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**DENSIFICATION RELATED PELLET
DIAMETER SHRINKAGE IN LOW
BURNUP THORIA-BASE FUELS**
(LWBR Development Program)

SEPTEMBER 1978

CONTRACT EY-76-C-11-0014

BETTIS ATOMIC POWER LABORATORY
WEST MIFFLIN, PENNSYLVANIA

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DENSIFICATION RELATED PELLET DIAMETER
SHRINKAGE IN LOW BURNUP THORIA-BASE FUELS
(LWBR Development Program)

G. L. Spahr

September 1978

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FOREWORD

The Shippingport Atomic Power Station located in Shippingport, Pennsylvania was the first large-scale, central-station nuclear power plant in the United States and the first plant of such size in the world operated solely to produce electric power. This program was started in 1953 to confirm the practical application of nuclear power for large-scale electric power generation. It has provided much of the technology being used for design and operation of the commercial, central-station nuclear power plants now in use.

Subsequent to development and successful operation of the Pressurized Water Reactor in the DOE-owned reactor plant at the Shippingport Atomic Power Station, the Atomic Energy Commission in 1965 undertook a research and development program to design and build a Light Water Breeder Reactor core for operation in the Shippingport Station.

The objective of the Light Water Breeder Reactor (LWBR) program has been to develop a technology that would significantly improve the utilization of the nation's nuclear fuel resources employing the well-established water reactor technology. To achieve this objective, work has been directed toward analysis, design, component tests, and fabrication of a water-cooled, thorium oxide fuel cycle breeder reactor for installation and operation at the Shippingport Station. The LWBR core started operation in the Shippingport Station in the Fall of 1977 and is expected to be operated for about 3 to 4 years. At the end of this period, the core will be removed and the spent fuel shipped to the Naval Reactors Expended Core Facility for a detailed examination to verify core performance including an evaluation of breeding characteristics.

In 1976, with fabrication of the Shippingport LWBR core nearing completion, the Energy Research and Development Administration established the Advanced Water Breeder Applications (AWBA) program to develop and disseminate technical information which would assist U. S. industry in evaluating the LWBR concept for commercial-scale applications. The program will explore some of the problems that would be faced by industry in adapting technology confirmed in the LWBR program. Information to be developed includes concepts for commercial-scale prebreeder cores which would produce uranium-233 for light water breeder cores while producing electric power, improvements for breeder cores based on the technology developed to fabricate and operate the Shippingport LWBR core, and other information and technology to aid in evaluating commercial-scale application of the LWBR concept.

FOREWORD (Cont)

All three development programs (Pressurized Water Reactor, Light Water Breeder Reactor, and Advanced Water Breeder Applications) have been administered by the Division of Naval Reactors with the goal of developing practical improvements in the utilization of nuclear fuel resources for generation of electrical energy using water-cooled nuclear reactors.

Technical information developed under the Shippingport, LWBR, and AWBA programs has been and will continue to be published in technical memoranda, one of which is this present report.

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In-reactor densification of ThO_2 and $\text{ThO}_2\text{-UO}_2$ fuel of low burnup and low power operation (hence low temperature) was assessed by measuring fuel pellet diameter changes. Pellet diameter changes ranged from nil in a large grain, low temperature thoria pellet (98.9 percent theoretical density) to -1.06 percent in a small grain, moderate temperature ThO_2 - 30 w/o UO_2 pellet (93.8 percent theoretical density). A correlation was established between quantity of small pores ($<2.3 \mu\text{m}$ diameter) and as-fabricated fuel grain size. An empirical equation, based on densification (pore closure) plus fuel swelling, was formulated for pellet diameter change as a function of initial grain size and fuel burnup. Predicted diameter changes were in good agreement with observed diameter changes in ThO_2 pellets, but the agreement with binary fuel was less satisfactory. It is concluded that densification of thoria-base fuels can occur at low burnup and low temperature and should be considered in performance assessments, even with high density fuel.

DENSIFICATION RELATED PELLET DIAMETER SHRINKAGE
IN LOW BURNUP THORIA-BASE FUELS
(LWBR Development Program)

G. L. Spahr

I. INTRODUCTION

In pressurized water reactors the nuclear core is generally a fuel rod system with sintered oxide fuel pellets, usually UO_2 -base oxide fuel. Typically, pellet fabrication procedures entail agglomeration of the oxide fuel powder with organic binders and lubricants, cold pressing to obtain green pellets, and sintering to obtain finished pellets of 90 percent or higher theoretical density. Completely dense (100 percent theoretical) pellets are not attained during fabrication.

The fuel rod system for the Light Water Breeder Reactor (LWBR) uses ThO_2 and $\text{ThO}_2\text{-UO}_2$ fuels in Zircaloy-4 cladding tubes. LWBR sintered fuel pellet densities are in the range of 95 - 99 percent of theoretical oxide densities. Densification of oxide pellets can occur in-reactor as the result of radiation-induced residual pore closure. As a consequence of in-reactor densification, fuel pellet dimensional shrinkage can occur resulting in fuel stack length shrinkage with possible formation of axial interpellet gaps potentially resulting

in cladding collapse into these gaps. An initial review of this problem in pressurized water reactors was reported (Reference 1) following which the densification phenomenon was the subject of considerable study (References 2 and 3).

In-reactor densification also can cause pellet diameter shrinkage which can lead to increased fuel-cladding gaps if the pellet shrinks in diameter faster than the cladding and/or to increased cladding diameter shrinkage for designs where the cladding is supported by the fuel. The case where the fuel-cladding gap is increased leads to higher than normal fuel temperature because of the poor thermal conductance of the gap. Because of higher fuel temperature the stored energy in the fuel is increased, resulting in higher cladding temperature and loss of cladding strength during postulated loss-of-coolant accident (LOCA) conditions. Increased cladding shrinkage can lead to reduced lateral support of the fuel rods.

This report presents an assessment of pellet diameter changes related to in-reactor densification of thorium and $\text{ThO}_2\text{-UO}_2$ at low power (hence, low temperature) and low burnup. Although fuel axial dimensional changes have been extensively investigated, radial dimensional changes which are important to design of rod mechanical support systems generally have not been as widely reported.

Experimental techniques are discussed briefly for ceramographic determination of fuel structure, grain size, and porosity in addition to presentation of pellet diameter measurements, uncertainty evaluations, and measurement error corrections. A correlation between fraction of small porosity ($<2.3 \mu\text{m}$ diameter) and fuel grain size was established from measurements made on as-fabricated unirradiated ThO_2 and $\text{ThO}_2\text{-UO}_2$ pellets.

An equation of exponential form, similar to one used to represent density changes in Reference (4), was developed with an added linear term for 1-dimensional fuel swelling to predict the pellet diameter changes. The exponential form of the equation was based on other radiation-induced densification studies (References 4 and 5) and a hot pressing model from Reference 6. It was assumed that the irradiation-assisted closure of small pores (arbitrarily chosen as $<2.3 \mu\text{m}$ diameter) was the predominate cause of in-pile fuel densification. To assume only 1 size category of active pores is a simplification of the probable fuel behavior, for which it has been suggested (Reference 7) that each pore size category tends to shrink at a characteristic rate. In support of the

simplifying assumption of 1 active pore size category, UO_2 fuel pellets with concentrations of small pores (1 - 3 μm diameter or less) have been found (References 4, 8, and 9) to densify much more readily under irradiation than pellets with predominantly larger pores (about 10 μm diameter or larger).

Experimental results derived primarily from pre- and post-irradiation measurements of pellet diameters show that densification resulting in fuel dimensional shrinkage can occur in ThO_2 and $\text{ThO}_2\text{-UO}_2$ fuels operating at low temperature and low burnup. The observed trend in diameter change of ThO_2 pellets agrees well with an empirical correlation based on initial fuel grain size and fuel burnup. It is assumed to be independent of temperature. $\text{ThO}_2\text{-UO}_2$ data, however, show less satisfactory agreement with the correlation.

