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Coated Particle Fuel Elements for UHTREX

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Coated Particle Fuel Elements for UHTREX*

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ABSTRACT

The Ultra High Temperature Reactor Experiment (UHTREX) of the Los Alamos Scientific Laboratory utilizes a helium-cooled, graphite-moderated reactor to be operated at a thermal power of 3 MW. Fuel elements are required for the start-up and early operation that will permit check-out of this high-temperature (2400°F), high-pressure (500 psi) reactor and evaluation of its operational characteristics with minimum contamination of the entire reactor system.

The initial fuel elements for UHTREX contain coated UC_2 particles in a graphite matrix. The fuel particles have triplex (three-layer) pyrocarbon coatings. The fuel element stock, 1 in. o.d. x 0.5 in. i.d., is fabricated by warm extrusion of a mixture of fuel particles, graphite flour, carbon black (Thermax) and resin binder (Varcum). The extruded stock is successively cured in air to 250°C, baked in vacuum to 800°C, heated in helium to 1700°C and cut into 5.5-in.-long fuel elements. The fuel elements are then leached with 20% HCl in helium at 1600°C to remove uranium exposed by the slight amount of damage to fuel particles that occurs during the mixing and extrusion steps and by the machining operation.

The coated fuel particles and other ingredients are described and details of the equipment and procedures used for UHTREX fuel element fabrication are presented. Properties of the fuel elements are reported.

*This work was performed under the auspices of the U.S. Atomic Energy Commission.

INTRODUCTION

The Ultra High Temperature Reactor Experiment (UHTREX) of the Los Alamos Scientific Laboratory utilizes a helium-cooled, graphite-moderated reactor to be operated at a thermal power of 3 MW. The objectives of UHTREX include: 1) operation of the reactor with simplified fuels, 2) testing of coolant gas cleanup systems for such operation, and 3) generation of very high temperature helium (2400°F).

The reactor, shown in Fig. 1, has a cylindrical graphite core that is surrounded by a graphite and carbon reflector and by low-density carbon insulation, all encased in a spherical steel pressure vessel.¹ The core, which is 70 in. o.d. x 23 in. i.d. x 39 in. high, has 312 fuel channels in the form of 1.1-in.-dia radial holes at 15° intervals in 13 horizontal planes with 3-in. vertical separations. The fuel elements are hollow cylinders of graphite loaded with 93% enriched uranium; the cylinders are 1 in. o.d. x 0.5 in. i.d. x 5.5 in. long. Four fuel elements rest end-to-end in each of the 312 fuel channels, for a total of 1248 elements in a complete core loading. A fuel loader, mounted on the side of the pressure vessel, moves fuel elements into and out of the core. The loader, a vertically aligned series of horizontal, motor-driven rams, operates in conjunction with an indexing mechanism that rotates the core. While the reactor is operating, a particular fuel channel may be loaded by one of the 13 loading rams after the core is indexed to one of the 24 angular positions. As a new fuel element is charged, a spent element is displaced into a slot in the center plug of the core, drops from the reactor, and is mechanically conveyed to a fuel handling cell.

Fuel elements are required for the start-up and early operation of UHTREX that will permit check-out of this high-temperature, high-pressure (500 psi) reactor and evaluation of its operational characteristics with minimum contamination of the system. Unclad graphite fuel elements containing pyrocarbon-coated UC_2 fuel particles were selected for this purpose. Although the details of the long-range fueling plan are not yet firm, some of the coated particle fuel elements may be kept in the reactor to burnups as high as 10%.

This paper describes the coated fuel particles for UHTREX and the fabrication of graphite matrix fuel elements containing them. More-detailed descriptions are given in reference 2.

PYROCARBON-COATED UC_2 FUEL PARTICLES

GENERAL DESCRIPTION

Pyrocarbon-coated UC_2 fuel particles were selected for the fuel elements because of compatibility with the graphite matrix, good thermal stability at the maximum UHTREX temperature, and the relatively advanced state of knowledge about their fission product retention capabilities. The diameter range of the coated particles was chosen to be 350 to 420 μ , with a nominal 100- μ pyrocarbon coating on 147- to 208- μ UC_2 particles. These dimensions represent a compromise between the requirements of good particle strength and fission product retention and the need to keep the particle size and volume fraction of particles as small as possible to avoid difficulties in the extrusion process used for fuel element fabrication.

SELECTION OF PYROCARBON COATING

Duplex Coating

At the time procurement of coated particles for UHTREX was started, duplex (two-layer) coatings, with an inner layer of laminar pyrocarbon and an outer layer of columnar pyrocarbon, appeared to have the best capability for retention of fission products. Irradiation testing at ORNL had shown such particles to have low rates of release of fission gases and good stability to high burnups.

