LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES
3 4456 0514403 7

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION • NUCLEAR DIVISION
for the
U. S. ATOMIC ENERGY COMMISSION

ORNL-TM-3361

Cy 37

PERFORMANCE OF BONDED BEDS OF COATED FUEL PARTICLES DURING IRRADIATION

P. E. Reagan
J. G. Morgan
E. L. Long, Jr.
J. H. Coobs

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.
UCN-7969
(3-67)

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.

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.

ORNL-TM- 3361

Contract No. W-7405-eng-26

REACTOR CHEMISTRY DIVISION
and
METALS AND CERAMICS DIVISION

PERFORMANCE OF BONDED BEDS OF COATED FUEL PARTICLES DURING IRRADIATION

P. E. Reagan E. L. Long, Jr.
J. G. Morgan J. H. Coobs

JUNE 1971

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES

3 4456 0514403 7

CONTENTS

	Page
Abstract	1
Introduction	1
Test Specimens	2
Experimental Method	2
Irradiation Tests	7
Uranium-Oxide-Fueled Bonded-Bed Experiment A9-15	7
Uranium-Carbide-Fueled Bonded-Bed Experiment B9-40	8
Postirradiation Examination	10
Uranium-Oxide-Fueled Bonded-Bed Experiment A9-15	10
Uranium-Carbide-Fueled Bonded-Bed Experiment B9-40	10
Discussion and Conclusions	13
Acknowledgments	16
References	17

PERFORMANCE OF BONDED BEDS OF COATED FUEL PARTICLES DURING IRRADIATION

P. E. Reagan E. L. Long, Jr.
J. G. Morgan J. H. Coobs

ABSTRACT

One bonded-bed experiment was operated to 35.5% heavy metal burnup at 1250°C with no evidence of failure or signs of potential failure. Another experiment was operated to 18% heavy metal burnup at temperatures up to 1700°C. Although the bonded bed remained intact the fuel migrated through the particle coatings in the direction of the highest temperature (the "Amoeba" effect). Failure of the coatings appeared to start at about 1500°C.

INTRODUCTION

One of the leading concepts of the High-Temperature Gas-Cooled Reactor (HTGR) fuel is based on a fuel stick made of coated fuel particles bound in a graphite matrix (a bonded bed). This is typified by the Fort Saint Vrain Reactor, where the bonded beds are inserted in holes in a graphite block.

Pyrolytic carbon coated fuel particles (primarily uranium oxide or uranium carbide in combination with thorium oxide or carbide) have been developed that will retain fission gases sufficiently well to meet the present reactor design criteria. The addition of a silicon carbide coating between layers of pyrolytic carbon provides an effective barrier to the metallic fission products, strontium, barium, and cesium.[1,2]

We are reporting here on two bonded-bed experiments that we conducted in-reactor under rather extreme conditions. The first experiment was conducted to determine whether the coated particles were damaged or weakened during the bonding process and to study the radiation stability of a bonded bed at a constant flux and temperature. The second experiment was conducted to determine the general physical integrity of a bonded bed at temperatures up to 1700°C, and to study the coated particle failure rate at temperature intervals from 1200 to 1700°C.

TEST SPECIMENS

The coated particles selected for the bonded-bed, in-reactor tests had performed well as loose coated particles irradiated to 20 atom percent burnup at 1200 to 1400°C. The results of the irradiation tests on these coated particles were reported in previous publications.[3,4,5,6]

The bonded beds of coated particles (fuel sticks) were prepared by injecting a phenolic resin binder into a blended bed of coated particles held in a mold. Graphite powder was mixed with the resin before injection to increase the residual carbon in the matrix, and maleic anhydride was added to the mixture to catalyze the hardening (curing) of the bonded bed. The cured bonded beds were then removed from the mold and carbonized by heating slowly to 1000°C in a stream of inert gas.

A sectioned view of the two types of coated particles irradiated in the bonded-bed assemblies is shown in Fig. 1. The physical properties of the coated particles in each experiment are given in Table I. A typical bonded bed as prepared for irradiation is shown in Fig. 2.

Some of the particle coatings may be damaged during the preparation of the bonded bed. Figure 3a shows a typical severely damaged coating. This coating cracked during the bed curing process. To prevent such damage, a sacrificial layer of anisotropic pyrolytic carbon, only about 5 μ thick, was added to the outer coating. This thin sacrificial layer, being bonded to the graphite matrix material, was cracked and separated from the main isotropic coating during curing of the bonded bed as shown in Fig. 3b.

EXPERIMENTAL METHOD

The experimental method has been described in detail in previous publications.[3,7] The only difference was in the specimens, which in the present tests were bonded beds encased in a graphite holder. In previous tests the loose coated particles were held in a very similar graphite holder. The loaded graphite holder was weld-sealed in an air-cooled, stainless steel capsule and the capsule was inserted in a lattice position in the Oak Ridge Research Reactor (ORR). The temperature of the bonded bed was monitored by tungsten 5% rhenium-tungsten 26% rhenium thermocouples located in the graphite holder. The thermocouple