II. DESCRIPTION OF TEST

Irradiation testing of Zircaloy-4 clad thoria base fuel rods was conducted, as part of the LWBR Development Program, in the Engineering Test Reactor (ETR) at the Idaho National Engineering Laboratory (INEL) in Idaho.

A. Test Rods

Four test rods identified as 81-90, 79-374, 79-513, and 79-514 were selected as typical rods for low burnup fuel (less than about 4.3 Mwd/KgM*) and low operating fuel temperature (less than about 1500°F centerline) to assess pellet diameter shrinkage. Detailed descriptions of the rods are given in Table II-1. Ranges of rod characteristics and operation are:

- rod length (6 to 90 inches)
- rod diameter (0.28 to 0.72 inch)
- fuel-cladding gap (about 4 to 12 mils, diametral)
- fuel composition (100 percent ThO_2 to ThO_2 - 30 w/o UO_2)
- operating time (about 1400 to 12000 hours)
- peak linear power (1.4 to 17.6 kw/ft)
- calculated peak centerline temperature (approximately 800 to 3850°F)
- peak burnup (0.9 to 8.2 Mwd/Kg U+Th).

B. Fuel Pellets

Fuel temperature and burnup vary axially in long fuel rods because of the axial neutron flux distribution in the reactor core. Thus, specific pellets at prescribed axial locations were selected in the 4 rods to provide the desired burnups and temperature. Rod axial power profiles and selected pellet positions (power levels) are shown schematically in Figure II-1. Three low temperature, low burnup ThO_2 pellets were chosen from 3 (79-513, 79-514, and 81-90) of the 4 rods. To extend the parameter ranges, a relatively high temperature but still moderately low burnup ThO_2 - UO_2 binary pellet was chosen from the fourth rod (79-374). An additional ThO_2 - UO_2 pellet was chosen from the combined thoria/binary fuel rod 79-513 to represent moderately high burnup at moderately low temperature operation. These 5 pellets are characterized in Table II-2. Ranges of pellet attributes and operating conditions are:

- pellet diameter (0.24 to 0.64 inch)
- pellet length (0.5 to 0.9 inch)

*Mwd/KgM = megawatt days per kilogram of metal (U+Th), and 4.3 Mwd/KgM corresponds to approximately 1×10^{20} fissions/cc fuel.

geometrical density (~94 to ~99 percent theoretical)
average grain size (ASTM #3.6 to 12.1, corresponding to average grain
diameters of about 100 μm to 5 μm)
peak fissioning rate (0.1 to 8.3×10^{12} fissions/cc-sec)
calculated peak centerline temperature (750 to 3700°F)
peak burnup (0.14 to 6.84 Mwd/Kg U+Th; 0.032 to 1.58×10^{20} fissions/cc
fuel).

TABLE II-1. CHARACTERIZATION OF FUEL RODS

Characteristics	Fuels			
Identification	81-90	79-374	79-513	79-514
Overall Length (in.)	6	45	90	90
Fuel Stack Length (in.)	3.5	36	29/15*	29/15*
Diametral Fuel-Clad Gap (in.)	0.0044	0.0119	0.0045/0.0061*	0.0037/0.0039*
Fuel Pellets				
Composition	ThO ₂	ThO ₂ - 30 w/o UO ₂	ThO ₂ /16.9 w/o UO ₂ *	ThO ₂ /17.2 w/o UO ₂ *
Sintering: Thoria	1700°C-12 hrs.		1750°C-12 hrs.	1750°C-12 hrs.
Binary		1750°C-12 hrs.	1750°C-12 hrs.	1750°C-12 hrs.
Cladding**				
Diameter (in.)	0.284	0.718	0.568	0.568
Wall Thickness (in.)	0.020	0.031	0.029	0.029
Structure**	SRA	RXA	SRA	SRA
Operation				
Time (hours)	2544	1435	12,080	12,080
Peak Linear Power (kw/ft.)	1.4	17.6	1.7/7.2*	1.7/7.2*
Calc. Peak Centerline Temp. (°F)	800	3,850	755/1,570*	755/1,570*
Peak Burnup (Mwd/Kg U+Th)	0.92	1.97	0.15/7.34*	0.19/8.20*
(10 ²⁰ fissions/cc)	0.21	0.45	0.03/1.9*	0.04/2.2*

*Thoria/Ebinary (combined fuel stack).

**Cladding = Zircaloy-4, tube reduced, 60-70 percent cold work on last pass.

SRA = Stress Relief Anneal (950 F, 4 to 6 hours); RXA = Recrystallization Anneal (1250 F, 4 to 6 hours).

TABLE II-2. ATTRIBUTES OF FUEL PELLETS

Attribute	Thoria			Binary	
	81-90 Pellet #4	79-513 Pellet #41	79-514 Pellet #41	79-374 Pellet #12	79-513 Pellet #55
Diameter (in.)	0.240	0.506	0.507	0.644	0.504
Length (in.)	0.49	0.57	0.49	0.93	0.75
Geometrical Density (%TD)*	97.4	93.9	98.9	93.8	96.6
Average Grain Size (ASTM#)	11.5	12.1	3.6	10.5	8.7
Equivalent Grain Diam. (μ m)	7.5	6.1	117.0	10.7	20.0
Peak Fissioning Rate (10^{12} fission/cc-sec)	2.29	0.07	0.10	8.32	3.64
Calc. Peak Temperature ($^{\circ}$ F)	800	750	750	3700	1450
Peak Burnup (Mwd/Kg U+Th)	0.92	0.14	0.19	1.89	6.84
(10^{20} fissions/cc)	0.21	0.032	0.044	0.43	1.58

*%TD = percent theoretical density; geometrical density is pellet weight divided by pellet volume as calculated from pellet diameter and length.

Figure II-1. Schematic Power Profile of Test Rods

III. EXPERIMENTAL OBSERVATIONS

A. Grain Size and Porosity in As-Fabricated Pellets

1. Measurements

Density, grain size, and porosity counts were measured on unirradiated fuel pellet samples to characterize these properties in the as-fabricated condition. Data from 12 thorium pellets (4 each from 3 different batches) and from 25 binary pellets (ThO_2 - 1.7 w/o UO_2 from two different batches) were used to develop a correlation between fuel grain size and volume percent small porosity. The thorium data are given here in Table III-1 and the binary data in Table III-2. Thorium pellet batches T-1 and T-2 were sintered in hydrogen at about 1750°C, thorium batch T-3 at about 1790°C, and both binary batches in the range 1750-1790°C. Thorium batch T-1 contained mainly fine grain pellets (ASTM #9 - 12 or finer*), batch T-2 contained coarser grain pellets (larger* than ASTM #9), and batch T-3 contained pellets mostly with grain size ASTM #8 - 10. Binary pellets had grain sizes of ASTM #4.5 to 7.0, but only those in the range of ASTM #6 - 7 were used in this correlation since the binary pellets involved in the diameter shrinkage evaluation were smaller grain pellets (ASTM #8 - 11).

Geometric density, included for information, was calculated from pellet weight divided by pellet volume. Grain size was determined from 250 - 500X photomicrographs of etched surfaces, using both ASTM comparator method and line intercept method. Counting of pore volumes and size classes was accomplished using the Quantimet, quantitative television microscope, with an epidiastroscope which permits the evaluation to be performed on glossy, high magnification (generally 250 - 500X) photomicrographs of a polished ceramographic specimen. The Quantimet provided a lineal analysis, using 625 scanning lines. Each scanning line included 800 "picture points," so that the field examined (called the "standard frame") contained 500,000 picture points. The epidiastroscope calibration was 222 picture points per inch. Thus, the photomicrograph area in a normal field of examination was 3.60 inches x 2.82 inches = 10.15 square inches.

