180

3-1-78

LH. 1879

GA-A14690
UC-77

MASTER

ASSESSMENT OF GRADE H-451 GRAPHITE FOR REPLACEABLE FUEL AND REFLECTOR ELEMENTS IN HTGR

by
G. B. ENGLE

Prepared under
Contract EY-76-C-03-0167
Project Agreement No. 17
for the San Francisco Operations Office
Department of Energy

DATE PUBLISHED: DECEMBER 1977

~~DISTRIBUTION~~ OF THIS DOCUMENT IS UNLIMITED

GENERAL ATOMIC COMPANY

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161
Price: Printed Copy \$8.00; Microfiche \$3.00

GA-A14690
UC-77

ASSESSMENT OF GRADE H-451 GRAPHITE FOR REPLACEABLE FUEL AND REFLECTOR ELEMENTS IN HTGR

by
G. B. ENGLE

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

**Prepared under
Contract EY-76-C-03-0167
Project Agreement No. 17
for the San Francisco Operations Office
Department of Energy**

**GENERAL ATOMIC PROJECT 3224
DATE PUBLISHED: DECEMBER 1977**

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

GENERAL ATOMIC COMPANY

.

.

.

.

.

.

ABSTRACT

Experimental data for grade H-451 graphite are presented and assessed to support licensing of H-451 graphite for use as fuel element blocks in a high-temperature gas-cooled reactor (HTGR). Additional data from the literature on graphite grades similar to grade H-451 are presented to supplement the H-451 data. Evaluation programs at General Atomic Company (GA) covering characterization, irradiation, and oxidation studies, along with studies carried out in the Great Lakes Carbon Corporation H-451 development program, are reported.

Grade H-451 is a high-purity graphite manufactured by extrusion as logs 432 mm in diameter by 864 mm in length from a special blend of near-isotropic petroleum coke using conventional graphite manufacturing processes. Prototype, preproduction, and production logs have been evaluated by systematically measuring physical and mechanical properties, impurity content, and fatigue behavior. Controlled irradiation experiments, carried out in instrumented capsules, were utilized to provide data over the temperature and fluence life of fuel elements in an HTGR. Irradiation-induced creep coefficients were taken from literature values on H-451 and graphites similar to H-451. Oxidation rates in helium-water mixtures and the effect of oxidation burnoff on strength and elastic modulus of H-451 are reported.

Grade H-451 shows an improvement in radial strength and thermal conductivity over H-327, a needle-coke graphite, which is currently used for HTGR replaceable fuel and reflector elements. Grade H-451 also has a lower anisotropy factor. Overall, H-451 will provide better safety margins in the Fort St. Vrain (FSV) reactor and more flexibility in design for future HTGRs.

CONTENTS

ABSTRACT	iii
1. SUMMARY AND CONCLUSIONS	1-1
1.1. Introduction	1-1
1.2. Concept and Need for a Near-Isotropic Fuel Element Graphite	1-1
1.3. Development Program at GLCC	1-2
1.4. Characterization Program	1-5
1.4.1. Distribution of Properties Within Individual Logs and Between Logs Within a Lot	1-5
1.4.2. Statistical Distribution of Strength Within a Log	1-6
1.4.3. Definition of Minimum Strength and Strength Distribution Among Logs	1-7
1.4.4. Fatigue Behavior	1-8
1.5. Irradiation Program	1-9
1.5.1. Dimensional Changes	1-9
1.5.2. Tensile Strength and Elastic Modulus	1-9
1.5.3. Fatigue Behavior	1-10
1.5.4. Thermal Expansivity	1-10
1.5.5. Thermal Conductivity	1-10
1.5.6. Irradiation-Induced Creep	1-11
1.6. Oxidation Program.	1-12
1.6.1. Oxidation Rate of H-451 Graphite	1-12
1.6.2. Effect of Oxidation on Tensile Strength and Elastic Modulus	1-12
2. INTRODUCTION	2-1
3. CONCEPT AND NEED FOR A NEAR-ISOTROPIC FUEL ELEMENT GRAPHITE . .	3-1
4. DEVELOPMENT PROGRAM AT GLCC	4-1
4.1. Filler Cokes	4-1
4.2. Processing	4-4

4.3.	Prototype and Preproduction Development	4-4
4.4.	Conclusions	4-6
	REFERENCE	4-6
5.	CHARACTERIZATION PROGRAM	5-1
5.1.	Introduction	5-1
5.2.	Distribution of Properties Within Individual Logs and Between Logs Within a Lot	5-1
5.2.1.	Sampling	5-1
5.2.2.	Bulk Density	5-2
5.2.3.	Mechanical Properties	5-2
5.2.4.	Thermal Properties	5-4
5.2.5.	Impurity Content	5-6
5.2.6.	Conclusions	5-14
5.3.	Statistical Distribution of Strength Within a Log	5-15
5.3.1.	Sampling.	5-16
5.3.2.	Conclusions	5-16
5.4.	Definition of Minimum Strength and Strength Distribution Among Logs	5-17
5.4.1.	Sampling	5-17
5.4.2.	Experimental Results	5-18
5.4.3.	Application of Results to Acceptance Testing	5-20
5.4.4.	Conclusions	5-26
5.5.	Fatigue Behavior	5-27
5.5.1.	Sampling	5-28
5.5.2.	Experimental Procedure	5-28
5.5.3.	Experimental Results.	5-28
5.5.4.	Conclusions	5-31
	REFERENCES	5-31
6.	IRRADIATION PROGRAM	6-1
6.1.	Introduction	6-1
6.2.	Sampling	6-3
6.3.	Dimensional Changes	6-3
6.3.1.	873 to 1673 K (600° to 1400°C)	6-3
6.3.2.	573 to 873 K (300° to 600°C)	6-7

6.4.	Tensile Strength, Elastic Modulus, and Strain at Fracture. . .	6-9
6.4.1.	823 to 1623 K (550° to 1350°C)	6-9
6.4.2.	627 to 873 K (350° to 600°C)	6-10
6.4.3.	Between-Log and Within-Log Variations in Strength Increase.	6-11
6.5.	Fatigue Behavior	6-13
6.5.1.	Sampling and Procedures.	6-13
6.5.2.	Experimental Results and Discussion.	6-13
6.6.	Thermal Expansivity.	6-15
6.6.1.	823 to 1623 K (550° to 1350°C)	6-15
6.6.2.	623 to 823 K (350° to 550°C)	6-17
6.7.	Thermal Conductivity	6-17
6.7.1.	923 to 1623 K (650° to 1350°C)	6-17
6.7.2.	523 to 923 K (250° to 650°C)	6-19
6.7.3.	Effect of Change in Temperature.	6-20
6.8.	Irradiation-Induced Creep.	6-20
6.8.1.	Conclusions.	6-21
	REFERENCES	6-21
7.	OXIDATION PROGRAM	7-1
7.1.	Oxidation Rate of H-451 Graphite	7-1
7.1.1.	Reaction Rate Theory	7-1
7.1.2.	Experimental Procedure and Sampling	7-3
7.1.3.	Determination of Reaction Rate Constants	7-4
7.1.4.	Application of Rate Constants	7-6
7.2.	Effect of Steam Oxidation on Tensile Strength and Elastic Modulus	7-8
7.2.1.	Experimental Procedure and Sampling	7-8
7.2.2.	Experimental Results	7-9
	REFERENCES	7-22
8.	ACKNOWLEDGMENTS	8-1
	APPENDIX A: SAMPLING DIAGRAMS	A-1
	APPENDIX B: RESULTS OBTAINED DURING ASSESSMENT PROGRAM	B-1

FIGURES

1-1.	Comparison of dimensional changes for H-451 and H-327 graphites: (a) axial, (b) radial	1-4
------	---	-----

6-1.	H-451 graphite irradiation data	6-2
6-2.	Irradiation-induced dimensional change in H-451 graphite (design curves): (a) axial direction, (b) radial direction . .	6-5
6-3.	Dimensional changes in H-451 graphite irradiated with changes in temperature (from Ref. 6-3)	6-6
6-4.	Fatigue stress limits for survival of H-451 graphite to 10^5 cycles, normalized to the unirradiated tensile strength, versus the fast neutron fluence at an irradiation temperature of 1173 to 1263 K (900° to 990°C)	6-16
7-1.	Comparison of rate constants as a function of P_{H_2O} (from Ref. 7-2).	7-7
7-2.	Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial edge and center specimens) oxidized at 1073 K (800°C)	7-14
7-3.	Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial specimens) oxidized at 1273 K (1000°C)	7-15
7-4.	Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial center specimens)	7-16
7-5.	Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length radial specimens) oxidized at 1073 K (800°C)	7-17
7-6.	Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length radial specimens) oxidized at 1273 K (1000°C)	7-18
7-7.	Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length axial center specimens)	7-19
7-8.	Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length axial specimens) oxidized at 1273 K (1000°C)	7-20
7-9.	Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length radial edge specimens) oxidized at 1273 K (1000°C).	7-21
A-1.	Sampling diagram of H-451 for within-log property measurements: slabs and sections	A-3
A-2.	Sampling diagram of H-451 for within-log property measurements: center section showing specimen locations	A-4
A-3.	Sampling diagram of H-451 for within-log property measurements: edge section showing specimen locations	A-5
A-4.	Orientation of orthogonal axes for definition of Poisson's ratio data in Table B-4	A-6
A-5.	Locations of slabs (shaded) for statistical strength testing H-451 graphite (log 5651-90)	A-7

A-6.	Coring plan for center zone of A and B sections of slabs (see Fig. A-5)	A-8
A-7.	Coring plan for edge zone of A and B sections of slabs (see Fig. A-5)	A-9
A-8.	Sampling diagram of H-451 logs for tensile strength.	A-10
A-9.	Sampling diagram of H-451 log sample core (see Fig. A-8): axial specimens	A-11
A-10.	Sampling diagram of H-451 log slab (see Fig. A-8): radial specimens	A-12
A-11.	Coring diagram for unirradiated H-451 fatigue specimens; edge zone, from slab 2 (see Fig. A-5)	A-13
A-12.	Sampling diagram for irradiation specimens: center zone, from slab 3 (see Fig. A-1)	A-14
A-13.	Sampling diagram for irradiation specimens: outer zone, edge section from slab 3 (see Fig. A-1)	A-15
A-14.	Sampling plan of graphite log showing slab designated for oxidation tests.	A-16
A-15.	Sampling plan of H-451 showing sectioning of a slab from Fig. A-14	A-17
A-16.	Sampling diagram of H-451 graphite: center section of slab from Fig. A-15	A-18
A-17.	Sampling diagram of H-451 graphite: edge section of slab from Fig. A-15	A-19
B-1.	Fatigue test data: in air log-log plot of normalized peak stress versus number of cycles to failure with $R = -1$; lower x/y tolerance limits represent limits above which x% of all data would fall, with y% confidence; axial specimens; open circles represent run-outs (from Ref. 5-10).	B-22
B-2.	Constant life fatigue diagram (Goodman diagram) for H-451 graphite: axial direction (from Ref. 5-10).	B-23
B-3.	Dimensional change in H-451 graphite: axial direction; irradiation temperatures 875 K to 975 K and 1075 K to 1175 K (602° to 702°C and 802° to 902°C) (from Ref. 6-3)	B-24
B-4.	Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1275 K to 1375 K and 975 K to 1075 K (1002° to 1102°C and 702° to 802°C) (from Ref. 6-3)	B-25
B-5.	Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1175 K to 1275 K (902° to 1002°C) (from Ref. 6-3).	B-26

- B-6. Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1475 K to 1575 K and 1575 K to 1675 K (1202° to 1302°C and 1302° to 1402°C) (from Ref. 6-3) B-27
- B-7. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 875 K to 975 K and 1475 K to 1575 K (602° to 702°C and 1202° to 1302°C) (from Ref. 6-3) B-28
- B-8. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 975 K to 1075 K and 1275 K to 1375 K (702° to 802°C and 1002° to 1102°C) (from Ref. 6-3) B-29
- B-9. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 1175 K to 1275 K (902° to 1002°C) (from Ref. 6-3) B-30
- B-10. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 1075 K to 1175 K (802° to 902°C and 1302° to 1402°C) (from Ref. 6-3) B-31
- B-11. Dimensional changes in near-isotropic graphite irradiated at 633 K to 773 K (360° to 500°C) (data from Refs. 6-4 and 6-6). B-32
- B-12. Dimensional changes in near-isotropic graphite irradiated at 773 K to 873 K (500° to 600°C) (data from Refs. 6-4 and 6-6). B-33
- B-13. Changes in tensile strength and elastic modulus of H-451 graphite as a function of fast neutron fluences. Error bars denote $\pm$ one standard deviation (from Ref. 6-3). B-34
- B-14. Fractional changes in sonic elastic modulus of extruded near-isotropic pitch coke graphite (Dragon code No. 100) (from Ref. 5-7) B-35
- B-15. Change in the sonic elastic modulus (E) and strength (S) of near-isotropic graphite irradiated at 623 K to 973 K at (350° to 700°C) (from Ref. 6-8). B-36
- B-16. Calculated percent increases in static elastic modulus of near-isotropic graphites (solid parts of curves correspond to data in Refs. 6-6 and 6-8; broken parts are extrapolated). . . B-37
- B-17. Fatigue test data irradiated to 3.0×10^{25} N/m² ($E > 29$ fJ)_{HTGR} at 1173 K (900°C). Log-log plot of normalized peak stress versus number of cycles to failure, with stress ratio, $R = -1$. Lower x/y tolerance limits represent the limits above which x% of all data would fall, with y% confidence; axial specimens; open circles represent run-outs (from Ref. 5-10). B-38

B-18.	Percent change in thermal expansivity [295 K to 773 K (22° to 500°C)] of near-isotropic graphites as a function of fast neutron fluence: irradiation temperature 865 K to 1205 K (592° to 932°C) (from Ref. 6-3)	B-39
B-19.	Percent change in thermal expansivity [295 K to 773 K (22° to 500°C)] of near isotropic graphites as a function of fast neutron fluence: irradiation temperature 1250 K to 1705 K (592° to 932°C) (from Ref. 6-3)	B-40
B-20.	Change in thermal expansivity of near-isotropic graphite irradiated at 350° to 550°C (data from Ref. 6-4)	B-41
B-21.	Thermal conductivity at the irradiation temperature for H-451 and other near-isotropic graphites as a function of fast neutron fluence (from Ref. 6-3)	B-42
B-22.	Irradiation-induced changes in thermal resistivity of near-isotropic graphite (data points from Refs. 6-6 and 6-8; curves from Ref. 6-10).	B-43

TABLES

1-1.	Comparison of H-451 and H-327 properties	1-3
4-1.	Microstructural analysis of calcined cokes used to manufacture H-451 graphite	4-3
4-2.	H-451 lots manufactured by GLCC and log distributions during development program	4-5
4-3.	Programs utilizing H-451 graphite logs	4-7
5-1.	Summary of boron equivalent test data on H-451, lot 426, log 6484-34	5-8
5-2.	Impurity content: H-451, lot 426	5-12
5-3.	Summary of lithium analyses: H-451	5-13
5-4.	Impurity content: H-451	5-14
5-5.	Statistical analyses of GA and GLCC axial strength data.	5-19
5-6.	Statistical analysis of radial strength data	5-20
5-7.	Components of variance: axial strength data	5-21
5-8.	Criteria for assigning logs to strength categories A, B, or C (axial data)	5-24
5-9.	Yields of logs in each strength category (based on axial strengths)	5-25
5-10.	Statistical analyses of fatigue data for unirradiated H-451 graphite	5-30

6-1.	Within-log variation in irradiation-induced shrinkage of H-451 graphite, Lot 426	6-8
6-2.	Between-log variations in irradiation-induced shrinkage of H-451 graphite, Lot 426	6-8
6-3.	Between-log variations in irradiation-induced strength increase of H-451 graphite, Lot 426	6-12
6-4.	Within-log variations in irradiation-induced strength increase of H-451 graphite, axial direction	6-12
6-5.	Results of statistical analysis of fatigue data for H-451 graphite irradiated at 1173 to 1263 K (900° to 990°C) (stress ratio, R = -1)	6-14
6-6.	Increase in thermal expansivity of near-isotropic graphite irradiated between 623 and 823 K (350° and 550°C)	6-18
7-1.	Experimentally determined Langmuir-Hinshelwood constants for H-451 graphite.	7-4
7-2.	Summary of UTS and elastic modulus data and comparison of population means of nonoxidized and oxidized specimens.	7-11
7-3.	Reduction in ultimate tensile strength and elastic modulus with oxidation	7-12
B-1.	Bulk density: H-451, Lot 426	B-3
B-2.	Ultimate tensile strength: H-451, Lot 426	B-4
B-3.	Elastic modulus in tension: H-451, Lot 426	B-5
B-4.	Poisson's ratio: H-451, Lot 266	B-6
B-5.	Flexural strength: H-451, Lot 426	B-7
B-6.	Ultimate compressive strength and elastic modulus in compression: H-451, Lot 478.	B-8
B-7.	Thermal expansivity: H-451, Lot 426	B-9
B-8.	Anisotropy factor: H-451, Lot 426	B-10
B-9.	Thermal conductivity: H-451, Lot 426, MLC	B-11
B-10.	Mean tensile strength of unirradiated H-451 graphite specimens used in fatigue study	B-11
B-11.	Irradiation conditions and mean tensile strength of irradiated H-451 graphite used in fatigue study (all specimens MLC location)	B-12
B-12.	Summary of irradiation-induced changes in thermal expansivity of H-451 graphite irradiated in capsules OG-1, OG-2, and OG-3	B-13
B-13.	Summary of irradiation-induced changes in thermal expansivity of H-429 graphite irradiated in capsules OG-1, OG-2, and OG-3	B-14

B-14.	Summary of irradiation-induced changes in thermal expansivity of TS-1240 graphite irradiated in capsules OG-2 and OG-3	B-15
B-15.	Summary of irradiation-induced changes in thermal expansivity of S0818 graphite irradiated in capsule OG-3	B-16
B-16.	Summary of irradiation-induced changes in thermal conductivity of H-451 graphite irradiated in capsules OG-1, OG-2, and OG-3 (all specimens from MLC of parent log)	B-17
B-17.	Summary of irradiation-induced changes in thermal conductivity of TS-1240 graphite irradiated in capsules OG-2 and OG-3 (all specimens from MLC of parent log)	B-18
B-18.	Summary of irradiation-induced changes in thermal conductivity of S0818 graphite irradiated in capsule OG-3 (all specimens from MLC of parent log).	B-19
B-19.	Calculated thermal conductivity at irradiation temperature of near-isotropic graphite irradiated between 573 and 873 K (300° and 600°C)	B-20
B-20.	Allocation of graphite samples for oxidation studies (number of specimens)	B-21

1. SUMMARY AND CONCLUSIONS

1.1. INTRODUCTION

Grade H-451 graphite, developed exclusively by the Great Lakes Carbon Corporation (GLCC) for HTGR fuel element blocks, has been evaluated extensively in preparation for its introduction into the high-temperature gas-cooled reactor (HTGR). This report summarizes the data on grade H-451 and data from the literature on graphites similar to H-451 that were used as a basis for preparing the Graphite Fuel Element section of the Fuel Design Data Manual (GA-A14429).

The five major sections of this report describe: (1) the concept and need for a near-isotropic fuel element graphite, (2) the development program at GLCC, (3) the characterization program, (4) the irradiation program, and (5) the oxidation program. Appendix A contains sampling plans for the program, and data obtained are presented in the figures and tables of Appendix B. This report is supplemental to the Materials Specification for Replaceable Fuel and Reflector Element Graphites (Number 900739) and the Graphite Fuel Element section of the Fuel Design Data Manual (GA-A14429).

1.2. CONCEPT AND NEED FOR A NEAR-ISOTROPIC FUEL ELEMENT GRAPHITE

The primary incentive for developing a near-isotropic graphite to replace needle-coke graphite grade H-327 for HTGR fuel elements was to improve dimensional stability, strength, and thermal conductivity and thus improve performance, safety margins in FSV, and flexibility in design for future HTGRs.

The data in Table 1-1 show a comparison of properties between grades H-451 and H-327 in the unirradiated state and after irradiation to end-of-life HTGR fluences. The major improvement is in the radial strength and thermal conductivity in the irradiated state. The thermal expansivity of H-451 is higher than that of H-327 in the unirradiated state, but the thermal expansivity of H-451 decreases with irradiation whereas it remains unchanged for H-327. Grade H-327 has a lower radial elastic modulus than H-451 but H-327 has a higher modulus in the axial direction. A comparison of dimensional changes is shown in Fig. 1-1. Grade H-451 has a lower anisotropy factor for dimensional change, and radial expansion is delayed to beyond the end-of-life HTGR fluences at peak HTGR temperatures.

In addition to the improved properties and irradiation behavior, the near-isotropy in properties and dimensional changes greatly simplifies design calculations over the more anisotropic needle-coke grade H-327.

1.3. DEVELOPMENT PROGRAM AT GLCC

GLCC developed two similar near-isotropic petroleum cokes, designated A and C, from two sources for production of near-isotropic graphites. The cokes were examined by metallographic techniques at GA and judged to have satisfactory microstructures for fuel element graphites. A prototype grade H-429 was prepared with coke A and submitted for early irradiation experiments. Cokes A and C were blended with coke B, a regular production coke, to develop grade H-451. A number of preproduction lots were manufactured, using cokes A₁ through A₃ in combination with cokes B₁ through B₃, and evaluated at GA. Preproduction lot 426, manufactured with cokes A₃ and B₂, was chosen for production. Approximately 98 H-451 production logs were evaluated for strength, and about 300 H-451 logs have been dispersed for various studies on programs at GA, ORNL, BNL, and the European HTGR programs.

TABLE 1-1
COMPARISON OF H-451 AND H-327 PROPERTIES

Property	Units	Unirradiated		Irradiated (a)			
				Temperature, K (°C)			
				1073 (800)		1273 (1000)	
		H-451	H-327	H-451	H-327	H-451	H-327
Ultimate tensile strength,	MPa						
Axial		12.8	11.2	23.0	17.5	21.5	16.7
Radial		12.0	6.4	21.5	8.4	20.2	8.1
Elastic modulus,	GPa						
Axial		7.9	10.3	19.7	20.1	17.8	18.6
Radial		6.9	4.0	17.2	7.8	15.5	7.2
Thermal conductivity,	W/m·K						
Axial		65	73	39	37	41	41
Radial		58	51	34	28	37	29
Thermal expansivity	K ⁻¹ (°C ⁻¹)						
α x 10 ⁶							
[295 to 1273 K (22° to 1000°C)]		4.4	2.0	3.4	1.9	2.7	1.8
Axial		5.2	3.7	4.0	3.6	3.1	3.4
Radial							

(a) End-of-life fluence for HTGR: $8 \times 10^{25} \text{ N/m}^2$ (E > 29 fJ)_{HTGR}

Fig. 1-1. Comparison of dimensional changes for H-451 and H-327 graphites: (a) axial, (b) radial.

1.4. CHARACTERIZATION PROGRAM

1.4.1. Distribution of Properties Within Individual Logs and Between Logs Within a Lot

A number of H-451 production lots were evaluated to determine the distribution of properties within single logs and between logs in a single lot. Properties measured were bulk density, mechanical properties including strength, elastic modulus, strain-at-fracture, and Poisson's ratio in tension, flexural strength, strength and elastic modulus in compression, thermal expansivity, thermal conductivity, and impurity content. Measurements were made at four locations within a log: midlength center (MLC), midlength edge (MLE), end center (EC), and end edge (EE).

Mean property values along with standard deviations and distributions of the values within the logs and lots are given in Section 5. The H-451 logs are weakest at their midlength center location. This characteristic makes it mandatory to sample the H-451 graphite for acceptance strength testing at or near the midlength center of each log.

The radial strength of H-451 is about 80% greater than that of H-327 (the needle-coke graphite used to manufacture FSV fuel elements). The thermal conductivity in the radial direction is also higher than that of H-327. These benefits resulting from the use of near-isotropic coke will improve in-service safety margins involving secondary stresses.

