cross sections. The information was obtained by fitting the appropriate transmutation equations to these data via the DBUFIT-I⁽²⁾ least squares analysis code. The data have been separated into two groups according to the type of neutron energy spectrum in which the sample was irradiated. Data from each of these groups have been analyzed separately for cross section ratios. Samples from the interior rods of a Saxton fuel bundle are from the "asymptotic" region which comprises the largest portion of the fuel. Samples from the edge of a bundle are influenced by extra water moderator resulting in a different neutron spectrum than exists in the interior of the bundle and are designated as being from the "water slot" region.

The data from the asymptotic region of the core and the best least squares fit to this data are presented graphically in Figure 2.24. One-standard-deviation uncertainties are attached to the plotted data points. The best least squares fit for each isotope is also shown bounded by its one standard deviation uncertainty. The fit was forced through the zero exposure data point as is evident from the accompanying figure. The cross section ratios obtained from the least squares analyses of both sets of data are given in Table 2.7.

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FIGURE 2.24. Plutonium Isotopes Versus ^{235}U for Saxton Asymptotic Data

TABLE 2.7. Cross Section Ratios Derived
from the SAXTON Core II
Burnup Data

<u>Cross Section Ratio</u>	<u>Asymptotic Data</u>	<u>Water Slot Data</u>
$\hat{\sigma}^{25} = \hat{\sigma}_c^{25} / \hat{\sigma}_f^{25}$	0.2878 ± 0.0026	0.2774 ± 0.0035
$\hat{\sigma}_c^{28} / \hat{\sigma}_a^{25}$	0.0315 ± 0.0016	0.0320 ± 0.0011
$\hat{\sigma}_a^{49} / \hat{\sigma}_a^{25}$	$1.278 + 0.8400/N^{49*}$	$2.010 + 0.2068/N^{49*}$
$\hat{\sigma}^{49} = \hat{\sigma}_c^{49} / \hat{\sigma}_f^{49}$	0.5074 ± 0.0062	0.5140 ± 0.0060
$\hat{\sigma}_a^{40} / \hat{\sigma}_a^{25}$	$1.77 + 0.0287/N^{40*}$	$0.852 + 0.275/N^{40*}$
$\hat{\sigma}_a^{41} / \hat{\sigma}_a^{25}$	2.37 ± 0.20	2.54 ± 0.21

The superscripts are $25 = {}^{235}\text{U}$, $28 = {}^{238}\text{U}$, $49 = {}^{239}\text{Pu}$,
 $40 = {}^{240}\text{Pu}$ and $41 = {}^{241}\text{Pu}$.

* N^{49} and N^{40} are the isotopic concentrations of ${}^{239}\text{Pu}$ and
 ${}^{240}\text{Pu}$, respectively, in units of 10^{20} atoms/g U+Pu.

References

1. W. R. Smalley. Saxton Plutonium Program Semiannual Progress Report for the Period Ending December 31, 1969, WCAP-3385-22. Westinghouse Electric Corporation, Pittsburgh, Pennsylvania, March 1970.
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PuO₂ PARTICLE SIZE REACTIVITY EFFECT

D. F. Newman and C. R. Gordon

Fuel element development for plutonium recycle has led to fuels composed of the mixed oxides of plutonium and uranium. These fuel elements are presently fabricated by distributing the plutonium throughout the uranium as particles. Due to self-shielding properties of the particle, the reactivity of the fuel is a function of particle size. Since a change in particle size is probable either in the event of a reactor transient or during normal fuel irradiation and burn-up, knowledge of the effect that plutonium particle size has on the reactivity of a reactor system is important for reactor safety and the economic utilization of plutonium.

A series of experiments with a 1.0-inch square light water moderated lattice containing UO₂-PuO₂ fuel rods have been conducted in the Physical Constants Testing Reactor (PCTR). These experiments provide reactivity data as a function of PuO₂ particle size distributed throughout the natural UO₂ diluent in 0.5-inch diameter, zircalloy clad, fuel rods. The concentration of natural boron, dissolved in the water moderator, corresponding to k_{∞} of unity was measured as a function of PuO₂ particle size.

PCTR Water Tank Experiment

The PCTR water tank, containing 0.5-inch diameter fuel rods in a light water moderated 1.0-inch square lattice pitch was installed in the central PCTR test cavity as shown in Figure 2.25. The central lattice assembly, shown in Figure 2.26, was inserted in the innermost region of the water tank. A 20-inch long center cell, consisting of a set of four rods, could be removed from the central lattice assembly and replaced with an aluminum void can. The central region end

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FIGURE 2.25. Water Moderated $\text{UO}_2\text{-PuO}_2$ Lattice Installed in the PCTR

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FIGURE 2.26. Central Lattice Assembly with Removable Center Cell and Aluminum Void Can

buffers are shortened duplicates of the center cell fuel such that the total length of the assembly does not exceed the 37.5-inch length of the test cavity.

Ten different sets of four center cell fuel rods with known PuO_2 particle size diameters were used in the PCTR water tank experiment. These fuel rod sets can be categorized into five different PuO_2 particle size diameters: 0, 52, 107, 195, and 328 microns. The change in reactivity of the PCTR due to the substitution of a void for each of the ten sets of center cell rods was measured with natural boron dissolved in the water moderator at concentrations of 1200, 1300, and 1400 weight-parts-per-million (wppm). The boron concentration corresponding to the null-reactivity condition, for each center cell fuel composition, was obtained by interpolation of the center cell reactivity worth versus boron concentration data.

Foil activation measurements were made in the central cell having the approximate boron concentration for null reactivity. These activation measurements will be used to infer the relative reaction rates in all central cell constituents. Bare and cadmium-covered foils were irradiated separately in the moderator and in special separable fuel rods having plutonium particles sized at zero microns and 328 microns in diameter with boron concentrations of 1300 and 1200 wppm, respectively. A similar set of foil activation measurements were made in the unpoisoned lattice using both the zero micron and 328-micron particle sized fuel rods.

RESULTS AND CONCLUSIONS

The measurement in the PCTR consists of simultaneously determining the matched spectrum incident on the central cell and the proper central cell constituents such that the change in the PCTR reactivity is zero when the central cell is removed

leaving a void. The k_{∞} of the void as seen by the PCTR is unity. With no change in neutron distribution or reactivity upon re-insertion of the central cell, the k_{∞} of the central cell material is thus implied to be unity.⁽¹⁾

The measured value of the concentration of natural boron dissolved in the water moderator at the null-reactivity condition is listed for each center cell fuel composition in Table 2.8. The lattice region surrounding the center cell was identically poisoned with boron in the water, such that the neutron energy distribution in the center cell duplicated the neutron spectrum that would have been present in the center of a bare, just critical, array of identical cells. Calculated predictions of neutron spectrum equilibrium in the region of the center cell were confirmed by gold cadmium ratio measurements.

TABLE 2.8. $\text{UO}_2\text{-PuO}_2$ Lattice Composition at $k_{\infty} = 1$

Effective Spherical PuO_2 Particle Diameter, microns	Fuel Density, gm/cm^3	Enrichment, wt% Pu/Pu+U	Boron Concentration in Water for Null-Reactivity Condition, wppm Boron
0	8.474	2.106	1274
0	8.571	1.980	1225
0	8.704	2.060	1294
52	8.601	2.251	1352
107	8.638	2.237	1334
107	8.739	2.443	1380
107	7.630	2.476	1226
194.6	8.718	2.222	1277
194.6	8.657	2.260	1262
327.5	8.691	2.178	1179
327.5	8.798	2.287	1236

Fuel fabrication specifications for all the particle sized rods were to be $2.000 \pm 0.002\%$ by weight PuO_2 homogeneously mixed with natural uranium oxide. Unexplained differences between reactivity measurements made in the PCTR during 1967 with these particle sized fuel rods, in both graphite and water moderated lattices, indicated the possibility of an increased plutonium content in all rods except those containing zero micron PuO_2 particles. One rod containing zero micron PuO_2 particles and one rod containing 328-micron PuO_2 particles were dissolved and chemically analyzed.⁽²⁾ Extensive gamma scan measurements with a high resolution GeLi detector were made to determine the relative plutonium enrichment in each set of fuel rods.⁽³⁾ The relative enrichment results were normalized to the chemical analysis data to obtain the percent by weight of Pu in U+Pu for each set of rods listed in Table 2.8.

