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MASTER

RESULTS FROM USAEC PLUTONIUM UTILIZATION PROGRAM
CONDUCTED BY BATTELLE-NORTHWEST

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1.0 INTRODUCTION

This paper presents significant results obtained from the USAEC Plutonium Utilization Program since the last IAEA Panel on Plutonium Utilization held in 1968. The paper will present topics on fuels developments and on reactor physics. A second paper, *Thermal Reactor Plutonium Fuel Irradiation Programs in the United States*, will cover major irradiation programs being conducted in the United States which will demonstrate the utilization of plutonium in current power reactors. It has become clear with the U.S. nuclear industry that recycling of plutonium in thermal power reactors will be required by 1975. After that date there will be a rapid rise in available plutonium. Storage of this plutonium for later use in fast reactors is not required nor economically justified. Therefore, the U.S. industry is taking the necessary steps required to be ready to use the plutonium when it becomes available.

2.0 PLUTONIUM FUEL CHARACTERISTICS

2.1 Introduction

The technical feasibility of utilizing plutonium in thermal reactors has been successfully demonstrated through the success of numerous experimental programs (Table I). The irradiation program in the Plutonium Recycle Test Reactor (PRTR) was characterized by testing advanced mixed-oxide fuel concepts to moderate burnups under specific power conditions higher than those currently employed in commercial power reactors^[1,2]. Experiments are being conducted in the Saxton reactor to evaluate the performance of mixed-oxide fuel at linear power and burnup levels consistent with modern PWR technology^[3].

The success of these and other experimental programs is attested to by the universal acceptance of plutonium utilization in light water reactors. These efforts have been culminated by the initiation of demonstration programs in commercial power reactors (Table II) with the rapid full-scale implementation of plutonium utilization in commercial plants anticipated^[4,5,6].

Table I

MAJOR DOMESTIC EXPERIMENTAL UO_2 - PuO_2 IRRADIATIONS

<u>Reactor</u>	<u>No. of Rods</u>	<u>Clad</u>	<u>Fabrication Method</u>	<u>Peak Burnup, MWD/MTM</u>	<u>Peak Rod Power, KW/Ft.</u>	<u>Peak Heat Flux, BTU/Hr-Ft²</u>
PRTR	~1860	Zirc.	Swaged	~17,000	~17	390,000
PRTR	~ 700	Zirc.	Vipac	~18,500	~16	370,000
PRTR (High Power Density)	~1730	Zirc.	Pellet-Vipac-Swaged	~13,000	~21.5	500,000
EBWR	1296	Zirc.	Vipac	~ 3,000	8	200,000
Saxton-Core II	638	Zirc/SS	Pellet-Vipac	~29,000	16	530,000
Saxton-Core III	~ 250	Zirc.	Pellet	~38,000*	~19	630,000
Dresden-I	4	Zirc.	Pellet	~13,000	~10	230,000

*December 1970 - Goal Peak Burnup ~50,000 MWD/MTM on Rods from Saxton Core II Program.

Table II

MAJOR DOMESTIC PLUTONIUM DEMONSTRATION PROGRAMS

<u>Reactor Type</u>	<u>Reactor</u>	<u>No. of Units</u>	<u>Program Objective</u>
BWR	Big Rock Point	16 Assemblies 32 Rods	Fuel Concept Evaluation
	Big Rock Point	3 Assemblies 204 Rods	Performance Demonstration
	Dresden I	11 Assemblies 99 Rods	Performance Demonstration
	Vermont-Yankee	4 Assemblies 48 Rods	Performance Demonstration
PWR	San Onofre	4 Assemblies 720 Rods	Performance Demonstration

2.2 Plutonium Fuel Types

Plutonium recycle fuel evaluation studies have included a variety of fuel types and fabrication processes^[1,2]. Pellet fuel, because of a vast amount of satisfactory UO_2 experience, is the reference design for plutonium utilization in commercial reactors. Packed particle fuel (vipac) is considered to be the most promising alternate fuel form^[7]. Under the operating conditions presently employed or projected for commercial power reactors in the near future, neither pellet nor packed particle mixed-oxide fuels appear to be confronted with inherent performance limitations.^[8]

2.3 Irradiation Behavior - Effects of Plutonia Additions to UO_2

Although the small PuO_2 additions to UO_2 for thermal reactor fuel has an insignificant effect on most irradiation performance characteristics, there are important considerations related to performance that are unique with plutonium enriched fuels.

2.3.1 Plutonium Uniformity

Sintered mixed-oxide fuel material produced from powder prepared by the coprecipitation process is essentially uniform $(U, Pu)O_2$ solid solution with small localized PuO_2 nonuniformity^[9]. However, mechanical mixing of UO_2 and PuO_2 powders prior to pressing and sintering, which is the most commonly used method of preparing mixed-oxide fuels, results in the formation of non-uniform solid solution with localized regions of high PuO_2 concentration^[10]. Mechanically mixed and heterogeneously enriched mixed-oxide is also used for packed-particle fuels^[2,11,12].

Uniform solid solution forms rapidly in regions of mechanically mixed mixed-oxide fuel operating above columnar grain growth temperatures ($\sim 1700^\circ C$) by a vaporization-condensation process, whereas homogenization occurs more slowly in the equiaxed grain growth regions ($1400 - 1700^\circ C$) by solid state diffusion. Therefore, localized regions of high plutonium concentration persist in a significant volume of the fuel in a rod operating under normal conditions. The high fission density associated with these localized regions could affect such fuel performance characteristics as swelling and fission gas release although there is no definitive supportive evidence.

2.3.2 Fuel Clad Mechanical Interaction

Core-clad mechanical interaction phenomena, which are more severe in pellet fuels than in particle fuels^[13], are sensitive to fuel type and design and should not be affected by PuO_2 additions to UO_2 except as the PuO_2 additions may affect swelling behavior. Likewise, fuel cladding reaction in Zircaloy-clad fuels^[14,15] is probably related to stoichiometry and oxygen partial pressure in the rod and not plutonium concentration.

Postirradiation length measurements and diameter profiles were obtained on vipac fuel rods and rods containing hot-press and cold-press-sinter pellet fuel irradiated in PRTR to compare the dimensional behavior of the three fuel types^[8]. The vipac rods contained crushed, pneumatically impacted UO_2 - PuO_2 fuel vibrationally compacted to a smear density of nominally 86% TD. The hot-press and the cold-press-sinter pellet density was approximately 93% TD and the rods were assembled with a nominal 0.3 mm diametral gap.

Observations related to the rod dimensional data are summarized as follows:

- Cladding ovality and length changes, which seem to be related, are more pronounced in the cold-press-sinter pellet rods than in the hot-press pellet and vipac rods.

The fuel in the vipac rods provides cladding support and thereby reduces the amount of ovality. Considerably less ovality occurred in the hot-press pellet rods than in the rods which contained cold-press-sinter pellets although both were assembled with a nominal 0.3 mm diametral gap. It is suspected that the hot-press pellet fuel, because of its high stoichiometry ($\text{O/M} = \sim 2.15$) and reduced thermal conductivity, operated at higher temperatures than the cold-press-sinter fuel. The increased expansion resulted in closure of the fuel-clad gap and prevented cladding collapse and distortion.

- Although the Zircaloy cladding may be calculated to be free-standing, ovality results from creep which occurs early in the irradiation.

