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# THORIA FUEL IRRADIATION

## PROGRAM TO IRRADIATE 80% $ThO_2$/20% $UO_2$ CERAMIC PELLETS AT THE SAVANNAH RIVER PLANT

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## ABSTRACT

This report describes the fabrication of proliferation-resistant thorium oxide/uranium oxide ceramic fuel pellets and preparations at the Savannah River Laboratory (SRL) to irradiate those materials. The materials were fabricated in order to study head end process steps (decladding, tritium removal, and dissolution) which would be required for an irradiated proliferation-resistant thorium based fuel. The thorium based materials were also to be studied to determine their ability to withstand average commercial light water reactor (LWR) irradiation conditions.

This program was a portion of the Thorium Fuel Cycle Technology (TFCT) Program, and was coordinated by the Oak Ridge National Laboratory (ORNL) under the Consolidated Fuel Reprocessing Program (CFRP). The fuel materials were to be irradiated in a Savannah River Plant (SRP) reactor at conditions simulating the heat ratings and burnup of a commercial LWR. The program was terminated due to a de-emphasis of the TFCT Program, following completion of the fabrication of the fuel and the modified assemblies which were to be used in the SRP reactor.

The reactor grade ceramic pellets were fabricated for SRL by Battelle, Pacific Northwest Laboratories (PNL). Five fuel types were prepared:

- 100% $UO_2$ pellets (control)
- 80% $ThO_2$/20% $UO_2$ pellets
- Approximately 80% $ThO_2$/20% $UO_2$ + 0.25 CaO (dissolution aid) pellets
- 100% $UO_2$ "hybrid" pellets (prepared from sol-gel microspheres)
- 100% $ThO_2$ pellets (control)

All of the fuel materials were transferred to SRL from PNL and were stored pending a subsequent reactivation of the TFCT Programs.

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## THORIA FUEL IRRADIATION: PROGRAM TO IRRADIATE 80% $ThO_2$/20% $UO_2$ CERAMIC PELLETS AT THE SAVANNAH RIVER PLANT

### INTRODUCTION

In 1977, an evaluation of fuel cycles using thorium was initiated by the Energy Research and Development Administration (ERDA). The objective of the Thorium Fuel Cycle Technology (TFCT) Program was to develop the technical information required to support future decisions on establishing fuel recycle facilities.[1] The primary criterion for evaluating thorium fuel cycles was the extent to which the potential for international proliferation of nuclear materials was reduced. A reference case thorium fuel cycle was defined[2] to serve as a common basis for development work at the participating national laboratories, universities, and commercial facilities.

Characteristics of the reference fuel cycle for the TFCT program were defined to be:

- Fissile uranium would be denatured by mixing with U-238.
- Chemical processing plant design would be based on the assumption that plants are located in secure areas.
- Plutonium would be recycled within these secure areas.
- Thorium would be recycled with recovered uranium and plutonium.
- The head end of the chemical processing plant would handle a variety of core and blanket fuel assembly designs for light water reactors (LWRs) and heavy water reactors (HWRs).
- The fuel form would be a homogeneous mixture of uranium and thorium oxide powders pressed into pellets.
- Fuel cladding would be zircaloy.
- MgO would be studied as an additive to improve the thorium-dissolving characteristics.

Thorium was being considered as the fertile component of nuclear reactor fuel in the expectation that its use would provide increased proliferation resistance compared to the use of U-238 as the fertile material in reactor fuel. The reason is that U-235 (which constitutes the initial fissile material) and U-233 (which is produced when thorium is irradiated) can be denatured, or made unusable for feasible explosive devices, by mixing with U-238, whereas plutonium cannot be denatured.

The proliferation-resistant composition for the proposed thorium oxide based fuels for LWRs utilizes a composition of 80%

$ThO_2$-20% $UO_2$.[3] The response of such fuels during the fabrication operations, reactor irradiation, and subsequent chemical reprocessing is considerably different from that of uranium oxide based fuels. Thus, procedures developed for uranium operations must be modified substantially. In particular, thorium oxide based fuels are much more difficult to dissolve during chemical reprocessing than uranium oxide based fuels. Thorium based fuels require the use of fluoride ion as a catalyst (approximately 0.05 molar) in concentrated nitric acid to affect dissolution. Grinding of the fuel to a small particle size is required, and even so, dissolution is still usually two to three times slower than with uranium based reactor grade ceramic materials. The very corrosive dissolver solution requires selection of corrosion-resistant dissolver materials and the fluoride catalyzed solution may complicate the subsequent vitrification processes of waste management. Techniques to remove tritium prior to dissolution will be significantly more difficult for thorium oxide based fuels than for uranium oxide fuels, since $ThO_2$ based fuels do not form powders during oxidative roasting.