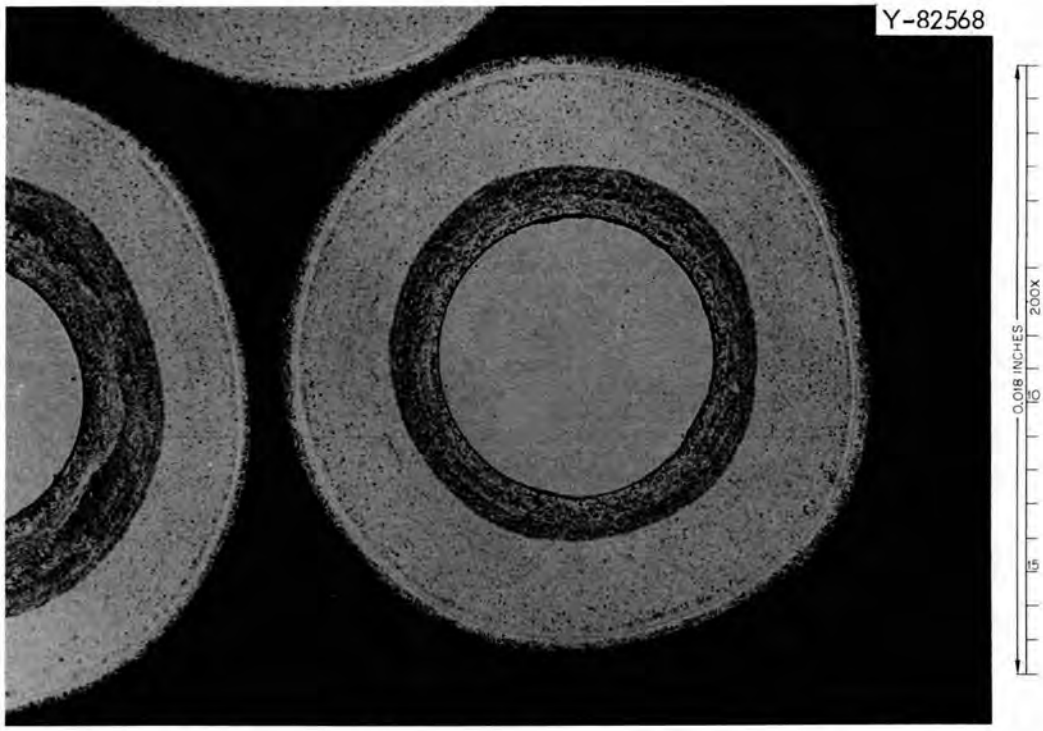


Fig. 1a. Sectioned View of Pyrolytic Carbon Coated Uranium Oxide Particles Not Encased in a Bonded Bed.

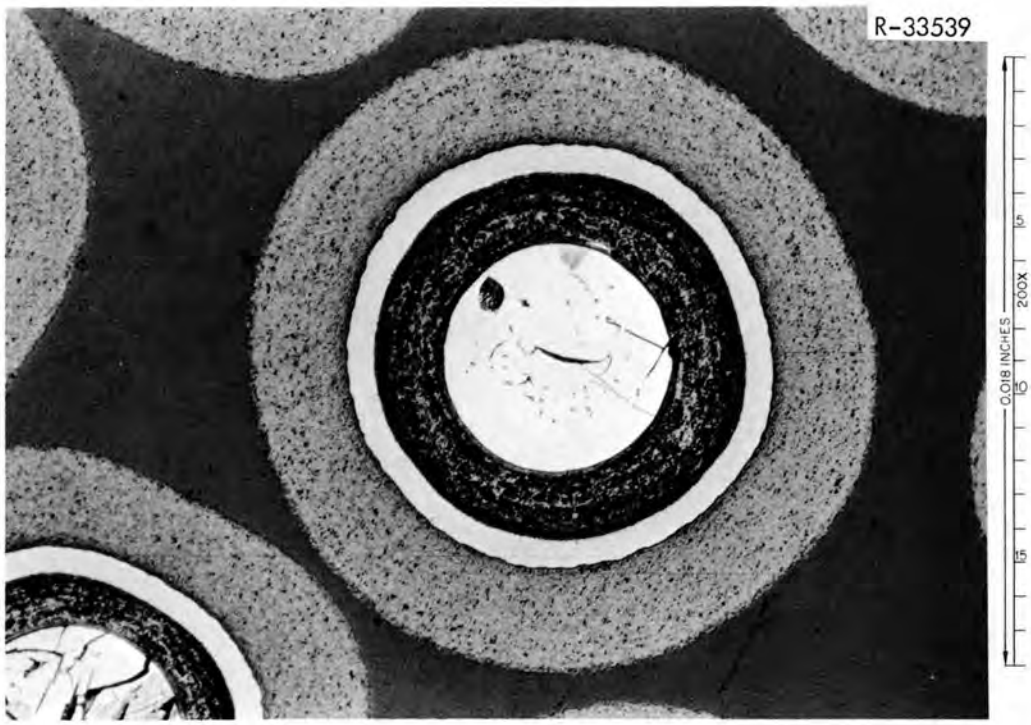


Fig. 1b. Sectioned View of Pyrolytic Carbon and Silicon Carbide Particles Not Encased in a Bonded Bed.

Table I. Data

	Experiment A9-15	Experiment B9-40
Fuel	UO_2	UC_2
Batch No.	OR-1010	OR-793
Fuel Core	212 μ	235 μ
Coating		
Porous Carbon Buffer	43 μ	43 μ
SiC Barrier	None	22 μ
Isotropic PyC	80 μ	69 μ
Anisotropic PyC (Sacrificial)	<5 μ	<5 μ
Overall Diameter	483 μ	480 μ
Number of Coated Particles in Bed	3450	5930
Coating Gas		
Buffer	C_2H_2	C_2H_2
Isotropic	C_3H_6	C_3H_6
SiC	--	CH_3SiCl_3
Deposition Rate (μm/min)		
Buffer	25	25
Isotropic	10	8
SiC	--	2.6
Coating Temperature (°C)		
Buffer	1050	1050
Isotropic	1250	1250
SiC	--	1600