Experiment-theory correlation of the $k_\infty = 1$ compositions is in progress. Analysis of the activation measurements in both the poisoned and unpoisoned lattice will yield relative reaction rates in all central cell constituents. This information is necessary to determine the value of k_∞ for the unpoisoned lattice. Using calculated corrections to k_∞ for enrichment and fuel density differences between different sets of fuel rods, the PuO_2 particle size reactivity effect will be determined for a 1.0-inch water lattice of UO_2 -2.0 wt% PuO_2 rods both for the case with neutron leakage (unpoisoned lattice) and the case without neutron leakage (poisoned lattice).

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GRANIT: A Code for Calculating Position Dependent Thermal
Neutron Spectra in Doubly Heterogeneous Systems by the
Integral Transport Method

Computer Code Synopsis

C. L. Bennett

1. Name: GRANIT
2. Computer: The GRANIT Code was programmed to operate on the UNIVAC 1108 computer.
3. Problem Solved: The code computes the position dependent thermal neutron spectrum in doubly heterogeneous systems over the energy range 0 to 0.683 eV by the integral transport method, using 30 speed points and a maximum of 30 space points. Dilute granular composite (particulate structured) material regions are allowed. Both diluent and particle spectra are computed as a function of the radial position of the granular material in the cylindrical system. The computed spectra is used to determine various one-thermal-group cross sections and reaction rates in the system for as many as five independent thermal energy ranges.
4. Method of Solution: The scalar thermal neutron spectrum is computed as a function of position in the system by solving the integral transport equation with isotropic scattering. The code uses a combination of Gauss iteration, overrelaxation, and extrapolation to accelerate convergence. Granular regions are handled by expanding the normal region-to-region collision probabilities, using an extension of the approximate method of Lane, Nordheim and Sampson, to explicitly include particle and diluent interactions. Speed dependent macroscopic parameters are constructed separately for particle, diluent and homogeneous cell materials from input nuclide

concentrations and the microscopic parameters either available on the library tape or input directly. Point, region, and cell averaging, over the energy range of interest of both microscopic and macroscopic parameters, are performed using the computed spectrum as a weighting function.

5. Restrictions on the Complexity of the Problem: Number of speed points ≤ 30 . Number of particle types in a granular region ≤ 2 . Particle shapes - spheres or infinite cylinders. Number of space points ≤ 30 (each granular point counts as two space points for one particle mixture and three space points for two particle mixtures). A position dependent seven-group distribution function can be specified to describe particle size and/or shape variation for each of the two allowable particle compositions. Number of mixtures ≤ 10 . The system geometry must be cylindrical. Either a vacuum, reflecting, or an isotropic albedo outer boundary condition may be specified. Number of isotopes used in the cell ≤ 20 . Number of isotopes used in edit ≤ 20 . Further restrictions on the type of granular mixtures are dictated by the various assumptions of the particle size methods used. These are as follows:
 - (1) the particles must be blacker than the diluent;
 - (2) the particles must be relatively dilute, less than or equal to about 10% by volume;
 - (3) the particle must be small enough such that the flat flux and source approximation in the particle does not introduce appreciable errors.
6. Typical Running Time: Typical running times for 30 speed and space point problems are about 30 to 45 seconds per case on the UNIVAC 1108.

7. Unusual Features of the Program: The code explicitly computes position dependent diluent and particle spectra in granular regions, in addition to the position dependent spectra in homogeneous regions, for doubly heterogeneous systems by using the well known THERMOS method with a redistributed first flight collision matrix. The code is structured such that future improvements to the particle effect assumptions can easily be incorporated.
8. Related and Auxiliary Programs: The GRANIT code is a special use version of THERMOS. The auxiliary programs LIBP and RESUME, respectively, update or make a new library tape and produce a résumé of the library data.
9. Status: GRANIT is in production use on the UNIVAC 1108 computer at Pacific Northwest Laboratory, Richland, Washington.
10. References:
 - C. L. Bennett. GRANIT: A Code for Calculating Position Dependent Thermal Neutron Spectra in Doubly Heterogeneous Systems by the Integral Transport Method, Battelle-Northwest, Richland, Washington, 1970. (To be Published).
 - R. K. Lane, L. W. Nordheim, and J. B. Sampson. "Resonance Absorption in Materials with Grain Structure," Nucl. Sci. and Eng., vol. 14, p. 390. 1962.
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 - C. L. Bennett. "Multiple Region Interaction Probabilities for Particle Size Theory," Reactor Physics Quarterly Report, July, August, September, 1968, BNWL-921. Battelle-Northwest, Richland, Washington December 1968.

- - H. C. Honeck. THERMOS, A Thermalization Transport Theory Code for Reactor Lattice Calculations, BNL-5826. Brookhaven National Laboratory, Upton, New York, September 1961.
- 11. Machine Requirements: 64K memory, normal input, output, and program units, 1 unit for library.
- 12. Programming Language Used: FORTRAN-IV
- 13. Operating System: UNIVAC-1108 computer with FORTRAN-V compiler and CSCX operating systems.
- 14. User Information: The code may be obtained from Pacific Northwest Laboratory in Richland, Washington.
- 15. Material Available: Magnetic Tape Transmittal
 - GRANIT Source deck (2180 cards)
 - LIBP Source deck (504 cards)
 - RESUME Source deck (64 cards)
 - Sample Problem
 - Library Data Tape
- 16. Acknowledgment: This code is based on work performed under U.S. Atomic Energy Commission Contract AT(45-1)-1830.

3.0 HIGH TEMPERATURE REACTOR PHYSICS PROGRAM

MEASUREMENT OF k_{∞} FOR A ThO_2 - $^{235}\text{U}_2$ HTGR LATTICE AS A FUNCTION OF TEMPERATURE

T. J. Oakes

The analysis of the first High Temperature Gas-Cooled Reactor (HTGR) experiment performed in the High Temperature Lattice Test Reactor (HTLTR) has been completed and a report of the work is in final form. The primary results obtained from the experiment was the variation of k'_{∞} , as a function of the temperature of the test lattice, over the range from ~ 20 °C to 1000 °C. Further refinements to the corrections made to the experimental measurements have resulted in small changes in the measured values reported previously.⁽¹⁾ The revised values are listed in Table 3.1.

TABLE 3.1. $k'_{\infty}(T)$ for a ThO_2 - $^{235}\text{UC}_2$ -C Lattice

<u>Temperature, °C</u>	<u>k'_{∞} (Measured)</u>	<u>k'_{∞} (Calculated)</u>
20	1.102 $\pm$ 0.011	1.0929
150	1.088 $\pm$ 0.013	1.0780
300	1.074 $\pm$ 0.013	1.0646
550	1.047 $\pm$ 0.014	1.0484
700	1.033 $\pm$ 0.015	1.0417
1000	1.028 $\pm$ 0.013	1.0285

The final report of this work, together with a companion report which describe the HTLTR and derives the relationships required to determine k'_{∞} from the experiments, is being prepared for journal publication.

Reference

1. T. J. Oakes and C. R. Richey. Reactor Physics Quarterly Report, April, May, June, 1970, BNWL-1381-2, p. 255.
Battelle-Northwest, August 1970.

MEASUREMENT OF $k_{\infty}(T)$ FOR A ThO_2 - $^{233}\text{UO}_2$ HTGR LATTICE IN HTLTR

E. P. Lippincott

The analysis of the second HTGR experiment⁽¹⁾ performed in the High Temperature Lattice Test Reactor (HTLTR) has been completed and the report is in draft form. The primary result from the experiment was the variation of k for the ^{233}U -enriched lattice as a function of temperature, over the range from ~ 20 to 1000°C .

The experimental results are compared with calculations in Table 3.2.

TABLE 3.2. Comparison of Calculation and Experiment

Temperature, $^\circ\text{C}$	Calculated, k_{∞}	Measured, k_{∞}
20	1.045	1.055 ± 0.001
150	1.035	1.045 ± 0.002
300	1.023	1.035 ± 0.001
500	1.013	1.029 ± 0.001
750	1.006	1.024 ± 0.001
1000	1.001	1.023 ± 0.001

There is a significant difference between the calculated and measured values of k_{∞} both in absolute value and in the variation with temperature. Further study is needed to establish the reasons for these differences.

Reference

1. E. P. Lippincott. Reactor Physics Quarterly Report, January, February, March, 1970, BNWL-1381-1, p. 3.6.
Battelle-Northwest, May 1970.