The impurity content was found to be highest in the center of the log; thus, testing for acceptance should parallel the procedure outlined for strength. The impurity content is a function of the degree of purification in each graphitization furnace run during manufacture. Mean ash contents were found to vary from 25 to 300 ppm depending on the graphitization furnace run.

In general, impurities that catalyze oxidation reactions, such as iron, titanium, and vanadium, follow the ash content. Lithium could not be detected in H-451 graphite at the lowest level of sensitivity of the analytical techniques used (0.005 ppm).

An analytical technique was worked out in collaboration among four laboratories, under the guidance of the ASTM Committee on Nuclear Graphite, for measuring the nonburnable boron equivalent impurity content in H-451 graphite. The results of round-robin tests provide a value of about 0.04 ppm of nonburnable boron equivalent content in a sample from a highly purified log.

1.4.2. Statistical Distribution of Strength Within a Log

A log of H-451 graphite was destructively tested to determine the systematic variation of strength within a log, the relationship between tensile and flexural strength, and the determination of a model applicable to H-451 strength distribution. The data were analyzed using Weibull's statistical model for brittle materials.

The H-451 graphite was found, as in the earlier characterization work, to be stronger in the axial direction than in the radial, and weaker along the central axis than at the edges. The variation of strength within a test zone was lowest for axial specimens. Variation in flexural strengths was less than the variation in tensile strengths.

Within experimental scatter, the data were equally well represented by normal (Gaussian) distribution or by a distribution predicted by the Weibull model. The value of the Weibull modulus, m , was lower for tensile data than for flexural data. Such a difference is contrary to predictions based on the Weibull model. Small specimens averaged 3% and 8% stronger than the larger specimens for axial and radial directions, respectively. The observed "size effect" was much lower than expected from the Weibull model,

which would predict differences of 29% and 38% for axial and radial directions, respectively. The mean flexural strength showed good correlation with the mean tensile strength of companion specimens (correlation coefficient 0.9). Flexural strength averaged 52% and 55% higher than the corresponding tensile strength for axial and radial directions, respectively. These values are in good agreement with values of 51% axial and 64% radial predicted by the Weibull model.

While there is agreement between the experimental flexural-to-tensile strength ratios and the ratios predicted by the Weibull model, the experimentally derived size effect and the difference in Weibull modulus between tensile and flexural data are inconsistent with the Weibull model. Thus it was concluded that the H-451 graphite log tested did not meet the criteria which would permit it to be treated as a Weibull solid.

1.4.3. Definition of Minimum Strength and Strength Distribution Among Logs

Tensile strength was measured on eight axial specimens taken from the midlength center from each of 98 production logs which were taken from three production lots. The axial strengths showed considerable lot-to-lot and log-to-log variations.

Acceptance criteria were derived for assigning logs to a minimum strength category based on axial tensile tests, if each log is sampled. A statistical model incorporating a separate lot-to-lot variance, log-to-log variance, and within-log variance was adopted for this purpose.

Acceptance criteria which can be used for assigning a lot to strength category A (minimum strength 10.3 MPa), B (minimum strength 8.3 MPa), or C (minimum strength 5.5 MPa) were derived. Calculations were made for either four or eight replicate specimens per log, and two alternative definitions of "minimum strength" were considered. The first definition would require

90% of the material at the midlength center of the log to exceed the specified minimum strength, with 90% confidence (90/90); the second would require 99% of the material to exceed the specified minimum, with 95% confidence (99/95).

Application of these criteria to the axial test data from the 98 logs provided information for acceptance/rejection of the logs, for assigning each log to a strength category, and subsequently for assigning each log to a specific location within the reactor core, i.e., strong logs to positions of high stress and weak logs to positions of low stress. It also provided a criterion for rejecting weak or defective logs during quality testing at a confidence level to be established by the designers.

Analysis of the data, using the acceptance criteria, showed that 5 logs would be rejected for a 4-specimen test at 90/90 confidence level whereas 18 logs would be rejected for an 8-specimen test at 99/95 confidence level. When the analysis was applied to specific lots the results showed that most of the rejected logs were in one weak lot which had a mean strength significantly below the other two.

1.4.4. Fatigue Behavior

Grade H-451 graphite was tested to determine its fatigue life. Fatigue tests were run over a range from compression/tension cycling (R values -1 and -0.5) and in tension (R values 0 and 0.5).

The H-451 graphite showed cyclic fatigue life curves (S-N curves) generally resembling those of metals, but with more scatter. For reversed stress cycling ($R = -1$), the homologous stress limits (maximum applied fatigue stress divided by the tensile strength) for 50% specimen survival to 10^5 cycles averaged 0.63 in the axial direction and 0.74 in the radial direction. Corresponding homologous stress limits for 99% specimen survival (99/95 tolerance limits) were 0.48 and 0.53. The fatigue life and the homologous stress limits increased as R increased.

1.5. IRRADIATION PROGRAM

1.5.1. Dimensional Changes

Dimensional changes were measured from 873 to 1623 K (600° to 1350°C) to $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} in a series of capsules irradiated in the ORR. Additional data at 573 to 873 K (300° to 600°C) on graphites similar to H-451 were taken from the literature. The dimensional data cover the temperature and fluence range of the HTGR.

The dimensional changes of H-451 graphites at 573 to 1623 K (300° to 1350°C) fall within an envelope of 0% and -3% in the axial direction and 0% and -1.5% in the radial direction up to $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR}, which is about 10% above the lifetime fluence of the H-451 graphite in an HTGR. When the irradiation temperature was increased after an initial irradiation period, the shrinkage rate increased to that of the higher temperature. When the irradiation temperature was reduced after an initial irradiation period, the shrinkage rate also was reduced, but remained higher than the isothermal rate at the lower temperature.

Differences in shrinkage between MLC and MLE specimens from the same log were not statistically significant, nor were there statistically significant differences in shrinkage among specimens taken from the same location in the log but from different logs.

1.5.2. Tensile Strength and Elastic Modulus

Tensile strength increased and elastic modulus increased during irradiation at 823 to 1623 K (550° to 1350°C). There was an increase in the

standard deviation of strength values but no change in the coefficient of variation. The increase in strength is related to the increase in elastic modulus by

$$\frac{S}{S_o} = \left(\frac{E}{E_o} \right)^{0.64} \quad (1-1)$$

Within-log variation and log-to-log variation of fractional strength increase were not significant.

1.5.3. Fatigue Behavior

Irradiation increased the homologous stress limits for fatigue endurance of H-451 by about a factor of 2 when the stresses were normalized by dividing by the unirradiated strength.

1.5.4. Thermal Expansivity

Irradiation induced an increase of about 20% in thermal expansivity of H-451 at 623 to 823 K (350° to 550°C) over the life of the fuel element. The thermal expansivity increased by about 10% at 873 to 973 K (600° to 700°C) at low fluences and returned to the original values at HTGR end-of-life. At 923 to 1673 K (700° to 1400°C) the thermal expansivity decreased 20% to 50% over the life of the fuel element.

1.5.5. Thermal Conductivity

Changes in thermal conductivity of H-451 graphite due to irradiation were measured at 923 to 1623 K (650° to 1350°C) to HTGR end-of-life fluences, and data at 523 to 923 K (250° to 650°C) were obtained from UKAEA data on graphites similar to H-451. Thermal conductivity decreased rapidly at fluences of 0 to $2 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} and then showed

only a small decrease between 2 and $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR}. Irradiation-induced reduction in thermal conductivity was temperature-dependent, with the largest reduction occurring at the lowest irradiation temperatures.

A method is described for calculating the irradiated thermal conductivities at the temperature of irradiation. When the irradiation temperature was either increased or decreased after an initial period, the new conductivities assumed the saturation level of the new irradiation temperature.

1.5.6. Irradiation-Induced Creep

A systematic program designed to measure irradiation-induced creep coefficients of H-451 graphite is in progress at ORNL, but only one capsule has been completed to date. Therefore, the creep coefficients have been derived from the literature.

A wide variety of graphites have been measured in a variety of experiments during the past 20 years. A few H-451 specimens were included in experiments at RCN Petten in the Netherlands, where a number of graphite grades were measured in a restrained shrinkage experiment; thus, a few H-451 data points were available for comparison. RCN and JRC Euratom, Petten, have completed the first phase of an experiment where the creep coefficient of an H-451 specimen was measured in tension. In this experiment, which was taken to about $3.3 \times 10^{25} \text{ N/m}^2$ [$(E > 29 \text{ fJ})$ _{HTGR} at 1123 K (850°C)], both the transient and steady-state creep coefficient were measured.

An equation for calculating the creep strain in design has been derived that covers the range 773 to 1473 K (500° to 1200°C) up to 10^{26} N/m^2 ($E > 29 \text{ fJ}$)_{HTGR} and creep strains up to 2.5% in tension and 2% in compression.

1.6. OXIDATION PROGRAM

1.6.1 Oxidation Rate of H-451 Graphite

The chemical reactions of water vapor with H-451 are represented by the Langmuir-Hinshelwood equation. Rate constants were derived to satisfy the Langmuir-Hinshelwood equation for low water concentrations (≤ 300 Pa) and high water concentrations (≥ 300 or ≤ 3300 Pa)

The OXIDE computer code is used to determine the oxidation rates for reactor service. Rate constants derived for H-451 by the Langmuir-Hinshelwood equation are used as input for the OXIDE calculations. High water level rate constants have been derived for accident conditions, where high moisture levels are possible. Low water level rate constants have been derived and should be used for normal steady-state reactor operation, where water concentrations in the range 10 to 1000 Pa are expected.

1.6.2. Effect of Oxidation on Tensile Strength and Elastic Modulus

A large number of small specimens taken from a parent log of H-451 graphite were oxidized to burnoffs up to 20%, and the changes in tensile strength and elastic modulus were determined.

The tensile strength and elastic modulus of H-451 graphite were decreased by an average of 3.6% in center axial specimens when oxidized to burnoffs up to 2.0% at either 1073 K or 1273 K (800° or 1000°C), and no significant reduction in elastic modulus in these same specimens was observed when they were oxidized to burnoffs of up to 2.0% at 1273 K (1000°C). These results are of particular importance because the maximum localized burnoff in the HTGR fuel element blocks has been calculated to be 1%.

The rate of reduction of tensile strength and elastic modulus for center axial specimens is greater at 1073 K (800°C) than at 1273 K (1000°C). This conclusion is not important for design because the maximum burnoff of the fuel element blocks in an HTGR is calculated to occur at approximately 1273 K (1000°C). About 0.1% to 0.2% burnoff is calculated for fuel element blocks operating at 1073 K (800°C).

In general, the reduction rate of elastic modulus with burnoff was greater than that of ultimate tensile strength (UTS). The higher reduction rate of the elastic modulus is important in design because a lower elastic modulus decreases the secondary stresses, i.e., thermal stresses resulting from gradients and differential irradiation-induced strain. The rate of reduction of tensile strength and elastic modulus for axial specimens oxidized at 1273 K (1000°C) is greater at the edge than at the center of a log. This does not impose a limitation in design because the tensile strength for acceptance of a log for fuel element blocks is always measured at the center of the log where it is a minimum. The tensile strength at the edge of a log oxidized up to 2% burnoff is never less than the tensile strength at the center at 0 to 2% burnoff. In general, the reduction in tensile strength and elastic modulus of radial specimens was higher than that of axial specimens. The higher seismic-induced stresses in an HTGR fuel element are in the radial direction and this phenomenon could affect design. However, the reduction in elastic modulus of the radial specimens was greater than the strength loss, and, thus, the overall effect on seismic performance should be negligible.

2. INTRODUCTION

Grade H-451 near-isotropic graphite is the reference candidate graphite for replaceable fuel and reflector elements in HTGRs. Grade H-451 graphite has been under development at GLCC for approximately 7 years. Concurrent with GLCC's development program, an intensive evaluation program has been carried out on H-451 at GA. The raw material type, formulation, and processing have been frozen, and several production lots have been manufactured and evaluated.

The data generated from the evaluation programs have been published in a series of GA reports. The purpose of this report is to collect, analyze, and sort the data from the GA reports, including data on graphites similar to H-451 from other U.S. or European programs and the open literature, and to present the total available data set on H-451 graphite for use in preparing the H-451 section of the Fuel Design Data Manual (GA-A14429), which is required for design and safety analysis of HTGRs. The following topics are discussed:

Concept and Need for a Near-Isotropic Fuel Element Graphite

Section 3 explains the logic and need for developing a near-isotropic graphite in the U.S. for HTGR fuel elements.

Development Program at GLCC

Section 4 describes the scope of the H-451 development program at GLCC.

Characterization Program

In Section 5, the characterization program is described. An extensive characterization program was required to (1) confirm that a suitable material was available for use in the core of HTGRs and (2) provide a data set on properties, purity, and fatigue. The characterization program was designed to evaluate the various development products from the GLCC program and select a formulation and process for production. The quality of the selected grade was established by determining the properties and distribution of properties representative of the grade. A materials specification has been written that defines requirements for fuel element graphite in HTGRs and it will be applied to ensure that future H-451 production meets the quality required.

Irradiation Program

Section 6 discusses the irradiation program, which was designed to provide the mean and distribution of properties of H-451 that are representative of the material in the fast neutron flux present during the service life of the fuel elements. These data are used directly to design the fuel elements and to calculate the thermal and stress gradients that occur in the fuel element graphite during its life in the reactor core. Tests on the filler cokes have been included in the materials specification to aid in standardizing the quality of the raw material and ensure that future H-451 production will exhibit the same irradiation behavior as that of the preproduction logs used to obtain the irradiation data set presented here.

Oxidation Program

The oxidation program is discussed in Section 7. The H-451 fuel element blocks may be exposed to steam oxidation during their life in the reactor; therefore, it is necessary to determine the effect of oxidation on their properties. The oxidation program was designed to

determine the rate of oxidation in H_2O -He mixtures and the effect of oxidation burnoff on mechanical properties, primarily strength and elastic modulus. The oxidation data are required for design and safety analysis.

Two documents are closely related to this report. One is the Materials Specification for Replaceable Fuel and Reflector Element Graphites (Number 900739), which contains raw material, processing, and property limits that must be met by the manufacturer to qualify H-451 for acceptance and subsequent use in HTGRs. The objective of the materials specification is to prevent the inclusion of defective or out-of-specification logs in the manufacture of fuel elements or replaceable reflectors. The specification also requires testing to ensure uniformity in raw materials from batch to batch over a long period, thus ensuring properties and irradiation behavior consistent with those of the materials utilized in the current evaluation programs.

The other document to be used with this report is the H-451 graphite section of the Fuel Design Data Manual (GA-A14429), which contains H-451 property data derived from this report, other domestic and European graphite programs, and the open literature. The material in the Fuel Design Data Manual is presented in a form suitable for use for the design and safety analyses of HTGR fuel elements.

3. CONCEPT AND NEED FOR A NEAR-ISOTROPIC FUEL ELEMENT GRAPHITE

The primary incentive for developing a near-isotropic graphite in the U.S. for HTGR fuel elements was to improve the dimensional stability and strength over the conventional HTGR needle-coke graphite (grade H-327). The development of H-451 was aimed at providing more flexibility in design, better safety margins for stresses, and stretch capability for future designs. The isotropy imparted to the graphitized logs by the use of near-isotropic petroleum coke was aimed at minimizing the dimensional contraction and delaying the subsequent expansion to fluences well beyond the service life in an HTGR. An additional goal of the program was to increase the thermal conductivity of the graphite, especially in the radial direction which is parallel to the thickness of the thin ligaments that separate the fuel and the coolant in the fuel element blocks.

4. DEVELOPMENT PROGRAM AT GLCC

Based on the incentives described in Section 3, the development of a near-isotropic graphite was initiated in 1969. To minimize the impact of development costs and to keep the price of the new product competitive, a decision was taken jointly by GA and GLCC to utilize commercially available materials with conventional extrusion and processing as in the manufacture of H-327, while at the same time emphasizing improved isotropy in the filler coke structure. Thus, the development work and future production could be accomplished employing existing raw materials, equipment, and plants.

The structure of Gilsocoke was used as a guide in procuring a new petroleum-based, special, near-isotropic coke for the GLCC development program. Gilsocoke was used in Europe to manufacture graphites for the UK Advanced Graphite Reactors. Graphites manufactured with Gilsocoke have shown satisfactory irradiation behavior. However, Gilsocoke is a single-source raw material that is no longer commercially available. Also, Gilso-coke presented difficulties when used to fabricate large-diameter logs, imparting a high degree of hardness to the graphitized products.

4.1. FILLER COKES

GLCC procured a special batch of near-isotropic coke and designated it coke A₁. Subsequent batches from supplier A were procured and are designated A₂ and A₃.

A commercial coke that is used routinely in the production of other commercial graphite articles in GLCC plants was also considered for blending with Coke A. This coke is a special type of sponge coke and has a structure that is less isotropic than coke A, but it is not a needle coke.

The second coke, designated B₁, was considered for blending with coke A₁ to improve the manufacturing characteristics and the microstructure of the final product.

A second special near-isotropic coke was procured from a second supplier. This coke, designated C, was produced from the same generic type feedstock, the aim being to establish a coke similar to coke A but from a different supplier. This coke was used along with coke B₁ to make prototype logs during later stages of the development program.

The microstructures of the calcined cokes described above were classified at GA using metallographic techniques described by White and Price (Ref. 4-1). The results are given in Table 4-1. Coke A₁, which was used in grade H-429 and the early prototype H-451 logs, consisted of only 65% of the desirable heterogeneous fine isotropic (HFI) material, whereas cokes A₂ and A₃ consisted of 90% or more of the HFI material. Coke C₁ is comparable to cokes A₂ and A₃ in microstructure. Coke B₁ is classified as a regular or sponge coke; the analysis shows a range of constituents from HFI to coarse fibrous (CF) constituents. None of the cokes used in H-451 development and subsequent production logs contained fine fibrous (FF) constituents, and they contain only small amounts of CF material. However, there is some degree of anisotropy in the final graphitized product due to the small content of oriented constituents in the filler cokes.

Cokes that have near-perfect isotropy were not considered because they impart high thermal expansivity to the graphitized product and are even more difficult to process into large-diameter logs with conventional equipment. Needle-like constituents, on the other hand, impart low strength and anisotropic dimensional changes during service in a fast neutron environment. Thus, the near isotropic petroleum cokes used in H-451 appear to be optimum for HTGR fuel element graphites.

TABLE 4-1
MICROSTRUCTURAL ANALYSIS OF CALCINED COKES
USED TO MANUFACTURE H-451 GRAPHITE

Coke No.	Coke Designation	Percent of Constituent (a)				
		HFI	FI	CI	CF	FF
5361-60	A ₁	65	0	15	20	0
5361-61	A ₂	95	0	5	0	0
5361-65	A ₃	97	0	3	0	0
5361-62	B ₁	52	7	12	29	0
5361-67	C ₁	90	0	4	6	0

(a) Descriptions from Ref. 4-1:
HFI = heterogeneous fine isotropic
FI = fine isotropic
CI = coarse isotropic
CF = coarse fibrous
FF = fine fibrous

4.2. PROCESSING

Details of the manufacturing processes of H-451 are proprietary to GLCC. In general, H-451 is produced as logs 432 mm in diameter by 864 mm in length using conventional graphite manufacturing processes common to the manufacture of electric steel furnace electrodes. The calcined coke is crushed and sized and a specific formulation selected consisting of multi-sized fractions of the coke particles, with coal tar pitch as the binder. This formulation of calcined cokes and coal tar pitch is mixed thoroughly in the range 393 to 453 K (120° to 180°C), extruded into green logs, and baked slowly to about 1073 K (800°C) to carbonize the binder pitch. The baked logs are then impregnated with petroleum pitch and the logs heated to 2973 K (2700°C) or above to graphitize the structure. The purity of the logs is improved by conducting the graphitization process in the presence of chlorine gas. The resultant product is a high purity, near-isotropic, medium-grained nuclear graphite suitable for use in an HTGR core.

4.3. PROTOTYPE AND PREPRODUCTION DEVELOPMENT

Grade H-429 was formulated with Coke A₁ and extruded in the GLCC pilot plant as a 52-mm-diameter log. Grade H-429 was committed to early irradiation testing by GA during 1970-1972. Concurrent with the irradiation of H-429, GLCC manufactured a number of full-size experimental grades. Grade H-451 (lot 266), initially fabricated with coke A₁, was selected jointly by GA and GLCC as the most promising material for further development. A chronology of the development program is given in Table 4-2. In subsequent preproduction development, lots 408, 424, and 426 were fabricated using a blend of either cokes A₂ and B₁ or A₃ and B₂. Two lots were produced using a blend of cokes C₁ and B₃. Logs from lots 266, 408, and 426 were incorporated into GA's characterization and irradiation programs.

In 1975, the formulation and processing methods of lot 426 were jointly selected by GLCC and GA as models for the initial production of 350 logs of

TABLE 4-2
H-451 LOTS MANUFACTURED BY GLCC AND DISTRIBUTION OF LOGS DURING DEVELOPMENT PROGRAM

	GLCC Identification			Filler Coke Designation	No. Logs Produced	Number of Logs Evaluated										Remarks
	Extrusion Lot No.	Furnace No.	Graphi- tization No.			Characterization Program	Irradiation Program	Fuel Element Development	Oxidation Studies	Recycle Fuel Studies	Foreign Program	ORNL Program	BNL Program	FSV Fuel Element Test Program	Large Scale Mechanical Tests	
Prototype program	H-429 ^(a)	(b)	(b)	A ₁	1	1	1	--	--	--	--	--	--	--	--	
Preproduction program	266	(b)	(b)	A ₁	3	2	1	--	--	--	--	--	--	--	--	
	408	(b)	(b)	A ₂ + B ₁	40	5	1	6	2	--	--	5	--	--	4	
	410	(b)	(b)	A ₂ + B ₁	5	--	--	--	--	5	--	--	--	--	--	
	424	44	5698-C, 6399-C	A ₃ + B ₂	12	--	--	1	--	6	--	--	--	--	2	
	426	44	5698-C	A ₃ + B ₂	45	--	--	28	--	--	2	--	--	--	1	
	426	77	6003-C	A ₃ + B ₂	51	4	3	6	4	--	11	--	1	--	19	Chosen for production
	426	79	6399-C	A ₃ + B ₂	31	--	--	--	--	--	5	--	--	17	6	
	438	79	6399-C	C ₁ + B ₃	27	--	--	--	--	10	--	--	--	--	--	
	440	79	6399-C	C ₁ + B ₃	23	4	2	--	--	--	7	--	--	--	--	
Production program	472	51	1272	A ₃ + B ₅	63	--	--	--	--	6	2	--	--	2	--	Extensive tensile testing
	474	51	1272	A ₃ + B ₅	166	--	--	--	--	--	--	--	--	--	--	
	476	51	1272	A ₃ + B ₅	24	--	--	--	--	--	--	--	--	--	--	
	478	51	1272	A ₃ + B ₅	63	--	--	4	--	23	2	7	4	6	--	Extensive tensile testing
	480	51	1272	A ₃ + B ₅	15	--	--	--	--	--	--	--	--	--	--	
	482	51	1272	A ₃ + B ₅	124	--	--	2	--	51	2	--	--	6	--	Extensive tensile testing

(a) Prototype grade number, not an extrusion lot number.

(b) Not available.

H-451 for possible use in FSV as reload elements. Production lots 472, 474, 476, 478, and 482 were produced for the Fort St. Vrain reload segments using a blend of coke A₃ and B₁ and the same formulation and processing as for preproduction lot 426. Ninety-eight logs from production lots 472, 478, and 482 were sampled at the midlength center (MLC) of each log, and tensile strength was measured on eight axial and four radial specimens from each.

During the development program a number of the logs from each lot were distributed to evaluation programs within and external to GA. The programs utilizing H-451 graphite are given in Table 4-3. The use of H-451 preproduction logs for experiments involving fuel development, oxidation, recycle, FSV fuel test elements, large-scale mechanical testing, foreign programs, ORNL programs, and BNL programs occurred concurrently with the GLCC development program and the characterization and irradiation programs at GA. Thus, selection of material for each program depended on the availability of logs at the time of their need; i.e., evaluation did not wait until the development program was complete.