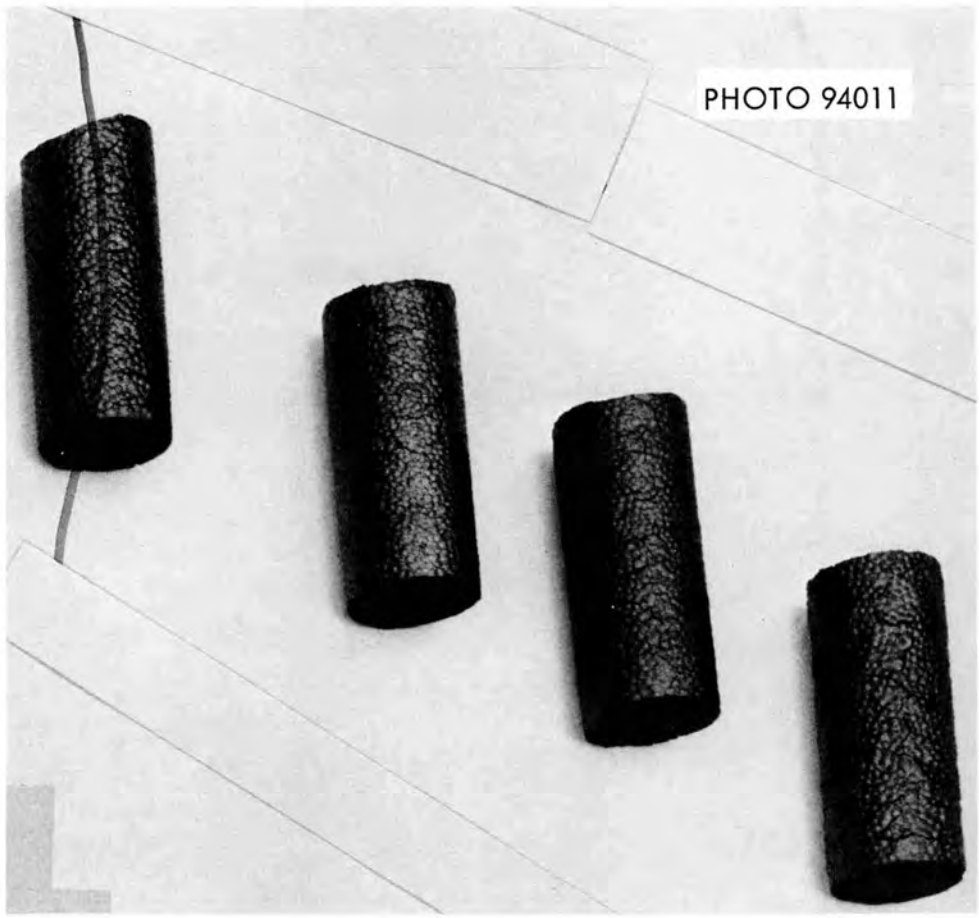


Fig. 2. Typical Bonded Bed Prepared for Irradiation Testing.

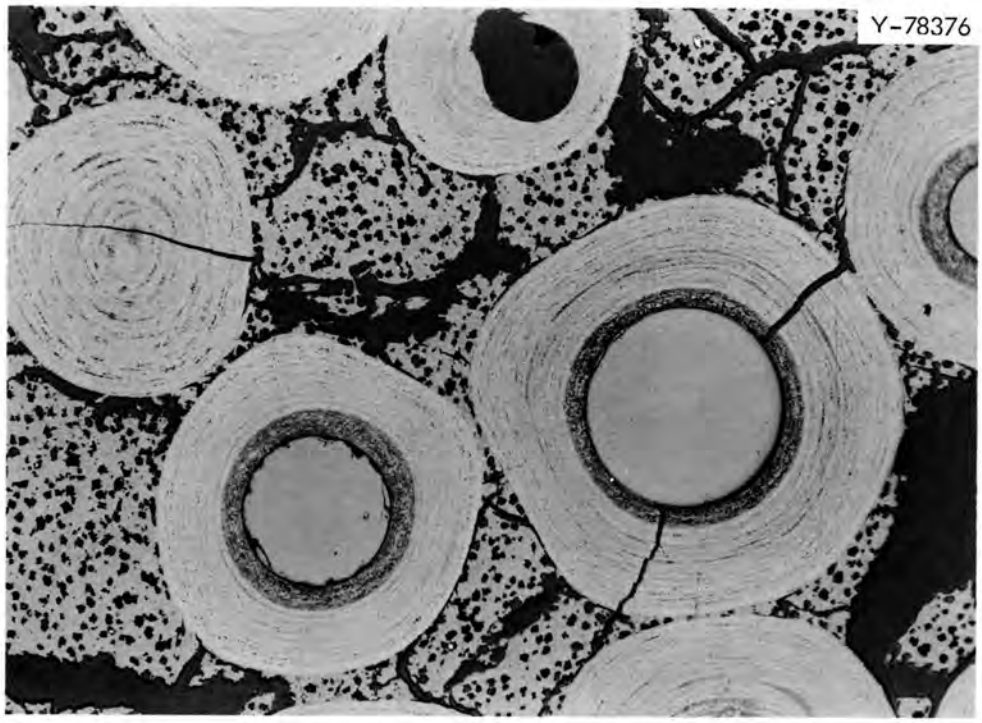


Fig. 3a. Early Development Work Produced Cracks in the Bonded-Bed Matrix that Propogated Through the Pyrolytic Carbon Coating.

