LA-6785-MS

Informal Report

UC-77

Issued: April 1977

# Irradiation Test HT-31: High-Temperature Irradiation Behavior of LASL-Made Extruded Fuel Rods and LASL-Made Coated Particles


P. Wagner

R. D. Reiswig

C. M. Hollabaugh

R. W. White

K. V. Davidson

D. H. Schell



los alamos scientific laboratory

of the University of California

LOS ALAMOS, NEW MEXICO 87545

An Affirmative Action/Equal Opportunity Employer

UNITED STATES

ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

CONTRACT W-7405-ENG. 36

This work was supported by the Gas Cooled Reactor Project of the US Energy Research and Development Administration, Division of Nuclear Research and Applications.

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161
Price: Printed Copy $3.50 Microfiche $3.00

IRRADIATION TEST HT-31: HIGH-TEMPERATURE IRRADIATION

BEHAVIOR OF LASL-MADE EXTRUDED FUEL RODS AND

LASL-MADE COATED PARTICLES

by

P. Wagner, R. D. Reiswig, C. M. Hollabaugh,

R. W. White, K. V. Davidson, and D. H. Schell



ABSTRACT

Three LASL-made extruded graphite and coated particle fuel rods have been irradiated in the Oak Ridge National Laboratory High Fluence Isotope Reactor test HT-31. Test conditions were about 9 x $10^{21}$ nvt(E >.18 MeV) at 1250°C. The graphite matrix showed little or no effect of the irradiation. LASL-made ZrC containing coated particles with ZrC coats and ZrC-doped pyrolytic carbon coats showed no observable effects of the irradiation.

## I. INTRODUCTION

Extruded fuel rods and coated particles made with ZrC and ZrC-doped pyrolytic carbon coats have been tested in the ORNL High Fluence Isotope Reactor[1] experiment HT-31. This is a summary of the fabrication history and irradiation test results on these materials.

The High Temperature Fuels Technology for Nuclear Process Heat Program at LASL had as its goals the identification, fabrication, and proof testing of nuclear fuels with higher temperature capabilities than those now being used in gas cooled reactors. This higher temperature capability could be utilized for higher processing temperatures of the graphite-base fuels and for higher reactor fuel operating temperatures. Details of the rationale for this program and descriptions of the research, development, and technology developed during the program have been described in a series of publications.[2-6]

The objectives of this experiment were:

(a) To test the irradiation behavior of fuel rods extruded at LASL. These fuel rods contained fertile, fissile, and inert particles. The graphite matrices were made using a fully graphitized flour, carbon black (Thermax), and a polyfurfural alcohol binder (Varcum). This formulation was tested in HT-29[6] with excellent results.

(b) To test the behavior of LASL-prepared ZrC-coated particles contained in the extruded graphite matrix. These particles had inert kernels; however, they were made with dimensions and coat designs of types to be used in fuel development. These test results will help to identify critical problem areas such as particle behavior in the restrictive graphite environment during fabrication and irradiation, and particle-matrix interaction (i.e. coat integrity after irradiation).

## II. MATERIAL DESCRIPTION

Three LASL-made extruded fuel rods were tested in the ORNL (HFIR) HT-31 irradiation experiment. These were identified as:

1. SNM 6517-01
2. SNM 6517-02
3. SNM 6517-03

All three fuel rods had the same coated particle composition; this was:

Fissile: $UC_2$ kernels with 6.36% enrichment. These particles were supplied by ORNL and were identified as lot SNM 6517 Or-2257H R-52C. These were TRISO coated (SiC) and had an overall mean diameter of 748 μm (Figure 1).

Fertile: $ThO_2$ BISO particles. These particles were purchased from the General Atomic Co. and were identified as Lot 6291 Composite No. 3 (Figure 2).

Inert particles to make up the particle fraction to 41 volume percent: These were LASL-made ZrC TRISO-type particles with ZrC-doped oLTI and were identified as lots 76-53HT and 76-54HT (Figure 3). 76-53HT had an inner LTI, 76-54HT did not.

The inert particles were made with carbon kernels. The ZrC layer was deposited using $CH_4$ and $ZrCl_4$[7] at a maximum temperature, in the coater, of 1720 K. The ZrC-doped LTI was deposited from a 6.4:1 propylene[8] to zirconium tetrachloride gas mixture at a maximum temperature of 1790 K. Weight per cent ZrC in the oLTI was estimated to be 17%. Crushing strengths on these particles were in the 2-4 kg range.

Fuel rod SNM 6517-01 was made of the materials that have been used in much of the extrusion development work. This had M3 graphite as the matrix flour and 85 parts of flour to 15 parts of Thermax as the matrix carbon filler. The extrusion was bonded with catalyzed Varcum 8251. The M3 graphite is an equiaxed reactor grade flour having a maximum particle size of 60 mesh (250 μm).

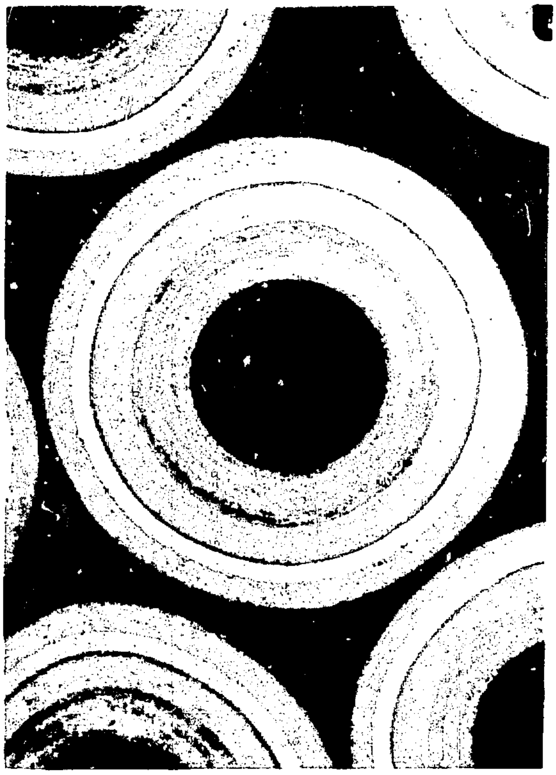


Figure 1. As-prepared fissile particle, SNM 6517 Or-2257H R-52 C (100 X).

