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IRRADIATION PERFORMANCE OF
THORIA-URANIA FUEL MATERIALS

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ABSTRACT

This report describes the work accomplished during the period October 1, 1964, to December 31, 1964, on the irradiation performance of thoria-urania fuel materials. The objectives of the program are as follows:

1. To establish, experimentally, particular physical properties of thoria-urania that are of interest to the reactor designer.
2. To present the physical property data in a form suitable for engineering application.

The thermal conductivity and gas retention and release characteristics of sol-gel thoria-urania fuel will be emphasized. Pelletized fuel will be examined to standardize the experimental equipment and to provide data for correlation with available information.

Equipment fabrication and initial operational trials constituted the major effort during this report period. The bench test equipment for initial examination of fuel permeability to fission gas was operated successfully at controlled temperatures using helium as the permeating gas. Sorbed gas release apparatus design was completed, and the system was fabricated. Several fuel specimens have been degassed, and the analytical equipment has been standardized. The unirradiated fuel thermal conductivity apparatus has been designed and partially fabricated. Capsule and specimen parts for the irradiated fuel thermal conductivity and fission gas release tasks are being fabricated. Hazards report calculations have been completed and are being integrated into the safety analysis of the irradiation experiments. The dynamic gas release experiment has been designed, and calculations for the hazards evaluation have been initiated.

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1. INTRODUCTION

Complete exploitation of the advantages of thorium as a fertile material in thermal and particular resonant spectrum reactors has been inhibited by the lack of quantitative physical property data for thoria and thoria-urania fuels. Thoria and thoria-urania solid solutions have not been investigated to the extent of pure urania. The meager information available has been deduced from extrapolation of urania data to thoria data or produced from the few isolated programs dealing with thoria itself. No reliable performance information on thoria-urania fuel is available, especially in the higher performance limits, and there is no knowledge of the material's ultimate performance limits.

This contract provides for a series of experiments directed toward establishing certain physical properties of thoria-urania fuel materials that are necessary for improved reactor core design. Gas content and gas release characteristics of thoria-urania fuel produced by the sol-gel method will be investigated to provide the core designer with insight into internal gas pressures to be expected during operation of this fuel under varying power conditions. Thermal conductivity during nuclear self-heating will be studied at power levels sufficient to raise central fuel temperatures to the melting point. The thermal conductivity experiments will be performed on sol-gel-produced powdered fuel and on pelletized thoria-urania to establish a degree of correlation to similar work carried out on urania pellets and to compare directly the performance capabilities of pellets and powdered fuel. The thermal conductivity data will enable the core designer to calculate internal fuel temperatures and temperature gradients under varying power conditions so that he can design for and predict thermal capabilities and core life more accurately.

The program is divided into six experimental tasks and an additional task providing supervision, reports, and technical support. The experimental tasks are as follows:

Task No.

Supporting Measurements

- 02 Gas Permeability of Irradiated Fuel
- 03 Sorbed Gas Release Determinations

Thermal Conductivity

- 04 Unirradiated Fuel Thermal Conductivity
- 05 Irradiated Fuel Thermal Conductivity

Gas Release

- 06 Dynamic Inpile Fission Gas Release Experiment
- 07 Static Gas Release Experiment

2. GAS PERMEABILITY OF THORIA-URANIA — TASK 02

(G. C. Robinson, Task Leader; A. C. Batten)

2.1. Task Objective

The principal objective of this task is to obtain information on the permeability of thoria-urania to fission product gases at both room and elevated temperatures. Permeability of sintered pellets and sol-gel powder thoria-urania will be examined, as well as fuel that has been structurally altered by high temperature exposure.

2.2. Task Description

Fuel rods are normally designed with a void volume in which gases released from the fuel can accumulate. The rate at which locally released gases permeate to expansion voids is of interest in predicting gas movement and pressure buildup within fuel rods. The accuracy of fission gas analysis of irradiated fuel specimens depends on the complete removal of all gases within the open pores of the fuel. Fission gases have been observed to emanate from fuel specimens long after the analysis has been completed, indicating that not all of the gas was initially released to the sampling system.

During core operation it is anticipated that thermal excursions will cause variations in fission gas pressures, resulting in large variations of driving force for movement of the gas through the structure. The local pressure buildup due to a pulse of fission gases released to open porosity depends on the capability of the fuel to dissipate these gases.

The initial program effort concerns the design and construction of an apparatus to test the gas permeability of thoria-urania fuel specimens. The magnitude of the variables involved will be determined by preliminary bench-scale tests on both pellet and vibratory compacted fuel. Information from these tests will then be used in designing an apparatus to make precision measurements for permeability calculations.

2.3. Work Accomplished During This Report Period

2.3.1. Summary

1. The bench test apparatus for low pressure measurements was assembled.
2. A vibratory compacted fuel specimen was prepared, and initial test runs on the low pressure apparatus were made.
3. Preparations for vapor deposition of copper onto the periphery of fuel pellets were begun.
4. The bench test apparatus for high pressure measurements is being assembled.

2.3.2. Bench Test Design

Since fuel pellets are pressed and sintered, the resultant structure contains less open porosity than is found in vibratory compacted fuel. As a result, it is anticipated that the permeability of these fuel types will differ greatly. It is necessary to establish a range of expected gas flow through both fuel types so that the proper metering and instrumentation can be provided on the final permeability apparatus. It is also necessary to establish a relationship between low and high pressure flow measurements to determine the need for a more complex apparatus. Simplified tests referred to as bench tests will be used to obtain information necessary for designing the final permeability apparatus.