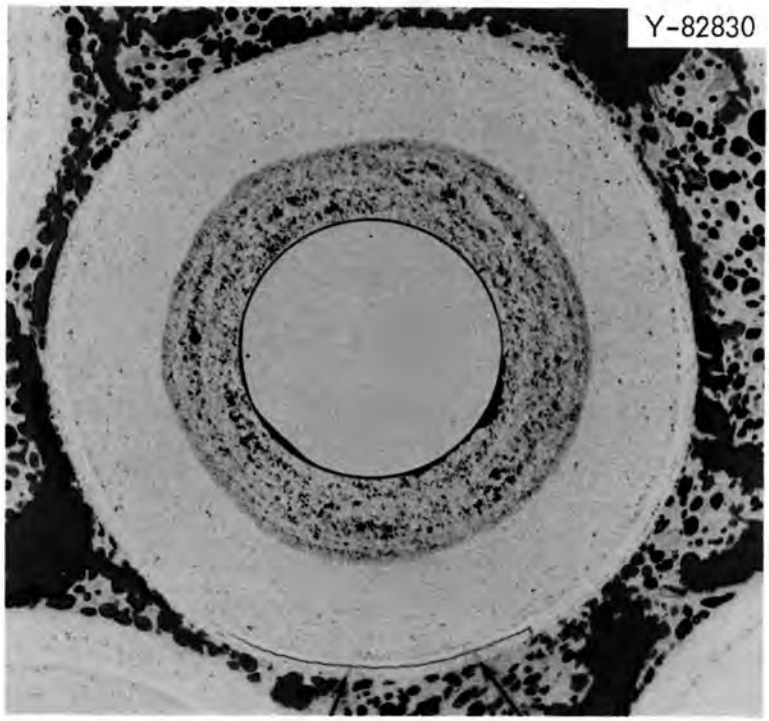


Fig. 3b. Thin Sacrificial Layer of Anisotropic Carbon Separated from the Isotropic Pyrolytic Coating (at arrows) During the Bonding Process.

located in the center of the bonded bed gave the maximum bonded-bed temperature. This center thermocouple was encased in a thin rhenium foil to avoid reaction with the graphite. The graphite holder was thermally insulated from the steel capsule wall by a blanket of metal oxide wool (≈ 51% Al_2O_3, 45% SiO_2, and 4% ZnO_2). A low flow (0.5 cc/sec) of high-purity helium was passed over the bonded bed during irradiation to sweep fission gases released by the bonded bed to a sampling station outside the reactor, where gas samples were analyzed with a gamma-ray spectrometer. The fission-gas release data, given in Table II, is expressed as a fractional release, R/B, which is the ratio of the release rate to the birth rate. The bonded-bed temperature was controlled by air flow over the capsule, and the neutron flux was controlled by adjusting the capsule position in the reactor lattice.

Table II. Irradiation Test Data

	Bonded Bed		
	Uranium Oxide Fueled		Uranium Carbide Fueled
Burnup, at. % Heavy Metal	35.5		18.0
Irradiation Temperature, °C	1250		1350-1700
Fractional Fission-Gas Release, R/B	Average		Maximum
	First Week	Last Week	at 1700°C
^{85m}Kr	2.7×10^{-8}	5.3×10^{-8}	2.3×10^{-1}
^{88}Kr	3.2×10^{-8}	4.0×10^{-8}	1.8×10^{-1}
^{87}Kr	1.2×10^{-8}	2.7×10^{-8}	1.5×10^{-1}
^{133}Xe	na	8.8×10^{-8}	3.8×10^{-1}
^{135}Xe	na	1.9×10^{-8}	1.8×10^{-1}

na = not available

IRRADIATION TESTS

Uranium-Oxide-Fueled Bonded-Bed Experiment A9-15

Experiment A9-15 contained a bonded bed of pyrolytic carbon coated uranium oxide particles from batch OR-1010. The physical data for the coated particles in this bonded bed are given in Table I, and the

irradiation test data including fission-gas release are given in Table II.

This bonded-bed experiment was irradiated for a little more than 4800 hours at 1250°C and to 35.5 at. % burnup. The fission-gas release during the first week of the test was very low, the fractional release being in the 2×10^{-8} range, which indicated that no particle coatings were damaged during the bonding process.

Coating failures during irradiation of loose coated particles have been noted by the release of bursts of fission gas. There were no bursts of fission gas released from this bonded-bed experiment. The fission-gas release increased very little during the 6-1/2-month irradiation period. Our fission-gas release data, as given in Table II, show two of the krypton isotopes doubled during the test, but it should be pointed out that the accuracy of our measurements in the 10^{-8} range are no better than 50%.

Uranium-Carbide-Fueled Bonded-Bed Experiment B9-40

Experiment B9-40 contained a bonded bed of pyrolytic carbon, silicon carbide coated uranium carbide particles from batch OR-793. The physical data and irradiation test data are given in Tables I and II respectively.

This experiment was conducted primarily to determine the rate of failure of particle coatings in a typical HTGR-design bonded bed as a function of temperature by measuring the fission-gas release rates as the temperature was increased stepwise. A history of the bonded-bed temperature and the fission-gas release is given in Fig. 4. This bonded bed was first irradiated for 2526 hours at 1350°C, accumulating 13.3 atom % burnup. The temperature was then increased in 50 to 100° steps. After being held at each step for 24 hours, the temperature was decreased and held at 1350°C again for 48 hours to determine what damage had occurred at the higher temperature, as indicated by fission-gas release. This procedure was followed up to 1700°C as shown in the upper part of Fig. 4. The fission-gas release did not reach equilibrium at any elevated temperature above 1450°C. At 1700°C we decided to hold constant temperature longer than 24 hours in order to attain an