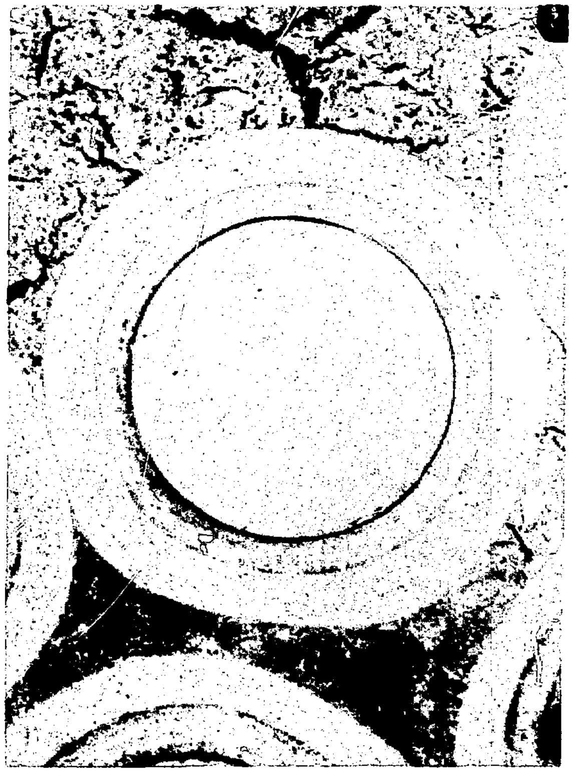


Figure 2. As-prepared fertile particle, Lot 6291 Composite No. 3 (100 X).

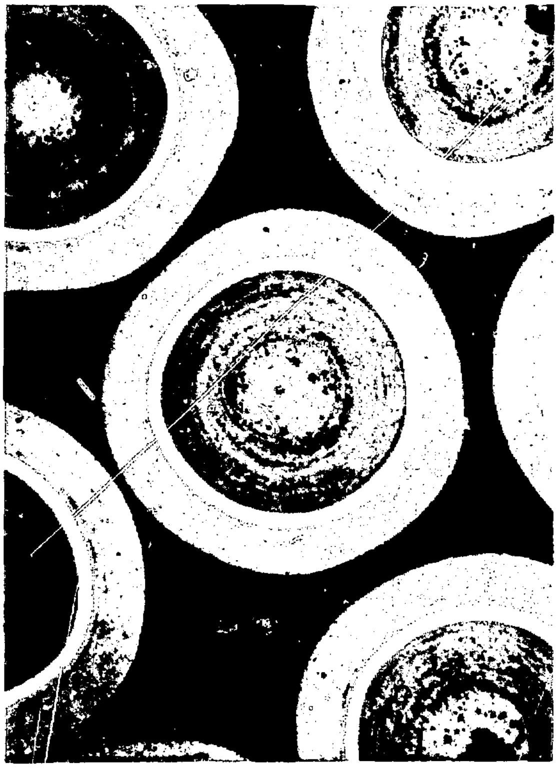


Figure 3. As-prepared inert particle, LASL 76-54HT (100 X).

The particle size distribution is such that 55% of the flour will pass through a 200 mesh screen and 33% will pass through a 325 mesh screen. Thermax is a carbon black having spherical particles less than a micrometer in diameter. Varcum 8251 is a partially polymerized furfuryl alcohol that is normally catalyzed with maleic anhydride to make it thermosetting. The normal mixing procedure was to mix thoroughly the M3, Thermax, and Varcum together. This was done by alternately passing the mixture through a meat chopper and extruding it. This mixture was hand mixed with the coated particles and inert shim particles. The mixture was then extruded through the appropriate die two times with the extrusions being crumbled and hand mixed between extrusions. The third extrusion was kept and processed. This was cured in air to 523 K using a 63-hour heating cycle. It was then transferred to a vacuum oven where it was heated to 1113 K over a period of 48 hours. Final heat treatment to 2073 K was in a graphite resistance furnace in an argon atmosphere at a pressure of about 30 cm Hg, absolute. The sample was then cut to length and leached.

Fuel rod, SNM 6715-02, was identical to the SNM 6517-01 fuel rod with respect to composition, mixing, extrusion, and processing. The only difference was that all the particles were coated with a thin (10-μm) layer of carnauba wax prior to mixing with the matrix material. This wax formed a barrier between the particles and matrix. The fugitive material left the extrusion during thermal processing leaving a gap between the particles and the matrix. The purpose was to minimize the particle-matrix interaction.

