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COATED-PARTICLE FUELS DEVELOPMENT AT OAK RIDGE NATIONAL LABORATORY
FOR PERIOD MAY 15, 1966, TO JANUARY 15, 1967

J. H. Coobs
O. Sisman

This document has been reviewed
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COATED-PARTICLE FUELS DEVELOPMENT AT OAK RIDGE NATIONAL LABORATORY
FOR PERIOD MAY 15, 1966, TO JANUARY 15, 1967Compiled by J. H. Coobs and O. Sisman
D. B. Trauger, Director, AGCR ProgramFor presentation at the Twelfth AEC Coated-Particle Working Group
Meeting, Nevada Test Site, March 1967*Las Vegas,*

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COATED-PARTICLE FUELS DEVELOPMENT AT OAK RIDGE NATIONAL LABORATORY
FOR PERIOD MAY 15, 1966, TO JANUARY 15, 1967

Compiled by J. H. Coobs and O. Sisman

INTRODUCTION

This report presents the highlights of the development work at the Oak Ridge National Laboratory during the period May 15, 1966, to January 15, 1967, on graphite-base fuel concepts involving pyrolytic-carbon-coated fuel particles for gas-cooled civilian power reactor applications. Major emphasis has continued on the testing and evaluation of pyrolytic-carbon-coated oxide fuel particles. The mathematical model for coated-particle behavior has been modified to include the effects of irradiation-induced creep and is being used to guide the irradiation program and to correlate results from irradiation experiments. In addition to some details on the revision of the mathematical model, the work described includes pyrolytic-carbon coating studies, out-of-reactor characterization of coated particles, irradiation testing, and postirradiation examination. The results of more or less routine evaluation tests on materials prepared or received during the report period have not been included except where they pertain to irradiation tests or other experiments described in the report.

CHARACTERIZATION OF COATINGS AND COATED PARTICLES

Mathematical Simulation of Coated Particles

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We have previously described a mathematical model for predicting coated-particle behavior^{1,2} which aids in identifying significant features controlling coated-particle performance and in the planning and

¹J. W. Prados and J. L. Scott, Mathematical Simulation of Coated Particle Behavior, pp. 53-59 in "GCRP Semiann. Progr. Rept. Sept. 30, 1965," USAEC Report ORNL-3885, Oak Ridge National Laboratory.

²J. W. Prados and J. L. Scott, "Mathematical Model for Predicting Coated-Particle Behavior," Nucl. Appl. 2, 402-414 (1966).

interpretation of irradiation tests. Agreement between the model's predictions and results from a high-burnup irradiation test at low fast fluence have been encouraging.³ However, our model neglects the effect of creep, which may be significantly enhanced by fast-neutron irradiation at normal fuel temperatures.⁴ Such creep could relieve coating stresses resulting from fast-neutron damage, and thus an elastic model's predictions of failure under high-fast fluence, low-burnup conditions may be overconservative. Hence, we have extended our analysis of stress and strain in spherical pyrolytic-carbon coatings to include creep, and have performed calculations to demonstrate its influence on mathematical-model predictions of coated-particle failure.

The analysis of stress and creep in a spherical pyrolytic-carbon shell, subjected to internal pressure and possible anisotropic dimensional changes^{1,2} poses formidable mathematical difficulties due to the nonlinearity of the governing equations. However, an iterative numerical technique for creep analysis, proposed by Mendelson et al.,⁵ provides a means of obtaining solutions well within the accuracy of existing data by a method of successive elastic approximations. The working equations which are presented elsewhere⁶ must be solved by trial and error for each time increment. In addition, a second trial-and-error solution is required to determine the fission-gas pressure within the particle coating as a function of fuel-kernel properties and burnup, and inner-coating free volume.^{1,2}

These calculations have been programmed for the CDC-1604A computer at ORNL, and results obtained for some typical coated-particle

³J. H. Coobs et al., "Testing of a Design Analysis for Coated-Particle Fuels," Trans. Am. Nucl. Soc. 9, 421 (1966).

⁴R. J. Price and J. C. Bokros, "Mechanical Properties of Neutron-Irradiated Pyrolytic Carbons," USAEC Report GA-6926, General Atomic, May 1966.

⁵A. Mendelson et al., "A General Approach to the Solution of Practical Creep Problems," J. Basic Eng., 81(D), 585-98 (1959).

⁶J. W. Prados, "Calculation of Creep in Spherical Pyrolytic-Carbon Shells Under Combined Radiation Damage and Internal Pressure," Trans. Am. Nucl. Soc. 9, 382-83 (1966).

dimensions, properties, and operating conditions. At present, the functional dependence of coating creep on temperature, stress, fast-neutron flux and fluence are poorly defined, as are the appropriate criteria for coating failure (i.e., limiting stresses and/or strains). We have performed calculations using the radiation-creep relation suggested by Price and Bokros⁴ [steady creep rate approximately $1 \times 10^{-27} \text{ (psi)}^{-1} \text{ (neutrons/cm}^2 \times 0.18 \text{ Mev)}^{-1}$] to predict fast dose and burnup at failure for a typical two-layer coated particle under the assumption that failure will occur when the tangential tensile stress in the outer coating exceeds 30,000 psi. Similar calculations were performed for the same particle, assuming 20% higher creep rate, 100% higher creep rate, and zero creep. The curves relating to fast-dose and burnup at failure for each creep assumption are known as failure loci,^{1,2} and are shown in Fig. 1 for each creep relation assumed. (These are the curves that converge on the abscissa near 20% burnup.) The relation between fast dose and burnup for several types of fuel kernels in different flux spectra are shown in the same figure; the intersection of one of these latter curves with a given failure locus represents the predicted burnup and fast dose at failure for the given fuel kernel and the coating represented by the failure locus. The significance of these relations is discussed more fully elsewhere.^{1,2}

In the present case, it is interesting to note that radiation creep exerts little influence on predicted coated-particle performance for fuel kernels with high ²³⁵U content in low fast-flux environments (conditions typical of many coated-particle irradiation tests). However for kernels with high thorium content, where appreciable fast fluence may be absorbed before significant fuel burnup can occur (conditions more typical of HTGR operation) radiation creep may serve to extend life significantly beyond that predicted by an elastic model. In fact, with sufficiently high creep rates, fast-neutron-induced failure would not be predicted at all, as indicated by termination of the failure loci for the higher creep rate assumed in the present case. This failure-locus termination may be explained by reference to Fig. 2, which contains curves of

240- μ -DIAM, FULLY-DENSE (Th,U)C₂ KERNEL

40- μ INNER PyC LAYER, 37% AVAILABLE VOLUME

40- μ HIGH-DENSITY OUTER PyC LAYER, ANISOTROPY

FACTOR = 1.4

Fig. 1. Influence of Irradiation Creep on Predicted Failure Conditions for a Typical Coated Particle.

Fig. 2. Effect of Creep on Pressure Buildup in Coated Particle.

calculated fission-gas pressure as a function of thermal fluence (proportional to irradiation time) for the same coating dimensions and properties considered in Fig. 1, with various assumed creep coefficients. These curves are only valid for a specific fuel composition (thorium-to-uranium ratio of 3:1) and a specific reactor flux spectrum (fast: thermal ratio of unity). Increasing degrees of creep allow a more rapid increase of fission-gas volume, and hence retard the pressure buildup for a given burnup (and gas-production) rate. For fuel kernels with significant initial fissile content, the burnup rate falls during irradiation; with sufficiently rapid coating creep, the gas production will eventually drop too low to compensate for the increasing volume and the pressure will begin to decrease. If the creep is rapid enough to accommodate radiation-induced dimensional changes, the overall stress level will fall also. Under these conditions, the coating will not fail until, perhaps, some limiting strain is reached. Although data on limiting strains (or even proof of their existence) are lacking at present, it is worthwhile to note that the maximum creep strains represented by conditions of Fig. 2 are in the neighborhood of 3%, still below the maximum values measured by Price and Bokros on pyrolytic carbon coating materials.⁴

This study has demonstrated, qualitatively, the significant influence that radiation-induced pyrolytic-carbon creep can exert on the operating life of a coated-particle fuel. As far as we can determine, the effect of creep is beneficial and may provide a significant increment in coated-particle fuel performance over that predicted from elastic theory in combination with the results of accelerated-burnup, low-fast-fluence irradiation tests.

Most proof tests of coated particles to date have been conducted in such a way that the beneficial effects of creep would not be revealed. Tests at much higher fast fluences for a given fuel burnup will be required to show these effects. In addition, we badly need additional data on the mechanical behavior of pyrolytic-carbon coating materials in the range of temperature and fast-neutron flux levels anticipated for high-temperature gas-cooled reactor fuel operation.

Pyrolytic-Carbon Coating Studies

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Coatings Deposited from Propane in a 1-in.-diam Fluidized Bed⁷

A previous systematic investigation⁸ showed that the properties of pyrolytic-carbon coatings deposited in fluidized beds could be varied in a controllable manner by regulating process parameters. The principal parameters upon which coating properties depended were deposition temperature and space velocity or volume flow rate of the hydrocarbon reactant relative to the surface area of the charge. However, the only carbon source considered was methane. This imposed a practical lower limit of about 1300°C on the deposition temperature and an upper limit on the carbon supply rate of about 20 g-at. per minute per square meter of surface area being coated in the case of 200- μ -diam particles. The lower coating temperature limit of 1300°C is imposed by the kinetics of methane decomposition, and the upper limit on the carbon supply rate is fixed to avoid entrainment of the particles in the gas stream. To use a lower coating temperature or a higher carbon supply rate requires a higher hydrocarbon.