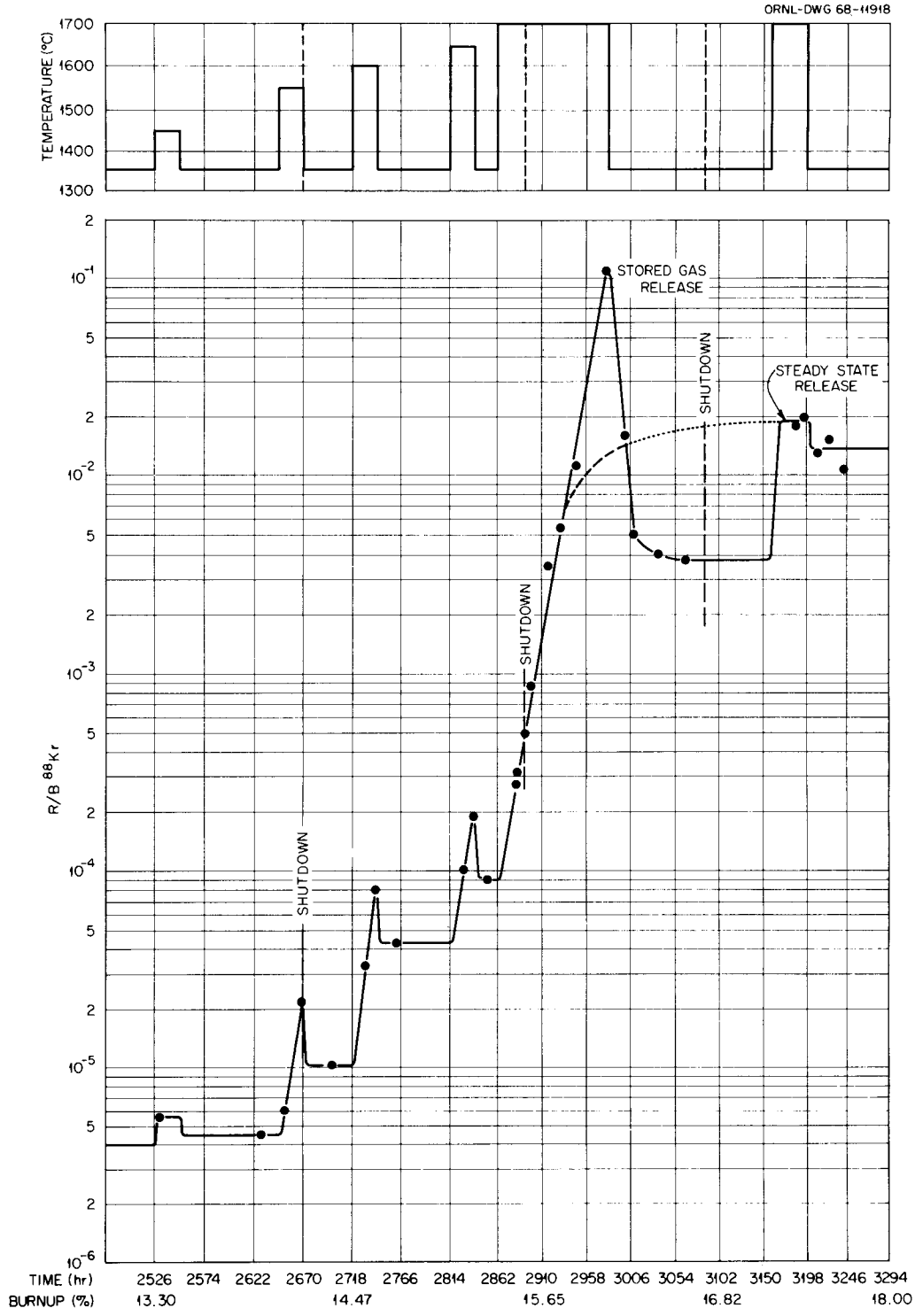


Fig. 4. Fission-Gas Release During Irradiation of a Bonded Bed Containing Pyrolytic Carbon and Silicon Carbide Coated Uranium Carbide Particles, Experiment B9-40.

equilibrium gas release rate. However, on reaching 1700°C the fission-gas release rates increased to a much higher level than we expected. The fission gas released at 1700° contained what appeared to be stored gas, rich in long-lived ^{133}Xe. The dots in Fig. 4 represent fractional release of ^{88}Kr that was collected and analyzed in sample bottles and the connecting lines represent the overall fission-gas release activity that was continuously recorded and normalized to the ^{88}Kr release rates.

POSTIRRADIATION EXAMINATION

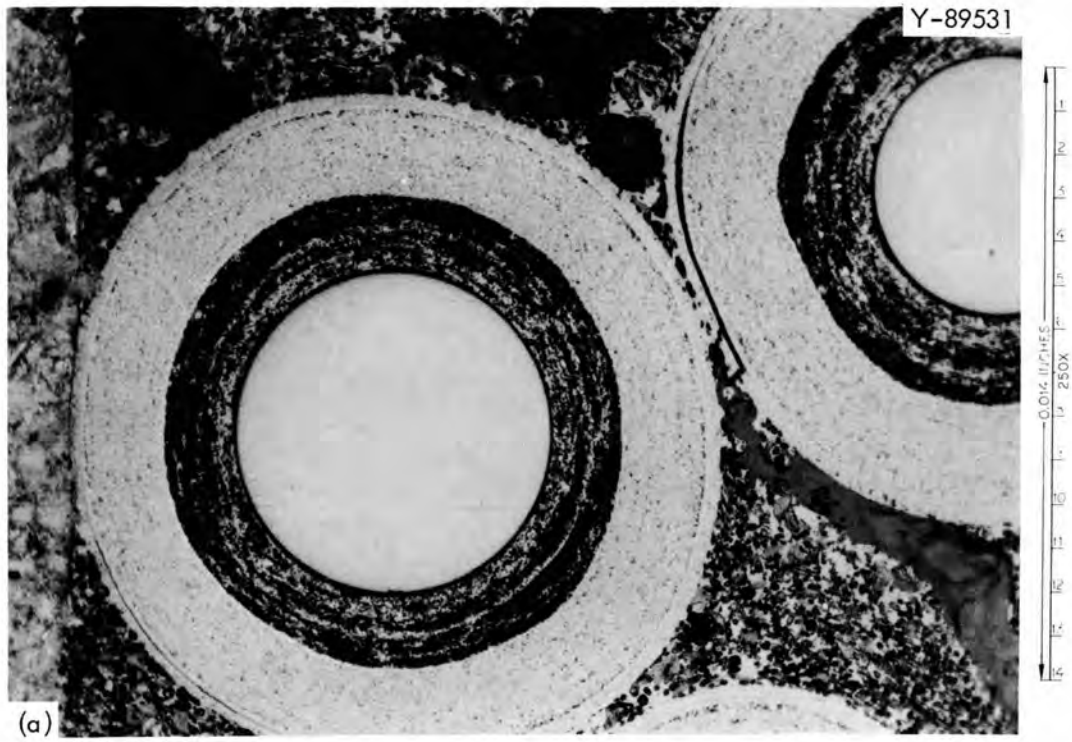
Uranium-Oxide-Fueled Bonded-Bed Experiment A9-15

Postirradiation examination showed this bonded bed to be intact. There were many discontinuous cracks, particularly near the ends, but these cracks were also noted in the unirradiated bed, shown in Fig. 2. In general, this bonded bed showed no serious damage caused by irradiation.