Data obtained from the Quantimet were adjusted to correct for the probability that the largest segmental plane of a pore (diametral plane) has not been intersected and for the higher probability that a larger pore will intersect

*Larger ASTM grain size numbers indicate smaller equivalent grain diameters, which are related by:

$$D = \frac{2 \times 25^4}{\sqrt{\pi}} 2^{-(G-1)/2}$$

where D is the equivalent grain diameter in μm and G is ASTM grain size.

some random plane, compared to a smaller pore. This is known as the Scheil correction, which was adapted from Reference 10 by R. M. Berman for computer programming. Volume percents of pores cumulative to and between pore diameters of 0.0, 0.46, 1.14, 2.29, 3.43, 4.58, 5.72, 6.86, 9.15, 11.44 μm , etc., were calculated using the computer program. These pore sizes correspond to arbitrarily selected numbers of Quantimet picture points. Pore sizes ranging up to 2.3 μm were selected as being most important to fuel densification.

2. Empirical Correlation

Grain size versus volume percent pores less than 2.3 μm diameter is shown in Figure III-1. Use of an arbitrary limiting pore size of 2.3 μm gives a good correlation and is consistent with the 1 - 3 μm limiting diameter range quoted in other work (References 4, 8, and 9). There are 24 thoria data points with 2 data points (edge average and center plus mid-radius average) from each of the 12 pellets. The 25 binary data points are so closely spaced on Figure III-1 that only the mean and 2σ range for median values (between edge and center locations) are shown. The best-fit line through the 49 data points is shown on Figure III-1, with a resulting equation of:

$$P_o = 0.636 (G-6.2) \quad (\text{III-1})$$

where

P_o = volume percent pores less than 2.3 μm diameter

G = ASTM grain size.

This correlation, which shows a definite trend for more small diameter pore volume in oxide with smaller equivalent grain diameter, is consistent with a study by Smid, Reference 11. Smid's study showed that ThO_2 and $\text{ThO}_2\text{-UO}_2$ with larger grain sizes will contain coarser pores as a result of impingement and growth of smaller pores during grain boundary migration (grain growth) in the fuel fabrication process.

B. Post-Irradiation Measurements

Irradiation test rods were sectioned transversely to obtain segments containing the selected pellets. The segments were impregnated with plastic (HYSOL) to prevent movement or loss of fuel fragments during further processing. The segment was transversely sectioned to remove a piece containing only the desired pellet. These sections were mounted using HYSOL in metallographic rings.

Pellets were ground and polished for microscopic examination and pellet diameter measurement. Crack area measurements and grain size measurements were made from photomicrographs. Pellet diameter measurements on each of 2 or 3 planes were made from each mounted sample using a PICOMM television microscope at 250X magnification.

1. Pellet Diameter

The PICOMM television microscope used to remotely measure irradiated pellet diameter combines a precision positioning head with an optical microscope to view samples placed on a stationary table. To facilitate remote measurements, the microscope image (at 250X) was relayed via closed circuit television for viewing on a monitor screen. Electronic control and instrumentation of the x-y coordinates of the positioning head provided a digital output of the microscope tube position relative to the sample. Thus, after alignment of the sample using eyepiece cross lines, the difference in opposite edge positions of the pellet gave the diameter of the pellet. Six diameter measurements at 30-degree intervals were made on each sample using a machined 12-sided sample holder. Measurement uncertainties are discussed in Appendix 1. Correction for pellet cracking is discussed in Appendix 2.

In general, 3 transverse planes in each of the 5 pellets were selected for diameter measurements. These planes were at pellet mid-length, quarter length, and about 0.02 - 0.04 inch from the end. In 2 of the pellets, only 2 planes were measured. In 1 of these pellets plus a third pellet, measurements on 1 plane were inconsistent with measurements on the other plane(s) of the same pellet. Thus, measurements on 11 planes (out of a possible 15) were used in the data base for the 5 pellets.

2. Grain Size, Porosity, and Structure

Ceramographic examination of the pellet sections was carried out to (a) assess post-irradiation porosity, (b) measure post-irradiation grain sizes, and (c) inspect for gross structural changes. Porosity examination was performed on the as-polished fuel sample before etching. Examinations for grain size determination and structural change were performed after etching the fuel sample in a hot (180 - 200°F) supersaturated solution of ammonium fluoride in water at 1-minute intervals for a few minutes total immersion time.

a. Porosity Evaluation. Qualitative examination of post-irradiation porosity in the 5 pellets revealed essentially uniform porosity distribution throughout the pellet cross-sections, with apparently no large bias in size

distribution. There did appear to be a tendency in the thorium pellets for the larger size porosity to exist as intergranular or "necklace" porosity around the prior (after cold pressing) green pellet granules. This same type porosity was observed in an unirradiated thorium retainer pellet for rod 81-90 and is shown in Figure III-2.

A quantitative evaluation of pore size distribution before and after irradiation was not obtained on the specific pellets irradiated because the quantitative pore analysis (by Quantimet microscopy) is a destructive examination requiring pellet sectioning and mounting (in plastic) for ceramographic preparation. An attempt to compare post-irradiation concentration of small pores ($<2.3 \mu\text{m}$ diameter) in 2 irradiated pellets with the concentrations measured from 2 similar unirradiated fuel pellets was not fruitful because of pellet-to-pellet scatter within the fuel batch. As shown in Figure III-1 there is considerable scatter in the concentrations of small pores in individual pellets with the same average grain size after a common fabrication procedure. Since the same pellet cannot be measured before and after irradiation, several replicate pellets would be needed to obtain a comparison of average pre- and post-irradiation porosities.

b. Grain Size. Grain size determinations were made using the ASTM Standards comparison method or linear intercept method on photomicrographs of 100 to 500X magnification. Post-irradiation grain sizes were measured on each pellet. Where there was a measurable radial variation, grain sizes were measured across the pellet radius at the edge, mid-radius, and center positions. The data are summarized here in Table III-3. Pre-irradiation grain sizes were available from both retainer samples or representative pellets from the same batches as the test rod pellets, and these data also are summarized in Table III-3. In some cases only grain size ranges are known for pellets representing the fuel batch. For both pre-irradiation and post-irradiation data the average grain sizes are tabulated along with the distributions and ranges of grain sizes. Note that average pre- and post-irradiation grain sizes are very nearly the same, except for the binary pellets in rod 79-513 in which case there is considerable overlap of pre- and post-irradiation ranges. Thus, both qualitative examinations and grain size measurements indicate no significant grain growth during irradiation under the conditions of these test rods, confirming low temperature operation.

c. Structural Changes. Except for pellet cracking, no gross structural changes such as grain growth, grain reorientation, formation of central voids or incipient melting were observed during examination of the various planes in the

5 pellets. This result is indicative of the low power, low temperature operation of the fuel rods. High temperature due to high power output rates can lead to structural changes which can affect fuel densification and dimensional changes.

Pellet cracking was observed in 3 of the 5 pellets. In sections from 1 thorium pellet (from rod 81-90) the few cracks observed comprised about 1 percent of the PICOMM planar area (area = $\pi D^2/4$, where D = average PICOMM diameter). Sections of 2 binary pellets showed more extensive cracking, especially in rod 79-374. Maximum crack area was over 3 percent of the PICOMM planar area in the 79-374 pellet. Details of the crack measurements are reported in Appendix 2.

C. Change in Pellet Diameter

Post-irradiation pellet diameter measurements are discussed above. The pellet average diameters measured are reported in Table III-4. Also reported are estimates of uncertainty (standard deviation). As indicated in Appendix 1, from 6 to 18 diameter measurements on each pellet were used to calculate the averages and standard deviation estimates.

