GENERAL ATOMIC

GA-A13506

HTGR FUEL PERFORMANCE

by
D. P. HARMON
R. C. DAHLBERG
P. SCHLEIFER
R. FINKBEINER*

This is a preprint of a paper to be presented
at the International Nuclear Industries Fair,
October 7-11, 1975, Basel, Switzerland

Work supported in part
by the U.S. Energy Research and Development Administration,
Contract AT(04-3)-167, Project Agreement No. 17

*General Atomic Europe

GENERAL ATOMIC PROJECT 3224 OCTOBER 1, 1975

NUCLEX 75

Fachtagung Nr. 6

Séance technique No. 6

Technical Meeting No. 6

Fuel Elements for Proven Tower Reactor Systems - Performance and Development

HTGR Fuel Performance

D. P. Harmon
R. C. Dahlberg
P. Schleifer
General Atomic Company

R. Finkbeiner
General Atomic Europe

HTGR FUEL PERFORMANCE

D. P. Harmon
R. C. Dahlberg
P. Schleifer
General Atomic Company

R. Finkbeiner
General Atomic Europe

SUMMARY

The reference fuel for the thorium cycle HTGR is composed of BISO-coated fertile particles and TRISO-coated fissile particles. These particles are blended together and bonded into fuel rods with a carbonaceous matrix. The fuel rods are then inserted into graphite fuel blocks.

Extensive research, development, and test work applicable to HTGR fuel has been underway at GA and numerous other facilities around the world for over 12 years. Results from irradiation tests have demonstrated reliable performance to and well beyond current HTGR design conditions. A wide range of coated particle microstructures and geometries has proven successful, indicating that considerable latitude is available in both fissile and fertile particle designs. Tests on the irradiation behavior of fuel rods have shown that with proper design there are no detrimental rod matrix-particle coating interactions.

A comprehensive testing program has been under way on several near-isotropic graphites to determine the behavior of its physical, thermal, and mechanical properties with irradiation. The tests have shown that near-isotropic graphites will satisfy current HTGR core designs and should provide additional design margins for increasing core power density and temperatures as required for advanced fuel designs.

ZUSAMMENFASSUNG

Der Referenzbrennstoff für den im Thorium-Brennstoffzyklus betriebenen HTGR besteht aus Brutstoffpartikeln mit BISO-Beschichtung und Spaltstoffpartikeln mit TRISO-Beschichtung. Diese Partikeln werden gemischt und zusammen mit einem graphitischen Matrixmaterial zu Brennstoffstäben geformt. Die Brennstoffstäbe werden in Bohrungen des Brennelementblockes aus Graphit eingesetzt.

Ausgedehnte Forschungs- und Entwicklungsarbeiten und experimentelle Untersuchungen werden seit über 12 Jahren bei General Atomic und zahlreichen anderen in- und ausländischen Forschungseinrichtungen durchgeführt. Ergebnisse von Bestrahlungstests haben das zuverlässige Betriebsverhalten dieses Brennstoffs unter schärferen Beanspruchungen als den von der Reaktorauslegung vorgegebenen demonstriert. Ein erfolgreiches Bestrahlungsverhalten konnte an einem breiten Bereich von Mikrostrukturen und geometrischem Aufbau der Partikeln nachgewiesen werden, so dass ein beträchtlicher Spielraum hinsichtlich der Auslegung der Spalt- und Brutstoffpartikeln existiert. Bestrahlungstests an Brennstoffstäben haben nachgewiesen, dass bei geeigneter Auslegung keine unerwünschten Wechselwirkungen zwischen Brennstabmatrix und Partikelbeschichtung auftreten.

Ein umfangreiches Untersuchungsprogramm wurde an verschiedenen nahezu isotropen Graphiten durchgeführt, um das Verhalten der physikalischen, thermischen und mechanischen Eigenschaften unter Bestrahlung zu bestimmen. Diese Untersuchungen haben gezeigt, dass nahezu isotrope Graphite die an sie sowohl von der derzeitigen Reaktorauslegung her gestellten anforderungen als auch die bei erhöhter Leistungsdichte oder höheren Temperaturen sich ergebenden anforderungen, erfüllen.

