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FISSION-GAS RELEASE AND IRRADIATION DAMAGE TO AVR PYROLYTIC-CARBON COATED THORIUM-URANIUM CARBIDE PARTICLES

P. E. Reagan

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REACTOR CHEMISTRY DIVISION

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JANUARY 1967

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ABSTRACT

Pyrolytic carbon coated thorium-uranium carbide particles, prepared commercially for use in the German AVR, were irradiated to 10 atom percent heavy metal burnup at 1300°C in the ORR. These particles were a blend of several batches of duplex coated particles that were selected as representative of the coated particles to be used in the first AVR fuel loading. The fractional fission-gas release for ^{88}Kr was 5×10^{-6} at the beginning of the test and increased with burnup to 4×10^{-5}. No bursts of fission gas were released during the test and no broken coatings were found on postirradiation examination. Metallographic examination revealed some damage to the inner coating, but nothing that indicated potential failure of the coated particles.

INTRODUCTION

The AVR (Arbeitsgemeinschaft Versuchs Reactor), nearing completion at Julich, West Germany, will be a 46 Mw(th) pebble-bed, gas-cooled reactor. The first fuel loading for this reactor will be pyrolytic carbon coated thorium-uranium carbide particles contained in 6-cm-diameter graphite spheres.[1,2] A capsule containing a sample of these particles was irradiated in the B9 facility in the Oak Ridge Research Reactor (ORR), to measure the fission-gas release rates, and to determine what damage was done by irradiation at high temperature. This report describes the physical properties, fission-gas release rates, and postirradiation examination of these particles. The work was conducted under the terms of a memorandum of understanding between the German project, Euratom and the USAEC.

TEST SPECIMEN

The coated particles selected for this test were a blend from several batches of coated particles. The particles were coated by the Carbon Products Division of the Union Carbide Corporation.[3] These particles were designated FV-1A by the manufacturer, as Batch NCC-222 by Oak Ridge National Laboratory, and were irradiated in experiment B9-28. The average coated particle diameter was 437 μ. The total coating thickness was 112 μ. Other physical properties and preirradiation evaluation data are given in Table 1, including coating surface contamination. These particles had been subjected to production-type inspection and were certified by the manufacturer to be typical of production-run coated particles. Therefore, preirradiation inspection at ORNL was limited to metallographic examination and microradiography. No unusual particles were removed. A radiograph of typical particles from Batch NCC-222 is shown in Fig. 1. While the fuel particles were nearly round, the coatings varied from round to polyhedral.

TEST METHOD

About 21,000 coated particles, containing 0.12 grams ^{235}U were placed in a graphite holder. These particles occupied an annulus between concentric graphite cylinders as shown in Fig. 2. This graphite holder was mounted in an Inconel capsule about 1 inch in diameter and 2-3/4 inches long. The holder was thermally insulated from the Inconel capsule walls by a 10-mil layer of pyrolytic carbon sheet and a layer of aluminum oxide-silicon oxide wool. The capsule was inserted in the air-cooled B9 facility tube in the ORR.[4] An exploded view of the in-pile capsule assembly is shown in Fig. 3 and a cut-away view of the ORR showing the air-cooled B9 facility tube, inserted in the reactor lattice, is shown in Fig. 4. The temperature of the particles was monitored by five thermocouples of tungsten, 5 percent rhenium vs tungsten, 26 percent rhenium located in the graphite holder. One of these thermocouples was in the center of the inner graphite cylinder surrounded by the coated particles and was considered to be at the maximum coated particle temperature. The temperature of the Inconel capsule wall was monitored by three thermocouples of platinum vs platinum 10 percent rhodium. After

Table 1. Physical Properties of Batch NCC-222 Coated Particles

Type of coating	Duplex
U content, wt %	5.55
Th content, wt %	25.4
Th/U ratio	4.59
Pour density, g/cm^3	1.66
Tapped density, g/cm^3	1.72
Fuel Material	$(Th,U)C_2$
^{235}U enrichment, at. %	93
Particle dimensions,[a] μ	
Total coated particle	
Average	437
Standard deviation	52
Minimum	341
$(Th,U)C_2$ particle	
Average	214
Standard deviation	37
Minimum	148
Coating Thickness	
Average	112
Standard deviation	13
Minimum	86
Average coated particle weight, g	1.15×10^{-4}
Density of coated particle,[b] g/cm^3	2.67
Coating density,[c] g/cm^3	1.93
Crushing load,[d] g	
Average	1778
Standard deviation	700
Minimum	840
Surface contamination, % of total U	
As received	8.7×10^{-3}
After first leach	9.8×10^{-3}
After first leach and thermal cycle	14.1×10^{-3}
After first leach, thermal cycle, and second leach	11.9×10^{-3}
Fuel removal, % of total U	
First leach	2.3×10^{-3}
Second leach	0.5×10^{-3}

[a]Measured on 25 randomly selected particles from a microradiograph of a riffled sample.

[b]Measured with helium pycnometer.

[c]Determined by burnoff method.

[d]Twenty-five particles were crushed from each lot.

