GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

H.C. \$ 3.00; RM 65

JOHN JAY HOPKINS LABORATORY FOR PURE AND APPLIED SCIENCE

P.O. BOX 608, SAN DIEGO, CALIFORNIA 92112

AEC RESEARCH AND
DEVELOPMENT REPORT

GA-6830

PUBLIC SERVICE COMPANY OF COLORADO
330-MW(E) HIGH-TEMPERATURE GAS-COOLED REACTOR
RESEARCH AND DEVELOPMENT PROGRAM

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
SEPTEMBER 30, 1965

Prepared under
Contract AT(04-3)-633
for the
San Francisco Operations Office
U.S. Atomic Energy Commission

December 31, 1965

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.

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.

INTRODUCTION

The objective of the research and development program work reported here is to develop and verify the information required to design, construct, operate, and maintain the Public Service Company of Colorado power plant as provided in U. S. Atomic Energy Commission Contract AT(04-3)-633.

Part I of this report includes the work described in Appendix B of the contract; this work consists largely of component development and testing, nuclear analysis, and fuel development and testing. Part II covers the work described in Appendix K of the contract; this work includes the development of the fuel transfer machine, the series-steam-turbine-driven circulator, the control rod drives, the steam generator, and coated-particle production processes.

.

.

.

.

.

CONTENTS

INTRODUCTION	i
------------------------	---

Part I

TASK I. COMPONENT DEVELOPMENT	1
---	---

Reactor Vessel	1
PCRV Model Tests	1
Prestressing System	2
Penetrations and Components	2
Concrete Tests	3
Reactor Thermal Barrier	3
Class A Thermal Barrier	3
Class B Thermal Barrier	4
Metal-Impure Coolant Compatibility Test	6
Primary Coolant System Flow	11
Compressors and Auxiliary Circulators	11
Compressors	11
Auxiliary Circulators	12
Valves	12
Steam-Water Dump Valve	12
Helium Service Valves	13
Instrumentation and Electrical	13
Moisture Detectors	13
Auxiliary Equipment, Manipulators, and Tools	13

TASK II. NUCLEAR ANALYSIS	15
-------------------------------------	----

Fuel Cycle Analysis	15
Equilibrium Fuel Cycle	15
Approach to Equilibrium	19
Fuel Zoning and Power Distribution	23
Transverse Power Distribution	23
Axial Power Distribution	25
Feed Compositions	29
Kinetics Analysis	29

TASK III. FUEL DEVELOPMENT	32
Fuel Element Design	32
Preliminary Design Study	33
Design	44
Materials	52
Particle Beds	52
Graphite Development	57
Control Materials	60
Coated Particle Irradiations	60
Graphite Irradiations.	61
Chemistry	63
Fission-product Release Studies	63
Mathematical Treatment of Metallic Fission-	
product Release from HTGR Fuel Blocks	79
Sorption and Diffusion of Fission Products	90
Helium Purification System	109
Behavior of Sorbent Beds	109
Decontamination Studies	109

Part II

TASK I. FUEL TRANSFER MACHINE	113
Bearing Tests.	114
TASK II. SERIES-STEAM-TURBINE-DRIVEN HELIUM	
CIRCULATOR	116
Rotor Dynamics Simulation, Lubrication and Seal System . .	123
Static Aerodynamics, Diffuser and Inlet.	129
Full-Scale Test Facility for Helium Circulator	
Development Program	129
TASK III. CONTROL ROD DRIVE	135
TASK IV. STEAM GENERATOR.	141
Materials	141
Metal-Coolant Compatibility Test	141
Heat Transfer and Fluid Flow	141
Steam/Water Flow Stability	141
Tube Vibration Studies (Including Flow Pulsation	
and Impedance).	143

Design and Analysis	144
Design Study	144
High-temperature Design Basis	147
TASK V. COATED PARTICLE DEVELOPMENT	150
High-density Isotropic Coatings	150
Buffer Coating Studies	150
Coated Particle Preparation.	152
Dounreay Fast-flux Experiment	152
Coated Particles for Packing Studies	152
Coating Evaluation Techniques	152
Coating Density Determination by X-ray Absorption	153
Coating Density Determination by the Electron Microprobe	156
Coating Thickness Determination	156

PART I

TASK I
COMPONENT DEVELOPMENT

REACTOR VESSEL

PCRVR Model Tests

PCRVR Model No. 2 is roughly a shortened 1/4-scale model of the anticipated reactor vessel for the PSC plant. It has penetrations in both the top and bottom heads which are close in size ratio to any which may be expected in the PSC vessel. The internal dimensions of the model are: 8 ft 0 in. in diameter, 10 ft 0 in. in height, with a concrete thickness of 2 ft 1-1/2 in. for the side wall and 4 ft 0 in. for the top and bottom heads. A toroidal transition from the cylindrical liner to the top head and a conical transition to the bottom head are being tested to ascertain the relative advantages of each, as well as to determine experimentally the validity of certain analytical assumptions.

The design of Model No. 2 was initiated in mid-July and was essentially completed in September. Details of tendon configuration are firm, but the actual tendon forces are awaiting the outcome of detailed analysis expected shortly. It was originally planned to use the Société d'Études et d'Équipements d'Entreprises F. 16 tendon, but the vendor recommended that a tendon of half the load capacity (F. 04) be used to ease installation in the short radius and 180° total curvature of the horizontal tendons.

The model site was prepared and a foundation constructed during August. An igneous rock aggregate called andesite, available in the vicinity of the PSC reactor site, was chosen and has been procured. Cement from the Ideal Cement Company in Colorado was chosen and has also been procured.

The concrete in the lower portion of the model will be installed by intrusion grouting of preplaced aggregate to assure a good fit of the concrete to the bottom head of the liner and bottom penetrations. The upper portion will be placed using premixed concrete in the conventional manner.

A bid package for the construction of the model has been issued and orders have been placed for the tendons, reinforcing steel, tendon tubes, and bearing plates.

The steel liner is being constructed of 1/4-in.-thick Type A441 plate with the exception of the torus cone and head sections, which are 3/16-in. Type A212. Anchors that attach the liner to the concrete are generally spaced at 8-in. centers on the cylinder and at 6-in. centers on the heads. These anchor spacings should inhibit liner buckling up to high strain levels. The liner, which was fabricated at General Atomic, is being prepared for sensor attachment.

Calculations have indicated that an ambient air temperature of 70°F around the model will be necessary to maintain the desired concrete ΔT of 50°F with the inner concrete surface at 150°F during the hot tests. Additional calculations indicated that this temperature can be maintained by properly insulating the building around the model and providing fans for ambient air temperature control. A fan will also be required for circulating the air underneath the model.

Prestressing System

Essentially no effort was expended in this area during the reporting period other than the specification of the more or less standard type tendons, anchors, and tendon sheaths to be used in Model No. 2.

Penetrations and Components

Design and fabrication of the penetrations for Model No. 2 have been completed. These include 37 refueling nozzle penetrations in the top head and the following penetrations in the bottom head:

- One superheater penetration
- Two helium circulator penetrations
- Four economizer-evaporator penetrations with insulation well
- Four economizer-evaporator penetrations without insulation well
- One reheater penetration with insulation well
- Two reheater penetrations without insulation well
- One access opening penetration

These penetrations and closures are designed so that the primary stress does not exceed the yield stress of the material at the 1500 psi predicted ultimate failure pressure for the main PCR structure. Thus the penetrations and closures are more conservative than ASME code for the PCR normal pressure of 600 psi and test pressure of 720 psi.

The penetrations are anchored to the concrete by means of bearing rings welded to the penetrations and are sized to provide adequate bearing area on the concrete under the full axial pressure load on the penetration. All the penetrations are sealed with standard welded pipe caps

or flat heads, except the 30-in. access opening. This opening uses a double closure with each head bolted in place and sealed with a neoprene O-ring gasket.

Concrete Tests

Initial evaluation of seven possible aggregate sources in the area of the Colorado site narrowed the choice to two. These two were an igneous rock called andesite and a sedimentary rock called dolomite. Further testing of the aggregates indicated the andesite to be superior from the standpoint of durability and soundness. Concrete tests indicated that the andesite would meet the PSC requirements as to a compressive strength of approximately 6000 psi. Chemical tests of the aggregate are in progress to provide data for use in shielding calculations.

Standard Type II portland cement from the Ideal Cement Company in Colorado, the primary local source of supply, was tested and found to be suitable for both the standard premix type of concrete and for the intrusion grout.

REACTOR THERMAL BARRIER

The reactor thermal barrier logically falls into four separate types which, for convenience, have been designated Class A, B, C, and D. The Class A thermal barrier, which will be used to insulate the PCRV inner liner will, in general, be exposed on the hot side to temperatures approximately equal to reactor primary coolant inlet temperature, 754°F. The Class A barrier is currently expected to be an insulation of the metallic, multilayer type. The Class B thermal barrier will be used, if required, in the reactor outlet plenum and also to insulate the core support structure. Class B thermal barrier will be exposed on the hot side to temperatures as high as the local maximum primary coolant exit temperature. The Class B barrier will probably be of a nonmetallic type. The Class C thermal barrier will be used, if required, on the reactor primary coolant outlet gas ducting and will be exposed to mixed exit gases of about the average exit temperature of 1425°F. The Class D thermal barrier will be used as necessary to insulate the concrete from the high temperature water or steam in the steam generator penetrations. Major effort is currently directed toward development of Class A and Class B thermal barriers.

Class A Thermal Barrier

Current practice in the use of Type A thermal barrier is limited to experience in England and France. In France, pumice concrete insulation in steel casings has been specified for use in the EDF-3 and EDF-4 reactors.

In England, reflective metal sheet insulation has been specified for use in the Oldbury and Wylfa reactors. Our analysis of the limited information available on these insulation systems indicate that they are either expensive or are of unknown reliability.

Several conceptual designs for a thermal barrier of metal sandwich type of construction are under consideration. A specification for a Class A thermal barrier testing program is in preparation. Performance of the Class A barrier is naturally tied to the performance of the liner cooling water system. Some preliminary studies of two means of attaching the cooling coils to the liner have been made. In the one the tubes are welded directly to the steel liner, and in the other the tubes are welded to fins that are in turn welded to the liner. The effective liner temperatures and temperature differences using metal baffle insulation material with a thermal conductivity of 0.32 Btu/hr-ft-°F are given in Table 1.

Class B Thermal Barrier

Early in the quarter various conceptual studies were made of means of supporting high-temperature insulations above the reactor core and outlet primary coolant plenum. These included spring-loaded insulating domes and column-supported flat roofs. These studies were redirected when, during the course of the quarter, it was decided to establish primary coolant downflow through the core as the reference design. Subsequently, Class B studies have been based on the concept of incorporating the Class B barrier as part of the core support structure.

Thermal insulation and expansion calculations have been made on two candidate core support plate insulation configurations. The first configuration consists of three layers of insulation: (1) 2-1/2 in. of graphite with a thermal conductivity of 4.50 Btu-ft/hr-ft²-°F, (2) 2-1/2 in. of porous carbon with a thermal conductivity of 1.40 Btu-ft/hr-ft²-°F, and (3) 2-1/2 in. of dense alumina with a thermal conductivity of 1.84 Btu-ft/hr-ft²-°F. In conjunction with a 2-in. carbon steel support plate, this configuration provides adequate insulation both during steady-state normal operation and emergency depressurized primary coolant conditions. In addition, the over-all thermal

Table 1
LINER COOLING TUBE SYSTEM

Insulation Thickness (in.)	Welded Directly				Welded to Fins			
	Without Nuclear Heat		With Nuclear Heat		Without Nuclear Heat		With Nuclear Heat	
	Temp ^a (°F)	ΔT^b	Temp ^a (°F)	ΔT^b	Temp ^a (°F)	ΔT^b	Temp ^a (°F)	ΔT^b
2.5	131	51	---	---	134	54	---	---
3.0	126	47	130	50	128	49	131	51
3.5	121	43	---	---	123	45	---	---

^a—Effective liner outside temperature from a concrete heat-transfer standpoint.

^b—Difference between effective liner temperature and concrete exterior temperature.

expansion of the various layers is nearly matched at their respective temperatures to provide little relative movement from layer to layer during temperature changes.

The second configuration consists of four insulating layers: (1) 2-1/2 in. of graphite, (2) 2-1/2 in. of dense carbon with a thermal conductivity of 3.45 Btu-ft/hr-ft²-°F, (3) 2 in. of porous carbon, and (4) 2 in. of dense alumina. The principal advantage of the second configuration is that there is less temperature differential across each layer, and as a consequence, there would be less bowing or curvature developed within the insulation blocks in each layer.

Metal-Impure Coolant Compatibility Test

An experiment to evaluate the effect of CO contaminant in helium on six structural metallic materials at 1650° and 1850°F was started in May 1965. The 1500-hr exposures were completed on August 2, 1965. Mechanical testing and metallographic work were completed in September 1965.

The test was based on the thesis that in a graphite-moderated, helium-cooled reactor with a steam generator in the primary coolant loop, a steam leak would result in the formation of CO and H₂. These contaminants in helium can damage metallic materials at elevated temperature. Previous General Atomic work at 1100° and 1375°F showed this to be true to a limited extent. In March 1965 it was decided to extend the work to higher temperatures and to other candidate materials. The environment was 3000 ppm CO and H₂ in helium at 1650° and 1850°F. The materials were as follows:

1. 0.065-in. René 41 strip (precipitation hardened)
2. 0.065-in. Inconel X strip (precipitation hardened)
3. 0.060-in. Hastelloy X strip (solution annealed)
4. 0.060-in. Type 304 stainless strip (solution annealed)
5. 0.076-in. Type 430 stainless strip (CR 1450°F annealed)
6. 0.061-in. Incoloy 800 strip (solution annealed)

The data are presented graphically in Figs. 1, 2, 3, and 4. The graphs show the change in mechanical properties caused by the environment. It is apparent that the least affected materials, propertywise, at 1650°F are Type 430 stainless steel and Hastelloy X. At 1850°F, the least affected material was Hastelloy X.

Metallographic examination showed more clearly that Hastelloy X is the best candidate material at both temperatures and that Type 430

Fig. 1--Effect on René 41 and Inconel X of exposure to impure helium at 1650° and 1850°F

Fig. 2--Effect on Type 430 stainless steel and Incoloy 800 of exposure to impure helium at 1650° and 1850°F

Fig. 3--Effect on Hastelloy X and Type 304 stainless steel 800 of exposure to impure helium at 1650° and 1850°F

Fig. 4--Reaction of CO with metal specimens during the test

stainless, a cheaper material, was not significantly changed at 1650°F. René 41 and Inconel X overaged because of exposure to temperature above their aging temperature. They both showed severe intergranular attack and carburization followed by oxidation. Type 304 stainless steel showed grain growth and sensitivity at both temperatures and intergranular attack at 1850°F. Incoloy 800 showed grain growth and intergranular attack at both temperatures. Hastelloy X and Type 430 stainless were not attacked significantly at either temperature, however, Type 430 at 1850°F showed grain growth and formation of martensite and, possibly, an intermetallic compound.

Figure 4 graphically shows the change in gas environment. For the first 500 hr almost all of the CO was consumed by the specimens. After this time the CO consumption diminished to a relatively small amount, indicating that the diffusion rate of carbon into the metal was becoming very small. Metallographic examination showed that René 41 and Inconel X were the primary absorbers and that at some time after 500 hr they had developed an oxide layer that prevented further diffusion of the carbon.

The conclusion drawn at this time is that Hastelloy X is the best of the six materials at both temperatures as insulation and as a structural material. Type 430 stainless steel is satisfactory for thermal insulation to 1650°F, but should not be considered as a structural material for reasons other than those presented here.

PRIMARY COOLANT SYSTEM FLOW

Preliminary test specifications were completed for testing a primary coolant system utilizing downward flow through the core and steam generators. The testing program will utilize a primary coolant system model built to a scale of 0.3 which yields dimensions of about 8.85 ft inside diameter and 18.6 ft inside height. For the first three stages of testing the general model would be only a half cylindrical model with each test studying flow conditions in various regions of the system. The first stage of testing will study the flow characteristics and mixing of exit coolant from the bottom of the reflector blocks to the entrance to the steam generator.

COMPRESSORS AND AUXILIARY CIRCULATORS

Compressors

Purified Helium Compressor. An investigation is being made to explore various sources and types of compressors that would be suitable for use in the helium purification circuit.

Low-pressure Helium Transfer Compressor. A proposed test program for a nonlubricated reciprocating compressor in helium under simulated operating conditions has been prepared including a schematic for the test facility, an operating procedure, and a cost estimate. An inquiry specification for a test compressor is in preparation. Principal areas of investigation in the testing program are over-all performance, piston-ring wear in high-purity helium, and leakage rate.

Auxiliary Circulators

Preliminary design of the auxiliary circulator was initiated. A number of aerodynamic design studies for the compressor have been completed.

VALVES

Steam-Water Dump Valve

A preliminary computer program has been developed to determine the dump time and pressure conditions as a function of valve size and dump-line size.

In this approach, the steam generator inventory was dumped from the economizer-evaporator inlet header to a dump tank from which the steam was vented directly to the atmosphere. The dump valves closed when the pressure in the failed steam generator dropped to a point about 50 psi above the primary coolant loop pressure. A pressure control valve in the tank vent line maintained dump tank pressure above primary coolant loop pressure. This pressure differential precluded the entry of primary coolant helium into the dump tank and the possible release of activity to the atmosphere.

The present concept eliminates the possibility of activity release if the dump valve fails to close and the tank vent controller leaks. This is accomplished by dumping into a closed pressure suppression tank. The dump tank is normally half full of cold water, and the dumped steam is mixed with the cold water by ejectors. A review of pressure suppression systems confirms that it is possible to design such a system for PSC requirements with a reasonable tank size.

Discussions with engineers of potential dump valve vendors indicated that standard valves and actuators could meet the flow requirements as well as the rapid opening and closing times specified. The vendors do not have facilities to test the dump system under these conditions. Therefore, investigation of such facilities in other areas is continuing.

