D 5/8

DEVELOPMENT OF A (U, Zr) C-GRAPHITE PULSED REACTOR FUEL ELEMENT*

J. A. REUSCHER, C. H. KARNES, R. H. MARION

Reactor Design and Development Division,
Sandia Laboratories, Albuquerque, New Mexico 87115, U.S.A.

SUMMARY

A new core is being designed to improve the performance of the Annular Core Pulse Reactor (ACPR). This design utilizes a two region core with the inner region containing high enthalpy fuel elements around the 228 mm diameter central irradiation cavity. An outer core region of uranium-zirconium hydride fuel elements is placed around the high-enthalpy inner core region to provide an adequate negative temperature coefficient for the entire core. A program has been conducted to develop a high-enthalpy fuel element using a graphite matrix fuel containing (U-Zr)C solid solution particles.

Since the (U, Zr)C-graphite fuel is subjected to a reactor period of two milliseconds with a large temperature gradient across the fuel diameter, fracture due to thermal stresses is the primary failure mode. The development program optimized this class of fuel for pulsed reactor application. A variety of graphite-based fuels were fabricated by the Materials Technology group of the Los Alamos Scientific Laboratory, and the details of this fabrication have been previously reported. Fuel specimens were fabricated by both extrusion and hot pressing using low thermal expansion graphite flour with uranium densities from 345 to 800 mg/cc. Mechanical and thermophysical characterization studies were performed to determine elastic modulus, fracture strength, thermal expansion, enthalpy and thermal conductivity in order to perform thermal and structural analyses. Fuel specimens with a diameter of 33 mm were tested without fracture in the ACPR to peak temperatures in excess of 2000 °C with radial peak-to-minimum energy deposition profiles of 2.0 to 3.0.

The outer diameter of the fuel element was fixed at 37.3 mm to be compatible with the existing core grid configuration. Since the (U, Zr)C-graphite must operate at high temperatures (2000 °C to 2300 °C) in the pulse mode an insulating liner is required to prevent excess temperatures in the 0.51 mm thick stainless steel clad. A filament wound graphite sleeve was chosen as the insulating liner because of its mechanical and thermal characteristics. The sleeve is one millimeter thick with an inside diameter of 34.0 mm. The thermal behavior of the fuel element design was analyzed with two dimensional heat transfer calculations. The fuel element design was verified by assembling a section of cladding with a 127 mm long fuel region and graphite liner. A water region was placed around the clad container and the apparatus was pulsed in the ACPR to peak fuel temperatures from 600 °C to 2300 °C. The fuel and clad temperatures were measured, the clad temperature rise varied from 50 °C to 120 °C.

The program has successfully developed a (U, Zr)C-graphite fuel element with a high volumetric enthalpy which can be used in a water cooled pulse reactor. Extensive tests have demonstrated the feasibility and safety of the fuel element design.

* This work was supported by the U.S. Energy Research and Development Administration.

1. Introduction

A project has been conducted to improve the pulse and steady-state performance of the Annular Core Pulse Reactor (ACPR). The current ACPR is a TRIGA reactor which uses uranium-zirconium hydride fuel and has been in operation since 1967. The ACPR contains a 23 cm diameter dry irradiation cavity which is utilized for a variety of experiments. The current reactor produces a pulse with a maximum energy release of 108 MJ and a pulse width at one-half maximum power of 4.5 milliseconds. The steady-state power is 600 kW. During the last few years, the ACPR has been employed in fast reactor safety experiments and the performance improvement of the reactor will increase its usefulness for such experiments. The Upgrade of the ACPR involves a new core with an inner region of high enthalpy fuel surrounded by an outer region of U-ZrH fuel. A (U,Zr)C-graphite composite was one of the candidate fuels considered for the central region. The existing grid structure will not be changed to save time and expense; the new fuel elements will have the same outer dimensions as the current ones. The neutron fluence in the irradiation cavity is expected to be increased by at least a factor of two since the fission density in the fuel surrounding the cavity will be significantly increased. The steady-state power of the reactor will be increased to 2 MW.

Since the fuel is subjected to temperatures in excess of 2000°C with a reactor period of about 2 milliseconds, the fuel experiences large thermal stresses and fractures may occur. One objective of the fuel element design was to develop a fuel configuration that remains intact for large temperature gradients. The fuel material consists of (U,Zr)C solid solution particles in a graphite matrix. A material development program in conjunction with an extensive in-pile test program has been conducted to eliminate fracturing of the fuel. The fuel element has been designed for maximum temperatures in excess of 2000°C with a modest temperature rise in the clad.