We therefore systematically studied the effects of varying deposition conditions on coating properties with propane as the carbon source. The potential advantages of using the higher hydrocarbon are less energy absorption on decomposition, three times as much carbon per unit volume, less hydrogen generation per mole of carbon, and more favorable decomposition kinetics at lower temperatures. Coating was done in a 1-in.-diam

⁷ Summation of University of Tennessee M.S. Thesis of R. L. Beatty, Pyrolytic Carbon Deposited from Propane in a Fluidized Bed, USAEC Report ORNL-TM-1649, Oak Ridge National Laboratory (November 1966).

⁸ R. L. Beatty, F. L. Carlsen, and J. L. Cook, Pyrolytic-Carbon Coatings on Ceramic Fuel Particles, Nucl. Appl., 1(6): 560-566 (December 1965).

fluidized bed into which a stationary axial probe was inserted, as shown in Fig. 3. The fluidized charge consisted of six graphite disks and 55 g of 420- to 500- μ -diam thoria particles. Deposition temperatures between 1200 and 1900°C were studied, and the propane supply rate was varied from 0.05 to 3.2 cm³/min per square centimeter of particle surface (0.7 to 43 g-at. of carbon per minute per square meter).

Results shown in Fig. 4 are plotted in the form of contour maps and represent the coating deposited on fluidized substrate materials. Anisotropy and crystallite size were measured on disk coatings, and density values were obtained from particle coatings. The deposition efficiency represented the weight of carbon deposited on the particles, disks, and probe divided by the total weight of carbon supplied. The microstructure montage consists of a cross-section photomicrograph of one coated particle from each batch used in the experiments for determining the contour maps.

Attention should be called to one particular feature of the results. The coating deposited at 1200°C at a propane supply rate of 3.2 cm³/min·cm²

Fig. 3. Fluidized-Bed Coating Chamber with Stationary Probe and Water-Cooled Gas Injector.

Fig. 4. Effect of Deposition Conditions on Properties and Microstructures of Pyrolytic-Carbon Coatings Deposited from Propane and on Process Characteristics of the Coating Operation.

is completely random in orientation, has a density of 1.98 g/cm^3 , and is deposited at a rate of nearly $9 \text{ } \mu\text{/min}$. To achieve comparable coating properties from methane requires a deposition temperature in excess of 2000°C and a flow rate that limits the deposition rate to approximately one-tenth the rate achieved here.

The significance of this feature is emphasized by the micrographs of two coating batches shown in Fig. 5. The same $(\text{Th,U})\text{O}_2$ sol-gel microspheres were used in both batches. For batch OR-502, shown on the left, the outer coating was deposited from methane and required about 1 hr at 2000°C , while the outer layer of batch OR-526 was deposited from propane at 1250°C in less than 10 min. Note that large grains developed in the oxide kernels during the high-temperature coating operation. These grains behaved in a brittle manner and made polishing more difficult but are not considered detrimental to the irradiation stability of the oxide. If the low-temperature coating shown here performs as well under irradiation as the high-temperature methane coating, a large reduction in coated particle cost should be possible.

The purpose of the stationary probes was to determine the axial uniformity of the deposition rate and coating characteristics as functions of controllable variables. The probe montage of Fig. 6 is arranged according to deposition temperature and propane supply rate so that trends in the deposition rate profiles may be noted as functions of these variables. Four general patterns of behavior are observed:

1. At the lower coating temperatures the probes coated at all but the lowest propane supply rate show a sudden drastic increase in coating thickness near the top. This point of sudden increase in coating buildup rate probably represents the top of the fluidized bed. These probes, however, show relatively uniform coating thicknesses up to the point of sudden thickness increase; this suggests that the coating deposition rate on the particles is fairly constant throughout the volume of the fluidized bed.

PHOTO 84629

BRIGHT FIELD
ILLUMINATION

POLARIZED
LIGHT

BATCH NUMBER OR-502 100X, AS POLISHED
COATING TEMPERATURES 1050°C, 1150°C, 1050°C, 2000°C
TOTAL DEPOSITION TIME ONE HOUR
OUTER COATING DENSITY 1.9 g/cm³

BATCH NUMBER OR-526 100X, AS POLISHED
COATING TEMPERATURES 1050°C, 1200°C
TOTAL DEPOSITION TIME TEN MINUTES
OUTER COATING DENSITY 2.0 g/cm³

Fig. 5. Metallographic Appearance of High- and Low-Temperature Isotropic Coatings on Sol-Gel (Th,U)O₂ Particles.

Fig. 6. Effect of Deposition Conditions on Axial Coating Profile of Stationary Probe Immersed in Fluidized Bed.

2. The probes coated at the lower temperatures and the lowest propane supply rate appear to be uniformly coated over their lengths, as do those coated at intermediate temperatures and all but the highest propane supply rates. Again a fairly constant particle coating rate throughout the fluidized bed is suggested.

3. The probes coated at the higher temperatures and all but the highest propane supply rates show a markedly thicker coating at the bottom that gradually decreases in thickness toward the top. For each of the higher deposition temperatures the shift of the maximum coating rate toward the bottom of the probe becomes more pronounced with decreasing propane supply rate. This suggests that at high temperatures and low propane supply rates most of the coating on the particles is deposited in the lower part of the fluidized bed.

4. At the intermediate and higher temperatures and the highest propane supply rates, the zone of maximum deposition rate appears to be shifted upward from the probe tip.

The trends shown by these probe-deposit profiles are significant in that they emphasize the nonuniformity of the deposition rate over the fluidized bed volume under certain conditions. Also notable is the dependence of the deposit profile on the coating conditions.

Coatings Deposited from Other Hydrocarbons

The investigation of propane as a carbon source for pyrolytic-carbon coatings was very encouraging. With the low-temperature propane process the density of isotropic coatings can be increased approximately 5% over the density obtainable with the high-temperature methane process. Further, the deposition rate for high-density isotropic coatings can be increased by about an order of magnitude over the rate obtainable with methane.

Several features of the propane process are not ideal, however, and an improvement in any of these features would give the low-temperature coating process an even greater advantage over the high-temperature methane operation. Though high-density isotropic coatings can be deposited at a high rate from propane, the soot and hydrogen generation rates are high, and the cooling effect on the coater system is excessive. The cooling is due to the highly endothermic nature of propane decomposition (24.8 kcal/mole at 298°K) and to the high heat capacity of propane.

To alleviate the heat loss problem, experiments were conducted with propylene (C_3H_6), which is essentially dehydrogenated propane, as the carbon source. Since this dehydrogenation step is probably the first step in the cracking of propane, the carbon deposition mechanism with propane and the desirable coating properties should be maintained. Since the first dehydrogenation step of propane cracking is avoided, the cracking of propylene is slightly exothermic (4.9 kcal/mole at 298°K), and substituting propylene for propane should greatly reduce heat absorption.

Evaluations are not complete, but we conclude from the data that propylene can be used to advantage instead of propane. The optimum coating conditions appear to be essentially the same for the two gases. The optimum coating temperature is about 1250°C, and the concentration of reactant should be high, with undiluted propylene being very convenient to use. Under these conditions isotropic coatings with a density of 2.0 g/cm³ can be deposited at a rate of about 10 μ/min. The cooling effect of propylene is, in fact, very much less than that caused by propane, and the hydrogen-generation rate is reduced by 25%. However, the soot generation is about the same.

Additional hydrocarbons have been investigated. We conclude from preliminary data that they may yield high-density isotropic coatings, but more experiments are needed. Allene (C_3H_4) and methyl acetylene (C_3H_4) cause a slight temperature rise in the coating apparatus, while 1,3-butadiene (C_4H_6) has a cooling effect comparable with that caused by propylene.

Heat Treatment of Pyrolytic-Carbon-Coated Oxide Particles

H. Beutler, R. L. Hamner, and J. W. Prados
Metals and Ceramics Division

We conducted a series of high-temperature heat treatments on multiple lots of pyrolytic-carbon-coated UO_2 and ThO_2 particles in the temperature range 2000 to 2600°C. These experiments were undertaken (1) to identify high-temperature failure mechanisms for pyrolytic-carbon-coated oxide particles that might be significant in reactor hazards evaluation, (2) to determine whether residual carbon contained in the oxide kernel has a significant effect on coated-particle thermal stability, (3) to investigate the feasibility of graphitizing a carbon matrix fuel element containing coated particles, and (4) to obtain estimates of the mechanical properties (particularly high-temperature rupture strength and creep behavior) for pyrolytic-carbon coating materials.

General Effects of Heat Treatment

Nine batches of coated oxide particles were selected for heat treatment. The fuel particles included conventionally prepared and sol-gel-derived UO_2 , sol-gel ThO_2 , and sol-gel $(\text{Th},\text{U})\text{O}_2$. Coating conditions and characteristics of the coated particles are given in Table 1. Two batches of particles (LC-1 and HC-1) having widely different amounts of carbon in the fuel particles were included to study the effects of carbon. These two batches were mixtures of coated UO_2 and coated ThO_2 particles for comparing the effects of heat treatment under the same conditions.

The heat treatments were conducted in a graphite resistance furnace under a flowing argon atmosphere. The samples were heated in graphite crucibles for 2 min at successive 100°C intervals in the range 2200 to 2600°C. The general results of these treatments are summarized in Table 2.

Migration of fuel appeared as isolated spots in the porous layers as if there had been vaporization and condensation. The pattern of the migration is illustrated in Fig. 7. Once fuel migration began after a given heat treatment, the degree of migration increased with increasing temperature but did not extend beyond the porous layer.

