~~CONFIDENTIAL~~

Paper to be Presented at the RDT/SNS Meeting on Materials Development for Space Reactor Power Systems, U. S. Atomic Energy Commission, Washington, D. C., April 23-24, 1968.

SPECIAL REREVIEW FINAL DETERMINATION	Reviewers	Class.	Date
	KAW	U	12-7-81
	HFC	U	12-7-81
Class: <u>U</u>			

STATUS OF URANIUM NITRIDE DEVELOPMENT PROGRAM
AT THE OAK RIDGE NATIONAL LABORATORY*

Part II

Compiled by

S. C. Weaver and J. L. Scott

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

CLASSIFICATION CANCELLED
DATE 11-27-73
For the Atomic Energy Commission

TED REDMON *TR* for the
Chief, Declassification Branch

PRELIMINARY DATA ONLY

This document contains information of a preliminary nature and is subject to revision or correction and therefore does not represent a final report. The information is not to be abstracted, reprinted or otherwise given public dissemination without the approval of the ORNL patent branch, Legal and Information Control Department.

*Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

~~RESTRICTED DATA~~

"This document contains restricted data defined in Executive Order 11652. Its transmission or disclosure to unauthorized persons is prohibited by law."

MASTER

~~CONFIDENTIAL~~

DISSEMINATION OF THIS DOCUMENT IS UNLIMITED
GOVERNMENT AGENCIES AND THEIR CONTRACTORS & EMPLOYEES
GG

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

~~CONFIDENTIAL~~

2

CONTENTS

Page

Continuation of CAMEL-UN Irradiations-----	3
Irradiation Testing at the Low Intensity Test Reactor-----	10
Irradiation Testing at the Oak Ridge Research Reactor-----	10

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

3

STATUS OF URANIUM NITRIDE DEVELOPMENT PROGRAM
AT THE OAK RIDGE NATIONAL LABORATORY

Part II

CONTINUATION OF CANEL-UN IRRADIATIONS

S. C. Weaver E. L. Long, Jr.
R. L. Senn B. H. Montgomery

When Pratt and Whitney-CANEL ceased operation in July, 1965, the Oak Ridge National Laboratory assumed responsibility for continuation of the CANEL uranium-nitride irradiation program at the Materials Testing Reactor. The program, as received by ORNL, included six capsules undergoing irradiation testing, four that had been tested and removed, and five capsules that were completely constructed and ready for insertion. All 15 capsules were designed to test UN at SNAP-50 Reference Design No. 3 conditions. These conditions, together with design limitations, are shown in Table 1.

Table 1. SNAP-50 Reference Design No. 3 Conditions

Thermal power level	2 Mw
Maximum fuel temperature	1260°C
Maximum power density	0.4 kw/cc
Maximum fuel burnup	1.5 at. % U
Maximum fission-gas release	20%
Maximum diametral pin swelling	2%
Maximum fuel density decrease	8%

A summary of the conditions and status of all 15 capsules is presented in Table 2. It is to be noted that eight capsules operated successfully for design irradiation times of approximately 10,000 hr. Four of the five untested capsules were modified at ORNL so as to achieve higher power densities and, therefore, burnups than initially intended. In two of these capsules (Nos. 57-612 and -665) the centerline temperatures were maintained at 1300°C and the cladding temperature was reduced to approximately 980°C. In the third (capsule 57-669) the