RESUME

Le combustible de référence pour le cycle HTGR au thorium est composé de particules enrobées fertiles du type BISO et de particules enrobées fissiles du type TRISO. Ces particules sont melangées et amalgamées par une matrice de carbone sous forme de crayons de combustible. Ces crayons sont ensuite insérés dans elements de combustible en graphite.

Un programme de recherche et de développement dans le domaine du combustible HTGR est en cours depuis plus de 12 ans à General Atomic et dans de nombreux autres laboratoires dans le monde. Les résultats d'essai d'irradiations ont démontré un comportement sûr du combustible et cela même au-delà des conditions d'utilisation prévues pour l'HTGR d'aujourd'hui. Un vaste ensemble de particules enrobées, tant en ce qui concerne la microstructure que la géometrie, s'est avéré entièrement satisfaisant, ce qui indique une marge de manoeuvre très importante pour le choix des types de particules enrobées fissile et fertiles. Les essais de comportement sous irradiation de crayons de combustible ont

demontré que pour un dessin adéquat il n'y avait pas d'interactions défavorables entre la matrice de carbone et l'enrobage de particules.

D'autre part, un programme d'essai étendu est en cours sûr plusieurs types de graphites quasi-isotropiques pour déterminer leur comportement physique, thermique et mécanique sous irradiation. Ces essais ont demontré que de tels graphites sont satisfaisants pour des coeurs HTGR courants et qu'ils devraient par ailleurs fournir une marge suffisante pour augmenter la puissance spécifique du coeur et les températures, comme l'exigent des conceptions avancées du combustible.

INTRODUCTION

Development of the high-temperature gas-cooled reactor (HTGR) has been under way in the United States since 1957. The initial fuel development effort for the Peach Bottom and Fort St. Vrain reactors was directed towards demonstrating the coated fuel particle concept and the multihole graphite fuel element design. Development of fuel for the large commercial HTGRs, termed LHTGRs, draws heavily upon General Atomic's experience in developing and manufacturing fuel for these reactors, especially for the 330 MW(e) Fort St. Vrain power station. However, optimization of the LHTGR fuel has led to a number of changes from Fort St. Vrain fuel, which are summarized in Table 1. These differences are principally the result of three major design goals: (1) complete separation of fissile and fertile material, (2) simplification of the fuel fabrication process, and (3) improved fuel performance.

LHTGR fuel consists of small, right-cylinder rods 1.56 cm in diameter by 6.3 cm long. These contain an intimate blend of coated fuel particles and isotropic graphite shim particles bonded together in a close-packed array. Fuel rods are formed by injecting the close-packed particle bed with a matrix material consisting of an organic binder and graphite filler, which when carbonized and heat treated yields a relatively strong carbonaceous bonding media. The fuel rods are carbonized and heat treated in situ in the graphite fuel element. The use of isotropic graphite shim particles and the cure-in-place (CIP) process for LHTGR rods represent design improvements that are attractive in both fresh fuel manufacturing and in the remote refabrication process. This commonality between fresh and recycle fuel will reduce overall development costs and reduce risks associated with the refabrication plant.

Two types of coated fuel particles are used in LHTGRs: fissile and fertile. The fissile particle has a dense UC_2 fuel kernel consisting of highly enriched (93%) uranium; the fertile particle has a ThO_2 kernel. The design of the coated particles using TRISO coatings on fissile particles and BISO coatings on fertile particles facilitates separation of the bred U-233 from the nuclear poison U-236, generated primarily in the fissile kernel. During reprocessing, a burn-leach step is used to separate thorium and U-233 from the fissile particle.

Since the coatings also serve as the primary barrier to fission product release, it is important to ensure a high degree of coating integrity under all normal operating conditions. This is done in two ways: (1) design and specification of coated particle parameters to ensure a high probability of surviving irradiation-induced stresses, and (2) thermal design of the reactor core to eliminate significant failure due to thermochemical interactions between fuel and fission products with structural coating layers. The results of extensive irradiation testing conducted by GA have demonstrated reliable performance to and beyond current LHTGR design conditions.