Pre-irradiation diameters obtained from hand micrometer measurements of the pellets also are reported in Table III-4. The diameters reported are either average or median values. Estimates of uncertainty in pre-irradiation diameter measurements, as reported in Table III-4, are described in Appendix 1.

Measured diameter change in the irradiated pellets was taken as the difference in average pre-irradiation and post-irradiation diameters reported as percent diameter change in Table III-4. The changes ranged from essentially no change in pellet #41 from rod 79-514 to slightly more than 1 percent reduction of diameter in pellet #12 from rod 79-374. No average pellet diameter increase was observed. Total uncertainty in the percent diameter change was taken as twice the estimated standard deviation, s_R (%), which was calculated according to procedures in Appendix 1 and reported in Table III-4.

TABLE III-1. UNIRRADIATED THORIA FUEL PELLETS

Batch	Pellet	Geometrical Density (% TD)*	ASTM Grain Size**		v/o Pores <2.3 μ m Diameter**	
			Edge	Interior	Edge	Interior
T-1	21	96.8	>12	9.95	4.10	1.68
	22	97.6	>12	10.90	3.17	3.08
	23	97.5	>12	10.70	3.50	3.70
	24	97.2	9.65	9.53	2.63	1.52
T-2	21	97.4	8.59	8.41	2.03	0.92
	22	97.3	8.97	8.72	2.32	0.91
	23	97.6	9.94	8.78	1.78	0.93
	24	97.6	7.18	8.26	1.50	0.93
T-3	9	97.7	9.71	9.48	2.39	2.15
	10	98.4	9.49	9.90	1.80	1.52
	11	98.5	8.37	8.98	1.66	1.54
	12	98.3	9.85	9.96	2.79	2.21

*%TD = percent of theoretical density.

**Edge = average of 2 evaluations.

Interior = average of 1 center plus 2 mid-radius evaluations.

TABLE III-2. UNIRRADIATED BINARY FUEL PELLETS

Batch	Pellet	Geometrical Density (% TD)	ASTM Grain Size			v/o Pores <2.3 μ m Diameter		
			Edge	Center	Median	Edge	Center	Median
B-1	1	97.0	6.5	6.5	6.5	0.10	0.14	0.12
	2	96.8	6.3	6.5	6.4	0.10	0.13	0.115
	3	97.1	6.3	7.0	6.65	0.07	0.13	0.10
	4	96.9	6.5	6.5	6.5	0.10	0.11	0.105
	5	96.9	6.0	6.5	6.25	0.11	0.15	0.13
	6	97.0	6.0	6.5	6.25	0.07	0.07	0.07
	7	97.0	6.3	6.5	6.4	0.10	0.13	0.115
	8	97.0	6.3	6.5	6.4	0.05	0.16	0.105
	9	96.9	6.5	6.0	6.25	0.07	0.08	0.075
	10	97.1	6.0	6.5	6.25	0.10	0.07	0.085
	11	96.9	6.3	6.0	6.15	0.11	0.09	0.10
	12	97.0	6.0	5.5	5.75	0.08	0.10	0.09
	17	97.0	6.3	6.0	6.15	0.05	0.18	0.115
	18	97.0	6.3	6.5	6.4	0.05	0.05	0.05
	19	97.0	6.0	6.0	6.0	0.06	0.07	0.065
	20	97.1	6.0	6.5	6.25	0.04	0.06	0.05
	21	96.8	5.5	5.5	5.5	0.08	0.13	0.105
	22	97.0	6.0	5.5	5.75	0.06	0.17	0.115
	23	96.9	6.0	6.0	6.0	0.14	0.12	0.13
	24	96.8	5.8	6.0	5.9	0.11	0.11	0.11
B-2	40	96.5	6.3	6.0	6.15	0.02	0.06	0.04
	45	96.7	6.5	6.5	6.5	0.03	0.06	0.045
	46	96.6	6.3	7.0	6.65	0.07	0.14	0.105
	47	96.7	6.8	7.0	6.9	0.09	0.16	0.125
	48	96.5	6.3	6.5	6.4	0.06	0.04	0.05

TABLE III-3. SUMMARY OF GRAIN SIZES

Rod	Irradiated Pellet	Comments	Pre-Irradiation (μm)*				Post-Irradiation (μm)*			
			Edge	Mid-Rad.	Center	Average	Edge	Mid-Rad.	Center	Average
81-90	4	1	8.4-15.1	4.7	3.8-5.3	7.5 (11.5)	12.2	6.3	4.3	7.3 (11.6)
79-374	12	2	14.1	-----	8.1	10.7 (10.5)	10.7-15.1	-----	9.0	11.5 (10.3)
79-514	41	3	-----	92.0-143.0	-----	117.0 (3.6)	121.0	112.0	128.0	121.0 (3.5)
79-513	41	3	-----	<6.3	-----	<6.3 (>12)	7.3	6.3	5.0	6.1 (12.1)
79-513	55	3	-----	15.1-27.0	-----	20.0 (8.7)	33.3	22.0	27.0	27.0 (7.8)

COMMENTS

1. Pre-irradiation data are from 3 pellets from the same batch as pellet #4. Post-irradiation measurements are from the mid-length plane.
2. Pre-irradiation data are from 1 pellet representative of the binary pellets in this rod. Post-irradiation values are from the plane near the pellet end.
3. Pre-irradiation data are the overall ranges of measurements on representative pellets. Post-irradiation values are from the mid-length planes.

*Grain sizes are given as grain diameters in μm . Average values in parentheses are the equivalent ASTM grain size numbers, (ASTM No.).

TABLE III-4. MEASURED AND CALCULATED DIAMETER CHANGES IN INDIVIDUAL IRRADIATED PELLETS

	Thoria			Binary	
	81-90 Pellet #4	79-513 Pellet #41	79-514 Pellet #41	79-374 Pellet #12	79-513 Pellet #55
Ave. Pellet Diameter (in.):					
Pre-Irrad.	0.2396	0.5059	0.5066	0.6442	0.5043
Post-Irrad.	0.2384	0.5054	0.5066	0.6374	0.5037
Est. Uncertainty (in.):					
Pre-Irrad.	0.0002	0.0003	0.0003	0.0001	0.0003
Post-Irrad.	0.0005	0.0004	0.0003	0.0019	0.0005
Diameter Change (%)	-0.50	-0.10	0	-1.06	-0.12
Uncertainty (%)	0.17	0.10	0.09	0.21	0.13
Diameter Change Calc. (%)	-0.53	-0.11	+0.01	-0.69	-0.42
Uncertainty (%)	0.26	0.02	0.00	0.19	0.08
Pre-Irradiation Grain Size:					
μm: Average	7.5	6.1	117.0	10.7	20.0
Range	3.8-15.1	4.7-7.0	112.0-128.0	3.0-14.1	17.3-22.0
ASTM No.: Average	11.5	12.1	3.6	10.5	8.7
Range	9.5-13.5	11.7-12.7	3.3-3.7	9.7-11.3	8.4-9.1
Burnup (Mwd/Kg U+Th)	0.92	0.15	0.19	1.89	6.84
(10 ²⁰ fissions/cc)	0.21	0.032	0.044	0.43	1.53

Figure III-1. Grain Size - Small Porosity Correlation for Thoria and Binary Fuel with Average Grain Sizes of ASTM No. 6 to 12

81-90 Retainer Pellet 50X

Figure III-2. Necklace Porosity in Unirradiated Thoria

IV. ANALYSIS OF PELLET DIAMETER SHRINKAGE

A. Development of Empirical Equation

As a first approximation it is assumed that fuel burnup is the primary irradiation parameter. This appears justifiable because of the observed densification (pellet diameter shrinkage) at low burnup and both low and moderately high fuel operating temperature and heat output. Also, fuel densification models (References 4, 5, and 8) for UO_2 have been developed with fuel burnup as primary independent variable. Figure IV-1 shows measured ThO_2 and ThO_2-UO_2 pellet diameter change as a function of fuel burnup. Operating parameters and fuel characteristics were given in Table II-2; data were given in Table III-4.