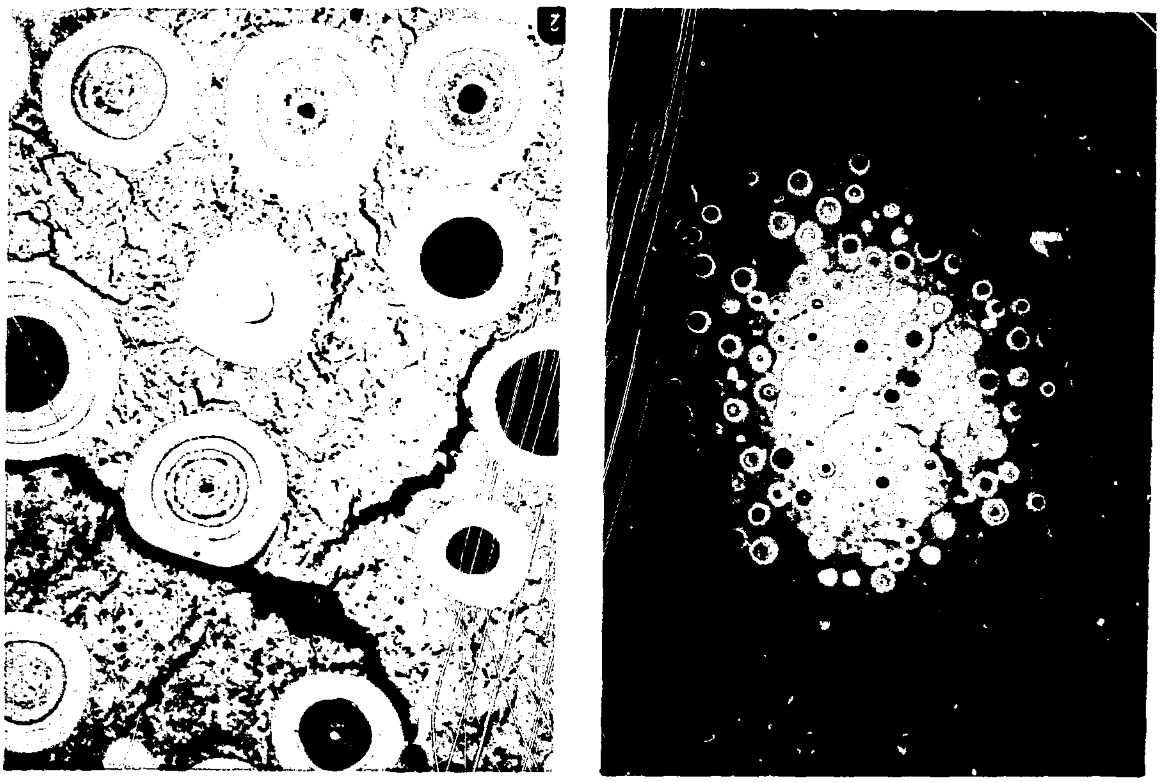
Fuel rod SNM 6517-03 differed from the other two in that the M3 flour was replaced by a Speer S0969 graphite flour. It is understood that this flour has been used by the Speer Carbon Co. to make HTGR fuel blocks for the General

Atomic Co. This experiment was designed to test the suitability of making fuel rod extrusions with a flour used for the fuel block fabrication. It would be hoped that such a fuel rod would have similar properties and behavior, under irradiation, to the fuel element. If this turns out to be the case, a loaded fuel block would have essentially a homogeneous behavior under HTGR operating conditions.

All fuel rods were heat treated to a maximum temperature of 2073 K. All fuel rods were then leached for 4 hours in a 10 $Cl_2/CO$ gas stream at 950°C to remove exposed U or Th. Graphite matrix densities averaged about 1.55 g $cm^{-3}$. Macrophotographs of these fuel rods are shown in Figures 4-6.

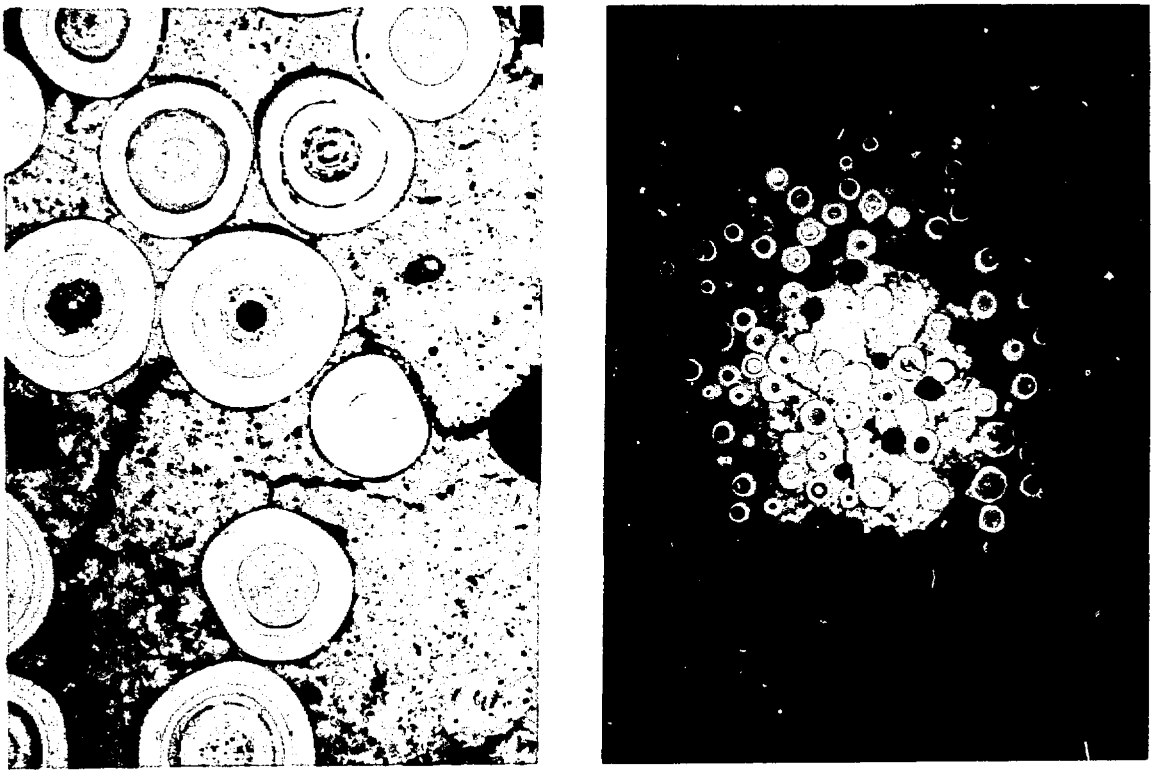
Chemical analysis of the fuel rods yielded the following:

SNM 6517-01 contained .0120g $cm^{-3}$ U & .0247g $cm^{-3}$ Th
SNM 6517-02 contained .0112g $cm^{-3}$ U & .0366g $cm^{-3}$ Th
SNM 6517-03 contained .0117g $cm^{-3}$ U & .0338g $cm^{-3}$ Th



a. (40 X) b. (~ 7X)

Figure 4. As-prepared fuel rod SNM 6517-01.



a. (40 X) b. (~ 7X)

Figure 5. As-prepared fuel rod SNM 6517-02.

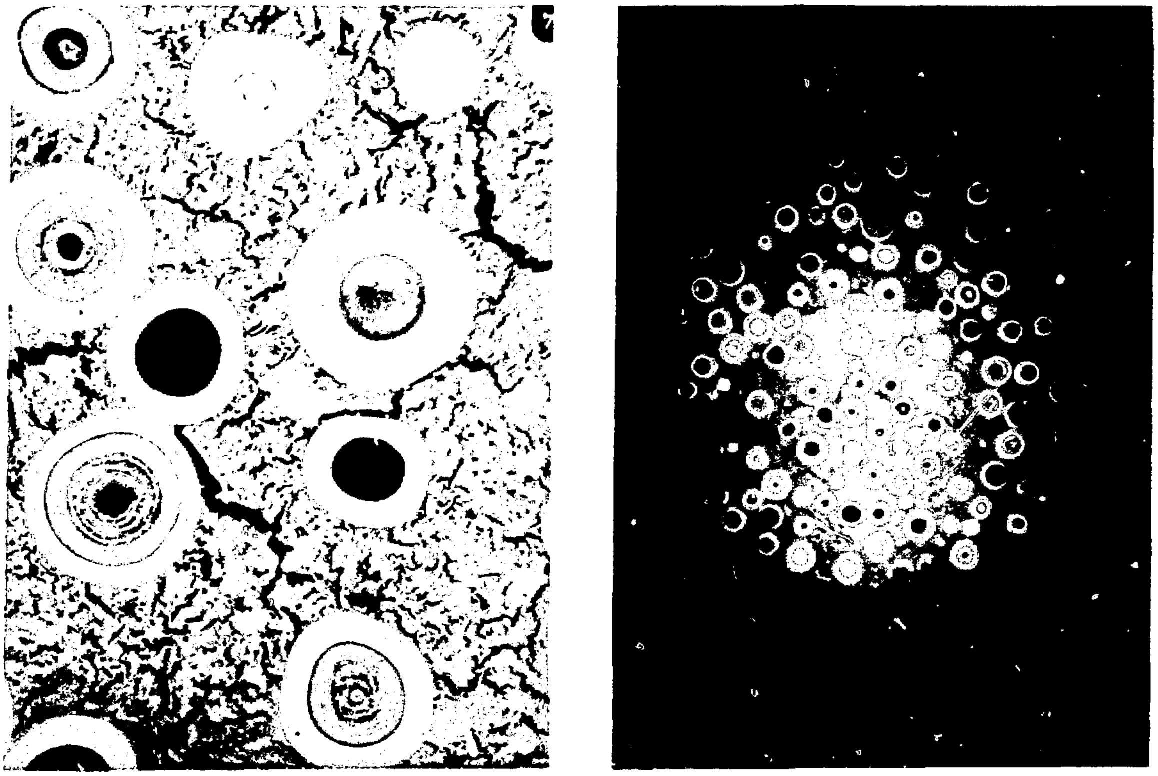
## III. IRRADIATION CONDITIONS

The HT-31 experiment was carried out in the ORNL High Fluence Isotope Reactor (HFIR)[1] in the target (HT) position. HT-31 operated for four HFIR cycles. The LASL fuel rods were located in the upper half of the capsule. Design operating temperature for the LASL fuel rods was 1250°C. The basic irradiation conditions for our materials are compiled in Table I. More detailed information on the HT-31 irradiation parameters may be obtained from the Oak Ridge National Laboratory.

## IV. RESULTS OF THE POST IRRADIATION EXAMINATION

### A. Visual Inspection of the Fuel Rods

Photographs of the extruded rods were made at ORNL after removal from the capsule and before mounting for metallographic examination. These are



a. (40 X) b. (~ 7X)

Figure 6. As-prepared fuel rod SNM 6517-03.

TABLE I

IRRADIATION TEST CONDITIONS FOR LASL FUEL RODS IN HT-31

| Fuel Rod | Flux (E>.18Mev) ($n\ s^{-1}\ cm^{-2}$) | Fluence (E>.18Mev) ($n\ cm^{-2}$) | T (Design) ($^oC$) | Burnup (% FIMA) $^{235}U$ | $^{238}U$ | $^{232}Th$ |
|---|---|---|---|---|---|---|
| SNM 6517-01 | $1.12 \times 10^{15}$ | $8.65 \times 10^{21}$ | 1250 | 84.77 | 18.00 | 8.59 |
| SNM 6517-02 | 1.15 | 8.89 | 1250 | 84.80 | 18.30 | 8.75 |
| SNM 6517-03 | 1.17 | 9.00 | 1250 | 84.83 | 18.50 | 8.88 |

compared with photographs made at LASL of unirradiated fuel rods in Figure 7. Although the lighting conditions and the magnifications were not exactly the same for the two sets of photographs, it is clear that changes in the appearance of the fuel rods as a result of the high temperature irradiation, if any, were very slight. Handling characteristics after irradiation appeared to be no different than they were before the tests. The samples were neither crumbly nor friable and they maintained their structural integrity throughout the hot cell manipulations. The chipped edges seen in Figure 7 are consequences of the many handling operations in loading and unloading the capsule and during the PIE.