2.3.2.1. Low Pressure Apparatus

Assembly of the bench test apparatus for low pressure measurements was completed. In this apparatus, flow is measured by determining the pressure rise in a known volume that has been previously evacuated. Such a method is very sensitive to low flow rates. Figure 2-1 shows the assembled low pressure apparatus, and a schematic diagram of the apparatus is shown in Figure 2-2. The apparatus is operated in the following manner: after the specimen is attached to the apparatus with bite-type pressure fittings, the chambers on both sides of the specimen are evacuated to a pressure of approximately 5 microns of mercury. The valves on the vacuum pump line are then shut off, and helium gas is admitted into chamber A. The desired constant gas pressure in chamber A, as measured by the manometer, is regulated

by the metering valve. The gas in chamber A passes through the fuel specimen into the evacuated chamber B. The rate of pressure rise in chamber B, measured by the absolute pressure gage, determines the flow rate of the gas passing through the fuel specimen.

For elevated temperature studies, the fuel in the specimen is heated by a heater cord wrapped around the fuel tube. A thermocouple on the surface of the tube determines equilibrium temperature conditions.

2.3.3. Test Results

Figure 2-3 shows results of 45 initial test runs with helium gas on a vibratory compacted UO_2 fuel specimen in the low pressure apparatus. Except for the three data points shown, all other values of permeability K fell within the straight line envelope shown. The permeability coefficient K was calculated from the relationship

$$K = \frac{(\text{flow rate})}{(\Delta \text{pressure}) (\text{area/length})}$$

where all the factors were experimentally measured. (The theoretical considerations involved in permeability measurements are given in BAW-3376-1, Appendix A.) The measured gas flow varied from 0.016 to 0.074 $\text{cm}^3\text{-atm/sec}$ for pressure drops of 20 - 116 cm mercury respectively. The data were obtained over a $\bar{p}$ range of 0.540 - 1.27 atmospheres, where $\bar{p}$ is the mean pressure of the gas in the fuel. The straight line permeability relationship indicates that accurate gas flows were measured with the apparatus.

Further tests runs will be made with the $\bar{p}$ range extended to ten atmospheres. This will provide sufficient data overlap between the high and low pressure apparatus to permit comparison of the two flow measurement techniques.

2.3.4. Specimen Design

The vibratory compacted fuel specimen consisted of a half-inch OD tube approximately 30 inches long with fuel located at the center 10 inches. The fuel powder was held in place by microporous stainless steel filter discs (20 micron porosity) supported by an internal sleeve extending to each end of the tube, where the sleeve and tube were welded. The tube was attached to the apparatus by bite-type

pressure fittings. To prevent plugging the filter disc with fine particles of fuel during vibratory compaction, a solid plug was used while the specimen was vibrated. The plug was then removed, and the filter disc was placed in position without disturbing the compacted fuel. As a further precaution, after the test runs are completed, the downstream filter disc will be removed and examined for evidence of partial plugging from imposed high gas pressures.

For vibratory compacted fuel rods, it is assumed that the fuel powder is tightly compacted against the tube walls to effect a seal. To evaluate the effectiveness of the seal, particularly at room temperature, test runs will be made at various elevated temperatures up to 1000 F. Zircaloy-2 was chosen for the specimen tubing because this material has a coefficient of expansion less than that of the fuel. At elevated temperatures, the seal at the tube wall will not relax, but could become tighter if the room temperature seal was not complete. Since specific permeability coefficients B_0 and K_0 are constant characteristics of the porous medium, any change in these coefficients as a result of temperature can be construed to be a change in the effective seal at the tube wall, or a change in the structure.

Pellet fuel specimens will consist of a fuel pellet brazed to the wall of Zircaloy-2 tubing. Prior to brazing, the peripheral surface of the pellet will be coated with copper or other suitable metal which can be brazed. The brazed joint will provide a seal at both room and elevated temperatures, since the joint will be placed into compression as the temperature is raised. Equipment is presently being prepared for vapor-depositing copper onto the periphery of UO_2 fuel pellets.

2.4. Planned Work for Next Report Period

During the next report period, pellet fuel specimens will be prepared, and assembly of the high pressure apparatus will be completed. Test runs on both apparatus with both types of fuel specimens will be completed. In addition, an attempt will be made to determine if thermal transpiration (thermomolecular pressure difference) occurs in the fuel specimen under conditions of a thermal gradient along the length of the fuel.

Figure 2-1. Low Pressure Gas Permeability Apparatus,
Assembled (WAS-65-415)

Figure 2-2. Low Pressure Gas Permeability Apparatus

Figure 2-3. Helium Permeability of Vibratory Compacted UO_2 Fuel

3. SORBED GAS RELEASE DETERMINATIONS — TASK 03 (C. J. Halva, Task Leader; J. W. Ewing, R. H. Dodd)

3.1. Task Objective

The objective of this task is the qualitative and quantitative determination of absorbed and adsorbed gases released from unirradiated sol-gel fuel as a function of temperature.

3.2. Task Description

Samples of sol-gel fuels of varying composition, size, and preparation history will be pyrolyzed at different temperatures under vacuum. The amount of sorbed gases evolved will be measured and collected; the collected gases will be analyzed using gas chromatographic and mass spectrometric methods. The amount and nature of these gases will be studied to determine whether changes in production methods are necessary to improve sol-gel thorium. The experimental approach to this task is described in report BAW-3376-1.

3.3. Work Accomplished During This Report Period

3.3.1. Summary

No major modifications have been made in the experimental approach or design of the program. Fuel production has been delayed because of difficulties in obtaining a uniform final product. The fuel should be produced satisfactorily during the next report period, however. Some changes in the vacuum extraction system have been made, and additional modifications are planned to upgrade the system. The gas chromatograph has been set up and calibrated for all gases except H₂O and CO. The mass spectrometer modifications to accept gaseous samples have been completed.

3.3.2. Progress on Sorbed Gas Studies

3.3.2.1. Sol-Gel Fuel Production

The production of sol-gel fuel for this program has been delayed by difficulties in the setup of the pilot plant. Lack of uniformity in the compositions of sol, gel, and oxide was the major problem; present indications are that this has been resolved by careful adjustment of the sol-gel formation conditions. The last several batches of fuel produced have had uniform composition between the various phases (sol, gel, and oxide).