To develop an empirical equation for predicting in-pile diameter shrinkage it is assumed that only small pores (diameters $<2.3 \mu m$) close. This is similar to assumptions used for UO_2 fuels where a much larger data base has been used to develop correlations. In general, smaller porosity of 1 - 3 μm diameter or less has been found (References 4, 8, and 9) to be much less stable under irradiation than pores of about 10 μm diameter or larger. Considerable data developed during the fabrication of thoria and ThO_2-UO_2 pellets for LWBR has demonstrated a direct relationship between fuel grain size and the fraction of small diameter porosity as shown in Section III.A. Pellets having small grains tend to have a larger fraction of small pores. The correlation derived between volume percent small pores and grain size is:

$$P_o = 0.636 (G-6.2) \quad (III-1)$$

where

P_o = volume percent of pores $<2.3 \mu m$ diameter

G = ASTM grain size number.

It seems reasonable that dimensional shrinkage due to closure of small pores should be asymptotically limited by the amount of porosity and that the approach to maximum shrinkage should vary with fuel burnup. Fission product swelling, a function of burnup, should be considered as an opposite effect to the densification. Larger pores may tend to close at higher burnups and higher temperatures, but the swelling may increase also under these conditions (Reference 12).

It was proposed by Chubb, et al, (Reference 5) that the pore closure should be an exponential function of burnup, to explain experimental data obtained from stack length decreases measured in UO_2 fuel rods in commercial PWR-type power

reactors. Collins and Hargreaves (Reference 4), also, derived an exponential form of empirical equation relating decrease in porosity (volume change) with fuel burnup. This equation was derived to fit measured stack length changes in UO_2 fuel pins from the WAGR (Windscale Advanced Gas-Cooled Reactor) reactor and direct density measurements in UO_2 fuel from SGHWR (Steam Generating Heavy Water Reactor) reactors. Mechanisms for densification due to pore closure are postulated (References 5, 7, and 13) to be fission-induced pore (vacancy aggregate) sintering, or fragmentation assisted by radiation-enhanced diffusion of vacancies to grain boundary sinks or external surfaces. An exponential form of volume change versus burnup was derived from theoretical considerations of very fine porosity in Reference 7, in which coarser pores were calculated to close according to summations of a power law (cubic equation) with respect to fuel burnup. In addition to these mechanistic approaches, a hot pressing model (Reference 6) was found to result in an exponential form of equation for pore closure versus burnup if the effective internal hydrostatic pressure due to fuel thermal gradient and fuel-cladding interaction forces is the dominant driving force for in-reactor densification.

Overall fuel volume changes during irradiation must include the effect of fuel swelling in addition to fuel densification. In References 5 and 7 it was assumed that fuel swelling is an independent, additive effect. Further it was assumed that irradiation-induced swelling is a linear function of fuel burnup.

Based on the models and data from other densification studies discussed above, an empirical equation consisting of an exponential pore closure term plus a linear swelling term was formulated to describe the observed diameter changes of irradiation LWBR thorium-base fuel pellets. The suggested equation is:

$$\Delta D(\%) = -a' P_o (1 - e^{-bB}) + cB \quad (\text{IV-1})$$

where

P_o = volume percent of small porosity ($<2.3 \mu\text{m}$ diameter)

B = fuel burnup.

a' , b , c = constants.

Substitution of the porosity-grain size correlation, Equation (III-1) into Equation (IV-1) gives:

$$\Delta D(\%) = -a (G-6.2)(1 - e^{-bB}) + cB \quad (\text{IV-2})$$

where a , b , c are constants.

The empirical coefficient "c" was evaluated from fuel stack length change data from 3 LWBR test rods with freestanding cladding and 0.008 to 0.009 inch fuel-cladding gap. Both fuel stack and cladding length increases in these rods followed essentially "free growth" behavior, indicating negligible fuel-cladding interaction. The thorium-uranium fuel stack length increases fit a linear relationship with burnup, where $\Delta L(\%) = 0.233 B$, with $B = \text{burnup in } 10^{20} \text{ fissions/cc}$. Thus, assuming isotropic swelling, the diameter change due to fission product swelling also is $\Delta D(\%) = 0.233 B$ and the coefficient "c" is 0.233 per $10^{20} \text{ fissions/cc}$ or 0.053 per Mwd/Kg metal.

Fitting of the pellet diametral changes measured, using known values of the fuel grain sizes and burnup and calculated values of porosity ($<2.3 \mu\text{m}$ diameter) concentrations, provided estimates of the empirical coefficients a' , a , and b in Equations (IV-1) and (IV-2). These constants are given below.

<u>Constant</u>	<u>Value</u>	<u>Units</u>
a'	0.5	None
a	0.318	None
b	2.0	$(10^{20} \text{ fissions/cc})^{-1}$
b	0.457	$(\text{Mwd/Kg Metal})^{-1}$
c	0.233	% per $10^{20} \text{ fissions/cc}$
c	0.053	% per Mwd/Kg Metal

Although the inventories of pore concentrations and size distributions could be directly relatable to in-pile densification mechanisms as proposed, and thereby provide basic variables to describe the densification, fuel grain size is a much more practical variable to measure and control. Also, the correlation of the grain size with a single group of limiting diameter pores allows a quantitative link with some "average" effect of "small" pore closure. Thus, the empirical relationship represented by Equation (IV-2) is suggested as a means of describing the observed pellet diameter changes related to irradiation induced densification of thorium-base fuel. An example of the diametral change predicted as a function of fuel burnup by Equation (IV-2) for a fuel of grain size ASTM #10 ($12.6 \mu\text{m}$ diameter) is shown in Figure IV-2.

B. Parametric Sensitivity

Sensitivity of the pellet diameter change calculation to fuel burnup and fuel grain size is shown in Figure IV-1. Parametric calculations of diameter change are shown as a function of burnup for various grain sizes. Note that maximum diameter shrinkage is strongly influenced by grain size, presumably due

to closure of small diameter pores. For grain sizes of ASTM #8 - 12 (6 - 25 μm diameter) the maximum shrinkage is reached at burnups of about 3.5 to 6.1 Mwd/KgM (0.8 to 1.4×10^{20} fission/cc fuel). Fuel swelling causes the pellet diameter to increase again with continued burnup after the effective small pore concentration has been exhausted.

C. Comparison of Measured and Calculated Diameter Shrinkage

A comparison of calculated and measured diameter changes is shown in Figure IV-3. Calculated diameter change uncertainty is based on 2 factors - the reported range of ASTM grain size and the estimate of uncertainty ($\pm 10\%$) in the calculated burnup of the fuel.

The thoria data appear to correlate better than the binary data. Calculated diameter change underpredicts the measured change in the binary pellet from rod 79-374 which operated at a higher temperature and fissioning rate than any of the other rods. (See Table II-2.) In UO_2 fuel it has been found (Reference 8) that densification occurs rapidly during irradiation at fission rates in the range 9.6 to 14.1×10^{12} fission/cc-sec and centerline temperatures in the range 1670 to 2415°F, but at lower fission rates ($< 7.8 \times 10^{12}$ fission/cc-sec) and temperatures ($< 1310^\circ\text{F}$) densification occurs at a much slower rate.

Also, judging from large cladding ridges formed at pellet interfaces there was apparently pellet-cladding contact in rod 79-374 during the in-pile exposure. The applied coolant pressure may therefore have caused diametral creep of the fuel (a volume-conserving process independent of the amount of small porosity).