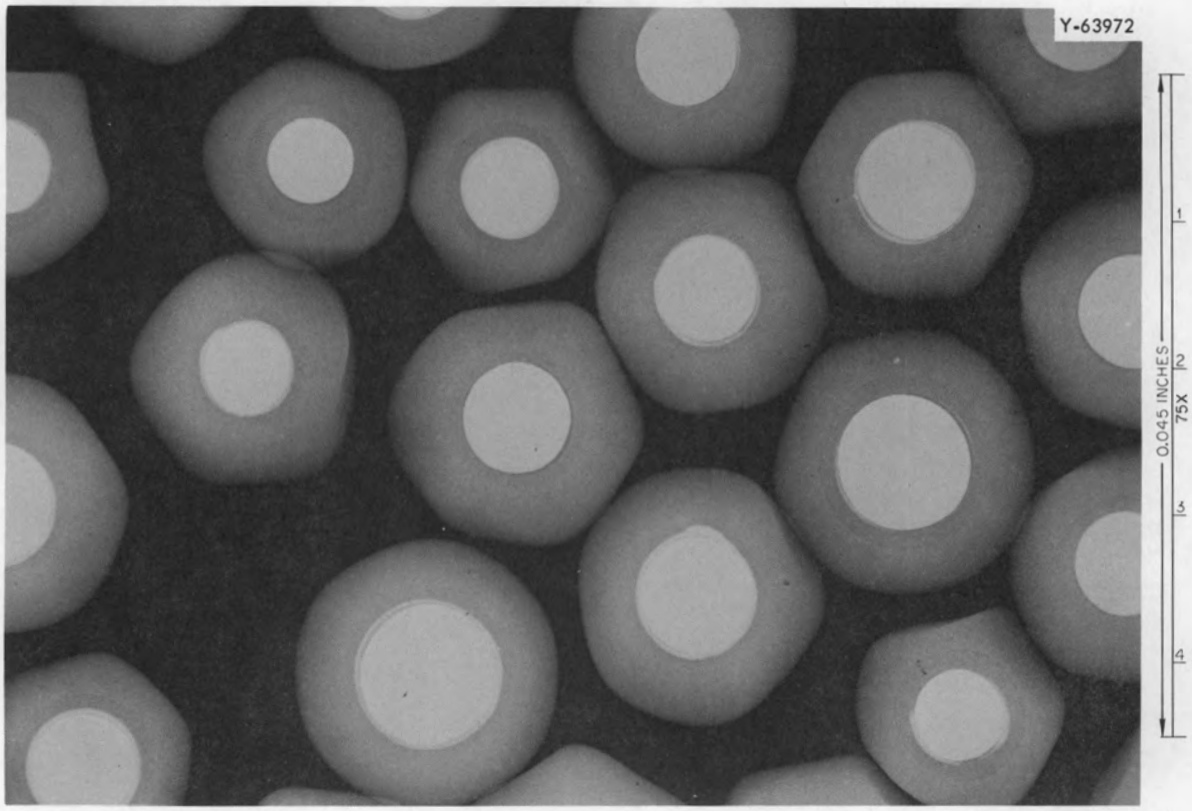

Fig. 1. Typical Particles from Batch NCC-222 as Inspected by Radiography.

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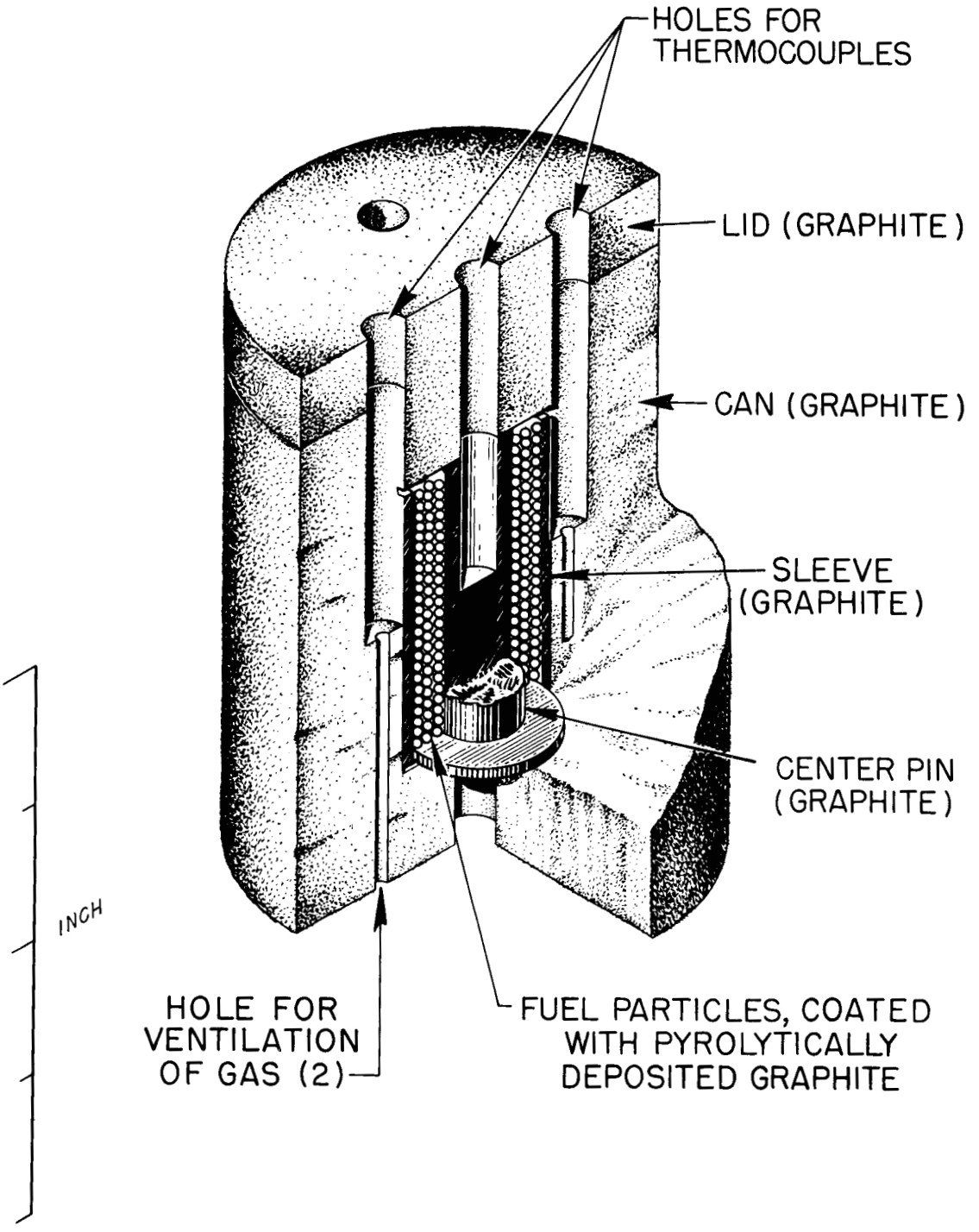

Fig. 2. Coated Particle Holder with Thermocouple Wells.