Table 1. Conditions for Deposition of Pyrolytic-Carbon Coatings on Particles
Used for Heat-Treatment Studies

Batch	Core Particle	Average Core Particle Diameter (μ)	Coating Layer	Coating Gas	Hydrocarbon Flow Rate ($\text{cm}^3/\text{min} \cdot \text{cm}^2$)	Application Temperature ($^{\circ}\text{C}$)	Average Deposition Rate (μ/min)	Coating Thickness (μ)	Coating Density (g/cm^3)
OR-491	UO_2^a	412	1	C_2H_2	6.6	1080	72	28	0.7
			2	C_3H_8	0.45	1150	0.4	10	2.0
			3	CH_4	1.0	2000	2.6	104	1.9
OR-493	UO_2^a	412	1	C_2H_2	6.6	1080	72	28	0.7
			2	C_2H_2	0.45	1150	0.4	10	2.0
			3	C_2H_2	4.5	1100	72	23	0.7
			4	CH_4	1.0	2000	2.3	81	1.9
OR-494	UO_2^a	412	1	C_2H_2	6.6	1080	72	28	0.7
			2	C_2H_2	0.45	1150	0.4	10	2.0
			3	C_2H_2	4.5	1100	72	55	0.7
			4	CH_4	1.0	2000	2.3	53	1.9
YZP-28	UO_2	208	1	C_2H_2	3.57	1000	22	48	0.7
LC-1	UO_2^b	400	2	CH_4	3.57	1600	4.4	61	1.52
			1	C_2H_2	6.0	1050	56	36	0.7
	ThO_2	280	2	C_3H_8	5.5	1250	18	49	2.0
			1	C_2H_2	6.0	1050	56	36	0.7
HC-1	UO_2^c	400	2	C_3H_8	5.5	1250	18	49	2.0
			1	C_2H_2	6.0	1050	54	31	0.7
	ThO_2	280	2	C_3H_8	5.5	1250	18	44	2.0
			1	C_2H_2	6.0	1050	54	31	0.7
HB-23	UO_2	148	2	C_3H_8	5.5	1250	18	44	2.0
			1	C_3H_8	2.3	1020	9.0	49	1.1
			2	CH_4	1.20	1320	0.03	30	2.05
OD-32	$(\text{Th}, \text{U})\text{O}_2$	286	3	CH_4	0.05	1900	0.13	60	2.0
			1	C_2H_2	6.0	1050	50	25	0.7
			2	C_2H_2	0.5	1150	0.6	10	2.0
			3	C_2H_2	5.0	1080	50	25	0.7
OD-46	$(\text{Th}, \text{U})\text{O}_2$	281	4	CH_4	0.6	2000	1.5	65	1.9
			1	C_2H_2	6.0	1050	50	25	0.7
			2	C_2H_2	0.5	1150	0.6	10	2.0
			3	C_2H_2	5.0	1080	50	25	0.7
OR-638	UO_2	350	4	CH_4	0.6	2000	1.5	65	1.9
			1	C_2H_2	7.8	1050	58	75	0.6
			2	C_3H_8	5.3	1250	0.6	19	2.0
OR-640	UO_2	350	3	CH_4	0.84	2000	1.7	75	1.9
			1	C_2H_2	7.8	1050	58	75	0.6
			2	C_3H_8	5.3	1250	13	80	2.0

^aConventionally prepared fuel; all other particles sol-gel derived.

^bCarbon content 200 ppm.

^cCarbon content 6800 ppm.

Table 2. General Effects of Heat Treatment on Coated Oxide Particles

Batch	Effect of 2-min Heat Treatment at Indicated Temperature				
	2200°C	2300°C	2400°C	2500°C	2600°C
OR-491	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Complete failure; particles fragmented
OR-493	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Complete failure; particles fragmented
OR-494	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating; some particles ruptured	Complete failure; particles fragmented
YZP-28	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Complete failure; particles fragmented
LC-1					
UO ₂	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Complete failure; particles fragmented
ThO ₂	No apparent change	No apparent change	Gap formed between fuel kernel and coating	Gap formed between fuel kernel and coating; fuel migrated into porous layer	Gap formed between fuel kernel and coating; fuel migrated into porous layer
HC-1					
UO ₂	Fuel migrated into porous layer	Fuel migrated into porous layer	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Fuel migrated into porous layer; gap formed between fuel kernel and coating	Complete failure; particles fragmented
ThO ₂	No apparent change	No apparent change	Gap formed between fuel kernel and coating	Gap formed between fuel kernel and coating; fuel migrated into porous layer	Gap formed between fuel kernel and coating; fuel migrated into porous layer
HB-23	No apparent change	No apparent change	Gap formed between fuel kernel and coating	Gap formed between fuel kernel and coating; fuel migrated into porous layer	Gap formed between fuel kernel and coating; fuel migrated into porous layer
OD-32	No apparent change	No apparent change	Gap formed between fuel kernel and coating	Gap formed between fuel kernel and coating; fuel migrated into porous layer	Gap formed between fuel kernel and coating; fuel migrated into porous layer
OD-46	No apparent change	No apparent change	Gap formed between fuel kernel and coating	Gap formed between fuel kernel and coating; fuel migrated into porous layer	Gap formed between fuel kernel and coating; fuel migrated into porous layer

Fig. 7. Microradiograph of Coated Particles (LC-1) After Heat Treatment for 2 min at 2500°C . Note expansion of outer coating and migration of fuel into inner coating of particles with UO_2 cores (larger particles).

Except for one batch (OR-494) there was no evidence of failure through the 2500°C heat treatment. The only batches that survived the 2600°C heat treatment were those with kernels of ThO_2 or $(\text{Th},\text{U})\text{O}_2$ and one batch of coated UO_2 particles (HB-23) that had a relatively high coating thickness-to-kernel diameter ratio (0.87). In these batches there appeared to be little or no reaction between the fuel kernel and the coating upon metallographic examination. The only unusual phenomenon observed in these particles was the formation of a gap in some of the HB-23 particles between inner and outer coating layers, as shown in Fig. 8.

Other results of interest in the heat treatment of coated oxide particles were the effects of carbon and the dimensional changes; these are discussed below.

PHOTO 85048

Fig. 8. Effects of Heat Treatment on Microstructure of Coated Particles. Bright field. 200 $\times$ (Original reduced 51.5%)

Effects of Residual Carbon Content in Sol-Gel UO_2 Microspheres on Their Thermal Stability

Carbon is the only significant impurity in sol-gel-derived UO_2 microspheres. We have demonstrated that this residual carbon can be reduced to below 100 ppm; however, from economic considerations it is desirable to relax limits on carbon impurities as far as possible. This is particularly justifiable in the case of oxide microspheres that are subsequently coated with pyrocarbon. To investigate the effect of residual carbon content on the thermal stability of coated UO_2 microspheres we

heat treated two batches having low and high carbon contents (LC-1, 200 ppm; HC-1, 6800 ppm). The coatings for both batches, which consisted of a porous buffer layer and a high-density isotropic outer layer, were identical. Heat treatment conditions and results are summarized in Table 3. The behavior of the coated UO_2 particles in both batches was remarkably similar with respect to fuel migration and dimensional stability. This suggests that the residual carbon content has no significant effect on the thermal stability of the coated oxide particles.

Table 3. Dimensional Changes of Coated UO_2 Particles Due to Heat Treatment

	LC-1	HC-1
Original dimensions, μ		
Fuel particle diameter	469	435
Total particle diameter	635	597
Dimensional changes, ^a %		
After heating at 2200°C for 1 hr		
Fuel particle diameter	+0.8	-1.3
Total particle diameter	+1.1	+0.7
After heating at 2200°C for 30 hr		
Fuel particle diameter	+0.4	-0.7
Total particle diameter	+1.8	+1.03

^a Average values obtained from six particles.

Dimensional and Structural Changes

For comparison we measured the dimensional changes of both core and coating of individually identified UO_2 and ThO_2 particles of batch LC-1 after heat treatment at 2200 and at 2500°C by microradiography. These coated particles are described in Table 1. Conditions of the heat treatment and results of the measurements are summarized in Table 4. Slight swelling of the fuel kernel (<1%) was noted after heating at 2500°C for 2 min. After a prolonged heat treatment for 10 min at 2500°C a drastic distortion of the fuel particles was observed that made dimensional

Table 4. Dimensional Changes of Coated UO_2 Particles After Heat Treatments at 2200 and 2500°C^a

Heat Treatment		Average Original Particle Dimensions		Average Dimensional Changes		Number of Observations
Temperature (°C)	Time	Core Diameter (μ)	Particle Diameter (μ)	Core Diameter (%)	Particle Diameter (%)	
2200	1 hr	469	635	+0.77	+1.13	6
	30 hr	469	635	+0.36	+1.78	6
2500	2 min	447	635	+0.67	+25.3	1
	10 min	397	582	(b)	+32.9	5

^aNo dimensional changes of particle and coating could be detected on coated ThO_2 particles with identical coatings exposed to the same heat treatment.

^bFive of ten heat-treated particles failed; pronounced deformation of core occurred; dimensional change of core was not measurable.

measurements impossible. Large expansion of the total particle diameter (up to 42%) and thinning of the outer coating were observed after heating at 2500°C. A typical expanded coating is compared with its original configuration in Fig. 9. A slight but positive expansion of the outer coating was noticed after heating at temperatures as low as 2200°C for 1 hr, but after a prolonged heat treatment (30 hr) at the same temperature the average expansion of the coating was not significantly greater if data scatter and experimental errors are considered. No dimensional changes of core or coatings could be detected for the ThO_2 particles.