B. Metallographic Preparation of the Fuel Rods

Transverse sections through the irradiated fuel rods were mounted, ground, re-impregnated with mounting resin, re-ground and polished on vibratory diamond laps. This preparation was sufficient to do the examinations of the particles and matrices reported in the following section. It has been our experience, at LASL, that metallographic mounts of ZrC coats often show cracks. By careful polishing experiments we have established that these apparent cracks are artifacts produced by mounting and/or grinding stresses. The polishing procedure used for the fuel rods removed an insufficient amount of material to eliminate these apparent cracks. The cracks that are seen in the ZrC coats are indeed artifacts.

C. Examination of the Metallographic Fuel Rod Mounts

Mosaics of the fuel rods each consisting of some fifty photomicrographs are shown in Figures 8-10. These cross-sectioned views were made at 90° to the extrusion axis. The Bacon Anisotropy Factor for the graphitized matrix component of these fuel rods was about 1.3. It has been our experience that metallographic sectioning of fuel rods, of this degree of isotropy, parallel to the extrusion axis, does not tend to yield further information. For this reason, the irradiated fuel rods were examined only in cross section. The various types of particles - fissile TRISOs, fertile BISOs, and inerts with and without iLTIs in the TRISO configuration - are largely identifiable from their appearance. Some of the particles were sectioned far enough from the equatorial plane to disguise their identity; however, these have small apparent diameters and can be recognized.

Overall there has been little or no change in the matrix. There is a suggestion that the matrix graphite is more porous after irradiation although there is no quantitative verification for this observation. Vacancy clustering upon irradiation which leads to larger pores has been observed in graphite irradiation experiments and could be the cause of this apparent increase in porosity.

There is no indication of any interaction between the matrix and the particle coats in any of the fuel rods. We think this a direct consequence of having the high density graphite matrix. The high volume fraction of filler flour used precludes having the particles completely surrounded by the poorly graphitized (i.e. binder) component of the matrix. Thus, only separated parts of the particle coats are in contact with the binder phase, and this is the component of the matrix that undergoes the maximum dimensional change with temperature and irradiation. The addition of the carnauba wax in SNM 6517-02 proved to be unnecessary; moreover, the uncoupling of the particles from the matrix could have added to the temperature drop between the fuel and the coolant causing the fuel to operate at a higher temperature.

Individual particles were examined using polarized light. We were especially interested in seeing if the combination of elevated temperature fuel rod processing and high temperature irradiation had introduced optical anisotropy in the ZrC-doped low temperature isotropic pyrolytic carbon coats of LASL's inert particles. No evidence of anisotropy was found. The coats appeared to be totally unaffected by irradiation experienced in the tests. A small amount of roughness at the inner surfaces of some of the ZrC coats on the irradiated particles has been noted. Examination of archive photomicrographs at LASL have revealed that this roughness was present in the particles in the as-prepared condition and was not a consequence of the irradiation.

SUMMARY

The behavior of the LASL fuel rod matrices and ZrC-coated particles in the HT-31 experiment was excellent. The matrices may have experienced pore-volume growth; however, this was very slight. Measurements on possible attendant fuel rod swelling because of this were not available. The particles showed no effect at all of having been extruded and heat-treated in the graphite matrix, and subsequently having been irradiated for $\sim 9 \times 10^{21}$ nvt at $1250^{o}C$. The visual evidence is presented in the photomicrographs. Since the examination of individual particles and of the graphite matrix at higher magnifications and in greater detail than has been described here, did not add to the information included in this report, the text has been confined only to the pertinent data. There were too few particles exposed by the metallography to make meaningful dimensional coat thickness measurements.

## ACKNOWLEDGMENTS

The cooperation on the part of the Oak Ridge National Laboratory staff in performing these experiments is gratefully acknowledged. The help of J. H. Coobs, F. J. Homan, and W. P. Eatherly in planning the experiments was invaluable. The cooperation of E. L. Long, Jr., R. L. Beatty, T. N. Tiegs, and the hot-cell crew who prepared the test capsule and did the PIE is especially appreciated. The excellent mosaics of the irradiated fuel rods were supplied to us by T. N. Tiegs.

