DECLASSIFIED

CONFIDENTIAL

UNCLASSIFIED

GA - 5918 (Pt. 2)

This document contains Confidential-Restricted Data relating to civilian applications of atomic energy

ADVANCED, GRAPHITE-MATRIX DISPERSION TYPE FUEL SYSTEMS AND RESEARCH ON GRAPHITE

MASTER

Quarterly Progress Report for the
Period Ending November 14, 1964

By

J. C. Bokros

N. L. Sandefur

W. V. Goeddel

J. L. White

R. E. Norman

Classification Cancelled

UNCLASSIFIED

Or Changed To

November 15, 1964

By Authority Of

DOC

By

C. S. Patton

Date 10/27/71

General Atomic Division
General Dynamics Corporation
San Diego, California

AEC Research and Development Report

GROUP 1

Excluded from automatic downgrading
and declassification

RESTRICTED DATA

This document contains restricted data as defined in the Atomic Energy Act of 1954. Its transmittal or the disclosure of its contents in any manner to an unauthorized person is prohibited.

UNITED STATES ATOMIC ENERGY COMMISSION
Division of Technical Information

CONFIDENTIAL

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

UNCLASSIFIED

DECLASSIFIED

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.

031557030

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

This report has been reproduced directly from the best available copy.

Printed in USA. Charge \$0.35. Available from the Division of Technical Information Extension, P. O. Box 62, Oak Ridge, Tennessee 37831. Please direct to the same address inquiries covering the procurement of classified AEC reports.

0317281030

~~CONFIDENTIAL~~

UNCLASSIFIED

GA - 5918 (Pt. 2)
ADVANCED CONCEPTS FOR FUTURE APPLICATION-
REACTOR EXPERIMENTS (M - 3679, 38th Ed.)

ADVANCED, GRAPHITE-MATRIX, DISPERSION TYPE FUEL SYSTEMS
AND
RESEARCH ON GRAPHITE

QUARTERLY PROGRESS REPORT FOR THE PERIOD
ENDING NOVEMBER 14, 1964

Written by:

J. C. Bokros, W. V. Goeddel, R. E. Norman,
N. L. Sandefur, and J. L. White

November 15, 1964

Prepared under
Contract AT(04-3)-167
Project Agreement No. 12
for the

San Francisco Operations Office
U.S. Atomic Energy Commission

Class:	SPECIAL REVIEW	Reviews	Class.	Date
4		JRP	4	10-28-71
ABN			4	11-20-81

~~RESTRICTED DATA~~

This document contains restricted data as defined in the Atomic Energy Act of 1954. Its transmittal or the disclosure of its contents in any manner to an unauthorized person is prohibited.

GROUP 1

Excluded from automatic downgrading
and declassification

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

UNCLASSIFIED

~~CONFIDENTIAL~~

UNCLASSIFIED

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, 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.

0315087030

PREVIOUS REPORTS IN THIS SERIES

GA - 4022 (Pt. 2) — April 1, 1962, through March 31, 1963 (C - RD)

GA - 5016 (Pt. 2) — April 1, 1963, through March 31, 1964 (C - RD)

GA- 5606 (Pt. 2) — April 1, 1964, through August 14, 1964 (C-RD)

0317228030

DECLASSIFIED
UNCLASSIFIED

CONTENTS

I. INTRODUCTION	1
II. SUMMARY	3
III. FUEL MIGRATION STUDIES	4
IV. APPLICATION OF HOT-WORKING PROCESSES TO FABRICATION OF GRAPHITE-MATRIX FUEL BODIES	5

UNCLASSIFIED
DECLASSIFIED

DECLASSIFIED
UNCLASSIFIED

I. INTRODUCTION

The purpose of this program is to obtain substantial improvements in the materials required for service in high-temperature reactors of advanced design. The approach is based on gaining a more complete understanding of material properties and behavior in order to provide effective guidance for the development of improved materials. Two principal lines of effort may be distinguished: (1) "Advanced, Graphite-Matrix Fuel Systems," which is concerned with the various materials directly involved in the fuel systems, and (2) "Research on Graphite," which is concerned with the graphitic materials employed either in the fuel system or in other structural components of the high-temperature reactor.