- The creep rate may be enhanced during neutron irradiation.
- There was no evidence of general or local clad swelling in any of the rods.
- There was no evidence of ridging at pellet interfaces in either the hot-press or cold-press-sinter pellet fuel rods.

2.3.3 Thermal Conductivity and Restructuring Kinetics

Small PuO_2 additions to UO_2 do not have a significant effect on the thermal conductivity of UO_2 ^[16]. Hence, fuel restructuring kinetics, which are affected by temperature and temperature gradient in the fuel, should not be significantly different for UO_2 and UO_2 - PuO_2 fuels operating under comparable conditions. Restructuring phenomena are more dependent on fuel type. Structural evidence of certain operating conditions in the fuel can be quickly eradicated by time-temperature-dependent mass transport and diffusion phenomena during subsequent reduced power operation^[2].

2.3.4 Fission Product Migration

The transport processes which affect fission product migration in oxide fuels during irradiation are not affected by small PuO_2 additions in UO_2 ^[17]. Fission products such as zirconium, niobium, cerium, and praseodymium, whose oxides are thermodynamically more stable than UO_2 and PuO_2 , do not redistribute significantly (Figure 1). Conversely, fission product cesium, ruthenium, and rhodium, whose oxides are thermodynamically less stable than UO_2 and PuO_2 , migrate significantly during irradiation (Figure 1).

2.3.5 Plutonium Redistribution

A significant change in radial plutonium concentration can occur rapidly in mixed-oxide fuel rods during irradiation^[17]. For mixed-oxide fuel operating above columnar grain growth temperatures ($\sim 1700^\circ\text{C}$), radial plutonium redistribution is coincident with the fuel restructuring phenomena and occurs as a result of the preferential evaporation of

FIGURE 1. RADIAL FISSION PRODUCT DISTRIBUTION IN A UO_2 -2 wt.% PuO_2 FUEL ROD AFTER IRRADIATION IN PRTR FOR 230 HOURS. THE DISTRIBUTION OF FISSION PRODUCTS WAS DETERMINED BY MICRO-GAMMA SCANNING.

gross amounts of uranium oxide from the central hot region of the fuel to the cooler peripheral regions near the cladding^[17]. Thermal diffusion can also cause plutonium enrichment in the high temperature region of the fuel^[18].

Electron microprobe scanning analyses of a vibrationally compacted UO_2 -2 wt.% PuO_2 fuel specimen irradiated for only 230 hours in PRTR show that the average plutonium concentration in the region of the fuel that operated at columnar grain growth temperatures and above, i.e., $>1700^\circ\text{C}$, is about 10% higher than the nominal as-fabricated concentration (Figure 2). The plutonium concentration in the once-molten fuel region is about 18% higher than the nominal value. Plutonium redistribution effects in mixed-oxide fuels have been shown to be stoichiometry dependent^[16].

2.3.6 Fission Gas Release

Fission gas release from oxide fuel is not significantly affected by adding plutonium to UO_2 or burnup level and is most sensitive to fuel temperature^[7]. Fission gas release fractions for pellet and particle fuels are comparable when evaluated on the basis of volumetric average fuel rod temperature. The higher surface area of particle fuels should result in a higher release by recoil-knockout; however, the contribution of this release mechanism to the total is relatively small under normal operating conditions.

Measurements made on mixed-oxide rods during the irradiation show that the release of sorbed gases and moisture from the fuel does not contribute to internal pressure^[19]. The released gases apparently react rapidly with Zircaloy cladding and/or the lower temperature fuel. Internal gas pressure resulting from the release of fission gases increases in a stepwise manner during reactor shutdown-startup power cycles with no significant changes occurring during steady power operation. This behavior, which is the same for UO_2 and UO_2 - PuO_2 fuels, is consistent with the diffusion-trapping release mode^[20] and the postulated containment of released fission gases in the central cavity of the rod until the fuel is cooled and cracking occurs^[21].

FIGURE 2. RADIAL PLUTONIUM CONCENTRATION IN AN IRRADIATED UO_2 -2 wt.% PuO_2 FUEL SPECIMEN AS DETERMINED BY ELECTRON MICROPROBE SCANNING. FUEL ROD WAS IRRADIATED FOR 230 HOURS IN PRTR.

2.3.7 Defect Behavior

Presently, there is no defect experience with mixed-oxide fuels in commercial power reactors. Comparative experiments to investigate the performance of defective and intentionally defected Zircaloy-clad mixed-oxide fuels were conducted in the Plutonium Recycle Test Reactor (PRTR)^[22] and Engineering Test Reactor (ETR)^[14].

A defect test was performed on a mixed-oxide fuel rod in the ETR pressurized loop facility. The pellet-containing rod was nominally 14.4 mm OD clad in 0.76 mm thick Zircaloy-4 cladding with a 68-cm active fuel length. The UO_2 -4 wt.% PuO_2 pellets were assembled in the cladding with a 0.25 mm diametral gap. Peak burnups of approximately 3000 MWd/MTM were achieved. The system pressure was 2000 psi at an average coolant temperature of 255°C.

The rod was intentionally defected by drilling a 0.51 mm 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.

The gross gamma scan of a companion nondefected rod shows a smooth fission product activity distribution that is related to the axial power profile. The gamma scan of the defected rod shows two localized variations in the fission product activity curve at and above the peak power position. 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 defected rod and companion nondefected rods show that the cladding has plastically deformed and become oval shaped during irradiation in the pressurized loop. The maximum ovality increase in the defected rod was 152 μm . Cladding collapse on the defected rod must have occurred during the relatively short predefect irradiation of 35 effective full power days since no collapse type deformation is expected to occur after defecting because of pressure equalization. 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.

The results of the defect experiments conducted to date show that (1) the defect performance of $\text{UO}_2\text{-PuO}_2$ fuels is essentially the same, although there is evidence that plutonium redistribution is enhanced in defected mixed-oxide fuels possibly as a result of a change in stoichiometry, i.e., increased O/M^[23], (2) defect behavior, i.e., activity release, is most sensitive to fuel type, fuel temperature, and power history^[23,24], and (3) the defect performance of both pellet and particle oxide fuels is excellent under normal operating conditions. It appears that the defect behavior of ceramic fuels is more sensitive to surface heat flux than to fuel type^[23,24]. Consequently, as more severe operating conditions are imposed on the fuel, defect behavior could become a performance limitation.

2.4 Transient Behavior

The behavior of oxide fuels when subjected to accidental high energy, short duration, power excursions is an important safety consideration in both thermal and fast reactors. Experiments were conducted to investigate and compare the transient behavior of the different plutonium-enriched fuel types^[8,25] and to investigate the possible effect of large PuO_2 particles or agglomerates in mixed-oxide fuels^[26].

The results of comparative transient experiments performed with unirradiated pellet and particle fuel pins show that^[8,25]:

- The cladding failure threshold energy of approximately 270 cal/gm fuel is essentially the same for both pellet and particle fuels.
- Both pellet and particle fuels fail by clad melting; however, because of higher fuel temperatures in particle fuels at comparable energy depositions, more fuel is expelled from the pin (Figure 3).
- The extent of Zircaloy-water reaction is comparable for both pellet and particle fuels and increases with increasing total energy deposition.