Coated oxide particles can only survive such heat treatments if the outer coating is able to retain the equilibrium CO pressure necessary to suppress the carburization reaction. For coated particles containing UO_2 cores the equilibrium CO pressure is estimated to be about 18 atm at 2200°C and 150 atm at 2500°C. On the other hand, the equilibrium CO

Fig. 9. Comparison of Microradiograph of Coated UO_2 Particle Before and After Heat Treatment for 10 min at 2500°C .

pressure in coated ThO_2 particles is estimated to be about 35 atm at 2500°C . We attribute the observed expansions of the outer coating on UO_2 particles to creep resulting from internal pressurization.

In a subsequent series of experiments we attempted to investigate the creep behavior of outer coatings under more refined experimental conditions. A graphite holder containing 16 particles in separated compartments was inserted into a preheated furnace stabilized at the required temperature. After the desired exposure, the holder was rapidly retracted from the hot zone of the furnace. With this technique it was possible to limit and control heatup and cool-down times. We investigated the effect of heat treatment time on the dimensional changes of two outer coatings having similar properties but deposited under different conditions. Both coatings were of high-density isotropic structure and identical in dimensions; they were deposited over a porous inner coating on the same batch of UO_2 particles. The outer coating of batch OR-638 was deposited from methane at 2000°C , whereas the outer coating of batch OR-640 was deposited from propylene at 1250°C . Coating conditions and particle specifications are given in Table 1. Results of heat treatments at 2300°C ranging from 1 min up to 300 min are summarized in Table 5, and a graphical representation of the data is given in Fig. 10.

The most significant dimensional change of both coatings occurred within 1 min of exposure, while no measurable additional expansion was observed after exposures of 300 min, as was observed in the experiment

Table 5. Effect of Heat Treatment Time on Dimensional Changes of Coated Particles Heated at 2300°C^a

Batch	Heating Time (min)	Expansion of Outer Coating Diameter ^b (%)	Contraction of Coating Thickness ^c (%)	Number of Failed Particles Observed
OR-638	1	0.28	2.2	
	3	0.14	2.13	
	10	0.32	6.36	
	30	0.19	3.18	
	100	0.20	3.47	
	300	0.25	2.88	
OR-640	1	29.6	25.5	1
	3	27.0	18.64	
	10	33.7	22.66	1
	30	28.0	23.48	1
	100	25.89	18.42	
	300	27.9	18.94	1

^aData represents average values for eight observations.

^bChange in total particle diameter versus original diameters.

^cChange in coating thickness versus original thickness.

Fig. 10. Effect of Annealing Time on Dimensional Change of Coated UO₂ Particles Heated at 2300°C.

previously discussed. This behavior is somewhat surprising since the large expansion and concurrent thinning of the outer coating observed in some coatings result in a drastic increase in stress level (up to 90%, within experimental accuracy). The coatings appear dimensionally stable in spite of increased stress. The initial expansions measured for coatings deposited from propylene at 1250°C were approximately 100 times those of the coatings deposited from methane at 2000°C. Large variations in expansion behavior of individual particles were observed. Furthermore some particles failed, whereas others survived the heat treatment. We were not able to relate these variations to size or coating thickness of the original particles.

We investigated structural changes of heat-treated particles by metallography. Polarized-light examination revealed an increase in preferred orientation in the pyrolytic-carbon coating due to heat treatments. A comparison between as-coated and heat-treated particles of batch LC-1 is shown in Fig. 11. A pronounced increase in preferred orientation is evident from the intensity of the Maltese cross. This effect was observed with initially isotropic coatings derived from methane, propane, and propylene. No change in preferred orientation or dimensional change was observed in identical coatings deposited on thoria particles, as was discussed earlier. Therefore, the observed reordering of crystallites must be associated with a strain effect rather than temperature or heat-treatment time. Previous heat-treatment studies by Carlsen⁹ on unstressed disk specimens of isotropic pyrolytic carbon did not reveal any changes of preferred orientation due to high-temperature anneals. Our preliminary data suggest that the degree of change in preferred orientation is closely related to the total amount of strain induced and is independent of temperature in the range investigated. By comparing the difference in behavior of low-temperature coatings deposited from propane on ThO₂ and

⁹F. L. Carlsen., Effects of Heat Treatments on Pyrolytic Carbon, pp. 28-33, GCRP Semiann. Progr. Rept. Sept. 30, 1965, USAEC Report ORNL-3885, Oak Ridge National Laboratory.

Fig. 11. Comparison of Microstructures of Coated ThO_2 and UO_2 Microspheres Before and After Heat Treatment at 2500°C for 10 min. 100x (Original reduced 40.5%)

UO₂ particles, as well as coatings of various thicknesses deposited on such particles, it appears that a certain critical stress level (estimated to be 1000 psi) is required to initiate the structural change. Once this level is exceeded the induced strain appears to be directly related to the stress level. High-density isotropic coatings deposited from methane appear to behave similarly; however, much higher stress levels (by a factor of 10) are required to produce the same effect.

One of the major causes leading to coated-particle failure during irradiation is excessive internal pressurization¹⁰ due to fission-gas buildup. The internal CO pressurization of coated oxide particles exposed to high temperature is a unique means of simulating fission-gas buildup and determining mechanical properties. The temperatures required, however, are well in excess of actual operating conditions; furthermore, only limited thermodynamic data on the oxide-carbon reactions in the temperature range of interest are available. We estimated lower rupture stress limits for coated particles that survived the heat treatments on the assumption that overpressurization alone would cause failure. The values obtained for both low- and high-temperature isotropic coatings varied between 8,000 and 10,000 psi. In isolated instances failure occurred at much lower calculated stresses. Room-temperature data¹¹ obtained from bend tests on pyrolytic-carbon disks indicate actual rupture stresses in the neighborhood of 30,000 psi for similar coating materials. A value nearer this level was calculated in the case of a high-density granular coating deposited from methane at 2000°C (HB-23) that survived a heat treatment at 2800°C for 1 min. In this case we estimated the maximum tensile stress perpendicular to the radius after 40% strain to be 28,500 psi.

¹⁰J. W. Prados and J. L. Scott, Mathematical Model for Predicting Coated-Particle Behavior, Nucl. Appl., 2(5): 402-414 (October 1966).

¹¹J. C. Bokros and R. J. Price, Deformation and Fracture of Pyrolytic Carbons Deposited in Fluidized Beds, USAEC Report GA-6332, General Atomic Division, General Dynamics Corporation (to be published in Carbon).

IRRADIATION TESTING OF COATED PARTICLES

In-Pile Sweep Tests for Fission Product Release
and Coating Failure Criteria

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The A9 and B9 capsule design has been modified to study the release and migration of fission solids. This was done by isolating the graphite holder within a gas gap and deposition plates and passing the exit helium through a cooling coil and filter assembly adjacent to the test capsule (see Fig. 12). Two of the experiments described below were built to include fission solid release studies. One of these was with UO_2 particles coated with pyrolytic carbon; the other was with UC_2 particles with a pyrolytic carbon coating containing a silicon carbide barrier layer.

Fig. 12. Gas-Gap Fission Solid Capsule.

Several experiments were performed to study the optimum coating thickness, and optimum ratio of inner porous coating layer to outer dense coating layer. One experiment was operated to test the Prados-Scott model for calculating the safe operating limits for a specific coating.

Descriptions of the particles and the irradiation test data, including fractional fission gas release, are given in Table 6.

Experiment A9-5

Capsule A9-5 contained 50 pyrolytic carbon coated uranium oxide particles from batch OR-YZ71, plus 4900 particles from batch OR-343 to provide enough fuel to bring the OR-YZ71 particles up to the desired irradiation temperature. The OR-YZ71 coated particles had a low-density isotropic inner coating and a high-density isotropic outer coating on 420-435 μ diameter UO_2 cores. All the OR-YZ71 particles were coated in the same coating run, but during deposition of the outer coating, some coated particles were removed at intervals to obtain different thicknesses of the outer coating. The inner coating thickness was 50 μ and the outer coating varied as shown in Table 7. The object of the experiment was to see at what flux and temperature the coatings would fail because of thermal stresses and internal pressure.

These coated particles were irradiated at 1250°C to 1.2 atom percent uranium burnup at a rate of 3.9% per month. The capsule was then inserted into a higher neutron flux for an additional 9% burnup at 1450°C, at a rate of 5.2% per month. Three bursts of fission gas were released at 8% burnup and another burst at 8.75%. The fractional fission-gas release for ^{88}Kr was in the 4×10^{-7} range at the beginning of the test and increased to 2.2×10^{-5} during the test. The average fractional fission-gas release data for the last week of the test are given in Table 6.

When the coated particles were examined after irradiation, all the OR-YZ71 particles appeared to have sound coatings and no cracked or broken coatings could be found, even in the thinnest coatings. However, we found four cores and several coating fragments from the OR-343 particles.

