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IRRADIATION OF REFRACTORY FUEL COMPOUNDS, UO_2 AND UC, AT HIGH SPECIFIC POWER TO HIGH BURNUPS

FINAL REPORT

by

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Irradiation of Refractory Fuel Compounds, UO_2 and UC, at High Specific Power to High Burnups

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ABSTRACT

Thirteen fuel capsules, containing UO_2 and UC fuel pins, were designed for irradiation at power levels from 20 to 50 kw/ft, and to achieve burnups between 10,000 and 80,000 MWD/MTU. Four of the capsules were irradiated at approximately 30 kw/ft to burnups of approximately 10,000 and 60,000 MWD/MTU. The high incidence of UO_2 pin failure (7 out of 12) in these capsules resulted in re-evaluation and termination of the program. None of the UC fuel pins in these capsules failed. Four other capsules received partial irradiation, but were not examined.

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SECTION 1

INTRODUCTION

Program Description

A joint Westinghouse (NES)-NASA program* was undertaken in mid-1963 for the irradiation of refractory fuel compounds UO_2 and UC at high specific power to high burnups in the NASA Plumbrook Reactor Facility (PBRF). These fuels were recognized as having the potential of being utilized at high specific power and burnups in a variety of reactor types. However, additional irradiation data on both uranium carbides and oxides were required to establish with confidence the performance limitations of these fuels. Accordingly, an irradiation program was designed with the following variables: a) fuel rod power level, b) fuel burnup, and c) degree of restraint provided by the cladding. A series of UO_2 and UC fuel pins, in capsules of four, was scheduled for irradiation at power levels between 20 and 50 kw/ft to burnups between 10,000 and 80,000 MWD/MTU.

Westinghouse designed and fabricated the fuel-bearing capsules (a total of thirteen) and prepared a design and safeguards analysis manual and an operating procedures manual. The capsules were irradiated in holders designed to fit in the "L" holes in the primary beryllium reflector of the PBRF.

The majority of post-irradiation examinations were conducted at PBRF. In addition to disassembly, this included photography, dimensioning, fission gas collection, sectioning of fuel pins, preliminary mounting of metallographic specimens, density measurement of fuel samples, and burnup analysis by cesium-137 radiochemistry and uranium and plutonium isotopic distribution by mass spectrometry. Final preparation and photography of metallographic specimens and analysis of fission gas was done by WAPD in its own hot cells.

*Contract No. NAS3-3392-PB, July 23, 1963.

A first interim report[1] on the irradiation performance of capsule I has been published. Subsequent capsule irradiations and program changes are the primary subject of this report.

SECTION 2

CAPSULE DESIGN AND IRRADIATION PARAMETERS

2.1 Capsule and Fuel Pin Design

Table 1 summarizes the design parameters of the fuel capsules and pins fabricated for the program.

Each fuel pin was composed of a 4-inch long column of 0.300-inch diameter, 0.5-inch long, undished pellets. The UC was arc cast and hyperstoichiometric (4.9 w/o C max.). The UO_2 was nominally stoichiometric, sintered to 96 percent of theoretical density. In each pin there was a suitable diametral gap to accommodate thermal expansion of fuel operating at nominal powers of from 20 to 50 kw/ft.

The pins were clad with annealed AISI 348 stainless steel. In each capsule the cladding on the UO_2 pins was alternately 10-, 20-, and 30-mils thick and the cladding on the UC pin 20-mils thick. Each pin was surrounded by either a NaK (Capsules 1-6) or Na (Capsules 7-13) annulus contained in an outer 50-mil thick can of Type 304 stainless steel. In addition to the fuel pins, a number of tensile wires representing a variety of potential cladding materials were placed in the capsules. Three tensile wires were fitted into the NaK or Na annulus of each can, and four tensile wires were trapped in each of the four flow dividers in the capsule, for a total of 28 wires. In addition to the fuel in each pin, there was a tubular spacer above the fuel to provide a plenum for fission gas, as well as to restrict the free axial expansion of the short fuel column. There was a deformable expansion marker below the fuel intended to record the maximum relative axial motion of cladding and fuel.