The work discussed in this report was performed as part of a program to provide irradiated ceramic oxide materials with the reference case $ThO_2/UO_2$ composition (80%/20%) for chemical reprocessing studies by the Savannah River Laboratory (SRL) and to provide the Pacific Northwest Laboratory (PNL) with samples for evaluation of irradiation performance.

## SUMMARY

Approximately 13 kg of full-size reactor grade ceramic oxide pellets were prepared by PNL and loaded into zircaloy rodlets for irradiation in a Savannah River Plant (SRP) reactor. The types of fuel materials are shown in Table 1.

**TABLE 1**

**Fuel Materials for the Thorium Irradiation Programs**

| Type | Description | No. Rodlets | Kg |
|---|---|---|---|
| I | 100% $UO_2$ Pellet | 6 | 2.6 |
| II | 80% $ThO_2$/20% $UO_2$ Pellet | 6 | 2.6 |
| III | 79.75% $ThO_2$/20% $UO_2$ + 0.25% CaO Pellet | 3 | 1.3 |
| IV | 100% $UO_2$ Sol-Gel Microsphere Hybrid Pellet | 3 | 1.3 |
| V | 100% $ThO_2$ Pellet | 12 | 5.2 |
| | Total | 30 | 13.0 |

The Thorium Irradiation Program was terminated prior to the irradiation of the tests materials, due to a de-emphasis of TFCT Programs. The fuel materials were prepared by PNL, shipped to SRL, and stored.

The objective of this report is to summarize the preparation of the fuel materials by PNL and to describe the preparations for the irradiation of those materials at SRP.

# DISCUSSION

## Background

### Material Requirements

The reference thorium fuel cycle[2] specified a fuel form consisting of a homogeneous mixture of thorium oxide and uranium oxide powders pressed into a pellet and contained within a zircaloy cladding. The oxide powders could be either coprecipitated from mixed thorium-uranium solution or mechanically mixed from pure thorium oxide and uranium oxide blend; the mechanically mixed powders were most closely related to current uranium oxide fuel technology. Small amounts of other constituents, such as magnesium oxide or calcium oxide, might be added as dissolution aids. The fuel pellets were to be formed by cold pressing and sintering techniques. Alternative fuel forms being considered consisted of sol-gel microspheres compacted into cladding by vibrating or cold pressed and sintered into hybrid pellets.

Limits on U-235 enrichments of about 20 percent, and on U-233 enrichments of about 12 percent, were placed on the constituent uranium oxide to ensure a proliferation-resistant fuel cycle. These limits required the thorium oxide based fuels to have uranium oxide contents in the range of 20 to 30 percent to achieve the 4 percent enrichment typical of LWR operation.

Diameters of fuel pellets for LWR operation range from 0.32 to 0.42 in. (typically 0.370 in.) and length/diameter ratios range from 1.0 to 1.5. The zircaloy rods are typically 0.430 in. OD, with 0.027-in.-wall thickness.

The fuel rods in pressurized water reactors operate generally at average power ratings of 6 to 9 kW/ft, with maximum ratings under normal operation up to 14 kW/ft. The maximum power ratings limit central fuel temperatures to not much more than 1500°C, the approximate threshold for restructuring of the uranium oxide due to recrystallization effects. Maximum exposures of fuel in LWRs are generally in the range of 30,000 to 35,000 MWD/MTHM (megawatt days/metric ton heavy metal).

### Existing Materials

A survey of available thorium oxide/uranium oxide irradiated fuels was conducted to determine if these could be utilized in the TFCT Program. The fuel samples available to SRL were:

- 95% $ThO_2$/5%$UO_2$ Elk River fuel, irradiated to approximately 12,000 MWD/MTHM
- 97% $ThO_2$/3%$UO_2$ Dresden fuel
- Westinghouse-Bettis irradiated test samples of various types

The Elk River and Dresden fuels did not meet the anti-proliferation guidelines (approximately 80% $ThO_2$/20%$UO_2$) and were, therefore, unsuitable for the reprocessing and post-irradiation studies. Some of the irradiated Westinghouse samples were of the desired formula, but not enough of the materials were available (<1 kg) for the head end, dissolution, and solvent extraction studies.

### Request for Quotations

Requests to present proposals to supply irradiated $ThO_2$/$UO_2$ (80%/20%) fuels were transmitted to four commercial fuel vendors:

- Babcock & Wilcox, Lynchburg, VA
- General Atomics, San Diego, CA
- Nuclear Fuel Services, Erwin, TN
- Westinghouse Electric, Pittsburgh, PA

An irradiation program was also developed with PNL to prepare reactor grade ceramic pellets at PNL and to subsequently irradiate them in a SRP reactor.