2. Fabrication Processes for (U,Zr)C-Graphite

The (U,Zr)C-graphite materials were fabricated by the Materials Technology Group at Los Alamos Scientific Laboratory (LASL) using both extrusion and hot pressing. Various combinations of graphite flour, carbide content, porosity, pressure, and temperature were used to produce fuel specimens which were resistant to fracture by thermal stresses. The details of the various fabrication processes are too lengthy to be included in this paper and can be found in the Quarterly Reports for the ACPR Upgrade.[1-8] The various lots of (U,Zr)C-graphite samples are listed in Table 1, along with pertinent process information.

2.1 Extrusion

Cylindrical samples in both solid and annular shapes were produced by the extrusion process. The extrusion process was developed at LASL for the Rover nuclear rocket program and the same basic techniques were used for the intial (U,Zr)C-graphite samples. Graphite flour (-200 mesh or -325 mesh KX-88, or 9553) is mixed with < 1 μm carbon black, furfuryl alcohol binder, and 3.5 μm (avg. size) ZrC and UO_2 (avg. size ~ 5 μm) particles. The mixture is extruded at room temperature and then subjected to the following thermal processing: (1) binder cure at 250°C, (2) outgas at 450°C, (3) binder carbonization at 850°C, (4) UO_2 reduction at 1600° to 1900°C, (5) (U,Zr)C-solid solution formed at 2200°C, and (6) graphitization at 2650°C. In order to eliminate the continuous carbide phase substructure, later material was extruded starting with (U,Zr)C solid solution powder which eliminates the UO_2 reduction and solid solution formation stages in the process. The carbide phase occupied 17 to 35 vol. percent of the resulting fuel. After thermal processing, the extruded material was cut into lengths varying from 3.2 to 76 mm.

2.2 Hot Pressing

Samples were prepared by hot pressing a mixture containing from 14 to 35 volume percent (U,Zr)C (average particle size ~ 3.5 μm), with the balance made of -325 mesh (< 45 μm) KX-88 graphite flour and voids. The pressing was performed at approximately 2650°C and pressures up to 24.8 MPa (3600 psi). Each pressing was cut into a number of pellets 3.18, 12.7, and 25.4 mm long. Low thermal expansion graphite flour (9553) and 350 μm long graphite fibers were also used to fabricate some samples.

3. Material Property Measurements

Mechanical and thermophysical properties were obtained for many of the lots of (U,Zr)C-graphite samples.

3.1 High-Temperature Mechanical Properties

The fracture strengths of several lots of (U,Zr)C-graphite are given in Table 2 up to 2400°C. These values were determined with tensile specimens, some of which failed in the grips producing minimum values of fracture stress and strain. The elastic modulus and the strain to failure were also measured for most lots of material.

3.2 Thermophysical Properties

Thermal expansion and thermal diffusivity measurements were conducted for the various lots of (U,Zr)C-graphite. The thermal expansion coefficients and thermal diffusivities are listed in Table 3.

3.3 Thermal Stress Resistance

The thermal stress resistance of the various lots of material can be ranked by considering the measured thermophysical and mechanical properties.[9] Such a procedure results in Lot SL017 being the best extruded material and Lots SL007, SL022, and SL024 as the best hot-pressed material.

4. Results of In-Pile Experiments

Extensive in-pile experiments were conducted in the ACPR with the various lots of (U,Zr)C-graphite to examine the fracture resistance of the specimens under pulsed conditions. The fuel pellets contained sufficient U-235 so that the energy depositions in these experiments approximated those which would occur in the ACPR Upgrade core. The in-pile experiments were divided into pellet tests which provided information on failure thresholds and element section tests which simulated the fuel element design.

4.1 Pellet Tests

Both solid cylinders and dual-slotted annuli were irradiated in a graphite holder to temperatures as high as 2800°C. A 25.4 mm thick polyethylene cylinder was placed around the graphite holder to enhance the energy deposition in the test sample. The specimens were tested in lengths from 3.2 to 76 mm and the pulse temperature was increased in steps with visual examinations between pulses. The observed failure thresholds were correlated with two-dimensional thermal stress calculations using the measured properties of the particular lot of (U,Zr)C-graphite. The peak-to-minimum energy deposition ratio in these experiments was as large as 2.5.

The thermal stress resistance determined from the in-pile reactor pulse tests agreed with the ranking determined from the material properties. Based on the results of the in-pile tests and the materials characterization, it was concluded that either as extruded fuel (similar to Lot SL017) or a hot-pressed fuel (similar to Lot SL007) could be fabricated which would not fracture in the upgraded ACPR core.