4.4. CONCLUSIONS

GLCC developed proprietary sources of near-isotropic cokes for use in developing grade H-451 as a candidate for advanced HTGR fuel elements. Grade H-451 was developed using conventional graphite processing and a blend of a near-isotropic coke and a special sponge coke. A number of preproduction lots and one production batch were manufactured for evaluation by GA and other reactor programs in the U.S. and Europe.

REFERENCE

- 4-1. White, J. L., and R. J. Price, "The Formation of Mesophase Microstructures During the Pyrolysis of Selected Coker Feedstocks," Carbon 12, 321 (1974).

TABLE 4-3
PROGRAMS UTILIZING H-451 GRAPHITE LOGS

Program	Use or Measurements
Characterization	Measurement of physical, mechanical, and chemical properties on small specimens cut from full-size logs. Variations of properties within logs, from log-to-log within lots, and from lot-to-lot were investigated. Fatigue characteristics were determined.
Irradiation	Same as above except fewer specimens measured due to limited resources and high cost of irradiation experiments.
Fuel element development	Full-size machined blocks used to determine characteristics of H-451 graphite and its interaction with fuel sticks during carbonization-in-place (CIP) experiments.
Oxidation studies	Measurement of reaction rates of H-451 in He-H ₂ O mixtures. Parametric studies of effect of H ₂ O and H ₂ concentration, flow rate, and temperature. Studies of the effect of oxidation on strength reduction.
Recycle fuel element studies	Full-size machined blocks used to determine effect of material on crushing, burning, and fuel separation.
Large-scale mechanical tests	Use of full-size or scaled machined blocks for seismic testing. Intermediate-size specimens used for dowel pin, thermal stress, and miscellaneous mechanical tests.
ORNL programs	Oxidation, irradiation creep, and fuel recycle tests.
BNL program	Oxidation tests.
Foreign programs: KFA, CEA, RCN Petten, and Dragon	Oxidation and irradiation tests including creep, in-reactor thermal stress tests, full-size machined elements for strength proof testing, and machining tests.

5. CHARACTERIZATION PROGRAM

5.1. INTRODUCTION

Characterization of grade H-451 includes determination of: (1) distribution of properties within individual logs and between logs within a lot, (2) statistical distribution of strength within a log, (3) strength distribution among logs, and (4) fatigue behavior.

5.2. DISTRIBUTION OF PROPERTIES WITHIN INDIVIDUAL LOGS AND BETWEEN LOGS WITHIN A LOT

5.2.1. Sampling

Sampling diagrams are shown in Appendix A. Properties were measured on specimens taken parallel to the extrusion direction (axial)* and perpendicular to the extrusion direction (radial)*. In the experiments to determine property distributions within a log, the logs were sampled at four different locations within each log: midlength center (MLC), end center (EC), midlength edge (MLE), and end edge (EE). Properties measured on logs in the unirradiated state were (1) bulk density, (2) tensile properties, including strength, elastic modulus, strain at fracture, and Poisson's ratio in tension, (3) flexural strength, (4) strength and elastic modulus in compression, (5) thermal expansivity, (6) thermal conductivity, and (7) impurity content.

*The terms axial and radial are used throughout this report to designate specimens whose longitudinal axis (axis along which measurement was made) was parallel to the longitudinal axis of the parent log and parallel to the radius of the parent log, respectively.

5.2.2. Bulk Density

The bulk density of graphite is defined as the weight of the log divided by the bulk volume (outer dimensions of the log). For characterization, the bulk density was determined on the tensile strength specimens prior to their use in strength tests. Density measurements were also made on whole as-graphitized logs at the GLCC production plant. Production acceptance will be based on whole log densities measured at GLCC.

The densities of four logs from preproduction lot 426 are presented in Appendix B, Table B-1 (Ref. 5-1). The data include measured and calculated whole log density values. The latter were obtained by averaging the mean values from the center and edge specimens. The bulk densities measured on whole logs in the manufacturer's plant were about 0.01 Mg/m^3 higher than those measured at GA on small specimens located throughout the log. There was a small but significant increase in density from the center to the edge of some logs. However, several logs had no gradient. The density was, in general, lowest at the MLC position and increased slightly near the edges and ends of the logs. Again a few exceptions were observed. The variation in density for H-451, lot 426, ranged from 1.695 to 1.753 Mg/m^3 .

The mean of the small-specimen measurements agrees well with that of measurements by GLCC on whole logs. The mean bulk density for the four logs from lot 426 was $1.73 \pm 0.01 \text{ Mg/m}^3$. Additional whole log density values were obtained on 98 logs from production lots 472, 478, and 482. The mean bulk density of these logs was $1.74 \pm 0.025 \text{ Mg/m}^3$.

5.2.3. Mechanical Properties

The mechanical properties measured were ultimate tensile strength, elastic modulus in tension, strain at fracture in tension, Poisson's ratio in tension, ultimate flexural strength, ultimate compressive strength, elastic modulus in compression, and fatigue behavior.

5.2.3.1. Tensile Strength, Elastic Modulus, and Strain at Fracture. The ultimate tensile strength data for preproduction lot 426 are summarized in Appendix B, Table B-2 (Ref. 5-1). The tensile strength of H-451 is highest in the axial direction, lowest at MLC for axial and radial directions, and increases from the center to the edge. The mean tensile strength of H-451, lot 426, ranged from 13.65 ± 1.60 MPa at MLC to 19.00 ± 1.85 MPa at MLE in the axial direction and from 10.75 ± 2.10 MPa at MLC to 15.22 ± 2.25 MPa at EE in the radial direction.

The observed pattern of lowest axial or radial strength at the MLC of each log makes it mandatory to test each log as near the MLC as possible during acceptance/rejection testing, as required by Specification 900739 (within slab 3, section AC/BC; see Appendix A, Fig. A-1). This ensures that each log is tested at its weakest location. Testing specimens of either axial or radial direction is acceptable. Variations in tensile strength were observed from lot to lot at the same location in each log (see Section 5.4).

Data on elastic modulus measured in tension for preproduction lot 426 are given in Appendix B, Table B-3 (Ref. 5-1). The elastic modulus followed the same pattern within logs as the tensile strength. The mean modulus of H-451, lot 426, ranged from 7.93 GPa at MLC to 8.82 GPa at MLE in the axial direction and from 6.89 GPa at MLC to 7.45 GPa at EE in the radial direction.

The strain at fracture for H-451 was in the range 0.25 to 0.35% (Ref. 5-1). The strain at fracture was lowest for both axial and radial specimens at the MLC of all logs and increased from the center to the edges. A systematic variation in the strain at fracture from log to log or lot to lot was not observed in H-451.

5.2.3.2. Poisson's Ratio. Poisson's ratio data are summarized in Appendix B, Table B-4 (Ref. 5-1). Poisson's ratio values, $\bar{\nu}_{ij}$, are identified in standard tensor notation according to the orthogonal axes (Ref. 5-2) within a log (see Appendix A, Fig. A-4). Poisson's ratio was measured on H-451 in the axial and radial directions at several locations within a log.

Poisson's ratio decreased with increasing stress during initial loading and reached a constant value, independent of stress, after two or three loadings. When stresses were applied in a radial direction (σ_1 or σ_2) to MLC specimens, $\bar{\nu}_{13} = \bar{\nu}_{23} = 0.110 \pm 0.009$ and $\bar{\nu}_{12} = \bar{\nu}_{21} = 0.108 \pm 0.008$. When stresses were applied in the axial direction (σ_3) to MLC specimens, $\bar{\nu}_{31} = \bar{\nu}_{32} = 0.127 \pm 0.013$. When stresses were applied in the axial direction (σ_3) to MLE specimens, $\bar{\nu}_{32} = 0.121 \pm 0.005$ and $\bar{\nu}_{31} = 0.114 \pm 0.006$. When stresses were applied in the axial direction (σ_3) to EE specimens, $\bar{\nu}_{32} = 0.110 \pm 0.010$ and $\bar{\nu}_{31} = 0.117 \pm 0.006$.

5.2.3.3. Flexural Strength. The flexural strength data for H-451 lot 426 are summarized in Appendix B, Table B-5 (Ref. 5-3). The flexural strength ranged from 20.2 to 25.3 MPa in the axial direction and from 17.4 to 21.7 MPa in the radial direction.

5.2.3.4. Compressive Strength and Elastic Modulus. Data on the compressive strength and elastic modulus are summarized in Appendix B, Table B-6. The mean ultimate compressive strength measured on a log from production lot 478 was 47 MPa in the axial and radial directions. The within-log variation was 2.5 MPa and the log-to-log variation was 1.5 MPa. Elastic modulus in compression averaged 7.6 MPa.

5.2.4. Thermal Properties

5.2.4.1. Thermal Expansivity and Anisotropy Factor. Thermal expansivity data are summarized in Appendix B, Table B-7 (Ref. 5-1). The mean thermal expansivity between 300 and 773 K (27° and 500°C) of H-451, lot 426, ranged from 3.9 to $4.1 \times 10^{-6} \text{ K}^{-1} (\text{°C}^{-1})$ in the axial direction and 4.5 to $4.6 \times 10^{-6} \text{ K}^{-1} (\text{°C}^{-1})$ in the radial direction. There was no significant variation of thermal expansivity as a function of specimen location within the logs of lot 426.

The anisotropy factor is defined as

$$\text{anisotropy factor (AF)} = \frac{\alpha(\text{radial})}{\alpha(\text{axial})} \quad (5-1)$$

where $\alpha(\text{radial})$ is the thermal expansivity in the radial direction and $\alpha(\text{axial})$ is the thermal expansivity in the axial direction. The anisotropy data are shown in Appendix B, Table B-8 (Ref. 5-1, 5-3). The mean anisotropy factor for the logs in lot 426 is 1.15.

5.2.4.2. Thermal Conductivity. Data on thermal conductivity are summarized in Appendix B, Table B-9 (Ref. 5-1). The mean thermal conductivity at the MLC of logs from lot 426 was 65 ± 4 W/m·K in the axial direction and 63 ± 4 W/m·K in the radial direction at 1073 K (800°C).

5.2.3.3. Specific Heat. The specific heat of H-451 graphite is given by the equation (Ref. 5-4)

$$\begin{aligned} C_p = & (0.54212 - 2.42667 \times 10^{-6} T - 9.02725 \times 10^1 T^{-1} \\ & - 4.34493 \times 10^4 T^{-2} + 1.59309 \times 10^7 T^{-3} \\ & - 1.43688 \times 10^9 T^{-4}) \times 4186 \end{aligned}$$

where C_p = specific heat at constant pressure (J/kg·K),
T = temperature (K).

5.2.5. Impurity Content

5.2.5.1. Neutron-Absorbing Impurities. Two types of neutron-absorbing impurity elements, burnable* and nonburnable*, are of importance to the neutronic design and operation of HTGRs. The impurity content of the non-burnable poison elements is expressed in boron equivalents:

$$\text{BE (boron equivalent)} = \frac{\bar{\sigma}_x A_B}{A_x \bar{\sigma}_B} \quad (5-2)$$

where $\bar{\sigma}_x$ = effective neutron capture cross section of impurity element x,

$\bar{\sigma}_B$ = effective neutron capture cross section of natural boron,

A_x = atomic weight of impurity element x,

A_B = atomic weight of natural boron.

The boron equivalent values for each element are then multiplied by their respective concentrations in the graphite, and the products are summed to obtain the boron equivalent level, in parts per million, for the graphite:

$$\sum_i \text{BE}_x C_x = \text{BE}_G \quad (5-3)$$

where BE_x = boron equivalent of impurity element x,

C_x = boron equivalent concentration of all impurity elements in graphite.

* Nonburnable impurity elements are those that remain in some proportion to their original content at the end-of-life for the fuel element block, whereas the burnable impurities are completely transmuted to other isotopes.

The neutronic designers of HTGRs require knowledge of the nonburnable impurities listed in Table 5-1, column 8. Those defined as burnable poisons are: boron, cadmium, europium, gadolinium, samarium, and lithium; the balance are defined as nonburnable. It is desirable to keep the total nonburnable impurity content below 0.5 ppm. However, it is a formidable and expensive task to perform chemical analyses for all required nonburnable elements and obtain the sensitivity required to detect the elements at a level satisfying the requirement of 0.5 ppm. A recommended practice for determining BE involving low-temperature ashing of the graphite and subsequent analysis of the ash by emission spectroscopy is under development by ASTM Committee C5:05, "Nuclear Applications," a subcommittee of C-5, "Industrial Carbon and Graphite." A round-robin specimen of H-451 taken from log 6484-34 of lot 426 was prepared by GA and analyzed by AirCo Speer, ORNL, BNL, and GA, who have representatives on Committee C5:05. The results of the round-robin tests are presented in Table 5-1, columns 4 through 7, with the permission of the organizations listed above.

The agreement among the four laboratories was excellent. The GA data (column 7) were obtained by emission spectroscopy on the ash of a 0.100-kg sample. The ash was prepared at about 473 to 523 K (200° to 250°C) over a 258-hr period. Low-temperature ashing prevents the evaporation of volatile impurities, thus improving the accuracy of the data. The values for each element represent, in most cases, the lower limit of detection and, in some cases, the actual value of the impurity element. Thus, the calculated boron equivalent concentration represents an upper limit of detection; i.e., the boron equivalent concentration, in most cases, is less than the value shown.

An attempt was made to select and eliminate elements that have either (1) a low boron equivalent factor (equal to or less than 0.0001), or (2) a high probability of occurring at insignificant concentrations.

Using the GA data (Table 5-1, column 7), a boron equivalent of 0.131 ppm was obtained for nonburnable elements. This excludes chlorine, which is included in column 8 but not in column 7 because no data were available.

TABLE 5-1
SUMMARY OF BORON EQUIVALENT TEST DATA ON H-451, LOT 426, LOG 6484-34

Column													
1	2	3	4	5	6	7	8	9	10	11	12	13	14
Element	Symbol	Boron Equivalent Factor	Round Robin Test Data (a) (ppm) Laboratories				Elements Requested For Analyses (b)	Element NOT Requested (b)	Analyses for Purposes Other Than Boron Equivalent (c)	Elements NOT Recommended for Analyses by ASTM Committed C5:05	Elements With Boron Equivalent < 0.0001	Elements for Boron Equivalent Analyses (d)	Boron Equivalent (ppm)
			I	II	III	IV							
Aluminum	Al	0.00012	0.3	0.45	0.2	0.3	X					X	0.000036
Antimony	Sb	0.0062	--	<0.45	--	<0.02	X					X	0.00012
Argon	A	--	--	--	--	--		X					
Arsenic	As	--	--	<0.45	--	<0.120		X					
Barium	Ba	0.00042	0.02	0.045	0.009	0.060	X					X	0.000025
Beryllium	Be	0.000016	<0.005	--	--	<0.001	X				X		0.000000016
Bismuth	Bi	0.000026	<0.01	<0.045	--	<0.006	X				X		0.000000016
Boron	B	1.0000	0.2	0.45	0.05	0.18	X					X	0.18
Bromine	Br	--	--	--	--	--		X					
Cadmium	Cd	0.855	<0.03	<4.5	0.007	<0.030	X			X		X	0.02565
Calcium	Ca	0.00020	0.2	0.45	0.2	0.30	X					X	0.00006
Carbon	C	--	--	--	--	--		X					
Cerium	Ce	0.0001	<0.2	--	--	<0.005	X				X		0.0000005
Cesium	Cs	0.0127	--	--	<0.02	<0.30	X			X			0.038
Chlorine	Cl	0.0136	--	--	--	--	X					X	
Chromium	Cr	0.00086	0.2	0.14	0.2	0.18	X					X	0.00015
Cobalt	Co	0.0114	0.02	<0.45	0.04	<0.012	X			X			0.00014
Copper	Cu	0.00099	0.07	0.14	0.001	0.030	X			X			0.000030
Dysprosium	Dy	0.1048	<0.05	<4.5	<0.018	0.000	X			X			0.0000
Erbium	Er	0.0692	<0.02	<4.5	<0.004	<0.002	X					X	0.00014
Europium	Eu	0.459	0.01	4.5	0.002	0.005	X			X			0.0023
Fluorine	F	--	--	--	--	--		X					
Gadolinium	Gd	1.57	<0.01	<4.5	<0.007	<0.005	X			X			0.0079
Gallium	Ga	--	<0.01	--	--	<0.003		X					
Germanium	Ge	--	<0.02	<0.045	--	<0.012		X					
Gold	Au	--	--	--	--	<0.018		X					
Hafnium	Hf	0.0475	<0.05	<0.45	<0.004	<0.060	X			X			0.0029
Helium	He	--	--	--	--	--		X					
Holmium	Ho	0.0222	<0.02	<4.5	<0.018	<0.002	X					X	0.000044
Hydrogen	H	--	--	--	--	--		X					
Indium	In	0.135	--	--	<0.007	<0.003	X			X			0.00041
Iodine	I	--	--	--	--	--		X					
Iridium	Ir	--	--	--	--	<0.030		X					
Iron	Fe	0.00068	4.0	2.2	2.6	1.8	X		X				0.0012
Krypton	Kr	--	--	--	--	--		X					
Lanthanum	La	0.00114	<0.01	<4.5	--	<0.005	X					X	0.0000057
Lead	Pb	0.000001	0.07	<0.45	0.014	<0.018	X				X		0.000000018
Lithium	Li	0.171	<0.005	<0.45	<0.007	<0.003	X					X	0.00051
Lutetium	Lu	0.048	--	<4.5	<0.001	<0.005	X					X	0.00024
Magnesium	Mg	0.00005	0.1	<0.045	0.07	0.060	X				X		0.000003
Manganese	Mn	0.0041	0.1	<0.045	0.07	0.030	X			X			0.00012
Mercury	Hg	--	--	--	<0.002	<0.030		X					
Molybdenum	Mo	0.0014	0.06	0.045	--	0.024	X			X			0.000034
Neodymium	Nd	0.00573	<0.03	<4.5	--	<0.05	X					X	0.00029
Neon	Ne	--	--	--	--	--		X					
Nickel	Ni	0.00114	0.2	0.14	0.2	0.12	X			X			0.00014
Niobium	Nb	0.00088	--	<0.045	--	<0.018	X					X	0.000016
Nitrogen	N	0.002	--	--	--	(e)	X					X	NA
Osmium	Os	--	--	--	<0.004	--	X				X		
Oxygen	O	0.000004	--	--	--	--	X				X		NA
Palladium	Pd	--	--	--	<0.001	<0.012		X					
Phosphorus	P	0.0001	--	--	--	--	X				X		0.0001

TABLE 5-1 (Continued)

Column													
1	2	3	4	5	6	7	8	9	10	11	12	13	14
Element	Symbol	Boron Equivalent Factor	Round Robin Test Data ^(a) (ppm)				Elements Requested for Analyses ^(b)	Element NOT Requested ^(b)	Analyses for Purposes Other Than Boron Equivalent ^(c)	Elements NOT Recommended for Analyses by ASTM Committee C5:05	Elements With Boron Equivalent < 0.0001	Elements for Boron Equivalent Analyses ^(d)	Boron Equivalent (ppm)
			Laboratories										
			I	II	III	IV							
Platinum	Pt	--	--	--	<0.001	<0.012		X					
Potassium	K	0.00081	0.1	--	<0.017	<0.030	X					X	0.000024
Praseodymium	Pr	0.00156	--	<4.5	--	<0.050	X					X	0.000078
Protactinium	Pa	--	--	--	--	--							
Radium	Ra	--	--	--	--	--		X					
Radon	Rn	--	--	--	--	--		X					
Rhenium	Re	--	--	--	<0.001	--		X					
Rhodium	Rh	--	--	--	<0.004	<0.024		X					
Rubidium	Rb	0.00033	00	00	<0.004	<0.120	X					X	0.000040
Ruthenium	Ru	--	--	--	<0.002	--		X					
Samarium	Sm	0.941	<0.1	<4.5	<0.007	<0.050	X			X			0.047
Scandium	Sc	0.0074	--	<0.45	--	<0.002	X					X	0.000075
Selenium	Se	--	--	--	--	--		X					
Silicon	Si	0.000085	20.0	4.5	26.0	3.0	X				X		0.00026
Silver	Ag	0.0322	0.01	--	0.028	0.001	X			X			0.000032
Sodium	Na	0.00034	0.2	0.22	0.10	0.90	X			X			0.0031
Strontium	Sr	0.000836	--	<0.045	0.006	<0.0120	X					X	0.00001
Sulfur	S	0.00029	--	--	--	--	X					X	0.00046
Tantalum	Ta	0.0159	--	<0.45	<0.007	<0.120	X			X			0.0019
Tellurium	Te	--	--	<4.5	--	<0.060		X					
Terbium	Tb	0.0137	0.050	--	--	<0.050	X					X	0.00069
Thallium	Ts	0.00043	0.1	0.45	0.2	0.060	X					X	0.000026
Thorium	Th	--	--	--	--	--		X					
Thulium	Tm	0.0487	<0.02	<4.5	<0.004	<0.005	X					X	0.00024
Tin	Sn	0.00029	0.2	<0.045	0.07	0.060		X					
Titanium	Ti	0.0018	--	<0.045	--	0.180	X		X				0.00032
Tungsten	W	0.00857	--	--	<0.004	<0.120	X			X			0.0010
Uranium	U	--	--	--	<0.004	<0.060		X					
Vanadium	V	0.00147	0.05	<0.45	0.001	0.018	X		X				0.000026
Xenon	Xe	--	--	--	--	--		X					
Ytterbium	Yb	0.00715	<0.005	<0.45	--	<0.002	X					X	0.000014
Yttrium	Y	--	<0.005	--	--	<0.003		X					
Zinc	Zn	0.00027	0.1	--	0.070	<0.060	X			X			0.000016
Zirconium	Zr	0.0001	0.1	<0.45	0.070	0.060	X				X		0.000006
Silver ¹⁰⁹	Ag ¹⁰⁹	0.053	--	--	--	--	X			X			

(a) Work done by ASTM Analytical Task Force; C5:05 Nuclear Graphite Committee.

(b) Elements requested by neutron designers.

(c) Measured for purpose of controlling oxidation, but included in BE calculation.

(d) Elements not listed in columns 10, 11, and 12.

(e) N₂ measured on log 6884-34 = 45 ppm

Note: Ash content of analytical specimen used for this experiment = 60 ppm.

No analysis was done for phosphorus or nitrogen, but values of 1.6 ppm for phosphorus (Ref. 5-1) and 45 ppm for nitrogen (see Table 5-1, footnote e) were obtained on different specimens and included in the BE calculation. A BE value of 0.238 ppm was obtained for burnable elements (boron, calcium, europium, gallium, lithium, and samarium). The only element that contributed significantly to the nonburnable BE value was cadmium, whereas for the burnable BE value boron and samarium were the big contributors. It was assumed that chlorine (BE factor = 0.0136) did not contribute greatly to the total BE.

By eliminating elements in Table 5-1, column 9 (not requested by the neutronic designers), column 10 (to be determined for purposes of checking on oxidation resistance), column 11 (not recommended for analyses by ASTM C5:05 work due to low probability of occurrence), and column 12 (BE factor <0.0001), we arrive at the remaining list of elements in column 13 which are required. Again using data from Table 5-1, column 7, a BE of 0.093 ppm was calculated for nonburnable elements and 0.18 ppm for burnable. The elimination of cadmium in accordance with the ASTM Committee C5:05 recommendation accounts for the large reduction in nonburnable BE. If cadmium is included in column 13, the value becomes 0.128 ppm, in excellent agreement with the value of 0.131 when all elements are included.

The nonburnable BE concentration is well below the desired limit of 0.5 ppm for this specimen and it made little difference whether it was calculated using impurity data for elements in column 8 or column 13. The BE value calculated from the round-robin data significantly increases the sensitivity of analysis and gives an upper limit of 0.131 ppm on the nonburnable BE in comparison with an upper limit value of 0.9486 ppm reported in Ref. 5-1 and 0.238 ppm for the round-robin data versus 3.2922 ppm from Ref. 5-1 for burnable BEs.