ThO₂-PuO₂ HTGR EXPERIMENT IN HTLTR

R. I. Smith

Efforts began in the third quarter of FY-1970 to obtain suitable fuel for an HTLTR experiment using ThO₂ and PuO₂ in an HTGR lattice. Previous studies by Gulf General Atomic have established the practicality of such an HTGR enriched with plutonium rather than ²³³U and a definitive experiment was needed to provide data with which to check the computational methods used in the earlier analysis. Four kilograms of high exposure plutonium were obtained from the Critical Mass Laboratory at Hanford. The isotopic composition of the material is listed in Table 3.3.

TABLE 3.3. Isotopic Composition of High Exposure Plutonium

<u>Pu Isotope</u>	<u>wt%</u>
238	0.070 ± 0.003
239	72.4 ± 0.1
240	23.1 ± 0.1
241	3.53 ± 0.03
242	0.962 ± 0.009

The plutonium contained a significant amount of ²⁴¹Am, a daughter of ²⁴¹Pu. Since the gamma-ray activity from the ²⁴¹Am presented a personnel exposure problem to ORNL in the planned microsphere fabrication, the plutonium nitrate

solution was purified by ion-exchange to remove the ^{241}Am and any other impurities. A reduction of ^{241}Am from ~ 0.66 wt% in plutonium to ~ 0.028 wt% was achieved.

The purified solution was reconcentrated to ~ 223 grams/liter and shipped to ORNL. The particle specifications were for PuO_2 spheres, $>96\%$ theoretical density, with a size distribution between 149 and 250 microns, and with 80% of the particles >177 microns in diameter before coating. A pyrocarbon coating of ~ 100 microns thickness has been specified.

Fabrication of the microspheres is in progress at ORNL, with the process yield of satisfactory particles averaging about 80%.

MSBR EXPERIMENTS IN HTLTR

E. C. Davis

A series of reactor lattice physics experiments are to be conducted in the High Temperature Lattice Test Reactor (HTLTR) in support of the Molten Salt Breeder Reactor Program (MSBR). The details of the experimental program are being worked out jointly between BNW and ORNL personnel.

The MSBR experiments will be similar to those currently being performed in support of the High Temperature Gas-Cooled Reactor Program (HTGR). The MSBR lattice will differ from the HTGR lattices by having larger fuel channels and a heavier fuel loading, as shown in Table 3.4.

TABLE 3.4. Comparison of MSBR and HTGR Lattices

Reactor Type	Fuel Description				
	Channels (blocks)	Channel Diam., in.	Volume Fraction	C/ ^{233}U	C/Th
MSBR #1	4	0.781	0.136	$\sim 9,700$	~ 150
HTGR #2	25	0.470	0.296	$\sim 11,000$	~ 200

Fabrication of the fuel blocks is about 70% completed and specification of the fuel blend (ThO_2 -microspheres-graphite) has been completed. Loading of the fuel into the graphite blocks will begin in a few weeks.

REACTIVITY MEASUREMENTS AND THE INHOUR EQUATION

E. P. Lippincott

The experiments in multizone critical assemblies (such as the HTLTR and PCTR) require a determination of neutron life-time for interpretation of reactivity measurements for the composite loading. An experimental method has been devised for this determination which uses a series of constant reactivity differences obtained from various pairs of rising reactor periods. In the method the reactor is treated as a large bare thermal reactor. In this case we write⁽¹⁾

$$D\nabla^2\phi - \Sigma_a\phi + \frac{(1-\beta)k_\infty\Sigma_a\phi}{1 + \tau B^2} + \frac{p\Sigma\lambda_i C_i}{1 + \tau B^2} = \frac{1}{v} \frac{\partial\phi}{\partial t} \quad (1)$$

where the parameters in this equation are given by suitable averages over the reactor. The time dependence of the concentrations of delayed neutron precursors, C_i , is given by:

$$\frac{\partial C_i}{\partial t} = -\lambda_i C_i + \frac{k_\infty}{p} \beta_i \Sigma_a \phi \quad (2)$$

where λ_i is the decay constant and the delayed neutron fraction is:

$$\beta = \sum_i \beta_i \quad (3)$$

Assuming the reactor is on its stable period, $1/\omega$

$$\phi = \phi_o e^{\omega t} \quad (4a)$$

$$C_i = C_{io} e^{\omega t} \quad (4b)$$

Then from Equation (2),

$$C_{io} = \frac{k_{\infty} \beta_i \Sigma_a \phi_o}{p(\omega + \lambda_i)} \quad (5)$$

Substituting in Equation (1)

$$D\nabla^2 \phi_o - \Sigma_a \phi_o + \frac{(1-\beta) k_{\infty} \Sigma_a \phi_o}{1 + \tau B^2} + \frac{k_{\infty} \Sigma_a \phi_o}{(1 + \tau B^2)} \sum_i \frac{\lambda_i \beta_i}{\omega + \lambda_i} = \frac{\omega}{v} \phi_o \quad (6)$$

$$vD\nabla^2 \phi_o - v\Sigma_a \phi_o + \frac{vk_{\infty} \Sigma_a \phi_o}{1 + \tau B^2} = \omega \phi_o + \frac{vk_{\infty} \Sigma_a \phi_o}{1 + \tau B^2} \sum_i \frac{\omega \beta_i}{\omega + \lambda_i} \quad (7)$$

or calling the operator on the left, M,

$$M\phi = \lambda \phi \quad (8)$$

$$\text{where } \lambda = \omega + \frac{vk_{\infty} \Sigma_a}{1 + \tau B^2} \sum_i \frac{\omega \beta_i}{\omega + \lambda_i} \quad (9)$$

$$= \omega + \frac{k}{\ell} \sum_i \frac{\omega \beta_i}{\omega + \lambda_i} \quad (10)$$

$$\text{where } k = \frac{k_{\infty}}{(1 + \tau B^2)(1 + L^2 B^2)} \quad (11)$$

and the neutron lifetime is given by,

$$\ell = \frac{1}{\Sigma_a v(1 + L^2 B^2)} \quad (12)$$

Thus we have

$$M\phi = \frac{k\omega}{\ell} \left(\frac{\ell}{K} + \sum \frac{\beta_i}{\omega + \lambda_i} \right) \quad (13)$$

$$= \frac{k}{\ell} \rho \phi \quad (14)$$

where ρ is given by the inhour equation

$$\rho = \frac{\ell\omega}{\ell\omega + 1} + \frac{1}{\ell\omega + 1} \sum_{i=1}^m \frac{\omega\beta_i}{\omega + \lambda_i}. \quad (15)$$

In the HTLTR, k is very close to 1, and so one can take for a small perturbation,

$$\delta M\phi = \left(\frac{k}{\ell} \right) \Delta\rho\phi \quad (16)$$

which leads to

$$\frac{\Delta\rho^{\text{cell}}}{\Delta\rho^{\text{Cu}}} = \frac{\int_{\phi^+} (\delta M)^{\text{cell}}_{\phi} dV}{\int_{\phi^+} (\delta M)^{\text{Cu}}_{\phi} dV} \quad (17)$$

to a very good approximation. In calculating ρ by Equation (15), ℓ is not known. However, if one assumes $\ell\omega \ll 1$, then

$$\Delta\rho = \rho_2 - \rho_1 = \ell(\omega_2 - \omega_1) + \sum_{i=1}^m \frac{\omega_2\beta_i}{\omega_2 + \lambda_i} - \frac{\omega_1\beta_i}{\omega_1 + \lambda_i} \quad (18)$$

which for measured ω , is of the form

$$\Delta\rho = \ell\Delta\omega + y,$$

that is, $\Delta\rho$ is a linear function of ℓ in this range.

From measured pairs of ω , a family of straight lines is obtained on a plot of $\Delta\rho$ versus ℓ which, if the above equations and the data were exact, would intersect at a point determining $\Delta\rho$ and ℓ . An example of such a plot is shown in Figure 3.1.