Helium Service Valves

As a result of inquiries to potential helium service valve suppliers, one vendor has supplied available information on thermal cycle testing of their bellows seal valves. Their testing facilities are being investigated to determine whether they are adequate or may be readily adapted to the proposed testing program.

INSTRUMENTATION AND ELECTRICAL

Moisture Detectors

Preliminary design criteria for the PSC plant primary coolant moisture monitoring system have been prepared. Two different types of instruments are being considered: (1) an indicating instrument with continuous display of moisture content on a recorder, and (2) a trip instrument for initiating various actions at different moisture levels. Effort thus far has been devoted to optically determined dewpoint-type instruments, and two operating modes for the detectors in the trip instruments have been considered. In one mode the mirror temperature in the detector would be manually adjusted to the frost or dewpoint temperature corresponding to the moisture content at which trip action is desired. In the other possible mode, the mirror temperature is automatically cycled slightly above and below the actual frost or dewpoint temperature of the gas sample in the detector with a resultant continuous indication of moisture content and trip at the set point(s). The recording instrument obviously must operate by the second mode. However, for trip instruments there are advantages and disadvantages to either mode, and a choice between the two will require further study.

Several different methods of providing cooling for the instrument mirrors are under consideration. These include the use of liquid nitrogen (available from a supply provided for the helium purification system) and Freon 22 or Freon 12 refrigeration units.

A preliminary design of a moisture detector PCRV penetration that incorporates a cooling water system for the sample gas has been prepared.

AUXILIARY EQUIPMENT, MANIPULATORS, AND TOOLS

A preliminary study was made to determine the nature of abnormal situations that might possibly arise during a refueling operation and the nature of special tools required to deal with these situations. The results of this study indicate that, in general, the short hexagonal-block fuel

element appears to present fewer handling problems than the rod type of fuel element. This is due primarily to the rugged and stable configuration of the hexagonal block and secondarily to the proposed method of grappling the element via a central hole in the block. The short block geometry of the fuel element appears to make it less vulnerable to massive fracture as a result of handling mishaps or dropping than is the case with a rod type of element.

A general purpose manipulator with one or more specific function attachments appears to be able to handle all the conceivable situations. Among these situations are a tipped element requiring righting, a stuck element, and a damaged element with chips and fragments broken off which require retrieving. The larger of these could be picked up by the manipulator and deposited in a receptacle provided for this purpose. Smaller fragments may be handled more easily with a vacuum attachment.

A broken control rod cable would require, in addition to a tool to cut off and retrieve the cable end, special facilities built into the auxiliary transfer cask for retrieving the control rod. A determination of the unusual situations involving the fuel transfer machine will have to await the detail design and testing of the machine.

It is apparent, as a result of this study, that the first requirement is a high quality remote viewing device to help determine the nature of an off-standard situation and to monitor and allow guidance of any remedial action. Studies are in process on both a conventional optical periscope system and a closed-circuit television system.

A literature search for information concerning the use of television cameras for remote reactor viewing yielded no useful information. Inquiries have been transmitted to potential suppliers outlining requirements and pertinent environmental conditions.

Some preliminary testing has been done on possible light sources to be used in conjunction with the viewing device. These include a G-E Quartzline lamp in conjunction with a polished stainless steel reflector, and a Sylvania Sungun motion picture light. Initial test results look promising.

TASK II

NUCLEAR ANALYSIS

FUEL CYCLE ANALYSIS

Equilibrium Fuel Cycle

The 330-Mw(e) HTGR uses a graded fuel cycle and operates on the U^{235} -Th- U^{233} fuel cycle. The design fuel residence time in the reactor is six years with annual refueling, so that roughly 1/6 of the core is replaced each year. Although these requirements can be achieved with a variety of Th and U loadings (usually characterized by the C/Th and C/U ratios), the reference fuel cycle has been chosen as a compromise between economic and engineering considerations. Figures 5 and 6 show the variation of age peaking, conversion ratio fissions per initial fissile atom (fifa), and relative fuel cycle cost (with private ownership) as a function of C/Th and average core carbon atom density. The latter parameter is of importance if the use of less-expensive, low-density graphite is considered. Clearly, fuel cycle cost and age peaking factor are very sensitive to changes in the C/Th ratio. Although the fuel cycle cost decreases with light loadings, the resulting lower conversion ratio and larger fifa make the age peaking factor increase considerably. The limit on the maximum fuel temperature combined with a gross radial peaking of 1.15 require an age peaking factor of 1.4 or less. This leads directly to the reference fuel cycle with a C/Th = 200 and C/U = 2350 in the nominal equilibrium feed. The variation in the material content of 1/6 of the core during its six-year residence time is shown in Table 2. It is clear that the majority of the power generated after three years of operation comes from the bred U^{233} .

The estimated burnup after six years of irradiation is 86,000 Mwd/t or 1.24 fifa. The material variation in the entire core during a yearly cycle is shown in Table 3.

Given in Table 4 is a neutron balance for the equilibrium fuel cycle. The principal neutron losses are associated with the leakage, fission products, and burnable and control poison. The increase in conversion ratio through the cycle is due to the increasing fraction of fuel absorptions in U^{233} and to the reduction in losses to control poison, which more than compensates the increased losses to fission products.

Fig. 5--Variation of fuel cycle cost and age peaking factor with carbon density and C/Th ratio

Fig. 6--Variation of conversion ratio and fuel burnup with carbon density and C/Th ratio

Table 2

MATERIAL CONTENTS OF 1/6 OF THE CORE

Residence Time (years)	Weight (kg)						
	Th	Pa ²³³	U ²³³	U ²³⁵	Total U	Total Pu	Total Fissile
0	2725	0.0	0.0	226	242	0.0	226
1	2692	4.27	23.5	158	211	0.463	181.9
2	2659	4.22	41.5	110	192	0.674	152.0
3	2626	4.17	52.9	78	178	0.776	131.4
4	2594	4.12	60.1	55	167	0.827	115.6
5	2563	4.07	64.5	40	160	0.850	105.0
6	2532	4.02	67.0	29	155	0.859	96.5

Table 3

MATERIAL CONTENT OF EQUILIBRIUM CORE

Material	Weight (kg)	
	Beginning of Cycle	End of Cycle
Thorium	15859	15666
Pa ²³³	14.6	24.9
U ²³³	248.7	309.5
U ²³⁵	667.6	470.7
Total U	1156.7	1063.3
Pu ²³⁹	1.82	2.11
Pu ²⁴¹	0.68	0.86
Total Pu	3.63	4.45
Total fissile	918.8	783.2

Table 4

EQUILIBRIUM CYCLE NEUTRON BALANCE

	Beginning of Cycle	End of Cycle
K_{eff} (without burnable poison or control and with the rapidly saturating fission products)	1.066	1.010
Conversion ratio	0.590	0.643
Neutron productions per absorptions in a fissile nuclide	2.01	2.04
Losses per absorption in a fissile nuclide, %:		
Leakage	9	9
Absorptions in Th^{232} , U^{234} , U^{238} , Pu^{240}	60	66
Absorptions in Pa^{233} , Np^{239}	1	1
Absorptions in U^{236} , Np^{237} , Pu^{242}	3	4
Absorptions in rapidly saturating fission products (Xe^{135} + Sm^{149})	4	5
Absorptions in remaining fission products	10	15
Absorptions in moderator	2	2
Absorptions in burnable poison and control	13	2
Absorptions in U^{233}	31	43
Absorptions in U^{235}	68	55
Absorptions in Pu^{239} , Pu^{241}	1	2

Approach to Equilibrium

Inherent in the graded fuel cycle is the requirement that most of the first and the last cores have to be removed before achieving their full exposure. If one-sixth of the core is removed after one year and if the reactor is refueled one year prior to the end of its 30-year life, there results a penalty of about 7% on the fuel cycle cost during the entire life of the reactor. However, if the first sixth is removed after two years, i.e., one-sixth is allowed to spend seven years in the reactor (and correspondingly at the end of life), this penalty is reduced to about 3%. Thus, it is proposed that

the initial core be operated for two years without refueling, with annual refueling thereafter.

The three primary considerations in determining the approach to equilibrium are:

1. Fuel cycle cost,
2. Requirements on the control system, and
3. Power peaking.

Assuming a constant equilibrium feed at every reload, one way is to build a complete mockup of the equilibrium core. In this case the zone that is to be removed after two years is given a fissile content equivalent to four-year-old fuel and fuel which comes out after six years obtains the nominal equilibrium feed. Additional excess reactivity resulting from the lack of fission products in the first core is compensated for by an increased thorium loading in the zones to be removed after two, three, and four years of operation. In this case the age peaking of the equilibrium cycle has to be expected right from the beginning. This type of approach to equilibrium should be the most economical and would not materially modify the control requirements relative to the equilibrium cycle. A second approach would be a completely uniformly loaded core which results in very low age peaking during the first years of operation. Subsequently the age peaking factor slowly increases to its equilibrium value. In this case the initial core consists of only fresh equilibrium fuel and obviously large amounts of excess reactivity must be controlled. Further, this type of approach to equilibrium is not very economical.

The initial core loading has been chosen as a compromise between these two extremes and corresponds to a partial mockup. It combines the advantages of both extremes in that the end-of-cycle excess reactivity is close to the desired 1% and, in addition, a comparatively low age peaking is obtained during the first five years. To further reduce the age peaking, the initial core will be more heavily loaded (average C/Th = 165) than the equilibrium core (C/Th = 200). The age peaking factors for the three approaches to equilibrium are shown in Fig. 7. The nominal fuel loadings of the initial core are shown in Table 5; a summary of the quantities of fuel that are discharged from the initial core is given in Table 6. Table 7 shows some physics parameters during the first eight years. At the end of six years, the end-of-cycle k_{eff} falls below the desired 1% excess reactivity margin because of the higher-than-average burnup at this point. This will be compensated for by a slight modification of the feed fuel in preceding and succeeding reloads.

Fig. 7--Age peaking factor versus core lifetime

Table 5
FUEL LOADINGS IN THE INITIAL CORE

Period until zone is discharged (years)	Weight (kg)		
	Th ²³²	U ²³⁵	Total U
2	3710	100	107
3	3660	160	171
4	3460	200	214
5	3090	230	246
6	3090	230	246
7	3090	230	246
Total	20000	1150	1231

Table 6
QUANTITIES OF FUEL DISCHARGED DURING
THE APPROACH TO EQUILIBRIUM

Fuel	Quantity of Fuel (kg)						Equilibrium
	Discharged After Year 2	Discharged After Year 3	Discharged After Year 4	Discharged After Year 5	Discharged After Year 6	Discharged After Year 7	
Th ²³²	3630	3450	3310	2920	2880	2850	2533
Pa ²³³	5.3	5.0	4.9	4.4	4.5	4.4	4.0
U ²³³	52.9	66.5	74.5	72.0	75.9	78.1	67.0
U ²³⁵	53.1	62.0	56.7	47.4	35.3	26.9	29.0
Total U	125	164	179	176	171	167	155
Pu ²³⁹	0.2	0.3	0.4	0.4	0.4	0.3	0.4
Pu ²⁴¹	0	0.1	0.2	0.2	0.2	0.2	0.2
Total fissile	106	129	132	120	112	106	96

Table 7
SOME PHYSICS PARAMETERS DURING
THE APPROACH TO EQUILIBRIUM

Year	k_{eff}^a	k_{eff} End	Average Conversion	Age Peaking
1	1.10	---	---	1.20
2	---	1.01	---	1.20
3	1.06	1.01	0.66	1.22
4	1.06	1.01	0.64	1.23
5	1.06	1.01	0.63	1.26
6	1.06	1.01	0.64	1.36
7	1.06	1.01	0.63	1.29
8	1.07	1.02	0.62	1.28
Equilibrium	1.07	1.01	0.62	1.34

NOTE: Without control or burnable poison.

^aBeginning with rapidly saturating fission products.

FUEL ZONING AND POWER DISTRIBUTION

Transverse Power Distribution

The core is divided into 37 roughly hexagonal regions each containing two control rods and from 60 to 84 fuel elements. Each region is serviced through a single nozzle and will be replaced in its entirety on refueling. Figure 8 shows the arrangement of the regions within the core. The numbers inside the regions denote the fuel age in years at one particular time of the equilibrium cycle just prior to refueling. The particular distribution of ages has the interesting property of a 60° rotation. That is, a year of operation is equivalent to rotating Fig. 8 60° in the clockwise direction. It is quite important, if flux tilting is to be minimized, to ensure that groupings of "young" and "old" fuel do not occur predominantly in any area of the core.

The use of a graded fuel cycle requires that fresh fuel blocks and irradiated blocks adjoin. This results in "age peaking," and thus, the total transverse peaking is a combination of radial peaking and age peaking. In addition, flux tilting depends on the azimuthal fuel age distribution. One design objective is to maintain the flux tilting below 1.1 at all times. In order to minimize flux tilting across the core, the following criteria have been used for choosing the distribution of the regions shown in Fig. 8.

1. Except for the central region, no regions of equal age should adjoin.
2. Each ring should contain an equal number of each age.

The radial power distribution has been flattened by using five different fuel zones. The zone boundaries have been chosen so that they coincide with the region boundaries in each ring, and the gross radial peaking is maintained below 1.15. The central region, which forms the central fuel zone, will be loaded so that its power density never greatly exceeds the core average. Currently, it is planned to operate the central control rod pair by automatic control and thus accommodate a large fraction of the reactivity swing in a yearly cycle.

Since the axial power distribution is tipped toward the top of the core, there is some concern that the top mounted rods might invert this distribution. If the distribution were inserted in a rodded region, the transverse peaking in this region would have to be maintained at about unity if excessive fuel temperatures are to be avoided.

The outer rings of fuel blocks are the only ones that are not loaded uniformly. In order to avoid power peaks at the core-reflector interface

Fig. 8--Refueling regions and distribution of ages

because of the inleakage of cold neutrons from the reflector, the outer four rows of fuel holes will have a reduced uranium and increased thorium loading. Figure 9 shows the gross radial power distribution.

Some control rod programs that result in little transverse flux tilting have been developed. The preliminary burnup calculations indicate that four control rod pairs, the central one and three in the outer ring of regions, will be sufficient to control the reactivity during a yearly cycle. In the outer ring three central rod pairs are inserted, in a roughly symmetric manner, into regions that are three or more years old, again to obviate problems with the axial power distribution. Figure 10 shows the relative peak power in each region and relative average power in each region for an unrodded and a rodded core at the end of an equilibrium cycle, just before refueling. For the rodded core, the rods are inserted into the shaded regions of Fig. 10. Because the fresh blocks contain about 70 ppm of burnable boron poison, which burns out nearly completely during the first year, this condition is believed to indicate the maximum transverse peaking. The peaks within the young regions occur at the boundaries and are caused by a neighboring lightly loaded (burned out) zone feeding softer spectrum neutrons into a more heavily loaded zone.

The fraction of the core having a transverse peaking factor that is in excess of any given value is shown in Fig. 11 for both the rodded and unrodded core. Comparing the power peaking for the rodded and unrodded core, it is clear that the proposed control rod program does not adversely affect the transverse power distribution.

Axial Power Distribution

The most advantageous axial power distribution in a gas-cooled reactor, from the standpoint of fuel temperature, is exponential in shape and decreases from inlet to outlet end. This desired exponential power shape has been approximated by zoning the fertile and fissile material into five different zones, such that the distribution remains relatively time invariant with burnup. The axial fuel zones are chosen to coincide with the fuel block boundaries. Having 12 fuel blocks in one stack, the zoning is such that each of the first three blocks has a different composition. The next two zones consist of four and five fuel elements.

The resulting strong variation of fuel loading per block implies that different sizes of fuel holes have to be used in different blocks. This affects the nuclear calculations in two ways: (1) the average moderator density varies, and (2) the effective cross sections change, particularly the self-shielded thorium, as a function of hole size. These effects have been incorporated into the nuclear calculations.

Fig. 9--Gross radial power distribution

Fig. 10--Transverse power distribution

Fig. 11--Integral power curve in the equilibrium core at end of cycle

Burnable poison has been included to control initial excess reactivity; it has been zoned together with the fertile and fissile fuel such that its perturbation to the power shape with burnup is minimized. Figure 12 shows the ideal exponential power shape together with a typical approximation using five zones.

Feed Compositions

Considering the five radial and five axial feed compositions, a total of 25 types of fuel blocks is required. The compositions and locations of these 25 zones are shown in Fig. 13. No attempt has yet been made to associate the fuel elements of relatively similar composition. This will decrease the number of different fuel blocks significantly with a minimal change in power distribution.

Kinetics Analysis

The control system reactivity worth should be capable of covering the following reactivity losses:

1.	Loss in reactivity in reaching operating temperature (temperature defect)	7% ΔK
2.	Loss in reactivity due to the buildup of rapidly saturating fission products (Xe, Sm, etc.) and Pa decay . .	3% ΔK
3.	Loss in reactivity due to fuel depletion and fission product buildup, or "maximum expected excess reactivity"	$\sim 3\%$ ΔK
4.	Shutdown margin	$\sim 3\%$ ΔK
Total		<u>16%</u>

Considering an uncertainty of 10% in the calculation of the control system worth and one stuck control rod pair, the control system should be capable of inserting from 18% to 19% ΔK .

Several studies have been performed in order to determine a control system that minimizes the space required for the rods in the core and supplies sufficient strength. In the required design, 3 in. OD hollow cylindrical rods containing 30 wt-% B_4C in graphite will be used. The total worth of the 37 rod pairs is calculated as 18.4% ΔK .

Fig. 12--Axial power distribution in the equilibrium core

Fig. 13--Feed composition and fuel zones

TASK III

FUEL DEVELOPMENT

FUEL ELEMENT DESIGN

A short, block type of fuel element has been selected as the reference configuration for the PSC plant. Each fuel block is hexagonal in shape and is approximately equal in length to the width across the flats of the hexagonal cross section.