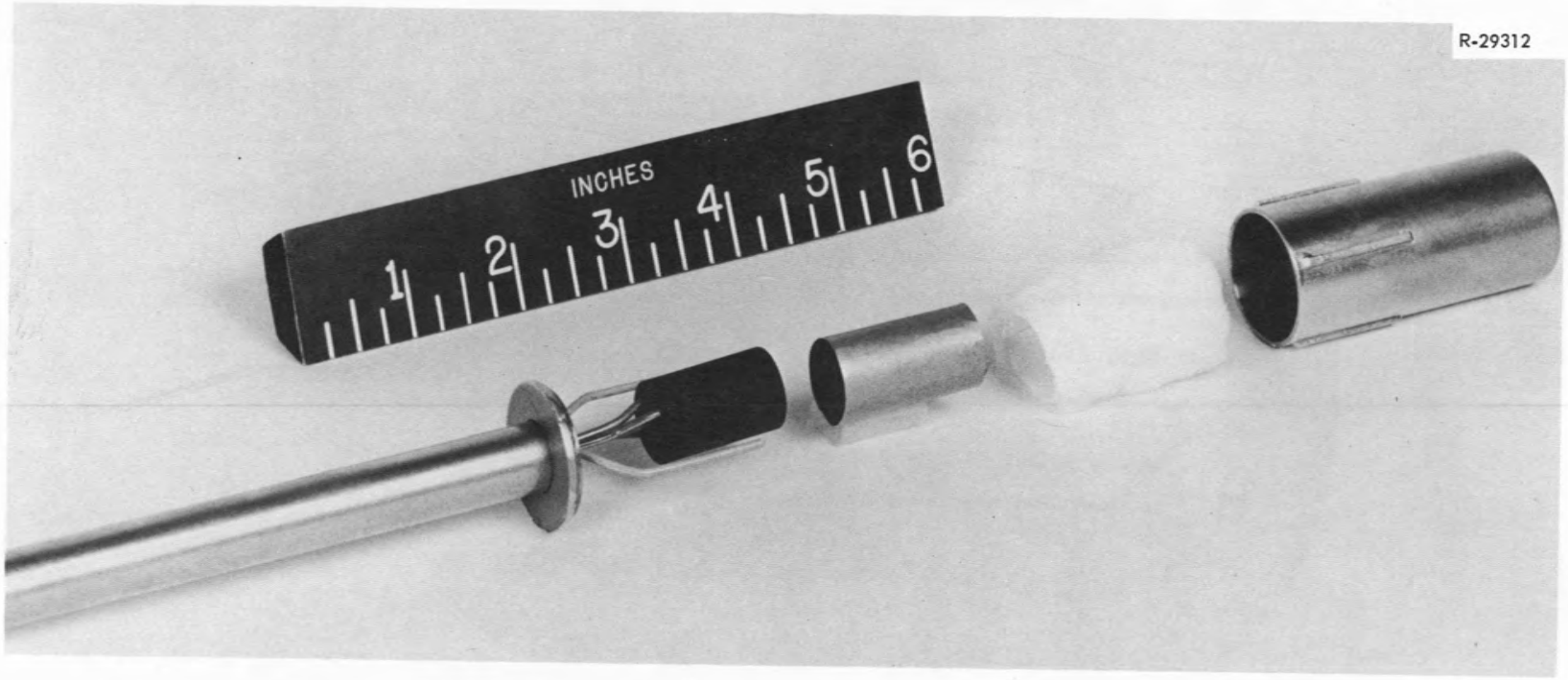

Fig. 3. Exploded View of the In-Pile Capsule.

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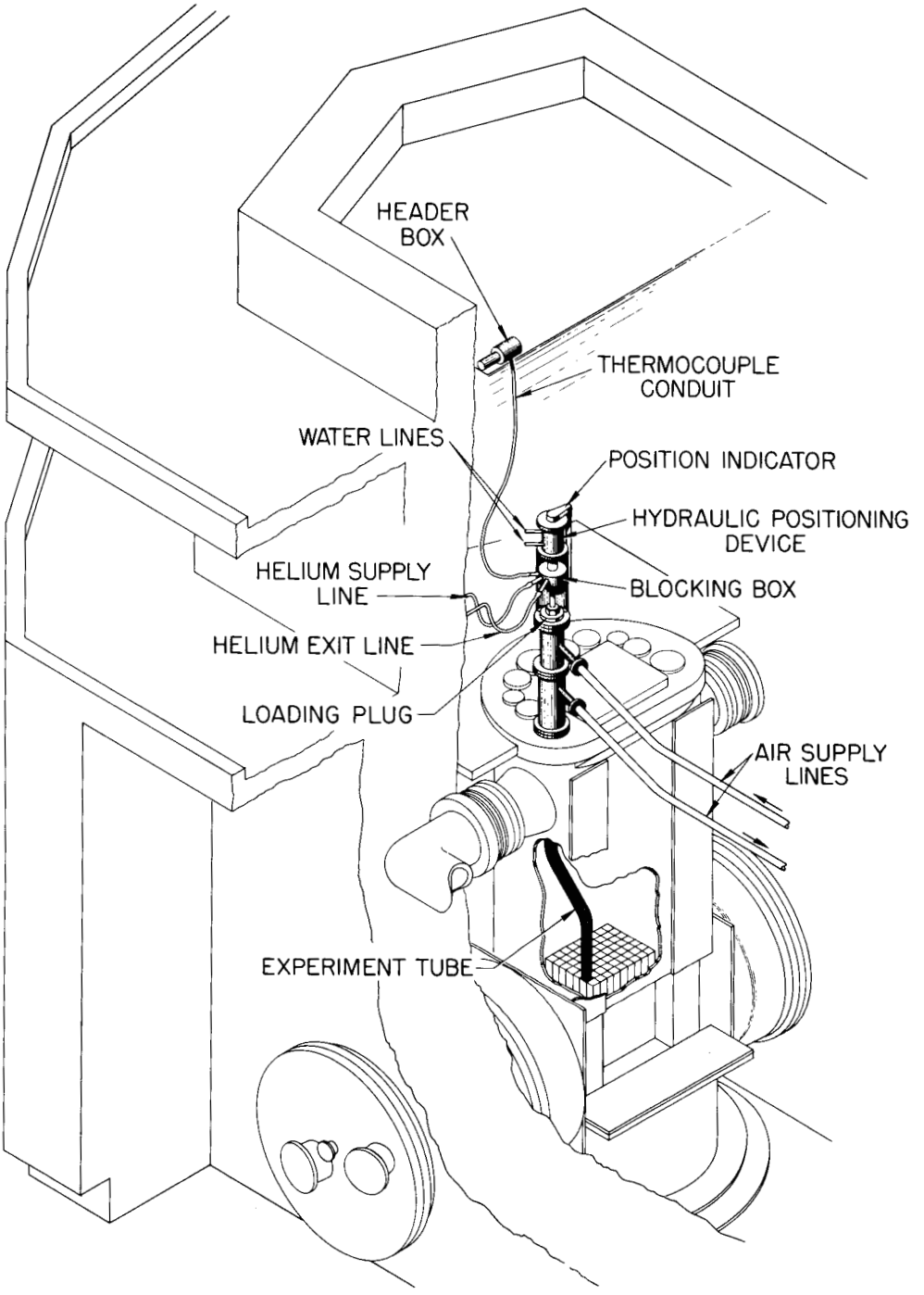