It should be noted that the results reported on the boron equivalent impurity content were obtained on a preproduction log of high purity (ash content = 60 ppm). Impurity content will be distributed among logs within

a production batch, the purity depending on the degree of purification within each graphitization furnace lot. The distribution of impurity content is discussed further in Section 5.2.5.2. Based on this experiment, log 6848-34, lot 426, is of high purity and would easily qualify as a fuel element in the core of an HTGR.

5.2.5.2. Ash, Impurities Affecting Oxidation Rate, and Lithium. The total ash content, impurities that catalyze oxidation reactions, and lithium are important for design and safety analyses. The data for H-451 graphite, lot 426, are summarized in Tables 5-2, 5-3, and 5-4.

Impurity content for four logs of lot 426 are shown in Table 5-2. The ash content for these logs averaged 45 ppm and ranged from <25 to 89 ppm with small variations within the log (Ref. 5-1). Elements known to catalyze oxidation reactions, such as iron, vanadium, titanium, aluminum, silicon, and calcium, ranged from 0.5 to 22 ppm. Sulfur content was about 1 ppm. The distribution of these elements was uniform within each log and paralleled the distribution of the ash. Lithium (see Table 5-3) was not detected using the most sensitive methods available. The lithium content was well below 1 ppm and may be below 0.005 ppm.

As mentioned previously, the purity of the logs is a function of the degree of purification imparted in each graphitization furnace. Data on ash content for a number of the preproduction lots as a function of their graphitization run are given in Table 5-4. The mean ash content in furnace runs 6399-C and 6003-C was 23 and 50 ppm, respectively, whereas it was 255 ppm in run 5698-C. Graphitization furnace run 6399-C contained logs from 3 lots; lot 426 logs contained coke A, and lots 438 and 440 logs contained coke C; however, all logs were low in ash. Production lots 472, 478 and 482 were relatively high in ash content. These logs were graphitized in a different and less efficient purification facility than the preproduction lots. Since the data presented here show that the manufacturing process is

TABLE 5-2
IMPURITY CONTENT: H-451, LOT 426^(a)

Log No.		Ash		Mean Value (MLC, MLE, EC, EE) (ppm)											No. of Replicates
GA	GLCC	GLCC ^(b)	GA	B	Fe	Va	Ti	S	Al	Ca	Si	Na	Mg	Pb	
6484-33	92	34	<25	3.0	<4.0	<0.5	<1.0	<2.0	4.0	<20	28.8	<10	<0.5	< 6.0	8
6484-34	198	24	<25	4.8	<7.4	<0.5	<2.1	<2.1	11.3	<27.5	36.3	<11.3	<0.6	<10.3	8
6484-40	155	42	89	<0.8	<1.0	<0.5	<1.0	<1.0	< 1.0	<20.0	<10.0	<10.0	2.0	< 6.0	8
6484-41	184	13	41	<1.4	<1.0	<0.5	<1.0	<1.0	< 1.0	<20.0	<10.0	<10.0	<0.5	< 6.0	8
Mean lot (4 logs)			<45	<2.5	<3.2	<0.5	<1.3	<1.5	< 4.3	<21.9	<21.3	<10.3	<0.9	< 7.1	32

(a) From Ref. 5-1.

(b) Measured by GLCC; 1 specimen per log.

TABLE 5-3
SUMMARY OF LITHIUM ANALYSES: H-451 (a)

GA Log No. / (GLCC Log No. / GLCC Lot No.)	No. of Replicates	Content (ppm)
5651-86/ 52/408	4	<1.0 ^(b)
	1	<0.07
	1	<0.02
6484-33/ 92/426	8	<1.0 ^(b)
	2	0.04 ^(b)
6484-34/ 198/426	2	<0.005 ^(b)
	8	<1.0 ^(b)
	1	0.07
	1	<0.005
	1	0.074
	1	<0.005
	1	<0.01
	1	<0.005
	1	<1.0
	8	<1.0 ^(b)
6484-40/ 155/426	1	<0.01
	1	<1.0
	8	<1.0 ^(b)
6484-41/ 184/426	1	<0.01
	1	<0.01

(a) From Ref. 5-1.

(b) All specimens have this value.

TABLE 5-4
IMPURITY CONTENT: H-451

GLCC Graphitization Furnace Run No.	GLCC Lot No.	Ash (ppm)		No. Logs Tested ^(a)
		$\bar{x}$	σ	
Preproduction				
5698-C	426	255	438	45
6399-C	426	23	6.5	31
6003-C	426	50	75	51
6399-C	438	21	9.1	25
6399-C	440	26	13	23
Production				
1272	472 ^(b)	304	258	98
	478 ^(b)			
	482 ^(b)			

(a) One specimen tested per log.

(b) Lots 472, 478, and 482 values averaged together.

capable of producing material with extremely low impurity content, it is the function of the material specification to maintain the impurity content required for reactor design and safety analyses in future production.

5.2.6. Conclusions

The bulk density averaged $1.74 \pm 0.02 \text{ Mg/m}^3$ for production logs. The mechanical properties exhibited near-isotropic characteristics with the lowest strength values at the MLC of the logs.

Thermal expansivity values were intermediate between needle-coke graphites and high-strength isotropic graphites. The anisotropy factor was 1.15. The intermediate thermal expansivity and the anisotropy factor

result from the structure of the filler coke used. The near-isotropic cokes improve the thermal conductivity in the radial direction (major direction of heat flow in reactor service) over needle-coke graphites.

The impurity content of logs from each graphitization lot depends on the thoroughness of purification during the graphitization process. The ash content can range from <50 to 300 ppm, depending on the degree of purification. The nonburnable boron equivalent value of a highly purified log was 0.131 ppm and the burnable boron equivalent value 0.238 ppm.

5.3. STATISTICAL DISTRIBUTION OF STRENGTH WITHIN A LOG

A detailed knowledge of the statistical distribution of tensile and flexural strength within an H-451 log was obtained by destructively testing a complete log (Ref. 5-5). Factors sought were (1) the systematic variation of strength with orientation and location throughout a log, (2) for a given orientation and location, the statistical distribution of strengths, the mean, standard deviation, and the coefficient of variation which characterize the specimen population, (3) the relation between flexural strength and tensile strength, and (4) the validity of the Weibull statistical theory (Ref. 5-6), including the dependence of strength on specimen size and the Weibull modulus, if applicable. The Weibull theory, in which the strength of a brittle solid is assumed to be controlled by flaws, has potential uses in the engineering design of load-bearing structures made from brittle materials because it relates the probability of failure to the volume of material under load, the stress gradient, and multiaxial stress states. It can be combined with finite element stress analysis to predict the probability of failure as a function of position in components under complex stress states. Previous studies of the strength of H-327 graphite (Ref. 5-7) and near-isotropic Gilsocoke-based graphites (Ref. 5-8) showed that while the ratios relating the strength of flexural specimens and internally pressurized tubes to the tensile strength were correctly predicted by the Weibull model, the dependence of strength on specimen volume was overestimated.

5.3.1. Sampling

Sampling diagrams are shown in Appendix A, Figs. A-5, A-6, and A-7. A log from preproduction lot 408, log 5651-90, was used. Tensile specimens of two sizes and flexural specimens were taken in both axial and radial directions from locations covering most of the log. More than 2000 specimens were tested to failure.

5.3.2. Conclusions

Complete data sets are reported in Ref. 5-5. The following paragraphs summarize the results.

Axial tensile specimens averaged 19% stronger than radial specimens, and axial flexural specimens averaged 13% stronger than radial specimens. Axial specimens taken near the edge of the log averaged 16% and flexural specimens 7% stronger than those taken near the center. The coefficient of variation for axial tensile strengths from the same zone in the parent log averaged 11.2%, compared with 14.7% for radial tensile strengths. In flexure, the coefficient of variation averaged 7.9% and 9.0% in the axial and radial directions, respectively.

Within experimental scatter, the data were equally well represented by a normal (Gaussian) distribution or by a distribution predicted by the Weibull model. The value of the Weibull modulus m was lower for tensile tests (mean values of 10.7 for axial specimens and 8.1 for radial specimens) than for flexural tests (mean values of 15.8 for axial specimens and 13.6 for radial specimens). Such a difference is contrary to predictions based on the Weibull model. Tensile specimens of two sizes (volumes 9847 mm³ and 724 mm³) were tested. The small specimens averaged 3% and 8% stronger than the large specimens for the axial and radial directions, respectively. The observed size effect was much lower than expected from the Weibull model, which predicted differences of 28% and 38% for the axial and radial directions, respectively.

The mean flexural strength showed good correlation with the mean tensile strength of companion specimens (correlation coefficient 0.9). Flexural strength averaged 52% and 55% higher than the corresponding tensile strengths for the axial and radial directions, respectively. These values are in good agreement with values of 51% axial and 64% radial predicted by the Weibull model. While there is agreement between the experimental flexure-to-tensile strength ratios and the ratios predicted by the Weibull model, the experimentally derived size effect and the difference in Weibull modulus between tensile and bend data are inconsistent with the Weibull model. Thus, the H-451 graphite log tested does not meet the criteria which would permit it to be treated as a Weibull solid.

When considering the minimum strength of H-451 logs for acceptance/rejection tests, the data in Ref. 5-5 confirm the results from Ref. 5-1 that the logs are weakest at the midlength center (MLC). Thus, testing specimens from the MLC zone constitutes the most conservative criterion for eliminating weak or defective logs.

5.4. DEFINITION OF MINIMUM STRENGTH AND STRENGTH DISTRIBUTION AMONG LOGS

Ninety-eight full-size (432 mm diameter by 813 mm long) production logs were manufactured, using the same formulation, raw materials, and procedures as preproduction lot 426. The 98 logs were a portion of a 354-log production order for possible use as FSV reload elements. The 354-log order was manufactured in 6 extrusion lots, taken through the baking process and stored at GLCC. The 98 logs were graphitized and distributed to GA for tensile strength testing, and to ORNL and BNL for experimental purposes. The 98 logs were taken from three of the production extrusion lots (472, 478, and 482). All logs were graphitized in furnace run number 1272.

5.4.1. Sampling

The logs were sampled for tensile testing by GLCC at their Morgantown plant in accordance with a sampling plan devised by GA. The sampling diagram is given in Appendix A, Figs. A-8, A-9, and A-10. A rod 16 mm in

diameter by 356 mm long was cored from the center and a 25-mm slab was taken from one end of each log. A 102-mm core was taken for axial specimens, as shown in Fig. A-9. The GA axial strength specimens were taken as close to the midlength center (MLC) of each log as possible (within 51 mm of the center; see Fig. A-8) while preserving the logs for subsequent machining into fuel element blocks. Axial test specimens 6.35 mm in diameter by 23 mm long were machined from the axial core, as shown in Fig. A-9, and radial specimens of the same size were machined from the end slab, as shown in Fig. A-10. A single tensile test specimen (12.8 mm in diameter by 76.2 mm long) was taken from each center core, as shown in Fig. A-8, for measurement by GLCC.

Ultimate tensile strength was measured on eight axial specimens and four radial specimens from each log at GA, and one axial specimen was measured from each log at GLCC.

5.4.2. Experimental Results

Individual strength values for each log and the mean values grouped by extrusion lot are reported in Ref. 5-9. The average strengths and standard deviations for axial specimens tested by GA and GLCC from each extrusion lot are given in Table 5-5. The grand mean of 12.7 MPa for GA data compares with 13.3 MPa for GLCC's single specimens.

Correlation between the values for GLCC's single specimen and the GA average values was poor (Ref. 5-9). The difference may reflect both the spatial separation of the sampling zones and the inherent randomness of the single specimen measured by GLCC. Statistical tests showed no significant difference in mean values between A and B specimens (located closest to the log center) and the C and D specimens; therefore, they can be treated as coming from the same population.

The differences in average strength (GA data) between the three extrusion lots, as shown in Table 5-5, were statistically highly significant. The small differences in standard deviation between the different extrusion lots were not statistically significant.

TABLE 5-5
STATISTICAL ANALYSES OF GA AND GLCC AXIAL STRENGTH DATA

GLCC Extrusion Lot No.	No. of Specimens Tested by:		Average Tensile Strength and Standard Deviation (MPa)			
	GA	GLCC	$\bar{s}$	σ	$\bar{s}$	σ
472	111	14	12.3	2.9	13.0	2.2
478	439	56	13.6	2.8	13.8	2.7
482	224	28	11.1	2.7	12.6	4.1
All	774	98	12.7	3.0	13.3	3.1

There was no correlation between strength and the position of the log within the graphitization furnace. The differences between the average strengths of lots 472, 478, and 482 were accounted for by GLCC as differences in processing during extrusion.

The average radial strengths and standard deviations for each extrusion lot are given in Table 5-6. The grand mean was 15.8 MPa. The difference in strength between lots 472 and 478 was not statistically significant, but lot 482 was significantly weaker than the other two lots. In contrast to the axial strengths, the differences in standard deviations were statistically significant, with lot 482 showing more variability than the other two lots. This was attributable to an increase in both the log-to-log variability and the within-log (specimen-to-specimen) variability.

There was no statistically significant correlation between the radial strength and the axial strength of the same logs (correlation coefficient 0.11). The higher strength and reduced standard deviation of the radial specimens at the end-center position compared with the axial specimens at the midlength-center position are not unusual and have been observed in preproduction logs (Refs. 5-1, 5-5). The differences between the three lots show that material extruded in different lots can vary considerably in strength properties after baking impregnation and graphitization.

TABLE 5-6
STATISTICAL ANALYSIS OF RADIAL STRENGTH DATA

GLCC Extrusion Lot No.	No. of Specimens Tested	Average Strength (MPa)	Standard Deviation (MPa)
472	56	16.4	1.4
478	224	16.3	1.8
482	112	14.5	2.7
All	392	15.8	2.2

5.4.3. Application of Results to Acceptance Testing

The strength of axial midlength-center specimens was both lower and more scattered than the strength of radial end-center specimens. Variations between extrusion lots and variations between logs within the same lot both contribute to the scatter. The log-to-log variations appears to be too great to allow qualification of entire lots. Therefore, acceptance tests intended to ensure minimum strength requirements should be based on axial midlength-center specimens and each log should be tested.

5.4.3.1. Statistical Model. Acceptance criteria assume the following statistical model (Ref. 5-9):

$$x_{ij} = \mu + \alpha_i + \beta_j + \epsilon_{ij} \quad (5-9)$$

where x_{ij} is an axial strength measurement from the j^{th} log in the i^{th} lot, μ is the grand mean strength, α_i is the variation systematic to the i^{th} lot, β_j is the variation systematic to the j^{th} log from this lot, and ϵ_{ij} is the random variation (in the midlength-center sampling zone) within this log. It was further assumed that α , β , and ϵ are normally and independently distributed with means of zero and variances of σ_{lot}^2 , σ_{log}^2 , and σ_{WL}^2 , respectively. The latter three quantities are, in order, the variances attributable to

lot-to-lot variations, log-to-log variations, and within-log variations. It is implicit in the model that the log-to-log variance is the same for all extrusion lots and the within-log variance is the same for all logs.

Values for the three components of variance were calculated from the test data by the analysis-of-variance method. The calculations confirmed that both lot-to-lot and log-to-log variations were statistically highly significant. The values for σ_{lot}^2 , σ_{log}^2 , and σ_{WL}^2 are shown in Table 5-7; the corresponding standard deviations are also included.

The largest contributor to the spread in the axial strength measurements is the log-to-log variation among logs from the same extrusion lot (see Table 5-7), whereas the within-log variance $[2.67 \text{ (MPa)}^2]$ is low enough for log-by-log acceptance tests based on a few specimens per log to be practicable.

5.4.3.2. Acceptance Criteria. The purpose of acceptance testing is to ensure that material from the weakest location (midlength center) of the logs meets the minimum strength requirement S_{min} . Because of the variability

TABLE 5-7
COMPONENTS OF VARIANCE: AXIAL STRENGTH DATA

Source of Variation	Symbol	Variance (MPa) ²	Standard Deviation (MPa)
Lot-to-lot	σ_{lot}^2	1.22	1.10
Log-to-log	σ_{log}^2	5.22	2.29
Within-log (residual)	σ_{WL}^2	2.67	1.63

in graphite strengths, a statistical definition of minimum strength is required. The following two alternative definitions were considered:

90/90: At least 90% of the material at the midlength center of the log equals or exceeds a specified minimum strength, with 90% confidence.

90/95: At least 99% of the material at the midlength center of the log equals or exceeds a specified minimum strength, with 95% confidence.

An acceptance testing procedure which meets the needs can be derived from the statistical model of Eq. 5-9, if it is assumed that the present estimate of the within-log variance $[2.67 \text{ (MPa)}^2]$ is exact and will remain unchanged in future production runs. It is assumed that n replicate specimens from the midlength center of a log are tested and their average, $\bar{x}$, is used to estimate the mean log strength, $\mu + \alpha_i + \beta_j$. The minimum strength requirement $S_{\min}$ will be met if $\bar{x}$ meets the following criterion:

$$\bar{x} \geq S_{\min} + 1.282 \sigma_{WL} + \frac{1.282}{\sqrt{n}} \sigma_{WL} \quad (5-10a)$$

where a 90/90 definition of minimum strength is used, or

$$\bar{x} \geq S_{\min} + 2.326 \sigma_{WL} + \frac{1.645}{\sqrt{n}} \sigma_{WL} \quad (5-10b)$$

where a 99/95 definition of minimum strength is used. The numerical factors in Eq. 5-10 come from tabulated values for the standard normal distribution.

Because different components in the HTGR core experience different stress levels, it is desirable to devise an acceptance test that sorts the logs according to their strength. Therefore, the following strength categories were considered:

Category A: minimum strength 10.3 MPa

Category B: minimum strength 8.3 MPa

Category C: minimum strength 5.5 MPa

The value which $\bar{x}$ must exceed for a log to be assigned to category A, B, or C can be calculated from Eq. 5-10. Values are shown in Table 5-8 for four and eight replicate tests per log, and for 90/90 and 99/95 definitions of minimum strength.

The criteria listed in Table 5-8 were applied to each of the 98 logs. For the case requiring four replicate tests, the four specimens closest to the log center (subscripts A and B; see Figs. A-8 and A-9b) were used. The assigned category for each log is given in Ref. 5-9 for a four- or eight-specimen sampling plan, and for a 90/90 or 99/95 definition of minimum strength.

5.4.3.3. Yields of Logs by Category. A summary of the numbers of logs assigned to each category under four- and eight-specimen sampling plans, applying either a 90/90 or 99/95 definition of minimum strength, is given in Table 5-9. Results are presented for each extrusion lot.

Using a 90/90 definition of minimum strength and an eight-specimen sampling plan, only two logs would be rejected as failing to qualify for strength category C, and almost half the logs would qualify for category A. If a 99/95 definition were used, 18 logs would be rejected and about 20% of the remainder would qualify for category A. A typical order of logs for a replacement segment of an HTGR core would require about 25% of the accepted logs to qualify for category A and 70% to qualify for A or B. If a 99/95

TABLE 5-8
CRITERIA FOR ASSIGNING LOGS TO STRENGTH
CATEGORIES A, B, OR C (AXIAL DATA)

No. of Tests per Log	Definition of Minimum Strength S_{min}	Strength Category ^(a)	Value Which Average Measured Strength $\bar{x}$ Must Exceed (MPa)
4	90/90 ^(b)	A	13.5
		B	11.4
		C	8.7
	99/95 ^(c)	A	15.5
		B	13.4
		C	10.7
8	90/90 ^(b)	A	13.2
		B	11.1
		C	8.3
	99/95 ^(c)	A	15.1
		B	13.0
		C	10.3

(a) Categories correspond to the following minimum strengths:

A: 10.3 MPa
B: 8.3 MPa
C: 5.5 MPa

(b) At least 90% of material at midlength center exceeds specified minimum strength, with 90% confidence.

(c) At least 99% of material at midlength center exceeds specified minimum strength, with 95% confidence.

TABLE 5-9
YIELDS OF LOGS IN EACH STRENGTH
CATEGORY (BASED ON AXIAL STRENGTHS)

No. of Tests per Log	Definition of Minimum Strength S_{min}	Extrusion Lot	No. of Logs Assigned to Strength Category				
			A	B	C	Reject	Total
4	90/90 ^(a)	472	6	3	4	1	14
		478	27	22	6	1	56
		482	6	6	13	3	28
		A11	39	31	23	5	98
	99/95 ^(b)	472	1	5	3	5	14
		478	12	16	26	2	56
		482	1	5	7	15	28
		A11	14	26	36	22	98
8	90/90 ^(a)	472	6	3	4	1	14
		478	29	22	5	0	56
		482	7	6	14	1	28
		A11	42	31	23	2	98
	99/95 ^(b)	472	2	4	4	4	14
		478	14	20	20	2	56
		482	1	6	9	12	28
		A11	17	30	33	18	98

(a) At least 90% of material at midlength center exceeds specified minimum strength, with 90% confidence.

(b) At least 99% of material at midlength center exceeds specified minimum strength, with 95% confidence.

minimum strength definition were adopted in place of the 90/90 definition, the yields in the higher categories would fall just short of these requirements, based on the quality of the 98 logs considered in this experiment.

The effect on the yields of reducing the number of tests per log from eight to four is small. The number of rejects would increase from two to five under a 90/90 strength definition, and rejects would be increased from 18 to 22 under a 99/95 strength definition. There would be some overall downgrading because more borderline logs would be moved down in category than would be moved up.

The better quality of extrusion lot 478, indicated by its higher average strength in Table 5-4, is reflected in the yields listed in Table 5-9. Even using a 99/95 definition of minimum strength, only two of the 56 logs in lot 478 would be rejected and 25% would meet category A requirements.

5.4.4. Conclusions

The axial strengths (average value 12.7 MPa) from the MLC showed considerable lot-to-lot and log-to-log variations, while the radial strengths (average value 15.8 MPa) from the EC were more uniform. There were no statistically significant correlations between axial strength and radial strength in the same log, or between strength and the position of the log in the graphitizing furnace.

Acceptance criteria for assigning logs to a minimum strength category can be based on axial tensile tests, if each log is tested. A statistical model incorporating a separate lot-to-lot variance, log-to-log variance, and within-log variance can be adopted for this purpose.

Acceptance criteria which could be used for assigning a log to strength category A (minimum strength 10.3 MPa), B (minimum strength 8.3 MPa), or C (minimum strength 5.5 MPa) were derived. Calculations were made for either four or eight replicate specimens per log and two alternative definitions of

minimum strength were considered. The first definition would require 90% of the material at the midlength center of the log to exceed the specified minimum strength, with 90% confidence (90/90); the second would require 99% of the material to exceed the specified minimum, with 95% confidence (99/95). When these criteria were applied to the axial test data from the 98 logs, two logs would fail to qualify for category C, while almost half of the logs would qualify for category A if a 90/90 definition of minimum strength and eight specimens per log were used. Use of a 99/95 definition of minimum strength would increase the number of logs rejected to 18. Decreasing the number of tests per log from eight to four would cause a small reduction in the yields.

Using a 99/95 definition of minimum strength, only two of the 56 logs in the lot of highest average strength (478) would be rejected and about 25% would qualify for category A. This compares with a rejection rate of about 40% for logs from the other two lots.

These results provide the information required to write an acceptance specification for strength of a log by measuring the tensile strength on specimens taken at the MLC. The number of logs and specimens tested and the confidence required depend on the confidence required in design.

5.5. FATIGUE BEHAVIOR

The graphite fuel element blocks are subject to cyclic stresses from power cycling, operating vibrations, and possibly seismic shaking. Design of the components requires a knowledge of the cyclic fatigue behavior of graphite under loading cycles with different ratios of minimum stress to maximum stress (R value). The inherent scatter in mechanical properties of graphites requires that sufficient tests be made for statistical analysis. Uniaxial fatigue tests were performed on H-451 graphite with R values ranging from -1 to 0.5 (Ref. 5-10).