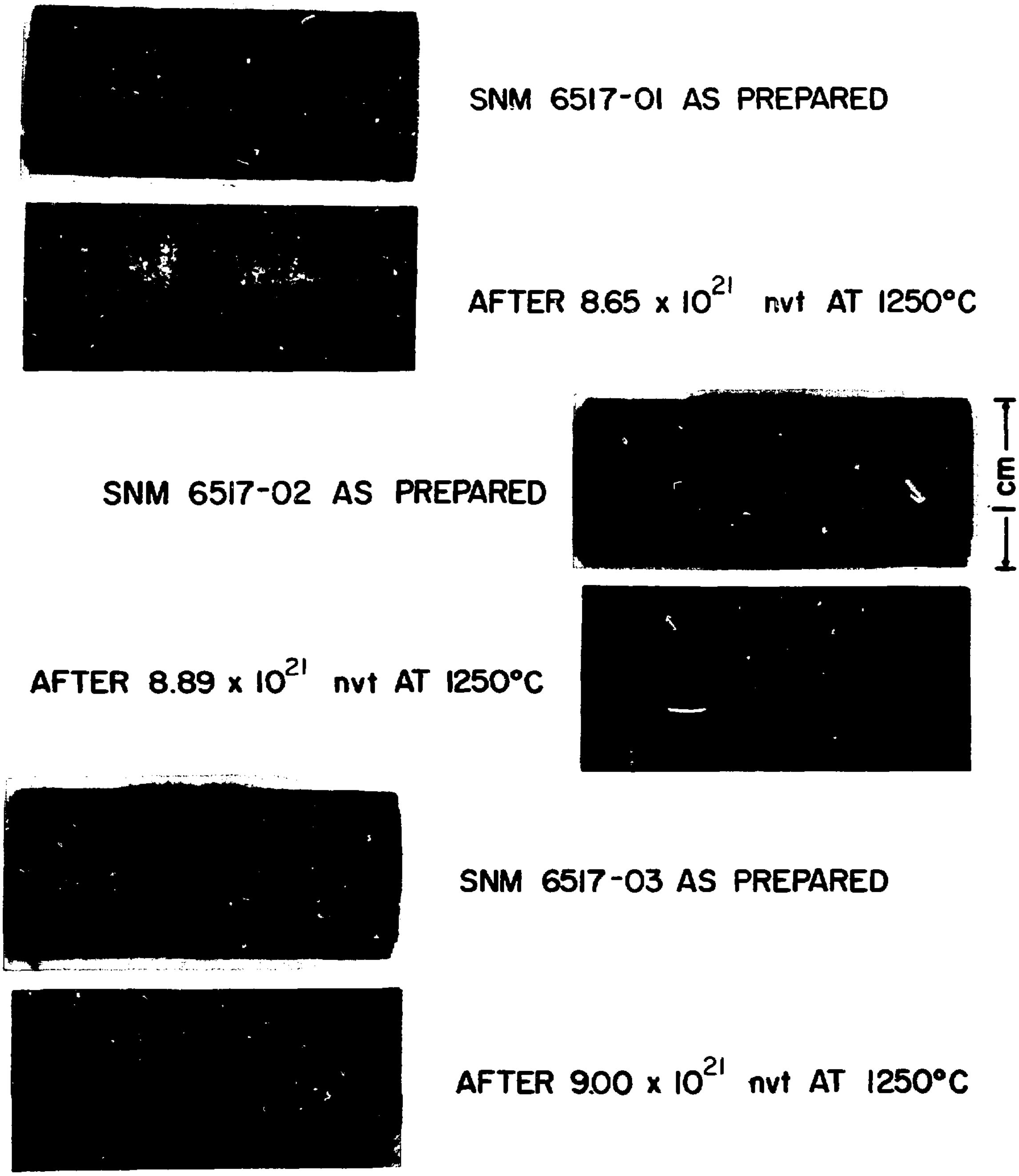



Figure 7. Effect of irradiation on external appearance of LASL extruded fuel rods. ORNL HFIR experiment HT-31.