Babcock & Wilcox (B & W) was the only commercial vendor to reply with a program proposal. In the B & W proposal, about 50 kg of thorium oxide/uranium oxide pellets fabricated by B & W and irradiated to 10,000 MWD/MTHM in a utility reactor were to be provided to SRL beginning about November 1981 (assuming a program starting September 1, 1978); post-irradiation examinations to characterize irradiation performance of the fuel would continue through January 1982. Projected cost was $2.3 million. The B & W proposal delayed delivery of irradiated material by about one year compared to the proposed PNL fabrication-SRP irradiation, and made no provision for supply of fuel at typical LWR exposures of about 30,000 MWD/MTHM. The plan worked out with PNL was chosen as the most cost-effective approach to meet the desired timing goals and was identified as the "Thorium Irradiation Program."

## Thorium Irradiation Program

### Introduction

In the Thorium Irradiation Program, thorium oxide/uranium oxide pellets were to be fabricated at PNL, placed in zircaloy rods, and shipped to SRL for irradiation in a SRP reactor. The irradiated materials would be used for tritium release and dissolution tests by SRL and for fuel performance studies by PNL.

PNL was responsible for fabrication of the test materials; for procurement of the enriched uranium; and for design, procurement, and loading of the zircaloy rods.[4] PNL was to supply prototype rods containing $ThO_2$ pellets needed for hydraulic and heat transfer tests of the irradiation assembly at SRL.

SRL was responsible for design and fabrication of the irradiation test assembly, and was to conduct all necessary test preparations, including preliminary hydraulic and heat transfer tests and long-term flow tests of the irradiation assembly.

### Program Details

Approximately 10 kg of PWR-size pellets of various material types and containing approximately 2 percent U-235 were to be exposed at average power ratings of 14 to 18 kW/ft within quatrefoil assemblies in the buckled zone of a Mark 16B-31 charge in P Reactor. The preliminary assembly design is shown in Figure 1. The test fuel, stacked within short-length rodlets, would form seven columns in two quatrefoil assemblies, with each column containing about three hundred pellets and weighing about 1.3 kg.

Following irradiation, the test assemblies were to be dismantled in the P Reactor disassembly basin at the reactor. Individual fuel rods would be transferred to the SRL High Level Caves for tests of tritium removal and fuel dissolution. In addition, post-irradiation examinations would be conducted to establish reactor behavior of the thorium oxide based fuels and to characterize microstructural features of significance to their dissolubility. Selected rods of primary interest to PNL would be shipped offsite for special characterizations provided by gamma scanning, profilometry, eddy-current testing, and neutron radiography. Post-irradiation examination operations conducted at SRL were to include burnup analyses, measurement of the quantities and composition of fission gases released, dimensional and density measurements of the irradiated fuel pellets, and characterization of microstructural features of the pellets as developed by optical microscopy.