A transverse section was cut at about midlength and examined metallographically. Forty-one of the coated particles were exposed in this section, and there were no failures nor indications of potential failures. The only significant change observed in the uranium oxide was the appearance of noticeable fission products (primarily molybdenum and tellurium)[8] as shown in Fig. 5b. These metallic fission products had collected in the grain boundaries, and equiaxed grain growth had occurred during irradiation. Fission-gas bubbles were distributed throughout the fuel. The inner region of the porous carbon buffer coatings had densified, with about one-third of the buffer thickness being affected. No change in the isotropic coating was noted. The thin sacrificial layer had separated from the isotropic coating as planned. The unirradiated particle in a bonded bed shown in Fig. 5a is included for comparison with the irradiated particle in a bonded bed shown in Fig. 5b. Separation of the thin sacrificial layer from the isotropic layer is evident in both irradiated and unirradiated bonded beds.

Uranium-Carbide-Fueled Bonded-Bed Experiment B9-40

Postirradiation examination revealed the bonded-bed assembly to be intact, as shown in Fig. 6. The several cracks, including one continuous



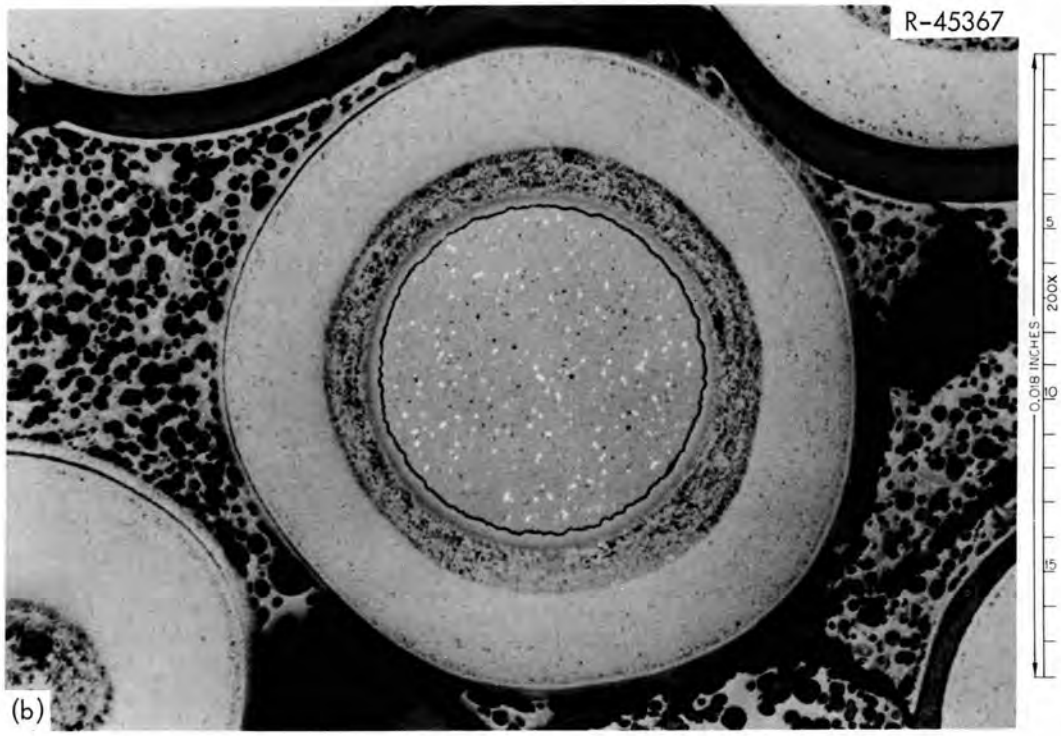


Fig. 5. (a) Unirradiated Coated Particles (b) Compared to Coated Particles After 35.5 atom % Burnup in Experiment A9-15.

Fig. 6. Bonded Bed of Pyrolytic Carbon, Silicon Carbide Coated Uranium Carbide Particles Irradiated at 1350 and 1700°C in Experiment B9-40.

crack around the middle, were present before irradiation. The loss of coated particles at both ends of the bed occurred during capsule disassembly. It is significant to note that the bed did not debond at 1700°C.

Postirradiation metallographic examination was made of a longitudinal section from the top to about midlength of the bonded bed. A cross section of typical damage is shown in Fig. 7. Damage to both the silicon carbide and pyrolytic carbon coatings appears to have been caused by reaction with the uranium carbide fuel. From the orientation of these coated particles in the bonded bed, it is evident that the uranium carbide attacked the coating and moved toward the center of the bonded bed. However, near the center of the bonded bed, where there was a smaller thermal gradient, the fuel moved out into the coating in all directions. The bonded bed annulus was approximately 5 particles thick. In some cases, where the fuel had completely penetrated the coating, the attack continued into the coating of the adjacent coated particle.