The fueled blocks are stacked end to end in parallel columns with unfueled graphite reflector blocks at the top and bottom of the individual columns to form a brickwork type of core arrangement. The fuel particles are contained within an array of holes drilled axially into each block. The coolant holes are drilled through the blocks parallel to the fuel holes and, when the blocks are installed in the core, they align with the coolant holes in the adjacent blocks above and below. Essentially all of the reactor coolant is directed through the holes within the blocks.

One of the major advantages expected for this type of element is the structural ruggedness of the short-block geometry. The shape is highly resistant to breakage during handling, both inside and outside of the reactor vessel. Also, the shape is more adaptable to semiautomatic machining operations and shows promise of reducing total fabrication costs from those expected for a cylindrical element. Graphite materials that do not meet the tensile strength requirements of the cylindrical element can be considered for the hexagonal block, making a wider selection of graphite materials possible with potentially lower core fabrication costs. In addition, preliminary thermal analyses have shown that maximum fuel temperatures and average graphite temperatures within the hexagonal block will probably be lower than the expected temperatures within the 20-ft cylindrical element. The lower fuel and graphite temperatures are expected to improve the retention of metallic fission products, such as strontium, barium, and cesium, within the fuel elements.

The preliminary design and performance studies leading to selection of the hexagonal-fuel-block design were reported in the HTGR Base Program quarterly report (GA-6677). For convenience, the preliminary design studies are briefly summarized in this first quarterly report.

Preliminary Design Study

Mechanical Design. A design and performance evaluation study has been completed to develop a preliminary hexagonal-block fuel element design. The study was based on the preliminary design conditions established for the PSC plant. For purposes of evaluation, performance of the fuel block was compared with the predicted performance of the 4.7-in.-diam., 20-ft-long cylindrical fuel element design.

Based on previous parametric studies of performance characteristics for various types of cylindrical fuel elements, the design having an annular fuel bed with an annular internal coolant passage has been found to perform satisfactorily with the added advantage that it has lower differential graphite contraction strains in the graphite structural sleeves. Since this type of element is representative of near-optimum performance for the cylindrical-element concept, it was selected for the comparison standard. Table 8 gives the preliminary core design parameters used for this study.

Table 8
CORE PARAMETERS USED IN FUEL ELEMENT
PERFORMANCE STUDIES

<u>Parameter</u>	<u>Value</u>
Total core flow rate	3.694×10^6 lb/hr
Fraction of coolant bypassing core (for side reflector and control rod cooling)	8%
Total core power	843 Mw(t)
Core inlet temperature	800°F
Core outlet temperature (mixed mean)	1427°F
Core pressure drop	5.5 psi max
Radial peak/average power	1.6
Coolant pressure	450 psia
Fuel bed area fraction for bottom fuel zone (max. power region)	0.27

Owing to core arrangement considerations, the two cores, though not identical, are quite similar in area. Table 9 is a list of the core layout parameters used for the study.

Table 9
CORE LAYOUT PARAMETERS

Item	Hexagonal Fuel Block	Cylindrical Element
Core area (with control rods), ft ²	300.03	295.85
Core area (no control rods), ft ²	290.20	286.02
Number of control rods	74	74
Fuel element pitch (cold), in.	14.212	4.700
Number of refueling regions	37	37
Number of fuel elements per reloading region	84	62 ^a
Number of fuel columns	247	2151
Total number of hexagonal fuel blocks	2964	---
Total number of hexagonal upper and lower reflector blocks	1235	---

^a—Plus two control rods.

The fuel block is basically a short block of graphite of hexagonal cross section that is about 14.2 in. across the flats and 15.6 in. long axially. Twelve of these blocks are stacked on top of each other in parallel columns to make up the active core length. Coolant holes, drilled through the blocks, are aligned with coolant holes in adjacent blocks above and below, when installed in the core, to provide continuous coolant channels. The fuel holes are drilled parallel with the coolant holes in the block. The fuel holes are all drilled from the top face and extend to within 1/4 in. of the opposite face. The fuel holes are filled with coated fuel particles and are contained within the hole by a plug at the upper end of the fuel hole. Additional blocks of graphite (with coolant holes) are added to make up the top and bottom end reflector regions. One column of hexagonal fuel blocks is equivalent in area to slightly more than nine of the 4.7-in.-diam cylindrical fuel elements in the core array. A core refueling region (except some regions adjacent to the side reflector) contains seven fuel-element columns.

The width dimension of 14.2 in. across the flats of the hexagonal-block element was selected for the study to retain the same refueling region size with the seven-column array of block-type elements as that established for the cylindrical elements containing 62 fuel elements. Each refueling region is centered under a control rod drive nozzle, which is also used for refueling access. Two control rods are utilized for each zone, and they are operated by a common drive assembly. The axial length of 15.6 in. was selected to provide for accurate, deep drilling of

the fuel and coolant holes and to provide for ease of element handling both inside and outside of the reactor vessel. Figure 14 shows a typical hexagonal-block element and a seven-block fuel region arrangement.

The fuel and coolant holes are located on a triangular array in which, ideally, there are three fuel holes for each coolant hole. Because of edge effects, the block contains 91 coolant holes and 240 fuel holes, giving a ratio of 2.64 fuel holes to coolant holes. There are two sizes of coolant holes in the block, 0.650 in. and 0.550 in. The 30 smaller coolant holes are located along the edges of the hexagonal block. These holes are smaller because they remove heat from only two fuel holes instead of three. The remaining 61 coolant holes are the larger size, and each coolant hole removes the heat from three fuel holes. The coolant holes are on a triangular pitch of 1.520 in. The fuel hole and coolant hole centerline locations remain constant in all fuel blocks. The fuel hole diameter varies, however, to accommodate the various axial fuel loadings. Because of the varying fuel hole size, the web thickness between fuel and coolant holes and adjacent fuel holes varies with each fuel zone.

Lateral alignment of blocks within each column is provided by three graphite pins in the top of the block which fit into three holes in the bottom of the adjacent block above. Each seven-column refueling region is arranged with the three pins oriented as shown in Fig. 14. The particular arrangement of the pins on the block face was chosen so that the pins would not interfere with the control rod holes within the central fuel block. Each graphite dowel pin is located over a coolant hole and has a hollow center to allow for normal coolant flow through the holes. The two large control rod holes through the central block of each region are also held in alignment by the dowel pins.

Thermal Design. The selection of the fuel and coolant hole arrangement and coolant hole diameters was based on a number of basic design requirements. One of the primary requirements is to allow the required reactor coolant flow rate to pass through the core within the specified maximum pressure drop. A second requirement is the ability to accommodate the required fuel loadings with a suitable fuel-hole-to-coolant-hole web thickness to maintain mechanical integrity.

A study was made of the interrelationship between coolant hole diameter and number of coolant holes in the core to meet the pressure drop and total flow requirements. The results showed that the core coolant channel voidage is decreased as the individual coolant hole diameters are increased. A coolant hole diameter range of 0.6 in. to 1.4 in. was selected as the range of interest for the fuel hole-coolant hole arrangement study.

Fig. 14--The hexagonal fuel block; shown at the top is a group of seven fuel columns forming a refueling region

After an examination of various fuel hole-coolant hole arrangements, it was seen that a large number of combinations should be considered to optimize the arrangement. The number of fuel holes for each coolant hole (FH/CH ratios) considered in the study were 1:1, 2:1, 3:1, 4:1, 6:1, and 12:1. Using the coolant hole diameter versus voidage information and a fixed fuel loading, fuel hole diameters and the graphite web thicknesses between fuel holes and between fuel and coolant holes were calculated for coolant hole diameters of 0.6, 1.0, and 1.4. For the 0.6 in. case, the maximum usable FH/CH ratio was 4.1. Beyond this, the web between adjacent fuel holes was too small. All the combinations listed above were possible for the other coolant hole sizes, although for some of the arrangements the web thickness was becoming quite small.

Thermal performance was analyzed for several of the possible fuel hole arrangements with each coolant hole size. All of the thermal analyses were performed for conditions at the bottom of the active core where high power and heavy fuel loading coincide to standardize the calculation procedure and make all the results directly comparable. A thermal analysis of the annular bed cylindrical fuel element was also made at the same location to provide comparable results. A summary of the thermal analysis is shown on Fig. 15. The figure shows maximum fuel temperatures and maximum graphite temperature differences. It is seen that only results for the 0.6- and 1.0-in. coolant holes are shown. The temperatures for the 1.4-in. coolant hole are not shown on the figure, other than one point, because the fuel temperatures consistently ran above 3400°F for the cases analyzed (4:1 and 6:1 FH/CH ratios). Such fuel temperatures are not acceptable, and thus, large coolant holes (above 1.0-in. diam) did not appear practical. A 0.65-in. diam coolant hole with three fuel holes per coolant hole was selected as a near optimum arrangement for the reactor conditions used in the study.

Local temperature profiles within a region of symmetry in the interior of the hexagonal block were calculated for various axial locations for elements experiencing peak power and for average power blocks. The axial temperature profile for the hottest fuel block is shown in Fig. 16. These temperature profiles were computed with a simplified axial power profile. It should be noted from Fig. 16 that the maximum fuel temperature occurs near the top of the active fuel region, indicating that the axial power shape selected for the study is not an optimum for the hexagonal-block element.

For comparison, axial temperature profiles for the cylindrical fuel element are shown in Fig. 17. It should be noted that maximum fuel temperature is almost a constant over the length of the element. A five-zone axial fuel loading profile was found to produce an approximate optimum power profile for this element.

Fig. 15--Parametric study of maximum fuel and graphite temperatures versus number of fuel holes per coolant hole for the hexagonal fuel block; a similar study for the annular-bed cylindrical fuel element is given for comparison

Fig. 16--Axial temperature profile for the hottest hexagonal fuel block

Fig. 17--Axial temperature profile for the hottest cylindrical fuel element

An analysis to determine the amount of coolant bypass flow through the clearance gaps between adjacent columns of hexagonal blocks within the core array was completed using preliminary data for gap widths at full power operating conditions. Total flow through the gaps was estimated to be 5.5% of the primary circuit helium flow.

Irradiation-induced Graphite Distortion. The effects of fast neutron exposure on the dimensional stability and structural properties of the fuel block graphite is an important factor in the over-all evaluation of fuel block performance. From available experimental data, the major effect of large fast-neutron exposures on graphite at HTGR temperature levels is to cause the material to contract. The data show that extruded graphite bodies contract at a higher rate in the parallel direction (i. e., along the axis of extrusion) than in the perpendicular or radial direction. Also, the rate of contraction is strongly temperature dependent.

During reactor operation, the HTGR fuel blocks will have temperature gradients present within the graphite bodies. As a result, the temperature dependence of the graphite contraction will cause differential contraction strains to be induced in the graphite bodies. The largest temperature differences are in the radial direction (i. e., parallel with the primary heat flow paths within the graphite). For extruded graphite bodies, the resulting internal differential contraction strains are greatest in the axial direction. As exposure accumulates, the graphite contracts more rapidly in the hottest parts of the body to produce a strain distribution similar to thermal strain differences but with the locations of tensile and compressive strains interchanged. The magnitude of the contraction strain differences is normally much greater than the differential thermal expansion, and thus strain in the higher temperature regions becomes tensile quite early with respect to the design fuel block lifetime.

A survey analysis of the local differential contraction strains within the hexagonal fuel block was conducted for comparison with the cylindrical type of element. At the location within the core where the largest differential strains were predicted, a maximum accumulated tensile strain of 0.26% $\Delta L/L$ was calculated. The corresponding peak tensile strain rate was calculated to be about 0.01% $\Delta L/L$ per 10^{20} nvt. Irradiation tensile strain tests have demonstrated that needle coke graphites of the type under consideration can accommodate 0.4% $\Delta L/L$ tensile creep strain under irradiation at strain rates up to 0.1% $\Delta L/L$ per 10^{20} nvt without failure. The maximum contraction-induced elastic tensile strain (maximum tensile stress) will occur when the differential contraction strain rate is at a maximum. Since the maximum tensile strain rate is about one tenth of the value observed experimentally without failure, it can be confidently concluded that elastic stress levels will be within acceptable limits for the graphite material.

Gross distortion of the fuel blocks due to fast-neutron exposure was estimated during the preliminary study. In general, gross axial contraction of the individual blocks does not present a serious design problem in that the fuel block columns are not restrained axially and are thus free to contract without separation at the end interfaces of the individual blocks.

Fuel columns located adjacent to the side reflector and those containing inserted control rods are, in addition, subject to regional fast-neutron exposure gradients and variations in temperature caused by local fission power differences. Fuel blocks adjacent to the side reflector were found to experience the greatest over-all distortion because of the steep decrease in fast-neutron exposure as the side reflector is approached.

Axial contraction of segments of a fuel block adjacent to the side reflector were evaluated incrementally across the block from the reflector interface inward. Figure 18 shows the resulting length change in a block located at a horizontal plane 2 ft above the bottom of the active core. Blocks in this region would bow, assuming that the contraction profile is linear across the block, to a centerline radius of about 1800 in. If it is assumed that the bowed block is essentially held vertical by other adjacent blocks with respect to a centerline tangent at the axial block midpoint, the radial deflection (between a chord line intersecting the centerline at the two ends and the vertical tangent to the centerline at the midpoint) would be about 0.02 in.

If the edge surfaces of adjacent blocks within a column were in contact at the point of minimum contraction next to the side reflector, gaps of about 1/8 in. would accumulate between blocks at the inner edges toward the core center. However, only blocks located within the lower one third of the active core length would experience this much contraction difference.

Blocks located about 10 ft above the bottom of the active core would accumulate axial contraction differences radially inward from one side to the other of about 0.04 in.

A free-standing column of hexagonal fuel blocks located adjacent to the side reflector was estimated to have an accumulated centerline deflection at the top of the column of 7 to 10 in. owing to differential axial contractions of individual blocks if all block end surfaces were allowed to come into contact.

The maximum radial contraction predicted for fuel blocks located in peak fast-neutron exposure regions is about 1% $\Delta L/L$, corresponding to about 0.14 in. of contraction across the flats of the hexagonal block.

Fig. 18--Estimated axial contraction of blocks adjacent to the side reflector at end of six-year life

DESIGN

Mechanical Design. The preliminary fuel block design was modified during the quarter to provide a stepped central hole to accommodate the fuel transfer machine grappling head. The fuel block grappling hole requires an area equivalent to six fuel holes and one coolant hole. Figure 19 shows the arrangement of holes within a typical fuel block containing the grappling hole.

Further study of the available graphite contraction data has been undertaken in an effort to better define, for fuel block performance analysis, the temperature dependency of contraction above 800°C. It appears that a very steep increase in contraction versus temperature at constant neutron exposure between 800° and 900°C might be required to best fit the currently available data, with the curve becoming essentially flat above 1000°C. Further contraction analyses are currently in process using the revised curves.

Thermal Design. An analysis of the over-all core thermal performance was completed this period using the preliminary PSC design conditions of system pressure, coolant weight flow, and temperatures. Temperature distributions within fuel blocks were calculated during the preliminary design study for various radial power peaking factors that are representative of fuel blocks of differing ages and radial locations within the core. The fraction of the core versus peaking factor (combined age peaking and radial peaking) is shown in Fig. 20 for the equilibrium, region-refueled core.

The simplified, axially stepped power distribution used for these analyses was developed to approximate the axial power distribution expected with five axial fuel loading zones arranged to provide a near optimum exponential axial power shape.

Axial coolant temperature profiles through the core for various peaking factors are shown in Fig. 21. The calculations were based on a uniform coolant hole size for all fuel blocks and end reflectors, with no orificing of low-power core regions.

The over-all predicted thermal performance of the core without orificing is summarized in Fig. 22, which shows the fraction of the total active fuel and the fraction of coolant channel surface above given ordinate temperature levels.

A study was initiated this quarter to evaluate the performance advantages and problems associated with orificing the PSC reactor

Fig. 19--Arrangement of fuel and coolant holes in a typical fuel block containing a central grappling hole

Fig. 20--Integral power curve for the equilibrium core at end of cycle

Fig. 21--Axial coolant temperature distribution

Fig. 22--Fraction of fuel bed and fuel block surface versus temperature

core. The objectives of orificing are to reduce the maximum fuel and graphite temperatures within the fuel blocks and to reduce the local peak exit coolant temperatures from higher power new fuel regions, thus providing a more nearly uniform reactor exit temperature. Coolant losses through mating block end faces from unorificed high-inlet-pressure new regions into older and orificed regions appear to be the most significant factor affecting orificed core performance compared to ideal performance with orificing.

Fission Product Transport. Preliminary work has been done during this period to evaluate the primary coolant circuit gaseous and plateout fission-product activities. Analytical work has been carried out during this period to develop a computer program (FREVP) to aid in predicting the release of metallic and condensable fission products from the particles and fuel blocks.

A report on the RAD-2* computer program for calculating fission-product activity distributions in a gas-cooled reactor system was completed this period. The RAD-2 program is used to evaluate the production of fission products and the release of the fission products from the fuel blocks assuming gaseous diffusion as the transport mechanism. Also, the program performs the bookkeeping calculations for radioactive fission product decay chains of any desired length.

Experiments are under way to measure the steam-oxidation rate and integrity (with respect to fission gas retention) of unirradiated and irradiated PyC-coated fuel particles.

Preliminary tests have utilized both irradiated and unirradiated PyC-coated (Th,U)C₂ particles from Sample 2 (see Table 14). The technique is to expose particles to a steam-helium gas mixture at temperatures in the range 1000° to 1500°C. In this work, the effluent gas is first dried to remove excess moisture and then allowed to pass through a gas chromatograph for analysis of carbon-oxidation products (CO and CO₂) as well as carbide-hydrolysis products (hydrocarbons). The gas also passes through an ionization chamber to detect any gaseous radioactive species (especially Kr⁸⁵), thereby yielding a precise measurement of the time of coating failure.

The experiments were conducted using 2.5% steam in helium flowing at 300 cm³/min. The system pressure was 5 psig. The temperature

*F. E. Vanslager, RAD-2, A Computer Program For Calculating Fission Product Radioactivities, General Atomic Report GAMD-6519, July 23, 1965.

was varied from 1176° to 1420°C. Nickel gauze, platinum gauze, a closed EGCR-graphite crucible, an open graphite boat, and an open quartz boat have been used as containers for the particles.