Transient tests were conducted on unirradiated oxide pellet-containing thermal recycle fuel pins to investigate the possible effects of large single 550 μm diameter PuO_2 particles, which could possibly be present in the fabricated fuel, on the transient behavior^[26]. The results are compared with the results of similar tests conducted on pins containing enriched UO_2 and $\text{UO}_2\text{-PuO}_2$ pellet fuel which did not contain abnormally large PuO_2 particles.

These tests demonstrate that below the energy deposition range of 200 to 213 cal/gm of fuel the energy generated in a single 550 μm diameter PuO_2 microsphere is readily absorbed by the surrounding UO_2 matrix and no external effects occur. Above this energy range, large PuO_2 particles near the surface of oxide pellets (0 to 800 μm) can penetrate the cladding (Figure 4). The cladding perforation threshold for thermal recycle pellet fuel containing 550 μm diameter PuO_2 agglomerates is in the energy deposition range of 200 to 213 cal/gm of fuel. Perforation occurs by localized melting (Figure 5) caused by the expulsion of PuO_2 particles located near the pellet surface through the cladding. Fuel loss to the water is limited to only the PuO_2 particle. Other tests have shown that the prompt cladding failure threshold due to melting for unirradiated pins containing enriched UO_2 and $\text{UO}_2\text{-PuO}_2$ pellet fuel is in

FIGURE 3. THE APPEARANCE OF ZIRCALOY-CLAD VIPAC AND PELLET-CONTAINING UO_2 PINS AFTER TRANSIENT IRRADIATION AT 275 cal/gm UO_2 .

FIGURE 4. POSTIRRADIATION APPEARANCE OF A ZIRCALOY-CLAD PIN SUBJECTED TO A TRANSIENT ENERGY DEPCSION OF 213 CAL/GM OF FUEL (CLADDING WAS PENETRATED BY TWO PuO_2 MICROSHERES LOCATED NEAR THE SURFACE OF UO_2 PELLETS.)

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200 μm

FIGURE 5. ZIRCALOY CLADDING PENETRATION CAUSED BY THE EXPULSION OF A PuO_2 MICROSPHERE LOCATED NEAR THE SURFACE OF THE UO_2 FUEL DURING TRANSIENT IRRADIATION.

the range of 225 to 274 cal/gm of fuel. Therefore, the presence of large 550 μm diameter particles in the fuel has slightly reduced the threshold energy deposition required for cladding perforation and the subsequent release of fission products. However, there were no indications of the effects of prompt fuel dispersal caused by the expulsion of the PuO_2 particles into the surrounding water when tested at these energy levels.

Because the presence of single 550 μm diameter PuO_2 particles in mixed-oxide fuels does not appear to significantly affect the cladding failure threshold energy from that of mixed-oxide fuels with the normal PuO_2 particle size and distribution, product specifications which limit the maximum PuO_2 particle size below 550 μm diameter do not appear to be warranted from the standpoint of transient fuel performance considerations. Specifications should be developed which limit the maximum PuO_2 particle size. These experiments have shown that such a limit is greater than 550 μm diameter.

3.0 PLUTONIUM FUEL FABRICATION

3.1 Fuel Fabrication Synopsis

Before the advent of the U.S. Atomic Energy Commission sponsored Plutonium Utilization Program at Pacific Northwest Laboratory in 1956, there was essentially no experience in producing plutonium-containing fuel elements. Largely through the success of this program, fabrication methods and techniques were developed which permit production of mixed-oxide fuels by either the pelletizing or the packed particle processes. The success of the program is attested to by the increased number of programs devoted to the commercial implementation of plutonium utilization in thermal reactors and the general acceptance of the need for and technical feasibility of plutonium utilization.

Solid mixed-oxide fuel pellets clad in Zircaloy is considered to be the current reference fuel design for the commercial utilization of plutonium in water cooled thermal reactors^[8]. This choice is a natural result of the vast amount of satisfactory experience accumulated with

UO₂ fuels using this basic design. Mixed-oxide pellet fuels can be prepared by sintering either coprecipitated (U, Pu)O₂ powder or a mechanical mixture of UO₂ and PuO₂. Variations in the basic pellet fuel design such as controlled density, controlled diametral gap, dishing, and coring are utilized to satisfy various performance criteria.

Advanced fuel concepts have been developed under the PUP, and packed particle fuel is considered to be the most promising alternate fuel form for commercial application^[8]. Packed particle fuels include the vibrationally compacted and swage-compacted concepts utilizing various feed materials such as sintered and crushed, fused, electrodeposited, pneumatically impacted, or Sol-Gel. Sol-Gel feed material can be as irregular or spherical shaped particles.

Packed particle fuels may possess performance advantages over pellet type fuel as higher heat ratings and burnups are demanded. Although most behavioral characteristics for pellet and particle fuels are similar, significantly reduced core-clad interaction and improved ability to accommodate swelling may constitute significant advantages for particle fuels^[8].

Sufficient technical information has been developed to permit establishment of process specifications for mixed-oxide fuel and cladding. Rigorous quality control and quality assurance procedures are employed during fabrication to provide fuel assemblies of the necessary quality. Establishing and achieving PuO₂ particle size and distribution requirements have been major considerations in mixed-oxide fuel production.

3.2 High Exposure Plutonium

Recycle plutonium produced in commercial power reactors contains significant amounts of the higher plutonium isotopes (Table III). The increased concentration of the higher isotopes results in increased gamma and neutron doses from high exposure plutonium which impose additional considerations related to the control of personnel exposure when handling this material during fuel fabrication^[27].

The photon spectrum from plutonium is a function of both reactor type and total exposure. There is less variation in spectrum from exposure than there is from reactor type^[28]. The isotopic composition of typical high exposure plutonium samples produced in different reactors is summarized in Table III.

Table III

ISOTOPIC CONTENT OF PLUTONIUM SAMPLES

Sample Source	Burnup, MWd/MTU	Isotopic Content, At. %					
		²³⁶ Pu	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
Yankee	8,000	4.0 x 10 ⁻⁷	0.144	87.0	9.56	3.06	0.208
Yankee	13,000	1.0 x 10 ⁻⁶	0.34	80.7	12.7	5.59	0.669
Yankee	24,000	3.0 x 10 ⁻⁶	1.00	67.7	18.9	10.0	2.51
Yankee	38,000	8.0 x 10 ⁻⁶	2.15	56.4	21.9	13.8	5.77
Dresden	20,000	1.4 x 10 ⁻⁶	0.403	69.55	21.46	6.862	1.719
Shippingport	20,000	1.4 x 10 ⁻⁶	1.06	51.2	28.9	11.0	7.78

Initially, the unshielded gamma dose rate from high exposure plutonium is due mostly to ²³⁸Pu^[28]. The higher energy 800 KeV radiations are very low in abundance but become the principal contributors to the dose rate from ²³⁸Pu when the lower energy 44 KeV and 100 KeV radiations are shielded out. Only about 1.3 mm of lead or lead glass shielding is required to eliminate almost all of the low energy radiation, leaving only the 800 KeV group^[28].

The unshielded dose rate from pure ²³⁹Pu is about a factor of 500 less than that from ²³⁸Pu. The penetrating components from ²³⁹Pu are at about 400 KeV.^[28]

The principal characteristics of the decay of ²³⁰Pu and ²⁴⁰Pu are similar and the difference factor of 100 lower for ²⁴⁰Pu is almost all due to the difference in half-life. The penetrating component from ²⁴⁰Pu (about 650 KeV) is both less energetic and less abundant than from ²³⁸Pu^[28].

