21-1293

ORNL-TM-4397
(Addendum)

An Irradiation Test of Candidate HTGR Recycle Fuels in the H-1 and H-2 Capsules

R. A. Olstad
E. L. Long, Jr.

MASTER

OAK RIDGE NATIONAL LABORATORY

OPERATED BY UNION CARBIDE CORPORATION • FOR THE U.S. ATOMIC ENERGY COMMISSION

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
Price: Printed Copy \$4.00; Microfiche \$2.25

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Energy Research and Development Administration, 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.

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.

ORNL-TM-4397 (Addendum)
UC-77 - Gas-Cooled
Reactor Technology

Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION

AN IRRADIATION TEST OF CANDIDATE HTGR RECYCLE FUELS
IN THE H-1 AND H-2 CAPSULES: ADDENDUM

R. A. Olstad and E. L. Long, Jr.

APRIL 1975

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, 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.

NOTICE This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
ENERGY RESEARCH AND
DEVELOPMENT ADMINISTRATION

MASTER

DISTRIBUTION OF THIS DOCUMENT UNLIMITED

.

.

.

.

.

.

CONTENTS

ABSTRACT	1
INTRODUCTION	1
IRRADIATION CONDITIONS	2
METALLOGRAPHY OF ROD H-2-5	3
METALLOGRAPHY OF ROD H-1-9	5
CONCLUSIONS	7
ACKNOWLEDGMENTS	11

AN IRRADIATION TEST OF CANDIDATE HTGR RECYCLE FUELS IN THE
H-1 AND H-2 CAPSULES: ADDENDUM

R. A. Olstad¹ and E. L. Long, Jr.

ABSTRACT

Two H-1 and H-2 capsule fuel rods, each containing Bisocoated ThC₂ and Triso-coated UC₂ particles, were examined metallographically after the examination of these capsules had been reported. Rods H-2-5 and H-1-9 operated at maximum center-line temperatures of approximately 1400°C and 1720°C, respectively. The metallography of rod H-2-5 revealed about 50 μm of ThC₂ kernel migration, no UC₂ kernel migration, some fission product attack of the SiC coatings, and graphitization of the inner pyrocarbon coatings at the cold side of the UC₂ particles. Rod H-1-9 exhibited extensive ThC₂ kernel migration (approximately 250 μm), no migration in most UC₂ particles, and considerable fission product attack of the SiC coatings.

INTRODUCTION

The design, operating conditions, and results of the H-1 and H-2 capsule irradiation tests have been reported in detail previously.² Since the time that report was prepared, two fuel rods (rods H-2-5 and H-1-9) that contained Triso-coated uranium carbide and Bisocoated thorium carbide particles have been examined metallographically. The metallography of these two fuel rods was not included in the original report because polishing procedures for ThC₂ had not yet been developed. The purpose of this Addendum is to report those additional metallographic observations.

¹Now with General Atomic Company

²R. A. Olstad, A. R. Olsen, R. B. Fitts, E. L. Long, Jr., and T. B. Lindemer, *An Irradiation Test of Candidate HTGR Recycle Fuels in the H-1 and H-2 Capsules*, ORNL-TM-4397 (July 1974).

IRRADIATION CONDITIONS

The H-1 and H-2 capsules operated normally for the first three irradiation cycles, but were inadvertently irradiated in an inverted position during the fourth cycle. During this last cycle the fuel samples in the top half of the capsules operated at temperatures much higher than the design temperatures.

The calculated temperatures of rods H-2-5 and H-1-9 at various times during the irradiation test are given in Table 1. The assumptions used in calculating these temperatures have been described in detail previously.² Rod H-2-5 operated at approximately the same temperatures before and after the capsules were inverted, but rod H-1-9 operated much hotter after the capsules were inverted. The calculated temperatures have a large uncertainty because of uncertainties in fission and gamma heat rates, gap widths, and fuel rod thermal conductivity. The temperatures for rod H-2-5 are uncertain by approximately $\pm 150^{\circ}\text{C}$, while those for rod H-1-9 are uncertain by approximately $\pm 235^{\circ}\text{C}$ during the last irradiation cycle.

Table 1. Calculated Temperatures of Rods H-2-5 and H-1-9

	Temperature, $^{\circ}\text{C}$			
	Rod H-2-5		Rod H-1-9	
	surface	center line	surface	center line
Beginning of First Cycle ^a	1010	1400	800	1150
End of Third Cycle	970	1350	860	1200
Beginning of Fourth Cycle	940	1300	1180	1720
End of Fourth Cycle	880	1200	1050	1480

^aThe H-1 and H-2 capsules operated for 97.1 and 85.6 effective full-power days, respectively, in their proper orientation during the first three cycles, but operated for 39.3 EFPD inverted during the fourth cycle.