Some minor modifications have been made in the requested U/Th ratios of the sol-gel oxide. The following runs are now scheduled for production at 10 kg of fuel per batch.

<u>Run No.</u>	<u>U/Th ratio of fuel, %</u>
1	1.00
2	2.00
3	3.00
4	4.00
5	5.00
6	7.50
7	Maximum

These are approximate compositions, and a variability of $\pm 10\%$ in the final ratios will be acceptable. Natural uranium will be used in these fuels.

3.3.2.2. Analytical Methods

The analytical methods for sol-gel fuel discussed in the previous quarterly report are now in routine use. Uranium content is determined by coulometric methods, thorium is determined volumetrically by ethylene diamine tetra-acetic acid, and colorimetric techniques are used to find nitrate content.

Triplicate samples are taken of the sol, gel, and oxide. The sol samples are taken directly from the blending tank, and the gel samples are obtained by riffing techniques, which are also used to select oxide samples.

Triplicate determinations are made for thorium and nitrate (when requested) on all samples, and duplicate determinations are made for uranium. The average error for thorium and uranium, based on the precision of the analytical results, is approximately $\pm 1\%$.

3.3.2.3. Vacuum Extraction System

Some modifications have been made in the vacuum extraction system. A quartz-enclosed carbon crucible is now used as a succesor and sample receptacle.

A platinum-rhodium thermocouple has been installed in the system. The temperatures read earlier with the optical pyrometer were 150 to 200 C low. This error in reading was probably due to absorption of the emitted light by the relatively thick walls of the Vycor heating chamber.

A thermocouple gauge has been installed in the system to give a continuous pressure reading. This gauge supplements the McLeod gauge, and is not intended to replace it, since the accuracy of the electronic device decreases at high pressures.

The glass lines leading to and from the Toepler pump are being replaced by more durable stainless steel lines. With this final modification completed, the vacuum extraction system will be ready for use.

3.3.2.4. Setup and Calibration of the Gas Chromatograph

A Barber-Colman series 5000 selecta system gas chromatograph has been set up, and its calibration is almost complete. Basically, the chromatograph consists of the components shown in block form in Figure 3-1. The carrier gas selected is commercial grade helium, because nitrogen, argon, and hydrogen are possible components of the sorbed gases in sol-gel fuel. A cold trap filled with liquid nitrogen was installed on the line to remove most of the impurities from the helium.

Calibration samples are placed into the injector (Figure 3-1) with a syringe. Samples of unknown gas collected with the vacuum extraction system will be injected directly from the sample collection bottle. Two columns, containing silica gel and a molecular sieve,

are located in the constant temperature bath. These two columns are necessary to separate the gases expected as parts of the fuel. Hydrogen, CO_2 , and H_2O are collected on the silica gel columns; CH_4 , nitrogen, argon, oxygen, and CO are trapped on the molecular sieve. The temperature bath containing the columns can be varied from -65°C to $+400^\circ\text{C}$. This versatility in temperature control enables the analyst to separate and identify the resultant chromatogram peaks more easily.

The detector is a thermal-conductivity cell which is also thermally controlled; it is a six-port unit with tungsten filaments for sensing devices. The eluted gases are finally exhausted through the last part to the atmosphere (Figure 3-2).

The signal received from the thermal conductivity cell is fed directly to the chart recorder, the response of which is variable from 1 to 5 millivolts with a 1 or 2-second cross chart pen speed. The recorder has a built-in Zener diode standardization completely separate from the input signal.

Calibration chromatograms of oxygen, nitrogen, argon, methane, CO_2 , and hydrogen have been made. As mentioned previously, the calibration samples are injected into the system with a syringe. During these experiments, gas samples were injected at various column temperatures while maintaining constant carrier gas flow rate and detector and injector temperatures. Early in the calibration runs, it was found that the best resolution occurred at sub-ambient column temperatures with the optimum temperature being between -25°C and -65°C . These subambient temperatures are achieved by using liquid carbon dioxide in the constant temperature bath.

The following tabulation summarizes the preliminary calibration results for the various gases. The values determined in this table were the T_i value (time after injection for the sample to appear on the chromatogram) and the Δt value (defined as time during which the sample gas is eluted from the column). Both these values are recorded in millimeters, but may be converted to seconds since the speed of the chart is known. Values for Δt are taken at the half-peak height location. (The value at the base line can be chosen; either method is satisfactory, providing the analyst knows which is used.) The following constants apply for all data in the tabulation:

Carrier gas:	helium
Flow rate of carrier gas	75 cc/minute
Injection temperature	200 C
Detector temperature	200 C
Chart speed	1/2 inch-minute

<u>Gas</u>	<u>Temperature, C</u>	<u>Ti, mm</u>	<u>Δt, mm</u>
Oxygen	-65	44.3	2.4
Nitrogen	-65	146.5	12.0
Argon	-65	40.8	2.5
CH ₄	-65	310.8	25.6
Oxygen	-50	34.8	1.5
Nitrogen	-50	83.3	5.5
Argon	-50	34.3	1.5
CH ₄	-50	147.0	10.0
CH ₄	-25	75	3.8
CH ₄	0	46.2	2.3
CH ₄	25	34.3	1.5
CO ₂	-5	47.3	19.0
Hydrogen	-65	21.3	2.0

Figure 3-3 illustrates graphically the effects of temperature on Ti and Δt values for methane. As predicted by theory, lowering the temperature increases the separation of the various gases, but also results in a broadening of the peaks.

Calibration values for CO and H₂O remain to be determined. Once these values are established, composite gas samples will be run. If no difficulties arise, the instrument will then be ready to analyze gas samples collected from sol-gel fuel. If other gases are found in the sol-gel, those peaks must also be calibrated. Figure 3-4 shows the gas chromatograph system.

3.3.2.5. Modification of Mass Spectrometer

The mass spectrometer has been modified to implement its use for gas analysis. Figure 3-5 diagrams the main component of the instrument after modification, and Figure 3-6 shows the completed unit. The mass spectrometer can now be used either for solids isotopic analyses (by thermal ionization) or for gas analysis.