Activity associated with the daughter products of ^{241}Pu and ^{236}Pu cause a substantial increase of dose rate from plutonium with time. Therefore, shielding requirements are dependent not only on the isotopic composition, but also on the time since chemical separation of plutonium from its daughter products.

^{241}Pu decays primarily by beta emission to form ^{241}Am . Because the half-life of ^{241}Am is longer than for ^{241}Pu , the americium activity associated with a plutonium source continues to increase for many years^[28]. A small amount of ^{237}U is also formed from ^{241}Pu , which causes an increase in dose rate shortly after chemical purification, but radiation from this source approaches equilibrium within a few weeks.

Decay of ^{236}Pu is complicated and no very long-lived nuclides are formed until the decay chain reaches stable ^{208}Pb . Even though the amount of ^{236}Pu is small (<1 ppb) in reactor produced plutonium, its daughter products, namely ^{208}Tl , can have an appreciable effect on dose rates from plutonium sources as measured through heavy shielding.^[28]

The calculated gamma dose rate from high exposure plutonium (38,000 MWd/MTM Yankee) 20 years after separation increases by a factor of 6 for unshielded material where L X-rays predominate, a factor of 36 through 1 cm of polyvinyl chloride where 40 to 100 KeV gammas predominate, and a factor of 6 through 1 cm of steel shielding where somewhat harder gammas predominate^[28].

Neutron dose measurements from high exposure plutonium are not well characterized nor are the neutron dose rates as well defined as the gamma dose rates^[29].

As with gamma radiation, the isotopic composition of plutonium has a significant effect on neutron dose rate. The specific neutron yield from spontaneous fission is highest for ^{238}Pu followed by ^{242}Pu and ^{240}Pu .

Because of secondary reactions, the neutron yield is also affected by the chemical form of the plutonium and impurities. Alpha particles from plutonium react with light elements such as beryllium, carbon, oxygen,

fluorine, and aluminum to produce neutrons. The (α , n) reaction with the oxygen in PuO_2 may increase the neutron yield and resultant dose rate by as much as 50% compared with plutonium metal^[29].

Because of its relatively high concentration, ^{240}Pu usually contributes the largest percent of neutrons emanating from high exposure plutonium. However, in high exposure PuO_2 where both spontaneous and (α , n) neutrons are produced, the specific neutron yield from ^{238}Pu is 10 times greater than for ^{242}Pu and 14 to 15 times greater than for ^{240}Pu ^[29]. Fortunately, the ^{238}Pu concentration is low in commercially produced plutonium.

Neutron doses measured on mixed-oxide fuel assemblies for commercial light water reactors have been low^[29]. However, when extrapolating the data obtained on higher exposure plutonium of the type expected to become available beginning the latter half of this decade, it appears that neutron shielding will become necessary for some fuel fabrication operations.

Although the theoretical gamma and neutron dose rates from commercially produced high exposure plutonium are high, particularly after a year or two of aging and daughter product formation, it is possible to reduce the dose rates to acceptably low levels for fuel fabrication and assembly by the use of practical shielding and material handling techniques. Studies have shown that the incremental fuel fabrication cost factor for mixed-oxide fuels containing high exposure plutonium is about 4% higher than the fabrication costs for fuel containing low exposure plutonium enrichment^[30].

4.0 REACTOR PHYSICS

The most rigorous methods of calculations do not predict "a priori", the criticality and other nuclear parameters of plutonium fueled systems to the desired accuracies. Normalization of methods, cross sections or calculated results to experimental data is required for nuclear design analysis. The impact of these uncertainties on the reactor operation will be substantially reduced by experience gained from the present U.S. demonstration programs and the probable mode of plutonium utilization where plutonium represents a small fraction (5% - 33%) of the reload fuel. The following subsections briefly summarize some recent comparisons of theory - experiment correlations conducted at Battelle-Northwest.

4.1 Batch Core Burnup Experiment in the PRTR

A major obstacle to the use of plutonium as fuel in present-generation power reactors has been the uncertainties which must be attached to the values of core reactivity lifetime predicted by calculation. To help reduce these uncertainties, a large-scale fuel burnup experiment (The Batch Core Experiment)^[31] was conducted in the Plutonium Recycle Test Reactor (PRTR). The basic fuel loading remained essentially unperturbed throughout the experiment, thus providing a unique set of data for use in checking methods of calculating reactivity lifetimes of reactor cores.

The PRTR Batch Core was a D₂O cooled and moderated, PuO₂-UO₂ fueled reactor system. The moderator and the coolant were separated by the process tubes which penetrated the core in the vertical direction. The fuel elements, which were clusters of 19 fuel pins clad in 1/2 in. diameter Zircaloy tubes, were loaded vertically inside the process tube. Each original fuel cluster contained about 29.44 kgs of uranium and 599.6 gms of plutonium. These fuels were arranged in a hexagonal lattice with 8 in. pitch in the core. In the standard configuration for power operation, about 80 elements were loaded, and operated at 60 MW.

The initial phase of the Batch Core Experiment comprised the loading of a new core of 55 UO₂-2 wt.% PuO₂ fuel elements and the performance of a series of critical tests to determine the physics parameters of this new core. Measurements included the critical loadings for boron concentrations ranging from zero to about 21 parts per million by weight (ppm) of ¹⁰B in the D₂O moderator, moderator level coefficients of reactivity over a 25-inch range of moderator heights at several boron concentrations, moderator void worths, thermal neutron flux distributions within the core, the effect of loss of coolant from fuel channels, fuel element worths, and kinetics parameters. At specified burnup levels during the Batch Core Experiment, interim critical tests were conducted on a basic 55 element core. These tests included measurements of core excess reactivity, fuel element worths, poison rod worths, and kinetics parameters^[32]. Some of the results from these measurements are shown in Tables IV and V. It is seen from Table IV that the deviation of calculated k_{eff} increases from the experimental k_{eff} of unity as burnup increases (¹⁰B decreases).

Table IV

CRITICALS VERSUS BURNUP

<u>Average Core (55 Elements) Burnups (MWd/MTM)</u>	<u>Unpoisoned Excess Reactivity (mk)</u>	<u>Experimental ¹⁰B (ppm) Concentration $k_{eff} = 1.0$</u>	<u>Poisoned Core Calculated k_{eff}</u>
0	255 ± 13	20.98	1.0066
1650	255 ± 12	17.30	1.0094
3560	194 ± 7	14.06	1.0139
4930	181 ± 6	12.33	1.01230
5330	167 ± 5	10.94	1.0216

Table V

CRITICALS AT END OF IRRADIATION

<u>Burnup (MWd/MTM)</u>	<u>No. of Elements</u>	<u>Excess Reactivity (mk)</u>
5330	55	167 ± 5
6139 ± 58	31	109 ± 2
6512 ± 61	19	50 ± 0.5

The extrapolations of calculated and experimental data to predict the end of life of the 80 element core have similar slopes. The calculated data predict a longer core life than the experimental data due to a combination of a larger initial excess reactivity and gentler slope. The calculated reactivity loss was -0.015 mk/MWd/MTM compared to an experimental value of -0.017 mk/MWd/MTM. The ²³⁹Pu and ²⁴⁰Pu isotopic number densities produced in fuel cluster burnup calculations agree with experimental values to within experimental uncertainties. Calculated ²⁴¹Pu and ²⁴²Pu number densities are lower than the experimental values and outside of the limits of experimental uncertainties.