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

4

Table 2. Conditions and Status of ORNL Uranium Nitride Irradiation Tests

ORNL Capsule Number	Insertion Date	Average Cladding Temperature ^a (°C)	Final Burnup ^b (at. % U)	Calc. Power Density (kw/cc)	Test Time Through 3-15-68 (hr Above 35 Mw)	Discharge Date	Current Status
<u>Capsules Tested in MTR</u>							
57-612	4-25-66	980	2.54	1.41	5205	1-23-67	PIE ^d Complete
57-642	9-2-64	1180	1.18	0.34	9966	1-31-66	PIE Complete
57-643	11-30-65	1180	0.85	0.57	4292	9-17-65	PIE Complete
57-649	3-14-66	1090	1.5 ^c	0.4 ^c	10,512	10-9-67	In PIE
57-652	12-21-64	1096 (1000 hr)	0.7 ^e	0.64 ^e	3180	6-14-65	PIE Complete
		1205 (2180 hr)					
57-656	12-21-64	1095 (1000 hr)	0.6 ^e	0.53 ^e	3180	6-14-65	PIE Complete
		1205 (2180 hr)					
57-658	3-2-65	1200	1.08	0.32	9779	7-18-66	PIE Complete
57-660	1-21-65	1095 (455 hr)	0.7 ^e	0.76 ^e	2635	6-14-65	PIE Complete
		1205 (2180 hr)					
57-662	3-25-65	1200	1.16	0.37	10,270	8-29-66	PIE Complete
57-664	5-25-65	1202	0.58	0.27	6209	4-4-66	PIE Complete
57-665	4-25-66	965	5.0	1.5 ^c	9583	10-9-67	In PIE
57-667	5-25-65	1210	0.78	0.36	6209	4-4-66	PIE Complete
57-669	4-25-66	980	3.0 ^c	0.8 ^c	10,352	11-20-67	In PIE
57-002	6-15-65	1200	1.12	0.32	10,173	11-20-66	PIE Complete
57-003	5-16-66	1200	4.0 ^c	0.8 ^c	11,985	3-4-68	At ORNL
<u>Capsules Under Test in ORR</u>							
05-UN1	3-5-67				5592	5-26-67	Pos. 06
TTS ^f		920	2.0 ^c	0.43			
MTS ^g		1100	2.0 ^c	0.75			
BTS ^h		1200	2.0 ^c	0.61			
05-UN2	12-15-67				220	7-69	Retracted
TTS		1	2.0 ^c	0.55			
MTS		1175	2.0 ^c	0.79			
BTS		1350	2.0 ^c	0.62			

^aValues cited for capsules under test are based on current conditions and do not represent a time average since the start of the tests.

^bBased on isotopic analyses unless otherwise stated.

^cDesign estimate.

^dPostirradiation examination.

^eBased on heat balance calculations. These values are probably high.

^fTTS: Top test specimen.

^gMTS: Middle test specimen.

^hBTS: Bottom test specimen.

ⁱIndicated cladding temperature surged to 1545°C due to loss of NaK around top test specimen. (See text.)

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

5

centerline temperature was reduced to 1100°C and the cladding temperature to 980°C, while in the fourth (capsule 57-003) the centerline temperature was increased to approximately 1350°C and the cladding temperature was maintained at 1200°C.

Schematic drawings of the fuel pins and capsule design are shown in Figs. 1 and 2, respectively. All fuel pins in these tests contained cold-pressed-and-sintered UN pellets clad with PWC-11 (Nb-1% Zr-0.1% C). A 5-mil tungsten reaction barrier was deposited by the chemical vapor deposition technique on the inside surface of the cladding. The fuel pellets were in the form of 0.188-in.-diam right-circular cylinders. The cladding dimensions were 0.25-in.-ID and 0.025-in. wall thickness. Most capsules contained three fuel pins with a fuel length of 0.65 in. However, three capsules contained single pins with a fuel length of 3.3 in. All fuel pins were immersed in lithium and the cladding surface temperature for pins in the unmodified capsules was 1200°C.

The results of postirradiation examinations are summarized in Table 3. It should be noted that except for capsule 57-003, in which the estimated burnup was 4 at. % U, the fuel pins performed better than the performance criteria designated in Table 1. Fission-gas releases were very small-6% or less. The volumetric increase of the fuel was approximately 1.7% per at. % U burnup at the maximum burnup of 5 at. % U.