The experimental results to date are given in Table 10. The results show that the unirradiated particles contained in nickel gauze and exposed to 2.5% steam at 1200°C ruptured in about 20 min, whereas the platinum and quartz contained particles ruptured in 30 to 138 min and 362 min, respectively. This behavior indicates that the container materials affect the reaction rate. Nickel is an excellent catalyst for the steam-graphite reaction, whereas platinum is believed to be inert. Silica, on the other hand is a poison, presumably because it either interferes with the natural catalysts present or forms a protective layer of silicon carbide on the carbon surface.

Irradiated particles in a platinum container showed coating failure in around 30 min when exposed at 1200°C. This suggests that metallic fission products catalyze the reaction and cause premature rupture, as expected.

Photomicrographs taken of the oxidized specimens (unirradiated) show that the oxidative attack is quite localized. At the beginning of the process, small pin holes develop, usually only 2 or 3 per particle. Next, the pin holes enlarge somewhat and penetrate to the kernel. The carbide kernel is then rapidly oxidized and/or hydrolyzed; and it increases in volume resulting in a fissure, and ultimately causes the particle coating to split into hemispheric sections. In some cases, however, the coating is not split open, and the particle appears covered with pot holes and craters.

Localized attack explains why coating failures are seen in a short time even though the oxidation rates (by CO and CO₂ analysis) are quite low, i. e., about 1% per hour for unirradiated particles and from 5% to 15% per hour for the irradiated specimens.

The above preliminary experiments yielded reaction rates and rupture times for individual particles. The next step in the investigation was an attempt to duplicate more closely the conditions in the PSC reactor.

In the PSC fuel block, steam must diffuse through and react with a graphite septum before it contacts the fuel particles. The minimum septum thickness is 0.15 in. (at the cold end) and the maximum is 0.25 in. (at the hot end). Accordingly, tests were run in which particles were encased in closed EGCR graphite crucibles having wall thicknesses of

Table 10

RESULTS OF STEAM-OXIDATION EXPERIMENTS ON PyC-COATED (Th, U)C₂ PARTICLES^a

Experiment No.	No. of Particles	Irradiated ^b	Sample Container	Temp. (°C)	Time to Rupture (min.)	Oxidation Rate (%/min)	Comments
6, 7, 8	4	No	Ni gauze	1200	20-22	---	Carbide oxidation products seen
10	1	No	Quartz	1200	362	---	Carbide oxidation products seen.
11, 23, J-3,	3	No	Pt gauze	1210	61, 138, 54	---	Xe ¹³³ analysis, TRIGA irradiated
24	5	No	Pt gauze	1220	30-100	---	Xe ¹³³ analysis, TRIGA irradiated
13, 20, 21, 25	4	Yes	Pt gauze	1220	74, 24, 29, 26	0.2	Kr ⁸⁵ analysis
19	4	Yes	Pt gauze	1230	20-40	---	Kr ⁸⁵ analysis
17	1	Yes	0.1 in. EGCR graphite crucible	1220	625	0.25	Kr ⁸⁵ analysis
18	3	Yes	0.1 in. EGCR graphite crucible	1230	318-348	0.25	Kr ⁸⁵ analysis
J-12	8	No	Pt gauze	1176	90, 185, 220, 240	---	Xe ¹³³ analysis. Two particles intact after 350-min exposure
J-13	8	No	0.15 in. EGCR graphite crucible	1210	>21.6 hr	0.05	
J-17	8	No	0.15 in. EGCR graphite crucible	1220	14-17 hr	---	Graphite can almost completely eroded, particles still intact
J-15	8	No	Pt gauze	1400	12-21	---	
J-18	8	No	0.15 in. EGCR graphite crucible	1410	330-430	---	

^aConditions: 2.5% steam, 5 psig He at 300 cm³/min.

0.10 and 0.15 in. Failures at 1220°C were noted after 5 to 10 hr in the 0.10-in. wall crucibles and in 14 to 17 hr for particles contained in 0.15-in. wall crucibles. Another experiment using a 0.15-in. wall crucible was terminated after 21.6 hr, and coating failure had not yet occurred. It is pertinent that in all of the crucible tests particle failure did not occur until a major fraction (50% to 100%) of the surrounding graphite had been oxidized. This suggests that in a major steam-leak accident the chief concern will be the integrity of the fuel block and not the oxidation of the fuel.

MATERIALS

Particle Beds

Fuel development studies are directed toward evaluating the physical behavior of loose coated-particle fuel systems in out-of-pile simulated reactor environments. These studies are intended to guide the design of fuel blocks employing loose, coated particles, and to guide the selection of fuel that will meet the performance requirements of the PSC reactor.

Packing and Vibration Studies. Packing studies are being performed to develop practical means of blending loose fissile and fertile particles and of loading such blends into fuel blocks without segregation of the two fuel types. In addition, packing densities are being measured for various blends of the reference coated particles. Vibration tests are being conducted to determine the extent of segregation in blended particle beds exposed to a vibration environment.

Segregation tests were conducted using coated fuel particles representative of the large (fertile) reference size, blended with glass beads in the small (fissile) reference size. (Particle parameters are given in Table 11). A mixture of 10 wt-% coated fuel particles with 90 wt-% glass beads was V-blended (the most effective blending method according to previous tests) and loaded into a 30-in. sampling tube. The bed was vibrated at 0.5 G (longitudinal at 120 cps) for a prescribed length of time. The particle column was then split into the standard 30 samples, which were analyzed for weight content of coated particles. Tests were run for various vibration times (8 to 64 hr).

An air vibrator used in the initial tests caused false vibration peaks >1 G. As a result, segregation was high after 16 hr of vibration. An electromechanical vibrator was installed that allowed better control and regulation of the G-level. The results of the subsequent tests are shown in Table 12.

Table 11

DATA ON PARTICLES USED IN PACKING TESTS

Type	Core		Coating		Over-all Size Range (μ)	Density (Air Pycnometer) (g/cm ³)
	Type	Size Range (μ)	Type	Average Thickness (μ)		
PyC-coated Particles (large reference size)	ThO ₂	300- 420	BISO	96	500-620	3.79
No. 80 Glass Beads	---	---	---	---	350-420	2.49
PyC-coated Particles (small reference size)	(Th, U)C ₂	150- 250	BISO	104	350-420	2.51

Table 12
DATA FROM SEGREGATION TESTS WITH 1/2-G VIBRATION

Vibration Time (hr)	Magnitude of Sample Deviation from 10 wt-% Coated Fuel Particles ^a		Packing Density (vol-% solids at end of test)
	Average	Maximum	
8	0.020	0.03	74.1
8	0.017	0.03	74.1
16	0.027	0.04	74.1
16	0.025	0.05	74.9
32	0.030	0.07	74.1
32	0.046	0.09	74.1
64	0.023	0.04	74.9

^aThe 30-in. particle column contained 10 wt-% coated particles (Batch 2702-91) and 90 wt-% glass beads (see Table 11).

The data reveal a very low amount of segregation which exhibited a normal, gradual increase with increasing vibration time, with the exception of the 64-hr test. In this test, the segregation decreased by a factor of about two from the 32-hr tests. The reasons for this behavior are not as yet known. One further point of interest involving the 64-hr test is the spatial distribution of the blend, shown in Fig. 23. The apparent cyclic distribution of the coated particles could be due to the presence of vibrational nodes and antinodes in the column of particles. This test (the longest run to date) is the only one in which this type of distribution has been observed.

It should also be noted that, although the average value of the 30 samples is 10.00%, there was only one 10.00% sample. However, there were six samples that deviated only ± 0.01 from the 10.00% average; 10 that deviated ± 0.02 ; 9 that deviated ± 0.03 ; and 4 that deviated ± 0.04 (all positive). This indicates a degree of coated particle saturation in the upper half and coated particle depletion in the lower half of the column. These effects will be investigated in greater detail using blends of reference coated fuel particles. If coated particle blends show the same limit in segregation, then it could be concluded that segregation due to a 0.5-G vibration environment does not cause excessive nonuniformity in the dispersion of fissile and fertile fuel. With the receipt of coated fuel particles in the small reference size, the tests on coated-particle-glass-bead blends have been terminated.

Fig. 23--Distribution of coated particles in 64-hr segregation test

Preliminary measurements of the packing density of blends of reference coated particles were made. A mixture of reference size coated particles (see Table 11) was blended and loaded into a graduated 10-cm³ cylinder. After the particle column was vibrated to a minimum volume, weight and volume measurements were taken. The procedure was repeated for three density measurements for each of five different blends. The results, shown in Table 13, reveal very little variation of solids content (by volume) with varying blend ratio. More detailed measurements of packing density are planned.

Table 13
PRELIMINARY PACKING DENSITIES OF
REFERENCE FUEL PARTICLES

Particle Ratio ^a (Fertile/Fissile)	Packing Density ^b	
	(g/cm ³)	(vol-% solids)
1.0:1	2.02	64.0
1.0:1	2.02	64.0
1.0:1	2.02	64.0
1.5:1	2.10	64.0
1.5:1	2.10	64.0
1.5:1	2.10	64.0
2.0:1	2.16	64.0
2.0:1	2.16	64.0
2.0:1	2.18	64.7
2.5:1	2.22	64.8
2.5:1	2.22	64.8
2.5:1	2.22	64.8
3.0:1	2.25	64.7
3.0:1	2.25	64.7
3.0:1	2.25	64.7

^a—Ratio of volume of solids for
Lot 2702-91 to that for Lot 2702-105
(see Table 11).

^b—Measured in a graduated 10-cm³
cylinder.

Thermophysical Properties. The thermophysical properties of particular interest include thermal expansion and thermal conductivity, since these parameters have a marked influence on the design and operation of the core. It is intended that apparent thermal conductivity and thermal expansion will be measured in helium to 1500°C using realistic particle bed geometries.

The techniques and equipment for thermal conductivity measurements have been developed in the past. During the present quarter, an apparatus for measuring the thermal expansion of a 30-in. column of loose, coated particles relative to that of 780-S graphite was designed and constructed. Temperature capabilities are in excess of 1500°C.

Graphite Development

The objective of the graphite development program is to evaluate and test conventional needle coke graphites for use in fuel blocks. The major effort is concerned with liaison with graphite vendors to procure material and with subsequent evaluation of the strength, purity, and thermal properties of these materials. Drop test studies are being conducted on reference hexagonal fuel blocks to determine the mode and extent of failure.

Graphite Evaluation. Conventional needle coke graphites are currently being evaluated for use as fuel block and reflector graphites. It appears that three of the major graphite vendors can manufacture needle coke graphites to order in diameters of 17 in. or larger that nearly meet HTGR specifications. The following samples have been received and are currently being evaluated:

Grade 9567	4-3/4 in. x 12-7/16 in. x 20 in.
Grade H-327	12-3/4 in. diam x 24 in.
Grade TS-688	
(EGCR Graphite)	20 in. x 20 in. x 20 ft

In addition to these samples, a log of Grade H-328 (17-in. diam x 24-in. long), which is an improved isotropic graphite, has been received. This material, while novel and not considered conventional, is also of interest to the HTGR program.

Five samples cut parallel to extrusion were made from each of two lots of TS-688 (EGCR) graphite that had been previously tested at ORNL. The samples were made to current "dog-bone" requirements and were pulled at a crosshead rate of 0.002 in./min with the best grips available. One set of samples had an average tensile strength of 1280 psi, and four of the five samples broke at the shoulder. The second set averaged

1510 psi, and only one out of the five samples broke at the shoulder. Tensile strengths of greater than 1500 psi were obtained by the ORNL investigators on samples cut from the same piece.

Battelle-Northwest has tested EGCR-type graphite in tension and obtained values in the range of 1800-2000 psi parallel to extrusion. BNW investigations indicate that the tensile value is sensitive to sample shape and that high values always result when cylindrical samples were used.

To further determine the true tensile strength of needle-type graphite, a round-robin testing program has been initiated. Tensile blanks will be cut at General Atomic in a systematic manner from an EGCR log supplied by ORNL, and these samples will be distributed among the three participating sites. Each tester will perform tensile tests using cylindrical samples. The results will then be evaluated by General Atomic. Test blanks are currently being machined for this study.

Coefficient of thermal expansion, electrical resistivity, X-ray, and density determinations will be made spanning a cross section of EGCR graphite that is 18 in. x 18 in. These studies will be made to determine whether a difference in graphiticity exists from outside to inside on a piece of large cross section.

Test Drops of Hexagonal Fuel Blocks. The type and extent of damage suffered by a reference hexagonal fuel block involved in a drop is being investigated. Two blocks of EGCR graphite, 14.2 in. across the flats and 15.6 in. long have been made. Both blocks were loaded with dummy fuel particles (Cu shot, glass beads, and ZrO_2) to a suitable bulk density (2.2 g/cm^3), and the fuel holes were capped with graphite plugs. A vertical frame was constructed to control the direction of fall. A timing device was provided so that, if the frame slowed down the free fall of the block, the equivalent drop height could be calculated. A lifting and release mechanism was also provided, and test drops on each block were performed.

For the first block, drops were made, flat face to flat face, from 1 to 8 ft at 1-ft intervals. The block began to crack after the 6-ft drop. It broke into two pieces on a line between two opposite points after the 8-ft drop (see Fig. 24), and about 10% of the "fuel particles" escaped. The timing device indicated that the guide frame did not slow down any of the drops.

The second block was dropped once from a height of 18 ft. It was also constrained by the guide frame so that it impacted flat face to flat

Fig. 24--Views of broken fuel block subjected to drop testing

face. No cracks or other damage could be observed on this block after it was dropped.

Two tests, especially since the mode of impact was constrained, are not conclusive. The results are encouraging, however, in that only 10% of the particles escaped from the block that split into two pieces after eight drops, and that the block given an 18-ft drop did not break.

Control Materials

Control material studies will be carried out in conjunction with a liaison program with various vendors. The aim of the program will be to determine the feasibility of producing B₄C graphite to a dimensional tolerance and to measure properties of interest.

An effort is being made to determine whether B₄C-graphite compacts can be manufactured to General Atomic fabrication specifications and a dimensional tolerance of $\pm 1/32$ in. without machining. The elimination of machining is desirable to reduce the over-all cost of the compacts, and at least one of the vendors has indicated that the compacts can be manufactured to this tolerance.

Strength measurements on B₄C-graphite samples that were previously irradiated in Capsules 1A1 and 2A2 under the Peach Bottom Program are planned. Modulus of rupture measurements on unirradiated and irradiated samples will be made.

Coated Particle Irradiations

Coated particle irradiation studies on the PSC program are intended to guide the selection of fuel materials that will meet the performance requirements of the Public Service reactor. BISO-coated particles have been selected as the reference fuel design based on their stability under high burnup in high-temperature irradiation environments in previous tests. A series of irradiation experiments is being conducted to test these particles under PSC design conditions with emphasis on evaluating stability under high fast-flux exposures.

Capsule P14. The objectives of the P14 experiment are to evaluate the effects of density and preferred orientation of the isotropic PyC layer of the reference BISO coatings. The density range covered for the isotropic coatings will be 1.90 to 2.15 g/cm³. The range of anisotropy factors will be 1.0 to 1.7. (The buffer layer of the coating will be kept constant on all particles as well as the T/D ratio. *) These coatings will be deposited on fissile particles. BISO coated fertile particles with two densities of isotropic PyC will also be included.

* Ratio of coating thickness to particle diameter.

face. No cracks or other damage could be observed on this block after it was dropped.

Two tests, especially since the mode of impact was constrained, are not conclusive. The results are encouraging, however, in that only 10% of the particles escaped from the block that split into two pieces after eight drops, and that the block given an 18-ft drop did not break.

Control Materials

Control material studies will be carried out in conjunction with a liaison program with various vendors. The aim of the program will be to determine the feasibility of producing B₄C graphite to a dimensional tolerance and to measure properties of interest.

An effort is being made to determine whether B₄C-graphite compacts can be manufactured to General Atomic fabrication specifications and a dimensional tolerance of $\pm 1/32$ in. without machining. The elimination of machining is desirable to reduce the over-all cost of the compacts, and at least one of the vendors has indicated that the compacts can be manufactured to this tolerance.

Strength measurements on B₄C-graphite samples that were previously irradiated in Capsules 1A1 and 2A2 under the Peach Bottom Program are planned. Modulus of rupture measurements on unirradiated and irradiated samples will be made.

Coated Particle Irradiations

Coated particle irradiation studies on the PSC program are intended to guide the selection of fuel materials that will meet the performance requirements of the Public Service reactor. BISO-coated particles have been selected as the reference fuel design based on their stability under high burnup in high-temperature irradiation environments in previous tests. A series of irradiation experiments is being conducted to test these particles under PSC design conditions with emphasis on evaluating stability under high fast-flux exposures.

Capsule P14. The objectives of the P14 experiment are to evaluate the effects of density and preferred orientation of the isotropic PyC layer of the reference BISO coatings. The density range covered for the isotropic coatings will be 1.90 to 2.15 g/cm³. The range of anisotropy factors will be 1.0 to 1.7. (The buffer layer of the coating will be kept constant on all particles as well as the T/D ratio.*) These coatings will be deposited on fissile particles. BISO coated fertile particles with two densities of isotropic PyC will also be included.

* Ratio of coating thickness to particle diameter.

The capsule will have five cells with individual temperature control; four are designed to operate at 1400°C and the fifth at 1000°C. All samples of coated particles will be tested at 1400°C, and three selected samples will also be tested at 1000°C. This lower temperature is being used because the results from the G9 capsule indicate that maximum dimensional effects may occur in the PyC at about 1000°C.

The capsule will be designed for a minimum of three irradiation cycles in the ETR. If it is performing well at the end of the third cycle, it will be left in for as many as six cycles in order to get a higher fast-flux exposure. Fast-flux exposure at the end of three cycles is expected to be about 3.7×10^{21} nvt. Design burnups will be 20% (FIMA) for the fissile particles, and 6% for the fertile particles over a four-cycle irradiation. If the capsule is terminated after three cycles, the burnups will be slightly below design, whereas if it should go a full six cycles, burnups will be somewhat higher than design. The capsule is scheduled to begin irradiation in January 1966 on ETR cycle 78.