4.2 Calculation of Multiplication Factors

A large number of "clean" plutonium fueled, H₂O moderated lattice experiments were reported at the 1968 IAEA panel meeting^[33]. Also, described was a set of analysis methods and how the results from the use of these methods compared with the lattice experiments. Since that time, changes in some of the cross section data were made (See Table VI). Also, improvements in the basic theory were developed as follows:

Slowing Down

- a. Include intermediate resonance formulation in HRG
- b. Allocate resonance cross section over more than one fine group in HRG.
- c. Upscattering correction for H in H₂O in HRG.
- c. Dancoff correction using Carlvik's method rather than ANL-5800 tables.

Thermalization

- a. Current calculation of diffusion coefficient in
BATTELLE-REVISED-THERMOS

The most significant changes in the cross section data were in the resonance integrals for ²³⁰U and ²³⁹Pu. The low value of the integral for ²³⁸U is arrived at by simply adding up all individually resolved resonances developed from resonance parameters. The modifications in the slowing down calculations also had an appreciable effect on the calculated k_{eff} 's, particularly for the tight lattices where resonance capture is high.

Tables VII, VIII, IX and X show comparisons between previous and current calculated k_{eff} 's for UO₂, Pu-Al and UO₂-PuO₂ lattice experiments^[34]. Generally the modified methods give k_{eff} results more nearly in line with experiments than the previously reported results. The lower ²³⁸U resonance integral results in considerable improvement in the correlation for the UO₂ Yankee experiments, particularly for the tight lattice cases. The Pu-Al data show much better correlation. However, the k_{eff} 's of the Al - 2 wt.% Pu experiments are still overpredicted by about 3% $\frac{\Delta K}{K}$. Because

Table VI

CHANGES IN BASIC DATA

<u>Quantity</u>	<u>Measured</u>	<u>Integral Value</u>	
		<u>Calculated from Master Library</u>	<u>Previous</u>
			<u>Current</u>
1. <u>Age in H₂O</u>	26.5 ± 0.3	25.5	26.5
2. <u>Resonance Integrals, I_∞</u> <u>(10⁷ to 0.5 eV)</u>			
a. ²³⁵ U I _{∞C}	140 ± 8	148	140
I _{∞f}	280 ± 11	286	280
α	0.50 ± 0.02	0.517	0.500
b. ²³⁸ U I _{∞C}	280 ± 12	282	269
c. ²³⁹ Pu I _{∞C}	-	215	177
I _{∞f}	310 ± 20	331	312
α	-	0.650	0.567
d. ²⁴⁰ Pu I _{∞a}	8370 ± 380	8424	8467
e. ²⁴¹ Pu I _{∞C}	-	164	164
I _{∞f}	557 ± 33, 524 ± 16	514	517
α	-	0.319	0.317
f. ²⁴² Pu I _{∞a}	1280 ± 60	1144	1112

Table VII

CHANGES IN CALCULATED VALUES OF K_{eff}
FOR UO₂ (YANKEE) CRITICALS

<u>Moderator/Fuel</u> <u>(Volume)</u>	<u>Calculated K_{eff}</u>		<u>ΔK_{eff}</u> <u>Current/Previous (10⁻³)</u>
	<u>Previous</u>	<u>Current</u>	
1.048	0.9731	1.0035	+30.4
1.405	0.9786	1.0017	+23.1
1.853	0.9828	1.0002	+17.4
3.357	0.9886	0.9989	+10.3
4.078	0.9888	0.9980	+ 9.2
4.987	0.9888	0.9969	+ 8.1
<u>Mean</u>	<u>0.9835</u>	<u>0.9999</u>	<u>+16.4</u>

Table VIII

CALCULATED VALUES OF K_{eff} FOR Al-Pu CRITICALS

Lattice Spacing (in.)	K_{eff}			
	Al-1.8 wt.% Pu		Al-2 wt.% Pu	
	Previous	Current	Previous	Current
0.75	1.0323	1.0034	1.0439	1.0214
0.80	1.0274	1.0069	1.0477	1.0288
0.85	1.0240	1.0054	1.0428	1.0294
0.90	1.0223	1.0051	1.0522	1.0394
0.95	1.0187	1.0029	1.0375	1.0257
Mean	<u>1.0249</u>	<u>1.0047</u>	<u>1.0448</u>	<u>1.0289</u>

Lattice Spacing (in.)	K_{eff} Al-5 wt.% Pu		
	Boron in H ₂ O (wppm)	Previous	Current
0.85	0	1.0232	1.0070
1.05	0	1.0234	1.0139
1.30	0	1.0125	1.0122
	<u>Mean</u>	<u>1.0197</u>	<u>1.0110</u>
1.05	0	1.0234	1.0139
	48	1.0206	1.0030
	101	1.0249	1.0099
	164	1.0222	1.0069
	229	1.0199	1.0056
	285	1.0193	1.0078
	<u>Mean</u>	<u>1.0217</u>	<u>1.0079</u>

Table IX

CALCULATED VALUES OF K_{eff}
FOR EBWR AND SAXTON FUELED CRITICALS

EBWR (UO_2 -1.5 wt.% PuO_2)

<u>Lattice Spacing</u> <u>(in.)</u>	<u>K_{eff}</u>	
	<u>Previous</u>	<u>Current</u>
0.55	0.9925	0.9936
0.60	0.9914	0.9910
0.71	0.9965	0.9940
0.80	0.9967	0.9938
0.90	0.9980	0.9958
0.93	0.9998	0.9979
<u>Mean</u>	<u>0.9958</u>	<u>0.9944</u>

SAXTON (UO_2 -6.6 wt.% PuO_2)

<u>Lattice Spacing</u> <u>(in.)</u>	<u>K_{eff}</u>	
	<u>Previous</u>	<u>Current</u>
0.52	1.0115	1.0013
0.56	1.0094	1.0011
0.735	1.0119	0.9957
0.792	1.0130	1.0007
1.04	1.0266	1.0163
<u>Mean</u>	<u>1.0145</u>	<u>1.0030</u>

Table X
CALCULATED VALUES OF K_{eff}
FOR UO_2 - PuO_2 FUELED CRITICALS

Lattice Spacing (in.)	<u>UO_2-2 wt.% PuO_2</u>					
	K_{eff}					
	8% ^{240}Pu		16% ^{240}Pu		24% ^{240}Pu	
	<u>Previous</u>	<u>Current</u>	<u>Previous</u>	<u>Current</u>	<u>Previous</u>	<u>Current</u>
0.80	0.9916	0.9854	-	-	0.9957	0.9877
0.93	1.0022	0.9937	1.0027	0.9955	1.0001	0.9921
1.05	0.9974	0.9887	1.0000	0.9923	1.0044	0.9942
1.143	1.0073	0.9976	1.0076	1.0007	1.0080	1.0006
1.32	1.0090	1.0016	1.0077	1.0036	1.0064	1.0016
1.386	1.0074	1.0004	1.0043	1.0011	1.0035	1.0035
1.611	0.9920	0.9973	-	-	-	-
<u>Mean</u>	<u>1.0010</u>	<u>0.9950</u>	<u>1.0045</u>	<u>0.9986</u>	<u>1.0030</u>	<u>0.9966</u>

<u>UO_2-4 wt.% PuO_2</u>	
	<u>K_{eff}</u>
0.80	0.9776
0.85	0.9925
0.93	0.9903
1.05	0.9985
1.143	0.9965
1.386	0.9946
1.60	0.9982
1.70	1.0040
<u>Mean</u>	<u>0.9928</u>
or	<u>0.9949</u> without 0.80 in. result.

of almost universal difficulty in predicting these experiments, the experimental data have been re-examined and the fuel is being analyzed, but so far no discrepancies have been uncovered.