5.5.1. Sampling

Specimens for unirradiated fatigue testing of H-451 were taken in the axial and radial directions a quarter of the way along the length of the log used for within-log strength testing (see Fig. A-5 in Appendix A) between 25 and 100 mm from the edge (see Fig. A-11 in Appendix A). For a given series of tests, specimens were taken from locations spread uniformly over the sampling zone to prevent bias from local inhomogeneities in strength. Cylindrical specimens 12.7 mm in diameter by 25.4 mm long were used.

5.5.2. Experimental Procedure

The first test series consisted of 30 to 35 control tensile tests of radial and axial specimens on standard fatigue specimens mounted in the fatigue machine. These tests established the mean tensile strengths of the control specimens (see Appendix B, Table B-10). Fatigue tests were run using R values of -1, -0.5, 0, and 0.5. Forty to fifty specimens were included in each series of tests. From seven to ten specimens were cycled at each of five maximum stress levels ranging from 0.6 to 1.0 times the mean tensile strength. The stress levels were selected to avoid an excessive number of run-outs beyond 100,000 cycles. Axial and radial specimens were measured.

5.5.3. Experimental Results

A typical S-N curve, taken from Ref. 5-10, in which the logarithm of the maximum stress S is plotted against the logarithm of the number of cycles at failure N is shown in Appendix B, Fig. B-1, for axial specimens with an R value of -1. The stresses are normalized by dividing by the mean tensile strength. The tensile data and first cycle failures are plotted at $N = 0.25$, and run-outs (specimens which survived 100,000 cycles without failure) are shown as open circles at $N = 100,000$.

The S-N curves have the same form as those of many metals, except that the scatter band is wider. The data do not extend to high enough values of N to confirm the existence of an absolute endurance limit.

The usual practice of analyzing fatigue data has been to deduce the N value corresponding to a particular failure probability from the dispersion of log (N) values at a given stress setting, assuming a Weibull or Gaussian distribution. This procedure has several disadvantages when applied to graphite; in particular, it provides no information on low failure probabilities in the high cycle regime. The analytical problem was approached in Ref. 5-10 by inverting the usual practice and assuming that for a constant life the failure stresses follow a specific distribution (Ref. 5-11). The following statistical model was assumed (Ref. 5-10):

$$\log_{10} \left(\frac{\sigma_{\max}}{S} \right) = \alpha + \beta \log_{10} (N) + \epsilon \quad (5-11)$$

where $\sigma_{\max}$ is the peak tensile stress, S is the mean tensile strength, α and β are constants, and ϵ is a random variable distributed normally with a mean of zero and a standard deviation s. Data in Ref. 5-10 were analyzed according to the model of Eq. 5-11, including the tensile tests at $N = 0.25$ but excluding the specimens which ran out beyond 10^5 cycles. The least-squares straight line through the data and the x/y lower tolerance limits (representing the limits above which at least x% of all observations would fall, with y% confidence) were calculated for the x/y combinations 90/90, 95/95, and 99/95 and are included in Appendix B, Fig. B-1. The data support a linear relation between $\log (\sigma_{\max})$ and $\log (N)$ assumed in Eq. 5-11. Results of the statistical analyses are summarized in Table 5-10.

For design, it is convenient to plot fatigue results on a constant life fatigue diagram (Goodman diagram), which relates fatigue life to the following interdependent loading cycle parameters: maximum stress, minimum stress, stress amplitude, and mean stress. The data in Appendix B, Fig. B-2, provide an example of a Goodman diagram, taken from Ref. 5-10, relating the

TABLE 5-10
STATISTICAL ANALYSES OF FATIGUE DATA FOR UNIRRADIATED H-451 GRAPHITE

Orientation	Stress Ratio R ($\sigma_{\min}/\sigma_{\max}$)	Intercept of Least Squares Line α ^(a)	Slope of Least Squares Line β ^(a)	Standard Deviation s ^(a)	Homologous Stress Limit $\sigma_{\max}/\bar{S}$ for Survival to 10^5 Cycles ^(a)	
					50% Probability	99/95 Lower Tolerance Limit
Axial	0.5	-0.023	-0.014	0.042	0.81	0.60
Axial	0	-0.025	-0.026	0.038	0.70	0.54
Axial	-0.5	-0.019	-0.037	0.052	0.63	0.44
Axial	-1	-0.032	-0.038	0.048	0.60	0.44
Radial	0.5	-0.010	-0.004	0.056	0.93	0.62
Radial	0	-0.019	-0.016	0.057	0.80	0.54
Radial	-0.5	-0.018	-0.021	0.059	0.75	0.50
Radial	-1	-0.025	-0.020	0.058	0.75	0.50

^(a) Based on Eq. 5-11.

loading cycle parameters for 50% survival to 10^2 , 10^3 , 10^4 , and 10^5 cycles in axial specimens of H-451 graphite.

When normalized by dividing by the mean tensile strength, the homologous stress limits for radial specimens were somewhat greater than those for axial specimens under similar conditions (Table 5-10).

Under reversed stress cycling ($R = -1$), the homologous stress limits for survival of axial specimens to 10^5 cycles averaged 0.63 for 50% survival and 0.48 for 99% survival (99/95 tolerance limit), in good agreement with Leichter and Robinson's results (Ref. 5-11). The homologous stress limits for radial specimens averaged 0.74 for 50% survival and 0.53 for 99% survival.

5.5.4. Conclusions

H-451 graphite showed cyclic fatigue life curves (S-N curves) generally resembling those of metals, but with more scatter. For reversed stress cycling ($R = -1$), the homologous stress limits (maximum applied fatigue stress divided by the tensile strength) for 50% specimen survival to 10^5 cycles averaged 0.63 in the axial direction and 0.74 in the radial direction. Corresponding homologous stress limits for 99% specimen survival (99/95 tolerance limits) were 0.48 and 0.53. The fatigue life and the homologous stress limits increased as R increased.

REFERENCES

- 5-1. Johnson, W. R., and G. B. Engle, "Properties of Unirradiated Fuel Element Graphites H-451 and TS-1240," ERDA Report GA-A13752, General Atomic Company, January 31, 1976.
- 5-2. Nye, J. F., Physical Properties of Crystals, Oxford University Press, London, pp. 1-8, 1967.
- 5-3. Engle, G. B., and W. R. Johnson, "Properties of Unirradiated Fuel Element Graphites H-451 and S0818," ERDA Report GA-A14068, General Atomic Company, October 8, 1976.

- 5-4. Butland, A. T. D., and R. J. Maddison, "The Specific Heat of Graphite: An Evaluation of Measurements," J. Nucl. Mater. 49, (1973-74).
- 5-5. Price, R. J., "Statistical Study of the Strength of Near-Isotropic Graphite," ERDA Report GA-A13955, General Atomic Company, May 24, 1976.
- 5-6. Weibull, W., "A Statistical Theory for the Strength of Materials," Hand. Ing. Vet. Akad., Proceedings, Royal Swedish Institute for Engineering Research, Stockholm, Nr. 151, 1939.
- 5-7. Price, R. J., and H. R. W. Cobb, "Application of Weibull Statistical Theory to the Strength of Reactor Graphite," Proceedings, Conference on Continuum Aspects of Graphite Design (CONF-701105), USAEC Technical Information Center, 1972, p. 547.
- 5-8. Brocklehurst, J. E., and M. I. Darby, "Concerning the Fracture of Graphite Under Different Test Conditions," Mater. Sci. Eng. 16, 91 (1974).
- 5-9. Engle, G. B., and R. J. Price, "Strength Testing of Production Grade H-451 Graphite; Lots 472, 478 and 482," ERDA Report GA-A14269, General Atomic Company, March 1977.
- 5-10. Price, R. J., "Cyclic Fatigue of Near-Isotropic Graphite: Influence of Stress Cycle and Neutron Irradiation," DOE Report GA-A14588, General Atomic Company, December 1977.
- 5-11. Leichter, H. L., and Robinson, E., "Fatigue Behavior of a High-Density Graphite and General Design Correlation," J. Am. Ceram. Soc. 53, 197 (1970).

6. IRRADIATION PROGRAM

6.1. INTRODUCTION

The properties of H-451 graphite during irradiation under HTGR conditions of fluence and temperature are required for design and safety analyses.

Irradiation behavioral studies of H-451 graphite began with the irradiation of grade H-429 in a series of BNWL capsules in the ETR. The H-429 specimens were later transferred to GA's OG capsule series and irradiation continued in the ORR. Irradiation of H-451 was initiated in the OG capsule series. Capsules OG-1, -2, and -3 were irradiated at 823 to 1623 K (550° to 1350°C) to a peak fluence of $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} (Refs. 6-1, 6-2, 6-3). By operating the three capsules in series it was possible to obtain complete data sets over the fluence and temperature range of the HTGR. The capsules were irradiated over a period of approximately 5 years, and specimens from the GLCC development program were inserted into the program as they became available. Specimens from H-451 lots 266, 408, and 426 were irradiated along with grade H-429.

Dimensional changes and changes in tensile strength, elastic modulus and strain at fracture, fatigue behavior, thermal expansivity, thermal conductivity, and irradiation creep were determined. Within-log and between-log variations in dimensional changes and properties were determined. Some of the specimens were interchanged within the capsules on subsequent irradiations to provide data on the effect on dimensional changes of increasing and decreasing the temperature during irradiation.

Figure 6-1 shows the fluences and temperatures of the HTGR covered by the irradiation data. In some instances the irradiation data exceed the temperature and fluence range of the reactor.

Capsule design and construction details are given in Refs. 6-1, 6-2, and 6-3. The irradiation data presented are exclusively for H-451 except for changes in thermal expansivity and conductivity, where data from grades

Fig. 6-1. H-451 graphite irradiation data

TS-1240 manufactured by Union Carbide Corporation and grade S0818 by AirCo Speer Division of Air Reduction Corporation were included. These grades were also manufactured for use as fuel element graphites in HTGRs and are similar to grade H-451. The TS-1240 and S0818 specimens were irradiated in the same crucibles with the H-451 specimens.

6.2. SAMPLING

Diagrams of the sampling procedures for irradiation testing are given in Appendix A, Figs. A-1, A-12, and A-13. The specimens were taken from slabs which were removed from the same logs as those used for the characterization studies. Unirradiated control specimens were taken from the same section of the log and from the same specimen population as those used for irradiation.

6.3. DIMENSIONAL CHANGES

Dimensional change data are reported as percent strain $[(\ell - \ell_o)/\ell_o \times 100]$, where ℓ is the postirradiation length and ℓ_o is the original (unirradiated) length.

6.3.1. 873 to 1673 K (600° to 1400°C)

6.3.1.1. Isothermal Dimensional Changes. In capsule OG-3, the third in the OG capsule series, 219 axial and 170 radial dimensional change specimens of H-451 graphite were irradiated. Many of these specimens were previously irradiated in capsules OG-1 and -2 (Refs. 6-1, 6-2); thus, the total fast fluence of some of these H-451 specimens exceeded the lifetime fluence for HTGR fuel element blocks.

Specimens were irradiated at average temperatures ranging from 887 to 1673 K (600° to 1400°C) to total fast neutron fluences of 1.6 to 9.2×10^{25} n/m² ($E > 29$ fJ)_{HTGR}. The data are shown in Appendix B, Fig. B-3 through

B-10. The closed symbols in the figures represent irradiation data from previous OG series capsules, whereas the open symbols represent the data from capsule OG-3.

Radial H-451 specimens were irradiated to fluences sufficient to show turnaround in dimensional change and, in two specimens, a small net radial expansion. The earliest change in linear dimensional change to net expansion occurred in H-451 graphite irradiated at approximately 1273 K (1000°C). Radial specimens appear to go to net expansion when exposed to approximately $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} neutron fluence. Higher irradiation temperatures create a greater initial contraction which delays crossover to net expansion until a fast fluence of greater than $9 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR}.

These data and related effects were incorporated into the design curves following the procedures outlined in Ref. 6-2. Isothermal sections through the design surface are shown in Fig. 6-2.

6.3.1.2. Effect of Changing Irradiation Temperature on Dimensional Changes.

After irradiation in capsule OG-1, two sets of four axial and four radial specimens were interchanged between two crucibles for the next two irradiation increments. The mean operating temperatures of the crucibles were 948 K (675°C) and 1323 K (1050°C), respectively. The dimensional changes are presented in Fig. 6-3; isothermal curves are included in the figure. When the irradiation temperature of the axial specimens was increased, the shrinkage rate increased to that of specimens irradiated isothermally at the higher temperature. The shrinkage rate of axial specimens decreased when their irradiation temperature was reduced, but remained higher than that of specimens isothermally irradiated at the lower temperature to the same fluence. The shape of the curves suggests that the new shrinkage rate may correspond to the lower temperature isothermal curve extrapolated to the higher strain of the stepped-down specimens. The data points for radial specimens are too closely clustered to draw firm conclusions, but the trends appear similar to those of the axial specimens.

Fig. 6-2. Irradiation-induced dimensional change in H-451 graphite (design curves):
(a) axial direction, (b) radial direction

Fig. 6-3. Dimensional changes in H-451 graphite irradiated with changes in temperature (from Ref.6-3)

6.3.1.3. Between-Log and Within-Log Variations in Dimensional Change.

Several groups of H-451 specimens were made up from three different logs from the same lot. In some cases, specimens from the MLC and MLE in the log were included. Four or five replicates were used for each lot and location. After postirradiation length measurements, the data were analyzed by the analysis-of-variance method to find whether the between-log and within-log variations were statistically significant. Summaries of the results of the analyses of variance are given in Tables 6-1 and 6-2. For H-451 graphite, neither the between-log nor the within-log variations were significant at the 95% confidence level. Differences in shrinkage between MLC and MLE specimens were not statistically significant for H-451 graphite.

6.3.2. 573 to 873 K (300° to 600°C)

Only a few data points are available on H-451 graphite below 827 K (600°C) from the OG capsule series. Therefore, the main body of data for low-temperature irradiation dimensional changes has been taken from UKAEA results (Ref. 6-4) on a near-isotropic graphite (code No. PI) irradiated in the Dounreay Fast Reactor (DFR) at temperatures between 633 and 873 K (360° and 600°C). The UKAEA results were collected and analyzed in a review paper (Ref. 6-5). The UKAEA graphite was manufactured with near-isotropic coke and had a thermal expansivity at 293 to 393 K (20° to 120°C) of $3 \times 10^{-6} \text{ K}^{-1} (\text{°C}^{-1})$ parallel to extrusion (axial) and $4.1 \times 10^{-6} \text{ K}^{-1} (\text{°C}^{-1})$ perpendicular to extrusion (radial). Therefore, its dimensional change behavior is expected to be similar to that of H-451.

The data points from Ref. 6-4 are given in Appendix B, Figs. B-11 and B-12. The figures also include Dragon data on extruded near-isotropic graphites (graphite reference numbers 100 and 120 from Ref. 6-6). A few GA data points on H-451 graphite are also included. Agreement between the three sets of data is good.

No irradiation data below 360° to 400°C have been reported for near-isotropic graphites. However, results for PGA (needle-coke) graphite irradiated in DIDO and PLUTO at 573, 623, and 723 K (300°, 350°, and 450°C) are available

TABLE 6-1
WITHIN-LOG VARIATION IN IRRADIATION-INDUCED SHRINKAGE OF H-451 GRAPHITE, LOT 426

Orientation	Crucible	Irrad. Temp, K (°C)	Fast Neutron Fluence x 10 ⁻²⁵ [N/m ² (E > 29 fJ) _{HTGR}]	Mean Shrinkage (%)		Is Variation Significant at 95% Confidence Level?
				Center of Logs	Edge of Logs	
Axial	6	1210(937)	2.68	0.71	0.75	No
Radial	8	1110(837)	2.01	0.18	0.18	No
Radial	3	1200(927)	2.83	0.36	0.30	No

TABLE 6-2
BETWEEN-LOG VARIATIONS IN IRRADIATION-INDUCED SHRINKAGE OF H-451 GRAPHITE, LOT 426

Orientation	Crucible	Irrad. Temp, K (°C)	Fast Neutron Fluence x 10 ⁻²⁵ [N/m ² (E > 29 fJ) _{HTGR}]	Mean Shrinkage (%)			Is Variation Significant at 95% Confidence Level?
				Log 6484-33	Log 6484-34	Log 6484-41	
Axial	6	1210(937)	2.68	0.71	0.79	0.69	No
Radial	1	910(637)	1.84	0.18	0.16	0.14	No
Radial	8	1110(837)	2.01	0.17	0.20	0.19	No
Radial	3	1200(927)	2.83	0.28	0.36	0.36	No
Radial	6	1270(997)	2.68	0.79	0.82	0.79	No

(Ref. 6-7). The probable behavior of near-isotropic graphite at 573 K (300°C) can be predicted from the PGA data by calculating the volumetric shrinkage rates at 573 and 723 K (300°C and 450°C) and assuming that the ratio between the rates at these two temperatures is the same for near-isotropic graphite and PGA. This procedure was applied to the near-isotropic 723 K (450°C) experimental data in Figs. B-11 and B-12. The calculated 573 K (300°C) volumetric shrinkages were then converted to linear shrinkage by assuming that the ratio of axial to radial shrinkage is the same at 573 K (300°C) as at 723 K (450°C).

6.4. TENSILE STRENGTH, ELASTIC MODULUS, AND STRAIN AT FRACTURE

6.4.1. 823 to 1623 K (550° to 1350°C)

Approximately 500 tensile tests measured at ambient temperature were conducted on H-451 graphite specimens from lots 266, 408, and 426 irradiated at several different fluences and temperatures in capsules OG-1, -2, and -3.

The percent increases in tensile strength and elastic modulus (referenced to unirradiated control specimens from the same location in the same log) are plotted as functions of fast neutron fluence in Fig. B-13. For H-451 graphite, the data for all orientations, locations, and lots fall in the same scatter bands.

The statistical spread in strength values is an important parameter in the mechanical design of graphite structures. Irradiation causes a significant increase in the standard deviation. However, the coefficient of variation (standard deviation divided by the mean) is unaffected by irradiation. The estimated coefficient of variation showed no definite trend with irradiation.

Design curves presently used for the irradiation-induced increase in elastic modulus (Ref. 6-2) are based on Dragon project data (Ref. 6-6) for change in sonic elastic modulus of a near-isotropic pitch coke graphite. The Dragon data cover a wide range of temperatures and fluences (see Fig.

B-14). The GA data on H-451 graphite are in excellent agreement with the Dragon curves. The design curves as shown in Fig. B-13 from Ref. 6-2 will continue to be used.

A design method for accounting for the irradiation-induced strength increase utilizing all the available data is recommended (Ref. 6-3). The increase in strength S is assumed to be related to the irradiation-induced increase in elastic modulus E as follows:

$$S/S_0 = (E/E_0)^k, \quad (6-1)$$

where the exponent k depends on the graphite grade. An average k value of 0.64 for axial and radial specimens was obtained from the current H-451 data. This k value is reflected in the design curves for the strength data plotted in Fig. B-13.

6.4.2. 627 to 873 K (350° to 600°C)

Measurements of changes in elastic modulus (by a sonic method) and strength (tensile and brittle-ring tests) were obtained on near-isotropic graphite irradiated in the DFR under the UKAEA program (Refs. 6-4, 6-8). Results from Ref. 6-8 for material irradiated at 623 to 713 K (350° to 440°C) are shown in Appendix B, Fig. B-15. The fractional increase in strength equals the square root of the fractional increase in the elastic modulus, indicating that the strain energy to failure remains constant. Later data (Ref. 6-4) show that this relation holds to about $3 \times 10^{26} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} ($2 \times 10^{26} \text{ N/m}^2$ NDE), but the strength may decrease somewhat at higher fluences.

Dragon has reported (Ref. 6-6) changes in the sonic elastic modulus of near-isotropic graphites irradiated at 673 and 873 K (400° and 600°C) to fluences of 4.5 to $6 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} (3 to 4×10^{25} NDE). The Dragon data are shown in Appendix B, Fig. B-14. The 673 K (400°C) increases are consistent with the higher fluence data also shown in Fig. B-14. Design curves for the static elastic modulus have been derived from the Dragon data

(Ref. 6-6) using a multiple of 0.8 to convert dynamic modulus to static modulus (unirradiated graphite) and a multiple of 0.6 to convert fluence ($E > 29 \text{ fJ}$) to fluence (NDE) (Ref. 6-3). For irradiated graphite, the sonic and static elastic moduli were assumed equal. Curves for temperatures other than 673 and 873 K (400° and 600°C) were obtained by interpolation. The calculated design curves are shown in Appendix B, Fig. B-16. For the low-temperature data, the increase in strength was assumed equal to the square root of the increase in modulus (Ref. 6-5):

$$\left(\frac{S}{S_o} = \frac{E}{E_o} \right)^{1/2} . \quad (6-2)$$

However, Eq. 6-1 was used in the design data manual (GA-A14428; see Section 2) for the temperature range 623 to 1673 K (350° to 1400°C).

6.4.3. Between-Log and Within-Log Variations in Strength Increase

Three groups of tensile specimens of H-451 graphite were made up from three different logs from lot 426, and three groups contained specimens from both the MLC and the MLE. Five or more replicate specimens were used from each log and location. The irradiation-induced percent strength increases (referenced to control specimens from the same log and location) were analyzed by the analysis-of-variance method. Individual strength values are given in Ref. 6-3. The results of the comparison are summarized in Tables 6-3 and 6-4. Among one group of specimens, those from one log showed a significantly smaller increase than those from the other two logs, but this was not the case for the other two groups (see Table 6-3). There were no significant differences in the strength increases between specimens from the center of the log and specimens from the edge of the log (see Table 6-4).

TABLE 6-3
BETWEEN-LOG VARIATIONS IN IRRADIATION-INDUCED STRENGTH INCREASE OF H-451 GRAPHITE, LOT 426

Orientation	Crucible No.	Irrad. Temp, K (°C)	Fast Neutron ₂₅ Fluence x 10 ⁻²⁵ [N/m ² (E > 29 fJ) _{HTGR}]	Mean Strength Increase (%)			Is Variation Significant at 95% Confidence Level?
				Log 6484-33	Log 6484-34	Log 6484-41	
Radial	8	1110(837)	2.01	28	32	39	No
Radial	3	1200(927)	2.83	57	35	60	Yes
Axial	6	1210(937)	2.68	41	56	48	No

TABLE 6-4
WITHIN-LOG VARIATIONS IN IRRADIATION-INDUCED STRENGTH INCREASE OF H-451 GRAPHITE, AXIAL DIRECTION

Lot No.	Crucible No.	Irrad. Temp, K (°C)	Fast Neutron Fluence x 10 ⁻²⁵ [N/m ² (E > 29 fJ) _{HTGR}]	Mean Strength Increase (%)		Is Variation Significant at 95% Confidence Level?
				Center of Logs	Edge of Logs	
408	1	940(667)	4.6	33	43	No
408	8	1150(877)	4.1	31	44	No
426	6	1210(937)	2.7	50	46	No

6.5. FATIGUE BEHAVIOR

6.5.1. Sampling and Procedures

The experimental procedures described for unirradiated specimens in Section 5.4.2 were also used to test irradiated specimens. Specimens were taken from the same regions as those for irradiated tensile strength testing.

Three groups of H-451 specimens were irradiated in graphite crucibles in capsules OG-1, -2, and -3 (Refs. 6-1, 6-2, 6-3). The specimens were irradiated at 1173 to 1273 K (900° to 1000°C) to different neutron fluence exposures. Ten specimens out of each group were tensile-tested in the fatigue machine to establish the mean tensile strength. The values are listed in Appendix B, Table B-11, together with the strengths of unirradiated companion specimens from the same location in the log and the irradiation conditions. All groups showed the expected increase in strength after irradiation. The remainder of the specimens in each group were used for tension-compression fatigue tests ($R = -1$). Eight specimens were cycled at each of four stress levels until they failed or reached 100,000 cycles without failure.