This plot was obtained as follows:

1. Set one control rod at a given position, e.g., 3 inches open. Adjust the remaining control rods to give a period of approximately 20 seconds.
2. Close the one control rod a given increment, e.g., 1 inch. Return the other control rods to their setting as previously and measure the resultant period.
3. Close a second control rod so that when the first control rod is replaced at 3 inches, a period of about 25 seconds is obtained. Then measure the period with the rod at 2 inches to obtain a second pair of ω s with the same $\Delta\rho$ as the first pair of ω s.
4. Repeat the above procedure with successively longer periods, obtaining 4 or 5 pairs of ω s at constant $\Delta\rho$.
5. Repeat one or two of the earlier measurements to check for drift and consistency. Since reactivity measurements that require only control rod movement can be done in rapid succession, such drifts are small and the data are usually very consistent.

To obtain the value of ℓ by a least squares method, rewrite Equation (18) to treat $\Delta\rho$ and ℓ as constants. Then the form $y = \Delta\rho + \ell X$ is obtained,

where $X = \omega_2 - \omega_1$

$$\text{and } y = \sum_{i=1}^m \left\{ \frac{\omega_2^{\beta_i}}{\omega_2 + \lambda_i} - \frac{\omega_1^{\beta_i}}{\omega_1 + \lambda_i} \right\}$$

FIGURE 3.1. Change of Reactivity Versus Neutron Lifetime

If n pairs of ω s are then measured with a constant $\Delta\rho$, the least squares expression for $\Delta\rho$ and ℓ are,

$$\sum_{i=1}^n y_i = n\Delta\rho + \sum_{i=1}^n X_i \ell$$

$$\sum_{i=1}^n X_i y_i = \sum_{i=1}^n X_i \Delta\rho + \sum_{i=1}^n X_i^2 \ell$$

These equations may then be solved for the least square value for $\Delta\rho$ and ℓ . The value of ℓ obtained from the data in Figure 3.1 is 2.7 msec. It can be seen that a large error in ℓ is obtained by this method but the error in a ratio of $\Delta\rho$ as in Equation (17) is negligible compared to the experimental error. For example, comparing a slow period with a fast period measurement from Figure 3.1, at $\ell = 2$ msec,

$$\frac{\Delta\rho_1}{\Delta\rho_2} = \frac{5.047}{5.010} = 1.007$$

and thus the error for this extreme case is $\pm 0.7\%$ compared to an experimental error of about the same magnitude.

By carrying out the measurement of ℓ in this manner, the true ℓ may not be obtained, but rather the value of ℓ which gives the best fit to the inhour equation. This value of ℓ , therefore, gives the most consistent set of reactivity values over the range used and therefore, to some extent, compensates for errors in the inhour equation arising from the approximations made in deriving it.

Reference

1. *S. Glasstone and M. C. Edlund. The Elements of Nuclear Reactor Theory. Van Nostrand, 1952.*

EVALUATING POTENTIAL DEFORMATION IN THE VERTICAL SAFETY RODS AT THE HIGH TEMPERATURE LATTICE TEST REACTOR

R. G. Clark

Several poison plates in each of the four vertical safety rods (VSR) became bowed during the last HTLTR experiment which was conducted at temperatures up to 1000 °C. This plastic deformation was obvious when the blades were removed for inspection. The modifications proposed to correct the condition were tested and evaluated by cycling a prototypical section of a VSR blade in and out of a simulated reactor operating at 1000 °C.

Some deformation, it was learned, can be expected after extruded cycling, even from an acceptable redesign of this type of VSR. An in-place surveillance method has been adopted to inform the operator when the end of the useful service life of the VSR blade is approaching.

The VSR housings have been modified; two windows were installed on each to view, in place, the poison plates in the upper half of each VSR blade; and a technique was demonstrated to measure, in place, any deformation that develops on a poison plate. The technique has been adopted for routine use and the equipment has been installed on the top of the reactor.

In this method, a standard 3 × 4 slide projector casts a horizontal grid pattern onto a VSR poison plate that has been positioned at a window. A camera, mounted at an angle of 45 degrees to the projection plane, records the grid pattern on film. Any horizontal line will be viewed as a straight line

as long as the surface it is projected upon remains flat. As curvature develops in the poison plate, the pattern will deviate from a grid of straight lines to one of curved lines as viewed by the camera.

Negatives of polaroid film taken of each poison plate will be viewed on a large optical comparator which gives a magnification of 50 and will be viewed on a 30-inch screen.

Developing deformation of as little as 10 miles has been easily detected during the calibration of the system, using a plate with a wedge shape that was machined to known elevations (deformation).

A procedure has been established to inspect routinely and visually the available plates on each rod which should detect any abnormal behavior of the poison plates during the experimental program at high temperatures. In addition, two poison plates on each VSR which are representative, or typical, will be filmed routinely and the negatives examined on the optical comparator.

Deformation of the poison plates in the VSRs can be measured if it occurs and the appropriate action taken before an unsafe condition can develop.

DETERMINING FEASIBILITY OF USING THE FAST NEUTRON CAVITY IN THE PCTR FOR CROSS SECTION EVALUATION

W. N. McElroy* and D. F. Newman

A project to determine the feasibility of using the Fast Neutron Cavity (FNC) in the Physical Constants Test Reactor (PCTR) for cross-section evaluation has been completed. The results indicate that the FNC can be used successfully for this purpose.

* *Now employed by WADCO Corporation, a Subsidiary of Westinghouse Electric Corporation*

The timely evaluation and definition of energy-dependent reaction cross section $\sigma(E)$ in the energy region between $\sim 10^{-5}$ and 10 MeV is important for both reactor physics and engineering use in the LMFBR program. Previous studies have demonstrated the feasibility of using a multiple foil activation iterative method for $\sigma(E)$ definition and evaluation.⁽¹⁻⁴⁾ The method involves irradiation of a selected set of detectors (usually 10 to 15) in different reference spectra, measurement of resultant reaction rates, and determination of the flux spectra by iterative solution for the different reference neutron environments. These reference spectra, and corresponding measured reaction rates for a single reaction, are used in an inverted iterative process to define and evaluate an energy-dependent cross section.⁽¹⁻³⁾

The accuracy of the method can be improved by the simultaneous irradiation of a single type of foil (for a specified reaction) in an appropriate set of reference spectra.⁽¹⁾ This analytical study establishes the feasibility of using the PCTR-FNC facility with a multiregion central core of beryllium and Al-30 wt% U (93% ^{235}U) to produce a suitable set of reference spectra. Results of some multiple foil/proton-recoil neutron flux-spectral measurements in the FNC central cavity have been previously reported.⁽⁵⁾

The calculated spectra for several positions in the central region and one position in the PCTR graphite reflector region are shown in Figure 3.2. Nine such spectra from these regions were selected as the set of reference spectra for this study.

Because of its importance to the LMFBR program, the $^{238}\text{U}(n,\gamma)^{239}\text{U}$ reaction was selected as a typical reaction which could be re-evaluated by this method in the PCTR/FNC. The existing SAND-II code⁽⁶⁾ cross section⁽²⁾ for this reaction

Neg 704325-1

FIGURE 3.2. Neutron Spectra in the PCTR/FNC Cross Section Evaluation Assembly

was used with the nine reference spectra to calculate a set of reaction rates which might have been measured during a simultaneous irradiation of identical ^{238}U foils in each location of the PCTR/FNC. These nine reaction rates and the corresponding nine reference spectra were then input to the SAND-II code with a number of different initial (and extreme) estimates of $\sigma(E)$ for the ^{238}U reaction--considered to be unknown. For each input form $\sigma^{(0)}(E)$, the SAND-II iterative procedure generated a solution $\sigma^{(K)}(E)$ where "K" is the number of iterations required to achieve the solution.

The $^{238}\text{U}(n,\gamma)^{239}\text{U}$ results are presented in Figure 3.3. The small inset figure shows the three extreme input forms used. The dashed curve represents an initial guess $\sigma^{(0)}(E)$ that has a $1/E^{1/2}$ shape (corresponding to a $1/v$ cross section). In Figure 3.3, the solution for this input is also represented by the dashed curve. Similar results are given for a constant and $E^{1/2}$ input form. The solid curve is the existing SAND-II cross section and is therefore the correct solution. As expected, it is approximated best by the solution based on the $1/v$ input form.

Although there are differences among the three solutions shown in Figure 3.3, the overall agreement is excellent considering the extreme differences in input forms. This implies that the nine reference spectra are sufficiently different to establish a rather unique solution over approximately six decades of energy by one irradiation. Thus, if the best available $\sigma(E)$ for the $^{238}\text{U}(n,\gamma)$ reaction (based on monoenergetic measurements and theoretical estimates) were used for $\sigma^{(0)}(E)$ together with reaction rates from a PCTR/FNC irradiation, the resulting $\sigma^{(K)}(E)$ solution could be expected to produce excellent integral consistency over the energy interval of $\sim 10^{-5}$ to ~ 3 MeV.