The calculated individual and mean values of k_{eff} for almost all of the UO_2 - PuO_2 data are within $\pm 1\%$. As stated previously, these correlations are with the low (269b vs. 282b) resonance integral for ^{238}U . There is no integral data to justify the low value. Also, other correlations performed by Battelle on Yankee reactor burnup data are improved if the higher resonance integral value of 282b is used^[35]. This is seen in Figure 6. The only difference in the two calculations is the ^{238}U resonance integral. The "1969 Cross Section Evaluation" curve is for 269b and the "Hybrid Cross Section" curve is for 282b. It is seen that the ^{239}Pu buildup at high exposures tracks much better when I_∞ of 282b is used.

Several infinite medium multiplication factor measurements (k_∞) have been made in the Physical Constants Test Reactor which has the advantage of eliminating leakage and core-reflector spectrum problems associated with small critical experiments^[36]. Table XI presents data on two such lattices. In another series of PCTR experiments, the change in k_∞ with PuO_2 particle size was determined and is shown in Figure 7. The latter is of interest since mechanically blended UO_2 - PuO_2 results in a powder containing finite particles of PuO_2 in UO_2 . It is seen from Table XI that for the 0.9 wt.% PuO_2 experiment, the calculated k_∞ of 1.105 compares quite favorably with the experimental value of 1.104, while the difference in the k_∞ values for the 2 wt.% PuO_2 experiment is almost 3%. It was determined that compensating differences in experimental and calculated " η " and " f " values of approximately 1.5% yielded the good agreement for the 0.9 wt.% PuO_2 lattice. Although there was some compensation between " η " and " f " for the 2 wt.% PuO_2 experiment, it was insufficient to result in agreement on k_∞ 's. The calculated " f " was 2% low while the experimentally determined " ηf " was approximately 3% low.

FIGURE 6. ZODIAC CALCULATED ^{239}Pu - ^{242}Pu ISOTOPIC CONCENTRATION VS. ANALYTICAL DATA

FIGURE 7. REACTIVITY DEFECT ASSOCIATED WITH PuO_2 PARTICLE IN UO_2

Table XI
COMPARISON OF MEASURED AND CALCULATED
VALUES OF k_{∞} FOR UO_2 - PuO_2 LATTICES

Fuel Type	Mod/Fuel	k_{∞}	
		Meas.	Calc.
UO_2 -0.9 wt.% PuO_2	3.693	$1.104 \pm .004$	1.105
UO_2 -0.9 wt.% PuO_2	3.693 (276.3 wppm B)	$1.000 \pm .002$	0.989
UO_2 -2.0 wt.% PuO_2	3.693	$1.325 \pm .005$	1.365

From Figure 7, it is seen that k_{∞} is also overpredicted for the particle size experiments while the effect of particle size, slope of k_{∞} with particle size, is well predicted. The k_{∞} 's for both sets of these experiments were determined from lattices poisoned with ^{10}B in the moderator to yield $k_{\infty} = 1.0$ using the PCTR poisoned lattice technique. The ^{10}B concentration was determined in each case (See Table XI for one example). These values of boron concentrations were used in k_{∞} calculations which yielded values within ± 0.006 of 0.99 for all cases. The problem with the high k_{∞} cases appears to be wrapped up in calculating and/or deducing from experiments values of " η " and " f ". Some evidence indicates that the calculated spectrum is too soft and the shielding of the 0.3 ev ^{239}Pu resonance is too large resulting in too high of calculated " η " values.

4.3 Reactor Kinetic Parameters of Lattices of Plutonium and Uranium in Water^[37]

The ratio β_{eff}/λ has been deduced from reactor noise measurements in several uniform light-water lattices in the Plutonium Recycle Critical Facility. These lattices included one in which the fuel was an aluminum-plutonium alloy, one in which the fuel was slightly enriched UO_2 , and several in which the fuel was a uniform mixture of UO_2 and PuO_2 . All of the noise measurements were performed in clean critical lattices, at low power, and in loadings which approximated circular cylinders. The fuel loadings were all moderated and infinitely-reflected with light water. The fuel rods and lattices are described in Tables XII and XIII.

Table XII
DESCRIPTION OF FUELS

Core No.	1*	2	3,4,6	5	7
Fuel Type	Al-1.83 wt% Pu	UO ₂ -1.5 wt% PuO ₂	UO ₂ -2 wt% PuO ₂	UO ₂ -2 wt% PuO ₂	UO ₂
Fuel dia. (in.)	0.500	0.372	0.508	0.508	0.440
Fuel length (in.)	43.83	48.50	36.0	36.0	36.0
Clad I.D. (in.)	0.506	0.372	0.508	0.508	0.440
Clad thickness (in.)	0.030	0.027	0.030	0.030	0.030
Clad material	Zircaloy-2	Zircaloy-2	Zircaloy-2	Zircaloy-2	Aluminum
g PuO ₂ -UO ₂ /rod	-	828.0	1128	1128	-
g PuO ₂ /rod	-	12.42	22.56	22.56	-
g UO ₂ /rod	-	815.58	1105.44	1105.44	825.6
g Pu-Al/rod	389.32	-	-	-	-
Average	²³⁹ Pu	93.97	91.41	71.76	-
Pu	²⁴⁰ Pu	5.58	7.83	23.50	-
Composition	²⁴¹ Pu	0.44	0.70	4.08	-
wt%	²⁴² Pu	0.014	0.03	0.66	-
Uranium Enrichment	None	0.16	natural	natural	2.32
wt% ²³⁵ U					

*The "aluminum" in Fuel No. 1 was composed of 97.21 wt% Al, 1.61 wt% Ni, 0.49 wt% Fe, 0.69 wt% Si.

Table XIII

DESCRIPTION OF CORES

<u>Core No.</u>	<u>1</u>	<u>2</u>	<u>3</u>	<u>4</u>	<u>5</u>	<u>6</u>	<u>7</u>
Number of Fuel Rods at Critical	483	482	151	252	394	319	378
Lattice Pitch (cm)	2.032	1.803	2.692	2.159	2.159	1.905	1.905
Lattice Pitch (in)	0.80 (hex)	0.71 (hex)	1.06 (square)	0.85 (hex)	0.85 (hex)	0.75 (square)	0.75 (square)
Effective Cylindrical Core Radius (cm)	24.0954	21.17227	18.617	17.976	22.485	19.0754	20.8601
Axial Buckling, m^{-2}	5.998	5.25	8.89	8.89	9.11	8.9	8.9

Values of β_{eff} and λ were calculated for these lattices with a code^[38] that uses standard perturbation-theory expressions^[39] for these quantities. Direct and adjoint fluxes for the perturbation theory expressions are obtained from a one-dimensional, multigroup, diffusion-theory code^[40]. The isotopic delayed neutron fractions that were used are from Ref. [39]. Average inverse velocities, prompt neutron spectra, and values of $\overline{v\sigma_f}$ were obtained from cell calculations using the computer codes BATTELLE-REVISED-THERMOS^[41] and HRG (Hanford Revised GAM)^[42].