6.5.2. Experimental Results and Discussion

A homologous stress S-N curve (analogous to the curve for unirradiated specimens in Fig. B-1) for axial specimens of H-451 graphite irradiated to $3.0 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} at 1173 K (900°C) is shown in Appendix B, Fig. B-17. Comparison between the results in Fig. B-17 and the unirradiated results (Fig. B-1) shows that the homologous stress limits for fatigue endurance are increased by neutron irradiation. The results of statistical analyses of the S-N curves for all the irradiated H-451 specimens are given in Table 6-5. The homologous stress limits for survival to 10^5 cycles are substantially higher than for unirradiated H-451 tested with $R = -1$ (Table 6-5), at least for the axial specimens. The homologous stress 99/95 lower tolerance limit is lower at higher fluences as the result of increased data scatter, but it remains above the unirradiated level. The increased scatter may be caused by slight warpage in the highly irradiated specimens, which interferes with accurate uniaxial alignment in the test rig.

TABLE 6-5
RESULTS OF STATISTICAL ANALYSIS OF FATIGUE DATA FOR H-451 GRAPHITE IRRADIATED AT
1173 to 1263 K (900° to 990°C) (STRESS RATIO, R = -1)

Orientation	Fast Neutron Fluence (10^{25} N/m ²) (HTGR)	Intercept of Least Squares Line $\alpha^{(a)}$	Slope of Least Squares Line $\beta^{(a)}$	Standard Deviation S(a)	Homologous Stress Limit $\sigma_{\max}/S$ for Survival to 10^5 cycles (a)	
					50% Probability	99/95 Lower Tolerance Limit
Axial	3.0	-0.010	-0.004	0.046	0.93	0.66
Radial	5.7	-0.020	-0.022	0.044	0.74	0.54
Axial	8.5	-0.033	-0.012	0.056	0.81	0.51

(a) Based on the model of Eq. 5-11.

If the fatigue stress limits are normalized by dividing by the unirradiated tensile strength instead of the irradiated tensile strength, the limits increase by more than a factor of two during irradiation. The stress limits for 50% survival to 10^5 cycles and the 99/95 lower tolerance limits normalized to the unirradiated strength are plotted against neutron fluence in Fig. 6-4.

6.6. THERMAL EXPANSIVITY

6.6.1. 823 to 1623 K (550° to 1350°C)

Thermal expansivity measurements were made on H-451 graphite (lots 266 and 426) and H-429 (Refs. 6-1, 6-2, 6-3). All except H-451 (lot 426) had been irradiated in previous capsules. Data on grades TS-1240 and S0818 are included (see Section 6.1). Measurements were made between room temperature and up to 100 K (°C) below the irradiation temperature.

The accumulated data on near-isotropic graphites H-451, H-429, TS-1240, and S0818 from capsules OG-1, -2, and -3 are summarized in Appendix B, Tables B-12 through B-15. The fractional change in thermal expansivity is plotted as a function of fast neutron fluence in Appendix B, Figs. B-18 and B-19. At the lowest irradiation temperatures [865 to 1045 K (592° to 772°C)] the thermal expansivity first increases by 5% to 10% and then slowly decreases. At higher irradiation temperatures, the thermal expansivity decreases by as much as 50% of its preirradiation value, with the decrease most marked at the highest irradiation temperatures. Data from all the near-isotropic graphites fall into the same band regardless of orientation and log location, with the exception of H-429, for which irradiation-induced changes in thermal expansivity are somewhat lower than the others.

For convenience in design calculations, the data for irradiation-induced fractional changes in thermal expansivity, $(\alpha_i - \alpha_o)/\alpha_o$, of the H-451 and other near-isotropic graphites were fit to the following polynomial (Ref. 6-9):

Fig. 6-4. Fatigue stress limits for survival of H-451 graphite to 10^5 cycles, normalized to the unirradiated tensile strength, versus the fast neutron fluence at an irradiation temperature of 1173 to 1263 K (900° to 990°C)

$$(\alpha_i - \alpha_o)/\alpha_o = (0.27830 - 4.2734 \times 10^{-4} T + 1.7815 \times 10^{-7} T^2)\phi - 2.0664 \times 10^{-2} \phi^2 + 1.3601 \times 10^{-3} \phi^3 \quad (6-4)$$

where T is the irradiation temperature ($^{\circ}\text{C}$) and ϕ is the neutron fluence [10^{25} N/m^2 ($E > 29 \text{ fJ}$)_{HTGR}]. Curves calculated from the equation are included in Appendix B, Figs. B-18 and B-19. The standard deviation of data points from the design curves is 0.063.

6.6.2. 623 to 823 K (350° to 550°C)

The thermal expansivity data of near-isotropic graphite samples irradiated in the DFR (Ref. 6-4) are reproduced in Appendix B, Fig. B-20. No irradiation temperature effect is apparent for the temperature range 623 to 823 K (350° to 550°C .)

The ratio of postirradiation thermal expansivity to preirradiation expansivity taken from Fig. B-20 is tabulated as a function of fast fluence in Table 6-6. The thermal expansivity of near-isotropic graphite irradiated at temperatures between 573 and 873 K (300° and 600°) are derived from the factors in Table 6-6.

6.7. THERMAL CONDUCTIVITY

6.7.1. 923 to 1623 K (650° to 1350°C)

Thermal diffusivity measurements were made on discs of H-451 (lots 266 and 426) graphite irradiated at three temperatures [(925, 1220, and 1615 K (652° , 947° , and 1342°C)] (Ref. 6-3). The H-451 specimens from lot 266 had been previously irradiated in earlier OG capsules (Refs. 6-1, 6-2). The measurements were made by the heat-pulse method and the thermal diffusivity value was converted to the thermal conductivity by multiplying by the density and heat capacity. Readings were taken every 100 K ($^{\circ}\text{C}$) between ambient temperature and 1073 K (800°), except for the specimens irradiated at 925 K (652°C), when the upper measurement temperature was limited to 773 K (500°C) to avoid the possibility of annealing.

TABLE 6-6
INCREASE IN THERMAL EXPANSIVITY OF NEAR-ISOTROPIC GRAPHITE
IRRADIATED BETWEEN 623 AND 823 K (350° AND 550°C)

Fast Fluence [10 ²⁵ N/m ² (E > 29 fJ) _{HTGR}]	Ratio of Postirradiation Expansivity to Preirradiation Expansivity α/α_0
0	1.00
1	1.09
2	1.13
3	1.15
4	1.18
5	1.19
6	1.20
7	1.21
8	1.22
9	1.21
10	1.20
11	1.20
12	1.19
13	1.18
14	1.18
15	1.17

The accumulated data on near-isotropic graphites H-451, TS-1240, and S0818 from capsules OG-1, -2, and -3 are summarized in Appendix B, Tables B-16, B-17, and B-18. The conductivity at the irradiation temperature is plotted as a function of fast neutron fluence in Fig. B-21. Irradiation rapidly reduces the thermal conductivity to a saturation level which increases with increasing irradiation temperature. The thermal conductivity of all the near-isotropic graphites tested falls into the same band after irradiation, even though some differences existed in their preirradiation conductivities. The irradiated values are in excellent agreement with the design equations currently in use (Ref. 6-2).

6.7.2. 523 to 923 K (250° to 650°C)

The thermal conductivity (at room temperature) of near-isotropic graphites irradiated in the DFR was measured by the UKAEA (Ref. 6-8). Changes in thermal resistivity for the grade (code-numbered PI, parallel to extrusion) are given in Ref. 6-8 and are replotted in Appendix B, Fig. B-22. Higher fluence results are contained in Ref. 6-4. A few data points at 807 to 923 K (435° to 650°C) on near-isotropic pitch-coke graphites were reported by Dragon in Ref. 6-6 and these points are also included in Fig. B-22.

The experimental data on near-isotropic graphites do not extend down to 573 K (300°C) and are not sufficient to show the shape of the curve at low fluences. However, a considerable amount of thermal conductivity data on PGA graphite irradiated between 523 and 923 K (250° and 650°C) is available from the AGR program (Refs. 6-10, 6-11). Curves taken from Ref. 6-10 showing the fractional change in thermal resistivity of PGA graphite are included in Appendix B, Fig. B-22. The solid curves correspond to experimental data, and the dashed curves are extrapolations. The data points for near-isotropic graphite are in good agreement with the PGA curves.

To obtain values useful for design, the fractional changes in thermal resistivity given by the smooth curves in Fig. B-22 were applied to the measured room temperature conductivity of H-451 graphite, interpolating

between the curves for calculations at intermediate temperatures. The conductivity at the irradiation temperature was calculated by assuming that for given irradiation conditions, irradiation adds the same increment to the thermal resistivity at all measurement temperatures (Ref. 6-12). The results of the calculations are given in Appendix B, Table B-19.

6.7.3. Effect of Change in Temperature

Two radial specimens of H-451 graphite which had been irradiated previously in capsule OG-1 at 1623 K (1350°C) were reirradiated in OG-3 at 923 K (650°C), and two specimens formerly irradiated at 873 K (600°C) were reirradiated at 1613 K (1342°C). In both cases the new conductivities were equal to the saturation level for the new irradiation temperature. This behavior is consistent with the currently used design model for the conductivity of graphite subjected to a changing irradiation temperature.

6.8. IRRADIATION-INDUCED CREEP

Two programs are currently under way on H-451: (1) compressive creep experiments at ORNL and (2) a tensile creep experiment at RCN and JRC Euratom, Petten. Creep coefficients for design have been taken from an analysis of restrained shrinkage experiments conducted at RCN, Petten, on H-451 graphite and a similar molded grade (AGL grade SMI-24) (Ref. 6-13). Irradiation creep in H-451 graphite may be represented by the equation:

$$\epsilon_{cr} = 4.16 \times 10^{-5} \sigma [1 - \exp(-5 \times 10^{-24} \gamma)] \exp(1.27 \times 10^{-3} T) + 6.2 \times 10^{-30} \sigma \gamma \exp(7.48 \times 10^{-4} T)$$

where ϵ_{cr} is the total creep strain (unitless),

σ is the applied stress, MPa,

γ is the fast neutron fluence, N/m^2 ($E > 29$ fJ)_{HTGR},

T is the temperature in K.

The equation applies between 773 and 1473 K (500° and 1200°C) to a fluence of 10^{26} N/m² ($E > 29$ fJ)_{HTGR} for tensile strains less than 2.5% or compressive strains less than 2%.

6.8.1. Conclusions

Several investigators have established that the form of the fast-neutron-irradiation-induced creep curve for graphites consists of a period of transient creep, which rapidly saturates, followed by steady-state creep, where the creep strain increases linearly with fluence. The only possible exception appears to be Gray's results (Refs. 6-14, 6-15) for compressive creep in isotropic graphites at 1073 K (800°C) where the creep rate slowed down at fluences above 6 to 7×10^{25} N/m² ($E > 29$ fJ)_{HTGR} when the compressive creep strain exceeded 2%. Another possible indication of reduced creep rates at high fluences may be inferred from the UKAEA restrained shrinkage results on isotropic graphites irradiated in the DFR.

REFERENCES

- 6-1. Price, R. J., and L. A. Beavan, "Final Report on Graphite Irradiation Test OG-1," USAEC Report GA-A13089, General Atomic Company, August 1, 1974.
- 6-2. Price, R. J., and L. A. Beavan, "Final Report on Graphite Irradiation Test OG-2," ERDA Report GA-A13556, General Atomic Company, December 15, 1975.
- 6-3. Price, R. J., and L. A. Beavan, "Final Report on Graphite Irradiation Test OG-3," ERDA Report GA-A14211, General Atomic Company, January 1977.
- 6-4. Kelly, B. T., et al., "Studies of Irradiation Damage in Carbons and Graphites Appropriate to MK II Gas-Cooled Reactors (AGRs) and the MK III Gas-Cooled Reactors (HTGRs)," Proceedings, Fourth UN Int. Conf. on Peaceful Uses of Atomic Energy, Geneva, 1971, v. 10, IAEA, Vienna, 1972, p. 399.

- 6-5. Price, R. J., "Property Changes in Near Isotropic Graphites Irradiated at 300°C to 600°C: A Literature Survey," ERDA Report GA-A13478, General Atomic Company, June 13, 1975.
- 6-6. Everett, M. R., et al., "Irradiation Performance and Selection of Graphites for HTGRs," Dragon Report DP-Report-877, April 1974.
- 6-7. Kelly, B. T., et al., "Dimensional Changes in Polycrystalline Graphite Under Fast-Neutron Irradiation," Phil. Trans. Roy. Soc. London, Ser. A, 260, 51 (1966).
- 6-8. Nettley, P. T., et al., "Irradiation Experiment with Isotropic Graphite," Proceedings, Symposium on Advanced and High Temperature Gas-Cooled Reactors, Jülich, October 21-25, 1968, IEAA, Vienna, 1969, p. 603.
- 6-9. Price, R. J., "Cyclic Fatigue of Near-Isotropic Graphite: Influence of Stress Cycle and Neutron Irradiation," DOE Report GA-A14588, General Atomic Company, December 1977.
- 6-10. Martin, W. H., and A. M. Price, "Determination of Dose and Temperature of Graphite Irradiation Experiments in the Dounreay Fast Reactor," J. Nucl. Energy 21, 359 (1967).
- 6-11. Bridge, H., et al., "Effect of High-Flux Fast-Neutron Irradiation on the Physical Properties of Graphite," Carbon 2, 83 (1964).
- 6-12. Price, R. J., "Thermal Conductivity of Neutron Irradiated Reactor Graphites," USAEC Report GA-A13157, General Atomic Company, October 8, 1974.
- 6-13. Veringa, H. J., and R. Blackstone, "Radiation Creep in Reactor Graphites for Reactor Applications," Carbon 14, 279 (1976).
- 6-14. Gray, W. J., "Constant Stress Irradiation Creep Experiments on Graphite," Proceedings of the Carbon - 72 Symposium, Baden-Baden, June 1972.
- 6-15. Gray, W. J., "Technical Activities Quarterly Progress Report, AEC Reactor Development and Technology Programs, Jan.-Mar., 1971," USAEC Report BNWL-1522-2, Pacific Northwest Laboratory, 1971, p. 8.1.

7. OXIDATION PROGRAM

Two types of water leaks are important in determining the effect of steam oxidation on the fuel element: (1) high water concentrations, up to 1000 kPa (10 atm), which may occur in the event of a large steam leak, and (2) low water concentrations, up to 50 Pa (500 μ atm), which could occur during normal operation. To assess the effects of water ingress as described above on the integrity of the fuel element graphite over the core life of the fuel elements, it is necessary to determine (1) oxidation rates of H-451 in steam-helium mixtures as a function of H_2O and H_2 concentration, reaction temperature, degree of burnoff, and sample configuration, and (2) changes in strength and elastic modulus due to oxidation burnoffs.

7.1. OXIDATION RATE OF H-451 GRAPHITE

7.1.1. Reaction Rate Theory

Reaction rate theory for steam-graphite reactions is discussed in detail in Refs. 7-1 and 7-2. The extent of the reaction of water vapor with the H-451 graphite in an HTGR is calculated (predicted) utilizing computer codes OXIDE-3 (Ref. 7-3) and GOP (Ref. 7-4) with appropriate input of reaction rate data.

The reaction of water vapor with graphite is controlled by chemical interactions and mass transport of the gaseous constituents within the pores of the graphite. Low concentrations of water vapor, up to 50 Pa (500 μ atm) water vapor and high concentrations, up to 1000 Pa (10 atm) are discussed.

Controversy still exists over the mechanisms involved in the reaction of water vapor with graphite, but there is general agreement that the chemical reactions can be represented by the Langmuir-Hinshelwood equation:

$$R = \frac{K_1 P_{H_2O} F_b F_c}{1 + K_2 P_{H_2}^n + K_3 P_{H_2O}} \quad (7-1)$$

where K_1 , K_2 , and K_3 are rate constants, n is a constant, P_{H_2O} and P_{H_2} are partial pressures of water vapor and hydrogen, respectively, F_b corrects the rate for the effect of burnoff, and F_c corrects the rate for the effect of catalytic reactions due to fission product impurities introduced into the graphite by irradiation. The effects of catalytic impurities have not been quantified in H-451 graphite; therefore, catalytic effects cannot be considered.

The oxidation of graphite by water vapor consists of three distinct processes* (Refs. 7-5, 7-6, 7-7): (1) sorption of water molecules on active carbon sites, (2) chemical reaction of the sorbed water to form H_2 and CO , and (3) inhibition of water sorption by competitive sorption of H_2 molecules. Process (1) can be expressed by

where C_f and $C(H_2O)$ denote free and occupied sites and i and j are reaction rate constants for the forward and reverse reactions. Following process 7-2, sorbed water reacts with the solid phase to form carbon monoxide and hydrogen as expressed by

* Transport considerations are neglected in this treatment.

where a is an integer having values from 0 to 2. If $a > 1$ (observed in the earlier stages of oxidation), the number of reaction sites increases as the reaction proceeds, leading to increased reaction rates. If $a < 1$ (observed only during the final stages of the oxidation process), the graphite becomes less reactive as the reaction proceeds. Thus, the reaction rate is affected by burnoff (i.e., the extent of oxidation).

Hydrogen molecules compete with water vapor molecules for the available sites (Refs. 7-5, 7-6). Thus, included in the denominator of Eq. 7-1 is a term which reflects the rates of adsorption of hydrogen on graphite (process 7-3) as expressed by

These three processes form the basis for the Langmuir-Hinshelwood equation:

$$\text{rate} = \frac{\frac{i_1 j_3}{j_1 + j_3} P_{H_2O}}{1 + \frac{i_2}{j_2} P_{H_2}^n + \frac{i_1}{j_1 + j_3} P_{H_2O}} \quad . \quad (7-5)$$

If j_1 , the rate of evaporation of water molecules from the surface, is negligible, $K_1 = i_1$, $K_2 = i_2/j_2$, and $K_3 = i_1/j_3$.

7.1.2. Experimental Procedure and Sampling

Instantaneous reaction rates are measured by specimen weight-loss determinations using automatic recording microbalances (Ref. 7-1).

Specimens were taken from near the center of a log of preproduction H-451 graphite. The density of the specimens ranged from 1.69 to 1.70 Mg/m³. The total metallic impurity content of the graphite was less than 200 ppm and iron ranged from < 1 to 6 ppm.

Values of the constant n and rate constants K_1 , K_2 , and K_3 are obtained from the slopes of isothermal plots of the reaction rate data derived from Eq. 7-1. Because all three rate constants increase with burnoff, values of the constants were corrected to a constant burnoff of 1%.

7.1.3. Determination of Reaction Rate Constants

Values were determined for reaction rate constants: (1) burnoff factor F_b , (2) hydrogen exponent n , and (3) K_1 , K_2 , and K_3 . The determinations of these constants from experimental data are given in Refs. 7-1 and 7-2. Recommended values are listed in Table 7-1.

7.1.3.1. Burnoff Factor F_b . The burnoff factor F_b is determined from plots of relative reaction rate versus percent burnoff, where the rate data are normalized to unity at 1% burnoff (Ref. 7-1).

TABLE 7-1
EXPERIMENTALLY DETERMINED LANGMUIR-HINSHELWOOD
CONSTANTS FOR H-451 GRAPHITE^(a)

Constant	Partial Pressure Water (Pa)	
	Low ≤ 300	High $\geq 300 \leq 3300$
$K_1 \text{ sec}^{-1} \text{ Pa}^{-1}$	218 exp (-253500/RT)	0.11 exp(-195400/RT)
$K_2 \text{ Pa}^{-0.75}$	1940 exp(-131900/RT)	8.2×10^{-8} exp(119700/RT)
$K_3 \text{ Pa}^{-1}$	1.02 exp(-47030/RT)	1.3×10^{-9} exp(131400/RT)
Hydrogen exponent n	0.75	0.75

(a) Langmuir Hinshelwood equation:

$$\text{rate (sec}^{-1}\text{)} = \frac{K_1 P_{H_2O}}{1 + K_2 P_{H_2}^{0.75} + K_3 P_{H_2O}}$$

The increase in reaction rate with burnoff is more rapid at lower temperatures. For example, at 5% burnoff, F_b is 4, 2.6, and 2.5 for 1093, 1173, and 1253 K, (820°, 900°, and 980°C) respectively.

In the reactor, because of high helium pressures, burnoff is largely surface oriented, and the F_b term is likely to be quite different from that found in these laboratory experiments.

7.1.3.2. Hydrogen Exponent n. The determined value of n is 0.695 ± 0.18 (Ref. 7-1), and this value is in accord with $n = 0.75$ currently used in the OXIDE-3 code (Ref. 7-3).

7.1.3.3. Rate Constants K_1 , K_2 , and K_3 . The rate constant K_1 is derived from the slope of the plot of $1/R$ versus $1/P_{H_2O}$, where R is the rate of reaction and P_{H_2O} is the partial pressure of water. It is essential that the partial pressure of hydrogen (P_{H_2}) be negligible when compared with P_{H_2O} in order to obtain a valid value for K_1 . Therefore, P_{H_2} was either excluded or held near zero during the experiments. Plots of $1/R$ versus $1/P_{H_2O}$ derived from experiments carried out under these conditions are reported in Ref. 7-2 for three temperatures [1093, 1173, and 1253 K (820°, 900°, and 980°C)]. The plots in Ref. 7-2 summarize the data on reaction rates using disk-shaped samples 31.8 mm in diameter by 1.57 mm thick. Previous work (Ref. 7-1) showed samples of these dimensions to exhibit minimal mass transport control of the reactions.

A least-squares fit (POLFIT) was used to generate the statistical data required to calculate the variances of slope and intercept. An apparent activation energy was calculated using POLFIT with each data point weighted by its variance using the methods given previously in Ref. 7-1.

The rate constant K_2 is derived from the slope of a plot of $1/R$ versus P_{H_2} . The slope is equal to $K_2/K_1 P_{H_2O}$; therefore, K_2 can be determined using known values of K_1 and n . The value of n (see above) was determined to be

0.695 ± 0.18. A similar value for n was used for deriving the constants used herein. Another assumption used in determining K_2 by this method is that errors associated with K_1 are negligible. As a precaution, propagational error theory was employed to examine the contributions of errors in K_1 to the variance of K_2 .

The experimental data used to obtain the required slopes are given in Ref. 7-2. A composite of data from several experiments was used to develop the 50-Pa water vapor isobar. The apparent activation energy and pre-exponential factor were estimated from the Arrhenius relation (Ref. 7-2).

Establishment of equilibrium for the competitive reaction

is slow, particularly at low temperatures, and, consequently, data at temperatures below 1123 K (850°C) are limited. When additional data are collected, values of K_2 can be determined at temperatures of interest down to about 1000 K (727°C).

The intercept values from plots of $1/R$ versus $1/P_{\text{H}_2\text{O}} \approx 0$ were used to determine K_3 . By rearranging the Langmuir-Hinshelwood equation, the intercept value of the above relation can be equated to K_1/K_3 . However, K_3 is also dependent on the errors associated with K_1 . The expression recommended for calculating K_3 over the partial pressures examined is given in Ref. 7-2.

7.1.4. Application of Rate Constants

A plot of log rate versus log $P_{\text{H}_2\text{O}}$ which compares both sets of derived constants for H-451, along with the constants presently used in the OXIDE code, is shown in Fig. 7-1. The OXIDE code constants were derived from a composite of data from other graphites (Ref. 7-1), primarily measured at low water concentrations; therefore, the OXIDE code constants closely match the H-451 constants derived herein for $P_{\text{H}_2\text{O}}$ less than 300 Pa. An additional

Fig. 7-1. Comparison of rate constants as a function of P_{H₂O} (from Ref. 7-2)

problem with the OXIDE code has been its predictions of a near-zero-order relation at partial pressures of water that are too low (see Fig. 7-1). The use of H-451 constants derived for high water vapor will improve the reliability of the OXIDE code. For accident conditions, where moisture concentrations in the range of 1 to 1000 kPa are possible, the high-water-level H-451 rate constants should be used. For normal steady-state reactor operation, where water concentrations in the range 10 to 1000 Pa are expected, the low-water-level H-451 constants should be used.