IRRADIATION PERFORMANCE OF COATED FUEL PARTICLES

A large number of coated particle samples have been irradiated by GA over the last 12 years. Almost 600 coated particle samples, 289 TRISO and 243 BISO, have been irradiated in 40 capsule experiments. Each sample contained between 300 and 5000 coated particles, all of which were examined before and after irradiation. Of these, 197 TRISO and 164 BISO samples demonstrated successful irradiation performance (>99% survival). A number of samples designed to test the limits of coating design and particle performance showed varying degrees of failure. These tests form the basis for the Fuel Product Specifications for coated particle fuels.

The initial irradiation test of the reference LHTGR fissile particle, UC_2 TRISO, was conducted in capsule P13L. Fuel particles in this test exhibited satisfactory performance to exposures of 1250°C and 7.8×10^{21} n/cm^2 $(E > 0.18 \text{ MeV})_{HTGR}$ with fuel burnups of 75% FIMA. These data provide the primary basis for the current fissile particle design. Early in the test program a very conservative approach to the all-uranium fissile particle design was taken because of lack of experience with the extremely high burnups. Therefore, fuel kernels of 100 μm diameter and thus relatively low power and fission product inventories per particle were investigated. Both economic and process studies indicated that larger kernels were desirable, and the initial irradiation results from P13L encouraged development in that direction. Larger diameter fissile particles were irradiated in capsules P13N (150 μm UC_2) and P13P (200 μm UC_2). In the latter test, LHTGR reference size particles (200 μm UC_2 TRISO) exhibited satisfactory performance to neutron exposures of 8.5 x 10^{21} n/cm^2 at 1350°C. A recently completed series of tests now undergoing postirradiation examination (capsules P13R and P13S) has demonstrated survival of reference design fissile particles from five different manufacturing batches to 12.1 x 10^{21} n/cm^2 at 1075°C (design) and ≥75% FIMA. This fluence is more than 50% beyond the expected peak exposure in commercial reactors.

The design of the reference fertile ThO_2 particles is based primarily on empirical irradiation results and dimensional change considerations. Initial design studies indicated BISO fertile particles having very thick coatings exhibit large shrinkages, particularily early in life, while particles with thin coatings may experience net expansion at high burnups. These considerations have led to the selection of the current nominal coating thicknesses; i.e., 85 μm buffer and 75 μm outer pyrocarbon layers. In a series of ThO_2 BISO design screening tests, it was demonstrated that many BISO particle designs exhibit satisfactory performance, providing the anistropy of the dense outer pyrocarbon layer is sufficiently low. More recent results from unbonded particle tests conducted in capsules P13R and P13S have shown that the 11 different ThO_2 BISO batches meeting current LHTGR design specifications had a 99.93% survival rate to 12.1 x 10^{21} n/cm^2 at 1075°C (design) and 5.6% FIMA.

A wide range of coated particle microstructural and geometrical designs have been successful in these experiments, indicating that considerable latitude is available in both fissile and fertile particle designs. The samples were designed using analytical stress models; some were deliberately under-designed to establish failure criteria. Over 100 of the TRISO samples and nearly 50 BISO samples were made in large-scale production equipment. Table 2 summarizes the completed GA irradiation tests of TRISO and BISO coated particles. The extent of GA successful demonstrations of coated particle performance is shown in Table 3.

THERMAL PERFORMANCE OF COATED FUEL PARTICLES

Out-of-pile testing is used to evaluate the stability of fissile-TRISO and fertile BISO particles at high temperatures both isothermally and in a thermal gradient. This work has provided the primary source of data for setting criteria in core design to limit failures from the two thermally activated failure mechanisms: kernel migration and fission product attack of the silicon carbide coating.