4.2 Element Section Tests

In order to experimentally verify the thermal characteristics of the fuel element design, element section tests were performed. A 200 mm long piece of stainless steel clad was fabricated into a pressure-tight vessel. The stainless steel vessel was positioned in an aluminum vessel filled with 700 cc of water. The clad was surrounded by a 14 mm thickness of water. Thermocouples were positioned in the fuel and on the clad; the pressure inside the clad and in the water region was monitored. A 25 mm thickness of polyethylene surrounded the aluminum container and enhanced the energy depositions in the fuel. The fuel region consisted of four 25.4 mm long, hot pressed (U,Zr)C-graphite pellets with 600 mg of U-235/cc. A filament wound graphite sleeve (Carbitex) was placed around the fuel to serve as an insulating barrier between the hot fuel and the clad. The experimental assembly was placed in the ACPR and subjected to pulses of various magnitudes. The fuel reached a peak temperature of 2300°C and the clad temperature did not exceed 150°C.

5. (U,Zr)C-Graphite Fuel Element Design

The design concept for a (U,Zr)C-graphite fuel element is shown in Fig.1. The various regions of the fuel element are identified in Table 4. Hot pressed pellets, 25 mm high, are stacked in the stainless steel cladding with a Carbitex liner. The total fuel length is 508 mm long and the fuel element length is 600 mm. An insulating spacer of graphite is provided at each end of the fuel stack. The fuel element is filled with an inert atmosphere of helium.

Two-dimensional heat transfer calculations were performed with TAC-2D[10] for both pulse and steady-state conditions. The calculations indicated that the fuel element could operate at a steady-state power of 16 kW without exceeding the heat flux corresponding to the Bernath correlation for the departure from nucleate boiling. In the upgraded core, the fuel element would operate at a steady-state power of 10 kW. For a pulse with a total energy release of 300 MW-sec in the upgraded core, the fuel reaches a temperature rise of 2400°C and the clad temperature does not exceed 140°C.

6. Conclusions

A (U,Zr)C-graphite fuel element has been developed for utilization in a water-cooled pulsed reactor. The fuel material has been developed to minimize cracking and fracture to improve the safety of the design. The design has been tested under conditions which simulate those which will exist in the upgraded core.

References

[1] Experimental Fast Reactor Safety Research Program - (Combined Quarterly Report, July-December 1974, SAND75-0068, Simulation Sciences Research Department, Sandia Laboratories, Albuquerque, New Mexico, February 1975.

[2] Experimental Fast Reactor Safety Research Program - Quarterly Report, January-March 1975, SAND75-0225, Simulation Sciences Research Department, Sandia Laboratories, Albuquerque, New Mexico, April 1975.

[3] Experimental Fast Reactor Safety Research Program - Quarterly Report, April-June 1975, SAND75-0449, Reactor Applications Research Department, Sandia Laboratories, Albuquerque, New Mexico, August 1975.

[4] Annular Core Pulse Reactor Upgrade - Quarterly Report, July-September, 1975, SAND75-0630, Reactor Research and Development Department, Sandia Laboratories, Albuquerque, New Mexico, January 1976.

[5] Annular Core Pulse Reactor Upgrade Quarterly Report, October-December 1975, SAND76-0165, Reactor Research and Development Department, Sandia Laboratories, Albuquerque, New Mexico, April 1976.

[6] Annular Core Pulse Reactor Upgrade Quarterly Report, January-March 1976, SAND76-0281, Reactor Research and Development Department, Sandia Laboratories, Albuquerque, New Mexico, July 1976.

[7] Annular Core Pulse Reactor Upgrade Quarterly Report, April-June 1976, SAND76-0371, Reactor Research and Development Department, Sandia Laboratories, Albuquerque, New Mexico, September 1976.

[8] Annular Core Pulse Reactor Upgrade Quarterly Report, July-September 1976, SAND76-0653, Reactor Research and Development Department, Sandia Laboratories, Albuquerque, New Mexico, January 1977.

[9] Thermal Stress Resistance of Graphite Matrix Fuels for Pulsed Reactors, by R. H. Marion, to be published.

[10] TAC-2D--A General Purpose Two-Dimensional Heat Transfer Computer Code, GA-8868, General Atomics, 1969, by J. F. Peterson.