Changeover requires only the time required to strip, clean, and modify the ion source and the consequent pumpdown. Vacuum and electrical connections can be changed in a few minutes. The complete gas inlet system is easily disassembled for physical and chemical cleaning. The entire changeover period takes 12 hours, barring complications.

3.3.2.6. Some Theoretical Considerations on Sorbed Gases

Sorbed gases in sol-gel fuels are probably present in one of three forms: physical entrapment by the fuel, adsorption, or absorption. Physical entrapment of gases is influenced by process history, density, and particle size of the fuel. The quantity of gas held in this form varies inversely as the density of the fuel, and directly with particle size. Smaller sized particles of any one batch have less gas encapsulated in voids than do larger particles, since processes obtaining the smaller fragments also release the entrapped gases in the larger voids. These assumptions are correct only if uniform (size and number) distribution of voids throughout the fuel is assumed. Gases held by entrapment in voids would be highly process-dependent; once all the production variables of the sol-gel process are fixed, the content will be uniform. Volatilization of impurities in the fuel (carrying through the entire sol-gel process) produces gases resembling those physically entrapped. Gases produced in this way might include organic compounds, borates, sulfates, and other volatile inorganic compounds vaporized during the firing process and physically contained by the fuel.

Gases may become physically attached to the fuel by adsorption processes. Certain definite adsorptive potentials have been postulated by Ross and Olivior¹. These potentials are caused by (1) London-type dispersion forces resulting from induced-dipole/induced-dipole and multipolar reactions, (2) induction forces brought about by operation of a surface electrical field on induced or permanent dipoles of resident molecules, (3) charge transfer between the adsorbed molecule and the adsorbent and surface atoms, and (4) dative bonding resulting from a chemical reaction between the adsorbate and surface atoms. Of these four, processes 1, 2, and 3 can be called physical, or van der Waals', adsorption, in which free valencies are not satisfied and the surface energy is not reduced to its lowest state². The dative

type bonding (commonly referred to as chemisorption) results from a chemical reaction between the adsorbate and surface atoms. This type of adsorption can satisfy the valency requirements and can reduce the surface energy—truly a chemical bond.

Physical adsorption plays the major role in the case of sol-gel fuels. Since both fired ThO_2 and UO_2 in solid solution are relatively stable chemically, chemisorption is expected only if impurities are present in the system. Chemisorption is important in steps prior to the actual oxide fuel, however. For instance, it is well known that CO_2 is rapidly absorbed by sol solutions. Whether this is true absorption, chemisorption, or both, remains to be determined. Chemisorption could cause the changes that occur in physical characteristics of gels exposed to CO_2 .

Bond² has listed another property which must be considered: the nature of the gas adsorbed. Generally, the sequence for adsorption is $\text{O}_2 > \text{C}_2\text{H}_2 > \text{C}_2\text{H}_4 > \text{CO} > \text{H}_2 > \text{CO}_2 > \text{N}_2$. It is possible to replace a gas in the sequence with one that is more strongly adsorbed. The exact sequence is not known for the complex sol-gel oxide system. Pressure, temperature, and gas composition phases are important in determining the amounts of various sorbed gases. The complex relationships of these variables are covered by Bond².

The third mechanism by which gases may be attached to fuels is absorption. Absorption is the process by which gaseous molecules penetrate the surface and occupy spaces in the lattice of the bulk material. Here again the process is probably very limited in the oxide fuel itself since most absorbed gases would be removed during initial evacuation of the vacuum extraction apparatus. Absorption would probably play important roles in the sol and gel steps described under chemisorption.

Invariably, all three types and mechanisms are important contributors to sorbed gas release. It would be almost impossible to assign values to the significance of each of the mechanisms without a specific experimental program directed toward such an objective.

3.4. Planned Work for Next Report Period

Sol-gel fuel of varying U/Th ratios will be supplied and subjected to chemical and sieve analyses and to density determinations. Some samples of these fuels will be pyrolyzed under varying conditions, and the eluted gases will be run on the chromatograph. The mass spectrometer will be calibrated, and some gas analyses may be made.

Figure 3-1. Gas Chromatograph System

Figure 3-2. Thermal Conductivity Detector

Figure 3-3. Effect of Column Temperature on Ti and Δt Values for CH_4

Figure 3-4. Gas Chromatograph With Pyrolysis Attachment

Figure 3-5. Mass Spectrometer Modified Gas Source System

Figure 3-6. Mass Spectrometer With Gas Source Attachment

4. UNIRRADIATED FUEL THERMAL CONDUCTIVITY —

TASK 04

(G. C. Robinson, Task Leader; J. M. Kerr, E. N. Harbinson)

4.1. Task Objective

The primary objective of this task is to establish a correlation between fuel structure and temperature in thoria-urania. This correlation will be used in estimating the operating temperatures of the fuel in the irradiated specimens of Tasks 05, 06, and 07. The secondary objective is the determination of the thermal conductivity of unirradiated thoria-urania to about 2500 C. These data are expected to provide a useful comparison with the in-reactor thermal conductivity information obtained in Task 05.

4.2. Task Description

In Task 04, an apparatus will be constructed which will be capable of (1) establishing microstructures, similar to structures obtained in-pile, in both pellet and compacted powder fuel specimens, (2) measuring temperatures associated with structural changes of interest, and (3) performing reliable and accurate calorimetry for thermal conductivity determinations. Initially, existing related technology will be studied in an effort to establish a basis for the conceptual design of the apparatus and to examine the experimental factors influencing structural changes.

The initial tests will be made on UO_2 pellets, so that the performance of the apparatus can be correlated with existing UO_2 structure/temperature correlations and thermal conductivity. After establishing the reliability and peculiarities of the equipment and completing the UO_2 tests, investigations on $\text{ThO}_2\text{-UO}_2$ will be conducted. Some of these specimens will be fabricated from the same batch of fuel, at the same time as the fuel for the irradiation specimens is fabricated, so that the effects of material and fabrication variables will be eliminated, or at least, minimized. Study of at least two U/Th ratios is planned.