Figure IV-1. Pellet Diameter Shrinkage Related to Burnup and Grain Size

Figure IV-2. Total Pellet Diameter Change as the Sum of a Densification Component and a Swelling Component for Fuel with ASTM No. 10 Grain Size

CALCULATED DIAMETER CHANGE: $-0.318(G-6.2)(1-e^{-0.457B})+0.053B$
 OR $-0.318(G-6.2)(1-e^{-2F})+0.233F$
 B IN Mwd/kg U+Th; F IN 10^{20} FISSIONS/CC

Figure IV-3. Correlation of Calculated and Measured Pellet Diameter Shrinkage

V. DISCUSSION

A. Maximum Diameter Change

1. Predicted from Empirical Correlation

It can be observed in Figure IV-1 that the maximum diameter changes calculated using Equation (IV-2) increase with increasing initial ASTM grain size number (i.e., decreasing grain diameter). This maximum diameter shrinkage is presented as a function of grain size in Figure V-1. Based on the correlations developed, the maximum pellet diameter shrinkage is shown to vary between zero (0) for grain sizes of about ASTM #6 or less (grain diameter of $\sim 50 \mu\text{m}$ or greater) to about 1.7 percent for a grain size of ASTM #13 (grain diameter of $\sim 4.5 \mu\text{m}$).

Also, it is noted that the fuel burnup for maximum predicted pellet diameter shrinkage increases with ASTM grain size number. Figure V-2 shows this relationship, which suggests for thoria-base fuels with grain sizes of about ASTM #8 - 13 that maximum densification will occur by $1 - 1.5 \times 10^{20}$ fission/cc or 4 - 6 Mwd/KgU+Th, which agrees with UO_2 test observations (Reference 5).

2. Densification Limit

If closure of residual small size porosity provides the means for fuel densification, then the maximum densification possible should be related to the concentration of these small pores. Assuming that pore closure is isotropic, 1/3 the initial (pre-irradiation) volume of small pores ($< 2.3 \mu\text{m}$ diameter) represents the maximum possible linear densification during irradiation exposure. Maximum diameter change discussed in the preceding section included the swelling effect. In this section, however, only the densification is considered so that the diameter change calculated as 1/3 P_o (linear densification) is assumed to be the limiting diameter change. This limiting value of 1/3 P_o is less than that calculated from the densification term in Equation (IV-2), which gives (at infinite burnup) a value of 1/2 P_o . However, since the burnup dependent swelling term in Equation (IV-2) would offset the densification, the 1/2 P_o value for diameter shrinkage would never be reached. Thus, in Figure V-3, the limiting value of 1/3 volume percent of pores $< 2.3 \mu\text{m}$ diameter is compared to the measured pellet percent diameter shrinkage minus calculated swelling. Small diameter porosity concentrations were calculated from Equation (III-1), and the swelling was calculated from the linear term in Equation (IV-2). For points falling above the correlation line, it appears that 1/3 the volume percent of small pores does

represent an upper limit for pellet diameter shrinkage due to in-pile densification. The data point for rod 79-51⁴ pellet lies at the origin because no pores <2.3 μm diameter were predicted for the large grains (ASTM #3.5) and no diameter change was measured at very low burnup. The data point below the line for rod 79-37⁴ indicates that some other mechanism in addition to pore closure may have caused the excess diameter change.

B. Diameter Change versus Stack Length Change

Although individual pellet lengths of the irradiated fuel were not measured, pellet stack lengths were measured or inferred from post-irradiation neutron radiographs. Comparison with pre-irradiation stack length measurements gave percent length changes for the pellet stacks or segments. Figure V-4 shows percent stack length change compared to percent diameter change. The comparison indicates that isotropic densification may be a good assumption for the rod 81-90 pellet. This is logical since the pellet stack length was short (3.5 inches) and the absence of any cladding ridges indicates negligible fuel-cladding interaction. In long fuel stacks the overall length change may not represent individual pellets in the stack and fuel-cladding interaction can lead to anisotropic dimensional changes.

Rod 79-37⁴ presents the greatest apparent departure from isotropic behavior as indicated in Figure V-4. Two possible reasons are: (1) length change of the 36-inch fuel stack does not represent the length change of the particular pellet on which diameters were measured, or (2) radial creep forces transmitted through fuel-cladding interaction caused anisotropic pellet diameter shrinkage as indicated in Figure V-3. A maximum cladding radial ridge height of 0.005 - 0.006 inch indicates significant radial interaction. Also, an increase of 0.008 - 0.009 inch in cladding ovality during in-pile operation coupled with pellet diameter measurements showing 0.003 - 0.006 inch ovality of the pellet support the possibility of radial interaction forces.

To account for anisotropy in the length change and pellet diameter change, a maximum possible diameter shrinkage was calculated as a weighted fraction of small pores for comparison to the measured diameter shrinkage minus calculated linear swelling (diametral densification). This is similar to the maximum isotropic densification ($1/3 P_o$) shown in Figure V-3. The weighted maximum diameter change is calculated by

$$\% \Delta D_{\text{max}} = P_o \frac{\% \Delta D_{\text{meas}}}{2 (\% \Delta D_{\text{meas}}) + \% \Delta L_{\text{stack}}},$$

where it is assumed that

$$\% \Delta V_{\text{meas}} = 2(\% \Delta D_{\text{meas}}) + \% \Delta L_{\text{stack}} ,$$

and P_o is the small pore ($< 2.3 \mu\text{m}$ diameter) volume percent calculated from the grain size correlation. If the densification is isotropic, then

$$\% \Delta D_{\text{meas}} = \% \Delta L_{\text{stack}} ,$$

and

$$\% \Delta V_{\text{meas}} = 3\% \Delta D_{\text{meas}} .$$

In this case the weighting factor

$$\frac{\% \Delta D_{\text{meas}}}{\% \Delta V_{\text{meas}}} = 1/3$$

and

$$\% \Delta D_{\text{max}} = 1/3 P_o$$

as in Figure V-3. If the densification is anisotropic, $\% \Delta D_{\text{max}} \neq \% \Delta L_{\text{stack}}$ and the weighting factor will be different than $1/3$, indicating possible anisotropy. Figure V-5 shows ΔD_{max} versus diametral densification. This procedure appears to have had some effect in smoothing out the scatter in the assumed isotropic comparison in Figure V-3, indicating that the weighting factor did tend to normalize limiting diametral densification due to pore volume reduction, thereby accounting for observed anisotropy in densification.

Figure V-1. Maximum Fuel Pellet Diameter Shrinkage as a Function of Initial Grain Size

Figure V-2. Fuel Pellet Burnup for Maximum Diameter Shrinkage as a Function of Initial Grain Size.

Figure V-3. Comparison of Assumed Isotropic Pore Closure with Diametral Densification

Figure V-4. Pellet Diameter Change Compared to Stack Length Change

Figure V-5. Maximum Diameter Shrinkage as a Weighted Fraction of Small Pores

VI. SUMMARY AND CONCLUSIONS

In support of the LWBR Development Program, irradiation tests of thoria base, Zircaloy-4 clad fuel rods were conducted in the Engineering Test Reactor (ETR) at the Idaho National Engineering Laboratory (INEL) in Idaho. Fuel pellet diameter changes were measured from 5 pellets (3 thoria and 2 binary) taken from 4 irradiation test rods. These 4 fuel rods represent generally low burnup and low temperature operation; however, a relatively high temperature pellet and a moderately high burnup pellet were included to extend the parameters.