Fig. 4. Fission Gas Release Experiment in the ORR.

the capsule was weld-sealed, the exact location of the thermocouples with respect to the coated particles was checked by radiography. Figures 3 and 5 show the location of the eight thermocouples within the capsule, with respect to the capsule wall and the coated particles. A low flow of helium (about 0.5 cc/sec) was passed over the particles during irradiation to sweep fission gases to a sampling station, where gas samples were collected in evacuated bottles and analyzed with a gamma-ray spectrometer. The bottles were filled through a hypodermic needle attached to the exit helium line, and were discarded after use to prevent cross-contamination. The release of gas, by the coated particles, is expressed as a fractional release, R/B, which is the ratio of the release rate to the birth rate. The particle temperature was controlled by adjusting the capsule position with respect to the reactor lattice. Figure 6 is a block flow diagram, describing the helium flow path, the cooling-air flow path, and the capsule positioning arrangement. The fuel burnup was calculated, based on the neutron flux which was determined during the test by passing helium containing one percent argon through the capsule and measuring the argon-41 activation.[5]

IRRADIATION TEST

The capsule assembly was installed in the reactor during the Aug. 2, 1965 shutdown but remained in a withdrawn position until the reactor was brought up to full power. It was then inserted stepwise into the reactor lattice until the coated particle temperature reached 1300°C (at an elevation of 1-1/2 inches above the reactor centerline). The thermal neutron flux measured at this elevation was 7.4×10^{13} n/cm^2·sec. As the test progressed, the capsule was kept at a constant temperature by gradually inserting it into a higher flux to compensate for capsule fuel loss by burnup, thus maintaining a constant burnup rate. Twice during the test, the temperature drifted up to near 1400°C for a few hours. Because of the high thorium-to-uranium ratio of the particles (4.59 to 1), it required almost 4000 hours of irradiation time to reach 10.1 atom percent heavy metal burnup. The coated particles were subjected to 17 thermal cycles as a result of regular reactor shutdowns. The thermal neutron dose was slightly more than 1×10^{21} n/cm^2·sec, and the fast dose (above 0.2 Mev) was 7×10^{20} n/cm^2·sec.

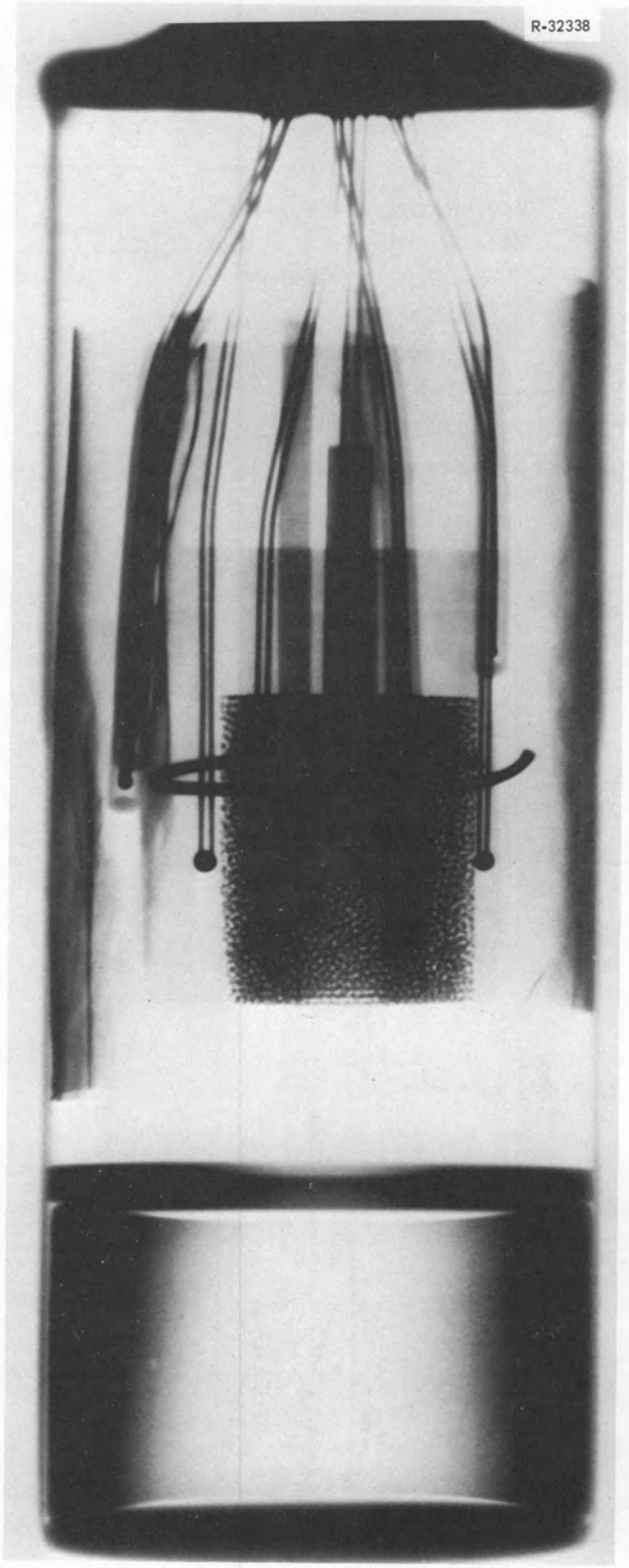

Fig. 5. Radiograph of Thermocouple Locations in Capsule B9-28. Mag. 4X. Reduced 20%.