The cause of the relatively large volume- and pin-diameter increases in capsule 57-003 has not been established. Metallographic examination of this capsule is now in progress. The capsule was irradiated for 11,985 hr at a cladding temperature of 1200°C. Capsule 57-665, irradiated to 5 at. % burnup, operated at a cladding temperature of only 980°C. Thus, it may be that the higher strength of the cladding at 980°C reduced the fuel swelling in 57-665 and caused most of the swelling to be expressed axially instead of radially.

The PWC-11 cladding performed very well under the service conditions of 980 and 1200°C. No failures or evidences of lithium corrosion were observed. The only detrimental effect was a 0.001-in. reaction zone at the tungsten barrier-cladding interface. This reaction zone consisted of a region of niobium-tungsten interdiffusion and a region containing a U-Nb intermetallic. At 980°C this reaction zone was less than 0.0001 in.

~~CONFIDENTIAL~~

CONFIDENTIAL
ORNL-DWG 66-10391B

Fig. 1. Schematic Drawing of CAMEL-UN Fuel Pins.

CONFIDENTIAL
ORNL-DWG 67-6367R

Fig. 2. Schematic Drawing of CANEL-UN Irradiation Capsule.

~~CONFIDENTIAL~~

8

Table 3. Results of Postirradiation Examination of CANEL-UN Capsules

Capsule Number ^a	Average Cladding Temperature (°C)	Burnup ^b (at. % U)	Calculated Power Density (w/cm ³)	Fission Gas Release (%)	Fuel Density Decrease (%)	Fuel Volume Increase (%)	Change in Pin Diameter (%)	$\left(\frac{\Delta r}{r} / \frac{\Delta V}{V}\right)$
57-612	980	2.54	1.41					
TTS				0.05	4-6 ^c	4-6 ^c	0.73	0.12-0.18
MTS				0.37	4-6 ^c	4-6 ^c	1.08	0.18-0.27
BTS				0.09	4-6 ^c	4-6 ^c	0.77	0.13-0.19
57-642	1180	1.18	0.34	0.34	4.0	4.1	0.80	0.20
57-643	1180	0.85	0.57	d	3.2	3.3	0.36	0.11
52-649	1135	1.5 ^e	0.4 ^e	0.038	2.66	2.74	0.59	0.21
57-652	1095 (1000 hr)	0.7 ^f	0.64 ^f					
TTS				g	h	h	nil	h
MTS				0.09	h	h	0.16	h
BTS				g	h	h	0.20	h
57-656	1095 (1000 hr) 1205 (1000 hr)	0.6 ^f	0.53 ^e					
TTS				0.03	h	h	0.24	h
MTS				0.11	h	h	0.12	h
BTS				0.13	h	h	0.48	h
57-658		1.08	0.32					
TTS				d	3.7	3.8	nil	0.00
MTS				0.14	3.8	4.0	0.18	0.05
BTS				0.09	3.5	3.6	0.12	0.03
57-660	1095 (1000 hr) 1205 (1000 hr)	0.7 ^f	0.76 ^f					
TTS				g	h	h	0.28	h
MTS				0.05	h	h	0.28	h
BTS				g	h	h	0.20	h
57-662	1200	1.16	0.37					
TTS				0.01	4.1	4.3	0.47	0.11
MTS				0.08	4.3	4.5	0.43	0.09
BTS				0.04	4.5	4.7	0.40	0.08
57-664	1202	0.58	0.27					
TTS				< 0.01	1.8	1.8	0.16	0.09
MTS				< 0.01	1.4	1.5	0.08	0.05
BTS				< 0.01	1.2	1.2	0.24	0.19
57-665	965	5.0 ^o	1.5 ^e					
TTS				1.36	7.1	7.7	1.00	0.13
MTS				5.96	7.9	8.5	1.52	0.18
BTS				1.78	7.7	8.3	1.27	0.15
57-667	1210	0.78	0.36					
TTS				< 0.01	3.0	3.1	nil	0.00
MTS				0.02	1.7	1.8	0.12	0.07
BTS				0.03	1.8	1.9	0.16	0.09
57-669	980	3.0 ^e	0.8 ^e					
TTS				i	6.1	6.5	0.52	0.08
MTS				i	6.9	7.4	0.36	0.05
BTS				i	5.6	6.0	0.36	0.06
57-002	1200	1.12	0.32					
TTS				< 0.02	2.4	2.5	0.39 ^j	0.16 ^j
MTS				< 0.02	2.2	2.3	0.55 ^j	0.24 ^j
MTS				0.06	1.9	2.0	0.71 ^j	0.36 ^j
BTS				d	k		0.75 ^j	
57-003	1200	4.0 ^d	0.8 ^e					
TTS				i	9.7	10.7	1.42	0.13
MTS				i	9.4	10.4	1.90	0.18
MTS				i	9.3	10.2	2.02	0.20
BTS				i	10.8	12.1	1.90	0.16