Table 1

SUMMARY OF FUEL CAPSULE DESIGN

Identification: Capsule - Pin	Fuel	Enrichment: U-235, %	348 Stainless Steel Clad Wall, 10^{-3} Inch	Capsule Material
1-1	UO_2	8.5	10	Aluminum
1-2	↓	↓	20	
1-3	↓	↓	30	
1-4	UC	6.8	20	
2-1	UO_2	8.5	10	Boronated Stainless Steel
2-2	↓	↓	20	
2-3	↓	↓	30	
2-4	UC	6.8	20	
3-1	UO_2	8.5	10	Boronated Stainless Steel
3-2	↓	↓	20	
3-3	↓	↓	30	
3-4	UC	6.8	20	
4-1	UO_2	12.5	10	Aluminum
4-2	↓	↓	20	
4-3	↓	↓	30	
4-4	UC	10	20	
5-1	UO_2	12.5	10	Aluminum
5-2	↓	↓	20	
5-3	↓	↓	30	
5-4	UC	10	20	
6-1	UO_2	12.5	10	Aluminum
6-2	↓	↓	20	
6-3	↓	↓	30	
6-4	UC	10	20	
7-1	UO_2	17.5	10	Aluminum
7-2	↓	↓	20	
7-3	↓	↓	30	
7-4	UC	14	20	
8-1	UO_2	17.5	10	Aluminum
8-2	↓	↓	20	
8-3	↓	↓	30	
8-4	UC	14	20	

Table 1 (Continued)

Identification: Capsule - Pin	Fuel	Enrichment: U-235, %	348 Stainless Steel Clad Wall, 10^{-3} Inch	Capsule Material
9-1	UO_2	17.5	10	Aluminum
9-2	↓	↓	20	
9-3			30	
9-4	UC	14	20	
10-1A[a]	UO_2	12.5	10	Stainless Steel
10-2A	↓	↓	20	
10-3A			30	
10-4A	UC	10	20	
11-1A	UO_2	12.5	10	Stainless Steel
11-2A	↓	↓	20	
11-3A			30	
11-4A	UC	10	20	
12-1A	UO_2	12.5	10	Stainless Steel
12-2A	↓	↓	20	
12-3A			30	
12-4A	UC	10	20	
13(10)-3[b]	UO_2	8.5	30	Stainless Steel
13(10)-4	UC	6.8	20	
13(11)-4	↓	↓	20	
13(12)-4			20	

[a] The A stands for the alternate (higher) enrichment finally selected for the fabrication of these fuel pins.

[b] The number in parenthesis refers to the capsule originally designated for fuel of this enrichment.

Each capsule contained four fuel pins, equally spaced on a one-inch diameter circle. Four aluminum flow diverters were screwed onto the capsule body, with recesses between the body and the diverters for insertion of tensile wires. The capsule bodies were constructed with either aluminum or stainless steel; 0.010-inch thick boronated stainless steel foil was wrapped and welded around several capsules as a flux suppressor.

The capsules were irradiated in tandem pairs in a holder designed to fit into the test holes of the reactor. The capsules were removable, thus allowing periodic changes to holders composed of boronated stainless steel, stainless steel, or aluminum as required for fuel power regulation.

Detailed descriptions of the irradiation holders, test capsules, and fuel pins are given in reference 2. Pre-irradiation dimensional and fuel pellet density measurements are included. Table 1 summarizes the composition of each of the 13 capsules fabricated for the program.