4.3. Work Accomplished During This Report Period

4.3.1. Summary

During this report period the design of the thermal simulation device was completed and drawings were issued for construction. Fabrication of the device is approximately 50 percent complete. The procurement of materials and equipment has been nearly completed.

4.3.2. Apparatus Design and Fabrication

The expected temperature profile across a specimen in the thermal simulation device was determined and is shown as curve 1 of Figure 4-1. Curve 2 shows a typical temperature profile expected in the pellet irradiation specimens of the irradiated thermal conductivity task. Comparison of the two curves in the temperature region of interest, i.e., 1900 to 2700 C, reveals that the thermal gradients in the outpile and inpile tests are about the same. Since columnar grain growth and grain growth rate are dependent on the thermal gradient, it is felt that a better correlation can be obtained between the fuel structures formed inpile and outpile if the respective thermal gradients are similar. This seems particularly important when low thermal gradients are present.

The temperature profile in a typical operating fuel rod is steeper than that expected in the present experiments as shown in Figure 4-1. Because void migration rates are highly temperature-dependent, the outer radius of columnar grain growth is sharply defined in a power reactor fuel rod. In specimens with lower thermal gradients, the zones of delineation between microstructures may not be as well defined.

Based on the analyses reported previously³, the detailed design of the thermal simulation device was completed and construction drawings were issued; the assembly drawing is shown in Figure 4-2. The device uses a central, tubular tungsten heater held in a vertical position by water-cooled clamps, detailed in Figure 4-3. The fuel is placed annularly around the heater with a radial gap of 0.060 inch. This arrangement should allow maximum heater temperatures with the minimum possibility of heater-fuel interactions. The cross sectional area of the expansion joint strips was determined to be that area

necessary to maintain power losses in the expansion joint to less than 1 percent and to conform to the available space. Thermal expansion of the tungsten heater causes deflection of the expansion joint, and the resisting force places an axial compressive load on the heater. Since the strength of the tungsten is quite low at operating temperatures, it is desirable to keep the resulting force to a minimum. It was determined by calculations that maximum expansion exerts a stress of 80 psi in the heater; available information indicates that the tungsten heater should be able to withstand this stress without buckling.

Fabrication of the thermal simulation device is about 50 percent complete, as shown in Figure 4-4. The micro-optical pyrometer traverses vertically in a fixed plane relative to the device. The power leads penetrate the top and bottom of the device, and the vacuum line and the main water coolant lines come into the device from the rear. The surface of the specimen cladding is cooled with demineralized water in a closed heat exchange loop. The heat exchanger is the large white drum located to the lower left of the device itself. The inert gas cleanup train is located to the rear and includes a cold trap and a zirconium chip furnace. Coolant water for electrical feedthroughs and chill plates is supplied from the right side of the device and is once-through tap water. The power supply is to be located remotely with controls in a relay rack which will be positioned to the right of the device. In order to correlate optical temperature measurements, tungsten-rhenium thermocouples are being procured, and an attempt will be made to place thermocouples on the ID of the electrode opposite a sight pipe location. Additionally, fine wire thermocouples will be passed into the sight wells to further verify the optical measurements. These thermocouples will be used on selected specimens to calibrate the device and the optical pyrometry.

Procurement is approximately 95 percent complete; fuel and insulator pellets are outstanding.

4.4. Planned Work for Next Report Period

In the next report period we plan to complete fabrication of the thermal simulation device and to initiate calibration tests.

Figure 4-1. Thermal Profiles of Typical Operating Fuel Rod, Outpile Test Specimen, and Inpile Irradiation Specimen

Figure 4-2. Thermal Simulation Device,
Assembly Drawing
(NDC-1164 E)

BILL OF MATERIAL				
ITEM	DWG. NO.	DESCRIPTION	QTY.	MAT'L.
1	NDC-1164 E	ASSEMBLY	—	—
2	NDC-1154 E	INERT GAS CHAMBER	1	AS NOTED
3	NDC-1152 E	UPPER FLANGE & CHILL PLATE	1	AS NOTED
4	NDC-1155 E	COOLING JACKET	1	AS NOTED
5	NDC-1156 B	TOP PLATE OF COOLING JACKET	1	AS NOTED
6	NDC-1164 B	SPECIMEN TUBE	1	AS NOTED
7	NDC-1161 D	ELECTRODE POSITIONER	2	AS NOTED
8	NDC-1164 E	4 x 8 x 1/4 THICK WINDOW	1	OPTICAL GLASS
9	NDC-1152 E	GLASS HOLD DOWN CLAMPS	20	AS NOTED
10	NDC-1164 E	1000 AMP WATER COOLED FEED THRU VACUUM TECH. INC. CAT. # 7554-218	2	COMM'L.
11	NDC-1164 E	ANKORITE O-RING # 50-276	1	COMM'L.
12	NDC-1164 E	ANKORITE O-RING # 50-262	1	COMM'L.
13	NDC-1164 E	ANKORITE O-RING # 50-250	1	COMM'L.
14	NDC-1164 E	3 SCH. 40 PIPE	AS REQD.	BRASS
15	NDC-1164 E	ANKORITE O-RING # 50-010	8	COMM'L.
16	NDC-1164 E	1/2 I.D. x 1/2 WALL TUBE	AS REQD.	TYGON
17	NDC-1164 E	3/8 I.D. x 1/2 WALL TUBE	AS REQD.	TYGON
18	NDC-1164 E	3/4 I.D. TUBE CLAMP	12	5.5.
19	NDC-1164 E	1/2 DIA. TUBE CLAMP	2	5.5.
20	NDC-1164 E	1/2 INCH HEX NUT	2	304 5.5.
21	NDC-1164 E	#4-20NC x 3/8 LONG SOCKET WD. CAP SCREW	20	304 5.5.
22	NDC-1164 E	#5-20NC x 1/2 LONG SOCKET WD. CAP SCREW	30	304 5.5.
23	NDC-1164 E	#10-32NC x 3/8 LONG SOCKET WD. CAP SCREW	10	304 5.5.
24	NDC-1164 E	#10-32NF x 1/2 LONG SOCKET WD. CAP SCREW	4	304 5.5.
25	NDC-1164 E	5/16-18NC x 3/8 LONG SOCKET WD. CAP SCREW	8	304 5.5.