Table 6. Fission-Gas Release from Pyrolytic Carbon Coated Fuel Particles During Irradiation

Capsule No.	Batch	Coating* Type and Thickness (μ)	Fuel Material Diameter (μ)	Burnup % H.M.	nvt Fast (above .2 Mev)	Irrad. Temp. ($^{\circ}$ C)	Average Fractional Fission-Gas Release, R/B				
							^{85m}Kr	^{88}Kr	^{87}Kr	^{133}Xe	^{135}Xe
A9-5	OR-YZ71 ^a OR-343	PyC Iso I + Iso II 105-215	UO ₂ 420	1.2 9.0 10.2	1.3×10^{20}	1250 1450	2.2×10^{-5}	1.7×10^{-5}	1.5×10^{-5}	1.6×10^{-5}	1.2×10^{-5}
A9-6	OR-348	Porous C + H.D. Iso 124	UO ₂ 218	7.8	1.1×10^{20}	1500	1.7×10^{-5}	1.3×10^{-5}	1.0×10^{-5}	1.7×10^{-5}	0.9×10^{-5}
A9-7	GA-327	PyC + SiC 90	(Th,U)C ₂ 196	3.2	1.9×10^{20}	1300	8.8×10^{-8}	6.4×10^{-8}	4.8×10^{-8}	9.0×10^{-8}	4.9×10^{-8}
A9-8	OR-653 OR-654	Porous C + H.D. Iso 98	UO ₂ (sol-gel) 270	16.8	2.4×10^{20}	1400	3.4×10^{-8}	2.3×10^{-8}	1.8×10^{-8}	0.9×10^{-8}	0.5×10^{-8}
B9-30	OR-491 OR-493 OR-494	Porous C + Iso 142	UO ₂ 412-429	1.9	2.6×10^{19}	1400	3.5×10^{-4}	2.7×10^{-4}	2.4×10^{-4}	b	b
B9-31	OR-YZ66	H.D. Laminar + H.D. Iso 123	UO ₂ (sol-gel) 202	23.4	3.1×10^{20}	1400	6.9×10^{-4}	5.7×10^{-4}	4.4×10^{-4}	3.6×10^{-3}	6.8×10^{-4}
B9-32	OR-596	Porous C + H.D. Iso 116	UO ₂ (sol-gel) 277	26.1	3.7×10^{20}	1400	1.2×10^{-4}	1.0×10^{-4}	0.8×10^{-4}	1.1×10^{-4}	0.7×10^{-4}

^aCoating thickness from 105 to 215 μ .^bData not available.

*PyC = Pyrolytic carbon; H.D. = High density; Iso = Isotropic.

Table 7. OR-YZ71 Particles in Capsule A9-5

Batch	Outer Coating Thickness, μ	Inner Coating Thickness, μ	No. of Particles Irradiated in Capsule
OR-YZ71A	55	50	5
OR-YZ71B	90	50	10
OR-YZ71C	130	50	15
OR-YZ71D	165	50	20

Postirradiation examination of the OR-YZ71 coated particles, including particles with all four coating thicknesses, showed no evidence of potential failures. Some of the impurities noted in the cores of the unirradiated particles were redistributed during irradiation and collected at the core-coating interface. No deleterious effects to the coatings as a result of the impurities in the fuel were noted. We have concluded that even the relatively thin coatings of the OR-YZ71 coated particles are sufficiently strong to withstand the fission rate, temperature, and burnup in this experiment. Typical irradiated particles with four different coating thicknesses are shown in Fig. 13.

Metallographic examination of the OR-343 coated particles revealed severe damage at the particle-coating interface. The damage was in the form of spiral-shaped cracks that originated at the interface and extended into the coating. In the most severe cases, the cracks had penetrated about one-third of the coating thickness. Examination of unirradiated OR-343 particles revealed similar but finer cracks at the particle-coating interface, and it appears that irradiation caused these cracks to enlarge and propagate to failure in a few cases. An irradiated particle that had moderate damage at the interface is shown in Fig. 14, along with an unirradiated particle with fine cracks at the interface.

Fig. 13. Typical Pyrolytic-Carbon-Coated UO_2 Particles (OR-YZ71) Irradiated in Capsule A9-5. As polished. 100X. Particles with four coating thicknesses are shown: (a) 105 μ , (b) 140 μ , (c) 180 μ , and (d) 215 μ . Original reduced 27.5%.

Fig. 14. Pyrolytic Carbon Coated Uranium Oxide Particles from Batch OR-343. 200X. (a) Unirradiated; (b) irradiated to 10.2% burnup at 1250 and 1450°C in capsule A9-5.

Experiment A9-6

Capsule A9-6, which contained particles from batch OR-348, was irradiated at 1500°C to 7.8 at. % uranium burnup. This capsule was equipped with deposition plates, a cooling coil, and a filter for studying the release of solid fission products. The fission-gas release rates increased continuously throughout the first month of the test, but no bursts of fission gas were detected. The average fractional fission-gas release for the last week of the test is given in Table. 6. It was about two orders of magnitude lower at the start of the experiment.

Metallographic examination of 97 coated particles from this capsule disclosed no failed coatings or evidence of potential failure. The only obvious microstructural changes observed as a result of the irradiation were confined to the UO_2 . The porosity noted in the unirradiated UO_2 was redistributed primarily along grain boundaries. In a few regions in several coated particles the UO_2 was apparently bonded to the inner buffer coating, but with no apparent deleterious effect to the coating. A typical irradiated particle is shown in Fig. 15.

Fig. 15. Typical Pyrolytic-Carbon-Coated UO_2 Particle from Batch OR-348 Irradiated in Capsule A9-6. As polished. 200X

The behavior of the solid fission products was investigated by disassembly and radiochemical analysis of the activities associated with various components of the capsule: a graphite cylinder, loaded with 0.823 g of coated particles, a concentric graphite shell, which was separated from the fuel holder by a gas gap, and an Inconel irradiation capsule. Each of the components of the assembly was either dissolved or thoroughly leached in acid for fission product recovery. These data, in fractions of the total amount generated by fission, based on the most abundant nuclide, are shown in Table 8. Since no appreciable fractions of the fission products were found downstream from the Inconel capsule, the data from these components (cooling coil, lead tube, etc) have been omitted from the table. The data indicate that more than half of the ^{89}Sr escaped from the fuel particles, whereas no significant amounts of ^{95}Zr and ^{144}Ce were found outside the particles. A fair amount of the ^{89}Sr was also found on the graphite deposition shell and the Inconel capsule. The ^{137}Cs which escaped was collected on the cooler Inconel capsule. In future experiments we intend to look for ^{140}Ba as well as some other of the shorter-lived fission products.

Table 8. Fission Product Release During Irradiation Experiment A9-6

Component	Temp. (°C)	Fractions of Total Formed*			
		^{89}Sr	^{95}Zr	^{137}Cs	^{144}Ce
Fuel Particles	1500	0.227	0.775	0.794	0.998
Graphite Holder	1500	0.377	2.46×10^{-6}	7.31×10^{-4}	1.28×10^{-3}
Graphite Deposition Shell	1200	0.0217	7.39×10^{-5}	1.98×10^{-4}	8.44×10^{-4}
Inconel Capsule	800	0.0714	1.13×10^{-5}	0.0478	9.35×10^{-6}
Total		0.697	0.775	0.843	1.000

* Based on ^{144}Ce .

Experiment A9-7

Capsule A9-7, which contained coated particles from batch GA-327, was irradiated to 3.2 at. % heavy-metal burnup at 1300°C. These particles are (Th,U)C₂ (3:1 thorium-to-uranium ratio) coated with pyrolytic carbon plus a barrier layer of silicon carbide (see Fig. 16). During the irradiation, a malfunction of the capsule position-control instrument caused the capsule to be inserted to the maximum neutron flux and the temperature was increased from 1300 to 1700°C. After 10 min at 1700°C, the capsule was returned to its original position and the temperature returned to 1300°C. No bursts of fission gas were released as a result of this temperature excursion. The fission-gas release rates remained in the 10⁻⁸ range throughout the test and the average fractional release given in Table 6 is for the last week of the test.

The results of postirradiation annealing studies on these coated particles are presented in another part of this report.

Fig. 16. Unirradiated Pyrolytic-Carbon- and Silicon Carbide-Coated (Th,U)C₂ Particles from Batch GA-327. 200X (Original reduced 3.5%)

Experiment A9-8

This experiment was conducted to test one of the new low-temperature, high deposition rate coatings. The coatings were deposited from propylene on sol-gel UO_2 particles. The porous inner layer was deposited from acetylene. Coated particles from two batches were used in this experiment. Batch OR-653 coated particles had an inner porous layer $45\ \mu$ thick with an outer high-density isotropic layer $50\ \mu$ thick; the inner layer for the OR-654 coated particles was $30\ \mu$, the outer layer was $72\ \mu$.

Based on alpha count, the coating surface uranium contamination was less than $1 \times 10^{-5}\%$ after leaching. These coated particles were irradiated for 2050 hr at 1400°C to 16.8 at. % heavy metal burnup. The fractional ^{88}Kr release remained near 2.5×10^{-8} until the last few days of the test, when it increased rapidly to 1×10^{-5} . When the particles were examined after irradiation, there was evidence of oxidation of the graphite holder, and a few particles seemed to have pit marks on the coating. The oxidation of the particles and consequent increase in fission-gas release seems to indicate that the capsule sprung a leak near the end of the experiment. Postirradiation metallography is in progress.

Experiment B9-30

Capsule B9-30, which contained particles from batches OR-491, -493, and -494, was irradiated at 1400°C to 1.8 at. % uranium burnup. After 159 hr of irradiation time, eight successive bursts of fission gas were released. Several of these bursts were of sufficient magnitude to drive the exit helium monitor upscale enough to cause the capsule to be withdrawn from the reactor flux. The test was therefore terminated.

The purpose of this test was to study the effect of ratio of thickness of inner porous carbon coating to the thickness of the dense pyrolytic-carbon outer coating. The coating thicknesses are given earlier in Table 1 and below in Table 9.