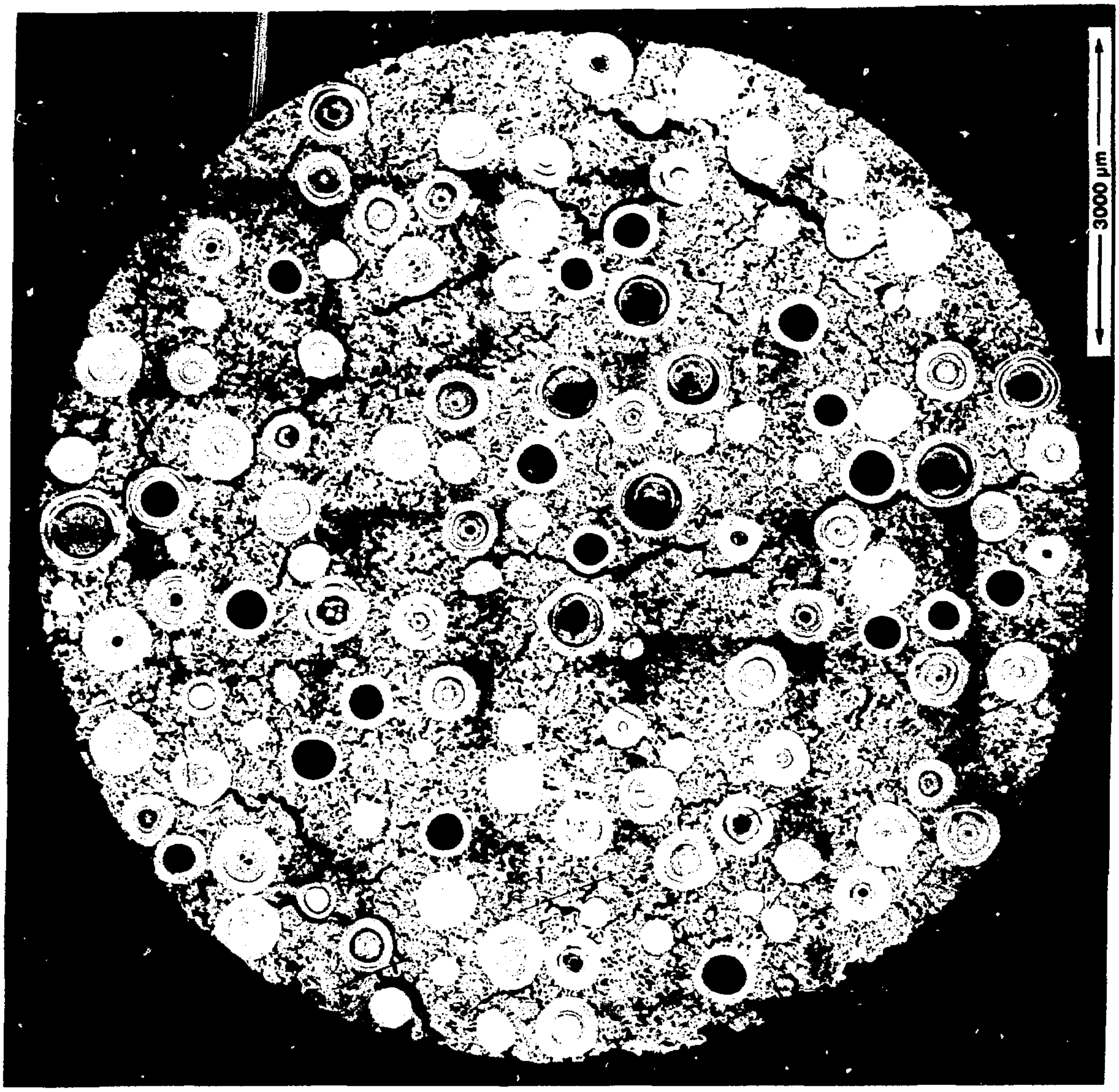



Figure 8. LASL fuel rod SMN 6517-01 after 8.65 x $10^{21}$ nvt at 1250°C (courtesy of the Oak Ridge National Laboratory.

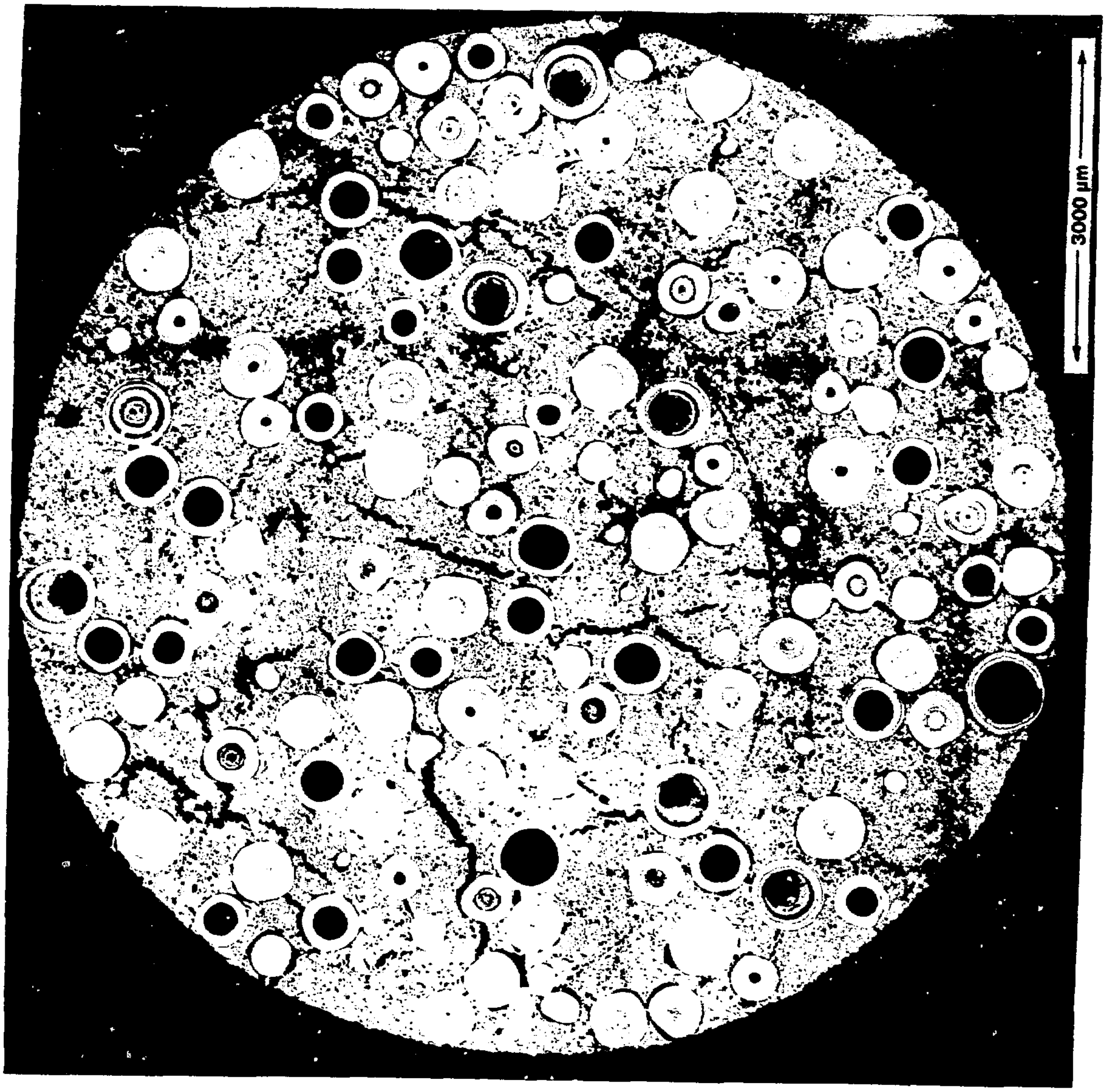



Figure 9. LASL fuel rod SMN 6517-02 after $8.89 \times 10^{21}$ nvt at 1250°C (courtesy of the Oak Ridge National Laboratory).