2.2 IRRADIATION DESIGN; BURNUP AND POWER CALCULATIONS

2.2.1 Original Irradiation Design

The original reactor test hole locations, calculated power levels and target burnups are presented in Table 2. Several options were provided by the adopted design to maintain constant power levels of the capsules throughout their irradiation.

a. Burnable poison sleeves (boronated stainless steel) were incorporated into several capsules (Table 1) to reduce the initial neutron flux; once burned out, these sleeves cannot be replaced. Several capsule holders (Table 2) were also built with boronated stainless steel sleeves; periodic replacement with fresh holders was possible.

b. As power declined, the holder sleeve around the capsule could be replaced with another holder sleeve that depressed the flux less (e.g., an aluminum holder could replace a stainless steel holder).

c. The reactor position of the capsules could be changed to higher flux regions.

TABLE 2

DESIGN POWER LEVEL AND BURNUP

Capsule No.	Scheduled Holder Material	Scheduled Reactor Position	Mean Initial Power, Kw/ft[a]	Peak Initial Heat Flux, $BTU/hr\text{-}ft^2$ [b]	Target Burnup, (UO_2)	MWD/MTU (UC)
1	SS	LA-1	32.19	1.64×10^6	8560	6420
2	B-SS	LA-11	19.07	1.07×10^6	25480	19080
3	B-SS	LA-11	19.07	1.07×10^6	40200	30150
4	B-SS	LA-11	28.7	1.57×10^6	10140	7610
5	B-SS	LB-1	28.7	1.69×10^6	25020	18770
6	B-SS	LB-1	28.7	1.69×10^6	46280	34080
7	B-SS	LC-1	38.05	2.24×10^6	10110	7580
8	B-SS	LC-1	38.05	2.24×10^6	25600	19200
9	B-SS	LC-1	38.05	2.24×10^6	40170	30130
10	SS	LA-1	35.89	1.94×10^6	59100	44300
11	SS	LA-1	35.89	1.94×10^6	68600	51450
12	SS	LC-11	47.17	2.74×10^6	80410	60310
13	Al	LC-11	39.14	2.30×10^6	57330	43000

[a] Calculated for UO_2 fuel (fission heat only). Power levels in UC pins are higher by a factor of 1.063.

[b] Calculated for UC fuel pins at the cladding-NaK interface (Dia. = 0.344 in.); includes gamma heat but not flux uncertainty factor of 1.25.

All these methods were employed, in various combinations, for the capsules actually irradiated. Another minor device used for power regulation involved exchanging the positions of the two capsules in a holder to take advantage of slightly different axial flux levels. To schedule the absorber and reactor position changes and to predict attainment of burnups, a calculational model for power and Uranium-235 depletion was developed. The model required data on the unperturbed neutron flux in various reactor positions and data on the perturbing effects of the fuel and absorber sleeves. As a result, a number of gold foil activation experiments were made with dummy capsules containing UO_2 fuel pellets in the low-power Mockup Reactor (MUR) at the Plumbrook site. The calculational model, the MUR experiments, and applications for determining a typical capsule scheduling sequence for irradiation in the reactor are presented in reference 3.

The design power levels in Table 2 represent initial estimates of neutron flux levels and perturbation effects. Subsequent critical experiments and calculations altered the numbers listed, and resulted in changes in the program.

2.2.2 Program Change and Final Termination

Two compelling factors led to an extensive re-evaluation (in the fall of 1966) and alteration of the original irradiation schedule, as listed in Table 2.

These factors were:

a. Safety Limitations

   Flux levels determined in MUR experiments were higher than originally anticipated. This, together with a large uncertainty factor (+25%) in flux levels, made it impossible to irradiate the 17.5 w/o (UO_2) capsules within the power levels dictated by fuel pin containment can stress limitations.

b. Excessive Incidence of Fuel Pin Failures

   Irradiation of capsules 10 and 11 (12.5 w/o, ∿30 kw/ft, 60,000 MWD/MTU) was terminated and preliminary post-irradiation examination revealed probable failure in five of the six UO_2 fuel pins. In addition, single failures occurred in both capsule 1 and 4, which were irradiated to much lower burnups (∿10,000 MWD/MTU). None of the UC pins failed.