Considerable work has been done to characterize kernel migration of carbide- and oxide-coated particles in a thermal gradient. The primary limiting property in thermal performance of the fuel under nominal conditions is the tendency of the fuel kernel to migrate in a thermal gradient toward the hot side of the particle. This migration can cause detrimental chemical interactions with the coating during irradiation. Empirical relationships, derived for both UC_2 and ThO_2 kernels, describe kernel migration rates in terms of the reactor environment. The temperature and thermal gradient dependence of kernel migration for both irradiated and unirradiated UC_2 particles have been shown to be similar.

A second failure mechanism operative in UC_2 TRISO particles is fission product attack of the silicon carbide layer. Results from in-pile tests have shown that at high burnups fission products, mainly rare earths, diffuse toward the cold side of the particle and accumulate on the inside surface of the silicon carbide coating. Under extreme temperature and high burnup conditions, failure of the silicon carbide layer has been observed. Out-of-pile studies of irradiated particles have been used to define the time-temperature-burnup limits for fuel survival and results of these studies are in very good agreement with the in-pile observations.

IRRADIATION PERFORMANCE OF FUEL RODS

A total of 171 pitch-bonded fuel rods fabricated by GA have been irradiated in 16 high exposure, instrumented, capsule experiments. As shown in Table 4, more than 110 of these rods have demonstrated successful performance. A number of rods have been designed for failure to study the effects of particle, matrix, and process variables on irradiation performance. The results of these tests provide a firm basis for specifications of fuel rod properties and processes.

Capsule tests most important to demonstrating the high exposure performance of LHTGR fuel rods are HRB-4, HRB-5, HRB-6, P13Q, P13R, and P13S. The latter three capsules are now undergoing postirradiation examination. However, in-pile fission gas measurements have shown that the fuel performed satisfactorily well beyond expected peak LHTGR fluences. Capsule P13Q was an integral body test of cure-in-place rods containing reference fissile and fertile particles irradiated to a peak exposure of 9.9 x 10^{21} n/cm^2. It had a low and nearly constant in-pile release (R/B Kr^{85m} $\sim$1 x 10^{-6}) throughout its operation indicating little or no coated particle failure occurred.

The three HRB capsules (HRB-4, -5, and -6) were the first test of fuel rods that were cured in place and contained isotropic graphite shim particles. They were irradiated to a peak exposure of 10.5 x 10^{21} n/cm^2 $(E > 0.18\ MeV)_{HTGR}$ at 1250°C. All rods were found to be in excellent condition after irradiation. Dimensional change measurements showed all rods exhibited both radial and axial shrinkage, and the dimensional change profiles were compatible with those of moderator graphite fuel elements. This is an important measure of fuel rod performance since net shrinkage is necessary to avoid mechanical interactions with the moderator block, but excessive shrinkage is undesirable because of its effect on fuel temperatures. Some matrix-coating interactions were observed in these rods as a result of a high matrix binder phase content; however, slight modifications in the matrix composition have eliminated this condition in rods fabricated subsequent to these tests.

A significant milestone in the overall HTGR fuel development program is the Fort St. Vrain (FSV) proof test capsule, F-30. This test contained FSV production material and irradiated to about 20% beyond expected peak exposure. All fuel rods were in very good condition after irradiation and exhibited dimensional changes that were in excellent agreement with model predictions. The irradiation performance of fuel rods in capsule F-30 gives a high degree of confidence in the FSV fuel. Calculated fission gas release from the FSV core based on empirical data from capsule F-30 indicates the circulating coolant activity should remain well below the design level throughout life.

Large numbers of fuel rods have been successfully irradiated under representative HTGR conditions in test elements in the Peach Bottom reactor. Irradiation data obtained from these tests substantiate the performance predictions based on accelerated capsule tests.