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Figure 4-3. Electrode Clamp, Detail Drawing
(NDC-1161 D)

Figure 4-4. Thermal Simulation Apparatus Assembly
Layout (WAS-65-422)

5. IRRADIATED FUEL THERMAL CONDUCTIVITY — TASK 05

(W. R. DeBoskey, Task Leader; L. B. Gross)

5.1. Task Objective

The objective of this task is to determine the thermal conductivity of thoria-urania fuel to the melting point by in-reactor self-heating. The correlation of temperature with structural alterations determined in the unirradiated fuel thermal conductivity task (Task 04) will be necessary to establish the temperatures achieved at particular locations in the irradiated specimens of this task. Comparisons will be made between thermal conductivity values determined by the electrical heating method of Task 04 and by the in-reactor self-heating of this task.

5.2. Task Description

Utilizing nuclear self-heating, the "heat rating $\left(\int \lambda d\theta\right)$ versus temperature" relationship for $\text{ThO}_2\text{-UO}_2$ to the melting point of the oxide will be developed. Experiment parameters will be chosen to bracket the melting point (three specimens) and, if possible, to bracket columnar grain growth (two specimens). This task will be accomplished by selective predetermined loading of four in-reactor capsules containing 20 fuel rod specimens as follows:

1. Five specimens of nominal 5 wt% UO_2 in ThO_2 , pellets.
2. Five specimens of nominal 5 wt% UO_2 in ThO_2 , vibratory compacted sol-gel powder.
3. Five specimens of nominal 10 wt% UO_2 in ThO_2 , pellets.
4. Five specimens of nominal 10 wt% UO_2 in ThO_2 , vibratory compacted sol-gel powder.

Properties such as density and isotopic composition will be initially determined for all fuel batches. Sources and batches of specimen

material will be the same as those used in the gas release experiments of Tasks 06 and 07.

Individual capsule irradiation will be of short duration (approximately three or four days), during which time neutron flux will be maintained as constant as practicable. The reactor space and capsule equipment console will be the same as those used for Task 07 capsules. Capsule instrumentation will be neutron monitors and thermocouples where practicable.

Post-irradiation examination of the 20 specimens will determine the relationship between temperature and heat rating $\left(\int \lambda d\theta\right)$ for bulk and powder materials of the two urania compositions investigated. Analysis of characterization and operating data, as well as radiochemical, isotopic, and ceramographic examinations will be performed. Assigning the temperature profile to the fuel specimen is a prerequisite achieved by noting the locations of various temperature-associated structural changes (such as melting) and changes in grain morphology.

Differentiation of the heat rating integral $\left(\int \lambda d\theta\right)$ with respect to temperature will yield thermal conductivity values as a function of temperature. Thermal data obtained from pellets will be utilized to establish the general shape and magnitude of $\int \lambda d\theta$ and thermal conductivity versus temperature curves. Application of sol-gel experimental data to the general curve will lead to specific knowledge of the thermal conductivity of sol-gel powder.

5.3. Work Accomplished During This Report Period

5.3.1. Summary

On the basis of reactor operational data and computer calculations, the neutron flux level expected in this experiment has been ascertained, and the fissile atom enrichments for the specimens have been determined. Continued evaluation of the expected specimen operating conditions has provided specific fuel temperature gradient information and has indicated the degree of coring necessary to compensate for center melting. Calculations of the expected operating conditions and engineering drawings of the experimental capsule have been completed and will be issued as a report for hazards evaluation. Procurement of capsule materials has been started.

5.3.2. Capsule Design

The neutron flux level in the large corner beryllium reflector hole RL-2 is axially distributed as follows when X is given in inches from midplane:

$$\phi = 6.94 \cos [0.0868(X - 0.665)] \times 10^{13} \text{ nv} \quad (1)$$

positive values of X being downward. Fluxes, so expressed, are 2200 meters/second values for a hole diameter of 1.812 inches. For this experiment, however, a larger hole is required, and an aluminum corner piece was manufactured for this purpose. The flux level in this new corner piece was determined relative to equation (1) by computer calculation. The new value is 0.542 of the equation (1) value for both situations water-filled; however, the content of the adjacent small holes affects the flux level in the large hole. The minimum neutron flux perturbation in the holes is 1.41 times the values obtained from equation (1).

The Bradley-Lewis correlation was used to calculate the perturbation of the neutron flux caused by introduction of the experiment into the irradiation site. The average neutron flux value in the fuel so determined, in conjunction with the episcadmium neutron flux contribution, was utilized to determine the power generation of various U-235 enrichments of the two urania-thoria compositions. These results are plotted in Figures 5-1 and 5-2; the particular enrichments required are summarized in Table 5-1.

Table 5-1. U-235 Loadings Required for the Experiment

Type	Enrichment	$\frac{W/F \text{ 235}}{U}$	
		5 wt% UO_2	10 wt% UO_2
Pellet	E - 1	0.143	0.087
	E - 2	0.100	0.060
Powder	E - 1	0.121	0.073
	E - 2	0.082	0.048
	E - 3	0.069	0.041
	E - 4	0.044	0.025

The radial temperature distributions in the fuel for the midplane-of-specimen power levels were calculated, and the results are graphically presented in Figures 5-3 and 5-4. Those pellet specimens which might experience center-melting will be cored to accommodate the volumetric increase on melting. Powder specimens possess sufficient free volume to accommodate the molten phase without coring.