After irradiation, the OR-491 and OR-493 coated particles were removed from the capsule separately from the OR-494 particles. No broken or damaged OR-491 or OR-493 coated particles were found, but many of the

Table 9. Data on Pyrolytic-Carbon-Coated UO_2 Particles
Contained in Capsule B9-30

	Batch OR-491	Batch OR-493	Batch OR-494
Average particle dimensions, μ			
Total diameter	713	703	704
Fuel particle	429	419	412
Porous coating thickness	28	61	93
Total coating thickness	142	142	146
Number of particles irradiated	5	10	570

OR-494 particles were damaged. It was estimated that from 5 to 10% of these particles had holes in the coating. These defects appeared to be limited to one hole per particle, they were about one-tenth the diameter of the particle, and there were no cracks extending from the holes into the coating surfaces.

All 15 coated particles from batches OR-491 and OR-493 were examined metallographically. Although none of the OR-491 or OR-493 coatings failed, in both cases about half of the inner porous carbon coating had been displaced after irradiation. There was no evidence of carbide formation in any of these particles. A typical irradiated particle from batch OR-491 is shown in Fig. 17. The fuel in the coated particles from batch OR-493 appeared to have moved through the inner porous coatings unilaterally; the outer coatings had not been affected (shown in Fig. 18). The microstructure of the fuel indicated that the UO_2 experienced temperatures in excess of the 1400°C reported; however, the appearance of the fuel could be misleading because of the relatively high impurity level in the UO_2 . These UO_2 particles were used in an earlier capsule, A9-5, and were known to contain as much as 3500 ppm aluminum and 200 ppm calcium.

Metallographic examination of 100 coated particles from batch OR-494 disclosed only two failures; however, numerous particles were observed that, for all practical purposes, had failed. These particles also showed unilateral movement of the UO_2 through the coatings. The fuel in these coated particles appeared to have operated at a higher temperature than

Fig. 17. Pyrolytic-Carbon-Coated UO_2 Particles from Batch OR-491.
(a) Unirradiated. (b) Irradiated to 1.86 at. % burnup at 1400°C in capsule B9-30. 200X

Fig. 18. Pyrolytic-Carbon-Coated UO₂ Particles from Batch OR-493.
(a) Unirradiated. (b) Irradiated to 1.86 at. % burnup at 1400°C in capsule B9-30. 200X

the fuel particles from batches OR-491 and OR-493. This was probably the result of the thicker inner porous layer. None of the damaged coated particles showed evidence of carbide formation, but small crystals of carbide were observed at the inner surface regions of the failed coatings. Several irradiated coated particles are shown in Fig. 19.

Experiment B9-31

This experiment was performed to test a special batch of coatings that were designed by J. W. Prados to withstand about 15% burnup without coating failure. The Prados-Scott model by which the dose at failure was calculated has been reported.¹² To test the model, we irradiated a capsule containing about 4000 coated particles with structural characteristics for which the model was readily applicable. These were sol-gel UO₂ particles, coated with a high-density laminar pyrolytic carbon inner coating and a high-density isotropic carbon outer coating (see Fig. 20). The average fuel particle diameter was 202 μ , and the average coating thickness was 123 μ . These particles (designated OR-YZ66) were irradiated at 1400°C in the B9 facility. After 1209 hr (15 at. % U burnup), the particles began to release bursts of fission gas at irregular intervals. The bursts continued, and at the end of the test (23% burnup) we had observed 197 bursts. Postirradiation examination of the coated particles showed that about 5 to 10% of the coatings had indeed broken and in many cases were completely separated from the uranium oxide particles as shown in Fig. 21. Metallographic examinations are in progress, and postirradiation burnup determinations are being made to check the values obtained by argon activation during irradiation.

¹²J. W. Prados and J. L. Scott, "Mathematical Model for Predicting Coated Particle Behavior," Nucl. Appl. 2(8): 402-414 (October 1966).

Fig. 19. Unirradiated OR-494 Particle and Five Irradiated OR-494 Particles from Capsule B9-30. Inner surface of the failed coating is covered with carbide crystals. As polished. 100X (Original reduced 17%)

Fig. 20. Unirradiated Pyrolytic-Carbon Coated Sol-Gel UO_2 Particles from Batch OR-YZ66. 200X.

Fig. 21. Pyrolytic-Carbon-Coated Uranium Oxide Particles from Batch OR-YZ66 Irradiated to 23 at. % U Burnup at 1400°C in Capsule B9-31. 30X.

The calculations had predicted coating failure at 14.2 at. % burnup. Since the first failure occurred at 15% burnup, the experiment seems to check the model very well. However, we would have expected to see a rather larger fraction of failures by 23% burnup. If the burnup values are verified by postirradiation analysis we must conclude that the model predicts a rather conservative value, and some changes may be in order in some of the parameters in the equations.

Experiment B9-32

The OR-596 fuel particles in experiment B9-32 were sol-gel UO_2 coated with a high-density isotropic outer layer deposited at low temperature from a 50-50 mixture of propylene and hydrogen. The porous inner coating was deposited from acetylene. In addition to testing this new type coating, this experiment was conducted to study the release of solid fission products during irradiation.

These coated particles were irradiated at 1400°C to a uranium burnup of 26%. The fission-gas release rate (R/B) was in the 10^{-7} range for ^{88}Kr at the start of the experiment, but increased gradually over the first twenty days to about 1×10^{-4} . It then remained nearly constant for the rest of the test.

The experiment has been removed from the reactor, and no damage to the coatings was evident when viewed at 30x. We are proceeding with metallographic examination of the coated particles, and analysis of the components of the experiment for solid fission product release.

Fission-Product Release from High-Burnup Coated UO_2 and UC_2
Particles by Postirradiation Annealing

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About 100 coated particles each of 8 types of coated particles were placed in separate compartments of an irradiation capsule (A9-4) and irradiated to a burnup of 14.5% of the uranium metal (in the A9 facility of the ORR) at an irradiation temperature of 1400°C. The four types of coatings represented are given in Table 10. Details of coating properties are given in Figs. 22 and 23. Samples of 10 coated particles from each batch were annealed at 1700°C and duplicate samples were annealed at 2000°C for a total of 19-1/2 hours for each sample in steps of 6-1/2 hours. The annealing procedure and apparatus were described previously.^{13,14}

Table 10. Four Types of Coatings Represented in the A9-4 Experiment

Number	Density, g/cm ³	Anisotropy Factor	Crystallite Size, (L_c), Å
1	2	1.5	30
2	1.5	1	50
3	1.9	1	115
4	2	1.5	120

Evaluation of Fuel Migration by Metallography and Microradiography

Microradiographs and metallographic photomicrographs were made on each of the eight batches of coated particles in the A9-4 experiment. The micrographs and radiographs were made on the as-coated particles, on

¹³M. T. Morgan, D. C. Evans, and R. M. Martin, Fission-Gas Release from High-Burnup UO_2 , pp. 103-106, GCRP Semiann. Progr. Rept. Mar. 31, 1963, USAEC Report ORNL-3445, Oak Ridge National Laboratory.

¹⁴M. T. Morgan et al., Fission-Gas Release from High-Burnup Coated Particles, pp. 149-152, GCRP Semiann. Progr. Rept. Sept. 30, 1963, USAEC Report ORNL-3523, Oak Ridge National Laboratory.

Fig. 22. Photomicrographs of Pyrolytic-Carbon-Coated UO₂ and UC₂ Particles Before and After Irradiation and After Postirradiation Heat Treatments at 1700°C and 2000°C, Respectively. Groups YZ-2, -4, -14, and -13.

Fig. 23. Photomicrographs of Pyrolytic-Carbon-Coated UO₂ and UC₂ Particles Before and After Irradiation and After Postirradiation Heat Treatments at 1700°C and 2000°C, Respectively. Groups YZ-11, -15, -18, and -19.

the irradiated coated particles before heating, and on the irradiated coated particles after the 1700°C and 2000°C anneals. Representative pictures are shown in Figs. 22 through 25.

All micrographs of unirradiated coated particles with the exception of YZ-2 are typical; being selected from a large group. The micrograph of the YZ-2 unirradiated coated particle is typical except for the porous lamina near the inner radius of the outer coating. This was produced during coating in a few particles believed to have been projected into the higher region of the coating tube where gas phase nucleation occurs. Micrographs of irradiated and heated particles are representative of groups of less than a dozen particles.

UO₂ Coated Particles. - Reaction areas in the UO₂ coated particles were restricted to the inner coating as seen in both the micrographs, Figs. 22 and 23 and the radiographs, Figs. 24 and 25. Spearhead-type attack occurred during irradiation in coated particles with low temperature outer coatings, and the UO₂ seemed to expand to fill the void areas. This may be seen in the YZ-2 and YZ-14 particles, Fig. 22. In the UO₂ coated particles with outer coatings deposited at 2000°C, spearhead attack is not evident and penetration of the inner coating is slight. The increased resistance to reaction of the YZ-11 and YZ-18 inner coatings is probably due to the 2000°C heat treatment received during the deposition of the outer coatings. It can be seen that the zone between the core and the inner coating is thicker in the YZ-11 and YZ-18 unirradiated coated particles than in the YZ-2 and YZ-14 coated particles.

The UO₂ particles lost crystalline detail during irradiation and void areas were redistributed as smaller voids. No carbide formation was evident. Dark or shady areas in the inner coating may be seen concentrated at reaction sites or distributed uniformly around the UO₂ particle. These areas became darker and expanded during heat treatment, but did not expand noticeably into the coating. The outer diameters of the shaded areas in the micrographs correspond with the diameter of the dense areas in the radiographs, indicating that uranium diffusion had occurred into the shaded areas. Darkening of these areas after heat treatment may be caused by fission products being driven out of the UO₂

PHOTO 84944

Fig. 24. Microradiographs of Pyrolytic-Carbon-Coated UO₂ and UC₂ Particles Before and After Irradiation and After Postirradiation Heat Treatments at 1700°C and 2000°C, Respectively. Groups YZ-2, -4, -14, and -13.