PERFORMANCE OF GRAPHITE

GA has conducted an extensive and continuing survey of the manufacture and testing of nuclear graphites. This work involves close liaison with technical personnel of the major carbon companies and of the other programs on nuclear graphite evaluation in the United States and in Europe. A comprehensive testing program has been under way for approximately 6 years on several near-isotropic graphites. Their behavior is being determined by measuring physical, thermal, and mechanical properties in the unirradiated state and as a function of irradiation to fast fluences and temperatures beyond the intended service life of graphite in an HTGR. These tests have shown that near-isotropic graphites will satisfy current HTGR core designs and should provide additional design margins for increasing core power density and temperatures as required for advanced fuel designs.

The mean tensile strength of near-isotropic graphites has been shown to increase with irradiation. Thermal expansivity has been found to remain unchanged at low irradition temperatures and to decrease during irradiation at higher temperatures. Thermal conductivity decreases rapidly at low fluences and saturates with further irradiation up to the fluences of HTGR service. The magnitude of the thermal conductivity change is reduced with increasing irradiation temperature. Irradiation-induced dimensional changes in the near-isotropic graphite elements have been shown to be within desired design limits over the intended life of the HTGR and net expansion will not occur. Irradiation creep measurements on similar graphites have shown relief of stresses within the graphite. Experiments in progress are planned to obtain full lifetime irradiation data on production near-isotropic graphites for the HTGR.

ACKNOWLEDGMENT

The results presented here draw on contributions from many members of the staffs of General Atomic Company and Oak Ridge National Laboratory.

TABLE 1
PRINCIPAL DIFFERENCES BETWEEN FORT ST. VRAIN AND LARGE HTGR FUEL

	Fort St. Vrain	LHTGR	Purpose of Change
Fuel Particles			
1. Fissile kernel	4:1 $(Th,U)C_2$	UC_2 (melted kernel)	Improve separability
2. Fertile kernel	ThC_2	ThO_2	Simplify process Improve performance
3. Fissile/fertile coatings	TRISO/TRISO	TRISO/BISO	Improve separability
Fuel Rods			
1. Carbonization technique	In Al_2O_3	In-block	Simplify process
2. Rod shimming method	Fuel particle blending	Graphite shim particles	Simplify process
3. Dimensions	Dia: 1.27 cm Length: 5.08 cm	Dia: 1.57 cm Length: 6.35 cm	Improve core design Simplify process

TABLE 2
COMPLETED GA IRRADIATION TESTS OF COATED PARTICLES

Test Number	Coated Particle Type(s)	Number of Samples			Peak Irradiation Test Conditions			Results: Samples Demonstrating Successful Performance[b,c]	
		TRISO[a]	BISO[a]	Total	Temperature (°C)	Burnup (% FIMA)	Fast Fluence E > 0.18 MeV (10^{21} n/cm^2)	TRISO	BISO
P13E to P13J	TRISO, BISO, other	34	27	70	900 to 1350	4 to 22	0.6 to 4.3	31	23
P13K	BISO, other		15	15	900, 1300	59	2.9		10
P13L	TRISO, BISO	15	15	30	900, 1300, 1500	75	7.8	4	3
P13M	TRISO, BISO	10	5	15	1350	70	6.9	9	3
P13N	TRISO, BISO	15	9	24	1350, 1500	68	5.2	10	5
P13P	TRISO, BISO	20	12	32	1050, 1350	70	8.5	17	0
P13R[e]	TRISO, BISO	9	12	21	1075	75	12.1	3	8
P13S[e]	TRISO, BISO	9	11	21	1075	75	11.8	5	9
P6 to P14	BISO, other		57	111	600 to 1450	7 to 18	0.1 to 5.6		29
P15	BISO		14	14	1200	24	3.8		11
P16	BISO		13	13	1300	15	2.5		10
P17	BISO		15	15	1175	27	4.8		15
P18	BISO		15	15	1125	26	8.4		15
P19	BISO		14	15	900, 1100	23	6.7		14
P20	TRISO	15		15	900, 1150	27	8.7	10	
P21	TRISO	15		15	900, 1200	17	4.3	11	
P22	TRISO	30		30	900, 1100	~23	7.7	13	
P23	TRISO, BISO	20	6	26	900, 1250	13	2.3	18	6
F-25	TRISO	9		9	1250	~15	4.3	7	
F-26	TRISO	9		9	1200	20	7.0	5	
F-27	TRISO, BISO	7	3	10	1250	17	5.7	4	3
F-28	TRISO	9		9	1250	~15	4.0	6	
F-29	TRISO	9		9	1150	24	8.5	5	
F-30	TRISO	16		16	1250	20	10.6	9	
FR-1	TRISO	10		10	550, 875	22	5.3	5	
FR-2	TRISO	10		10	550, 1200	7	1.0	10	
FR-3	TRISO	10		10	600, 1200	7	1.3	10	
HRB-2[d]	TRISO	8		8	750, 1100	11	7.9	8	
	TOTAL TESTS	289	243	597	Successful tests to 1500°C	Successful tests to 75% FIMA	Successful tests to 12.1	197	164