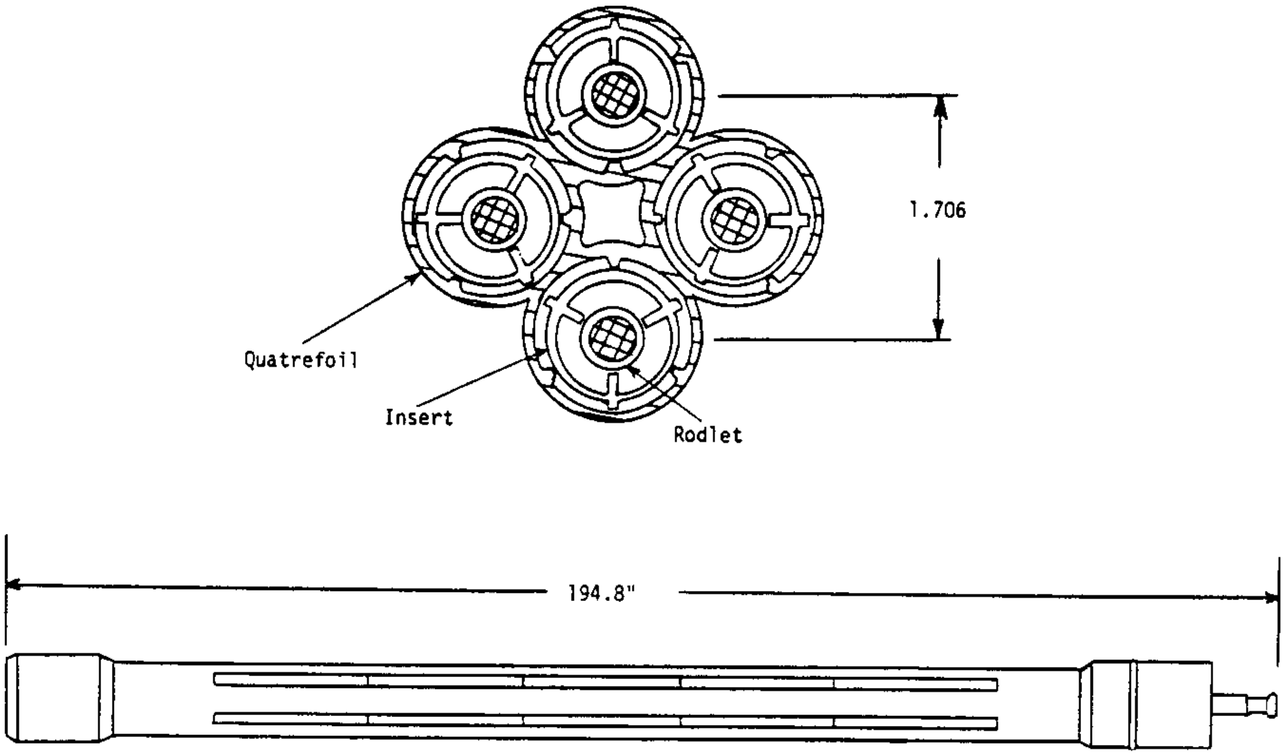


FIGURE 1. SRP Quatrefoil Assembly

The Thorium Irradiation Program proceeded through the fabrication of the pellets and zircaloy rodlets at PNL, testing of a modified reactor assembly at SRL, and completion of the irradiation plan in P Reactor. The program was canceled prior to the irradiation phase. The following sections describe the work completed at PNL and SRL prior to the program termination.

**Fuel Types for Irradiation**

Five fuel types were chosen to be irradiated in this program (Table 1). All of the fuel pellets were prepared and loaded into zircaloy rodlets by PNL. The low enriched uranium (2.07% U-235) was prepared at Oak Ridge National Laboratory (ORNL). A portion of this material was prepared as $UO_2$ powder, and shipped to PNL for preparation of the 100% $UO_2$ pellets. The remainder of the low enriched material was transferred to ORNL as an acid-deficient uranyl nitrate solution, from which sol-gel microspheres were prepared. The microspheres were then transferred to PNL, where they were used to prepare "hybrid" ceramic pellets. The high enriched uranium oxide (approximately 11.3% U-235 was also prepared at Y-12 and then shipped to PNL, where it was used to prepare the 80%/20% $ThO_2/UO_2$ and the approximately 80%/20% $ThO_2/UO_2$ plus 0.25 CaO pellets.

The dissolution process for thorium oxide fuels utilizes fluoride ions (about 0.05M) to catalyze thorium dissolution. The fluoride ion increases the corrosion rate of the dissolution equipment, unless fluoride resistant materials are employed. Previous work at SRL indicated that the addition of MgO to vibratory-packed $ThO_2$ increased dissolution rate after irradiation.[5] However, more recent work at SRL[6] indicated that the addition of MgO in reactor-grade ceramic pellets did not significantly increase dissolution rate. Sample pellets containing about 1.4% CaO (fabricated by General Electric Co. of Canada) dissolved quite readily in nitric acid, without the need for fluoride catalyst. These thoria pellets containing CaO were not, however, of reactor grade quality, which was attributed to excess CaO that collected at the crystal grain boundaries. Electron microprobe analysis indicated that about 0.5% CaO formed solid solution in the thoria matrix, with the remaining 0.9% at grain boundaries.

To evaluate CaO as a dissolution aid in reactor grade ceramic pellets, PNL fabricated $ThO_2/UO_2$ pellets containing 0, 0.33, and 0.66% CaO. PNL found that the addition of CaO caused the sintered pellets to have a less uniform grain structure and grain size, with some fine cracking. The completed samples were considered "marginally close to commercial grade." Subsequent dissolution tests at SRL indicated that CaO improved the dissolution rate significantly.

PNL then demonstrated improved pellets containing CaO (using a new lot of uranium oxide feed material). Therefore, pellets containing CaO were included in the test matrix.

### Fabrication of the Pellets by PNL

Development of the procedures and techniques used by PNL to fabricate the fuel pellets for the Thoria Irradiation Program is described in detail in References 7, 8, and 9. The actual fabrication, characterization, inspection, and specifications for the fuel materials are described in detail in Reference 4. The materials and fabrication steps for each fuel type are summarized in Appendix A. Additional information about each lot of powder (or microspheres) used to prepare the various pellet types is given in Table 5 on page 20.