The fast neutron fluence obtained in rods H-2-5 and H-1-9 was approximately 5.9×10^{21} n/cm² (>0.18 MeV). The burnups of the UC₂ particles in rods H-2-5 and H-1-9 were calculated to be 39 and 44% FIMA, respectively. The burnup of the ThC₂ particles was calculated to be approximately 0.4% FIMA in both rods.

METALLOGRAPHY OF ROD H-2-5

Rod H-2-5 was badly fragmented and debonded at the time the capsules were disassembled. Several random fragments from this rod were examined metallographically, but since their location in the rod could not be determined, the temperatures and temperature gradients in these fragments could only be roughly estimated.

Substantial migration of the ThC₂ kernels in the H-2-5 fragments was observed, but no migration of the UC₂ kernels could be detected. Fig. 1 shows adjacent ThC₂ and UC₂ particles as they appear under bright field and under polarized light. There is no observable migration of the UC₂ kernel, whereas the ThC₂ kernel has migrated approximately 20 μm under the same temperature conditions. A region of rejected carbon behind the migrating ThC₂ kernel appears white under polarized light. A ThC₂ kernel that migrated about 50 μm is shown in Fig. 2. The measured extent of migration of the ThC₂ kernels in the fragments of rod H-2-5 ranged from 7 to 67 μm. The approximate ThC₂ kernel migration coefficient determined from these data is reported elsewhere.³

A typical UC₂ particle in this sample is shown under bright field and polarized light in Fig. 3. The SiC coatings on the UC₂ particles showed some chemical attack and some cracking. There was also a very noticeable graphitization of the inner pyrocarbon layer on the cold side

³T. B. Lindemer and R. A. Olstad, *HTGR Fuel Kernel Migration Data for the Th-U-C-O System as of April 1, 1974*, ORNL-TM-4493 (June 1974).

Fig. 1. ThC₂ and UC₂ Particles Irradiated in Rod H-2-5. (a) Bright field. (b) Polarized light. Note that the ThC₂ kernel migrated and the UC₂ kernel did not.

Fig. 2. ThC_2 Particle Irradiated in Rod H-2-5. Note the considerable migration of the kernel up the temperature gradient.

of the particles, as indicated by its white appearance under polarized light. The SiC attack and graphitization of the inner pyrocarbon layer are shown more clearly under high magnification. Electron microprobe analysis has shown that rare-earth fission products are deposited in the attacked regions of the SiC and in the graphitized region of the inner pyrocarbon layer. This indicates that rare-earth fission products are involved in both these phenomena.

METALLOGRAPHY OF ROD H-1-9

Rod H-1-9 was also badly fragmented at the time of capsule disassembly, but the fragments were held in place by putting them in epoxy before removal from the capsule. The ThC_2 kernels in this rod migrated much more than they did in rod H-2-5. In most cases the ThC_2 kernels migrated through both the buffer and pyrocarbon coatings, and frequently migrated into the coatings of the adjacent particles. The measured extent of migration of the ThC_2 kernels in this rod varied from 50 to 350 μm . The kernel migration

Fig. 3. UC₂ Particle Irradiated in Rod H-2-5. (a) Bright field; showing fission product attack of the SiC. (b) Polarized light, showing graphitization of the inner pyrocarbon layer at the cold side of the particle. (c) Same particle at higher magnification. Note the fission products deposited in the SiC and inner pyrocarbon coatings.

coefficients determined from these data have been reported previously.³ Migration of UC_2 kernels was detectable in only a few of the UC_2 particles. The SiC coatings on the UC_2 particles showed considerable attack and some cracking.

A metallographic cross section of rod H-1-9 (Fig. 4) shows the extensive migration of the ThC_2 kernels throughout the rod. Figure 5 shows at increased magnification the migration of a ThC_2 kernel through its coating and into the outer pyrocarbon and SiC coatings of adjacent particles. The ThC_2 kernel shown in Fig. 6 has "devoured" the adjacent particle. The polarized light view of this ThC_2 particle [Fig. 6.(b)] shows only a small region of rejected graphite behind the kernel, but numerous graphite flakes have precipitated throughout the kernel.

The UC_2 particle shown in Fig. 7 exhibits both SiC attack and graphitization of the inner pyrocarbon layer at the cold side of the particle. The SiC coatings on the UC_2 particles in rod H-1-9 exhibited more rare-earth fission product attack than those in rod H-2-5 because of the higher operating temperatures of rod H-1-9.

CONCLUSIONS

1. Rod H-2-5, which operated at temperatures as high as $1400 \pm 150^\circ C$ exhibited: (a) 7 to 67 μm of ThC_2 kernel migration, (b) no UC_2 kernel migration, (c) rare-earth fission product attack of the SiC coatings on the UC_2 particles, and (d) graphitization of the inner pyrocarbon layers at the cold side of the UC_2 particles.