In the period covered by the present report, August 15 through November 14, 1964, the following investigations have been conducted and are reported in the unclassified portion of this report (GA-5918, Part 1):

1. Investigations of Pyrolytic Carbon Coatings Deposited in Fluidized Beds. The structures of pyrolytic carbons deposited in a fluidized bed are being characterized in order to establish the relationships existing between bed deposition conditions, coating structure, and the physical and mechanical properties relevant to reactor performance. The earlier studies are being extended to consider deposition temperatures greater than 1925°C and gases other than methane. The property studies include measurements of the effects of annealing and irradiation on the pyrolytic carbons.
2. Diffusion of Uranium and Thorium in Pyrolytic Carbon. The rates of diffusion of uranium and thorium in pyrolytic carbons typical of those employed in graphite-matrix fuel systems are being determined, with emphasis on relating diffusion to structure.
3. Fission-product Release from Carbide Fuel Systems. The release of metallic fission products through pyrolytic carbon coatings typical of those employed in graphite-matrix fuel systems is being determined in an effort to relate release rates to coating structure.
4. Characterization of Microstructure of Graphitic Materials. Methods of characterizing the degree of graphitization of carbons and graphitic materials are being investigated.

1
UNCLASSIFIED

DECLASSIFIED

0310587030

CONFIDENTIAL

2

5. Relation of Physical Properties of Graphitic Materials to Preferred Orientation. The relation of certain physical properties, including Young's modulus and the dimensional changes under irradiation, to the preferred orientation of various graphitic materials is being studied.
6. Chemical and Metallurgical Processes for Modifying the Microstructure of Graphite. The process of hot-working of graphitic materials is being investigated, with current emphasis on evaluating the variables of pitch content and additives in the graphite bodies.

This portion of the report describes work conducted on the following investigations:

1. Fuel Migration Studies. Studies of the rates and mechanisms of migration of uranium and thorium through pyrolytic-carbon coatings are being concluded. Particular importance is being placed on relating the coating structure and the particle composition to over-all thermal stability of the coated fuel particles.
2. Application of Hot-working Processes to Graphite Bodies Containing Fuel Carbides. The hot-working experiments described in Part 1 of this report are being extended to include graphite-matrix fuel bodies containing carbide particles. The limiting conditions for hot-working these bodies are being determined, and the properties of the resultant fuel compacts are being defined.

GROUP 1

Excluded from automatic downgrading
and declassificationCONFIDENTIAL
RESTRICTED DATA

0310587030

DECLASSIFIED
CONFIDENTIAL

II. SUMMARY

A topical report was issued describing the studies on the effect of coating structure on uranium migration through pyrolytic carbon. Results of these studies showed a definite correlation between the structure of the pyrolytic carbon and the migration rate through it. The lowest rates were observed in high-density, oriented laminar structures, while the highest rates were observed in low-density, isotropic structures. Migration rates through high-density, granular structures were also observed to be high. The results were discussed in terms of the structure of the deposits.

A topical report was also issued on the studies of the application of hot-working processes to the fabrication of graphite-matrix fuel bodies. Conclusions from this report were that it is possible to hot-work graphite bodies containing uncoated carbide particles without disrupting the particles. The properties of these hot-worked fueled graphite bodies were consistent with those observed for unfueled bodies (e. g. , porosity sharply reduced, large increase in anisotropy). Further work on the hot-working of fueled graphite bodies is being held in abeyance until improvements are made in the hot-working characteristics and properties of unfueled graphite bodies.

CONFIDENTIAL
DECLASSIFIED

031004700
CONFIDENTIAL

III. FUEL MIGRATION STUDIES

J. C. Bokros and N. L. Sandefur

EFFECT OF STRUCTURE ON URANIUM MIGRATION THROUGH PYROLYTIC CARBON

A topical report was prepared on the studies of the effect of coating structure on uranium migration through pyrolytic carbon. This report, GA-5793, "Uranium Migration Through Pyrolytic Carbons Deposited in a Fluidized Bed," by J. C. Bokros and N. L. Sandefur, summarizes the results of this phase of the research program and includes the work done during the present reporting period. The summary from this topical report is given below.

Summary of Report GA-5793: "Uranium Migration Through Pyrolytic Carbons Deposited in a Fluidized Bed"

A systematic study of the rate of uranium migration at 1850° and 2300°C, in a variety of pyrolytic carbon structures deposited from methane in a fluidized bed in the temperature range 1350° to 1925°C, showed a definite correlation between the structure of the pyrolytic carbon and the migration rate through it.

The lowest rates were observed in high-density, oriented laminar structures deposited at 1350° to 1450°C from high methane concentrations. The low migration rates in these structures were discussed in terms of their anisotropy, density, crystallite size, and annealability.

The high migration rates through the granular structures were shown to be due to preferential grain-boundary or rapid interlaminar diffusion in grains whose layer planes are inclined at large angles to the migration direction.

The migration resistance of the low-density isotropic structures is the poorest of all the structures studied. This is considered to be a consequence of an abundance of "a" direction short circuits through the structure.