^aTTS-top test specimen; MTS-middle test specimen; BTS-bottom test specimen.^bBased on isotopic analyses unless otherwise stated.^cEstimated values; preirradiation inspection data not available.^dSample lost.^eDesign estimate.^fBased on heat balance calculations. These values are probably high.^gNo sample taken.^hNot measured.ⁱData not yet available.^jThese numbers are based on conservative calculations and are probably high.^kAll pellets were split during handling of this pin and the resulting densities are thought to be erroneous.~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

in width. The reaction did not appear to be detrimental to fuel pin performance for the times and temperatures achieved in these experiments.

Metallographic examination revealed the presence of fission-gas bubbles in the microstructure of the fuel pellets. In the pins that operated at a cladding temperature of 1200°C the fission-gas bubbles migrated radially up the temperature gradient until they reached a grain boundary where they became trapped. This effect was particularly evident in regions near the surface of the pellets where the temperature gradient was largest. The result of this oriented bubble migration is that a preponderance of bubbles exists on grain boundaries parallel to the fuel surface. No bubbles were observed at grain boundaries near the center of the pellets where (1) the relatively flat temperature profile apparently did not provide a significant driving force for bubble migration and (2) fewer gas atoms were present due to the thermal flux depression.

Reducing the cladding temperature from 1200 to 980°C, while maintaining the same fuel centerline temperature of 1300°C, provided a means for assessing the effects of higher linear heat ratings. In these tests the mobility of the fission-gas bubbles near the periphery was significantly reduced. It was observed that under these conditions very few bubbles reached grain boundaries by radial migration, in spite of the fact that the temperature gradient was increased by a factor of four relative to that for specimens that operated at cladding temperatures of 1200°C. Because of the axial gradient, however, the gas bubbles were observed to line up at grain boundaries parallel to the end of the fuel column. Apparently the temperature of the pellets at the end of the fuel stacks, but near the radial center, was high enough so that the bubbles were mobile.

Electron microscopy of fracture surfaces of the fuel from capsule 57-612 (2.54 at. % U burnup) illustrated the distribution of fission gases. Near the periphery the fracture was intergranular and large bubbles were observed to have coalesced at the grain boundaries. At about midradius the fracture was transgranular and bubbles were observed within the grains, which were approximately 800 Å in diameter. Near the center the fuel appeared to have flowed plastically, indicating that

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

10

restraint by the cladding and the outer regions of the fuel had caused the fuel in the center to extrude axially.

IRRADIATION TESTING AT THE LOW INTENSITY TEST REACTOR

W. S. Ernst

In an earlier program, six capsules were irradiated in the Low Intensity Test Reactor; the cladding on three capsules was D-43 (Nb-10% W-1% Zr-0.1% C) and in the other three was T-111 (Ta-8% W-2.5% Hf). Five of the capsules contained two fuel pins each and a sixth capsule contained a single fuel pin with annular fuel pellets and a central thermocouple. The conditions and results of these tests are presented in Table 4.