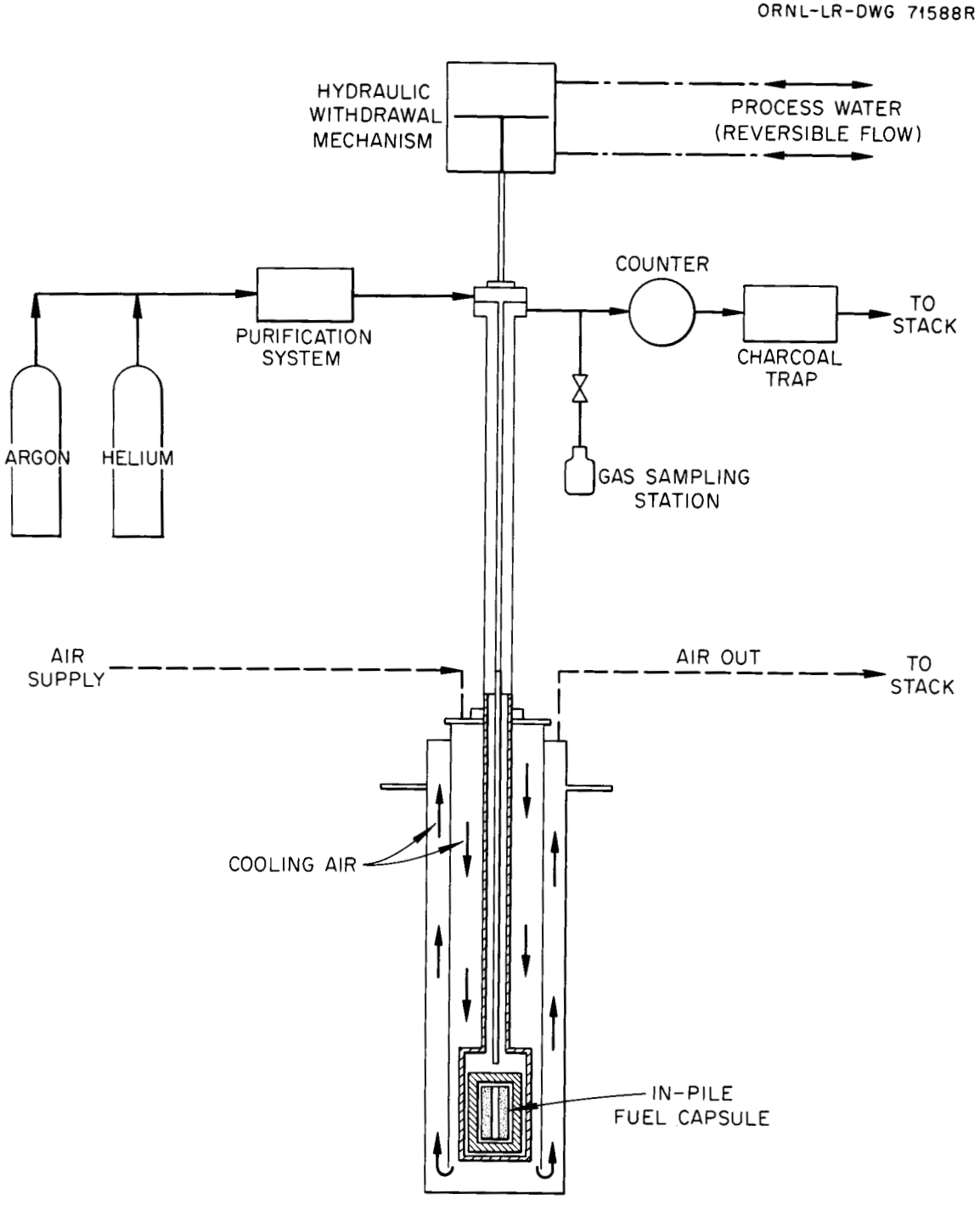

Fig. 6. Flow Diagram of Capsule B9-28.

The fractional fission-gas release, R/B, was below 5×10^{-6} for ^{88}Kr at the beginning of the test and increased continuously with burnup to approximately 4×10^{-5} at the end of the test, as shown in Fig. 7. The fractional release, for the five isotopes measured, increased about the same amount, and when plotted against their half-lives, as shown in Fig. 8; the slope of the lines remained nearly constant during the test. The offsets in the lines between the Xe and Kr are characteristic of fission-gas release observed from previous experiments with unbroken coated particles. There were no bursts of fission gas released during the test.

POSTIRRADIATION EXAMINATION

When the particles were examined at 30X, no broken or cracked coatings were found. The surface of the particles was smooth and showed no signs of damage (shown in Fig. 9). Metallographic examination of 106 particles from capsule B9-28 revealed no failed coatings or signs of potential failure. Typical particles before and after irradiation are shown in Fig. 10. Some damage was evident at the fuel-coating interface and was accompanied by movement of fuel into the damaged regions. Except for a loss in crystalline detail and the presence of fine porosity in the carbide particles, no other significant microstructural changes were observed.