Neg 704152-1

FIGURE 3.3. SAND-II Solution Uniqueness Study for the
 $^{238}\text{U}(n, \gamma)^{239}\text{U}$ Reaction

Energy dependent cross sections, usually based on monoenergetic measurements and theoretical estimates, are generally inconsistent with activation measurements which are inherently of an integral nature. Energy dependent cross sections determined as described here do not suffer from this inconsistency.

Below $\sim 4 \times 10^{-5}$ and above ~ 3 MeV the derived cross sections must be disregarded since the nine reference spectra do not have adequate reaction sensitivity in these regions. Irradiations in additional spectra would be required to provide reliable results for these regions.

The results of this study also show that if a previously undefined cross section were needed, such as for a tag gas used in fuel element leak detection, a smoothed energy-dependent cross section could be easily defined for immediate engineering application. That is, the derived cross section could be expected to give reliable estimates of reaction rates for engineering applications for most LMFBR type spectra. The PCTR/FNC facility has been shown to be suitable for cross section evaluations of this type.

References

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5. D. F. Newman and L. C. Davenport, "Neutron Flux Spectrum Measurements in the PCTR Fast-Neutron Cavity," Trans. Am. Nucl. Soc., Vol. 12, 714 (1969).
6. W. N. McElroy, et. al., "A Computer-automated Iterative Method for Neutron Flux Spectra Determination by Foil Activation," Vol. 1, AFWL-TR-67-41, (1967).

4.0 PHOENIX STUDIES

No contribution this reporting period

5.0 STEADY STATE AND TRANSIENT SUBCHANNEL CODE

DEVELOPMENT AND DATA ANALYSIS

D. S. Rowe and B. M. Johnson

The objective of this program 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 program includes both analytical studies for the continued development of computer programs and experimental studies for the verification and implementation of these computer programs.

COBRA-II COMPUTER PROGRAM

The objective of this portion of the program is to develop a mathematical model and computer program for predicting the steady-state and transient performance of rod bundle nuclear fuel elements. The approach being used is similar to that of the COBRA-II computer program. The bundle flow cross section is divided into flow subchannels that are assumed to contain one-dimensional flow and are coupled to each other by turbulent and diversion crossflow mixing. Both types of crossflow mixing carry mass, energy and momentum between the subchannels. Boiling and nonboiling conditions are considered by using an equation of state that gives density as a function of enthalpy, flow, heat flux, pressure, position, and time. The model neglects sonic velocity propagation; therefore, it is limited to transients with time duration greater than the time for a sonic wave to pass through the channel. A simplified transverse momentum equation that neglects temporal and spatial acceleration is used to simplify the numerical solution. This gives instantaneous response of the diversion crossflow to changes in subchannel pressure gradient. The equations of the mathematical model are solved by using a semiexplicit finite difference scheme that is stable for all time steps. This scheme

also gives a boundary value flow solution for both steady state and transients where the boundary conditions are the inlet enthalpy, inlet flow rate and exit pressure. Several successful computer solutions have been run using the above mathematical model and numerical solution. This has included rapid changes in heat flux, inlet flow rate, inlet enthalpy and system pressure. The ability of the boundary value solution to include high crossflow resistance has also been successfully demonstrated. High crossflow resistance has the effect of causing a downstream flow disturbance to be felt upstream. This effect cannot be considered by COBRA-II and other similar computer programs using an initial value flow solution.

The features of COBRA-III can be summarized as follows:

- It contains all the analysis capability of the COBRA-II program.
- It can consider transients up to, but not including, sonic velocity propagation effects.
- The numerical scheme performs a boundary value solution to more properly include the effects of crossflow resistance.
- The numerical solution is stable for all time steps.

Development of COBRA-III is continuing to improve its capability. A fuel heat transfer model is being developed to account for thermal response of the fuel. The ability to consider flow reversals is being developed together with a pressure drop boundary condition to eliminate the present need for a prior knowledge of the inlet flow.

EXPERIMENTAL STUDY OF FLOW STRUCTURE IN ROD BUNDLES

The objective of this experimental study is to obtain a more complete description of the turbulent flow structure of

single-phase flow in rod bundles. This study is needed to obtain a better understanding of intersubchannel mixing so that better mixing correlations can be developed for use in subchannel computer programs. Present mixing correlations do not adequately account for the effect of subchannel geometry.

This experimental study will investigate the turbulent flow structure in rod bundle geometry to define the effect of subchannel shape, gap spacing and subchannel interconnections. The transverse fluctuating velocity in the gap between subchannels will be the primary measurement in these experiments. These measurements will be made with a two-component laser-doppler velocimeter. 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. This measurement technique will enable measurements of velocities that have never been obtained in rod bundle geometries.

Design of the experimental apparatus is in progress. A common flow channel will be used to accommodate a variety of subchannel sizes and shapes. This will be accomplished by using a channel that consists of two common side plates containing glass windows for entrance and exit of the laser beam. Inserts of various thicknesses will be placed between the side plates to create rectangular flow channels. Subchannels will be formed within this channel by inserting rods, or sectors of rods, to form the desired subchannel shapes. Glass rods will be used where appropriate to enable passage of the laser beam. A flow straightening and calming section will be included to control the level of upstream turbulence.

6. PLUTONIUM CRITICALITY STUDIES

CRITICAL EXPERIMENTS WITH UNMODERATED PuO₂

S. R. Bierman and E. D. Clayton

Part of the research effort at the Battelle-Northwest-operated Critical Mass Laboratory is concerned with determining the criticality parameters of plutonium mixtures over the entire range of neutron moderation. The cumulative results of this effort up through the early part of 1969, both experimentally and theoretically, were recently published in Nuclear Applications.⁽¹⁾ The object of this paper is to present data from a later series of critical experiments in the undermoderated region of the neutron spectrum. In this region the plutonium mixtures are highly concentrated, typifying wet powders, precipitates, slurries and polymers encountered in plutonium fuels fabrication and reprocessing. The experiments were carried out using our remote split-table machine⁽²⁾ and essentially dry, unmoderated PuO₂ (0.04 H/Pu atomic ratio) containing 18.35 wt% ²⁴⁰Pu in the plutonium. The PuO₂ was in the form of 2 × 2 × 1 1/2-inch blocks, each compressed to a density of 5.762 ± 0.039 g Pu/cm³ and encased, for contamination control, in a heat-shrinkable polyolefin plastic. A detailed description of the fuel is presented in Table 6.1.

Experimental data were obtained from both bare and reflected parallelepipeds of PuO₂. The critical sizes of each of these assemblies are presented in Table 6.2. The dimensions of each assembly have had experimentally determined corrections made to them 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 fuel at a density of 5.762 g Pu/cm³.

TABLE 6.1. Description of Fuel

Dimension of Compacts	
Without Cladding, L × W × H	$5.13 \times 5.13 \times 3.81 \pm 0.02$ cm
Average Dimension of Stacked Compacts With Cladding, L × W × H	
	$5.54 \times 5.54 \times 4.09 \pm 0.04$ cm
Average Thickness of Cladding, cm	
Sides	0.38
Top	0.13
Bottom	0.13
Composition of Cladding, Atoms per Barn-cm	
H	4.614×10^{-2}
C	2.906×10^{-2}
O	2.309×10^{-2}
Composition of Compacts, Atoms per Barn-cm	
^{238}Pu	3.383×10^{-5}
^{239}Pu	1.092×10^{-2}
^{240}Pu	2.654×10^{-3}
^{241}Pu	7.269×10^{-4}
^{242}Pu	1.632×10^{-4}
O	3.094×10^{-2}
H	5.511×10^{-4}
PuO_2 Particle Size, mm	
Maximum	0.0400
Mean	0.0074
Minimum	0.0025
Plutonium Density, g/cm ³	5.762 ± 0.039

As was observed in previous experiments⁽³⁾ with PuO_2 -polystyrene fuel having an H/Pu atomic ratio of 5, the critical mass of each of these assemblies shown in Table 6.2 was not affected by the stacking voids and cladding material. In these relatively fast systems, the fuel density reduction caused by stacking voids and cladding was offset by the increased neutron moderation caused by the cladding material.