Minimum burnup and minimum calculated peak centerline temperature were 0.14 Mwd/Kg metal (0.03×10^{20} fissions/cc) and 750°F respectively (same pellet). Maximum burnup and maximum calculated peak centerline temperature were 6.84 Mwd/Kg metal (1.58×10^{20} fissions/cc) and 3700°F, respectively (different pellets). Fuel composition range was pure thoria to binary $\text{ThO}_2 + 30 \text{ w/o } \text{UO}_2$, and sintered pellet grain size range was ASTM #3.6 to 12.1 (about 6 to 120 μm diameter).

Measured pellet diameter shrinkage ranged from nil in a large grain thoria pellet (98.9 percent theoretical density) to 1.06 percent in a small grain binary pellet (93.8 percent theoretical density). Measurement uncertainties in percent diameter change were 0.1 to 0.2 percent.

In unirradiated fuel, a correlation was observed between concentration of small pores and as-sintered pellet grain size. The porosity-grain size correlation found is:

$$P_o = 0.636 (G - 6.2)$$

where P_o = volume percent small pores (diameter $< 2.3 \mu\text{m}$) and G = fuel grain size (ASTM No.) fitted over a grain size range of ASTM #6 - 12 (6 to 50 μm diameter).

An empirical equation, developed from the assumption that closure of small pores (less than 2.3 μm in diameter) is responsible for in-reactor densification, was found appropriate to calculate pellet diameter change. The form of the equation is as follows:

$$\Delta D(\%) = a' P_o (1 - e^{-bB}) + cB ,$$

where B = fuel burnup. The first term is the densification component and the second term is the swelling component. The exponential character of the densification term was adapted from other densification studies and from a hot

pressing model to account for the self-limiting effect of pore closure. The linear swelling term has been observed to be appropriate for the empirical description of fission product swelling. Substituting the grain size-porosity correlation gives:

$$\Delta D(\%) = -a (G-6.2) (1 - e^{-bB}) + cB$$

Observed fuel dimensional changes, assuming isotropic swelling, were fit to this equation to evaluate the empirical coefficients. The resultant empirical correlation is:

$$\Delta D(\%) = -0.318 (G-6.2) (1 - e^{-0.457B}) + 0.053B$$

or

$$\Delta D(\%) = -0.318 (G-6.2) (1 - e^{-2.0F}) + 0.233F$$

where B = fuel burnup in Mwd/KgU+Th and F = fuel burnup in 10^{20} fissions/cc.

Using this correlation it is shown that predicted maximum pellet diameter shrinkage increases with decreasing initial grain size below about 50 μm diameter to about 1.7 percent for a grain size of 4.5 μm diameter. Over the grain size range of 4.5 to 50 μm diameter, the burnup at which maximum diameter shrinkage due to densification is expected is 4 - 6 Mwd/KgU+Th ($1 - 1.5 \times 10^{20}$ fissions/cc).

One third the calculated or measured volume percent small pores (based on assumption of isotropy) is shown to be a reasonable upper limit for measured diameter shrinkage. A comparison of fuel stack length change and **individual pellet diameter shrinkage** shows that **isotropic dimensional shrinkage** probably is a reasonable assumption where negligible fuel-cladding interaction has occurred. However, increasing the degree of interaction appears to produce anisotropic deformation biased towards either length change or diameter change depending on the diametral stability of the cladding.

It is concluded from measured fuel pellet diameter changes that densification occurs in ThO_2 -base fuels, as it does in UO_2 fuels, at low power (low temperature) operation and at low burnups. Diameter changes measured correlate reasonably well with as-fabricated grain size and fuel burnup. Grain size dependence is introduced through its correlation with the concentration of small porosity, which closes during in-pile densification. These data and empirical correlations illustrate the importance of fabricating high density fuel with large grains, thereby minimizing small porosity and densification. Although the observed dimensional changes due to densification did not correlate with as-fabricated density (i.e., total porosity), it is obvious that a completely dense fuel with

no porosity (of any size) will not densify during irradiation. For a typical ThO_2 -base fuel of 97 percent theoretical density with grain size ASTM #8 (about 1 - 1.5 v/o pores <2.3 μm diameter), up to about 0.6 - 0.8 percent volumetric densification (0.2 - 0.3 percent diameter reduction assuming isotropy) could be expected. Fission product swelling would offset further densification.

VII. REFERENCES

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APPENDIX 1. UNCERTAINTY IN DIAMETER MEASUREMENTS FOR IRRADIATED PELLETS

I. POST-IRRADIATION DIAMETER MEASUREMENTS

Three categories of uncertainty in the PICOMM remote diameter measurements are: (a) instrument accuracy, (b) reproducibility of measurements, and (c) error introduced by rounding of pellet edges and by pellet cracking. Correction for pellet cracking is discussed in Appendix 2; other errors are discussed below.

A. Instrument Accuracy

Instrument accuracy was checked before and after all pellet sample measurements by measuring a calibrated micrometer scale with 0.0001 inch and 0.010 inch divisions over at least 0.200 inch gage. The accuracy determined in this manner was within 0.0002 inch total range (i.e., ± 0.0001 inch).

B. Reproducibility

Reproducibility of measurements depends on the sample characteristics, sample holder placement against square edges on the table, alignment of the pellet edges with the crosslines and operator technique. In this work it was found that sample characteristics combined with locating the pellet edge were the major contributors to reproducibility error. Specifically, the largest uncertainty arises in the ability to define the true edge of the pellet because of the ragged appearance (at 250X) of the ceramic pellet edge, especially if any edge chips or pellet cracks occur at the position of the measurement. The estimate of standard deviation in the average of diameter measurements made on the pellet was taken to represent the reproducibility error. These errors are given in Table 1.1 for the 5 pellets measured.

C. Pellet Edge Rounding

Error introduced by rounding of pellet edges* during grinding and polishing of the ceramographic sample was measured for an unclad, unirradiated ThO_2 pellet. Measurements indicated a PICOMM diameter bias of about -0.0007 inch, referenced to the average diameter measured beforehand using a hand micrometer. It was judged that the diameter measurement error due to pellet edge rounding in an

*Optical conditions and a limited depth of field do not allow focusing on the true diameter of the pellet with rounded edge when measuring the diameter positions.

irradiated clad pellet sample was much less than that in the unirradiated unclad pellet sample. This judgement was based on evidence that the unclad control pellet was unprotected by the relatively soft plastic mounting material (HYSOL), whereas the irradiated clad pellet edge was essentially protected from abrasive rounding by touching or proximity to the cladding. Thus, the edge rounding error in an irradiated clad pellet sample was assumed to be within the uncertainty measured from the diameter reproducibility.

D. Total Uncertainty

Because of the relative magnitudes of the errors discussed above and the nature of the diameter measurements (wherein the characteristic condition of the pellet edge at the measurement location was the predominate source of error), it was assumed that the total uncertainty was represented by the reproducibility as measured by the calculated estimate of standard deviation about the average of the individual diameter measurements on a pellet. The estimates of standard deviation and the number of measurements are given in Table 1.1 for the 5 pellets.

The reproducibility was assumed to be a random error about the average diameter. The pellet cracking error described in Appendix 2, however, is a biased error and was subtracted from the measured average diameter (best estimate of the true mean diameter).

TABLE 1.1. ESTIMATES OF STANDARD DEVIATION ($\bar{s}_D$) IN
POST-IRRADIATION PELLET DIAMETER MEASUREMENTS

<u>Rod</u>	<u>Pellet</u>	<u>$\bar{s}_D$ (inch)</u>	<u>n (no. of measurements)</u>
81-90	4	0.0005	12
79-513	41	0.0004	17
79-514	41	0.0003	18
79-374	12	0.0019	12
79-513	55	0.0005	6

II. PRE-IRRADIATION DIAMETER MEASUREMENTS

Pre-irradiation diameter measurements were obtained specifically on 4 of the 5 pellets irradiated. Specific diameter measurements on pellet #41 from rod 79-514 were not known. The pre-irradiation diameter assumed for this pellet

was the average of measurements made on a 50-pellet sample from the batch from which this pellet was taken. The standard deviation about the batch average was 0.0002 inch. All pre-irradiation diameter measurements were made with hand micrometers. Estimates of uncertainty in the pre-irradiation diameters of the 5 pellets are evaluated below and given in Table 1.2.