To examine the restraining effect of the D-43 and T-111 we plotted the ratio of the fractional radial increase ($\Delta r/r$) of the cladding to the fractional volume increase ($\Delta V/V$) of the fuel, as a function of cladding temperature. The results are shown in Fig. 3 and clearly indicate that the more creep-resistant T-111 cladding provided greater restraint than the D-43 cladding.

IRRADIATION TESTING AT THE OAK RIDGE RESEARCH REACTOR

S. C. Weaver

K. R. Thoms

V. A. DeCarlo

Based on the success and information generated by the CAMEL and LITR programs we have initiated a third UN irradiation program in the ORR. The capsules in this program are designed to test UN at more severe conditions. Some of the design features are (1) use of higher strength cladding (W-26% Re), (2) operation at fuel centerline temperatures of 1550°C, (3) operation to higher burnups, and (4) improved capsule instrumentation.

A schematic diagram of the capsule design is shown in Fig. 4 and specific design parameters are listed below.

~~CONFIDENTIAL~~

Table 4. Conditions and Results of LITR-UN Irradiation Tests

Pin No.	Power Density (kw/cm ³)	Exposure Time (hr)	Estimated Burnup (% U)	Temperatures			Fuel Density Decrease (%)	Fuel Volume Increase (%)	Change in Pin Diameter (%)
				End Cap Measured (°C)	Estimated Peak (°C)				
					Cladding	Fuel			
LN-1T	0.95	5397	2.0	695	1030	1135	5	5.3	< 0.4
LN-1B				780	1115	1220	7 ^a	14	5
LN-2T	0.52	8056	1.6	800 ^b	1135	1240	9.0	9.9	0.7
LN-2B				1050	1385	1490	c	c	1.6 ^d
LN-3T	1.02	7045	2.8	655	990	1095	5.3	5.6	< 0.4
LN-3B				970	1305	1410	8.2	8.9	0.8
LN-6T	0.95	5520	1.8	800	1135	1240	5.0	6.4	1.00
LN-6B				990	1325	1430	12.8	14.7	4.3 ^e
LN-4T	0.92	4540	2.0	780	1115	1220	c	c	c
LN-4B				980	1315	1420	c	c	c
LN-5	1.19	1310	2.0	820	1230	1315 ^f	c	c	c

^aThis value is thought to be inaccurate (too low).

^bWall thermocouple failed at startup; temperatures listed for LN-2T are rough estimates.

^cNot determined.

^dIn the fuel region, the radial increase was 1.6%, but in the ruptured zone an increase of 5.7% was measured.

^eIn fuel region where extensive grain growth occurred and a color change was noted.

^fMeasured fuel center temperature.

CONFIDENTIAL

~~CONFIDENTIAL~~

12

ORNL-DWG 67-1376

Fig. 3. Effect of Cladding Restraint on Diametral Swelling of UN Fuel Pins.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

13

CONFIDENTIAL
ORNL-DWG 67-983R

Fig. 4. ORR-UN Irradiation Capsule.

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

14

Scheduled irradiation time: 10,000

Estimated burnup: 3 at. % U

Cladding

Temperature:	1370°C
Wall thickness:	0.030 in.
Inside diameter:	0.305 in.
Outside diameter:	0.365 in.
Material:	W-26% Re

Fuel

Top test pin:	Annular cold-pressed and sintered pellets 0.105 in. central hole 0.300 in. OD
Middle test pin:	Single crystal and cold-pressed and sintered pellets, 0.250 in. OD immersed in lithium
Bottom test pin:	Cold-pressed-and-sintered pellets 0.300 in. OD
Fuel centerline temperature	1550°C

As indicated in Table 2, two capsules are now at the ORR. One is operating satisfactorily, but the second, having operated in an over-temperature condition, has been retracted. Attempts are being made to correct the problem which we attribute appears to be associated with a loss of NaK in the Nb-1% Zr can.

~~CONFIDENTIAL~~