TABLE 6.2. Experimental Data from PuO_2 Compacts 18.35 wt% ^{240}Pu Isotopic Content, 5.762 g Pu/cm^3 , 0.04 H/Pu Atomic Ratio, 50 °C

<u>REFLECTOR</u>	<u>CRITICAL DIMENSIONS, cm</u>			<u>CRITICAL MASS, KG OF Pu</u>
	<u>LENGTH</u>	<u>WIDTH</u>	<u>HEIGHT^(a)</u>	<u>18.35 WT% ^{240}Pu</u>
PLEXIGLAS	25.65	25.65	10.03±0.01	38.3±0.3
PLEXIGLAS	25.65	30.78	8.98±0.01	41.1±0.3
PLEXIGLAS	30.78	30.78	7.97±0.03	43.8±0.3
PLEXIGLAS	30.78	41.05	6.86±0.08	50.3±0.7
PLEXIGLAS	41.05	41.05	5.95±---	58.2±0.3
BARE	30.78	30.78	20.90±0.09	113.8±0.9
BARE	30.78	30.78	21.12±0.06	115.1±0.8

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

The critical sizes shown in Table 6.2 also reflect corrections made for temperature changes during the course of the experiments. Because of the relatively high plutonium density and ^{240}Pu content, a high specific heat generation (about 20 W/liter or 3 1/2 W/kg) was obtained and resulted in

temperature variations of as much as 30 °C during the course of a single experiment. To obtain consistent sets of data, the count rates in each critical approach were corrected to 50 °C by a technique developed by R. C. Lane and others⁽⁴⁾ at Aldermaston, U.K., and shown below:

$$K = \frac{N}{T_1 - T_2} \left(\frac{1}{C_1} - \frac{1}{C_2} \right),$$

where N = number of fuel blocks,

T_1 = temperature corresponding to count rate C_1 ,

T_2 = temperature corresponding to count rate C_2 .

K = change in reciprocal count rate per degree
change in temperature for N fuel blocks.

Although there was an insufficient amount of fuel to achieve criticality unreflected, two bare critical assemblies were built using driver regions of PuO_2 -polystyrene having an H/Pu atomic ratio of 5. [Experiments with this 5 H/Pu fuel have been previously reported.⁽³⁾] Each of the bare assemblies had the same cross-sectional dimensions but different amounts of driver fuel. Based on these measurements, a bare assembly of 5.762 g Pu/cm^3 of PuO_2 , $30.78 \times 30.78 \times 20.99 \pm 0.22$ cm containing 114.6 ± 1.4 kg of plutonium would be critical. With our calculational techniques^(5,6) we have been able to calculate this bare critical assembly to within 6 mk in k_{eff} . Consequently, it was felt that the extrapolation distance for the bare assembly could be calculated with a high degree of confidence. By using a calculated extrapolation distance of 3.57 cm for the bare assembly and the bare critical dimensions given above, the critical buckling for this essentially unmoderated PuO_2 material was determined to be $262.0 \pm 1.7 \text{ m}^{-2}$.

The reflected critical assemblies ranged over a series of slabs from about 10 to 6 cm in thickness to permit determination of 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 linear least squares fit extrapolation of this data to an inverse cross-sectional area of zero yields a critical thickness of 3.34 ± 0.10 cm for a slab of this essentially unmoderated PuO_2 material infinite in two dimensions. A corresponding extrapolation distance of 8.20 ± 0.22 cm was obtained for the reflected infinite slab by equating the critical dimensions of each reflected assembly to the critical buckling of $262.0 \pm 1.7 \text{ m}^{-2}$ determined for this material from 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.

For establishing criticality limits and for checking nuclear constants and calculational techniques, these slab data need to be expressed in equivalent spherical, cylindrical, and cubic geometries. Consequently, the experimental data were converted at the critical buckling of $262.0 \pm 1.7 \text{ m}^{-2}$ 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 ± 0.2 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 their theoretical density corresponding to an 0.04 H/Pu atomic ratio. These were obtained by making density corrections to the $5.762 \text{ g Pu/cm}^3 \text{ PuO}_2$ data.

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FIGURE 6.1. Extrapolation Distances and Measured Critical Height

TABLE 6.3. Critical Dimensions in Spherical, Cylindrical, Cubic and Slab Geometries at H/Pu Atomic Ratio of 0.04

GEOMETRY	PuO ₂ -WATER 5.762g Pu/cm ³ 18.35 WT% ²⁴⁰ Pu(a)		²³⁹ Pu-WATER 19.16g Pu/cm ³ 0.0 WT% ²⁴⁰ Pu	²³⁹ PuO ₂ -WATER 10.92g Pu/cm ³ 0.0 WT% ²⁴⁰ Pu	Pu-WATER 19.16g Pu/cm ³ 18.35 WT% ²⁴⁰ Pu(a)	PuO ₂ -WATER 10.92g Pu/cm ³ 18.35 WT% ²⁴⁰ Pu(a)
	λ	χ ^(b)	χ ^(b)	χ ^(b)	χ ^(b)	χ ^(b)
	cm	cm	cm	cm	cm	cm
BARE ASSEMBLIES						
INFINITE SLAB	3.62±0.2	12.17±0.28	3.71±0.09	6.38±0.14	4.13±0.00	7.07±0.16
SPHERE	3.60±0.2	15.81±0.20	5.20±0.07	8.41±0.11	5.74±0.07	9.22±0.12
INFINITE CYLINDER	3.58±0.2	11.28±0.20	3.60±0.06	5.96±0.11	3.99±0.07	6.57±0.12
CUBE	3.57±0.2	26.48±0.28	-----	-----	-----	-----
REFLECTED ASSEMBLIES ^(c)						
INFINITE SLAB	8.20±0.22	3.34±0.10	0.82±0.02	1.60±0.04	1.15±0.03	2.31±0.06
SPHERE	8.17±0.38	11.24±0.38	4.22±0.14	6.57±0.22	4.70±0.16	7.39±0.25
INFINITE CYLINDER	8.17±0.38	6.69±0.31	2.38±0.11	3.86±0.17	2.72±0.13	4.48±0.21
CUBE	8.13±0.38	17.36±0.54	-----	-----	-----	-----

(a) ISOTOPIC CONCENTRATION

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

(c) 5.762 PuO₂-WATER REFLECTED WITH PLEXIGLAS, OTHER SYSTEMS REFLECTED WITH WATER

Table 6.3 also shows values which have been corrected to zero ^{240}Pu content to show the relatively small effect that ^{240}Pu has on the critical size of this highly concentrated system. In this high neutron energy system the percent change in spherical critical mass per percent change in ^{240}Pu content is only 2.07 for the reflected case and 1.88 for the bare case, as compared to values of about 10 and 12 for a more thermalized system. A calculated neutron energy spectrum for the PuO_2 fuel used in the experiments is shown in Table 6.4.

TABLE 6.4. Calculated 18-Group Flux for PuO_2

5.762 g Pu/cm^3 , 0.04 H/Pu Atomic Ratio
18.35 wt% ^{240}Pu Isotopic Content

<u>Group</u>	<u>Lower Energy</u>	<u>Relative Flux</u>
1	7.79 MeV	0.0036
2	6.07	0.0103
3	4.72	0.0247
4	3.68	0.0398
5	2.87	0.0621
6	1.74	0.1596
7	1.35	0.0847
8	183.00 keV	0.4789
9	24.80	0.1260
10	3.36	0.0094
11	454.00 eV	0.0007
12	101.00	0.0002
13	37.30	
14	13.70	
15	5.04	
16	1.86	
17	0.683	
18	0	

As with previously published data^(2,3) for 15 and 5 H/Pu material, these data for essentially unmoderated systems are compared in Figures 6.2, 6.3, 6.4, and 6.5 with data from TID-7028⁽⁷⁾ for slab, cylindrical and spherical geometries. In all three geometries the critical sizes derived from the experiments are slightly larger than those shown in TID-7028. Calculated values for the spherical critical mass and radii, obtained by using GAMTEC-II⁽⁵⁾ 18-group constants with the transport theory code DTF-IV,⁽⁷⁾ are shown also in Figure 6.5.