### SRP Irradiation Plan Details

The irradiation of $ThO_2/UO_2$ fuel types in P Reactor was to utilize a modified quatrefoil assembly (Figure 1). The five different types of ceramic pellets were loaded into zircaloy rodlets, and the rodlets were to be placed into aluminum inserts in the two quatrefoil assemblies as shown below:

| Assembly 1 | No. Columns | No. Rodlets |
|---|---|---|
| 100% $UO_2$ pellets | 1 | 3 |
| 80/20% $ThO_2/UO_2$ pellets | 1 | 3 |
| 100% $UO_2$ Hybrid pellets | 1 | 3 |
| 100% $ThO_2$ pellets | 1 | 3 |

| Assembly 2 | No. Columns | No. Rodlets |
|---|---|---|
| 100% $UO_2$ Pellets | 1 | 3 |
| 80/20% $ThO_2/UO_2$ pellets | 1 | 3 |
| 80/20% $ThO_2/UO_2$ + 0.25 CaO pellets | 1 | 3 |
| Aluminum blank | - | - |

The rodlets (each 30 in. long) were to be removed from the assemblies, and after cooling for approximately 2 months were to be transferred directly to the High Level Caves in SRL (30 in. is the maximum length that can be transferred through the High Level Caves entrance ports). Because of the possibility of contamination of the reactor or storage basin and loss of the ability to measure fission product off-gases, it would have been undesirable to cut longer rodlets to less than 30 in.

The enrichment of the uranium, the placement of the assemblies (in the buckled zone), and the reactor irradiation cycle (P-11-2) were balanced such that the average power experienced by the fuel materials would be 16 to 17 kW/ft. The power rating was designed to produce heat ratings in the SRP irradiation which would be comparable to heat ratings in a commercial LWR reactor.

The volumetric average temperature of $UO_2$ operating at peak commercial conditions was selected as the basis for relating the operating conditions of $UO_2$ in a commercial reactor and the $ThO_2/UO_2$ in Savannah River. Commercial LWR power reactors normally operate at a peak power rating of 14 kW/ft. However, if the $ThO_2/UO_2$ fuels were irradiated at a heat rating of 14 kW/ft in the SRP Reactor, the average volumetric temperature would have been significantly cooler, due to the lower reactor coolant temperatures. Therefore, in order to simulate the desired fuel temperature, the test materials must be operated at a power level close to 18 kW/ft.

In a commercial LWR, most fuel will not experience sufficiently high temperatures to cause crystalline restructuring, although some of the fuel may restructure, especially if the reactor undergoes a high temperature fluctuation. It was, therefore, desirable that the $ThO_2$ irradiation program be designed to prepare samples of both unrestructed and restructed fuels. The estimated restructuring temperatures are shown in Table 2.

**TABLE 2**

**Estimated Restructuring Temperatures for $ThO_2/UO_2$ Materials**

| Fuel Type | Crystal Restructuring | Estimated Temp, °C |
|---|---|---|
| $UO_2$ | Equiaxial Grain Growth | 1500 |
| | Elongated Grain Growth | 1800-1900 |
| | Melting Point | 2800 |
| $ThO_2/UO_2$(80/20%) | Equiaxial Grain Growth | 1800 |
| | Elongated Grain Growth | 2100-2200 |
| | Melting Point | 3300 |

The fuel centerline temperatures are a function of power and burnup, and these are shown in Table 3.

**TABLE 3**

| Heat Rating (kW/ft) | Centerline Fuel Temperatures, °C | |
|---|---|---|
| | Burnup (MWD/MT) | |
| | 0 | 10,000 |
| 18 | 2000 | 1800 |
| 16–17 | <2000 | 1600 |
| 14–16 | 1800 | 1400 |

The data in Tables 2 and 3 indicate that the $ThO_2/UO_2$ fuel will undergo initial restructuring at approximately 1800°C, which corresponds to 14 to 16 kW/ft at the start of the irradiation (zero burnup).

The heat rating of the fuel materials is significantly affected by the axial placement of the rodlets in the fuel column, due to the axial profile of the neutron flux. Therefore, the axial placement of the rodlets was designed so that at least half of one rod would not experience a nominal heat rating of greater than 16 kW/ft. This would provide irradiated fuel samples of each type which had not restructured, and would, therefore, be representative of the normal operating conditions in a commercial LWR reactor.