Fig. 25. Microradiographs of Pyrolytic-Carbon-Coated UO₂ and UC₂ Particles Before and After Irradiation and After Postirradiation Heat Treatments at 1700°C and 2000°C, Respectively. Groups YZ-11, -15, -18, and -19.

into areas where uranium had diffused into the coating. White spots in the UO_2 are believed to be metallic fission product alloys.

UC₂ Coated Particles. - The UC₂ fuel showed an apparent fine porosity after irradiation. Heat treatment at 2000°C seemed to have consolidated the fine porosity into larger pores or voids. Some graphite flakes were seen around the periphery of the fuel particles. The particle-coating reaction areas in the carbide coated particles were not as local as the spearhead attack in the oxide coated particles, and were associated with a progressive, more extensive diffusion of fuel into the coating. Fuel diffusion into the inner coatings started during the manufacture of the particles and can be seen in the radiographs of the unirradiated carbide coated particles, Figs. 24 and 25. Penetration of the inner coatings was complete in all carbide coated particles after irradiation as indicated by the radiographs and by the spotted areas in the micrographs. Diffusion of fuel deep into the low-density outer coating was evident in the irradiated YZ-13 coated particles. Large voids and separation of the inner coating from the outer coating were apparent in the YZ-15 and YZ-19 coated particles after heat treatment at 1700°C and 2000°C.

Coating Stability

No coating failures were observed in the eight groups during irradiation at 1400°C. Coatings of the UC₂ particles failed during the postirradiation tests at both 1700°C and 2000°C, but no failures occurred with the UO₂ particles. Based on ⁸⁵Kr release, approximately 30% of the YZ-4 sample and 25% of the YZ-13 sample failed at 1700°C. At 2000°C, 80% of the YZ-4, 70% of the YZ-13, and 45% of the YZ-19 samples failed. Failures all occurred during heatup or within a few minutes after the annealing temperature was reached.

The two samples of UC₂ coated particles which did not fail at 1700°C (one of which also survived the 2000°C anneal) had undergone the heat treatment during fabrication. We think the migration of fuel into the coatings of UC₂ particles weakened the coating, and caused the high percentage of failures.

Postirradiation Annealing of Coated Particles from A9-7 Experiment

The GA-327 (Th,U)C₂ pyrolytic carbon coated particles with a silicon carbide barrier layer were annealed after irradiation. These coated particles were irradiated in the A9-7 experiment to a burnup of 3.2% heavy metal at a temperature of 1300°C. A sample of the irradiated particles was separated into three groups of 50 particles each for annealing at temperatures of 1300°C, 1700°C, and 2000°C, respectively. The 1300°C and 1700°C groups were annealed for 6-1/2 hours each. The 2000°C group was annealed in steps of 6-1/2 hours for a total of 32 hours.

Fission product release was negligible at 1300°C and 1700°C, except for ¹³⁷Cs, as shown in Table 11 below. Release rates at 2000°C were low during the first 6 hours but increased greatly during the rest of the 32-hour anneal. The results of the 2000°C anneal are shown in Fig. 26. The appearance of the SiC coating before and after the 2000°C anneal is shown in Fig. 27.

Table 11. Postirradiation Anneal of A9-7 Experiment
(Fission product fractions collected in
6-1/2 hrs in ppm or % as noted.)

Temp.	⁸⁹ Sr	⁹¹ Y	¹⁴⁰ Ba	⁹⁵ Zr	¹⁴⁴ Ce	¹⁰³ Ru	¹⁰⁶ Ru	¹³⁷ Cs
1300°C ppm	6.0	3.3	≤ 1.1	≤ 3.6	20	~4.4	150	2400
1700°C ppm	9.8	3.0	≤ 14	≤ 2.1	15	≤ 2.2	≤ 57	32000
2000°C ppm	8.4	4100	~12	1900	1200	1700	1700	39000
32 hrs at 2000°C %	0.8	48	0.08	2.0	39	3.3	3.2	29

Fig. 26. Fission-Product Release During Postirradiation Annealing at 2000°C of (Th,U)C₂ Particles Coated with Pyrolytic Carbon and Silicon Carbide Barrier Layer.

PHOTO 86376

BRIGHTFIELD ILLUMINATION

POLARIZED LIGHT ILLUMINATION

BEFORE IRRADIATION

POST-IRRADIATION

IRRADIATED AND ANNEALED
32 hr AT 2000°C

Fig. 27. Photomicrographs of Particles from Experiment A9-7 After Irradiation and After Heating.

High-Burnup Irradiation Tests and ORR Loop 1 Experiments

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ORR Loop 1 Experiment 15

Except for the results of metallographic examination, this experiment was reported previously.¹⁵ The test element in this experiment consisted of an upper region with an annular bed of coated particles in a graphite fuel body, and a lower region containing a fuel body which was sealed from the loop coolant, and consisted of a graphite cylinder with two axial holes that were filled with a mixture of two types of coated fuel particles. In one hole the particles were loose and in the other the particles were in a lightly bonded bed.

The irradiation lasted for 4920 hours. The fuel temperature in the upper, vented region was 1400°C at the inner diameter of the annular fuel bed and 1000°C at the outer diameter. The fuel in the lower, sealed region operated at 650-625°C. The heavy metal burnup was 12% in the upper capsule, and 9% in the lower capsule. The R/B for fission gas increased from the 10^{-6} range at the start of the irradiation to the 10^{-4} range at termination.

Postirradiation examination of the fuel from the upper region showed no broken coatings with the exception of a few coating fragments from the ThO₂ particles (GA-324). The character of the fractures and the core material involved indicate that the coating failures were mechanically induced rather than an irradiation effect. The only obvious microstructural changes noted in the duplex-coated (Th,U)C₂ particles were (1) densification of the inner buffer coating, (2) loss in crystalline detail of the carbide, and (3) localized intrusion of the carbide into the buffer coating in a few of the particles. Examination of the duplex-

¹⁵I. T. Dudley et al., "ORR Loop No. 1, Experiment 15," Coated Particle Fuels Development at Oak Ridge National Laboratory for Period November 15, 1965 to May 15, 1966, pp. 42-46, USAEC Report ORNL-TM-1528, Oak Ridge National Laboratory.

coated ThO_2 particles showed no microstructural changes except for a slight loss of integrity of the inner surface regions of the buffer coating, Figs. 28 and 29. Examination of the larger particles from batch GA-323, pyrolytic-carbon-coated $(\text{Th,U})\text{O}_2$, disclosed no unusual microstructural features except for a slight loss in integrity of the inner buffer coating and the presence of a few "shiny" crystals in the fuel-coating gap, see Fig. 30.

In the lower compartment numerous small cracks were found in the binder material of the bonded particles and these cracks extended into the peripheral regions of the coatings. Examination of an unirradiated bonded control disclosed similar but smaller cracks. In the fuel of the lower region, microcracks were found in the UC_2 particles, batch GA-322. Partial densification of the inner buffer coating and loss in crystalline detail were the only visible changes in the $(\text{Th,U})\text{C}_2$ particles, batch GA-321-2. Typical coated particles are shown in Figs. 31 and 32.

High-Burnup Coated-Particle Irradiation Test No. 1 in ORR Position F-9

A capsule irradiation experiment¹⁶ pertinent to the high-temperature gas-cooled reactor fuel program was designed to test coated-particle fuels to relatively high burnup under conditions expected in reactor service. The first fuel assembly of this design was irradiated in the ORR F-9 facility from July 14, 1965 to July 14, 1966. The objective of this test was to evaluate the fission-product retention and overall stability of coated particles to burnups of 50 at. % heavy metal at particle-surface temperatures between 1100 and 1400°C.

The fuel capsule (Fig. 33) contained two batches of pyrolytic-carbon coated fuel particles supported as monolayers in the central graphite holder. The upper compartment of the holder contained UO_2 particles from batch HB-23 and the lower compartment, General Atomic UC_2 particles from batch GA-320. Results of characterization of these particles are given in Table 12. It should be mentioned that the particles from batch

¹⁶A. W. Longest and J. A. Conlin, Design of an Irradiation Experiment to Test Coated-Particle Fuels to High-Burnup, USAEC Report ORNL-TM-1427, Oak Ridge National Laboratory.

Fig. 28. Appearance of Pyrolytic-Carbon-Coated (U,Th)C₂ Particles (Batch GA-321-2) Loaded in the Upper Portion of the Loop 1-15 Assembly. Etched. 200X. (a) Unirradiated. (b) Irradiated.

Fig. 29. Appearance of Pyrolytic-Carbon-Coated ThO_2 Particles (GA-324) Used in the Upper Section of the Loop 1-15 Assembly. As polished. 200X. (a) Unirradiated. (b) Irradiated.

Fig. 30. Appearance of Typical Pyrolytic-Carbon-Coated (Th,U)O₂ Particles (Batch GA-323) Used in the Upper Section of the Loop 1-15 Assembly. As polished. 200X. (a) Unirradiated. (b) Irradiated.

Fig. 31. Appearance of Pyrolytic-Carbon-Coated UC_2 Particles (Batch GA-322) Used in the Bottom Portion of the Loop 1-15 Assembly. Etched. 200X. (a) Unirradiated. (b) Irradiated.