HTGR Reference Fuel Tests. HTGR reference coated particles (BISO fissile and fertile particles in a packed bed) are being tested in ORNL Loop 1-15 to approximately 16% burnup (FIMA) over an 8-month period at a maximum fuel temperature of 1400°C.

On July 5, the experiment began irradiation in the ORR with the capsule in a retracted position to check the new coolant purification system. It was fully inserted on July 13, when impurity levels became acceptably low. After insertion, the maximum (center) thermocouple temperature rose to 1480°C, about 80°C above design maximum fuel temperature. This was attributed to design errors in power-density calculations. In addition, there was a 350°C difference in the graphite container temperature between the top (760°C) to the bottom (1110°C). On August 2, the beryllium filler block in an adjacent experiment was replaced with aluminum. This reduced the maximum thermocouple temperature to 1350°C, but the longitudinal temperature difference remained high (315°C). The experiment is scheduled to continue irradiation until March 1966.

Graphite Irradiations

Graphite irradiation experiments on the PSC program are directed toward obtaining more extensive data on conventional needle-coke graphites. The objectives are to determine the effect of temperature upon dimensional and property changes in the range 600° to 1200°C and to relate the results to current theory on irradiation damage. This work covers the design, monitoring, and interpretation of a series of graphite irradiation experiments.

Capsule G11. A series of capsule experiments is being planned to irradiate conventional needle-coke graphite in the temperature range 600° to 1200°C to a maximum exposure of about 9×10^{21} nvt ($E > 0.18$ Mev). The first of these experiments is designated G11. Thermal and mechanical design are under way, and present plans are to insert the capsule in the 113NW position of the ETR.

The graphite samples will consist of small (~ 0.875 -in.) wedge-shaped samples stacked in bundles of twelve. A thermocouple will be placed at the center of each bundle for temperature monitoring.

The bulk of the samples will consist of Grade TS-688 (EGCR) graphite, which is representative of a conventional needle coke that can be made in large cross section. This type of graphite has been studied extensively at Battelle-Northwest in the temperature range 450° to 800°C to neutron doses of about 16×10^{21} nvt ($E > 0.18$ Mev). In addition to the TS-688 material, samples from Grade 9567 and Grade II-327 will be included as a means to check the stability of material manufactured by other vendors. Grade H-328, an isotropic material, will be included to determine its behavior above 800°C .

The samples will be measured before and after irradiation to determine their dimensional stability, changes in the coefficient of thermal expansion (CTE), thermal conductivity, X-ray parameters, and weight.

Cooperative Graphite Irradiations. A cooperative program is being formulated among Battelle-Northwest, General Atomic, and Dragon for the irradiation of graphite in the Dragon Experimental Reactor. It is planned to irradiate isotropic graphites in the range 650° to 1650°C up to a dose of 5×10^{21} nvt (fast). Battelle-Northwest will coordinate the program for the United States, and General Atomic will consult and supply samples.

Samples of two isotropic graphites (H-319 and H-207-85) have been machined and sent to BNW. BNW will carry out further laboratory measurements before shipping the samples to Dragon for irradiation.

Samples of HLM-85 and 780-S graphite, which are being irradiated at Battelle-Northwest for General Atomic, are currently being examined. These experiments were designed to operate at 600° to an exposure of about 5×10^{20} nvt ($E > 0.18$ Mev).

CHEMISTRY

Fission-product Release Studies

Work has continued on the release of the metallic fission-product nuclides of cesium, strontium, cerium, and barium from PyC-coated fuel particles. The purpose of this work is to obtain information for use in evaluating the retention properties of coatings and predicting the release of metallic fission products from PSC fuel blocks. The work involves (1) the measurement of the fractional retention of metallic fission products by coated particles during irradiation tests conducted at reactor temperatures, (2) the determination of release constants by use of postirradiation annealing experiments, and (3) the determination of release constants by analysis of irradiation test data. The release constants determined in this work find important use in calculations to estimate the release of metallic fission products from PSC fuel as described in the section below entitled "Mathematical Treatment of Metallic Fission-product Release from HTGR Fuel."

In postirradiation examination of PyC-coated particles, three different methods are employed: (1) annealing experiments, (2) direct gamma counting, and (3) radiochemical analyses.

In the annealing method the particles are placed in a graphite container and annealed in a King furnace in a helium stream at temperatures in the range 1100° to 1800°C . During the anneal, cesium is collected on a cold finger, and the other metallic nuclides are collected in the graphite container. Temperatures are measured optically, and the uncertainties are believed to range from $\pm 25^{\circ}\text{C}$ at 1100°C to $\pm 50^{\circ}\text{C}$ at 1800°C . After the anneal, the quantities collected on the cold finger and graphite container are determined by gamma spectrometry for Cs^{137} , Ce^{144} , and Ba^{140} and by radiochemistry for Sr^{89} .

The annealing method is used to determine the fraction of metallic fission products retained by the coated particles during irradiation tests. In using the method for this purpose, the particles are annealed for around 200 hr at 1800°C , and the quantities released are measured. Previous experience has shown that the release of cesium, strontium, barium, and cerium is complete as a result of this treatment.

The annealing method is also used to measure release constants, in which case the particles are annealed at selected temperatures in the range 1100° to 1800°C . The cold finger and graphite container are analyzed at intervals of about 24 hr to determine the fractional release of the metallic fission products as a function of time. The resulting fractional release values are based on the quantities of the metallic fission products contained in the particles at the start of the annealing experiment.

In the direct gamma-counting method, quantities of Zr^{95} , Cs^{137} , and Ce^{144} in particles or collector samples are determined using a germanium-crystal detector. (In short-cooled irradiated particles, where the Ba^{140} (La^{140}) activity is relatively high, this nuclide can also be determined by the direct gamma-counting method.) By use of the germanium detector, it is possible to resolve the gamma peaks for Cs^{137} , Ce^{144} , Zr^{95} , and Nb^{95} . This method has high potential; it is straightforward, simple, and is believed to be more accurate than the other two methods. The major difficulty is the low counting efficiency of the small germanium crystal now in use; the method is therefore only applicable when activities are relatively high. This method is used in the analysis of collector samples from annealing experiments and also in the direct determination of the quantities of metallic fission products contained in irradiated particles.

In the radiochemical method for determining quantities of metallic fission products, samples are dissolved, chemical separations are performed, and the fission products are determined by beta and gamma spectrometry. The radiochemical method is necessary for the determination of the strontium nuclides Sr^{89} and Sr^{90} , which emit no gammas. The nuclide Sr^{89} (rather than Sr^{90}) is normally determined in this work because of its higher activity relative to the activity of Sr^{90} (one expects the release behavior of the two isotopes to be the same). The radiochemical method is used in the analysis of collector samples from annealing experiments and also in the analysis of coated particles.

The particles used in these measurements are first gamma counted using the germanium crystal detector to determine the Zr^{95} activity (zirconium is essentially completely retained under the conditions of irradiation tests), and the result is used to calculate the number of fissions. The amounts of the other fission products produced in the irradiation test are calculated from known fission yields and decay times.

Table 14 presents the characteristics and the irradiation conditions of four coated particle samples measured during the period. Measurements of the fractions of Cs^{137} , Ce^{144} , Ba^{140} , and Sr^{89} retained by Samples 1, 2, and 3 during the irradiation tests were performed using the three different methods described above (annealing experiments conducted at 1800°C , direct gamma-counting, and radiochemical analyses), and Table 15 gives the results.

Comparison of the data in Table 15 shows that the fractional-retention values obtained by the three methods vary somewhat; in particular, the values obtained by the radiochemical method are appreciably lower than the values obtained by the other two methods. The low values of the radiochemical data may possibly be due to the incomplete dissolution of the particle coatings.

Table 14

SAMPLE CHARACTERISTICS AND IRRADIATION CONDITIONS

Sample	Particle			Coating		Irradiation Condition			
	Type	Size (μ)	Th:O Ratio	Type	Total Thickness (μ)	Burnup (% FIMA)	Temp ($^{\circ}$ C)		
							Nom	Min	Max
1	UC ₂	150-250	---	BISO (2000) $\rho = 2.02$	102	20	1300	1170	1460
2	(Th,U)C ₂	250-350	3:1	BISO (2000) $\rho = 1.97$	102	5	1500	1270	1550
3	UC ₂	150-250	---	Triplex	103	20	1300	1170	1460
4	(Th,U)O ₂	300-420	1:1	BISO (1600) $\rho = \sim 1.8$	170	11.6	1450	1385	1520

Table 15
METALLIC FISSION-PRODUCT FRACTIONS RETAINED BY COATED
CARBIDE PARTICLES DURING IRRADIATION
(Determined by Three Methods)

Sample	Method of Analysis	Cs ¹³⁷	Ce ¹⁴⁴	Ba ¹⁴⁰	Sr ⁸⁹
1	1800°C Annealing Expt.	0.84	0.54	0.14	0.03 ^a
	Direct Gamma Counting ^b	0.95	0.78	c	d
	Direct Gamma Counting	0.81	0.56	c	d
	Direct Gamma Counting	0.90	0.71	c	d
	Radiochemical Analysis ^e	0.76	0.48	0.20	0.02
	Radiochemical Analysis	0.78	0.41	0.44	0.05
2	Direct Gamma Counting	0.87	0.59	c	d
	Direct Gamma Counting	0.90	0.63	c	d
	Direct Gamma Counting	0.88	0.63	c	d
	Radiochemical Analysis	0.71	0.38	0.24	0.03
	Radiochemical Analysis	0.77	0.34	0.20	0.02
3	Direct Gamma Counting	0.93	0.43	c	d
	Direct Gamma Counting	0.87	0.43	c	d
	Direct Gamma Counting	0.84	0.48	c	d
	Radiochemical Analysis	0.59	0.46	0.16	0.02
	Radiochemical Analysis	0.66	0.50	0.18	0.01

^aBy radiochemical analysis of graphite collector.

^bBy use of lithium-drifted germanium detector.

^cBa ¹⁴⁰ (La ¹⁴⁰) activity too low for direct gamma counting.

^dSr ⁸⁹ is a beta-emitter and cannot be determined by gamma counting.

^eRadiochemical analysis results are based on Zr ⁹⁵ activity as determined by lithium-drifted germanium detector.

The radiochemical data were not consistently low, however. The particles that were submitted for radiochemical analyses were first gamma counted on the germanium crystal detector to determine the Zr^{95} activity, and the values obtained agree within experimental error with the Zr^{95} activities determined in the radiochemical analyses.

As shown in Table 15, there is some variation in the fractional retention data determined by direct gamma counting. Several of the specimens were recounted, and the resulting data agree within $\pm 4\%$ with the data in Table 15. It therefore appears that the variation in data obtained by direct gamma counting may be due to variations in particle characteristics.

For the coated particles from Sample 1, average values of the fraction retained during the irradiations as determined by direct gamma counting (see Table 15) are 0.89 for Cs^{137} and 0.68 for Ce^{144} . Based on the radiochemistry data, the value of the fraction of Sr^{89} retained averages about 0.03. Based on data obtained by the annealing method, the fractional retention value for Ba^{140} is about 14%. These are taken to be the best values to use for the calculation of release constants.

The direct gamma-counting technique was used to determine the fraction of Cs^{137} retained in PyC-coated oxide particles (Sample 4). Two separate counts using different particles yielded the fractional retention values 0.72 and 0.70 -- the average is 0.71. This is considered to be more accurate than the value 0.38 determined by radiochemical analysis and reported in the HTGR Base Program Quarterly Report for the Period Ending August 31, 1965 (GA-6671). The rate of accumulation of fission products in coated particles during an irradiation test may be described by

$$\frac{dN}{dt} = B - RN - \lambda N,$$

where B is the production rate (product of fission rate and yield), R is the release constant,* λ is the decay constant, and N is the number of nuclei. Production rate and temperature are assumed constant. This equation integrates to

$$N = \frac{B}{(R + \lambda)} (1 - e^{-(R+\lambda)t}) .$$

* A discussion of R and empirical and theoretical equations describing R are presented in a later section of this report.

The total number of nuclei produced is Bt , and the fraction retained, F_{ret} , is N/Bt . Combining these relationships yields (in the case where $\lambda \ll R$)

$$F_{\text{ret}} \approx \frac{(1 - e^{-Rt})}{Rt}, \quad (1)$$

which describes the fractions retained in terms of R and t .

Table 16 gives release constants derived from irradiation test data using the above equation. The input data were the fractional retention values and irradiation times given above.

Table 16
RELEASE CONSTANTS DERIVED FROM IRRADIATION TEST DATA

Nominal Irradiation Temp ($^{\circ}\text{C}$)	Release Constant, R (fraction/hr)		
	Cs^{137}	Ce^{144}	Sr^{89}
1450	1.3×10^{-3}	---	---
1300	5.6×10^{-4}	1.9×10^{-3}	7.7×10^{-2}

Rates of release of the metallic fission products Cs^{137} , Sr^{89} , and Ce^{144} from irradiated particles have been determined in postirradiation annealing experiments.

Measurements were made on particles from Sample 1. Release fractions for Cs^{137} , Sr^{89} , and Ce^{144} were obtained as a function of annealing time at four different temperatures in the range 1100° to 1800°C . The results are given in Figs. 25 through 29.

It is of interest that in the annealing experiments, the cesium is always found on the cold finger, even in experiments conducted at 1100°C --no cesium is detectable in the graphite. This indicates that cesium has relatively high diffusivity in graphite and high volatility from graphite surfaces.

On the basis of Fick's law of diffusion one would expect, after a steady state is reached, the release of fission products from coated particles in an annealing experiment to be approximated by the relationship $F_{\text{ret}} = e^{-Rt}$, where F_{ret} is the fraction retained, R is the release constant, and t is the annealing time. The release data for Cs^{137} at 1800°C (see Fig. 26) follow this relationship nicely, and the data for Ce^{144} and Sr^{89} at 1800°C are approximately exponential. In general, however,

Fig. 25--Fractional release of Cs^{137} at 1100° and 1300°C ; results of annealing experiments using PyC-coated particles from Sample 1

Fig. 26--Fractional release of Cs^{137} at 1500° and 1800°C;
results of annealing experiments using
PyC-coated particles from Sample 1

Fig. 27--Fractional release of Ce^{144} at 1100°C and 1300°C; results of annealing experiments using PyC-coated particles from Sample 1

Fig. 28--Fractional release of Ce^{144} at 1500° and 1800°C; results of annealing experiments using PyC-coated particles from Sample 1

Fig. 29--Fractional release of Sr^{89} at various temperatures; results of annealing experiments using PyC-coated particles from Sample 1

the data at lower temperatures deviate from the exponential relationship. As shown in Fig. 26, Cs^{137} showed an unusual, delayed type of release behavior. The deviation from exponential behavior may be due to reasons such as nonuniform initial fission product concentrations, a transient (nonsteady) state of diffusion, existence of more than one diffusion mechanism, trapping mechanisms, and existence of layered (nonhomogeneous) sections in the particles.

The initial fission-product release behavior of particles in an annealing experiment is of primary interest, because the initial release behavior in the annealing experiments should represent the release behavior of the particles at the end of the irradiation test. Therefore, the curves in Figs. 25 through 29 were so drawn that they reflect the initial release behavior of the particles. The curves were also drawn to conform to exponential behavior.

Release constants (R) derived from the data are given in Table 17. The release constants represent the initial release rates, with the exception of the value for Cs at 1300°C , and they correspond to the initial slopes of the curves. In the case of Cs^{137} at 1300°C , the slope of the data obtained after the delay period was used. In deriving release constants from the data at higher temperatures, where the initial release rates were high and initial slopes difficult to measure, the exponential relationship given above was applied to the curve.

Table 17
EXPERIMENTAL RELEASE CONSTANTS^a

Temp. ^b ($^{\circ}\text{C}$)	Release Constant, (fraction/hr)		
	Cs^{137}	Ce^{144}	Sr^{89}
1100	3.0×10^{-6}	2.4×10^{-6}	2.2×10^{-3}
1300	8.3×10^{-5}	7.5×10^{-4}	5.0×10^{-3}
1500	4.5×10^{-3}	1.0×10^{-2}	3.2×10^{-2}
1800	3.0×10^{-2}	0.17	0.13

^a—For metallic fission products from coated carbide particles from Sample 1.

^b—Temperature uncertainties range from $\pm 25^{\circ}$ at 1100°C to $\pm 50^{\circ}$ at 1800°C

In Fig. 30 the release constants for Cs^{137} , Ce^{144} , and Sr^{89} are plotted against reciprocal temperature. In drawing curves through the data, the points at 1300°C and above were given more weight than the points at 1100°C . From the slopes of the curves, one obtains the apparent activation energies 74 kcal/mole for Cs^{137} , 70 kcal/mole for Ce^{144} , and 41 kcal/mole for Sr^{89} . The values for Cs^{137} and Ce^{144} appear reasonable for the activation energy of diffusion of these two isotopes in pyrocarbon, whereas the value for Sr^{89} is lower than expected.

The release constants for Cs^{137} determined in postirradiation annealing experiments and reported in Table 6.11, HTGR Base Program Quarterly Progress Report for the Period Ending August 31, 1965 (GA-6671) need to be revised by the factor $0.38/0.71$ as a result of the new determination for the fractional retention value (see above). The revised release constants are 3.2×10^{-6} f/hr at 1185°C , 1.9×10^{-5} f/hr at 1275°C , 3.4×10^{-4} f/hr at 1485°C , and 8.5×10^{-3} f/hr at 1800°C .

Figure 31 shows how release constants derived from irradiation test data (given in Table 13) fit to curves representing release constants derived from postirradiation annealing experiments. The curves for the Sample 1 particles were taken from Fig. 30. The Cs^{137} curve for the Sample 4 particles represents the release constants given above.