For pellets from rods 81-90 and 79-374 only the pre-irradiation diameter ranges were known. Thus, the median values were assumed for nominal pre-irradiation diameters and the diameter ranges were taken as the total uncertainty.

Only average pre-irradiation diameter values were known for the 2 pellets from rod 79-513 and the pellet from rod 79-514, which was discussed above. To establish an estimate of uncertainty for these 3 pellets, hand micrometer measurements were made on 5 unirradiated thoria control pellets. An average reproducibility of 0.0003 inch for the control pellets was assumed as the total uncertainty for the pre-irradiation diameters of the 3 irradiated pellets.

Table 1.2 below gives the pre-irradiation uncertainty estimates and types of nominal diameter for the 5 irradiated pellets.

TABLE 1.2. PRE-IRRADIATION PELLET DIAMETER UNCERTAINTIES

<u>Rod</u>	<u>Pellet</u>	<u>Nominal Diameter (type)</u>	<u>Uncertainty (in.)</u>
81-90	4	Pellet Median	0.0002
79-513	41	Pellet Average	0.0003
79-514	41	Batch Average	0.0003
79-374	12	Pellet Median	0.0001
79-513	55	Pellet Average	0.0003

III. CHANGE IN DIAMETER

The change in diameter, ΔD , is the difference in the average pre- and post-irradiation pellet diameters, $\bar{D}_1$ and $\bar{D}_2$, respectively. Thus, if R is the percent diameter change,

$$R = 100 \frac{\bar{D}_2 - \bar{D}_1}{\bar{D}_1}$$

It has been shown (e.g., Reference 1.1) that the variance (square of the standard deviation) of a function such as R is:

$$\begin{aligned}
 s_R^2 &= \left[\left(\frac{\partial R}{\partial \bar{D}_1} \right)^2 s_{\bar{D}_1}^2 + \left(\frac{\partial R}{\partial \bar{D}_2} \right)^2 s_{\bar{D}_2}^2 \right] \times 10^4 \\
 &= \left[\left(\frac{\partial R}{\partial \bar{D}_1} \right)^2 \frac{s_1^2}{n_1} + \left(\frac{\partial R}{\partial \bar{D}_2} \right)^2 \frac{s_2^2}{n_2} \right] \times 10^4 \\
 &= \left[\frac{\bar{D}_2^2}{\bar{D}_1^4} \frac{s_1^2}{n_1} + \frac{1}{\bar{D}_1^2} \frac{s_2^2}{n_2} \right] \times 10^4
 \end{aligned}$$

where

s_1 = estimate of standard deviation for pre-irradiation diameter measurements

n_1 = number of pre-irradiation diameter measurements

s_2 = estimate of standard deviation for post-irradiation diameter measurements

n_2 = number of post-irradiation diameter measurements.

The number of pre-irradiation measurements for the individual pellets was not known, so it was conservatively assumed that $n_1 = 2$ for all 5 pellets. The total error in the measured diameter change was taken as $2s_R$, with s_R calculated using the uncertainties tabulated in Tables 1.1 and 1.2 for s_2 and s_1 , respectively. Values of $2s_R$ are given in Table 1.3 below.

TABLE 1.3. TOTAL UNCERTAINTY IN DIAMETER CHANGE

<u>Rod</u>	<u>Pellet</u>	<u>$2s_R$ (%)</u>
81-90	4	0.17
79-513	41	0.10
79-514	41	0.09
79-374	12	0.21
79-513	55	0.13

REFERENCE

- 1.1 O. L. Davies, Statistical Methods in Research and Production.
Published Oliver and Boyd, London (1949) pp 37, 46.

APPENDIX 2. PELLET DIAMETER CRACK CORRECTIONS

A diameter measurement bias due to pellet cracking, unlike the measurement bias due to rounded pellet edges (discussed in Appendix 1) is not negligible and can account for several mils error. Also, pellet cracking is of opposite sign from edge rounding, in that pellet cracks result in an over-estimate of true diameter whereas edge rounding results in an underestimate of true diameter.

Pellet cracking (some extensive) was observed in the ceramographic samples of 3 of the 5 pellets examined. The ceramographic samples for the 2 thorium pellets from rods 79-513 and 59-514 did not reveal cracks. Minimal cracking was observed in the ceramographic sample for the thorium pellet from rod 81-90, whereas moderate to extensive cracking was observed in the samples of binary pellets from rods 79-513 and 79-374. Photomicrographs representing the degrees of pellet cracking observed are presented in Figure 2.1.

Crack area measurements were made from 25X - 50X enlargements of the pellet cross-sections. A graduated scale with 0.01 inch divisions was used to measure crack segment lengths and a 7X magnifier with a 0.005 inch division scale was used to measure crack widths. The total crack areas measured and the percent of planar area based on PICO-MM measured diameter are given in Table 2.1.

The PICO-MM diameter was corrected for pellet cracking by subtracting crack area (ΔA) from PICO-MM measured area (A_m) to obtain a corrected area (A_c), from which the corrected pellet diameter (D_c) was calculated.

Corrected areas and diameters are given in Table 2.1. Also tabulated are the diameter decrements ($\Delta D = D_m - D_c$) due to pellet cracking.

As a check on crack area measurements, the cracks in plane 2 of the binary pellet from rod 79-374 were remeasured by another technician using the same procedure. The results are shown in Table 2.1. The repeated cracking correction resulted in a diameter 0.001 inch larger than the diameter based on the initial cracking evaluation. From the repeat corrected diameter, a diametral shrinkage of 0.98 percent during irradiation was calculated. This value is within that calculated from the initial average of planes 1 and 2, which was 1.06 ± 0.21 percent.

TABLE 2.1. MEASURED AND CORRECTED PELLET DIAMETERS

Rod	Pellet	Composition	Plane*	PICO MM Measurement		Cracks			Corrected. Dimensions	
				Diam. D_m (in)	Area** A_m (in ²)	Area ΔA (in ²)	$\Delta A/A_m$ (%)	Diam. ΔD (in)	Area** A_c (in ²)	Diam. D_c (in)
81-90	4	ThO ₂	2	0.2394	0.0450	0.0004	0.9	0.0011	0.0446	0.2383
			3	0.2393	0.0450	0.0004	0.8	0.0009	0.0446	0.2384
			Ave.	0.2394	-----	-----	---	0.0010	-----	0.2384
79-513	55	ThO ₂ -UO ₂	2	0.5054	0.2017	0.0024	1.2	0.0017	0.1993	0.5037
79-374	12	ThO ₂ -UO ₂	1	0.6456	0.3273	0.0076	2.3	0.0076	0.3197	0.6380
			2	0.6482	0.3299	0.0113	3.4	0.0113	0.3186	0.6369
			Ave.	0.6469	-----	-----	---	0.0095	-----	0.6374
			2-Repeat	0.6482	0.3299	0.0103	3.1	0.0103	0.3196	0.6379

*Plane 1 - near pellet end; Plane 2 - quarter length; Plane 3 - mid-length.

**Cylindrical pellets: Area = $\pi D^2/4$.

81-90 Pellet #4, Plane 2 10X
a) Least Severely Cracked Pellet

79-374 Pellet #12, Plane 2 5X
b) Most Severely Cracked Pellet

Figure 2.1. As-Polished Photomicrographs Showing Degrees of Pellet Cracking Observed