(a) Samples with round, dense kernels, 150 to 500-micrometer nominal diameter.

(b) Less than 1% failure.

(c) A number of experimental particle batches designed to test the limits of coating parameters or particle performance have shown varying degrees of failure. No failures have been observed in samples containing particles designed to current specifications.

(d) Cooperative HNL-GA irradiation experiment in HFIR.

(e) Preliminary results, postirradiation examination in progress.

TABLE 3
NUMBER OF COATED PARTICLE SAMPLES SUCCESSFULLY TESTED TO INDICATED EXPOSURE

Type	Fast Fluence (10^{21} N/CM^2)							
	1	2	3	4	5	6	7	≥8
BISO	31	18	31	19	15	15	8	31
TRISO	21	21	19	38	29	9	23	38

Type	Burnup (% FIMA)												
	≤2	4	6	8	10	12	14	16	18	20	21-49	50-69	70-75
BISO	14	13	39	9	7	8	9	7	12	6	36	5	3
TRISO	15	2	13	7	9	27	9	25	11	6	37	22	15

Type	Temperature (°C)									
	500	600	700	800	900	1000	1100	1200	1300	≥1400
BISO		1		6	12	11	22	22	71	23
TRISO	6	3	7	1	24	30	36	29	63	

TABLE 4
COMPLETED GA IRRADIATION TESTS OF FUEL RODS

Capsule Number	Peak Irradiation Test Conditions		Results	
	Temperature (°C)	Fast Fluence (10^{21} n/cm²)	No. of Rods Tested	No. of Rods Demonstrating Successful Performance[a]
F-25	1150-1300	3.3-4.8	12	10
F-26	1250-1300	5.1-7.0	8	5
F-27	700	3.7-4.0	2	1
	1250-1350	4.8-5.7	5	3
F-28	700-900	1.8-2.5	2	2
	1200-1250	3.2-3.7	4	3
F-29	750	3.4-5.0	2	2
	1200	6.8-7.8	4	0
F-30	1050	5.3-10.6	10	10
	1250	4.1-6.5	3	3
HRB-2[b]	750	4.6-6.2	3	3
	1100	7.5-7.9	3	3
HRB-3[b]	1150	5-9	2	1
HRB-4[b]	1250	4-10.5	6	6
HRB-5[b]	1250	2-4.5	6	6
HRB-6[b]	1250	5-9	6	6
P13M	1050	8.1-8.4	6	6
	1350	2.8-4.8	6	6
P13N	1350	1.6-5.4	16	14
	1560	4.2-4.7	4	2
P13P	1050	5.0-7.5	8	8
	1350	2.5-8.5	13	8
P13R	1075	3.7-12.4	15	(c)
	1300	7.9-9.5	5	(c)
P13S	1075	3.5-12.1	15	(c)
	1500	7.5-9.1	5	(c)
	Successful tests to 1500°C	Successful tests to 10.6 x 10^{21} n/cm²	171	≥110[c]

(a) Criteria are good rod integrity and dimensional shrinkage.

(b) Excludes HNL rods.

(c) All rods irradiated in capsules P13R and P13S were intact and in very good condition; however, results not included since postirradiation examination is still in progress.