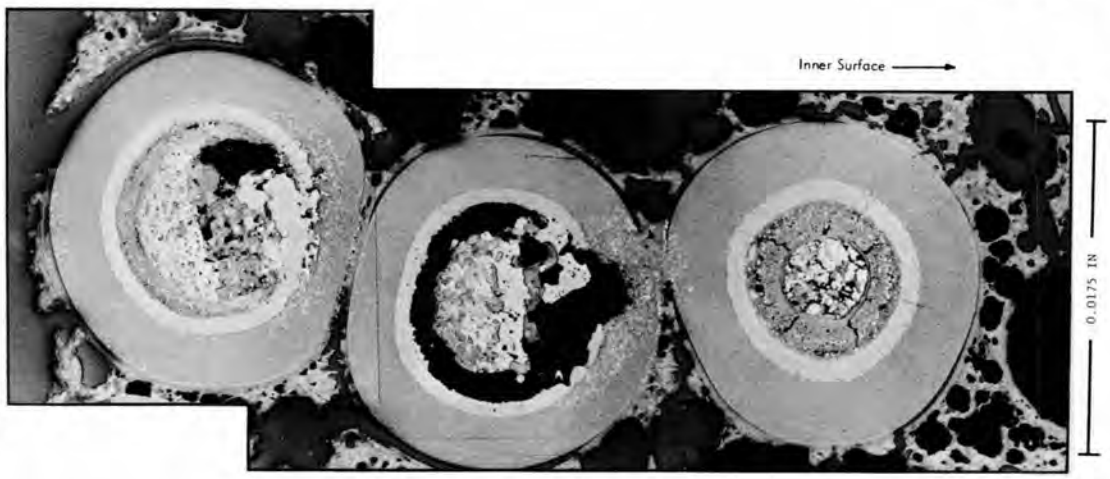
Most of the uranium carbide, graphite, and silicon carbide in a damaged coated particle could be identified optically by noting the appearance before and after etching (see Fig. 8). Three coated particles, typical of mildly damaged to severely damaged coatings, are compared in Fig. 9 with an unirradiated coated particle (upper left). The coated particle in the upper right shows fission-gas bubbles within the uranium carbide.

DISCUSSION AND CONCLUSIONS

At 1250°C, which is a little higher than present operating temperatures for this kind of fuel, the bonded bed performed very well to 35.5% burnup, and it appears that this bonded bed could have operated to a considerably higher burnup.

At temperatures above about 1500°C the fuel fails by the "Amoeba" effect. For the "Amoeba" effect to occur it is necessary to also have a temperature gradient in addition to the high temperature. It was only possible to make a very rough estimate of the temperature gradient in this experiment. The maximum gradient was estimated to be 15°C at the