5.3.3. Documentation

Engineering drawings of the experimental capsule have been completed; the general assembly drawing is presented as Figure 5-5. The drawings in this series are:

- NDC 1147F - Capsule for TUIP (Figure 5-5)
- NDC 1148E - Details of Specimens for TUIP
- NDC 1149E - Details of Capsule for TUIP
- NDC 1160E - Aluminum Lead Tube for TUIP

The draft report of calculations for the hazards analysis has been completed.⁴

5.3.4. Procurement

Procurement of materials for experiment construction has commenced. Purchase orders have been issued for stainless steel capsule tubing and miscellaneous hardware. Quotations have been received for pellet fuel, and a vendor was selected. A purchase order will be issued on receipt of authorization from the AEC. Quotations are being solicited for the necessary thermocouple and neutron flux instrumentation and ancillary equipment.

5.4. Planned Work for Next Report Period

The report of calculations for hazards analysis will be issued, and the necessary modifications will be performed in accordance with directives from the BAWTR Safeguards Committee. Construction of the experiment will commence.

Figure 5-1. Total Pellet Specific Power Generation Vs
U-235 Loading

Figure 5-2. Total Powder Specific Power Generation Vs
U-235 Loading

Figure 5-3. Calculated Temperature Distribution in Task 05/07 Pellet Specimens

Figure 5-4. Calculated Temperature Distribution in Task 05/07 Powder Specimens

Figure 5-5. Capsule for Thoria-Urania
Irradiation Program,
Design Two (NDC-1147 F)

6. DYNAMIC INPILE FISSION GAS RELEASE
EXPERIMENT — TASK 06
(W. R. DeBoskey, Task Leader; L. B. Gross)

6.1. Task Objective

The objective of this task is to determine the internal gas pressure in a thoria-urania fuel rod as a function of time during reactor operation. The effect on fission gas release of initial and terminal operation at power sufficient to significantly alter the grain structure will be examined.

6.2. Task Description

The internal gas pressures of a series of ThO_2 - UO_2 fuel rods during reactor operation will be determined by in-reactor monitoring. Four capsules will be irradiated: two capsules to undergo a power burst at the beginning of exposure, and the remaining two undergoing a power burst at the end of exposure. Four specimens (one in each capsule) will have nominal 5 wt% UO_2 in ThO_2 in the following forms:

1. Pellet (initial burst).
2. Pellet (final burst).
3. Vibratory compacted sol-gel powder (initial burst).
4. Vibratory compacted sol-gel powder (final burst).

Properties, such as density and isotopic composition, of the specimen materials and operating characteristics of the detection systems will be determined. Specimen material sources and batches will be the same as those used for Tasks 05 and 07.

Capsules will be irradiated individually for approximately 60 days. Gas pressure and neutron flux will be recorded as a function of time. Power bursts will exceed operation to that degree required for extensive recrystallization or center melting.

Post-irradiation examinations will include analyses of pre-irradiation and operating data, gas sampling, radiochemistry, and ceramic examinations. Data obtained from pellet investigations will provide value limits for the powder data, which is difficult to analyze.

6.3. Work Accomplished During This Report Period

6.3.1. Summary

Work performed during this report period included establishment of the design concept and completion of thermal and hydraulic calculations for the pellet experiments for the hazards evaluation. A fuel pin simulating PWR configuration and conditions is to be encapsulated to render it compatible with the operating characteristics and license requirements of the BAWTR. Since the BAWTR is a new reactor and therefore not fully characterized, it is necessary to assume some operational characteristics, such as the magnitude and variation of neutron flux and coolant availability. The design establishes the fuel surface temperature at approximately that expected in a water-cooled reactor fuel element.

6.3.2. Experiment Design

The general approach to the capsule design for the 06 task utilizes the concepts and calculational methods developed previously for the second design of Task 05/07. This concept limits, rather than controls, the fuel surface temperature, as previously discussed³. The fuel rod to be tested will simulate a water-cooled reactor element configuration and conditions. Accommodation of this fuel rod (typically 0.42-inch diameter with 0.025-inch cladding thickness) in the large diameter (1.78-inch) capsule, necessary for effective heat transfer to the coolant, requires an annular spacer having high thermal conductivity. NaK-bonded OFHC copper was selected for this purpose, as shown in Figure 6-1. Thermocouple instrumentation will be placed in the copper to monitor the heat output of the fuel.

The 15-inch fuel rod will be irradiated in the upper half of the axial neutron flux cosine. The calculated temperature profile for the expected operating conditions is presented in Table 6-1. A more detailed temperature profile of the nominal case fuel is shown in Figure 6-2.

Table 6-1. Capsule Temperature and Heat Rating Summary for Pellet Fuel

Location	Nominal case		Burst case		Safeguard case	
	Bottom	Top	Bottom	Top	Bottom	Top
Outer capsule OD, F	189	156	195	158	252	189
Outer capsule ID, F	252	184	265	188	340	229
Inner capsule OD, F	473	309	501	320	608	391
Inner capsule ID, F	531	335	564	348	686	428
Copper OD, F	544	341	578	354	704	436
Copper ID, F	594	358	635	373	777	461
Cladding OD, F	623	367	668	383	818	473
Cladding ID, F	745	406	803	426	978	526
Fuel surface, F	915	453	996	479	1220	594
Fuel center, C	2300	510	2630	580	3210	780
$\int \lambda d\theta$, w/cm	44.2	11.5	50.4	13.1	63.0	16.4

6.3.3. Fuel Rod Pressure Distribution

The pressure distribution in the fuel rod is a complicated function of temperature distribution and free volume distribution. By assuming isobaric conditions throughout the free volume, an end plenum volume was selected to maintain the pressure (resultant from 100% fission gas release) at a level consistent with reasonable material strength. However, the presence of a temperature gradient in a porous material (such as the fuel) or in a capillary (such as the tube from the fuel rod to the pressure transducer) would very likely produce an imbalance of pressure within the total system, due to thermomolecular flow. An estimate of the pressure imbalance for temperatures and dimensions expected in the nominal case in this experiment is as follows:

I. Reference situation	- Isobaric case:	315 psi
II. With thermomolecular flow	- In fuel, at cosine peak:	450 psi
	- In end plenum:	275 psi
	- At measuring head:	225 psi

To determine the relative effects of thermomolecular flow in this system, pressure taps will be located at the temperature extremes of the

pellet rods. The possibility of powder plugging the tap at the lower location precludes its use for powder rods.