7.2. EFFECT OF STEAM OXIDATION ON TENSILE STRENGTH AND ELASTIC MODULUS

The fuel element blocks are subject to mechanical stresses including compressive, due to the weight of the core components, and tensile, due to irradiation-induced strains and thermal gradient effects. The fuel element blocks may have to withstand additional stresses resulting from seismic events. The fuel element blocks are required to withstand these in-service stresses and retain their structural integrity. Thus, a knowledge of the effect of oxidation on the strength and elastic modulus is required.

7.2.1. Experimental Procedure and Sampling

The experimental work (Ref. 7-8) consisted of oxidizing large numbers of small test specimens uniformly and isothermally at 1073 and 1273 K (800° and 1000°C) in steam-helium mixtures, and subsequently measuring the strength and elastic modulus of the oxidized specimens. The strength and elastic modulus values of the oxidized specimens were compared with values of non-oxidized control specimens, taken from the same population of specimens used for oxidation, to determine changes due to oxidation burnoff.

In an effort to obtain data on material of highest and lowest strength, specimens were selected from quarter-length center (QLC) and quarter-length edge (QLE) positions of GA log 6384-34. Axial and radial specimens were selected as indicated in Appendix A, Figs. A-14 through A-17.

Groups of samples were randomized and allocated for oxidation according to Appendix B, Table B-19. Oxidation specimens were 634 mm in length. After oxidation, approximately 0.6 mm was removed from each end of each oxidized specimen and each specimen cut in half to provide two specimens 6.34 mm in diameter by approximately 2.3 mm in length.

The apparatus used for the oxidation measurements is described in Ref 7-8. Several of the cylindrical graphite specimens were oxidized simultaneously in each experiment. A gas mixture composed of 3% water vapor, 5% hydrogen, and the balance helium was used for oxidizing the specimens.

To determine the extent of oxidation, the oxidation process was interrupted periodically and the samples were cooled and weighed to ± 0.1 mg. The percent burnoff was calculated from changes in specimen weight.

It was anticipated that use of small-diameter samples in a hydrogen-rich gas mixture would ensure relatively uniform oxidation throughout the bulk of the samples. Uniformity of oxidation burnoff was confirmed by determining radial density profiles on a number of nonoxidized and oxidized specimens. Techniques for measuring tensile strength and elastic modulus are reported in Ref. 7-9.

7.2.2. Experimental Results

Complete data sets of ultimate tensile strength, elastic modulus, and strain at fracture are tabulated in Ref. 7-8.

7.2.2.1. Oxidation-Induced Bulk Density Profiles. Bulk density profiles for nonoxidized control specimens of H-451 and for three oxidized specimens are given in Ref. 7-8. There is large scatter in the data, which is due to experimental difficulties and the heterogeneous nature of the graphite.

The oxidation profiles are uniform for all specimens oxidized at 1073 K or 1273 K (800° or 1000°C). It was anticipated that some preferential oxidation on the outer surface of the specimens would occur at 1273 K (1000°C);

that is, the final density profile along the radius after oxidation should increase from the outer edge toward the center. In most cases, oxidation at 1273 K (1000°C) produced uniform profiles. It is concluded from the bulk density profile data that all UTS specimens were oxidized uniformly across the radius.

7.2.2.2. Ultimate Tensile Strength, Elastic Modulus, and Strain at Fracture.

The UTS and elastic modulus values for nonoxidized control material were highest in the axial direction and increased from the center to the edge of the log. The ultimate tensile strengths and elastic modulus were in agreement with other data obtained on the same log and other logs of H-451 graphite (Ref. 7-9).

Statistical analyses were performed on the data to (1) determine whether the distribution of UTS was Gaussian (using the chi-square analysis) (Refs. 7-10, 7-11), (2) compare population mean values of strength and elastic modulus for control and oxidized specimens [using a comparison of population means (CPM) test] (Ref. 7-12), and (3) determine the maximum, minimum, and most probable (mean) rates of property change with burnoff at 95% confidence (using the CONFID computer code, derived from a technique given in Ref. 7-12).

The UTS data followed a Gaussian distribution. Confirmation of a Gaussian distribution is important since both the CPM test and the CONFID computer code assume Gaussian distributions.

The results of the CPM and CONFID analyses are given in Tables 7-2 and 7-3, respectively. The rates of changes in UTS and E from the CONFID calculations are presented graphically in Ref. 7-8, where the most probable rate changes are shown as linear regression lines. A comparison of nonoxidized and oxidized UTS values showed that strength decreased with oxidation. The population means of the control and oxidized specimens were compared (see Table 7-2, column 7). No significant reduction in UTS or modulus (at 95% confidence) was observed in axial specimens oxidized up to 2% and 1% burn-off, respectively.

TABLE 7-2
SUMMARY OF UTS AND ELASTIC MODULUS DATA AND COMPARISON OF POPULATION
MEANS OF NONOXIDIZED AND OXIDIZED SPECIMENS

Location and Orientation (a)	Oxidation		Ultimate Tensile Strength				Elastic Modulus			
	Temp, K (°C)	Burnoff %	MPa		No. of Data Points	CPM Test Results (b)	GPa		No. of Data Points	CPM Test Results (b)
			Mean	Std. Dev.			Mean	Std. Dev.		
CA	--	0	12.9	1.8	19	--	7.5	1.1	8	--
CA	1073 (800)	0.17-0.36	13.4	1.7	12	NSD	7.2	0.6	4	NSD
CA	1073 (800)	0.49-0.69	13.2	1.5	16	NSD	7.9	0.77	13	NSD
CA	1073 (800)	0.86-1.60	12.5	1.4	37	NSD	6.9	1.1	19	SD
CA	1273 (1000)	0.99-1.30	13.4	1.2	14	NSD	7.1	0.6	8	NSD
CA	1273 (1000)	1.47-1.90	12.5	1.5	16	NSD	7.0	1.0	8	NSD
CA	1273 (1000)	5.38-7.83	10.7	1.3	20	SD	5.8	0.7	8	SD
CR	--	0	12.2	1.3	20	--	6.9	0.6	9	--
CR	1073 (800)	0.25-0.33	10.6	1.1	12	SD	--	--	--	--
CR	1073 (800)	8.4 -10.0	6.1	0.9	12	SD	--	--	--	--
CR	1273 (1000)	10.2-14.5	6.9	1.2	12	SD	--	--	--	--
CR	1273 (1000)	15.7-19.6	4.2	1.3	9	SD	--	--	--	--
ER	--	0	16.1	1.8	18	--	8.0	0.8	11	--
ER	1073 (800)	0.49-0.59	10.7	1.3	12	SD	--	--	--	--
ER	1073 (800)	10.5-13.8	7.4	1.1	12	SD	--	--	--	--
ER	1273 (1000)	3.4 - 4.8	13.0	1.2	9	SD	5.1	0.6	4	SD
ER	1273 (1000)	5.8 - 7.6	10.9	1.3	10	SD	4.3	0.4	4	SD
ER	1273 (1000)	3.4 - 7.6	11.9	1.6	19	SD	4.7	0.7	8	SD
EA	--	0	18.9	1.7	15	--	8.6	1.4	12	--
EA	1073 (800)	1.0 - 2.5	16.8	1.4	24	SD	--	--	--	--
EA	1273 (1000)	3.3 - 5.2	14.8	1.1	20	SD	6.3	0.7	8	SD

(a) CA = center axial specimens, CR = center radial specimens, EA = edge axial specimens, ER = edge radial specimens.

(b) NSD denotes no significant difference (95% confidence) between property averages of controls and oxidized samples,
SD denotes a significant difference (95% confidence) between property averages of controls and oxidized samples.

TABLE 7-3
REDUCTION IN ULTIMATE TENSILE STRENGTH AND ELASTIC MODULUS
WITH OXIDATION

Location ^(a)	Burnoff Temp, K (°C)	Burnoff Range %	% Reduction/1% Burnoff		
			Maximum (2.3% Probability)	Mean (Most Probable)	Minimum (2.3% Probability)
	Ultimate Tensile Strength				
CA	1073 (800)	0-2	11.17	5.60	0.0
CA	1273 (1000)	0-2	7.32	1.73	0.0
CA	1273 (1000)	0-8	3.85	2.82	1.70
EA	1273 (1000)	0-6	6.48	5.14	3.74
CR	1073 (800)	0-10	6.06	5.32	4.32
ER	1073 (800)	0-14	5.14	4.60	3.23
CR	1273 (1000)	0-20	3.92	3.59	3.18
ER	1273 (1000)	0-8	5.93	4.79	3.72
	Elastic Modulus				
CA	1073 (800)	0-2	14.04	10.46	0.01
CA	1273 (1000)	0-2	11.56	3.71	0.0
CA	1273 (1000)	0-8	4.88	3.26	1.45
EA	1273 (1000)	0-5	9.53	6.10	1.96
ER	1273 (1000)	0-6	9.31	7.83	6.11

- (a) CA: center axial specimens.
EA: edge axial specimens.
CR: center radial specimens.
ER: edge radial specimens.

The effect of oxidation on UTS for axial edge and center specimens oxidized at 1073 K (800°C) is shown in Fig. 7-2. The data in Fig. 7-2 indicate no decrease in strength occurred in the center specimens up to 2% burnoff, whereas a slight decrease was observed in the edge specimens. The UTS reduction rates for oxidation at 1273 K (1000°C) are shown in Fig. 7-3. Again, the edge specimens lost strength at a higher rate than the center specimens. The combined data for axial center samples at low burnoff are shown in Fig. 7-4. No strength reduction was observed for burnoffs up to 2% at either 1073 or 1273 K (800° or 1000°C).

The UTS data for radial edge and center specimens are presented in Figs. 7-5 and 7-6 for oxidation at 1073 and 1273 K (800° and 1000°C), respectively. The radial specimens lost strength at a faster rate than axial specimens, and in both cases edge specimens lost strength at a faster rate than center specimens.

The effect of oxidation on elastic modulus of axial center specimens at 1073 and 1273 K (800° and 1000°C) is given in Fig. 7-7. No effect of burnoff up to 1% was observed, but above 1% the reduction of the elastic modulus was significant. Changes in modulus of axial center and edge specimens may be compared in Fig. 7-8. The modulus of the edge specimens is reduced at a higher rate than that of center specimens. Figure 7-9 is a plot of elastic modulus versus burnoff for radial specimens oxidized at 1273 K (1000°C).

The trends for UTS and elastic modulus are summarized in Table 7-3 by comparing the relative reduction in each property at 1% burnoff. The results in Table 7-3 were obtained from CONFID, which can be used to calculate, at 95% confidence, the most probable (50% probability) or mean decrease in property and the maximum and minimum (2.3% probability) decrease in property. Reductions in strength and elastic modulus were greater when the specimens were oxidized at 1073 K (800°C) compared to 1273 K (1000°C), and changes were greater for edge specimens than for center specimens. This phenomenon was particularly significant in modulus data where the average decrease in edge specimens at 1273 K (1000°C) was twice that of center specimens.

Fig. 7-2. Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial edge and center specimens) oxidized at 1073 K (800°C)

Fig. 7-3. Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial specimens) oxidized at 1273 K (1000°C)

Fig. 7-4. Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length axial center specimens)

Fig. 7-5. Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length radial specimens) oxidized at 1073 K (800°C)

Fig. 7-6. Tensile strength versus burnoff for H-451 graphite (log 6484-34, quarter-length radial specimens) oxidized at 1273 K (1000°C)

Fig. 7-7. Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length axial center specimens)

Fig. 7-8. Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length axial specimens) oxidized at 1273 K (1000°C)

Fig. 7-9. Elastic modulus versus burnoff for H-451 graphite (log 6484-34, quarter-length radial edge specimens) oxidized at 1273 K (1000°C)

Changes in strain at fracture were measured on nonoxidized control specimens and on specimens oxidized to burnoffs up to 12%. The data were anomalous, showing no changes in axial center specimens at 4.3% burnoff, about 50% reduction in radial center specimens at 2.8% to 9.3% burnoffs, and about 40% reduction in radial edge specimens at 5.5% to 12% burnoffs. No firm conclusions can be drawn from these data.

7.2.2.3. Conclusions. The tensile strength of H-451 graphite decreased by an average of 3.6% in center axial specimens when oxidized to burnoffs up to 2.0% at either 1073 K or 1273 K (800° or 1000°C), and no significant reduction in elastic modulus in these same specimens was observed when oxidized to burnoffs of up to 2.0% at 1273 K (1000°C). The rate of reduction of tensile strength and elastic modulus for center axial specimens was greater at 1073 K (800°C) than at 1273 K (1000°C) and, in general, the reduction was higher in radial specimens than in axial specimens.

In general, the reduction rate of elastic modulus with burnoff was greater than that of tensile strength. The rate of reduction of tensile strength and elastic modulus for axial specimens oxidized at 1273 K (1000°C) was greater at the edge than at the center of a log.

REFERENCES

- 7-1. "Quarterly Progress Report for the Period Ending August 31, 1976, HTGR Fuels and Core Development Program," ERDA Report GA-A14046, General Atomic Company, September 29, 1976.
- 7-2. "Quarterly Progress Report for the Period Ending May 31, 1977, HTGR Fuels and Core Development Program," ERDA Report GA-A14418, General Atomic Company, June 1977.
- 7-3. Perroomian, M. B., A. W. Barsell, and J. C. Saeger, "OXIDE-3: A Computer Code for Analysis of HTGR Steam or Air Ingress Accidents," General Atomic Report GA-A12493, January 15, 1972.
- 7-4. Giberson, R. C., and G. L. Tingey, "Reactions of Gaseous Impurities in a High Temperature Gas Cooled Reactor," USAEC Report BNWL-974, 1968.

- 7-5. Johnstone, H. F., C. Y. Chen, and D. S. Scott, "Kinetics of the Steam Carbon Reaction in Porous Graphite Tubes," Ind. Eng. Chem. 44, 1568 (1952).
- 7-6. Walker, P. L., Jr., F. Rusinko, Jr., and L. G. Austin, "Gas Reactions of Carbon," Advanced Catalysis 11, 149 (1959).
- 7-7. Ergun, S., and M. Menken, "Reactions of Carbon with CO₂ and Steam," in Chemistry and Physics of Carbons, v. 1, P. L. Walker, Jr. (ed.), Marcel Dekker, New York, 1965, p. 229.
- 7-8. Velasquez, C., et al., "The Effect of Steam Oxidation on Strength and Elastic Modulus of Graphite H-451," DOE Report GA-A14657, General Atomic Company, December 1977.
- 7-9. Johnson, W. R., and G. B. Engle, "Properties of Unirradiated Fuel Element Graphites H-451 and TS-1240," ERDA Report GA-A13752, General Atomic Company, January 31, 1976.
- 7-10. Bevington, P. R., Data Reduction and Error Analysis for the Physical Sciences, McGraw-Hill, New York, 1969, pp. 84 and 314.
- 7-11. Johnson, N. L., and F. C. Leone, Statistics and Experimental Design, v. 1, John Wiley and Sons, New York, 1964, p. 224.
- 7-12. Bevington, P. R., op. cit., p. 104.

8. ACKNOWLEDGMENTS

The author gratefully acknowledges the contributions of R. J. Price, L. A. Beavan, and R. D. Burnette in helping to summarize the data and in commenting on the manuscript.

APPENDIX A

SAMPLING DIAGRAMS

Fig. A-1. Sampling diagram of H-451 for within-log property measurements: slabs and sections

Fig. A-2. Sampling diagram of H-451 for within-log property measurements: center section showing specimen locations

Fig. A-3. Sampling diagram of H-451 for within-log property measurements: edge section showing specimen locations

Fig. A-4. Orientation of orthogonal axes for definition of Poisson's ratio data in Table B-4

Fig. A-5. Locations of slabs (shaded) for statistical strength testing H-451 graphite (log 5651-90)

Fig. A-6. Coring plan for center zone of A and B sections of slabs (see Fig. A-5)

Fig. A-7. Coring plan for edge zone of A and B sections of slabs (see Fig. A-5)

Fig. A-8. Sampling diagram of H-451 logs for tensile strength

Fig. A-9. Sampling diagram of H-451 log sample core (see Fig. A-8): axial specimens

Fig. A-10. Sampling diagram of H-451 log slab (see Fig. A-8): radial specimens

Fig. A-11. Coring diagram for unirradiated H-451 fatigue specimens: edge zone, from slab 2 (see Fig. A-5)

Fig. A-12. Sampling diagram for irradiation specimens: center zone, from slab 3 (see Fig. A-1)

Fig. A-13. Sampling diagram for irradiation specimens: outer zone, edge section from slab 3 (see Fig. A-1)

Fig. A-14. Sampling plan of graphite log showing slab designated for oxidation tests

Fig. A-15. Sampling plan of H-451 showing sectioning of a slab from Fig. A-14

Fig. A-16. Sampling diagram of H-451 graphite: center section of slab from Fig. A-15

Fig. A-17. Sampling diagram of H-451 graphite: edge section of slab from Fig. A-15

APPENDIX B

EXPERIMENTAL RESULTS

TABLE B-1
BULK DENSITY: H-451, LOT 426

Log No.		Whole Log (a) Density ₃ (Mg/m ³)	Mean Bulk Density (Mg/m ³) (Standard Deviation, Mg/m ³) [No. of Replicates]				Mean Log (b) Density ₃ (Mg/m ³)
GA	GLCC		MLC	MLE	EC	EE	
6484-33	92	1.75	1.707 (0.008) [16]	1.737 (0.015) [16]	1.733 (0.013) [16]	1.753 (0.009) [16]	1.739
6484-34	198	1.73	1.711 (0.003) [16]	1.722 (0.013) [16]	1.709 (0.010) [16]	1.722 (0.004) [16]	1.719
6484-40	155	1.72	1.703 (0.002) [16]	1.726 (0.008) [16]	1.725 (0.007) [16]	1.717 (0.006) [16]	1.720
6484-41	184	1.72	1.695 (0.008) [16]	1.718 (0.013) [16]	1.685 (0.012) [16]	1.707 (0.018) [16]	1.707
Mean, lot 426		1.73 [4 logs]	1.704 [4 logs]	1.726 [4 logs]	1.713 [4 logs]	1.725 [4 logs]	1.721

(a) Measured by GLCC.

(b) Calculated from small specimens.

TABLE B-2
ULTIMATE TENSILE STRENGTH: H-451, LOT 426

Log No.		Mean Strength (MPa)(Standard Deviation, MPa) [No. of Replicates]							
		Axial				Radial			
GA	GLCC	MLC	MLE	EC	EE	MLC	MLE	EC	EE
6484-33	92	14.54 (1.5) [19]	20.16 (2.3) [15]	18.44 (1.3) [20]	20.14 (1.6) [19]	10.44 (1.5) [20]	14.04 (2.5) [19]	15.87 (2.4) [20]	17.47 (1.2) [20]
6484-34	198	14.18 (0.8) [20]	19.42 (1.8) [20]	14.10 (0.9) [20]	18.62 (1.9) [20]	10.38 (2.1) [18]	13.69 (2.9) [18]	12.31 (3.0) [20]	14.74 (2.4) [19]
6484-40	155	12.99 (1.6) [20]	18.04 (1.1) [19]	16.67 (1.0) [20]	16.83 (1.1) [19]	11.72 (1.4) [20]	14.13 (1.6) [20]	15.38 (0.8) [18]	15.14 (1.0) [20]
6484-41	184	12.93 (1.2) [20]	18.66 (1.6) [19]	11.86 (0.8) [20]	16.53 (1.4) [20]	10.44 (2.9) [20]	13.75 (1.7) [20]	12.33 (2.0) [20]	13.43 (1.9) [19]
Mean, lot 426		13.65 (1.6) [79]	19.01 (1.9) [73]	15.26 (2.7) [80]	18.01 (2.1) [78]	10.75 (2.1) [78]	13.91 (2.2) [77]	13.94 (2.6) [78]	15.22 (2.2) [78]

TABLE B-3
ELASTIC MODULUS IN TENSION: H-451, LOT 426

Log No.		Mean Modulus (GPa)(Standard Deviation, GPa) [No. of Replicates]									
		Axial					Radial				
GA	GLCC	MLC	MLE	EC	EE	MLC	MLE	EC	EE		
6484-33	92	8.13 (0.21) [8]	9.03 (0.28) [8]	8.41 (0.21) [8]	9.03 (0.21) [8]	6.76 (0.21) [8]	7.24 (0.41) [8]	7.79 (0.21) [8]	7.65 (0.21) [8]		
6484-34	198	8.20 (0.14) [8]	9.03 (0.21) [8]	8.13 (0.21) [8]	8.76 (0.55) [8]	6.83 (0.34) [8]	7.72 (0.76) [8]	7.51 (0.48) [8]	7.58 (0.28) [8]		
6484-40	155	7.86 (0.21) [8]	8.67 (0.21) [8]	8.55 (0.07) [8]	8.96 (0.76) [8]	7.03 (0.14) [8]	7.31 (0.21) [8]	7.45 (0.21) [8]	7.51 (0.35) [8]		
6484-41	184	7.58 (0.24) [8]	8.62 (0.21) [8]	7.31 (0.21) [8]	8.00 (0.48) [8]	6.89 (0.14) [5]	7.17 (0.28) [8]	6.82 (0.21) [8]	7.03 (0.35) [9]		
Mean, lot 426		7.93 (0.28) [32]	8.82 (0.28) [32]	8.13 (0.21) [32]	8.69 (0.62) [32]	6.89 (0.28) [29]	7.38 (0.48) [32]	7.38 (0.48) [32]	7.45 (0.41) [33]		

TABLE B-4
POISSON'S RATIO: H-451, LOT 266

Log No.		Location of Specimen in Log	Direction of Applied Stress	Direction of Transverse Strain	Poisson's Ratio (Standard Deviation) [No. of Measurements]				
GA	GLCC				$\nu_{13} = \nu_{23}$	$\nu_{12} = \nu_{21}$	$\nu_{31} = \nu_{32}$	ν_{32}	ν_{31}
5651-63	22	MLC	Radial	Axial Radial	0.120 (0.001) [3]	0.137 (0.001) [3]			
5651-63	22	MLC	Radial	Axial Radial	0.096 (0.019) [10]	0.104 (0.011) [10]			
5651-63	22	MLC	Radial	Axial Radial	<u>0.112 (0.002) [64]</u>	<u>0.107 (0.006) [64]</u>			
Mean Mean				Axial Radial	0.110 (0.009) [77]	0.108 (0.008) [77]			
5651-63	22	MLC	Axial	Radial			0.125 (0.009) [6]		
5651-28	18	MLC	Axial	Radial			0.130 (0.019) [24]		
5651-63	22	MLC	Axial	Radial			<u>0.125 (0.008) [48]</u>		
Mean				Radial			0.127 (0.013) [78]		
5651-28	18	MLE	Axial	Radial (a) Chord				0.123 (0.009) [12]	0.113 (0.007) [12]
5651-63	22	MLE	Axial	Radial (a) Chord				<u>0.121 (0.003) [64]</u>	<u>0.114 (0.006) [64]</u>
Mean Mean				Radial (a) Chord				0.121 (0.005) [76]	0.114 (0.006) [76]
5651-63	22	EE	Axial	Radial (a) Chord				0.110 (0.010) [40]	0.117 (0.006) [40]

(a) Perpendicular to radial direction.