2. Rod H-1-9, which operated at temperatures as high as $1720 \pm 235^\circ C$, exhibited: (a) 50 to 350 μm of ThC_2 kernel migration, (b) no observable migration in all but a few of the UC_2 particles, (c) substantial fission product attack of the SiC coatings, and (d) some graphitization of the inner pyrocarbon layers of the UC_2 particles.

R-64175

Fig. 4. Diametral Section of Rod H-1-9, Which Contained Biso-Coated ThC_2 and Triso-Coated UC_2 Particles. A portion of the graphite sleeve is attached to the rod. The rod operated at a maximum surface temperature of approximately 1200°C and a maximum center-line temperature of approximately 1700°C .

Fig. 5. ThC_2 Kernel Migrating Through Its Own Coatings and Into the Coatings of Adjacent Particles. These particles were near the edge of rod H-1-9 and operated at a maximum temperature of about 1350°C in a temperature gradient of about $1400^\circ\text{C}/\text{cm}$.

Fig. 6. ThC₂ Particle Irradiated in Rod H-1-9. (a) Bright Field, showing migration of kernel into adjacent particle. (b) Polarized light, showing precipitation of graphite flakes throughout the kernel. The maximum temperature and temperature gradient were approximately 1450°C and 1200°C/cm, respectively.

Fig. 7. Typical UC₂ Particle Irradiated in Rod H-1-9. Note the extensive fission product attack of the SiC coating.

ACKNOWLEDGMENTS

The authors would like to acknowledge F. J. Furman for applying coatings in the prototype production coater and loading the fuel assemblies, C. B. Pollock for applying coatings in the laboratory coater, and J M Robbins and F. P. Jeffers for characterizing the particles and fabricating the fuel rods. Gulf General Atomic provided the carbide fuel particles. M. K. Preston and J. T. East fabricated the capsules and assured quality control. Bettis Atomic Power Laboratory provided the hafnium-zirconium alloys, and R. G. Donnelly and co-workers fabricated the hafnium sleeves. The Idaho Nuclear Corporation provided information needed for the experiment design and monitored the fluxes during irradiation in the ETR. J. D. Jenkins chose the capsule flux monitors and set up the XSDRN calculations. A special acknowledgement is given to H. T. Kerr for interpreting the flux monitor data and the XSDRN calculations. R. A. Buhl

assisted in the postirradiation examination and set up the depletion and heat transfer calculations. Metallography of the irradiated samples was performed by N. M. Atchley, and the electron microprobe analyses were performed by T. J. Henson. Julia Bishop of the Metals and Ceramics Division Publications and Records Office prepared the manuscript for reproduction.

ORNL-TM-4397
Addendum
UC-77 — Gas Cooled
Reactor Technology

INTERNAL DISTRIBUTION

R. L. Beatty	A. P. Malinauskas
E. S. Bettis (consultant)	W. R. Martin
R. A. Bradley	B. H. Montgomery
A. J. Caputo	H. C. McCurdy
J. M. Chandler	M. T. Morgan
T. E. Cole	K. J. Notz
J. A. Conlin	(2) A. R. Olsen
J. H. Coobs	M. F. Osborne
D. A. Costanzo	P. Patriarca
J. E. Cunningham	R. L. Pearson
H. J. de Nordwall	W. H. Pechin
F. F. Dyer	H. Postma
W. P. Eatherly	P. L. Rittenhouse
R. B. Evans III	J M Robbins
J. I. Federer	J. L. Scott
D. E. Ferguson	J. D. Sease
(2) R. B. Fitts	G. M. Slaughter
C. L. Fitzgerald	J. W. Snider
R. J. Gray	K. R. Thoms
P. A. Haas	T. N. Tiegs
R. L. Hamner	D. B. Trauger
(3) M. R. Hill	K. H. Valentine
F. J. Homan	J. E. Van Cleve
J. D. Jenkins	V.C.A. Vaughn
M. S. Judd	T. N. Washburn
M. J. Kania	J. R. Wier, Jr.
(38) P. R. Kasten	G. V. Smith (consultant)
H. T. Kerr	John Moteff (consultant)
E. M. King	J. W. Prados (consultant)
W. J. Lackey, Jr.	(3) Central Research Library
(5) T. B. Lindemer	ORNL — Y-12 Technical Library
(15) E. L. Long, Jr.	Document Reference Section
A. L. Lotts	(5) Laboratory Records Department
R. S. Lowrie	Laboratory Records, ORNL R. C.
R. E. MacPherson	ORNL Patent Office

EXTERNAL DISTRIBUTION

USERDA DIVISION OF REACTOR RESEARCH AND DEVELOPMENT, Washington, DC 20545
Director

USERDA OAK RIDGE OPERATIONS OFFICE, P.O. Box E, Oak Ridge, TN 37830
Research and Technical Support Division
Reactor Division

For distribution as shown in TID-4500 Distribution Category, UC-77,
Gas-Cooled Reactor Technology (25 copies — NTIS)