6.3.4. BAWTR Irradiation Facility

The capsules will be irradiated in the large corner beryllium reflector hole RL-1. The adjacent small holes, RS-1 and RS-2, will be used to produce the terminal power bursts. Movable neutron-absorbing stainless steel rods will be positioned in the small holes; removal of these rods from the flux field adjacent to the specimen will increase the neutron flux to the specimen, and thus simulate a power burst.

6.4. Planned Work for Next Report Period

The thermal and pressure distribution analyses for powder-filled fuel rods will be performed. Applicable calculations will be assembled into a report for hazards analyses. The pressure readout instrumentation and system will be designed, and a bench model will be tested.

Figure 6-1. Capsule Axial Section, Schematic Diagram

Figure 6-2. Temperature Gradient in Nominal Case Fuel, Pellet

7. STATIC GAS RELEASE EXPERIMENT — TASK 07 (W. R. DeBoskey, Task Leader; L. B. Gross)

7.1. Task Objective

The objective of this task is to obtain quantitative and qualitative information on gases released during irradiation of thorium-uranium fuel at varying heat ratings. Heat ratings for these specimens will be essentially identical to those of the thermal conductivity specimens of Task 05, so that some confirmation of thermal conductivity data may be possible, provided that the operating heat rating of the specimens can be maintained constant during the 60-day irradiation.

7.2. Task Description

The "gas release versus operating heat rating ($\int \lambda d\theta$)" relationship for ThO_2 - UO_2 fuel will be developed from post-irradiation sampling data. Experiment parameters will be chosen so that the operating heat rating varies up to that required for melting. Some duplication of experiments for improved statistics will be accomplished by loading five in-reactor capsules with 24 fuel rod specimens as follows:

1. Five specimens of nominal 5 wt% UO_2 in ThO_2 , pellets.
2. Six specimens of nominal 10 wt% UO_2 in ThO_2 , pellets.
3. Six specimens of nominal 5 wt% UO_2 in ThO_2 , vibratory compacted sol-gel powder.
4. Seven specimens of nominal 10 wt% UO_2 in ThO_2 , vibratory compacted sol-gel powder.

Properties of the specimen materials, such as density and isotopic composition, will be determined. Specimen material sources and batches will be the same as those used for Tasks 05 and 06.

Capsules will be irradiated individually for approximately 60 days, during which time a constant neutron flux will be attempted. The reactor

space and capsule equipment console will be the same as those used for Task 05. Capsule instrumentation will be neutron monitors, and thermocouples if practicable.

Post-irradiation examination of the 24 specimens will determine the relationship between gas release and heat rating for bulk and powder materials for the two urania compositions investigated. Analysis of characterization and operating data, gas sampling, and radiochemical, isotopic, and ceramographic examinations will be performed. All specimens will be examined specifically to corroborate Task 05 data, and samples will be available for post-irradiation gas permeability measurements under Task 02.

Gas release data obtained from pellet specimens will be used to establish the general shape and magnitude of the curve representing the relationship between gas release and heat rating. Application of data from sol-gel specimens to this general curve will lead to specific knowledge of the gas release properties of sol-gel fuel.

Capsule design for this task will be similar in all respects to the thermal conductivity capsules of Task 05, reflecting the inter-relationship of these two tasks. The principal experimental difference, other than the differences in analyses, is the irradiation time. The capsules of Task 05 will be irradiated for only three to four days—time for generation of sufficient fission products for precise fuel burnup analysis. The capsules of this task will be irradiated for approximately two months—the time required for buildup of sufficient fission gases for precise analysis. Although these specimens will be used to corroborate the thermal conductivity specimens, it should be noted that the longer operating time lessens the degree of flux control. From this point of view, the irradiation time of the thermal conductivity experiment should be as short as possible, in direct antithesis to the gas release experiment.

7.3. Work Accomplished During This Report Period

Since the capsules of this task are similar in all respects to the thermal conductivity experiment capsules of Task 05, the design documentation and procurement statements made for Task 05 will apply here.

7.4. Planned Work for Next Report Period.

The report of calculations for hazards analysis will be issued and the necessary modifications performed in accordance with directives from the BAWTR Safeguards Committee. Construction of the experiment will commence.

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ABSTRACT

This report describes the work accomplished during the period October 1, 1964, to December 31, 1964, on the irradiation performance of thorium-uranium fuel materials. The objectives of the program are as follows:

1. To establish, experimentally, particular physical properties of thorium-uranium that are of interest to the reactor designer.
2. To present the physical property data in a form suitable for engineering application.

The thermal conductivity and gas retention and release characteristics of sol-gel thorium-uranium fuel will be emphasized. Pelletized fuel will be examined to standardize the experimental equipment and to provide data for correlation with available information.

Equipment fabrication and initial operational trials constituted the major effort during this report period. The bench test equipment for initial examination of fuel permeability to fission gas was operated successfully at controlled temperatures using helium as the permeating gas. Sorbed gas release apparatus design was completed, and the system was fabricated. Several fuel specimens have been degassed, and the analytical equipment has been standardized. The unirradiated fuel thermal conductivity apparatus has been designed and partially fabricated. Capsule and specimen parts for the irradiated fuel thermal conductivity and fission gas release tasks are being fabricated. Hazards report calculations have been completed and are being integrated into the safety analysis of the irradiation experiments. The dynamic gas release experiment has been designed, and calculations for the hazards evaluation have been initiated.