TABLE B-5
FLEXURAL STRENGTH: H-451, LOT 426

Log No.		Mean Strength (MPa)(Standard Deviation, MPa) [No. of Replicates]							
		Axial				Radial			
GA	GLCC	MLC	MLE	EC	EE	MLC	MLE	EC	EE
6484-40	155	20.9 (1.5) [20]	25.4 (1.2) [20]	23.7 (1.5) [20]	18.3 (4.6) [20]	17.1 (4.2) [20]	22.2 (1.2) [20]	21.7 (1.7) [20]	22.5 (1.0) [20]
6484-41	184	19.8 (1.6) [20]	25.1 (1.1) [20]	16.8 (1.3) [20]	22.3 (1.3) [20]	17.7 (1.4) [20]	21.1 (1.3) [20]	18.4 (1.1) [20]	19.6 (1.7) [20]
Mean, lot 426		20.3 (1.7) [40]	25.3 (1.1) [40]	20.2 (3.8) [40]	20.3 (3.9) [40]	17.4 (3.1) [40]	21.7 (1.4) [40]	20.0 (2.2) [40]	21.1 (2.0) [40]

TABLE B-6
ULTIMATE COMPRESSIVE STRENGTH AND ELASTIC MODULUS IN COMPRESSION: H-451, LOT 478

Log No.		Axial				Radial			
GA	GLCC	MLC	MLE	EC	EE	MLC	MLE	EC	EE
Ultimate Compressive Strength (MPa)									
7194-49	15	50.3	58.9	49.2	55.2	52.4	54.6	49.4	55.9
7194-52	72	51.8	58.9	50.0	58.3	48.3	54.5	52.2	57.8
7194-87	35	53.7	60.5	53.6	62.9	48.2	50.3	53.1	56.1
Lot mean		51.9	59.4	50.9	58.8	49.6	53.1	51.6	56.6
Standard deviation		2.7	1.9	3.1	5.0	2.7	3.0	3.1	2.2
Elastic Modulus in Compression (GPa)									
7194-49	15	6.0	6.8	6.2	6.9	5.2	5.4	5.4	5.7
7194-52	72	6.4	6.7	6.0	6.8	4.7	5.2	5.4	5.5
7194-87	35	6.6	7.3	6.9	7.7	5.0	5.4	5.7	6.1
Lot mean		6.3	6.9	6.4	7.1	5.0	5.3	5.5	5.8
Standard deviation		0.8	0.5	0.7	0.8	0.5	0.3	0.45	0.5

TABLE B-7
THERMAL EXPANSIVITY: H-451, LOT 426

Log No.		Mean Thermal Expansivity $\times 10^6 \text{ K}^{-1} (^\circ\text{C}^{-1})$ [Standard Deviation $\times 10^6 \text{ K}^{-1} (^\circ\text{C}^{-1})$] [No. of Replicates]							
		Axial				Radial			
GA	GLCC	MLC	MLE	EC	EE	MLC	MLE	EC	EE
6484-33	92	4.08 (0.14) [8]	3.86 (0.20) [8]	4.00 (0.18) [8]	3.88 (0.09) [8]	4.51 (0.29) [8]	4.65 (0.13) [8]	4.70 (0.14) [8]	4.70 (0.21) [8]
6484-34	198	4.10 (0.10) [8]	3.92 (0.19) [8]	4.31 (0.08) [8]	4.13 (0.12) [8]	4.63 (0.20) [8]	4.66 (0.25) [8]	4.55 (0.27) [8]	4.59 (0.10) [8]
6484-41	184	3.92 (0.07) [8]	3.90 (0.09) [8]	3.90 (0.13) [8]	3.89 (0.09) [8]	4.42 (0.18) [8]	4.50 (0.18) [8]	4.43 (0.16) [8]	4.59 (0.18) [8]
Mean, lot 426		4.03 (0.13) [24]	3.89 (0.16) [24]	4.07 (0.22) [24]	3.97 (0.15) [24]	4.52 (0.23) [24]	4.60 (0.20) [24]	4.56 (0.22) [24]	4.63 (0.17) [24]

TABLE B-8
ANISOTROPY FACTOR: H-451, LOT 426

Log No.		Mean Anisotropy Factor, $\frac{\alpha(\text{radial})^{(a)}}{\alpha(\text{axial})}$			
GA	GLCC	MLC	MLE	EC	EE
6484-33	92	1.12	1.18	1.12	1.17
-34	198				
-41	184				

(a) Mean of three logs.

TABLE B-9
THERMAL CONDUCTIVITY: H-451, LOT 426, MLC

Log No.		Orientation	Mean Thermal Conductivity, W/m·K (Standard Deviation, W/m·K) [Number of Replicates]					
GA	GLCC		295 K (22°C) ^(a)	473 K (200°C) ^(a)	673 K (400°C) ^(a)	873 K (600°C) ^(a)	1073 K (800°C) ^(a)	
6484-34	198	Axial	136 (17) [8]	120 (27) [8]	92 (7) [8]	75 (5) [8]	65 (5) [8]	
6484-34	198	Radial	129 (10) [8]	115 (6) [8]	88 (6) [8]	73 (6) [8]	63 (5) [8]	
6484-41	184	Axial	147 (17) [7]	125 (14) [7]	92 (4) [7]	76 (5) [7]	64 (3) [7]	
6484-41	184	Radial	139 (13) [7]	112 (7) [7]	88 (4) [7]	74 (5) [7]	63 (5) [7]	
Mean, Lot 426		Axial	141 (18) [15]	122 (21) [15]	92 (6) [15]	76 (5) [15]	65 (4) [15]	
		Radial	134 (13) [15]	114 (6) [15]	88 (5) [15]	73 (6) [15]	63 (4) [15]	

(a) Temperature of meas

TABLE B-10
MEAN TENSILE STRENGTH OF UNIRRADIATED H-451 GRAPHITE
SPECIMENS USED IN FATIGUE STUDY

Orientation	Mean Tensile Strength (± Standard Deviation) (MPa)
Axial	18.9 ± 1.8
Radial	13.9 ± 2.3

TABLE B-11
IRRADIATION CONDITIONS AND MEAN TENSILE STRENGTH OF IRRADIATED H-451 GRAPHITE
USED IN FATIGUE STUDY
(ALL SPECIMENS MLC LOCATION)

Orientation	Fast Neutron Fluence (10^{25} N/m ²)	Irradiation Temperature		Mean Tensile Strength ($\pm$ Standard Deviation) (MPa)	Mean Tensile Strength of Unirradiated Companions ($\pm$ Standard Deviation) (MPa)
	(E > 29 fJ) HTGR	K	°C		
Axial	3.0	1173	900	14.5 $\pm$ 2.1	10.8 $\pm$ 0.9
Radial	5.7	1223-1263	950-990	22.4 $\pm$ 2.6	11.5 $\pm$ 1.7
Axial	8.5	1173-1243	900-970	20.2 $\pm$ 1.9	10.8 $\pm$ 0.9

TABLE B-12
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL EXPANSIVITY OF H-451 GRAPHITE
IRRADIATED IN CAPSULES OG-1, OG-2, AND OG-3

Lot	GA Log No.	Orientation	Location in Log	Mean Irradiation Temperature		Fluence x 10 ⁻²⁵ (N/m ²) (E > 29 fJ) _{HTGR}	Average Percent Change in CTE 295-773 K (22°-500°C)
				K	°C		
266	5651-28	Axial	Midlength center	915	642	2.0	+1
				935	662	3.7	+3
				935	662	5.5	-3
				1045	872	1.9	+6
				1195	922	2.3	-4
				1205	932	4.5	-15
				1170	897	6.4	-24
				1270	997	8.5	-38
				1335	1062	3.0	-7
				1665	1392	3.2	-4
				1660	1387	6.1	-34
				1660	1387	9.0	-47
			Midlength edge	1045	772	1.9	+4
				1125	852	1.9	+2
				1110	837	3.6	+2
				1255	982	2.8	-11
				1260	987	5.3	-20
				1480	1207	6.3	-30
				1515	1242	9.2	-43
266	5651-28	Radial	Midlength center	915	642	2.0	+3
				935	662	3.7	+7
				935	662	5.5	0
				1045	772	1.9	+9
				1195	922	2.3	-9
				1205	932	4.5	-9
				1170	897	6.4	-23
				1270	997	8.5	-43
				1335	1062	3.0	-7
				1665	1392	3.2	-9
				1660	1387	6.1	-34
				1660	1387	9.0	-46
			Midlength edge	1045	772	1.9	+2
				1125	852	1.9	+2
				1110	837	3.6	+2
				1255	982	2.8	-11
				1260	987	5.3	-18
				1480	1207	6.3	-27
				1515	1242	9.3	-41
426	6484-34	Axial	Midlength center	970	697	1.8	+10
				1040	767	2.4	+11
				1655	1382	2.9	+15
		Radial	Midlength center	970	697	1.8	+10
				1010	737	2.4	+9
				1655	1382	2.9	+13

TABLE B-13
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL EXPANSIVITY OF
H-429 GRAPHITE IRRADIATED IN CAPSULES OG-1, OG-2, and OG-3

GA Log No.	Orientation	Mean Irradiation Temperature		Fluence ($\times 10^{25}$ N/m ²) (E > 29 fJ) _{HTGR}	Average Percent Change in CTE 295-773 K (22°-500°C)
		K	°C		
4974-04A	Axial	1165	892	3.0	-8
		1180	907	5.2	-4
		1170	897	7.2	-21
		1435	1162	5.6	-13
		1460	1187	8.6	-28
		1480	1207	11.4	-41
		1705	1432	6.6	-22
		1675	1402	8.8	-30
	Radial	1165	892	3.0	+2
		1145	872	5.6	+1
		1170	897	7.2	-19
		1435	1162	5.6	-9
		1460	1187	8.6	-29
		1480	1207	11.4	-39
		1705	1432	6.6	-17
		1675	1402	8.8	-27

TABLE B-14
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL EXPANSIVITY OF
TS-1240 GRAPHITE IRRADIATED IN CAPSULES OG-2 AND OG-3

GA Log No.	Orientation	Location in Log	Mean Irradiation Temperature		Fluence ($\times 10^{25}$ N/m ²) (E > 29 fJ) _{HTGR}	Average Percent Change in CTE 295-773 K (22°C-500°C)
			K	°C		
5651-73	Axial	Midlength- center	895	622	1.2	+7
			1040	767	1.7	+2
			1195	922	2.5	-1
			1250	977	4.9	-36
			1380	1107	2.9	+1
			1415	1142	5.9	-33
	Radial	Midlength- edge	880	607	2.3	+6
		Midlength- center	865	592	1.2	-1
			1080	807	1.7	-4
			1195	922	2.5	-4
			1200	927	4.9	-20
			1475	1202	2.9	-11
			1500	1227	5.9	-47
		Midlength- edge	880	607	2.3	+4

TABLE B-15
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL EXPANSIVITY OF
SO818 GRAPHITE IRRADIATED IN CAPSULE OG-3

Lot	GA Log No.	Orientation	Location in Log	Mean Irradiation Temperature		Fluence x 10 ⁻²⁵ (N/m ²) (E > 29 fJ) _{HTGR}	Average Percent Change in CTE 295-773 K (22°-500°C)
				K	°C		
4B	6484-22	Axial	Midlength center	1040	767	2.4	+3
				1655	1382	2.9	-12
		Radial	Midlength center	1010	737	2.4	+7
				1655	1382	2.9	-11

TABLE B-16
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL CONDUCTIVITY OF H-451 GRAPHITE IRRADIATED IN
CAPSULES OG-1, OG-2, AND OG-3
(ALL SPECIMENS FROM MLC OF PARENT LOG)

GLCC Lot No.	GA Log No.	Orientation	Mean Irradiation Temperature		Fluence $\times 10^{-25}$ (N/m ²) (E > 29 fJ) _{HTGR}	Mean Thermal Conductivity (W/m·K) $\pm$ Standard Deviation	
			K	°C		At 293 K (20°C)	At Irradiation Temperature
266	5651-28	Axial	--	--	0	145.2 $\pm$ 1.3	--
			875	602	2.0	34.7 $\pm$ 1.4	33.3 $\pm$ 2.7
			890	617	3.7	30.1 $\pm$ 0.9	32.5 $\pm$ 4.3
			900	627	5.5	29.6 $\pm$ 1.4	31.5 $\pm$ 4.4
			1195	922	2.8	48.8 $\pm$ 1.8	40.0 $\pm$ 1.7
			1205	932	5.3	41.3 $\pm$ 8.4	39.8 $\pm$ 3.0
			1210	937	7.7	43.6 $\pm$ 6.8	34.1 $\pm$ 5.9
			1625	1352	3.2	79.8 $\pm$ 4.2	41.1 $\pm$ 1.2
			1625	1352	6.1	80.9 $\pm$ 4.0	40.7 $\pm$ 0.6
			1620	1347	9.0	68.9 $\pm$ 9.2	34.5 $\pm$ 3.8
266	5651-28	Radial	--	--	0	123.0 $\pm$ 7.1	--
			875	602	2.0	32.0 $\pm$ 1.9	33.3 $\pm$ 2.7
			890	617	3.7	27.8 $\pm$ 3.1	29.5 $\pm$ 3.2
			900	627	5.5	25.9 $\pm$ 0.8	28.0 $\pm$ 4.7
			1195	922	2.8	42.9 $\pm$ 3.3	36.1 $\pm$ 2.8
			1205	932	5.3	34.2 $\pm$ 4.3	29.5 $\pm$ 2.0
			1210	937	7.7	31.2 $\pm$ 5.1	30.7 $\pm$ 6.1
			1625	1352	3.2	83.5 $\pm$ 7.2	39.3 $\pm$ 3.4
			1625	1352	6.1	65.7 $\pm$ 8.2	36.4 $\pm$ 2.0
			1620	1347	9.0	56.2 $\pm$ 2.8	33.1 $\pm$ 4.4
426	6484-34	Axial	--	--	0	136.0 $\pm$ 17.2	--
			925	652	1.8	34.0 $\pm$ 3.0	32.0 $\pm$ 6.6
			1220	947	2.4	60.8 $\pm$ 5.1	39.0 $\pm$ 6.7
			1615	1342	2.9	82.4 $\pm$ 1.1	32.5 $\pm$ 6.3
		Radial	--	--	0	128.9 $\pm$ 10.0	--
			925	652	1.8	29.6 $\pm$ 4.1	29.8 $\pm$ 6.7
			1220	947	2.4	58.9 $\pm$ 3.1	35.4 $\pm$ 6.4
			1615	1342	2.9	72.4 $\pm$ 1.3	31.9 $\pm$ 6.3

TABLE B-17
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL CONDUCTIVITY OF TS-1240 GRAPHITE
IRRADIATED IN CAPSULES OG-2 AND OG-3
(ALL SPECIMENS FROM MLC OF PARENT LOG)

UCC Lot No.	GA Log No.	Orientation	Mean Irradiation Temperature		Fluence x 10 ⁻²⁵ (N/m ²) (E > 29 fJ) _{HTGR}	Mean Thermal Conductivity (W/m·K) ± Standard Deviation	
			K	°C		At 293 K (22°C)	At Irradiation Temperature
1	5151-73	Axial	--		0	97.9 ± 8.4	--
			910	637	1.7	30.5 ± 1.2	34.1 ± 1.7
			915	642	3.5	26.5 ± 0.9	27.9 ± 5.0
			1220	947	2.5	45.5 ± 3.7	38.2 ± 2.4
			1220	947	4.9	51.5 ± 2.1	36.5 ± 4.0
			1620	1347	2.9	68.8 ± 3.4	37.2 ± 1.5
			1615	1342	5.8	66.0 ± 7.9	37.5 ± 4.6
	5651-73	Radial	--		0	103.3 ± 4.2	--
			910	637	1.7	28.5 ± 2.8	32.6 ± 0.3
			915	642	3.5	27.5 ± 0.8	25.1 ± 3.4
			1220	947	2.5	37.0 ± 2.2	29.9 ± 1.7
			1220	947	4.9	50.1 ± 1.4	37.6 ± 4.7
			1620	1347	2.9	64.5 ± 5.5	33.5 ± 1.2
			1615	1342	5.8	65.4 ± 2.8	37.0 ± 3.5

TABLE B-18
SUMMARY OF IRRADIATION-INDUCED CHANGES IN THERMAL CONDUCTIVITY OF SO818 GRAPHITE
IRRADIATED IN CAPSULE OG-3
(ALL SPECIMENS FROM MLC OF PARENT LOG)

AirCo Lot No.	GA Log No.	Orientation	Mean Irradiation Temperature		Fluence $\times 10^{-25}$ (N/m ²) (E > 29 fJ) _{HTGR}	Mean Thermal Conductivity (W/m·K) $\pm$ Standard Deviation	
			K	°C		At 293 K (22°C)	At Irradiation Temperature
4-B	6484-22	Axial	--		0	135.1 $\pm$ 5.0	--
			925	652	1.8	28.6 $\pm$ 0.8	26.7 $\pm$ 4.6
			1220	947	2.4	50.5 $\pm$ 0.8	36.7 $\pm$ 4.2
			1615	1342	2.9	64.5 $\pm$ 4.9	34.3 $\pm$ 4.4
4-B	6484-22	Radial	--		0	125.9 $\pm$ 5.0	--
			925	652	1.8	28.4 $\pm$ 0.8	27.0 $\pm$ 5.0
			1220	947	2.4	43.7 $\pm$ 4.2	32.3 $\pm$ 4.3
			1615	1342	2.9	65.8 $\pm$ 2.3	33.0 $\pm$ 4.2

TABLE B-19
CALCULATED THERMAL CONDUCTIVITY AT IRRADIATION TEMPERATURE OF NEAR-ISOTROPIC GRAPHITE
IRRADIATED BETWEEN 573 AND 873 K (300° AND 600°C)

Temperature K (°C)	Fast Neutron Fluence, (N/m ² x 10 ⁻²⁵) (E > 29 fJ) _{HTGR}	Thermal Conductivity (W/m·K)		Temperature		Fast Neutron Fluence, (N/m ² x 10 ⁻²⁵) (E > 29 fJ) _{HTGR}	Thermal Conductivity (W/m·K)	
		Radial	Axial	K	°C		Radial	Axial
573 (300)	0.9	11.7	13.4	773	500	0.9	28.9	0.080
	1.8	9.2	10.5			1.8	20.9	0.058
	2.7	7.9	9.2			2.7	18.4	0.052
	3.6	7.1	8.4			3.6	16.7	0.046
	4.5	6.7	7.9			4.5	15.1	0.042
	8.9	5.9	7.1			8.9	13.0	15.1
	13.4	5.9	6.7			13.4	11.7	0.033
673 (400)	0.9	19.7	23.0	873	600	0.9	32.2	37.2
	1.8	14.6	17.2			1.8	24.3	28.0
	2.7	13.0	15.1			2.7	20.5	23.8
	3.6	11.7	13.8			3.6	18.0	20.9
	4.5	10.9	12.6			4.5	16.7	19.7
	8.9	9.6	11.3			8.9	15.5	18.0
	13.4	8.8	10.5			13.4	13.8	16.3

ALLOCATION OF GRAPHITE SAMPLES ^(a) TABLE B-20
FOR OXIDATION STUDIES (NUMBER OF SPECIMENS)

Log-Slab- Section No.	Axial							Radial Orientation		
	Controls	1% ^(b) 1073 K (800°C)	2% 1073 K (800°C)	4% 1073 K (800°C)	1% 1273 K (1000°C)	2% 1273 K (1000°C)	4% 1273 K (1000°C)	Controls	4% 1073 K (800°C)	4% 1273 K (1000°C)
Quarter-Length Center										
6484-34-5AC	1	2	3	4	8	10	7	43	42	40
	5	13	6	15	14	12	9	49	48	50
	16	17	18	25	19	22	11	54	52	56
	20	26	21	28	32	24	27	60	55	59
	33	31	23	34	35	29	30	64	62	61
	37		36	38			39	66	65	63
6484-34-5BC	11	1	4	3	2	8	5	40	41	45
	14	7	15	6	12	13	10	47	43	48
	21	9	29	17	25	18	16	52	44	51
	24	28	30	19	27	22	20	57	54	61
	26	33	32	23	31		35	59	58	65
	38		34	36		39	37	62	63	67
Quarter-Length Edge										
6484-34-5AE	101			103			102	128	126	129
	106			107			109	132	127	130
	111			112			110	134	133	131
	116			114			113	136	137	135
	121			118			117	138	141	143
	122			119			124	139	142	144
6484-34-5BE	103			102			101	129	126	125
	104			106			105	131	132	127
	112			108			111	122	134	128
	115			113			114	136	137	135
	117			116			120	141	138	139
	123			122			124	144	142	143

(a) Samples cored from H-451 graphite log 6484-34.

(b) Planned percent burnoff and temperature of oxidation.

Fig. B-1. Fatigue test data: in air log-log plot of normalized peak stress versus number of cycles to failure with $R = -1$; lower x/y tolerance limits represent limits above which x% of all data would fall, with y% confidence; axial specimens; open circles represent run-outs (from Ref. 5-10)

Fig. B-2. Constant life fatigue diagram (Goodman diagram) for H-451 graphite: axial direction (from Ref. 5-10)

Fig. B-3. Dimensional change in H-451 graphite: axial direction; irradiation temperatures 875 K to 975 K and 1075 K to 1175 K (602° to 702°C and 802° to 902°C) (from Ref. 6-3)

Fig. B-4. Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1275 K to 1375 K and 975 K to 1075 K (1002° to 1102°C and 702° to 802°C) (from Ref. 6-3)

Fig. B-5. Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1175 K to 1275 K (902° to 1002°C) (from Ref. 6-3)

Fig. B-6. Dimensional change in H-451 graphite: axial direction; irradiation temperatures 1475 K to 1575 K and 1575 K to 1675 K (1202° to 1302°C and 1302° to 1402°C) (from Ref. 6-3)

Fig. B-7. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 875 K to 975 K and 1475 K to 1575 K (602° to 702°C and 1202° to 1302°C) (from Ref. 6-3)

Fig. B-8. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 975 K to 1075 K and 1275 K to 1375 K (702° to 802°C and 1002° to 1102°C) (from Ref. 6-3)

Fig. B-9. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 1175 K to 1275 K (902° to 1002°C) (from Ref. 6-3)

Fig. B-10. Dimensional change in H-451 graphite: radial direction; irradiation temperatures 1075 K to 1175 K and 1575 K to 1675 K (802° to 902°C and 1302° to 1402°C) (from Ref. 6-3)

Fig. B-11. Dimensional changes in near-isotropic graphite irradiated at 633 K to 773 K (360° to 500°C) (data from Refs. 6-4 and 6-6)

Fig. B-12. Dimensional changes in near-isotropic graphite irradiated at 773 K to 873 K (500° to 600°C) (data from Refs. 6-4 and 6-6)

Fig. B-13. Changes in tensile strength and elastic modulus of H-451 graphite as a function of fast neutron fluences. Error bars denote $\pm$ one standard deviation (from Ref. 6-3)

Fig. B-14. Fractional changes in sonic elastic modulus of extruded near-isotropic pitch coke graphite (Dragon code No. 100) (from Ref. 5-7)

Fig. B-15. Change in the sonic elastic modulus (E) and strength (S) of near-isotropic graphite irradiated at 623 K to 973 K at (350° to 700°C) (from Ref. 6-8)

Fig. B-16. Calculated percent increases in static elastic modulus of near-isotropic graphites (solid parts of curves correspond to data in Refs. 6-6 and 6-8; broken parts are extrapolated)

Fig. B-17. Fatigue test data irradiated to $3.0 \times 10^{25} \text{ N/m}^2$ ($E > 29 \text{ fJ}$)_{HTGR} at 1173 K (900°C). Log-log plot of normalized peak stress versus number of cycles to failure, with stress ratio, $R = -1$. Lower x/y tolerance limits represent the limits above which x% of all data would fall, with y% confidence; axial specimens; open circles represent run-outs (from Ref. 5-10).

Fig. B-18. Percent change in thermal expansivity [295 K to 773 K (22° to 500°C)] of near-isotropic graphites as a function of fast neutron fluence: irradiation temperature 865 K to 1205 K (592° to 932°C) (from Ref. 6-3)

Fig. B-19. Percent change in thermal expansivity [295 K to 773 K (22° to 500°C)] of near isotropic graphites as a function of fast neutron fluence: irradiation temperature 1250 K to 1705 K (592° to 932°C) (from Ref. 6-3)

Fig. B-20. Change in thermal expansivity of near-isotropic graphite irradiated at 350° to 550°C (data from Ref. 6-4)

Fig. B-21. Thermal conductivity at the irradiation temperature for H-451 and other near-isotropic graphites as a function of fast neutron fluence (from Ref. 6-3)

Fig. B-22. Irradiation-induced changes in thermal resistivity of near-isotropic graphite (data points from Refs. 6-6 and 6-8; curves from Ref. 6-10)

GENERAL ATOMIC

GENERAL ATOMIC COMPANY
P. O. BOX 81608
SAN DIEGO, CALIFORNIA 92138