It was anticipated that these experiments with essentially unmoderated PuO_2 would either verify or disprove that a reflected cube could have a smaller critical mass than a reflected sphere. The reflected critical mass of a cube of this essentially unmoderated PuO_2 was observed to be less than that for a reflected sphere of the same material. However, as with previous observations with 5 H/Pu and 15 H/Pu,^(3,2) this phenomenon was not so pronounced that it could not be accounted for by the inaccuracies in the measurements.

The results from the experiments to date indicate that the values for the critical sizes and masses of plutonium given in such references as TID-7028, should be increased over the intermediate-to-fast neutron energy range. However, additional data is needed at H/Pu ratios around 30 and 55, for a better definition of the variation in critical mass with concentration and ^{240}Pu content. Experiments are currently planned which will provide this data.

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

Neg 704563-2

FIGURE 6.3. Data Derived from Critical Slabs of PuO_2 -Polystyrene

Neg 702359-3

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

Neg 702359-1

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

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7.0 CALCULATIONS OF POTENTIAL ENVIRONMENTAL RADIOLOGICAL
CONSEQUENCES OF REACTOR ACCIDENTS

A progress report is in preparation.

8. NUCLEAR GRAPHITE

G. L. Tingey

IN-REACTOR CREEP

W. J. Gray

The second set of two capsules containing samples under a compressive stress was placed in the ETR reactor in July 1970 and is scheduled to be discharged in February 1971 after an additional exposure of about 5.3×10^{21} n/cm² (E > 0.18 MeV). Length-change data from the first exposure⁽¹⁾ are shown in Table 8.1. These data clearly demonstrate the effect of a compressive stress on the contraction rate but meaningful creep constants cannot be calculated until the second set of data is available. One can only say, at this point, that the observed creep strain is roughly linear with stress and near the expected magnitude.

RADIOLYTIC REACTIONS IN GRAPHITE MODERATED REACTORS

G. L. Tingey and D. E. Clark

Corrosion reactions, and chemical changes in general, occur in nuclear reactors by radiolytic reactions as well as the more common thermally induced reactions. In high temperature gas-cooled reactors, reactions may occur between impurities in the gas coolant and the graphite moderator or the fuel elements. In addition, homogeneous reactions are present in the gas phase producing changes in the coolant composition.

Studies of radiolytic reactions are normally carried out at ambient temperatures because it is generally considered that radiation-induced reactions have zero temperature coefficient. This assumption is approximately true for most reactions within a limited temperature range but should not be considered generally true without experimental verification.

TABLE 8.1. Length Changes of Graphite Under Compressive Stress

<u>Graphite</u>	<u>Irradiation Temp, °C</u>	<u>Stress, psi</u>	<u>ΔL, %</u>
AGOT	820	0	-0.210
		1000	-0.615
		2000	-0.652
		3000	-0.966
	560	0	-0.373
		1000	-0.228
		2000	-0.744
		3000	-0.957
H337	820	0	+0.065
		1000	-0.102
		2000	-0.493
		3000	-0.568
	560	0	-0.159
		1000	-0.282
		2000	-0.500
		3000	-0.699
AXF-8QBG1	820	0	+0.246
		1000	-0.008
		2000	-0.026
		3000	-0.457
	560	0	-0.014
		1000	-0.153
		2000	-0.259
		3000	-0.644

Fluence $\approx 1.5 \times 10^{21}$ n/cm² (E > 0.18 MeV) for all samples.

Unstressed length changes are averages of 10 to 20 samples.

Stressed length changes are for one sample (AGOT) or two samples (H337 and AXF-8QBG1).

Previous studies have been conducted at Battelle-Northwest in which the temperature coefficient of the radiolytic reaction of carbon dioxide with water vapor was measured.⁽²⁾ These studies were conducted in the temperature range from 100 to 400 °C and led to the following expression for radiolytic yield:

$$G(\text{CO}) = G(\text{H}_2\text{O}) = 17.3 \exp^{-1,700/RT} \text{ molecules/100 eV.}$$

Since HTGR temperatures exceed 1000 °C, calculations of radiolytic reactions required an extrapolation from 400 to 1000 °C. Although higher temperature radiolytic studies were desirable, they were difficult because of interference with the thermal reactions.

During the previous quarter, we have measured the yield of the radiolytic reaction of carbon dioxide and hydrogen to temperatures as high as 650 °C. These studies have been accomplished by diluting the gas mixture with helium to concentrations more nearly typical of the HTGR Gas coolant. This dilution with inert gas reduces the rate of the thermal reaction to an immeasurable value but the radiolytic reactions have approximately the same rate as observed at higher concentrations because of transfer of energy from the helium to the reactive gases.

Figure 8.1 shows the 100 eV yield of CO as a function of temperature for a gas mixture with no helium present and for mixtures containing 1000 ppm, 433 ppm, or 60 ppm carbon dioxide and hydrogen in helium. The data shown in the figure yield the following expressions for the rates of product formation:

$$P_{\text{H}_2} = P_{\text{CO}_2} = 0.5 \text{ atm; } P_{\text{He}} = 0$$

$$G(\text{CO}) = 0.356T^{1/2} \exp^{-1100/RT} \text{ molecules/100 eV}$$

FIGURE 8.1. Effect of Temperature and Reactive Gas Concentration on Product Yield from the Radiolysis of a Mixture of CO_2 and H_2 in Helium

$$P_{H_2} = P_{CO_2} = 10^{-3} \text{ atm}; P_{He} = 1 \text{ atm}$$

$$G(CO) = 0.0732T^{1/2} \exp^{-200/RT} \text{ molecules/100 eV}$$

$$P_{H_2} = P_{CO_2} = 4.33 \times 10^{-4} \text{ atm}; P_{He} = 1 \text{ atm}$$

$$G(CO) = 0.056T^{1/2} \exp^{-200/RT} \text{ molecules/100 eV}$$

$$P_{H_2} = P_{CO_2} = 6.0 \times 10^{-5} \text{ atm}; P_{He} = 1 \text{ atm}$$

$$G(CO) = 0.0247T^{1/2} \exp^{-200/RT} \text{ molecules/100 eV}$$

The reader will note the radiolytic yield of products increases with temperature only slightly when the system is diluted with helium but when there is no helium present the temperature coefficient is much greater. Although we had not anticipated a change in the temperature coefficient when helium was present, it now appears reasonable to expect some difference because different activated states are involved. When no helium is present the reactive gases are excited directly by the radiation and thus a variety of ionic and excited species are formed. In a mixture containing 1000 ppm CO_2 and H_2 , 98.8% of the energy is absorbed by the helium. Since no stable products are formed with excited helium much of this energy is transferred to the reactive gases. Since the excitation mechanism is different, the excited states are likely to be different; thus, a different temperature coefficient for reaction is observed.

The more dilute gases have a lower overall yield because a greater portion of the excitation energy of the helium is dissipated by photon emission.

With this new information, we are now able to evaluate the effect of radiolysis in the HTGR coolant much more accurately. Since measurements have been made at much higher temperatures, the extrapolation can be made with more confidence. In addition, with a lower temperature coefficient for the reaction, the temperature effect is smaller and thus less important.

Further work is currently underway to measure the temperature coefficient of a dilute mixture of CO and H₂O vapor in helium. With this data a more accurate calculation of gas compositions in the HTGR coolant will be possible.

EFFECT OF SAMPLE SIZE ON DIMENSIONAL STABILITY

W. J. Gray

Samples previously irradiated at low temperature (cold seeded) were discharged from the ETR reactor in July 1970 after high-temperature irradiation to a peak fluence of about 1×10^{21} n/cm² (E > 0.18 MeV).⁽¹⁾ Although many of the cold-seeded samples contracted more than the unseeded ones, at least one more exposure period will be required to definitely establish whether the observed effect on contraction rates is real and to determine whether the observed effect continues during the irradiation or is merely a transient one. These samples are scheduled to be recharged into the reactor in November 1970 for an additional exposure of about 3×10^{21} n/cm².

In a separate experiment,⁽³⁾ relatively large samples (1.75 in. diameter $\times$ 2.25 in. long) were irradiated in the GETR and ETR reactors to a peak fluence of about 8.5×10^{21} n/cm². Some of these samples were irradiated under a compressive stress of 50 psi. Preliminary analysis of the data indicated that the contraction rates and the post-turnaround behavior

of the stressed samples is not significantly different from smaller samples previously irradiated in an unstressed condition in the GETR and ETR reactors. Therefore, this experiment will be discontinued. Following more detailed analysis of the data, a final report will be issued.