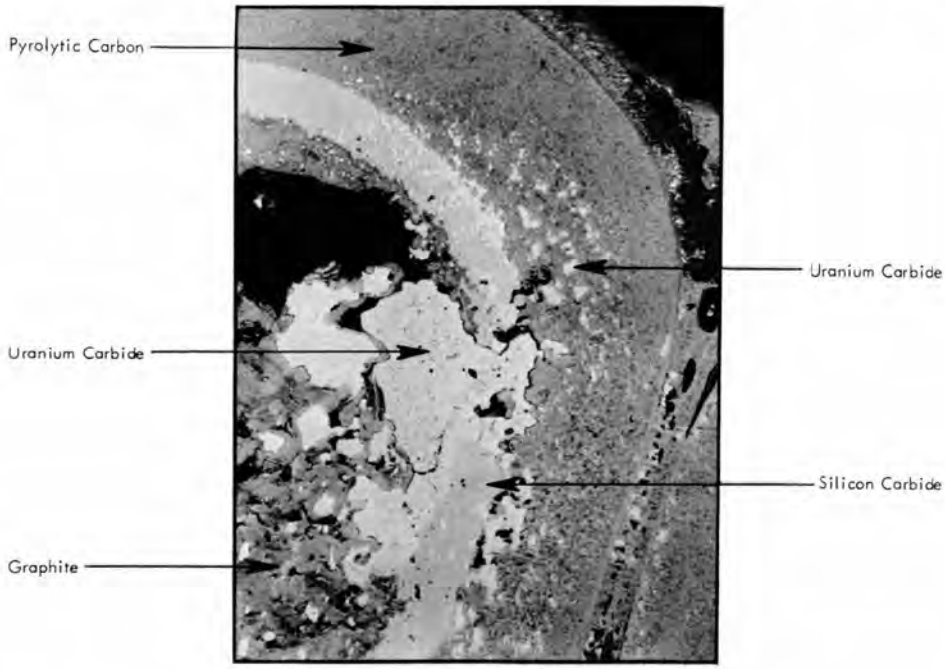
R-47163A



B9-40, BONDED BED, OR-793, UC_2, 3294 HR, 1350-1700°C

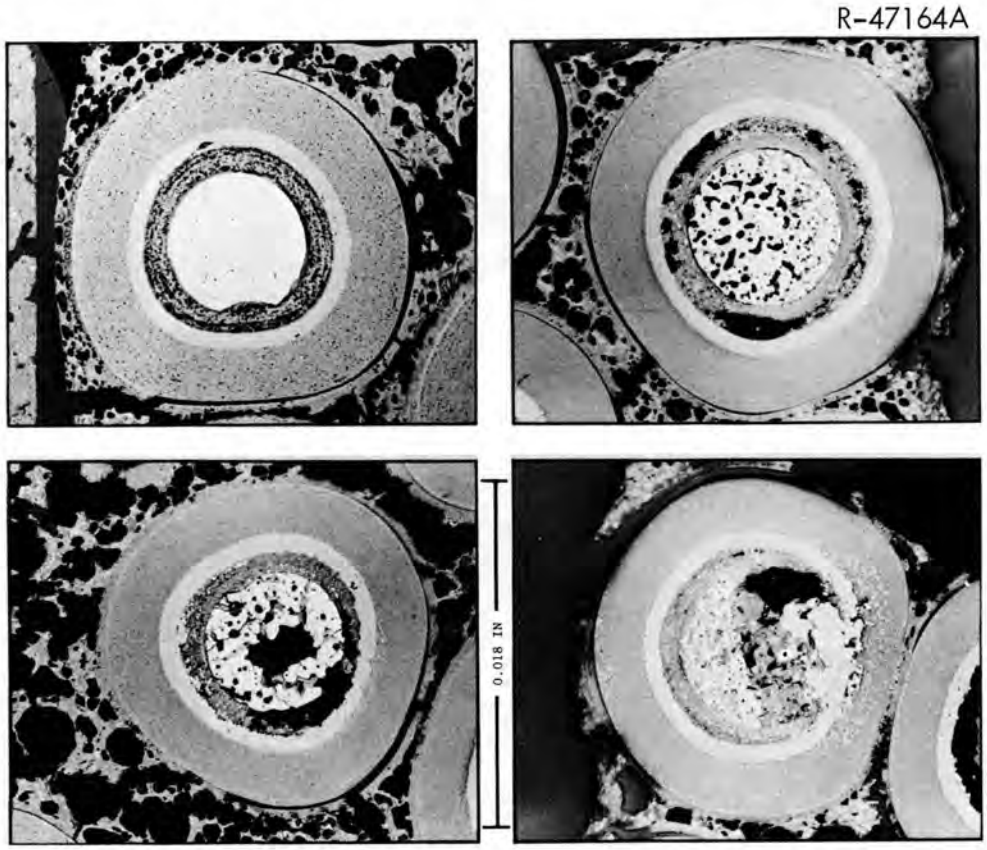
Fig. 7. Cross Section Showing Damaged Coated Particles in Bonded Bed Irradiated in Experiment B9-40.

R-47162A



B9-40, BONDED BED, OR-793, UC_2

Fig. 8. Higher Magnification of Outermost Coated Particle Shown in Fig. 7.



B9-40, BONDED BED, OR-793, UC_2, 3294 HR, 1350-1700°C

Fig. 9. Coated Particles from Bonded Bed Irradiated in Experiment B9-40 Compared with Unirradiated Coated Particle (Upper Left).

highest temperature. As observed in Fig. 7, the migration of the fuel was very pronounced, even entering adjacent fuel coatings in some instances. Migration of the fuel, as has been previously observed many times, is toward the hot core of the fuel element. The very innermost fuel particles (see Fig. 7) did not behave as badly as the fuel particles farther away, however, and this is because the temperature gradient is very small here even though the temperature is high. The fuel migration in this innermost coated particle seems to be fairly uniform in all directions.

The fuel migration in the high-temperature experiment was very drastic and caused excessive damage. It must be remembered, however, that these are accident conditions which would not be allowed to persist for a very long time in an operating reactor. The effects observed in this experiment were produced over many hours of operation.

ACKNOWLEDGMENTS

The authors express their appreciation to T. W. Fulton for gas analyses, to H. E. Robertson for postirradiation examination, to L. G. Shrader for metallographic specimen preparation, to R. J. Robson for engineering in-reactor facilities, and to O. Sisman for consultation throughout the tests.

REFERENCES

1. J. R. Gough and D. Kern, "Studies of Coated Fuel Particles for Dragon Reactor Experiment," Journal of Nuclear Energy, 21(8): 623-692 (1967).
2. E. H. Voice, "Silicon Carbide as a Fission-Product Barrier in Nuclear Fuels," D.P. Report 616 (Oct. 1965).
3. P. E. Reagan, F. L. Carlsen, and R. M. Carroll, "Fission-Gas Release from Pyrolytic Carbon Coated Fuel Particles During Irradiation," Nucl. Sci. Eng., 18(3): 301 (1964).
4. P. E. Reagan, J. G. Morgan, and O. Sisman, "Fission-Gas Release from Pyrolytic Carbon Coated Uranium Oxide Particles During Irradiation at 2000 to 2500°F," Nucl. Sci. Eng., 23(3): 213 (1965).
5. P. E. Reagan, R. L. Beatty, E. L. Long, Jr., "Performance of Pyrolytic Carbon Coated Uranium Oxide Particles During Irradiation at High Temperatures," Nucl. Sci. Eng., 28(1): 34 (1967).
6. P. E. Reagan, E. L. Long, Jr., J. G. Morgan, J. H. Coobs, "Irradiation Performance of Pyrolytic Carbon and Silicon Carbide Fuel Particles," Nucl. Appl. Tech., 8(5): 417-431 (May 1970).
7. R. M. Carroll and P. E. Reagan, "Techniques for In-Pile Measurements of Fission-Gas Release," Nucl. Sci. Eng., 21(2): 141-146 (1965).
8. B. D. Bradbury, J. T. Demont, and P. M. Martin, "Solid Fission Products in Irradiated Uranium Dioxide," British Report AERE-R-5149 (1966).

ORNL-TM- 3361

DISTRIBUTION

1. J. W. Anderson
2. W. D. Bond
3. J. M. Chandler
4. J. A. Conlin
5. J. H. Coobs
6. C. M. Cox
7. H. J. deNordwall
8. S. J. Ditto
9. W. P. Eatherly
10. D. E. Ferguson
11. W. R. Grimes
12. P. A. Haas
13. S. I. Kaplan
14. P. R. Kasten
15. R. E. Leuze
16. E. L. Long, Jr.
17. A. L. Lotts
18. R. S. Lowrie
19. J. G. Morgan
20. E. A. Nephew
21. P. Patriarca
22. A. M. Perry
23. P. E. Reagan
24. J. L. Scott
25. J. D. Sease
26. M. D. Silverman
27. O. Sisman
28. M. J. Skinner
29. J. W. Snider
30. V. C. A. Vaughen
31. T. N. Washburn
32. G. M. Watson
33. G. D. Whitman
34. R. G. Wymer
35. Gale Young

36-37. Central Research Library
38. Document Reference Section
39-41. Laboratory Records Department
42. Laboratory Records, ORNL R.C.
43. ORNL Patent Office
44-45. Division of Technical Information Extension
46. Laboratory and University Division, ORO