A typical UO_2 pin failure from the high burnup capsules 10 and 11 is shown in Figure 1. The outer can of fuel pin 11-3 has been longitudinally slit to reveal a failure at the top of the pin from a molten cladding. Also visible are two tensile wires (fastened at the top) which were irradiated in the liquid-metal annulus between the fuel cladding and the can.

The capsule irradiation goals listed in Table 2 were altered to assure containment can structural integrity and minimize the possibility of premature fuel pin failure. Capsules 7, 8, 9, 12 and 13 were deleted from the program; capsules 5 and 6, which had earlier experienced partial irradiation, were retained for further exposure. A target UO_2 burnup of 35,000 MWD/MTU was specified for capsules 3 (nominally 20 kw/ft) and 5 (∿30 kw/ft) and a burnup of 60,000 MWD/MTU was specified for capsules 2 (∿20 kw/ft) and 6 (∿30/ft).

The program was formally terminated by mutual consent of NASA and WAPD in late May, 1968. Up to this date irradiation of capsules 1, 4, 10, and 11 had been completed. Partial irradiation of capsules 2 and 3 had taken place in test hole LA-11 (cf. section 3), and irradiation of capsules 5 and 6 in position LA-1 had been resumed but not completed. There were several reasons for termination of the program. Since program conception in late 1962, the emphasis had changed on testing of UO_2 fuels for commercial power reactor applications. The establishment of absolute operating limits of molten UO_2 fuel was no longer of primary interest for water reactors. Furthermore, extensive tests on high-power UO_2 fuel had subsequently been reported in the literature[4,5,6]. Because the design of the Plumbrook capsules was formulated earlier, it was too late to incorporate changes (such as cored pellets) which would prevent fuel pin failures resulting from the effects of UO_2 center melting, gross fuel swelling, and possible molten fuel-cladding contact. The potential value of the Plumbrook capsules therefore amounted to an accumulation of data on fission gas release as a function of burnup, and the effect of high burnup and clad restraint on fuel swelling at moderate power levels. However, the occurrence of center fuel melting would tend to debase the post-irradiation data even if there were no fuel failures.

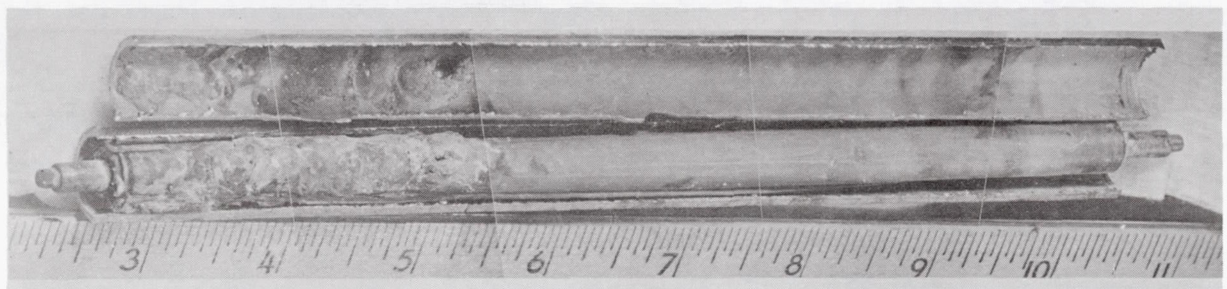

Figure 1. Failed UO_2 Fuel Pin 11A-3.

Calculations of peak power levels (based upon cycle-average flux data for LA-1 and LA-11) for six fuel capsules are given in Table 3. Instantaneous peak powers during the course of a reactor cycle would be approximately ten percent higher.

These calculations do not include the approximately 30 percent combined uncertainties or gamma heating. The presence of axial and radial flux gradients accounts for peak power levels considerably in excess of the average values. The peaks occur fairly close to the beginning of irradiation,--a condition dependent upon the employment of burnable-poison absorber sleeves. Peak power levels in the UC fuel pins are comparable (a factor of 1.063 higher), but center melting is not reached. This condition results from a comparatively high bulk thermal conductivity for UC fuel, enabling extremely high power levels to be achieved without compromising fuel pin integrity.