HIGH-TEMPERATURE IRRADIATIONS

W. J. Gray

Samples that were last discharged from the ETR reactor in November 1969⁽⁴⁾ were recharged together with some new samples in September 1970. The size of the irradiation capsule was increased from 2.0 in. OD to 2.5 in. OD. The new samples were of experimental graphites, supplied by M. C. Smith of the Los Alamos Scientific Laboratory, and a grade of POCO graphite that has a larger grain size than those previously irradiated.

ANALYSIS OF COOLANT-GRAPHITE REACTIONS

R. C. Giberson

The purpose of this task is to maintain and upgrade a computer program which has been developed to calculate graphite oxidation in high-temperature gas-cooled reactors. The code has found wide use in past years in predicting the results from both steady-state and simulated accident conditions in proposed and current HTGR's.

The most recent changes in the code have made it adaptable to the proposed 1100 MW_e HTGR. This simulation requires 10 "zones" of varying temperature and flow characteristics to adequately represent the reactor.

Additional data obtained on the effect of radiation on the CO₂ + H₂ reaction have been incorporated in the computer code. This information enables a more accurate assessment of the

effect of this reaction on the total system. The results show an increase in the H_2/H_2O due to the changes made. Similar data on the $CO + H_2O$ reaction are required to evaluate accurately the importance of radiolytic reactions.

MECHANISM OF IRRADIATION DAMAGE

W. C. Morgan and W. J. Gray

The objective of this study is to gain a better understanding of irradiation damage in polycrystalline graphite. Although considerable information is available concerning the gross effects of irradiation on a variety of graphites, more detailed knowledge of the fundamental mechanisms is required to develop graphites that are more suitable for specific reactor applications. Emphasis will be placed on obtaining information on the microstructure and changes in microstructure occurring during irradiation. This information is required to develop a refined model for predicting the behavior of polycrystalline graphites during irradiation.

Irrespective of arguments as to the relative importance of other structural parameters (such as graphitic and non-graphitic carbon phases, macro- and micro-crack systems, internal stresses, etc.), there is rather universal agreement that analysis of the thermal expansion behavior of artificial graphites will provide valuable insights into the structure and into the changes occurring during irradiation. The gross thermal expansion behavior can be easily explained on the basis of a single phase model;⁽⁵⁾ subtle differences in the coefficient of thermal expansion (CTE) could, however, be expected to result from effects of a nongraphitic phase and/or internal stresses. Thus, the lattice expansion needs to be determined rather precisely if the thermal expansion behavior is to be used to characterize different types of artificial graphites.

A theory of thermal expansion was derived by Riley⁽⁶⁾ and applied to the analysis of experimental data on lattice expansion of graphite; the resultant equations for the lattice CTE's are widely used for the analysis of the bulk CTE of artificial graphites. Riley's analysis, however, suffers from two defects:

- The data cover only the temperature range from 15 to 800 °C
- The analysis employs the Debye lattice vibration temperatures derived by Magnus⁽⁷⁾ in 1923.

Considerable data exist which suggest that the lattice CTE's derived by Riley may not be sufficiently precise to be used as the basis for analysis of bulk CTE when one is looking for these subtle differences. It should be noted that various evaluations of specific heat data and evaluations of thermal expansion data are available in the open literature. Examination of these, however, shows large areas of disagreement, especially in regard to the Debye temperatures derived from specific heat data.

One of the immediate goals of this study is, therefore, to refine the equations for the lattice CTE's. To accomplish this goal, we are proceeding with the following schedule:

- (1) Collection and evaluation of available specific heat data over the temperature range 100 to 2000 °K.
- (2) Analysis of specific heat data to obtain improved estimates of the Debye lattice vibration temperatures.
- (3) Collection and evaluation of graphite lattice spacings and thermal expansion data over the same temperature range.
- (4) Derivation of lattice thermal expansion coefficients using the above data and the theoretical method of Riley.

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9. CORROSION PRODUCT TRANSPORT STUDY

T. F. Demmitt

RDT terminated support of this program during September 1970. AECL personnel from Chalk River contacted BNW immediately upon receipt of the program termination notice to determine if an alternate agreement could be reached to allow continuation of the work under their sponsorship. The USAEC was advised of this interest. Discussions are still in progress on this subject.

CRUD GENERATOR STUDIES

An in-line, electrically activated crud generator device was designed, fabricated and tested for about 10 days in a high-temperature loop system. This experiment was designed to determine whether the corrosion product release rate from mild steel surfaces could be increased significantly by adjusting the electrical potential into a more active corrosion region. The experiment had to be terminated prematurely due to the program termination. The results obtained during the experiment were not entirely conclusive; however, there was no indication during the experiment that the crud release rate was increased significantly by variations in potential. The samples should be examined to assess the effect of potential on the corrosion that occurred. The efforts to evaluate the crud generator capabilities should also be continued. This work will not be performed unless an alternate funding source (e.g., AECL) is arranged.

IRON ANALYSIS PROCEDURE

A recently published colorimetric procedure for measurement of trace concentrations (0 to 5 ppb) of iron in water was adapted for use in analyzing the effluent samples from the crud

generator. This procedure employs Ferrozine as the indicator. The procedure appears adequate for measurement of iron concentrations in the 1 to 2 ppb range without preconcentration of the samples. The procedure was used to assess performance of the crud generator.

SYSTEM DESIGN

An engineering design analysis was performed to assess the facility modifications necessary to perform the scheduled experiments. The results of this analysis were discussed with AECL personnel (Charlesworth, Hollies, and Minns) when they visited BNW during July 1970. Detailed design work was then undertaken and was about 20% completed when the program termination notice was received. No further work will be performed on this project unless an alternate source of funding is arranged.

10. ATR PRESSURIZED WATER LOOP OPERATION AND MAINTENANCE

R. S. Hope

The Advanced Test Reactor began Cycle 3B shutdown on June 28, 1970. Startup for 3B began on July 1, 1970 and continued until Cycle 4 shutdown on July 18, 1970. Accumulated exposures for Cycle 3B were 3182.9 MWd or 14.47 effective days at 220 MW. Cycle 4A startup began on July 30, 1970 and continued until August 17, 1970. Accumulated exposures were 3348 MWd or 15.22 effective days at 220 MW. Cycle 4B startup began August 21, 1970 and continued until September 7, 1970. Accumulated exposures were 3356.5 MWd or 14.34 effective days at 234 MW. Cycle 5 startup began September 13, 1970 and is scheduled to operate until October 18, 1970.

The 1D-N loop has been operating at 2000 psig, 520 °F, 45 gpm, pH 10.00 $\pm$ 0.2 with O₂ concentration at less than 0.05 ppm. The loop test train contained stainless steel, Zircaloy-2, and molybdenum test specimens during Cycle 3. Toward the end of Cycle 3B, samples of loop water showed high radioactivity. Water was filtered and the filter was counted in a pulse height analyzer. The spectrum showed excessive ⁹⁹Mo along with some ⁵⁴Mg and a grade of ⁶⁰Co. It was decided that molybdenum samples supplied by the Naval Research Laboratories were corroding, thus giving the high activity. During Cycle 4 shutdown, the sample holder containing the molybdenum samples was transferred to the TRA hot cell where the samples were removed. The holder with its remaining stainless steel samples was then transferred back to the ATR where it was reinserted into the 1D-N test train for further irradiation. Zircaloy-2 corrosion coupons were also removed from the test train.

The 1D-N loop was filled, flushed, and drained five times prior to discharge of the test train. It was filled, flushed, and drained seven more times after the test train was removed before radiation levels were back to normal. The ion exchange resin was also changed out in the M-16 purification column.

The 1D-N pressurized water loop control panel is located on the ATR first basement. Two loop control panels for loops 2E and 2D are located on one side of the second basement while three loop control panels are located on the other side of the second basement. This arrangement required one reactor engineer to be at the 1D-N control panel, one engineer to be at the two control panels, and one engineer to be at the three control panels. This resulted in one man's full time being charged to one loop while the other men divided their time charges two and three ways. It was proposed that satellite instruments be installed on the second basement to record the pressure, flow and temperatures of the 1D-N loop and remove the requirement of one man full time at the first basement control panel. This proposal was accomplished by July 1, 1970 and now the reactor engineer for the 1D-N loop is only required to record data every 2 hours and make adjustments as necessary. This requires about half of one man's time.

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