General Atomic was selected, on the basis of competitive bidding, to supply the duplex-coated particles to LASL specifications. A pre-production quantity of these particles, containing about 5 kg of 93% enriched uranium, was prepared by General Atomic for use by LASL in development of fuel element fabrication procedures. The inner laminar and outer columnar pyrocarbon layers were about 30 and 70 μ thick, respectively. The particles contained 30.4 wt % uranium and had an average crushing strength of 3.0 lb per particle (ranging from 1.3 to 4.2 lb per particle). The external shape was somewhat irregular, due to propagation of irregularities ("dimples") in the UC_2 particles and faceting during coating.

These coated particles were used to establish modifications to an existing LASL extrusion process that were necessary for fabrication of loaded graphite stock with the UHTREX fuel element cross section and containing particles of this size and strength. Particle damage during blending and extrusion was determined by leaching mixtures of particles and dry carbon ingredients, following removal of any binder, in 8M nitric

acid solution at 95°C for 8 hr and determining the amount of uranium leached, for comparison with that leached from the starting coated particles by the same treatment. Blending and extrusion procedures that resulted in less than 0.1% of the uranium becoming exposed due to particle damage were developed using these particles. (The validity of the nitric acid leaching method for evaluating particle damage during blending and extrusion was established by neutron activation and post-irradiation heating experiments with samples of the same materials. The Xe^{133} released in these experiments corresponded to the amount of exposed uranium found by nitric acid leaching.)

Triplex Coating

Later developments in the coated particle field led to the decision to obtain particles with a triplex (three-layer) pyrocarbon coating,³ which had performed well at high temperatures and under thermal cycling conditions in irradiation testing at ORNL.⁴ The triplex coating consists of an inner buffer zone of porous pyrocarbon, a middle layer of isotropic pyrocarbon and an outer layer of granular pyrocarbon. The porous pyrocarbon catches fission recoils and dissipates stresses during irradiation by accommodating swelling of the fuel kernel and contraction of the balance of the coating. The porous layer also furnishes a void volume to minimize fission-gas pressure buildup. The isotropic pyrocarbon is less susceptible than oriented pyrocarbon to anisotropic dimensional changes that may result from fast neutron exposure and lead to cracking. The interface between the middle and outer layers prevents propagation of any cracks that may start in the middle layer.

A pre-production quantity of triplex-coated UC_2 particles, meeting revised LASL specifications and containing about 3 kg of depleted uranium, was obtained from General Atomic. These particles were used to check the applicability of the blending and extrusion procedures developed with the duplex particles. Although these triplex particles were somewhat weaker than the duplex, particle damage during fuel element fabrication was not significantly greater for them.

Finally, a production quantity of triplex-coated UC_2 particles containing 25 kg of 93% enriched uranium was obtained from General Atomic for the UHTREX fuel elements.

CROSSBLENDING, SAMPLING AND TREATMENT OF TRIPLEX COATED PARTICLES

The production quantity of enriched triplex particles was delivered to LASL as 44 individual coating batches. Each batch was examined microscopically, and the particles were found to have good shape and to be very uniform from batch to batch.

The 44 batches were incorporated into one crossblend which was sampled for analyses. The amount of exposed uranium in this crossblend was 0.06%. Although this was about what was expected to be exposed by damage to the particles through the second-extrusion stage in fuel element fabrication, it was desired to start fabrication with clean particles. Consequently, the crossblend was leached, in portions in favorable-geometry equipment, with 8M nitric acid solution at 90°C for 8 hr. The leached portions were washed and dried and were crossblended again into a final lot which had only 0.01% exposed uranium.

PROPERTIES OF TRIPLEX COATED PARTICLES

The enriched triplex-coated UC_2 particles are shown in Fig. 2. Properties of these particles are given in Table 1.

TABLE 1. Properties of Triplex Particles for UHTREX

Property		Value
UC_2 diameter, μ		147 - 208
Carbon content of UC_2 , wt %		9.9
Coating thickness, μ	- buffer layer	27
	- isotropic layer	35
	- granular layer	40
	- total	102
Coating density, g/cc	- buffer layer	1.1
	- isotropic layer	1.39
	- granular layer	1.92
Anisotropy factor of isotropic layer		1.1
Uranium, wt %		36.19
Oxygen, ppm		140
Nitrogen, ppm		340
Hydrogen, ppm		20 - 30
Spectrochemical impurities, ppm	- B	0.2
	- other individual elements	< 25
Particle density, g/cc		2.46
Crushing strength, lb/particle	- range	1.0 - 3.4
	- average	1.7