It is significant that points for Cs^{137} and Ce^{144} fit the curves within the temperature uncertainty of the data. The point for Sr^{89} fall considerably above the Sr^{89} curve, outside the realm of temperature uncertainty. This may reflect greater uncertainty in the Sr^{89} data, which were determined by radiochemical analyses.

The release data can also be compared as in Table 18, which gives experimentally determined fractional retention values and values calculated from postirradiation annealing data. The calculated values were obtained using Eq. (1) above and release constants taken from the curves in Fig. 31. The agreement in these values is satisfactory considering the uncertainty of the data, excepting perhaps the case of Sr^{89} .

The results of these experiments indicate the following:

1. The fractions of metallic fission products retained in coated carbide particles (Sample 1) during the irradiation test were about 0.89 for Cs^{137} , 0.68 for Ce^{144} , 0.03 for Sr^{89} , and 0.14 for Ba^{140} .
2. The fraction of Cs^{137} retained in coated oxide particles (Sample 4) during the irradiation test was about 0.71.

Fig. 30--Arrhenius plot of release constants (fraction/hr) for the release of Cs^{137} , Ce^{144} , and Sr^{89} from BISO-coated carbide particles from Sample 1

Fig. 31--Comparison of release constants; curves represent release constants obtained from postirradiation experiments; points represent constants obtained from irradiation test data

Table 18
COMPARISON OF EXPERIMENTAL AND
CALCULATED FRACTIONAL RETENTION VALUES

Sample	Nuclide	Irradiation Temp (°C)	Fraction Retained	
			Experimental ^a	Calculated ^b
4	Cs ¹³⁷	1450±50	0.71	0.95
1	Cs ¹³⁷	1300±100	0.89	0.98
1	Ce ¹⁴⁴	1300±100	0.68	0.86
1	Sr ⁸⁹	1300±100	0.03	0.32

^a—Fractional retention values determined by direct gamma counting of particles.

^b—Calculated using release constants determined in post-irradiation release experiments.

3. Release constants determined in postirradiation experiments can be used to predict the fraction of metallic fission products retained in coated particles under irradiation at high temperature.
4. The coated carbide particles of Sample 1 (100-μ BISO coatings, coating density = 2.02 g/cm³) showed slightly higher release rates than the coated oxide particles of Sample 4 (170-μ BISO coatings, coating density = ~1.8 g/cm³), indicating that the higher coating density of the carbide particles did not compensate for the lower coating thickness.

Mathematical Treatment of Metallic Fission-product Release from HTGR Fuel Blocks

FREVAP Code. The FREVAP code for estimating the release of longer-lived metallic fission products from HTGR fuel blocks has been developed to take into account the combined effects of the retention of metallic fission products by fuel particles and the rather strong sorption of these fission products by the graphite of the fuel blocks. Release calculations are made on the basis that the loss of fission-product nuclides such as of strontium, cesium, and barium is determined by their evaporation from the graphite surfaces and their transpiration induced by the flowing helium coolant. The code is devised so that changes of fission rate (fuel block power), fuel temperature, and graphite temperature may be incorporated into the calculation. Temperature is quite important in determining release because, in general, both release from fuel particles and loss by evaporation (transpiration) vary exponentially with the reciprocal of the absolute temperature.

The physical and logical basis for FREVAP is perhaps best indicated by giving the following equations upon which the program is based:

Fission product production is given by

$$\frac{dP_i(t)}{dt} = B_{ij} - \lambda_i P_i(t), \quad (2)$$

where t = Time (days),

$P_i(t)$ = The total quantity of a given fission-product isotope, i , of the fission-product element being considered (i. e., Sr, Cs or Ba) at time t (moles),

B_{ij} = Production rate of the isotope, i (the subscript j indicates the time interval to which B_{ij} applies), (moles/day), and

λ_i = Radioactive decay constant, (days^{-1}).

The equation for the buildup of the average concentration of a given fission product nuclide in the graphite of the fuel element is given by

$$\frac{A_G}{10^6} \frac{d\bar{C}_i(t)}{dt} = S_{ij} - \left(\frac{A_G}{10^6} \right) \lambda_i \bar{C}_i(t) - \left(\frac{\dot{n}_{\text{He}}}{p_{\text{He}}} \right) \zeta p_b X_i, \quad (3)$$

where A_G = The total quantity of graphite in active zone of the fuel block*
(m^2 BET surface area).

$C_i(t)$ = Average concentration of isotope i in the graphite (μ moles/ m^2
BET internal surface area).

t = Time (days).

S_{ij} = Release rate of isotope i from the fuel particles to the graphite
(j indicates the time interval to which S_{ij} applies).

$\dot{n}_{He}$ = Flow rate of He coolant (moles/day).

P_{He} = Pressure of He coolant (atm).

ζ = The rate of the partial pressure of the given fission-product
element in the coolant stream (in the turbulent region beyond
the boundary layer) to the partial pressure at the graphite-
coolant interface.

p_b = Partial pressure of the given fission-product element at the
graphite-coolant interface. This is assumed to be in equi-
librium with the fission product sorbed in the graphite at the
boundary (graphite-coolant interface).

X_i = Mole fraction of nuclide i of a given fission-product element.

Now,

$$X_i = \frac{(C_i)_b}{\sum_i (C_i)_b} = \frac{(C_i)_b}{C_b}, \quad (4)$$

where $(C_i)_b$ = Concentration of isotope i of a given fission-product element
at the boundary, and

$\sum_i (C_i)_b$ = Concentration of given fission-product element (sum of isotopes)
at the boundary $\equiv C_b$.

* A_G is the product of the mass of the graphite (g) and the specific
internal surface area (m^2/g) determined by the BET method. It is to be
noted that m^2 of BET internal surface area has been chosen as the "rational"
unit in which to express the sorption behavior of graphite (and activated
carbon). Accordingly, m^2 of BET area is used in place of mass (g) to
express the quantity of graphite and the concentrations of fission-product
isotopes in graphite.

From experimental studies of the sorption of metallic fission-product elements, the equilibrium pressure, p , of a given element at concentration C in graphite is given by

$$p = e^{\nu} C^{\mu} , \quad (5)$$

where μ and ν are empirical coefficients (μ is usually of the order of 2 to 6) that are functions of temperature. They are expressed by the equations

$$\nu = a + b \left(\frac{10^3}{T_G} \right) ,$$

$$\mu = c + d \left(\frac{10^3}{T_G} \right) ,$$

with T the temperature of the graphite ($^{\circ}\text{K}$), and a, b, c , and d being determined from experimental data.

Accordingly, the partial pressure of a given fission-product element at the graphite boundary (graphite-coolant interface) is given by

$$p_b = e^{\nu_j} C_b^{\mu_j} = e^{\nu_j} \left(\sum (C_i)_b \right)^{\mu_j} . \quad (6)$$

(As before, j indicates a specific time interval and this is characterized by a unique graphite temperature at the boundary, T_{G_j} .) Thus,

$$\nu_j = a + b \left(\frac{10^3}{T_{G_j}} \right) , \quad (7)$$

$$\mu_j = c + d \left(\frac{10^3}{T_{G_j}} \right) . \quad (8)$$

The concentration of the fission-product nuclide at the boundary will not, in general, be the same as the average concentration and is determined

by the diffusion and decay behavior of the individual isotopes; thus, we define

$$\eta_i \equiv \frac{(C_i)_b}{\bar{C}_i} = \text{ratio of concentration of a given isotope } i \text{ at the boundary to the average concentration.} \quad (9)$$

Thus, substituting (9) into Eq. (6) and the modified Eq. (6) into Eq. (3), we have the basic equation (set) for FREVAP,

$$\frac{A_G}{10^6} \left(\frac{d\bar{C}_i}{dt} + \lambda_i \bar{C}_i \right) = S_{ij} - \left[\frac{\dot{n}_{\text{He}}}{p_{\text{He}}} \beta e^{\nu_j} \right]^{\mu_j - 1} \eta_i \bar{C}_i \quad (10)$$

The first term on the right hand side of Eq. (10) is the source term and is equal to the rate of release of a given isotope i (in the time interval j) and is a function of time t . The second term on the right is the loss term (to the coolant) and represents the moles per day of a given fission-product isotope swept away by the coolant stream (i. e., loss by transpiration).

In the case of steady-state release, in which decay of isotope i is negligible in the time period required for diffusion from a coated fuel particle kernel and for diffusion through the coating, release from fuel particles is simply related to a release constant R and the quantity of fission product in the fuel particles.

In this case,

$$\frac{dN_i(t)}{dt} = B_{ij} - \lambda_i N_i(t) - R_j N_i(t), \quad (11)$$

where $N_i(t)$ = The quantity of isotope i in the fuel particles at time t (moles), and

R_j = The release constant for the fuel temperature of time interval j (day^{-1}).

R is determined empirically from in-pile release data and postirradiation annealing release experiments that serve to evaluate the constants R_0 and E_a ; thus, R is given by

$$\ln R = \ln R_0 - \frac{E_a}{1.987} \left(\frac{10^3}{T_F} \right), \quad (12)$$

where T_F = Fuel temperature ($^{\circ}\text{K}$),

R_0 = Empirical release constant at $T_F = \infty$ (day^{-1}), and

E_a = Apparent activation energy for release (kcal/mole).

The above constants depend on the nature of the coating on the fuel particles of interest. They are selected to fit the experimentally observed release behavior of coated particle fuels.

In terms of fundamental quantities for spherical fuel particles,

$$R = \frac{D}{\phi} \frac{4\pi(r_1 r_2)}{\frac{4\pi}{3} r_1^3 (r_2 - r_1)} = \frac{D}{\phi} \frac{3r_2}{r_1^2 (r_2 - r_1)}, \quad (13)$$

where D = Diffusion coefficient of fission product in coating (cm^2/day),

$\phi = \frac{\text{conc. of fission product in kernel}}{\text{conc. of fission product in coating}}$ (for equilibrium at a given temperature),

r_1 = Inner radius of the coating, and

r_2 = Outer radius of the coating.

From the above, it is seen that the activation energy for release, E_a , will depend on the energy of activation for diffusion and the temperature dependence of ϕ (the change in enthalpy for the fission product going from "solution" in the kernel to "solution" in the coating).

The source term S_{ij} is then given by the rate of fission-product release from fuel particles,

$$S_{ij} = R_j N_i(t), \quad (14)$$

where specifically

$$\ln R_o = \ln R_o - \frac{E_a}{1.987} \left(\frac{10^3}{T_{F_j}} \right).$$

The integrated fractional release, F_i , of a given fission-product isotope to the coolant (F_i = isotope i outside of fuel element/isotope inside fuel element, at any given time t) is

$$F_i(t) = \frac{P_i(t) - N_i(t) - \frac{A_G}{10^6} C_i(t)}{P_i(t)}. \quad (15)$$

However, when the fractional release is a small fraction of $P_i(t)$ ($\lesssim 10^{-3}$), it cannot be calculated accurately. In this case it is better to calculate the integrated fractional release as follows:

$$F_i(t) = \frac{\dot{n}_{He}}{P_{He} P_i(t)} \left\{ \zeta \sum_{j=1}^{n-1} e^{v_j} \int_{t_{j-1}}^{t_j} \left[\sum_i \eta_i C_i(\tau) \right]^{\mu_j-1} \eta_i C_i(\tau) e^{-\lambda_i(t-\tau)} d\tau \right. \\ \left. + e^{v_n} \int_{t_{n-1}}^t \left[\sum_i \eta_i C_i(\tau) \right]^{\mu_n-1} \eta_i C_i(\tau) e^{-\lambda_i(t-\tau)} d\tau \right\}. \quad (16)$$

Here, as before, j specifies a given time interval (between t_{j-1} and t_j), and n is the total number of intervals involved up to time, t (with the n^{th} interval, in general, not completed).

ζ is calculated from the following equation, which is based on the equality of fission-product diffusion across the laminar boundary layer to

the transpiration of the fission product by the turbulent helium coolant stream. Thus, transport across boundary layer = transport by helium stream,

$$S_c \frac{D_{12}}{R_g T_{He}} \frac{p_b(1-\zeta)}{\beta} = \frac{\dot{n}_{He}}{p_{He}} \zeta p_b, \quad (17)$$

where S_c = Area of graphite-coolant interface (cm^2),

D_{12} = Mutual diffusion coefficient of a given fission product and helium,

$D_{12} = (D_{12}^0/p_{He}) (T_{He}/273)^{1.75}$ (cm^2/day),

D_{12}^0 = Mutual diffusion coefficient of fission product and He at 1 atm and 0°C (cm^2/day),

T_{He} = Average temperature of coolant in boundary layer ($^\circ\text{K}$),

R_g = Gas constant ($\text{cm}^3\text{-atm/mole-}^\circ\text{K}$), and

β = Laminar boundary layer thickness, a function of Reynolds number (cm).

Other quantities are as defined above. A typical value for ζ is 0.7.

η_1 may be determined empirically from concentration profiles of fission-product isotopes in the fuel element sleeve or calculated from solutions of Fick's law for the diffusion of fission products in graphite which theoretical concentration profiles on the basis of empirically determined diffusion coefficients. The latter is complicated by the fact that diffusion is occurring in a medium (graphite) with a pronounced temperature gradient. A proper calculation must take into account the effect of the temperature gradient on concentration distribution and the effect of temperature gradient as a driving force for the transport of a given fission-product nuclide. Work is being carried out on the theoretical treatment, and some use is being made of observed concentration profiles for the estimation of η_1 .

The FREVAP-1 code has been developed in accordance with the above and utilizes a FORTRAN subroutine for the solution of differential Eq. (10). The integral release fraction is best calculated from Eq. (16). This is given in the output as INT F(I). Modifications of FREVAP are being developed to permit calculations involving more than one metallic fission-product sorbing material, such as graphite and activated carbon, at different temperatures.

Early in this quarter, calculations of cesium, strontium and barium fractional release were made prior to the development of the FREVAP code to calculate concentrations and release fractions of individual isotopes. In this case the concentrations and releases of stable isotopes and long-lived isotopes such as Cs^{137} (30 y) and Sr^{90} (28 y) are lumped together. The results of these preliminary calculations are given in Figs. 32, 33, and 34. It is to be emphasized that these releases are for long-lived or stable nuclides. The fractional release of Ba^{140} is expected to be 10^{-3} to 10^{-4} of the stable nuclide (Ba^{138}) release due to its decay upon diffusion through graphite of the fuel element prior to its evaporation. The input for the calculations were:

$$A_G = 1.90 \times 10^8 \text{ (m}^2 \text{ BET)},$$

$$\frac{\dot{n}_{\text{He}}}{p_{\text{He}}} = 3.30 \times 10^8 \frac{\text{moles}}{\text{atm. day}},$$

$$\zeta = 1.$$

For Cesium:

$$\begin{aligned} \ln R_O &= 15.54 & E_a &= 75(\text{kcal/mole}) \\ a &= 31.3, \quad b = -62.2, \quad c = -13.4, \quad d = 22.0 \\ \lambda_1 &= 0 & \lambda_2 &= 6.33 \times 10^{-5} \text{ day}^{-1} \\ \eta_1 &= 1 & \eta_2 &= 1 \end{aligned}$$

For Strontium:

$$\begin{aligned} \ln R_O &= 15.00 & E_a &= 65(\text{kcal/mole}) \\ a &= 13.81, \quad b = -44.9, \quad c = -1.42, \quad d = 6.73 \\ \lambda_1 &= 0 & \lambda_2 &= 6.77 \times 10^{-5} \text{ day}^{-1} \\ \eta_1 &= 1 & \eta_2 &= 1 \end{aligned}$$

For Barium:

$$\begin{aligned} \ln R_O &= 15.00 & E_a &= 65(\text{kcal/mole}) \\ a &= 7.469, \quad b = -35.08, \quad c = 1.458, \quad d = 2.253 \\ \lambda_1 &= 0 & \eta_1 &= 1 \end{aligned}$$

Fig. 32--FREVP estimation of fractional release of Ce^{137} from fuel block

Fig. 33--FREVAP estimation of fractional release of Sr^{90} from fuel block

Fig. 34--FREVAP estimation of fractional release of Ba^{138} from fuel block

The input temperatures and production rates (B_{ij}) for the time intervals t_{j-1} to t_j are given in Table 19.

Table 19
INPUT TEMPERATURES AND PRODUCTION RATES

j	Time Interval (days)	T_{Fj} ($^{\circ}\text{C}$)	T_{Gj} ($^{\circ}\text{C}$)	Production Rate (Moles/day)				
				Cs		Sr		Ba
				B_{1j} 133+135	B_{2j} 137	B_{1j} 88	B_{2j} 90	B_{1j} 138
1	0-365	1240	1145	0.6242	0.3121	0.2140	0.3013	0.3016
2	365-730	1220	1130	0.6242	0.3121	0.2140	0.3010	0.3016
3	730-1095	1140	1060	0.5393	0.2697	0.1849	0.2603	0.2606
4	1095-1460	1010	940	0.4585	0.2293	0.1572	0.2213	0.2216
5	1460-1825	945	875	0.3818	0.1909	0.1309	0.1843	0.1845
6	1825-2190	920	850	0.3070	0.1573	0.1053	0.1482	0.1484

The R values (R_0 and E_a) used in the calculations were based on postirradiation anneal release data from Sample 4 fuel particles and particularly in the case of cesium were somewhat optimistic in regards to the cesium retention capability of the fuel particle. Figures 35 and 36 give, respectively, (1) a comparison of the calculated release fractions for cesium based on R values corresponding to $\ln R_0 = 15.54$ and $E_a = 75$ kcal/mole to release fractions based on $\ln R_0 = 17.49$ and $E_a = 74$ kcal/mole from Sample 1 fuel particle release data obtained subsequently, and (2) the concentrations obtained in the two calculations.

Figures 37, 38, and 39 give the results of calculations for cesium isotopes in two groups: (1) stable $\text{Cs}^{133} + 2 \times 10^6\text{-y } \text{Cs}^{135}$ and (2) 30-y Cs^{137} . Included in the figures are integral release fraction, concentrations, and moles of isotopes retained in the fuel particles as a function of time. The input listed above (with $\ln R_0 = 15.54$ and $E_a = 75$ kcal/mole) was used. The release fraction results (Fig. 37) show there is little difference in the calculated release $\text{Cs}^{133} + \text{Cs}^{135}$ and Cs^{137} .