Fig. 32. Appearance of the Bonded Pyrolytic-Carbon-Coated $(U,Th)C_2$ Particles (Batch GA-321-2) Irradiated in the Lower Section of the Loop 1-15 Assembly. As polished. 200X.

Fig. 33. Cross Section of High-Burnup Experiment Fuel Capsule.

Table 12. Results of Characterization of Pyrolytic-Carbon-Coated Fuel Particles

	Batch Designation	
	HB-23	GA-320
Fuel material	UO ₂	UC ₂
Uranium content, wt %	17.8	20.4
Thorium content, wt %	0	0
Thorium-to-uranium ratio	0	0
Enrichment in ²³⁵ U, at. %	93	93
Particle dimensions, ^c μ		
Total diameter		
Average	407	379
Standard deviation	14	25
Minimum	385	353
Core diameter		
Average	148	125
Standard deviation	12	9
Minimum	126	111
Coating thickness		
Average	129	127
Standard deviation	7	11
Minimum	118	112
Pour density, g/cm ³	1.52	1.20
Tapped density, g/cm ³	1.54	1.21
Average coated particle weight, g	0.82×10^{-4}	0.51×10^{-4}
Density of coated particle, ^d g/cm ³	2.43	(a)
Coating density, ^e g/cm ³	2.03	(a)
Coating structure	Triplex	Biso ^b
Crushing load, ^f g		
Average	1210	1550
Standard deviation	160	337
Minimum	900	1100
Surface contamination, % of total U		
As received	0.16×10^{-3}	0.08×10^{-3}
After first leach	0.16×10^{-3}	0.08×10^{-3}
After first leach and thermal cycle	0.17×10^{-3}	(a)
After thermal cycle and second leach	0.13×10^{-3}	(a)
Fuel removal, % of total U		
First leach	$<0.01 \times 10^{-3}$	0.15×10^{-3}
Second leach	$<0.02 \times 10^{-3}$	(a)

^aNot measured.^bTwo-layer coating composed of a porous buffer layer and an outer layer of isotropic pyrolytic carbon.^cMeasured on 25 randomly selected particles from a microradiograph of a riffled sample.^dMeasured with helium pycnometer.^eMeasured by oxidizing coated particles and calculating from the pycnometer density of the coated particles^fBased on crushing of 25 coated particles.

GA-320, which were used in the experiment, received no leach treatment at the request of GA, while those from batch HB-23 did receive leach treatment prior to irradiation.

The graphite structure supporting the fuel particles was continuously swept during irradiation with an inert gas mixture which was sampled periodically for fission-gas release measurements. Typical fission-gas release rates are reported in Table 13 in terms of the ratio of release rate to estimated birth rate (R/B). Also reported in Table 13 are the operating temperature and estimated fuel burnup at the time of each sample. Note that the fission-gas release rate increased by two orders of magnitude in going from 6 to 14 at. % heavy metal burnup and remained at the higher level for the remainder of the irradiation.

The measured fuel temperature (particle surface) during the early part of the irradiation was kept slightly below the 1400°C maximum design temperature because the lower batch of particles was uninstrumented and, because of flux gradients in the reactor, may have been at a slightly higher temperature than that measured in the upper region. The last measured temperature before the central thermocouple failed on April 1, 1966 was 1100°C. We believe the fuel temperature was close to the 1100°C minimum design temperature at the end of the irradiation because the high neutron flux exposure could have caused a considerable downward drift in the W-5% Re vs W-26% Re thermocouple reading.

Postirradiation evaluation of this experiment is now in progress. Metallographic examination of 72 of the coated UO₂ particles (batch HB-23) disclosed no failures nor was there any evidence of potential failure, except for one unusual particle where the inner coating had been severely damaged. The only significant change was noted in the UO₂ fuel cores. The majority of the UO₂ particles contained shiny metallic inclusions and porosity normally observed after appreciable burnup; about 10% of the particles were essentially pore free. No crystalline detail could be resolved in the UO₂. Only slight damage was observed in the inner surface regions of the buffer coatings. Typical coated particles from batch HB-23 are shown in Fig. 34.

Examination of 163 coated UC₂ particles (batch GA-320) from the lower portion of the capsule disclosed no failures. However, in about

Table 13. Fission-Gas Release Measurements from the High-Burnup
Coated-Particle Irradiation Test No. 1

Date	Estimated ^a Fuel Burnup (at. % H.M.)	Sweep Gas	Measured Fuel Temp. (°C)	Ratio of Fission-Gas Release Rate to Birth Rate, R/B				
				^{85m} Kr x 10 ⁻⁶	⁸⁷ Kr x 10 ⁻⁶	⁸⁸ Kr x 10 ⁻⁶	¹³³ Xe x 10 ⁻⁶	¹³⁵ Xe x 10 ⁻⁶
7-16-65	.9	Neon	1282	-	1.52	2.97	-	-
7-19-65	1.8	Neon	1285	1.69	.65	.33	.76	.19
7-23-65	3.0	Neon	1291	1.72	.74	1.01	.82	.18
8-5-65	5.8	Neon	1290	1.78	.65	1.34	.86	.16
8-18-65	9.8	Neon	1321	5.41	2.21	6.29	3.31	.89
8-31-65	11.3	Neon	1199	28.6	33.4	.44	-	-
9-7-65	13.4	Helium-Argon	1360	237.0	-	-	102.0	62.5
9-16-65	15.8	Neon	1174	97.0	31.1	104.0	51.4	-
9-22-65	17.5	Helium-Argon	1371	12.0	-	-	291.0	5.5
10-6-65	21.1	Argon	1343	64.8	-	-	443.0	89.1
10-26-65	24.9	Argon	1243	379.0	-	-	194.0	42.0
11-11-65	27.5	Argon	1246	206.0	-	-	103.0	1.6
12-28-65	32.9	Argon	1252	459.0	-	-	462.0	76.1
1-12-66	35.9	Argon	1238	569.0	-	-	1206.0	129.0
2-3-66	39.6	Argon	1168	408.0	-	-	602.0	80.4
3-21-66	44.3	Argon	1107	345.0	-	-	531.0	64.8
4-29-66	47.8	Argon	--	176.0	-	-	549.0	53.0
5-30-66	51.3	Argon	--	263.0	-	-	207.0	67.7
7-14-66	54.0	Argon	--	109.0	-	-	83.6	72.7

^aThese burnups are estimated from measured flux and calculated flux depression. The final burnup as determined from isotropic analysis is 47-50% heavy metal.

Fig. 34. Appearance of Typical Pyrolytic-Carbon-Coated Sol-Gel UO_2 Particles from Batch OR-HB-23. (a) Unirradiated. (b) Irradiated. Original reduced 8.5%.

5% of the particles localized attack had occurred at the inner surface of the buffer coating and extended into the isotropic layer. Densification of the buffer coatings was observed in all of these coated particles. The attack mentioned above was noted only where the buffer coating had densified enough to cause separation between the buffer and isotropic layers. Slight pitting of the outer surface was also noted in a few of the coatings. As expected, no crystalline detail could be resolved in the fuel after irradiation. Typical coated particles from batch GA-320 are shown in Fig. 35.

Dimensional changes of the graphite, pyrolytic carbon, and ZrC components caused by the irradiation are given in Table 14. We found that the porous carbon insulators (or end spacers) exhibited the greatest shrinkages, followed by the pyrolytic carbon insulator sleeve. The ZrC included as a gamma heat source experienced a small growth in outside diameter. Note that the shrinkage of the outer graphite thermocouple sleeve, which operated in the temperature range of 760-940°C, was much less than that of the inner graphite sleeves which operated at higher temperatures.

The remaining postirradiation work planned for this experiment includes additional studies on the coated particles and determinations of the actual fuel burnup and neutron flux exposure. A tentative fuel burnup of 47-50 at. % heavy metal has been obtained from a uranium isotopic analysis of the GA-320 particles.

Fig. 35. Appearance of Typical Pyrolytic-Carbon-Coated Particles from Batch GA-320. (a) Unirradiated. (b) Irradiated. As polished.

Table 14. Dimensional Changes of Graphite and ZrC Components Irradiated In
Eigh-Burnup Coated-Particle Experiment No. 1
(Estimated fast-neutron ($E > 0.18$ Mev) exposure: 1.7×10^{21} neutrons/cm²)

	Estimated Operating Temp. Range (°C)	Outside Diameter Change (%)	Inside Diameter Change (%)	Length Change (%)	Wall Thickness Change (%)
Graphite Components					
Thermocouple sleeve of Speer 780S graphite	760-940	-0.1	-0.2	-0.2	(a)
Pyrolytic-carbon insulator sleeve	875-1125	-1.67	-2.4	-2.69	+7.5
Inner sleeve of Speer 780S graphite	975-1275	-0.7	-0.8	-0.6	---
Top end spacer of porous carbon (NC Grade 60)	~675	-1.8 -2.0 ^b	---	-0.9	---
Bottom end spacer of porous carbon (NC Grade 60)	~700	-2.7 -3.2 ^b	---	-2.1	---
Upper fuel holder of CHN graphite ^c	1050-1370	-0.7	---	-1.0	---
Lower fuel holder of CHN graphite ^c	1050-1400	-0.7	---	-1.0	---
ZrC Cylinders					
No. 5	1000-1340	+0.3	---	<+0.1	---
No. 6	---	+0.2	---	---	---
No. 7	---	+0.2	---	+0.1	---

^aNot detectable.

^bMinor outside diameter.

^cExperimental graphite obtained from GA.

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