The axial placement design for the rodlets was specified, based on axial power distribution.[10] The bottom of the lowest rodlet was specified to be 26.5 in. below the fuel core mid-plane. This required a 52.2-in.-long spacer below the bottom rodlet to the bottom of the fuel element. Including the 2-in.-long spacers between the 30-in.-long rodlets, the placement of the 3 rodlets was to be:

| Rodlet No. | Distance from Fuel-Core Mid-Plane, Inches | |
|---|---|---|
| | Bottom | Top |
| 1 | −26.5 | + 3.5 |
| 2 | + 5.5 | +35.5 |
| 3 | +37.5 | +65.5 |

## Reactor Physics Calculations

The enrichment of U-235 in Uranium was calculated by D. S. Cramer of the SRL Nuclear Physics Division to be 11.3% for 80% $ThO_2$/20% $UO_2$ and 2.07% for 100% $UO_2$ material. The two quatrefoils would have gone into 2 out of 6 vacant assembly positions in the buckled zone starting in the P-11.2 subcycle. The total material in both positions would cause a small flux tilt across the reactor of 0.5%. The computed local power pertubation of -4% is within the uncertainties normally experienced with coolant flow zoning. The product displacement was conservatively estimated, based on total capture in the quatrefoils, with no credit given for neutrons produced by fission in the rodlets. Peak power densities at 18 to 20 kW/ft were estimated for the rodlets in the P-11 cycle, based on an evaluation of the operating history of an earlier P-9 cycle. Corrections were included for the effects that were to be introduced by a new automatic backup shutdown safety system. Estimates were also made for worst-case fluctuations which may occur in each subcycle. Maximum power densities would not exceed 20 kW/ft, even for worst-case power fluctuations. A careful study of axial power distributions showed that requirements for specific ranges of power densities would be met by distributing rodlets out axially from the reactor mid-plane. Two data libraries of averaged cross-sections for the quatrefoil assemblies were completed. These libraries were to be used in subsequent design code calculations, safety analyses, and accounting calculations for billing this special irradation program.

Calculations provided values for decay heat generation for discharge and shipping operations at various post-irradiation times. Concentrations of Kr-85, I-131, and total noble gases were calculated. Dissolution in the High Level Caves three months after shutdown would have contributed less than 3% of the 1979 guidelines to the SRL yearly release. This release is principally Kr-85 and would not be significantly lower following longer cooling. Current techniques for trapping I-131 (efficiency 99.999%) were assumed.[11]

**Power Peaking in Fuel Rods**

The fuel rods to be irradiated in the $ThO_2/UO_2$ program were to be contained in two quatrefoils in the buckled zone of a Mark 16-B 31 charge. Vertically, the rods would have consisted of three rod sections about 30 in. long. An air gap of about 2 in. and zirconium end plugs were to separate fissile material in the rod sections. Power peaking in pellets near ends of the rod sections would occur as a result of peaking of the thermal flux. A much smaller power peaking would occur in adjacent fuel assemblies. The magnitude of this peaking was estimated so that this effect can be accounted for in limits calculations.

A two-dimensional transport theory code (TWOTRAN) was used to compute the power peaking effects. Averaged over the end pellet of a rod section, the calculated power peaking was less than 6% relative to the vertical center of the rod section. The design coolant velocity provides a much larger margin against nucleate boiling than this. One to three pellets at the ends of rod sections might have been less suitable for chemical studies than the remainder of the material, but this was acceptable.

Power peaking in adjacent assemblies was calculated to be 0.4%. Because of limitations in the model, a double value of 0.8% was recommended for limits calculations.[12]

**Enriched Uranium for the Fuel Pellets**

The enriched uranium oxide powders were prepared at ORNL (Y-12) and transferred to PNL for fabrication into the ceramic pellets. Three types of enriched $UO_2$ were prepared by ORNL, as shown in Table 4.

**TABLE 4**

**Enriched Uranium Production**

| Material Description | % U-235 Goal | Analytical Results ORNL | Analytical Results PNL |
|---|---|---|---|
| $UO_2$ Powder for 80% $ThO_2$ 20% $UO_2$ pellets | 11.3 ± 0.2 | 11.44 | 11.38 |
| $UO_2$ powder for 100% $UO_2$ pellets | 2.07 ± 0.04 | 2.16 | 2.19 |
| $UO_2$ microspheres for hybrid pellets | 2.07 ± 0.04 | 2.16 | 2.09 |

Calculations showed that the low enriched material was acceptable for the proposed irradiation, even though the U-235 content was slightly higher than goal (2.16 vs. 2.07). The enriched uranium contained approximately 4% more U-235 than the goal, which would result in only a 3% increase in the nominal power in the reactor. By locating the columns in selected quatrefoil locations, the test assembly power could have been reduced slightly. Additionally, this increase (3%) was within the uncertainties of the predicted power levels. The isotopic analyses obtained by PNL are also shown in Table 4.[4]

**Modified Quatrefoil Assembly Tests**

The modified quatrefoil design to be used for the $ThO_2/UO_2$ irradiation called for placing three 30-in.-long zirconium rodlets (containing the thoria/urania pellets) inside a ribbed aluminum insert, which would then fit into the standard quatrefoil tube. The aluminum inserts required for the modified assembly were extruded at SRP.