Sorption and Diffusion of Fission Products

Vaporization of Metallic Fission Products from Graphite.

Measurements are continuing on the vapor pressures of fission-product metals, such as cesium, strontium, and barium, sorbed on graphite. The main purpose of this work is to obtain basic data for use in calculations to predict the release of these metals from PSC fuel blocks.

The vaporization behavior of the barium-HLM-85 graphite system

Fig. 35--FREVAP calculation of fractional release of Ce^{137}

Fig. 36--FREVAP calculation of Ce in fuel block graphite

Fig. 37--Integral release fraction of Ce isotopes as a function of time

Fig. 38--Concentration of Ce isotopes as a function of time

Fig. 39--Ce isotopes retained in fuel particles as a function of time

has been investigated using a mass-spectrometric-Knudsen-cell technique. This work was done to check previous results that indicate the existence of a transition concentration at about 5 mg Ba/g C, above which the vapor pressure of barium roughly obeys Henry's law and below which the vapor pressure is strongly concentration dependent (see GA-6418).^{*} A radio-tracer-Knudsen-cell technique was employed in the previous studies.

In addition to this work on the barium-HLM-85-graphite system, studies using the Knudsen-cell technique are under way to investigate the vaporization behavior of the cesium-EGCR-graphite and strontium-EGCR-graphite systems.

In a typical experiment, 0.5 g of minus-200-mesh graphite loaded with 5 mg of barium (as barium nitrate) was placed in a tantalum Knudsen cell. The cell was installed in a mass spectrometer and heated rapidly to about 1000°K, and then slowly, over a period of about 2 hr, to about 1800°K to allow time to convert the barium nitrate to adsorbed barium. The barium ion signal was monitored during heating.

At temperatures around 1800°K, the cell barium content was slowly decreased by the effusion process. Periodically during cell depletion, measurements were made to determine the temperature dependence of the barium ion signal to obtain heats of vaporization.

Ion signals were converted to vapor pressures through use of the Knudsen equation and knowledge of the orifice size and the amount of barium effused from the cell.

Figure 40 is a log-log plot of barium concentrations against barium cell pressures. It will be noted that the pressure-concentration data can be fit fairly well to a straight line and that the slope of the line differs considerable from unity. This behavior is good evidence of Freundlich-type adsorption. (A slope of unity for a log-log plot of concentration versus pressure indicates Henry's law behavior.) The data in Fig. 40 were obtained with a single loading of the Knudsen cell. The data agree with vapor pressure data from another experiment using a different sample.

Another requirement of Freundlich behavior is that a plot of the heat of vaporization against the logarithm of the concentration be linear. This plot is shown in Fig. 41. All the points except one were derived from a single experiment. The data in Fig. 41 fit a straight line within the experimental uncertainty. At low concentrations, the data appear to fall below the line. This corresponds to a normal mode of deviation from Freundlich behavior.

^{*}TARGET, A Program for a 1000-Mw(e) High-temperature Gas-cooled Reactor, Quarterly Progress Report for the Period Ending May 31, 1965, General Atomic Report GA-6418, June 30, 1965.

Fig. 40--Vapor pressure of Ba adsorbed on HLM-85 graphite as a function of concentration at 1829°K

Fig. 41--Concentration dependence of heat of vaporization of Ba adsorbed on HLM-85 graphite

Based on the lines in Figs. 40 and 41, the Freundlich isotherms for barium in HLM-85 graphite are described by

$$\log P/0.66 = (-1.252 + 6534/T) \log C/0.30, \quad (21)$$

where P is the barium pressure in atm, C is the barium concentration in g Ba/g C, and T is the temperature in $^{\circ}\text{K}$. Isotherms calculated using this equation at 1000° , 1500° , 2000° , and 2500°K are shown in Fig. 42. The outlined area in Fig. 42 shows the temperature, pressure, and concentration region studied in this work.

The general equation for describing Freundlich pressure-concentration data can be written as

$$\log P/P_o = (A + B/T) \log C/C_o \quad (22)$$

where P_o , C_o , A , and B are constants. The isotherms representing this equation converge at C_o ; however, contrary to popular belief, C_o does not represent the monolayer concentration. To obtain a set of lines which converge at the "Freundlich monolayer concentration", one must use activities of the adsorbed species, not vapor pressures. The dependence of activity, a , on concentration is obtained by subtracting the vapor pressure equation for the pure substance, $\log P' = A' + B'/T$, from Eq.(22). This results in

$$\log a/a_o = (A + B/T) \log C/C_m, \quad (23)$$

where C_m is the "Freundlich monolayer concentration" and A and B are the same constants presented in Eq.(22). Ideally, a_o is unity; that is, the vapor pressure at the monolayer equals the vapor pressure of the pure substance.

Application of the thermodynamic relation $d\ln P/dT = \Delta H_2/RT^2$ to Eq. (22) yields

$$-4.576 B \log \frac{C}{C_o} = \Delta H_v,$$

where ΔH_v is the heat of vaporization. In this expression, C is the monolayer concentration when ΔH_v is the heat of vaporization at the monolayer concentration. Using the values for B and C_o given in Eq.(21) and 36 kcal/mole for the heat of vaporization of liquid barium (as given by Stull and Sinke *), one obtains 0.018 g Ba/g C for the monolayer concentration. This is a very reasonable value considering that one would estimate the monolayer

*D. R. Stull and G. C. Sinke, Thermodynamic Properties of the Elements, American Chemical Society, Washington, D. C., 1956.

Fig. 42--Calculated Ba vapor pressure isotherms as a function of Ba concentration showing region of study

concentration to be about 0.011 g Ba/g C on the basis of simple cubic packing, $10\text{m}^2/\text{g}$ for the BET surface area of graphite, and $4.5 \text{ \AA}$ for the atomic diameter of barium. The difficulty with this interpretation, in which liquid barium is assumed to be the adsorbed phase at the monolayer concentration, is that the vapor pressure of adsorbed barium (at the monolayer concentration) is about three orders of magnitude below the vapor pressure of liquid barium.

It is interesting to consider BaC_2 as the adsorbed species. Hoch* gives an equation that indicates a heat of vaporization of 48.8 kcal/mole for the reaction

If BaC_2 is the adsorbing species, then on the basis of Eq.(21), the associated monolayer is 0.0073 g Ba/g C (the concentration at which $\Delta H_v = 48.8 \text{ kcal/mole}$). The vapor pressure at this concentration and at the temperature 1829°K according to Fig. 40 is $1.2 \times 10^{-4} \text{ atm}$. Hoch's equation indicates that the vapor pressure of Ba over BaC_2 is 2.8×10^{-3} at 1829°K . These two values differ somewhat, but the agreement is much better than one obtains if liquid barium is assumed to be the adsorbed phase at the monolayer. It should be noted that the two vapor pressure values (1.2×10^{-4} and $2.8 \times 10^{-3} \text{ atm}$) are sufficiently close that the uncertainties in heat and vapor pressure data may be sufficient to resolve the difference. The monolayer value of 0.0073 g Ba/g C is well in line with the theoretical value 0.011 g Ba/g C presented above, particularly when it is considered that the latter value might be lowered somewhat owing to the fact that BaC_2 molecules have a larger effective diameter than do Ba atoms.

The present data are not in agreement with data from the previous studies cited above. The present data are believed to be more reliable, since they appear reproducible and were obtained under generally better experimental conditions (for example, background pressures were lower in the present studies).

Sorption of Cesium and Rubidium in Graphite. Work is continuing on the sorption of cesium and rubidium on graphite using the isopiestic method. The purpose of this work is to obtain information for use in predicting the release of fission-product cesium and rubidium from PSC fuel blocks.

*M. Hoch, "Heat of Formation of BaC_2 " J. Appl. Phys., Vol 29, 1959, p. 1588

Approximately 0.5 g of graphite having a known BET surface area is placed in the horizontal leg of an L-shaped tube constructed of Pyrex glass and stainless steel. After evacuating the system, cesium metal tagged with Cs^{137} to a known specific activity is distilled from an auxiliary glass tube into the Pyrex leg of the tube. The entire system is thoroughly out-gassed and sealed off at a pressure of $\sim 10^{-5}$ mm Hg.

During a run, the cesium pressure is fixed by maintaining the cesium metal in the Pyrex leg at a constant temperature. The amount of cesium on the graphite specimen is monitored with a calibrated gamma scintillation counter until an equilibrium condition has been established at the desired specimen temperature and cesium pressure. A diagram of the apparatus is given in General Atomic report GA-6415.*

Measurements to obtain sorption isotherms on the rubidium-HLM-85-graphite system have been completed. Isotherms were obtained at 700° , 800° , 900° , and 1000°C over a rubidium pressure range of 2×10^{-8} to 9×10^{-5} atm. Equilibrium conditions were established within a few hours, as compared to several days for equilibration of the cesium-graphite system. This indicates that the diffusivity of rubidium in graphite is considerably greater than that of cesium at the same temperature.

Figure 43 is a log P (pressure) versus log C (concentration) plot of these data. The data form a family of straight lines, indicating obedience to the Freundlich adsorption isotherm. These results can be interpreted by assuming that liquid rubidium is the adsorbed phase at the monolayer concentration. In this interpretation, the heat of adsorption (the heat for the sorption of rubidium from the liquid state) is obtained by subtracting the heat of vaporization of liquid rubidium (19.6 kcal/mole) from the total heat of sorption. If heats of adsorption determined in this manner are plotted against log C and the resulting line extrapolates to $\Delta H = 0$, a monolayer coverage of 4.0 mg Rb/g C is obtained. This value for a monolayer equivalent is unrealistic, since 0.37 mg Rb/g C corresponds to a monolayer, assuming an atomic radius for rubidium of 2.43 Å. This means that the isotherms in Fig. 43 converge at too high a concentration and may reflect uncertainties owing to insufficient data points.

Figure 44 compares cesium and rubidium isotherms at 800° , 900° , and 1000°C , where the concentrations are expressed as $\mu\text{moles metal/m}^2$ graphite. The cesium isotherms are those reported in General Atomic

*C. E. Milstead, A. B. Riedinger, and L. R. Zumwalt, Cesium-Graphite Sorption Isotherms Determined by an Isopiestic Method, General Atomic Report GA-6415, June 1, 1965.

Fig. 43--Rb sorption on HLM-85
graphite

Fig. 44--Rb and Ce sorption on HLM-85 graphite

report GA-6415. It can be seen that the sorption characteristics of these metals are very similar, as would be expected. The apparent difference in the 800° and 1000° C isotherms can be attributed to uncertainties in the relatively small amount of rubidium data.

Two runs were made to investigate the cesium-rubidium-graphite system (cesium and rubidium cosorbed on graphite) at 1000° C using HLM-85 graphite. A cesium-rubidium metal source was used. In the first run, the source was made up so that the cesium pressure was approximately four times that of rubidium, assuming these metals form an ideal (Raoult's law) solution. In the second run, approximately equal partial pressures were obtained from the source (which contained 0.67 mole fraction rubidium). In both runs the preferential sorption of cesium was shown. This was indicated by initial rapid sorption of rubidium followed by desorption as the cesium loading increased above that of rubidium. The sorption data from both runs are somewhat scattered and do not yield reliable isotherms.

Three runs are now in progress to obtain cesium sorption isotherms on EGCR graphite. Measurements are currently being made at 800°, 900°, and 1000° C using 6 to 10 mesh granules of the needle-coke graphite having a BET surface area of 2/5 m²/g. This surface area is considerably greater than that found for HLM-85 (0.7 m²/g) and needle-coke graphite, Grade CHN (1.1 m²/g), and H315A (0.3 m²/g). It is expected that this increased surface area will result in higher loadings of cesium at a given temperature and cesium pressure than observed with the other graphites.

Diffusion of Metallic Fission Products in EGCR Graphite. The diffusion of barium in EGCR graphite is being studied to obtain data for use in calculations to predict the release of Ba¹⁴⁰ from PSC fuel blocks. Information on the rate of diffusion of Ba¹⁴⁰ is particularly important, because this nuclide will decay appreciable (owing to its relatively short half life, 12.8 days) during passage through the graphite of the fuel block.

Three separate experiments for measuring diffusion coefficients for barium in EGCR graphite are under way. The three experiments are being conducted in separate furnaces under identical conditions except for operating temperatures, which are 900°, 1000°, and 1200° C.

The apparatus used in measuring diffusion coefficients consists of a graphite compact held inside a graphite sleeve. The sleeve is mounted in a King furnace and is resistance heated. The compact is first loaded with a mixture of barium oxide (tagged with 7.5-year Ba¹³³) and graphite powder and preconditioned by heating it in a separate graphite tube at 1200° C for several hours. The purpose of this is to react BaO with the graphite powder to form elemental barium uniformly distributed in the graphite compact.

The compact and sleeve are brought to a desired temperature and held for sufficient time to allow the tagged metal to be converted to the elemental form and to diffuse out of the compact into the graphite. Periodically the experiment is interrupted, and the sleeve is removed from the King furnace and cored to obtain a profile of the concentration of metal in the graphite. In coring, a hole is drilled into the sleeve using a special drill, and the activity of the cuttings is determined as a function of depth. Diffusion coefficients are determined from the concentration profiles.

The status of the diffusion experiments is given in Table 20. In the experiment at 900°C, analysis of corings show that 506 hr barium has not penetrated significantly into the graphite sleeve. In the experiment at 1000°C, barium has shown small penetration. The small depth of penetration allows only an approximation of the diffusion coefficient. The approximate value is 4×10^{-9} cm²/sec. In the experiment at 1200°C, barium has penetrated appreciably in a period of only 12.6 hr allowing a good estimate of the diffusion coefficient. The estimated value is 7.8×10^{-8} cm²/sec.

Table 20
STATUS OF BARIUM DIFFUSION EXPERIMENTS

Temp (°C)	Total Time (hr)	Ba in Sleeve (mg)	D (cm ² /sec)
900	506	0.078	---
1000	246	0.70	$\sim 4.0 \times 10^{-9}$
1200	12.6	0.76	$\sim 7.8 \times 10^{-8}$

The accuracy of these diffusion coefficients will improve as the depth of penetration increases at each temperature.

From the two values of the diffusion coefficient, the activation energy for barium is calculated to be around 55 kcal/mole.

The activation energy (55 kcal/mole) and diffusivity at 1000°C (4×10^{-9} cm²/sec) compare to 70 kcal/mole and 2×10^{-9} cm²/sec, respectively, found previously for barium in HLM-85 graphite. This comparison suggests that barium has about the same diffusivity at 1000°C in the two graphites.

Cesium Plateout on Metal Surfaces. Studies of the plateout of cesium on stainless steel and copper surfaces is continuing. The purpose of this work is to gain information for use in predicting the degree of cesium plateout on the surfaces of reactor components. An isopiestic method is used. The apparatus and experimental procedure are the same as those used in studies of the sorption of cesium and rubidium in graphite, except that a metal specimen is substituted for the graphite. This specimen is a strip of 0.002-in. stainless steel foil 3 cm by 20 cm loosely rolled up so that the geometric surface is considered to be 120 cm². Before a run is started the specimen is outgassed at the desired temperature before sealing off the system at a pressure of about 10⁻⁵ mm Hg.

A run measuring the plateout on Type 304 stainless steel was terminated after 70 days. The steel specimen was vacuum-baked for 18 hr at 1350°F before the run was started. A summary of the data obtained is shown in Table 21.

Table 21
SORPTION OF CESIUM ON TYPE 304 STAINLESS STEEL

Temp (°F)	P _{Cs} (atm)	Monolayers Cs ^a
1350	3.5x10 ⁻⁶	11.6
1350	1.5x10 ⁻⁷	9.0
1350	1.8x10 ⁻⁸	5.6
1100	3.0x10 ⁻⁶	11.9
1100	2.1x10 ⁻⁷	10.2
1100	1.8x10 ⁻⁸	9.4

^aOne monolayer Cs =
4 x 10¹⁴ atoms/cm²

The sorption levels found are approximately 7 times lower than those found in a previous run performed under similar conditions. The specimen showed no traces of oxidation at the termination of this run; ordinarily the specimens have a dull surface appearance at the end of a run. Thus, it must be assumed that the decrease in cesium plateout is due to a lower degree of surface oxidation of the specimen. The dependence of plateout level on cesium pressure is lower at 1100°F than at 1350°F. This behavior (lower dependence of plateout level on cesium pressure at lower temperature) has been observed in all plateout runs, regardless of the actual number of monolayers of cesium present.

One additional run is still in progress measuring the plateout of cesium on Type 304 stainless steel at 650°F.

An experiment to measure the plateout of cesium on "clean" copper at 650°F was completed after 109 days. The equilibrium loading with a cesium pressure of 6×10^{-6} atm was found to be 73 monolayers of cesium (a monolayer of cesium is equivalent to about 4×10^{14} atoms/cm²). The ease of removal of the cesium with water (see below "Decontamination Studies") indicates that cesium did not diffuse appreciably into the copper under the conditions of the plateout experiment.

Sorption of Strontium on Steel and Dust Particles. Experimental work is under way for measuring the relative affinity of graphite dust particles and steel surfaces for strontium nuclides. The purpose of this work is to obtain information for use in predicting how the fission product nuclides of strontium (Sr⁸⁹ and Sr⁹⁰), that escape from HTGR fuel elements distribute themselves in the reactor.

The work involves measuring the relative sorption of strontium on graphite dust particles prepared from EGCR graphite and on steel surfaces. Type 304 stainless steel is being used in the initial experiments.