DISCUSSION

The AVR (NCC-222) particles were manufactured after several irradiation tests had demonstrated that the duplex-type pyrolytic carbon coated thorium-uranium carbide particles of this type could be expected to operate well, under AVR conditions.[6] The ratio of the coating thickness to core diameter was 0.52, which is a good margin above the 0.30 to 0.40 rate well, under AVR conditions.[6,7,8,9] Some of the coated particles had small defects in the inner layer of pyrolytic carbon coating before irradiation. Examples of this are shown in the unirradiated particles at the bottom and lower righthand corner of Fig. 10a. Delamination and vertical cracks were found at the particle-coating interface. Irradiation damage appears to be limited to the inner coating, and in no case was it found to go beyond. Also each coated particle examined seems to

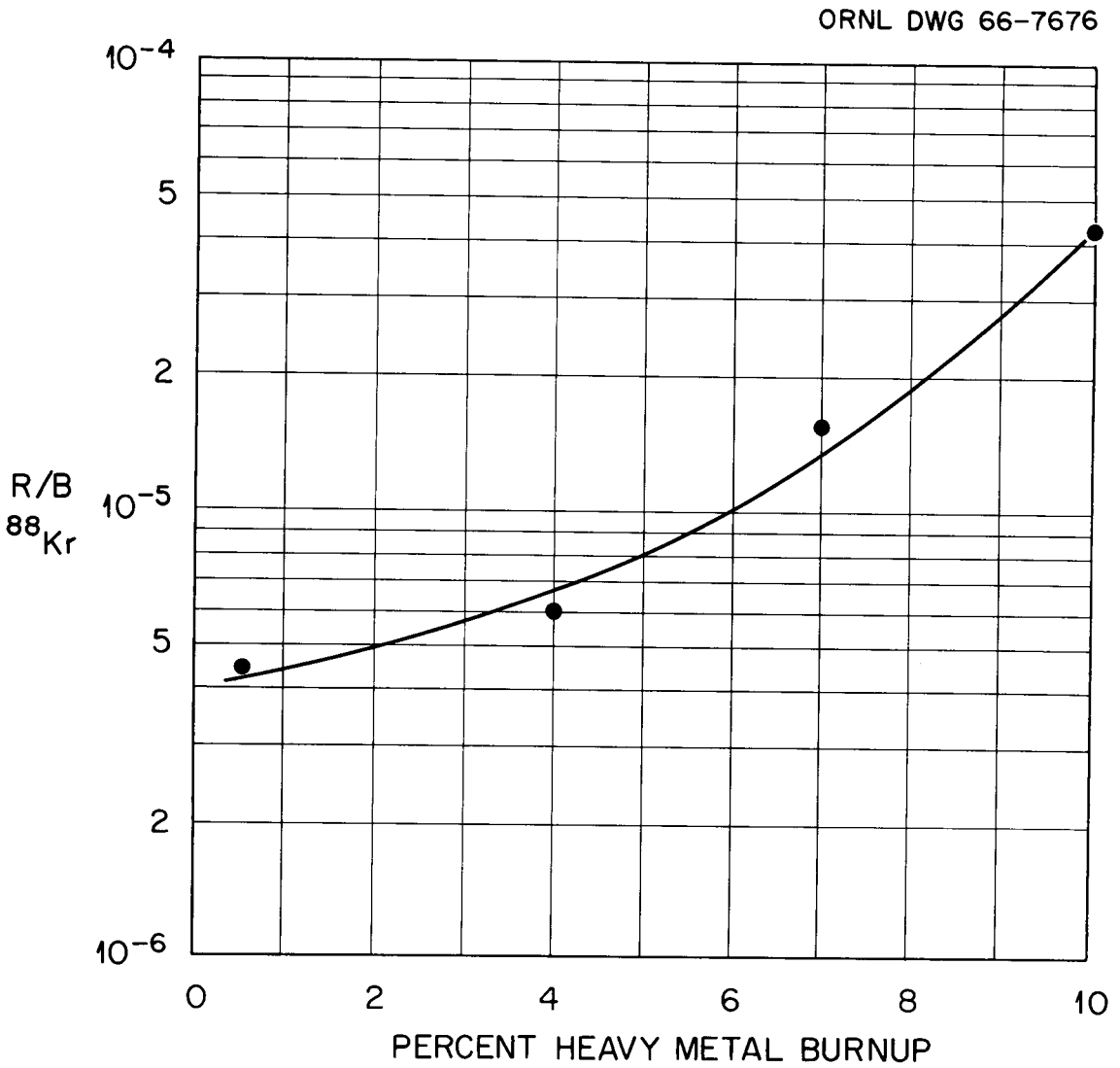

Fig. 7. Krypton-88 Fractional Release Increase with Burnup.