TABLE 1
Lot Identification

Lot	Process/ Geometry	Density (g/cc)	Porosity (%)	Carbide Content (vol %)	U Loading 93% Enriched (mg U/cc)	Type of Graphite Flour	Size of Graphite Flour (μm)
SL001	Extruded/ Solid	3.497	22.6	34.8	434	KX-88	<75
SL002	Extruded Solid	3.538	23.3	35.1	509	KX-88	<75
SL003	Extruded/ Outer Dual Annulus	3.560	18.6	35.8	332	KX-88	<75
SL004	Extruded/ Inner Dual Annulus	3.545	21.8	35.1	491	KX-88	<75
SL007	Hot Pressed Solid	3.55-3.80	10.0	31.8	359	KX-88	<45
SL008	Hot Pressed Solid	3.32-3.78	20.0	33.1	383	KX-88	<45
SL012	Extruded/ Inner Dual Annulus	3.414	19.4	30.2	519	9553	<45
SL013	Extruded/ Outer Dual Annulus	2.826	21.6	20.1	345	9553	<45
SL015	Hot Pressed Solid	3.597	~8	29.4	435	9553	<45
SL017	Extruded/ Solid	2.69	23.4	16.7	420	9553	<45
SL018	Hot Pressed Solid W/Fibers	3.50	10.0	29.2	432	9553	<45
SL019	Hot Pressed Solid	3.776	4	31.9	453	9553	<10
SL021	Hot Pressed Solid	3.024	10	17.1	423	9553	<45
SL022	Hot Pressed Solid	3.404	13	24.8	626	KX-88	<45
SL024	Hot Pressed Solid	3.808		13.7	736*	KX-88	<45

* 62.6% Enriched

Table 2

Mechanical Properties of Representative Lots of (U,Zr)C-Graphite

Temperature (°C)	Fracture Strength MPa (ksi)					
	SL007	SL012	SL015	SL017	SL019	SL024
22	55.1 (8.0)	50.1 (7.27)	38.0 (5.5)	35.3 (5.1)	20.6 (3.0)	20.7 (3.0)
1000	49.6 (7.2)	45.6 (6.62)	30.3 (4.4)	32.2 (4.7)	26.9 (3.9)	20.7 (3.0)
1500	46.9 (6.8)	37.1 (5.4)	27.5 (4.0)	37.2 (5.4)	21.8 (3.2)	17.2 (2.5)
2000	34.5 (5.0)	32.9 (4.8)	24.1 (3.5)	33.3 (4.8)	21.4 (3.2)	17.2 (2.5)
2400	10.3 (1.5)	10.4 (1.5)	9.2 (1.3)	16.5 (2.4)	7.8 (1.1)	6.9 (1.0)

Table 3

Thermophysical Properties

Lot Number	Thermal Expansion 20 - 1500° C ($\times 10^{-6}$/° C)	Thermal Diffusivity 1835° C cm^2/sec
SL001	6.6	.093
SL002	6.6	.100
SL003	6.7	.092
SL004	--	.094
SL007 & SL008	7.2	.082
SL012	5.8	.107
SL013	4.3	.108
SL015	7.7	.077
SL017	4.4	.122
SL018	8.5	.166
SL019	8.1	----

Table 4

Design Concept for (U,Zr)C-Graphite Fuel Element

Region	Description
1	Upper stainless steel end fitting.
2	6.4 mm wide expansion gap.
3	Graphite insulator, 25.4 mm long, 35.6 mm O.D.
4 thru 23	(U,Zr)C-graphite fuel region 25.4 mm long pellets, 33 mm O.D.
24	Graphite insulator, 25,4 mm thick, 35.6 mm O.D.
25	Lower stainless steel end fitting.

(UC-ZrC) GRAPHITE FUEL ELEMENT DESIGN

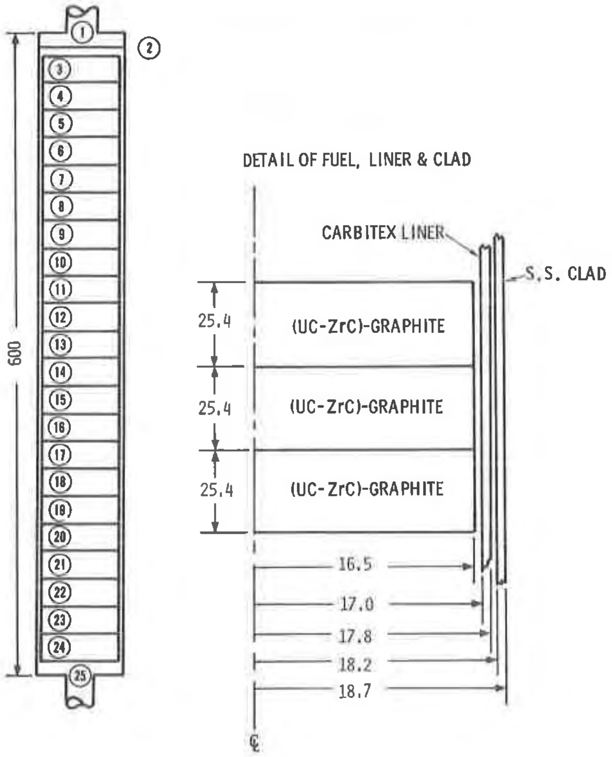

FIGURE 1. (U,ZR)C-GRAPHITE FUEL ELEMENT DESIGN

(ALL DIMENSIONS IN MM)