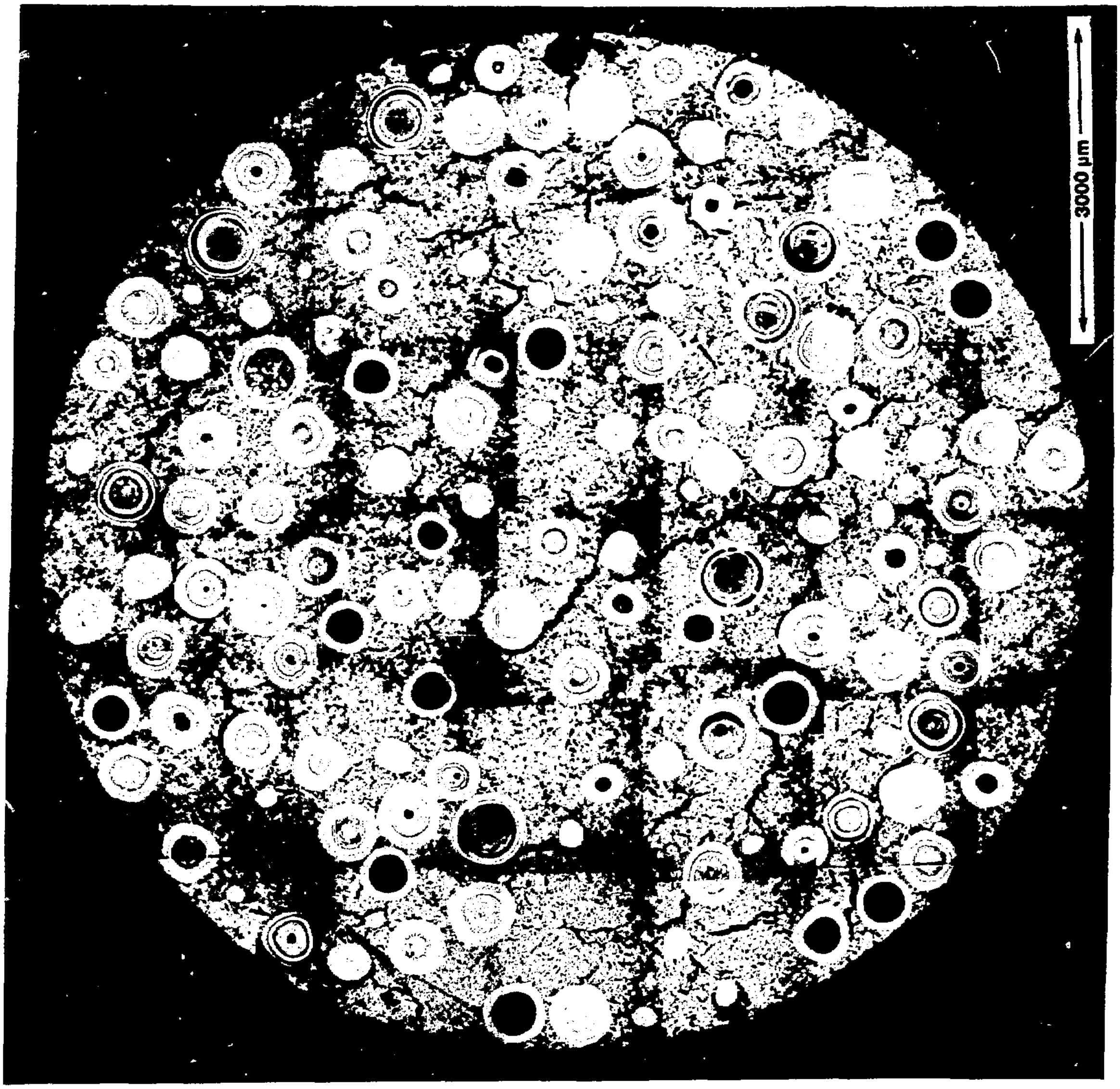



Figure 10. LASL fuel rod SNM 6517-03 after 9.00 x $10^{21}$ nvt at 1250°C (courtesy of the Oak Ridge National Laboratory.

REFERENCES


1. F. T. Binford and E. N. Cramer, Eds., "The High Flux Isotope Reactor," Vol. I and II, Oak Ridge National Laboratory report ORNL-3572 (1964).

2. J. D. Balcomb and P. Wagner, "Extending the Temperature Range of the HTR," Proc. BNES Intern. Conf. (London, Nov. 1974).

3. P. Wagner, C. M. Hollabaugh, and R. J. Bard, "ZrC, A Key Ingredient for High Temperature Nuclear Fuels" in "Gas-Cooled Reactors with Emphasis on Advanced Systems," Vol I, International Atomic Energy Agency, Juelich, FRG, 277 (1975).

4. C. Hollabaugh, R. Reiswig, P. Wagner, R. White, and L. Wahman, "A New Method for Coating Microspheres with ZrC and ZrC-C Graded Coats," J. Nucl. Mat. 57, 325 (1975) and Los Alamos Scientific Laboratory report LA-6012 (1975).

5. P. Wagner, "The Dependence of Thermal Conductivities of Binder Residues on Heat Treatment Temperature" Carbon 14, 71 (1976) and LA-6181 (1976).

6. R. D. Reiswig, P. Wagner, C. M. Hollabaugh, R. W. White, J. A. O'Rourke, K. V. Davidson, D. H. Schell, "Behavior of LASL-Made Graphite, ZrC, and ZrC-Containing Coated Particles in Irradiation Tests HT-28 and HT-29," Los Alamos Scientific Laboratory report LA-6211 (1976).

7. P. Wagner, L. A. Wahman, R. W. White, C. M. Hollabaugh, and R. D. Reiswig, "Factors Influencing the Chemical Vapor Deposition of ZrC," J. Nucl. Mat. 62, 221 (1976).

8. C. M. Hollabaugh, L. A. Wahman, R. D. Reiswig, R. W. White, and P. Wagner, "Chemical Vapor Deposition of ZrC Made by Reactions of $ZrCl_4$ with $CH_4$ and with $C_3H_6$," (accepted for publication) Nuclear Technology (1977).