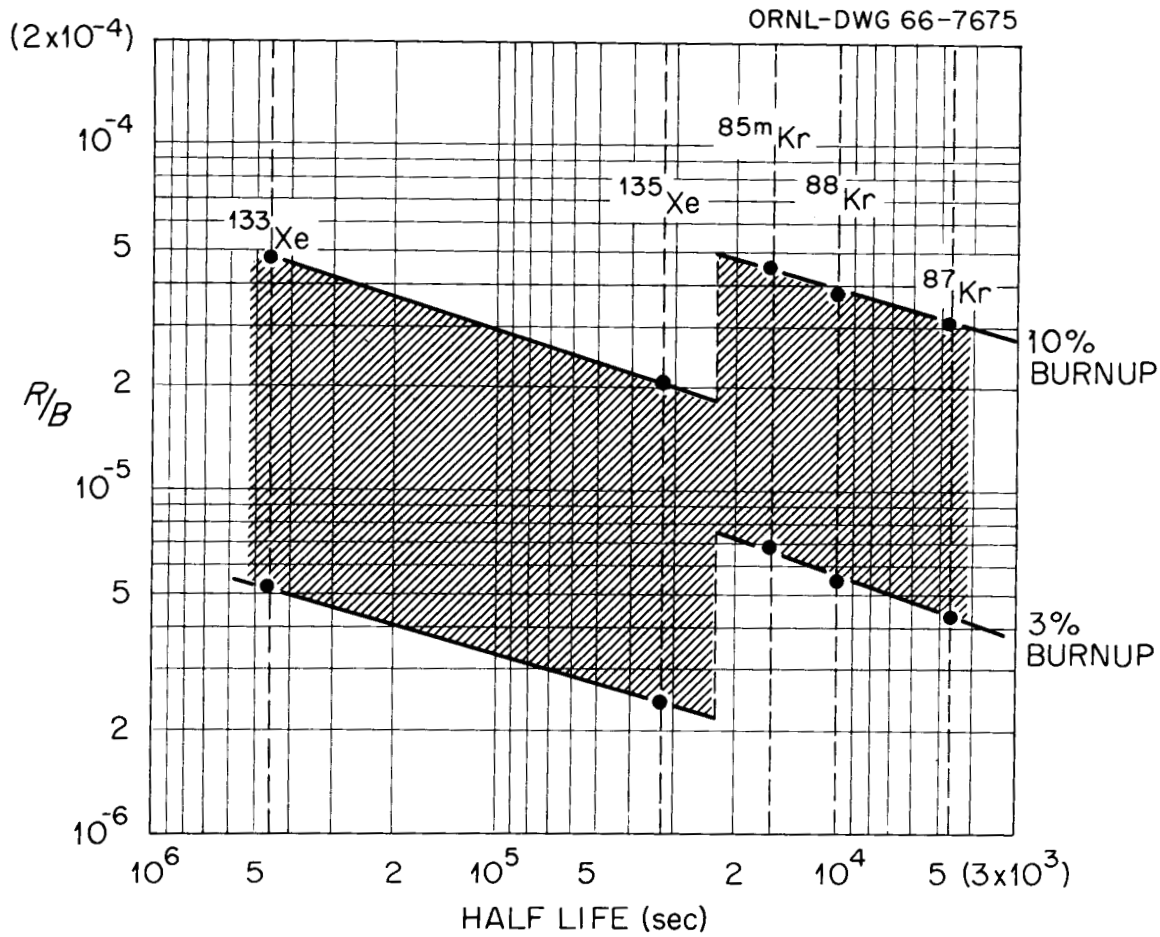

Fig. 8. Average Fractional Fission Gas Release at 3% Burnup and at 10% Burnup.

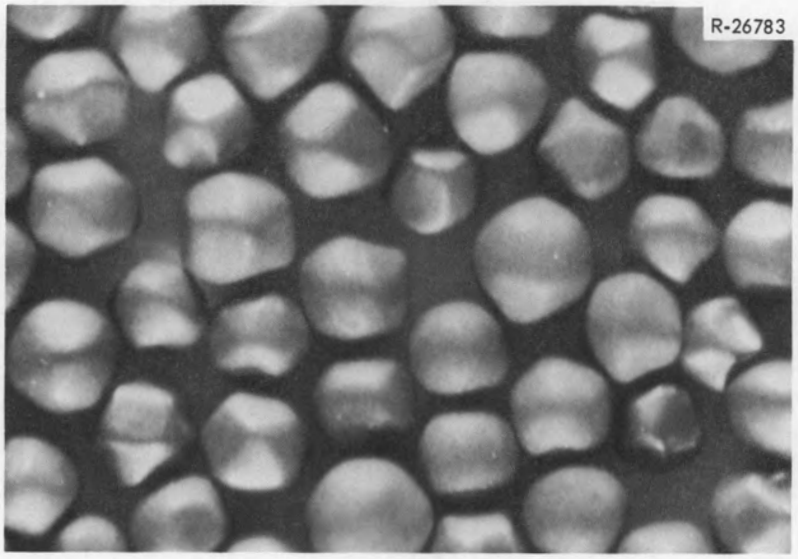

Fig. 9. Pyrolytic Carbon Coated Thorium-Uranium Particles from Batch NCC-222. Mag. 30X. Irradiated to 10.1% heavy metal burnup at 1300°C in capsule B9-28.