FABRICATION OF FUEL ELEMENTS

MATERIALS

The uranium-loaded graphite extrusions for the UHTREX fuel element stock were made from a formulation which yielded a high density body. The mix consisted of 85 wt % graphite flour, 15% carbon black (Thermax), triplex-coated UC_2 particles and a partially polymerized furfuryl

alcohol resin as the binder. Each of the individual components was analyzed for impurities, a sample of flour from each drum was radiographed for high density inclusions, and eight drums were screened and crossblended to obtain a large supply of relatively uniform graphite flour. The resin binder was prepared at the vendor's plant under LASL supervision and the drums of resin were stored at 10°C to minimize aging and composition changes. Maleic anhydride was employed as the catalyst for the resin.

PROCEDURES

The graphite flour and carbon black were blended dry in 55-gal drums to obtain a uniform distribution of the carbonaceous powders. The required weights of blended powders and coated particles were dry blended for 5 min in a 16-qt twin-shell blender at which time the binder resin, heated to 45°C, was introduced into the blend through a plastic-coated intensifier bar. The binder was forced into the mixer under low pressure gas and approximately 15 minutes were required to feed all of the binder into the blender. The blend, having the consistency of wet sand, was then loaded into stainless steel pans and covered tightly with aluminum foil to prevent evaporation of the resin. This material could be held for considerable time at or below room temperature without affecting the subsequent operations.

The fuel element stock was extruded in a 150-ton Elmes horizontal extrusion press. A sliding vacuum sleeve was fitted to the ram of the extrusion press so that the chamber and the extrusion charge could be evacuated prior to and during the actual extrusion operation. The

extrusion chamber was electrically heated to 45°C, the desired extrusion temperature. The extrusion die was relatively simple in design.

The tubes were extruded directly into a graphite fixture (62 in. long) located at the exit end of the extrusion die, as shown in Fig. 3. When the fixtures were filled with 34 extrusions, they were placed in a large, electrically heated, circulating air oven and heated through a closely controlled cycle for 63 hr to 250°C. The purpose of this initial heating operation was to polymerize the binder resin and drive off the volatiles due to polymerization. The heating cycle must be controlled closely in order to avoid a build-up of gas pressure within the extruded material which would result in a cracked or spalled element.

The cured extrusions were then transferred to another graphite fixture having holes sized to the cured dimensions of the extrusion. The baking cycle processed the extrusions to 850°C over a 48-hr cycle in a resistance-heated furnace operating at a vacuum of 10 torr or less. The baked extrusions were then transferred to another graphite fixture, having the holes sized to the baked dimensions of the extrusion, and heated to 1700°C. This furnace was induction heated and a dynamic helium atmosphere kept air out of the furnace and prevented the graphite from oxidizing. The 1700°C temperature was above the maximum design temperature of the fuel element in the reactor but well below the temperature at which graphitization of any of the element constituents or migration of the UC_2 into the pyrocarbon particle coating would occur.

The 59-in.-long graphitized extrusion possessed outer and inner

diameter dimensions within specification and did not require machining on these surfaces. Consequently, the only machining operation consisted of parting the extrusions into the 5.5-in.-long fuel element length, putting a 1/8-in. radius on the outside corners, and inscribing numbers on the outside cylindrical surface.

Some coated particles were damaged during blending and extrusion and about ten times as many were cut during machining of the ends and inscribing the numbers. Each fuel element was leached with 20% HCl in helium at 1600°C for 3 hr to remove the exposed uranium.

The uranium loading in the fuel elements was determined by gamma counting techniques utilizing the gamma rays emitted from U^{235} . The production inspection of the elements was accomplished with a special gamma scanner constructed for this project. The scanner was programmed to perform three counting periods of 100 seconds each for each of the fuel elements. With automatic operation of the scanner, standardization was accomplished about every 30 min. The counting standards were selected for each nominal load range. Portions of these standards were chemically analyzed for total uranium content.

The completed fuel elements were then subjected to a complete dimensional inspection. The data from each fuel element was recorded for comparison with dimensions of the same element after irradiation.

DISCUSSION

COATED PARTICLE DAMAGE

Table 2 lists typical results obtained from evaluation of coated particle damage by leaching fabrication samples with nitric acid solution, following removal of any binder.

TABLE 2. Typical Coated Particle Damage During Blending and Extrusion

<u>Sample Description</u>	<u>% Total U Leached</u>
Initial Coated Particles	0.01
As Blended	0.07
2nd Extrusion	0.08

Neutron activation and post-irradiation heating to 1200°C for 8 hr resulted in Xe¹³³ releases of 0.0026% and 0.0024% for two HCl-leached fuel elements containing triplex coated particles. These results compare with prior results ranging from 0.0025 to 0.0068% for four HCl-leached fuel elements fabricated from duplex coated particles and indicate the effectiveness of the high temperature leaching with HCl-helium for removing exposed uranium.