CONFIDENTIAL
031004700

DECLASSIFIED
CONFIDENTIAL

IV. APPLICATION OF HOT-WORKING PROCESSES TO THE FABRICATION OF GRAPHITE-MATRIX FUEL BODIES

J. L. White and J. M. Pontelandolfo

A topical report has been prepared to present the results of studies undertaken to determine the feasibility of using hot-working processes to fabricate graphite-matrix fuel bodies with improved properties. A summary of this report, GA-5882, "Hot-Worked Graphite-Matrix Fuel Bodies," by J. L. White and J. M. Pontelandolfo, is given below.

All work during the present reporting period is covered by the above-mentioned topical report. Since it has been shown that graphite bodies containing uncoated fuel particles can be hot-worked, under rather stringent temperature limitations, without disrupting the particles, no further work on the hot-working of fueled graphite bodies is anticipated until improvements are made in the hot-working characteristics and resulting properties of unfueled graphite bodies. For current work on this latter topic, see the unclassified section of this report.

Summary of Report GA-5882: "Hot-Worked Graphite-Matrix Fuel Bodies"

The objectives of this investigation were to learn the conditions under which a graphite matrix containing uncoated carbide particles can be hot-worked without adversely affecting the particles and to determine the properties of the resulting fuel bodies. The conventional HTGR-type fuel body, which uses fuel particles coated with pyrolytic carbon and a low pitch content in the graphite matrix, was found to be unsuitable for hot-working. By dispensing with particle coatings, by using graphite-matrix compositions which perform more favorably in hot-working,⁽¹⁾ and by using relatively low hot-working temperatures, it has been possible to prepare hot-worked fuel bodies containing either UC-ZrC or UC₂ particles. Micro-graphic observations disclose little or no disruption of the fuel particles.

The most serious problem encountered in this work is the disruption or complete disintegration of the fuel particles during hot-working. Because of the matrix deformations involved in hot-working, no opportunities can be given for fuel migration from the integral particles, e. g., by the relatively fast migration through a pyrolytic-carbon coating. The disruption processes with uncoated fuel particles of either UC-ZrC or UC₂ appear to

5
CONFIDENTIAL

DECLASSIFIED

~~UNCLASSIFIED~~

~~CONFIDENTIAL~~

6

be strongly temperature-sensitive and impose severe limitations on the maximum temperatures that can be employed in hot-working. Hot-worked bodies of good integrity have been prepared by holding the temperature below approximately 2450°C for UC-ZrC particles and below approximately 2400°C for UC_2 particles. Under these hot-working conditions, the times required to reach reasonable hot-working strains are long, e. g., 4 hr to reach 40% true strain with a body containing UC_2 particles.

The properties of the hot-worked fuel bodies are consistent with the properties observed for hot-worked graphite containing no fuel particles.^(1,2) The porosity is sharply reduced by hot-working, although the decreases in permeability at this point fall short of the levels desired for a retentive fuel body. Direct measurements of the fission-product release by postirradiation annealing show that there is, in general, an enhanced retentivity of hot-worked fuel bodies, but that this retentivity is selective, e. g., in one test, xenon retentivity is improved sevenfold while barium retentivity is adversely affected. The graphite matrix is highly oriented, and this preferred orientation is reflected strongly in the measured properties of compressive strength and thermal diffusivity.

Further work on the concept of a hot-worked fuel body should concentrate on improving the properties of the graphite matrix, in respect to obtaining both improved hot-working characteristics and better finished properties. Because the creep rate of graphite is quite stress-sensitive, some further reduction in hot-working times might be obtained by determining the optimum pitch content to avoid failure during hot-working at higher stress levels. The use of metal additives which form a liquid phase in the graphite matrix at hot-working temperatures⁽³⁾ is promising if reactions with the fuel particles can be avoided.

REFERENCES

1. Price, R. J., and J. L. White, "Research on Graphite, Annual Report, April 1, 1963, through March 31, 1964," General Atomic Report GA-5033. 94 p.
2. White, J. L., and R. J. Price, "Hot-working of Graphite for Graphite-matrix Nuclear Fuels," General Atomic Report GA-5348, July 17, 1964. 10 p.
3. Bokros, J. C., et al., "Advanced, Graphite-matrix, Dispersion Type Fuel Systems and Research on Graphite, Quarterly Progress Report for the Period Ending November 14, 1964," General Atomic Report GA-5918 (Part 1).

~~CONFIDENTIAL~~

~~RESTRICTED DATA~~

~~UNCLASSIFIED~~

0115087030

~~CONFIDENTIAL~~

UNCLASSIFIED

UNCLASSIFIED

~~CONFIDENTIAL~~

0371281030