Calculations indicated that coolant flow in the modified quatrefoil assembly would be sufficient to avoid nucleate boiling. A hydraulic test assembly was fabricated to measure the actual pressure drops and resulting flow in the quatrefoil assembly.

Nine zircaloy rodlets containing 100% $ThO_2$ ceramic pellets were fabricated by PNL to meet the same specifications as the rodlets to be irradiated. The prototype rodlets were inserted in a full-scale assembly, and a long-term hydraulic test was started in January 1980. Inspection of the rodlets and assembly detected no noticeable erosion, wear, or vibration damage[13,14] at the completion of the six-month flow test program. The coolant flow in the test facility was maintained at 110 gal/min, the same as that expected during the actual irradiation.

Two assemblies to be irradiated in P Reactor were completely fabricated. The design of the "spider", a device at the top of the assembly which aligns the tubes concentrically, was modified to prevent any upward displacement of the rodlets in the assembly.

The irradiation assemblies were tested for hydraulic characteristics, and the flow and temperature monitoring instrumentation and the reactor discharge procedure and discharge crane cooling flow were checked. All equipment and procedures were shown to meet specifications.[11]

**TABLE 5**

**Information on Fuel Powders Used for the Thoria Irradiation Program**

| Fuel Type | Description | Material | Vendor I.D. | PNL I.D. | Enrichment, % ORNL | Enrichment, % PNL |
|---|---|---|---|---|---|---|
| I | 100% $UO_2$ pellets | $UO_2$ powder | ORNL Lot No. 450806 | 2E-D-10 | 2.16 | 2.19 |
| II & III | 80/20% $ThO_2/UO_2$ pellets | $UO_2$ powder | Order No. BRM-2808, Isotope Order No. SS79-19-7 No ORNL Lot No. | 11E-D-1 | 11.44 | 11.38 |
| | | $ThO_2$ powder | PNL Purchase Order No. 22050 AF-T No Tenn. Nuclear Lot No. | TB-3* | | |
| IV | 100% $UO_2$ hybrid pellets | $UO_2$ microspheres | CGT-69-1-2, -3, -4, & -5 | E-D-11-H16 | 2.16 | 2.09 |
| | | | CGT-70-1, -2, -3, -4, & -8 | E-D-11-mix 3 | | |
| V | 100 % $ThO_2$ pellets | $ThO_2$ powder | See above | | | |

* Further information on $ThO_2$ powder, TB-3, can be found in Table 3 (p. 16) of "Fabrication of $ThO_2$ and $ThO_2/UO_2$ Pellets for Proliferation-Resistant Fuels", PNL-3210, October 1979.

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## APPENDIX A

## MATERIALS AND FABRICATION STEPS FOR FUEL MATERIALS PREPARED BY PACIFIC NORTHWEST LABORATORIES

**TYPE I: 100% $UO_2$, 2.19% U-235**

Material: $UO_2$ powder from ORNL

Press Feed Preparation:

Slugging — 4.3 ±0.1 g/cc
Granulation — 20 mesh; - 100 M fines reslugged and granulated
Lubricant Addition — 0.3% zinc stearate

Pellet Pressing: 5.25 ±0.5 g/cc

Sintering:

Ramp Rate — 100°C/hr to 700°C, 300°C/hr to 1700°C
Soak Time/Temperature — 8 hrs/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-50% $H_2$/20 CFH

Immersion Density: 94.46% TD

Resinter Density Change: + 0.4% of TD

APPENDIX A (CONTD)

TYPE II: 80% $ThO_2$/20% $UO_2$, 11.3% U-235

Materials: $UO_2$ powder from ORNL;
$ThO_2$ from Tennessee Nuclear Specialties, Jonesboro, TN

Preblending: 1 Kg per blend for 10 min.

Ball Milling: $ThO_2$ premilled 8 hr, each blend milled 8 hr.