FUEL ELEMENT DATA

TABLE 3. Uranium Loading and Carbon Density

<u>Uranium Loading</u>			<u>Average C Density,^b g/cc</u>
<u>mg/cc</u>	<u>g/element</u>	<u>Obtained,^a g/element</u>	
68	3.46	3.55	1.823
79	4.05	4.15	1.820
93	4.74	4.83	1.807
108	5.55	5.70	1.804
127	6.49	6.60	1.796

^aFrom a 1% sample

^bFrom a 10% sample

TABLE 4. Dimensions

Specification	Fuel Elements ^a		
	Max.	Min.	Average
O.D. 0.993 in. $\pm$ 0.010 in.	0.989 in.	0.985 in.	0.988 in.
I.D. 0.497 in. $\pm$ 0.005 in.	0.496 in.	0.493 in.	0.494 in.

^aFrom a 10% sample

TABLE 5. Strength

U Loading, g/element	Flexural Strength, psi	Compressive Strength, psi
3.46	5760	14,200
4.05	----	15,300
4.74	4810	12,500
5.55	----	13,400
6.49	4480	14,000

TABLE 6. Permeability

Type of Graphite	Pressure for Beginning of Leakage, psig	Leak Rate, ^a l/min.
CS 312 ^b	1	4.9
CS 312 ^c	1	9.5
AUC ^d	1	7.0
AUC ^e	1	8.4
UHTREX Element	18	0.01

^aMeasured from i.d. to o.d. at 25 psig of helium pressure.

^bMachined from outside of 4-in. round.

^cMachined from inside of 4-in. round.

^dWith as extruded surface on o.d.

^eWith 0.010 in. of as-extruded surface removed.

TABLE 7. Impurities

<u>Element</u>	<u>Content, ppm^{a,b}</u>
Li	< 1
Be	< 1
B	< 3
Na	20 - 40
Mg	30 - 50
Al	10 - 50
Si	200 - 300
Ca	40 - 100
Ti	50
V	20
Cr	5 - 40
Mn	< 5
Fe	80 - 200
Co	< 30
Ni	< 30
Cu	10
Zr	< 30
Cb	< 30
Mo	< 10
Cd	< 10
In	< 30
Ba	10
Sm	< 25
Eu	< 25
Gd	< 25
Dy	< 25
W	< 300
Pb	< 10

^aBy semiquantitative analysis of one sample from each loading.

^bContents of < were below the limits of detection of the method used.

SUMMARY

The request for a UHTREX fuel element which would operate at temperatures up to 1600°C in a helium atmosphere and provide reduced radioactive contamination to the reactor system during start-up operations was fulfilled.

Coated fuel particles with a triplex (three-layer) pyrocarbon coating on a UC_2 core were leached in HNO_3 to reduce uranium contamination and the leached particles were blended into a mixture of graphite flour and carbon black to which was added a thermosetting resin binder. The blended material was then extruded into long tubes and processed through the necessary thermal cycles to 1700°C. The outer and inner diameters of the resulting tubes conformed to the specified dimensions of the UHTREX fuel elements and the required lengths were machined out of the long tubes. The fuel elements were leached in HCl at 1600°C to remove uranium exposed by the machining operation and inspected for uranium content and final dimensions.

An evaluation of the resulting fuel elements indicated that residual exposed uranium was low. The carbon density, strength and permeability values were indicative of high quality fuel elements.

ACKNOWLEDGMENTS

The work was done for UHTREX Group K-4 by the following LASL groups: coated particle specification, procurement, treatment and evaluation - CMB-8, SP-1, CMB-1, N-1 and GMX-1; fuel element fabrication - CMB-6, CMB-3 and Shop Department; fuel element evaluation - GMX-1, Shop Department, N-1, CMB-11, CMB-1 and K-4.

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3. W. V. Goeddel, Nucl. Sci. Eng., 22, 201-218 (1964).
4. W. O. Harms, ORNL Coated-Particle Fuel Activity Letter for Period November 15, 1963, to May 15, 1964 - Part I, Oak Ridge National Laboratory Report ORNL-CF-64-5-17, May 6, 1964, p. 34.

FIGURE CAPTIONS

Figure 1 - Reactor Vertical Section

Figure 2 - Triplex Coated Particles for UHTREX

Figure 3 - Extrusion of UHTREX Fuel Element Stock

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