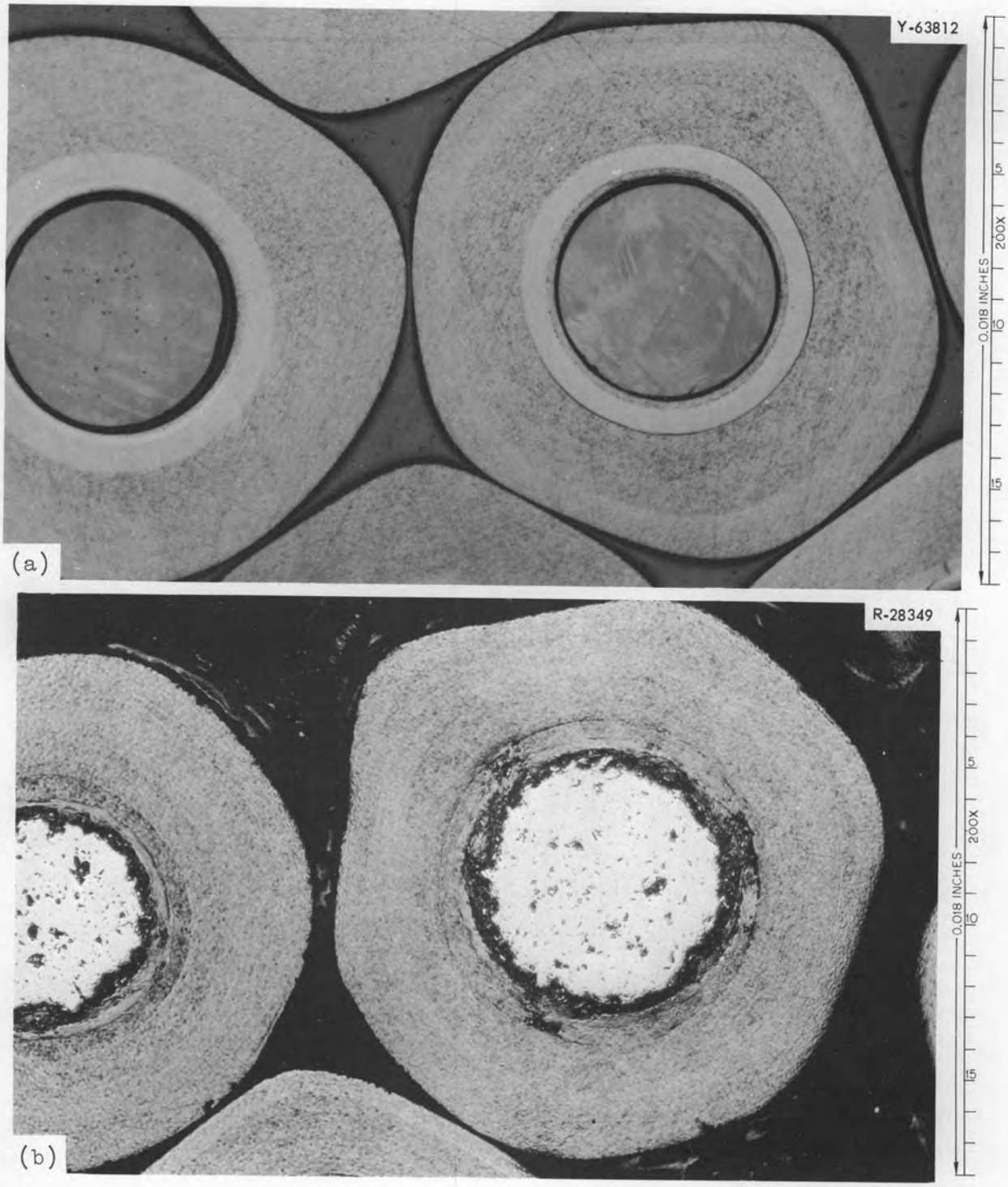

Fig. 10. Pyrolytic Carbon Coated Thorium-Uranium Carbide Particles from Batch NCC-222. Mag. 200X, as polished. (a) Unirradiated, (b) Irradiated to 10.1% heavy metal burnup at 1300°C in capsule B9-28.

have received about the same amount of damage at the core-coating interface.

The fission gas released by the particles came primarily from contamination in the coating. Based on the alpha count (after leaching) there was 1.42×10^{-5} grams of ^{235}U distributed in the coatings within the alpha range of the surface. This amount of fuel would yield about 6×10^{7} atoms/second of ^{88}Kr during irradiation. The measured release rate of ^{88}Kr was about 0.3×10^{7} atoms/second at the beginning of the test and increased to about 3×10^{7} atoms/second by the end of the test.

Throughout the irradiation period, the coated particles were subjected to thermal shock each time the reactor shut down. The temperature changes caused by both shutdown and startup are shown in Fig. 11. The greatest rate of change in temperature occurred immediately after a change in reactor power. The temperature decreased at about 400°C/min, maximum, after shutdown, and increased at about 340°C/min, maximum, during startup. Seventeen startup-shutdown cycles seemed to have had no adverse effect on these coated particles.

The performance of these coated particles was well within the AVR design criteria up to 10% burnup at 1300°C.[10,11]

ACKNOWLEDGEMENTS

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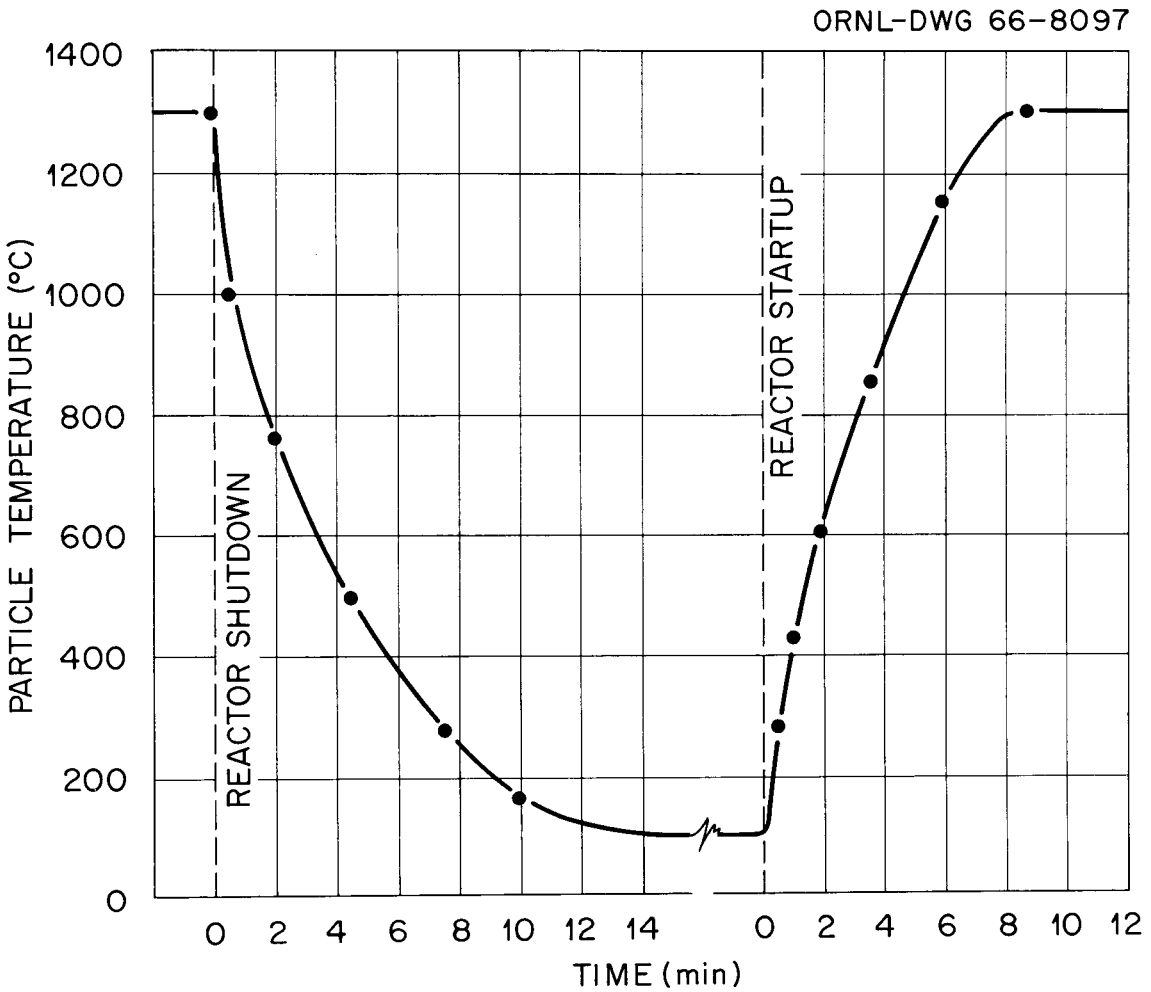

Fig. 11. Coated Particle Temperature after Reactor Startup and after Reactor Shutdown.

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