Blending: All 1 Kg milled blends cross-blended

Press Feed Preparation:

Slugging — N/A
Granulation — N/A
Lubricant Addition — 0.3% zinc stearate

Pellet Pressing: 6.1 ±0.05 g/cc

Sintering:

Ramp Rate — 150°C/hr to 450°C; 300°C/hr to 1700°C
Soak Time/Temperature — 8 hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-4%$H_2$/20 CFH

Immersion Density - Five Pellet Average: 94.50% TD

Resinter Density Change - Five Pellet Average: + 0.3% of TD

APPENDIX A (CONTD)

**TYPE III: $ThO_2$-20% $UO_2$-0.25% CaO, 11.3% U-235**

Materials: $UO_2$ from ORNL;
$ThO_2$ from Tennessee Nuclear Specialties

Preblending: 1 kg per blend for 10 min

Ball Milling: $ThO_2$ premilling 8 hr; each blend milled 8 hr

Blending: All 1-kg milled blends cross-blended

Press Fed Preparation:

Slugging — N/A
Granulation — N/A
Lubricant Addition — 0.3% zinc stearate blended with powder

Pellet Pressing: 6.0 ±0.05 g/cc

Sintering:

Ramp Rate — 100°C/hr to 700°C; 300°C/hr to 1700°C
Soak Time/Temperature — 8 hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-4%$H_2$/20 CFH

Immersion Density - Five Pellet Average: 95.00% TD

Resinter Density Change - Five Pellet Average: + 0.4% of TD

APPENDIX A (CONTD)

**Type IV - 100% $UO_2$ Hybrid Pellets, 2.09% U-235**

**RODLET NO. 1**

Material: $UO_2$ sol-gel microspheres from ORNL, PNL Batch No. 16

Preblending: N/A*

Ball Milling: N/A

Blending: N/A

Press Feed Preparation: Calcined at 550°C for 16 hr in Ar-4%$H_2$/80 CFH.

Slugging — N/A
Granulation — N/A
Lubricant Addition — N/A - Die lubricated

Pellet Pressing: 4.85 ±0.05 g/cc

Sintering:

Ramp Rate — 150°C/hr to 450°C; 300°C to 1700°C
Soak Time/Temperature — 8hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-50%$H_2$/20 CFH

Immersion Density - Five Pellet Average: 94.69% TD

Resinter Density Change - Five Pellet Average: + 0.3% TD

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* Not applicable.

APPENDIX A (CONTD)

Type IV (Contd)

RODLETS NO. 2 and 3

Material: $UO_2$ sol-gel microspheres, PNL Batch No. Mix (3)

Preblending: N/A*

Ball Milling: N/A

Blending: N/A

Press Feed Preparation: Calcined at 550°C for 16 hr in Ar-4%$H_2$/80 CFH. Mix (3) is a blend of calcines H12:H15:H17 in a ratio of 43:28:29.

Slugging — N/A
Granulation — N/A
Lubricant Addition — N/A - Die lubricated

Pellet Pressing: 4.85 ±0.05 g/cc

Sintering:

Ramp Rate — 150°C/hr to 450°C; 300°C/hr to 1700°C
Soak Time/Temperature — 8hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-50%$H_2$ 20 CFH

Immersion Density - Five Pellet Average: 95.05% TD

Resinter Density Change - Five Pellet Average: + 0.4% TD

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* Not Applicable.

**APPENDIX A** (CONTD)

**Type V - 100% $ThO_2$ (Dummy Pellets) Used for Flow Tests at SRL**

Material: $ThO_2$ from Tennessee Nuclear Specialties

Preblending: N/A

Press Feed Preparation:

Slugging — N/A
Granulation — N/A
Lubricant Addition — 0.5 wt % Sterotex blended with powder

Pellet Pressing: 6.0 ±0.05 g/cc

Sintering:

Ramp Rate — 150°C/hr to 450°C; 300°C/hr to 1700°C
Soak Time/Temperature — 5 hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-4%$H_2$/20 CFH

Immersion Density - Five Pellet Average: 94.33% TD

Resinter Density Change - Five Pellet Average: N/A

**Type V: 100% $ThO_2$ Pellets for Irradiation Tests**

Material: $ThO_2$ from Tennessee Nuclear Specialties

Preblending: N/A

Ball Milling: 14 hr 1 kg per mill

Blending: All milled powder cross-blended

Press Feed Preparation:

Slugging — N/A
Granulation — N/A
Lubricant Addition — 0.6% Carbowax plus 0.2% zinc stearate blended with powder

Pellet Pressing: 6.0 ±0.05 g/cc

APPENDIX A (CONTD)

**Type V (Contd)**

Sintering:*

Ramp Rate — 150%C/hr to 450°C; 300°C/hr to 1700°C
Soak Time/Temperature — 8 hr/1700°C
Cool Rate — 400°C/hr
Atmosphere/Flow Rate — Ar-4%$H_2$ 20 CFH; bubbled through $H_2O$ to 900°C

Immersion Density - Five Pellet Average: 93.77% TD

Resinter Density Change - Five Pellet Average: +0.2% of TD

* Entire pellet batch resintered using same heating cycle and atmosphere conditions to improve thermal stability. After the first sintering, the immersion density average was 93.4% TD.