TABLE 3

CALCULATED PEAK POWER LEVELS: IRRADIATED UO_2 FUEL PINS

Capsule No.	Peak, kw/ft	Peak Heat Flux, $BTU/hr\text{-}ft^2$	Design, kw/ft (avg.)	Effective Full Power Time, 10^6 sec
2, 3	28.8	1.16×10^6	∿20	2.16
5, 6	42.2	1.67×10^6	∿30	3.57
10, 11	34.9	1.41×10^6	∿30	0

The discovery that five of six UO_2 fuel pins failed in capsules 10 and 11 thus indicated probable failures in capsules 5 and 6. Unfortunately, there was no method available at that time short of destructive examination, to determine whether the fuel pins (doubly encapsulated) in the latter capsules were sound. In addition, it was calculated that some center melting would occur even in the nominal 20 kw/ft UO_2 fuel pins. (A minimum of ∿26 kw/ft peak would be sufficient). For these reasons the quality of data resulting from continued irradiation was considered to be insufficient to justify post-irradiation examinations. The program was accordingly discontinued.

At the time the program was terminated the irradiation of capsules 1, 4, 10 and 11 had been completed. A comprehensive report[1] on capsule 1 has been published. Examination of the other three capsules at the PBFR hot cells included disassembly and visual examination, dimensional measurements, fuel density determinations, fission gas collection, and burnup analysis. The data (available from NASA) have not been evaluated and therefore are not presented in this report. No further post-irradiation examinations will be performed by Westinghouse. All capsules and associated hardware are being disposed of by Westinghouse. Capsules 6 and 12, and holders 018 and 019 are being retained by NASA.

A brief summary of the irradiation history of the capsules is presented in the following section.

SECTION 3

IRRADIATION HISTORY OF THE FUEL CAPSULES

A summary of the irradiation history of each fuel capsule listed in Table 1 is given below:

3.1 Capsule 1

See section 3.5 below.

3.2 Capsules 2 and 3

Capsule Pair Loading Sequence:

Reactor Position	Capsule Holder Loading				
LA-1	Upper Position	2	2	3	Out
	Lower Position	3	3	2	Out
	Reactor Cycle (beginning)	59P	61P	72P	73P
	Time (EFPT), 10^6 sec	0	2.16	11.98	13.085
	Date	3/16/67	5/15/67	12/15/67	1/5/68
	Holder Material	B-SS	B-SS[a]	Al	------

[a]New B-SS Holder at this time.

Irradiation exposure is reported as effective full power time (EFPT). Capsules 2 and 3 were fabricated with boronated stainless steel (B-SS) sleeves (Table 1). However, it was necessary to remove the B-SS wrap on these capsules prior to the beginning of irradiation to fit them into the designated holder. The result was an increase in initial power of about 25%. Irradiation of capsules 2 and 3 was terminated after cycle 72P; no post-irradiation examinations were conducted.

3.3 Capsules 4, 5 and 6

Capsule Pair Loading Sequence:

Reactor Position	Capsule Holder Loading					
LA-11	Upper Position	4	5	Out	5	Out
	Lower Position	6	6	Out	6	Out
	Reactor Cycle (Beginning)	30P	36P	38(B)P	64P	73P
	Time (EFPT), 10^6 sec	0	4.43	6.435	6.435	14.42
	Date	2/25/65	7/15/65	8/27/65	8/16/67	1/5/68
	Holder Material	B-SS	B-SS[a]	-------	B-SS[a]	------

[a]New B-SS Holder

Upon completion of irradiation, Capsule 4 had an EFPT of 4.43×10^6 sec. and an estimated average UO_2 burnup of about 11,400 MWD/MTU. Post-irradiation examination at the PBFR hot cells revealed that fuel pin 4-1 (UO_2, 12.5 w/o, 0.010 in.clad) had failed. A section of the pin had melted and disintegrated about one-third of the way from the bottom. Irradiation of capsules 5 and 6 was terminated after cycle 72P and no post-irradiation examinations were conducted.