Calculations of β/λ were performed using four, six, or eight energy groups of neutrons (Table XIV). The four-group scheme made no differentiation between the spectra of prompt and delayed neutrons, whereas the six- and eight-group schemes did. In the six-group scheme all the delayed neutrons were assumed to occur in Group 2. In the eight-group scheme the delayed neutron spectrum that was used is that given by Keepin^[43], which is a composite of the data reported by Batchelor and Hyder^[44] and Bonner et al.^[45]

Calculated values of β_{eff} and λ are listed in Table XV. The calculated values of λ change only slightly with changes in the high-energy-group structure. However, calculated values of β_{eff} obtained with the four-group scheme are approximately 10% smaller than values obtained with six or eight groups. This is what one would expect, and illustrates the fact that the differences in age, τ , (and thus also in leakage, τB^2) between the prompt and delayed neutrons are significant. The refinement from six to eight energy groups, in which the delayed spectrum is described somewhat more exactly, produces a much smaller change than going from four to six energy groups.

Table XIV

ENERGY GROUP BOUNDARIES

gp	8-groups	6-groups	4-groups
1	10 MeV - 1.35 MeV	10 MeV - 498 keV	10 MeV - 11.7 keV
2	1.35 MeV - 498 keV	498 keV - 235 keV	11.7 keV - 2.38 eV
3	498 keV - 235 keV	235 keV - 11.7 keV	2.38 eV - 0.683 eV
4	235 keV - 111 keV	11.7 keV - 2.38 eV	0.683 eV - 0 eV
5	111 keV - 11.7 keV	2.38 eV - 0.683 eV	
6	11.7 keV - 2.38 eV	0.683 eV - 0 eV	
7	2.38 eV - 0.683 eV		
8	0.683 eV - 0 eV		

TABLE XV
CALCULATED VALUES OF β_{eff} AND λ

Core No.*	Fuel Type*	H/Pu Atom Ratio	$\beta_{eff} \times 10^3$			$\lambda, 10^{-5} \text{ sec}$		
			4	6	8	4	6	8
			Groups	Groups	Groups	Groups	Groups	Groups
1	A1-1.8 wt% Pu	810	-	2.42	2.37	-	6.942	6.914
2	UO ₂ -1.5 wt% PuO ₂	567	-	3.17	3.12	-	4.200	4.209
3	UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu)	686	-	-	3.55	-	-	4.890
4	UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu)	293	3.45	3.87	3.78	2.994	3.021	3.031
5	UO ₂ -2 wt% PuO ₂ (24% ²⁴⁰ Pu)	293	3.74	4.05	3.98	2.883	2.899	2.899
6	UO ₂ -2wt% PuO ₂ (8% ²⁴⁰ Pu)	243	-	3.94	3.83	-	2.718	2.697
7	2.35 wt% enr. UO ₂	330**	-	7.58	7.44	-	4.657	4.654

* Described in detail in Tables XII and XIII.

** H/²³⁵U atom ratio in this case.

Measured and calculated (8-energy groups) values of $\beta_{\text{eff}}/\lambda$ are compared in Table XVI. The measured and calculated values agree, to within the standard deviation of the measured value, in all cases except one. Even in this worst case, however, the disagreement is only approximately 6%. The $\beta_{\text{eff}}/\lambda$ for this odd case (core No. 6) has been measured^[46] independently using another method, in which the value for a delayed critical α is obtained by extrapolating subcritical measurements of α . Interestingly, the calculated value for $\beta_{\text{eff}}/\lambda$ falls between the two measured values.

Table XVI
CALCULATED AND MEASURED $\beta_{\text{eff}}/\lambda$ VALUES

Core No.*	Fuel Type*	H/Pu Atom Ratio	$(\beta_{\text{eff}}/\lambda)$ Calc, sec ⁻¹	$(\beta_{\text{eff}}/\lambda)$ Meas., sec ⁻¹
1	Al-1.8 wt% Pu	810	34	33 ± 3
2	UO ₂ -1.5 wt% PuO ₂	567	74	71 ± 6
3	UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu)	686	73	76 ± 3
4	UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu)	293	125	128 ± 8
5	UO ₂ -2 wt% PuO ₂ (24% ²⁴⁰ Pu)	293	137	133 ± 8
6	UO ₂ -2 wt% PuO ₂ (8% ²⁴⁰ Pu)	243	142	151 ± 3 (135 ± 1)**
7	2.35 wt% enr. UO ₂	330**	160	153 ± 8

* Described in detail in Tables XII and XIII.

** H/²³⁵U atom ratio in this case

*** Measured by a different method (Reference 46).

4.4 Calculations of Power Distributions^[47]

An analysis has been made of twelve experiments conducted as part of the joint USAEC-CNEN experimental program^[48]. This study involves six configurations for each of two fuel types, 2.35 wt% enriched UO₂ and 2 wt% PuO₂-UO₂ (8% ²⁴⁰Pu) rods. The six configurations were (1) a regular uniform loading of rods; (2) the same loading but with a water hole in the center (i.e., the central fuel rod was

removed); (3) water slab (a row of fuel rods removed); (4) water cross; (5) a 7 x 7 rod array surrounded by water slots; (6) a similar 9 x 9 rod array.

In these twelve experiments, spatial power distributions were measured by gamma-scanning selected fuel rods. The main interest was in the rod-to-rod power distribution, especially the effects of water slots on the power peaking. The series of experiments, ranging from the simplest (regular) array to one simulating a 7 x 7 or 9 x 9 bundle, provides a systematic test for the evaluation of calculational methods.

In the analysis of H₂O-moderated and reflected experiments (especially clean critical experiments) one generally assumes the reactor is composed of two regions: core and reflector. Few-group cross sections are calculated, assuming that an infinite medium spectrum applies in each region. The few-group cross sections are then used in a diffusion theory calculation of k_{eff} .

In an earlier study^[49], it was pointed out that such a two-region, infinite medium model generally does not predict the power distribution well, although it may yield a satisfactory value for k_{eff} . In general, this method shows a pronounced trend, such that if the power distribution is normalized at the center of the core, the power near the core-reflector interface is consistently underestimated. A simple modification, which resulted in considerably improved correlations, was reported in Reference [49]. This simple modification consisted in introducing an extra reflector region (one lattice unit thick, adjacent to the core) which is represented by cross sections averaged over a spectrum characteristic of the core.

The multigroup transport theory codes HRG and Battelle-Revised-THERMOS were used to generate four-group cross sections for core and reflector regions. These cross sections were used in the two-dimensional diffusion theory code 2DB in an x-y calculation of power distributions and k_{eff} .

Four mesh points per cell were used in the 2DB calculations: This mesh description was carried out two lattice units into the reflector; then the mesh points were more widely spaced. An axial buckling of 8.9 m^{-2} was used consistently.

In our current analysis we have compared three variations of our calculational model:

Model 1. The usual two-region, infinite-medium model (described in previous section);

Model 2. A simple modification consisting of an additional reflector region whose cross sections are obtained from cell calculations performed for the core (described in the previous section). In this model all water gaps also contained these modified reflector cross sections.