3.4 Capsules 7, 8 and 9

These capsules were not irradiated.

3.5 Capsules 10 and 11 (and 1)

Capsule Loading Sequence:

Reactor Position	Capsule Holder Loading						
LA-1	Upper Position	1	11	10	10	A-1[a]	Out
	Lower Position	10	10	11	11	11	Out
	Reactor Cycle (Beginning)	20P	27P(II)	39P	48P	53P	57P
	Time, (EFPT), 10^6 sec	0	3.23	11.57	19.26	23.47	25.67
	Date	7/1/64	11/15/67	10/25/65	7/1/66	9/12/66	----
	Holder Material	SS	SS	SS	Al	Al	

[a]Dummy Capsule

Capsules 1 and 10 were removed for gamma scanning for the duration of cycle 21P (7/29 - 8/6/64). Capsule 1 was removed for post-irradiation examination after cycle 27P (Part I) with an exposure of 3.23 x 10^6 sec. Average measured burnup (UO_2) was about 8700-10,400 MWD/MTU. Pin 1-2 (UO_2, 8.5 w/o, 0.020 in. cladding) failed with a large cladding rupture and loss of fuel. Although a definite explanation of the failure could not be found, the experimental observations on the UO_2 fuel pins confirmed predicted behavior based upon the design mean power level (Table 2). Fuel pin 1-4 (UC, 6.8 w/o, 0.020 in. cladding) achieved an average burnup of about 9300 MWD/MTU. Fission gas release was low (0.55%), and fuel volume swelling was approximately 4 percent as determined from metallographic and density measurements. Further details are given in NASA-CR-72019[1].

Capsules 10 and 11 were removed after accumulating 23.47 x 10^6 sec and 22.44 x 10^6 sec ($\sim$60,000 MWD/MTU), respectively. During cycle 56P, capsule 11 experienced a loss-of-coolant flow transient. The capsule was removed for disassembly and examination, which revealed two UO_2 fuel pin failures. Consequently no further irradiation was scheduled for capsule 10.

Fuel pins 10-1, 10-2, and 10-3 (UO_2, 12.5 w/o) all failed; fuel pins 11-1 and 11-3 (UO_2, 12.5 w/o) also failed. The failures were characterized by clad melting and various degrees of disintegration of the fuel pins (c.f. Figure 1). Neither UC fuel pin (10-4, 11-4) failed. No further post-irradiation examinations were performed after initial examinations at the PBRF Hot Cells.

3.6 Capsules 12 and 13

These capsules were not irradiated (Capsule 12 was used in MUR critical experiments).

SECTION 4

SUMMARY AND CONCLUSIONS

Thirteen fuel containing both UO_2 and UC fuel pins was designed for irradiation at lower levels from 20 to 50 kw/ft and to burnups between 10,000 and 80,000 MWD/MTU. Four of these capsules were irradiated at about 30 kw/ft to burnups of 10,000 and 60,000 MWD/MTU. The high incidence of UO_2 fuel pin failure (7 out of 12) and the fixed capsule design led to a re-evaluation, and ultimately to termination, of the program. None of the UC fuel pins failed in these capsules. Of these four capsules, one (number 1, about 30 kw/ft and 10,000 MWD/MTU) was fully examined and evaluated; the results were consistent with design predictions. Four other fuel capsules were partially irradiated, but received no post-irradiation examination. The remaining five capsules were not irradiated.

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The author wishes to acknowlege the major contribution to this program by R. E. Schreiber (WAPD) who was the responsible engineer until June, 1966. The author also expresses his appreciation to R. F. Barrows (NASA-Plumbrook) for his contributions and co-operation.

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