Model 3. A more detailed representation of the differences in spectrum in successive rows of fuel and water. This was accomplished with THERMOS calculations in slab geometry, with appropriate homogenized regions of core, reflector and gaps; editing was done over the proper spatial points to obtain average cross sections for each "row" of fuel and water. For analysis of the UO_2 loadings, five sets of core cross sections were used to represent fuel rods in various locations, and four sets of cross sections were used to represent water. For analysis of the $\text{UO}_2\text{-PuO}_2$ loadings, three sets of core cross sections and three sets of water cross sections were used.

Power distributions calculated using the three models described above were compared with measured distributions. The trend that is so evident in the regular lattices when using Model 1 (i.e., calculated power consistently underestimated near core-reflector boundary) was significantly reduced when the modified models were used. The simple modification used in Model 2 gives a better correlation than the more refined modification used in Model 3. This is evident especially in the case of the mixed oxide loading.

The goodness of each correlation is represented by a standard deviation, σ , defined by^[48]

$$\sigma = \sqrt{\sum_i^N (\bar{\delta} - \delta_i)^2 / (N - 1)}$$

where N is the number of rods measured,

$$\delta_i = \frac{P_{\text{calc}} - P_{\text{meas}}}{P_{\text{meas}}} \text{ for the } i^{\text{th}} \text{ rod,}$$

$$\text{and } \bar{\delta} = \sum_i^N \delta_i / N.$$

The definition of σ implies an "effective" normalization such that the average fractional deviation, $\bar{\delta}$, is zero. This definition thus makes σ independent of the particular choice of normalization, and provides us a meaningful, consistent measure for purposes of comparing methods.

The σ 's for the various cases are given in Table XVII. We can make the following general observations:

- In every case σ was significantly reduced when modifications were made to the simple two-region, infinite medium model (Model 1).
- In most cases, Model 2 gives the best correlation.

Table XVII
STANDARD DEVIATION (%) IN POWER DISTRIBUTIONS

	UO_2			PuO_2-UO_2		
	<u>Model 1</u>	<u>Model 2</u>	<u>Model 3</u>	<u>Model 1</u>	<u>Model 2</u>	<u>Model 3</u>
Regular	2.09	1.18	1.05	3.37	1.86	2.17
H ₂ O-hole	1.48	0.98	1.14	2.48	1.78	-
H ₂ O Slab	1.95	1.48	1.56	1.60	1.38	1.13
H ₂ O Cross	1.57	1.37	1.21	2.13	1.40	-
7 x 7	1.96	1.72	2.16	2.26	1.69	1.78
9 x 9	2.91	2.67	3.12	2.29	1.90	-

The simple modification incorporated in Model 2 of our analysis significantly improved power distribution correlations for the twelve configurations that were analyzed. This improvement, together with the simplicity of this method, makes this method attractive for calculating power distributions in H₂O cores. The simplicity of the model comes about because cross sections for the modified water regions (water gaps and the reflector adjacent to the core) are obtained directly from cell calculations for the core. No additional calculations are necessary.

4.5 The Effect of Plutonium-241 Decay on the Analysis of Plutonium Critical Experiments^[50]

Often there is a considerable lapse of time between measurement of plutonium isotopic densities and use of the plutonium in critical experiments or power reactors. In addition, the same plutonium is often used in subsequent experiments extending over several years. In many cases the isotopic densities need to be adjusted to account for the decay of ²⁴¹Pu over these periods of time. This isotope decays with a half-life of ~14.5 years, resulting in a gradual depletion of fissile plutonium and a corresponding buildup of ²⁴¹Am which also must be accounted for.

For purpose of illustration, the effect of ^{241}Pu decay on reactivity has been calculated for several light-water-moderated lattices of UO_2 -4 wt% PuO_2 fuel rods^[51]. The plutonium isotopic compositions were measured in 1967, and the fuel was used in critical experiments in 1969. Originally, the plutonium contained ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu , in the amount of 0.28, 75.38, 18.10, 5.08, and 1.15 wt%, respectively. In the 2-year period from the isotopic measurements to the critical experiments, the ^{241}Pu content diminished by 10%, and an amount of ^{241}Am corresponding to this decay was formed.

Changes in cell-average thermal " $\bar{\eta}f$ " values due to this 10% loss of ^{241}Pu and buildup of ^{241}Am were calculated using the BATTELLE-REVISED-THERMOS^[41] code, and are shown in Table XVIII. The results show that calculations in which the effects of ^{241}Pu decay are ignored result in multiplication factors that are too large. Some specific results of exchanging ^{241}Pu for ^{241}Am are:

1. A significant reduction in " $\bar{\eta}f$ ".
2. The change is mainly in " $\bar{\eta}$ " since " f " changes very little.
3. The buildup of ^{241}Am has a larger reactivity effect than the depletion of ^{241}Pu .

The reason an exchange of ^{241}Pu for ^{241}Am changes the " $\bar{\eta}f$ " value is clear when one compares the thermal cross sections of the two isotopes. The capture cross sections for the two isotopes are of the same order of magnitude but the fission cross section of ^{241}Pu is approximately 300 times that of ^{241}Am . This explains also why " $\bar{\eta}$ " changes significantly, whereas " f " does not. The value of " $\bar{\eta}$ ",

$$\bar{\eta} = \frac{\nu}{1 + (\bar{\Sigma}_c / \bar{\Sigma}_f)_{\text{fuel}}}$$

reflects the change in total fissions in the fuel rod, which is significant, whereas the value of " f " reflects the change in total absorptions in the rod, which is less significant. The values of " $\bar{\eta}$ " and " f " are flux- and volume-weighted averages over the specified regions.

TABLE XVIII

THE EFFECT OF PLUTONIUM-241 DECAY ON CALCULATED VALUES OF " $\bar{n}$ " and "f"

Atom Ratio H/Pu	Parameter	Calculated Parameters Using Original Densities	Reduced ^{241}Pu Density by 10%		Added Corresponding Amount of ^{241}Am	
			Value of Parameter	% Decrease from Original	Value of Parameter	% Decrease From Original
123.5	$\bar{n}$	1.7886	1.7369	0.10	1.7791	0.53
	f	0.94741	0.94724	0.02	0.94738	0.00
	$\bar{n}f$	1.6945	1.6926	0.11	1.6855	0.53
152.8	$\bar{n}$	1.7951	1.7932	0.11	1.7858	0.52
	f	0.93478	0.93452	0.03	0.93470	0.00
	$\bar{n}f$	1.6780	1.6758	0.13	1.6692	0.52
203.3	$\bar{n}$	1.8027	1.8009	0.10	1.7939	0.49
	f	0.91258	0.91234	0.03	0.91254	0.00
	$\bar{n}f$	1.6451	1.6430	0.13	1.6370	0.49
288.7	$\bar{n}$	1.8107	1.8087	0.11	1.8023	0.46
	f	0.87537	0.87493	0.05	0.87522	0.02
	$\bar{n}f$	1.5850	1.5825	0.16	1.5774	0.48
413.9	$\bar{n}$	1.8150	1.8132	0.10	1.8070	0.44
	f	0.84403	0.84359	0.05	0.84385	0.02
	$\bar{n}f$	1.5319	1.5296	0.15	1.5248	0.46

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