The apparatus consists of a closed loop which contains (1) a helium circulator, (2) a helium purifier, (3) a strontium source that consists of a mixture of Sr⁸⁵-tagged strontium and graphite powder contained in a closed graphite crucible, (4) a Type 304 stainless steel containment tube, (5) a charcoal trap, and (6) the specimens, which are dust particles (105-125μ) contained inside a stainless steel tube of small inside diameter. The strontium source and specimens are contained in separate furnaces. Helium is recirculated over the source held at a temperature sufficient to yield a strontium vapor pressure of 10^{-5} to 10^{-7} atm. The system pressure is about 10 cm Hg above atmospheric, and the helium flow rate is about 200 cm³/min. The vapor-laden helium passes over the dust particles held at a desired temperature (800°C in the initial run). Excess vapor in the gas stream is removed by the charcoal trap. The buildup of strontium on the steel and graphite is monitored by a gamma scintillation counter. The experiments will be terminated when steady-state conditions are reached, and the steel and dust specimens will be analyzed by gamma spectrometry to determine the relative sorption of strontium.

In a variation of the dust experiment, EGCR graphite particles impregnated with Sr⁸⁵-tagged strontium are annealed in small sealed steel tubes for a sufficient time for strontium to equilibrate between the graphite particles and the surface of the steel. At the end of the experiment the tube is quenched and the relative sorption of strontium on the graphite and steel is determined by gamma spectrometry. Vapor pressures inside the sealed

tube at equilibrium can be obtained from Knudsen-cell studies or from the loop experiments described above. Use of the sealed-tube technique will make possible conducting several experiments simultaneously. Apparatus for conducting the sealed-tube experiments is currently being assembled.

It is expected that these experiments will yield data on the relative affinity, on a surface area basis, of dust particles and steel surfaces for strontium vapor.

In addition, these experiments should yield independent measurements of the vapor pressure of strontium sorbed on EGCR graphite to supplement measurements using the Knudsen-cell technique.

HELIUM PURIFICATION SYSTEM

Behavior of Sorbent Beds

Preparations are being made for the measurement of the dynamic adsorption coefficient of tritium on activated carbon at low temperatures (-250° to -320°F). A license amendment authorizing use of the required amount of tritium gas has been approved by the California Bureau of Radiological Health.

In addition, apparatus is being designed for studying the effectiveness of charcoal beds for adsorbing gaseous fission products, such as cesium, tellurium, and iodine, at relatively high temperature (600° to 800°F).

Decontamination Studies

Development of Decontamination Methods. Work is beginning on the development of specific, efficient methods for the decontamination of PSC reactor components. In this work, emphasis will be placed on vacuum-cleaning, steam and high-pressure liquid spraying, and possibly electrolytic cleaning, rather than on chemical cleaning as in the past. The trend away from chemical cleaning methods was prompted largely by the intricate nature of some of the components to be decontaminated. The fuel-handling machinery, for example, is generally complicated and contains parts, such as relays, wiring, and limit switches, which could be damaged by harsh chemical cleaning methods.

Decontamination of Cesium from Stainless Steel and Copper Surfaces. Experiments were conducted to determine the ease of removal of cesium that had plated out on relatively clean stainless steel and copper surfaces. The metal specimens (see Table 22) used in this work resulted from studies

Table 22
STAINLESS STEEL AND COPPER SPECIMENS USED IN DECONTAMINATION STUDIES

Specimen ^a	Material	Pretreatment of Specimen	Monolayers ^b Cs Deposited	Run Temp ^c (°F)	Length of Run (Days)
V-4	304 SS	Degreased; vacuum bake at 1020°C, 22 hr	3.79	1350 1100 840	69
V-8	304 SS	Degreased; vacuum bake at 650°F, 18 hr	26.1	650	24
V-10	Copper	Degreased; vacuum bake at 900°C, 18 hr	73	650	109
V-11	304 SS	Degreased; vacuum bake at 1350°F, 18 hr	9.47	1350 1100	70
Tube from V-10	304 SS	HNO ₃ -HF etch; vacuum bake at 900°C, 18 hr	---	650	109

^a—Specimens V-4, V-8, and V-11 were 2-mil-thick foils having a total surface area of 120 cm². Copper specimen V-10 was a 10-mil thick foil having a total surface area of 60 cm². Specimen V-10 was a Type 304 stainless steel tube, 3/4 in. in diameter.

^b—A monolayer of cesium is equivalent to about 4×10^{14} atoms/cm².

^c—Temperatures at which plateout experiments were conducted.

of cesium plateout on metal surfaces. The cesium was tagged with Cs^{137} ; the agents used a commercial descaling and decontamination agent.

The treatments used and their effectiveness in removing the cesium are described in Table 23. Each specimen was successively exposed to the various treatments.

The results indicate that water was fairly effective in removing cesium from stainless steel and copper surfaces but that the use of the descaling and decontamination agent was not effective in removing what remained. The effectiveness of water in removing cesium from the copper surface (more than 99% of the cesium was removed by the water treatment at 75°F for 16 hr) indicates that cesium did not diffuse appreciably into the copper during the previous plateout experiment, which was conducted at 650°F for 109 hr.

Table 23

RESULTS OF DECONTAMINATION TESTS ON STAINLESS STEEL AND
COPPER SPECIMENS CONTAMINATED WITH CESIUM

Treatment			Cesium Remaining After Treatment ^b (%)				
Agent	Temp (°F)	Time (min)	Stainless Steel Specimen V-4	Stainless Steel Specimen V-8	Copper Specimen V-10	Stainless Steel Specimen V-11	Stainless Steel Tube from V-10
Water	75	5	NP	23	NP	56	NP
Water	75	30	8.4	7.8	NP	37	15
Water	150	30	6.3	NP	NP	NP	NP
Water	150	30	4.5	NP	NP	NP	NP
Water	75	16 hr	2.5	3.3	0.18	3.7	1.1
Agent ^a (15:1)	150	30	NP	2.6	NP	1.2	NP
Agent ^a (5:1)	150	30	1.9	0.64	0.06	1.1	NP
Agent ^a (Undiluted)	150	30	NP	0.61	0.06	0.8	NP
Water	75	16 hr	NP	0.55	0.05	NP	NP
Water	75	72 hr	1.7	0.54	0.05	0.7	NP

^a—Descaling and decontamination agent.

^b—NP = not performed on specimen.

PART II

TASK I
FUEL TRANSFER MACHINE

The conceptual design of a fuel transfer machine was based on the hexagonal block type of fuel element. Basic design criteria for the pickup and handling mechanisms have been developed as follows:

1. Pickup surfaces in the fuel block will be provided near the bottom of a central, blind hole in the graphite which penetrates nearly through the block to allow pickup stresses in the graphite block to be primarily compressive.
2. Sufficient overtravel will be provided to enable the machine to cope with unusual situations with regard to fuel block location.
3. The grapple head will have the capability of automatically engaging any block with 5/8 in. of nominal position with the current block design.
4. The grapple head can float 1 in. in any direction and tilt 5° out of the horizontal plane.
5. The grapple head itself will have a maximum lifting and seating force capability of 1000 lb with strain-gauge sensing to measure these forces.
6. Pickup ledge area under load in the fuel block is about 1/4 in.².
7. Various sections of the grapple head will have plug-in type electrical connections to simplify disassembly for maintenance.
8. The main lift boom will be of the telescoping type with telescoping drive shafts.
9. The grapple head will operate with an overhead clearance above the core of about 6 ft with a 3 in. by 30° chamfer on the insulation at the bottom of each refueling nozzle penetration.

10. The grapple head will have a 30-in. maximum reach to allow replacing the first layer of reflector blocks beyond the normal fuel patch at the outer boundary of the core. This will eliminate the necessity of additional refueling nozzles at the outer edge of the core to handle these reflector blocks.

Bearing Tests

For some time, tests have been under way to evaluate the performance of dry-film lubricants in helium under the temperature and chemical impurity level at which certain critical-service bearings in the fuel transfer machine would be expected to operate. In these tests, a radial load of 440 lb is applied at a speed of 53 rpm, the direction of rotation being reversed each hour. The temperature is continuously held at 500°F and the helium impurity to a level of 5 ppm of oxygen or less. Prior to the beginning of this quarter, three tests were run and a fourth was started.

Test No. 1 was performed with a bearing lubricated with Lubeco CL-6002-M390 dry film. The bearing was operated to complete destruction within 50 hr.

Test No. 2 was performed with a bearing lubricated with Everlube 811 dry film. At 150 hr the test was stopped for examination of the bearing although the torque was still at an acceptable level. It was found that the bearing had developed excessive internal clearance (about 0.025 in.). Since the bearing tester was not equipped for measuring bearing clearance, the relationship of wear to running time was not known. However, from observation it could safely be concluded that the first bearing (Lubeco lubricated) was serviceable for 20 hr, and the second bearing (Everlube lubricated) for 100 hr.

Test No. 3 was performed with a bearing coated with Lubeco 6021 dry-film lubricant. After operation under the above conditions for 83 hr, the test was discontinued. The friction torque, which had measured 20-30 in.-lb at the beginning of the test, had increased to 30-55 in.-lb.

The bearing was removed from the test rig and disassembled for inspection. It was found that the internal diametral clearance had increased by slightly less than 0.001 in. On the S-Monel separator it was apparent that wear had occurred along the line of contact with the balls. A series of photomicrographs of the balls, races, and separators was taken, but proved inconclusive in distinguishing between wear failure and solid film lubricant particulate matter deposition.

Just before the beginning of the present quarter, testing of a bearing coated with Everlube 811 (Test No. 4) was started. This bearing operated

for 205 hr, at which time the system torque had increased from an initial value of 20 in. -lb to 60 in. -lb.

Test No. 5 was performed with a bearing identical to bearings Nos. 1 through 4, but without any lubricants, to obtain more specific data on the behavior of the bearing materials during breakdown. This test was terminated after one hour of operation, at which time the system torque had reached 90 in. -lb from an initial value of 20 in. -lb.

Photomacrographs of this bearing were used to distinguish between true wear and deposits of solid-film-lubricant particulate matter that were previously indistinguishable in earlier photomacrographs.

In addition to identifying that true wear had started (but was not excessive) on the previous tests, a microprobe examination of one of the balls from Test No. 3 was made. This examination identified elemental constituents found not only in the solid-film lubricant but also those from the separator and races indicating metal pickup on the balls.

Test No. 6, the evaluation of DF-700 (A Marlin Rockwell Corporation proprietary coating), was initiated late in the quarter. Preliminary indications give considerable promise for this coating. For this reason, an evaluation of the bearing test program that was initiated in the previous quarter has been suspended pending the completion of Test No. 6.

TASK II

SERIES-STEAM-TURBINE-DRIVEN HELIUM CIRCULATOR

Before the beginning of the quarter, the design of the helium compressor was essentially complete for application to an HTGR using a steel pressure vessel for the steam generator and a separate one for the reactor. The rapid progress toward a prestressed concrete reactor vessel (PCRv) and a later service date for the first large-sized HTGR made it apparent that the research and development program on the helium circulator should be reexamined. It also appeared that all future HTGR plants would use the PCRv, thereby necessitating changes in the dimensional and physical characteristics of the helium circulator. It therefore seemed appropriate also to consider whether other anticipated future design changes and improvements could be examined at the same time. The development program was reevaluated on the basis of the specific requirements for a large HTGR employing a PCRv, and the following factors were taken into account:

1. Optimization of the circulator design and plant operating parameters.
2. Simplification of the circulator design and its accessory system, considering the requirements of the PCRv installation.
3. The extension in the time schedule when production units will be required for installation in a PCRv plant. This extension may be sufficient that advancements in circulator design can be incorporated into the machines at an earlier date than would have been possible if the machines were to be used in the steel vessel plant as had been previously planned.

The results of the evaluation indicated that a large portion of the past research and development effort would be applicable to the new requirements, although future detailed design work should be directed toward a circulator that is compatible with the PCRv, has simpler auxiliaries, is more reliable, and incorporates water-lubricated bearings and seals. Also, the design work should employ a single-stage compressor and include further study on the diffuser section. In keeping with the results of the evaluation, the helium circulator program was redirected toward a design satisfying these general criteria.

For a plant employing a PCRVR, there are a number of changes in the circulator design requirements. The casing closures are considerably different; there is a much greater emphasis on the closure size and the diameter of penetration hole in the pressure vessel required for installation and removal of the circulator. A new circulator design was therefore required. Preliminary investigations also showed that a single-stage machine is feasible if the system pressure is increased to 575 psi.

The former circulator accessory system required that the three fluids within the casing of the machine--oil, steam, and helium--must be kept separate from each other under operating conditions. The best method of ensuring this separation was through the use of a buffer fluid, helium, between the compressor and the lubricating system. The buffer gas and lubricant on both sides of the machine, and the buffer gas and steam were designed to leak into three separate chambers along the axis of the machine at a rate that was controlled by their pressure levels and by the seal and bearing clearances. The constituents of the three leakage flows were separated in the accessory system and were then returned to their respective systems. Since the pressure in the steam side of the system varied with the load, and since the leakage rate was a function of pressure difference, the pressure level of the lubricant had to be kept at a value approximately equal to that existing in the fluid contiguous to it. This resulted in two lubricating systems that leaked into each other. The equipment required to circulate the various fluids, control the leakage, and separate the fluids that mix with each other became somewhat complex.

The use of water for the lubricant eliminates one of the three fluids. Since some leakage of water into the steam side can be allowed, the system can be considerably simplified. A water-helium seal will still be required, but it will be the only one requiring a low leakage and a buffer gas. Furthermore, a source of high-pressure water is already available from the feed-water system. The separation of water from helium is no more difficult than separating oil from helium. It may prove to be easier.

The use of water as the lubricant instead of oil introduces no particular difficulties for the bearings if hydrostatic lift is used for startup and for operation at low speeds. Since startup and low-speed operation presented rather severe conditions for oil lubrication also, a combined hydrostatic-hydrodynamic bearing system would have been required with the previous circulator design as well as with the new design.

Another pertinent issue is the constraint that the PCRVR application places on the circulator, particularly on the degree of design flexibility that is allowed. This is discussed below for the various components and the circulator.

The aerodynamic design of the compressor and turbine is determined by the primary coolant system characteristics and has been optimized for the PCRV plant by General Atomic. The static aerodynamic components are determined by the plant arrangement. The performance of one of these components, the diffuser, has been set nominally at a reasonably low level in the preliminary design because of difficulty in obtaining vendor agreement as to what is achievable (there is a tendency to use conservative numbers). The diffuser is a more important component for this circulator, which is a single-stage machine, than would normally be the case in most multistage compressors, because the amount of kinetic energy lost in the diffuser is of the same magnitude as that lost in the blading. Since this loss must be made up in the blading, the tendency is to assume conservative design values for the diffuser efficiency to be sure the over-all performance will be adequate. However, if sufficiently early in the schedule the component is proven by testing to be more efficient than assumed, the advantages of this improvement can be factored into the final design. It is also important to investigate other possible geometric configurations so that more freedom in component arrangement results for the entire reactor.

The casing of the machine and its closure, which forms the seal for the primary coolant gas circuit, will determine the size of the hole required in the PCRV. The other half of the seal is part of the reactor vessel liner. Therefore, a major part of the circulator design and its envelope will be specified by General Atomic to meet these conditions.

The reference circulator and its installation are shown in Fig. 45. This design has a single-stage axial compressor driven by a single-stage steam turbine in series with the main steam turbine-generator set. An increase in primary coolant system pressure for this reference design from 450 psia to 575 psia decreases the pumping power and increases the gas density. This reduces the required pressure rise and blade annulus of the compressor, so that a single-stage compressor is adequate for the design and has margin for possible future increases in requirements.

Figure 46 shows the variation of pressure rise and axial velocity as a function of system pressure with compressor diameter as a parameter. Since the outer diameter of the blading determines the size of the hole in the vessel, it should be minimized. A 3.5-ft diameter, therefore, was selected.

The aerodynamic design of the compressor for the reference case is shown in Table 24.

Fig. 45--Installation of helium circulator preliminary design

Fig. 46--Variation of pressure rise and axial velocity as a function of minimum system pressure for various compressor outer diameters

Table 24

COMPRESSOR AERODYNAMIC AND MECHANICAL DESIGN

Inlet pressure	565.1 psia
Inlet temperature	755°F
Pressure Rise	9.9 psi
Outer diameter	3.5 ft
Inner diameter	2.35 ft
Rotational speed	5730 rpm
Blade tip speed	1050 ft/sec
Gas axial velocity	525 ft/sec
Mass flow rate	482 lb/sec
Blade cent. stress	18,036 psia
Blade bend. stress	5,980 psia
Blade chord length	5.96 in.

There is a margin of approximately 13% increase in speed permissible with this design, which allows for a flow increase of 13% and a pressure rise of 28% for exigencies and changes to the primary system.

The turbine design is shown in Table 25. This design is much more favorable than was previously possible.

The turbine and compressor wheels are attached to the shaft by means of curvic couplings and both are overhung on each side of the bearing housing. The bearing housing is cantilevered from the casing, and water-lubricated bearings are used. Tilting-shoe bearings with hydrostatic pockets in each pad are used for both the journal and thrust bearings. The design basis is that sufficient redundancy can be accomplished by providing for adequate hydrostatic lift from only half the pads at the maximum load condition--startup to 25% speed. The hydrostatic action would be required at part load, so that under normal operating conditions prevailing in the power plant, 70% to 100% percent load, the bearings would be operating hydrodynamically. The thrust bearing will probably be a 16-in.-OD bearing with eight pads. The loads are such that there

will be approximately 50 psi at design speed with a minimum film thickness of 0.001 in. The hydrostatic action will be started at the part-load condition, when the minimum film thickness reaches 0.0005 in.

Table 25

TURBINE DESIGN DATA

Enthalpy drop	21 Btu/lb
Steam flow rate	301 lb/sec
Inlet pressure	820 psia
Inlet enthalpy	1351 Btu/lb
Outlet pressure	645 psia
Outlet enthalpy.	1330 Btu/lb
Outer diameter	25.7 in.
Inner diameter	22.0 in.
Blade tip speed	642 ft/sec
Steam through flow	250 ft/sec

The turbine end sealing system is simplified in that a labyrinth is used to restrict the steam leakage and the visco, or screw-back, seal to restrict the water flow on the bearing side. The steam-water mixture that results from the leakage past these seals is condensed in a heat exchanger in the accessory system and returned to the feedwater system at a convenient point. On the compressor side, a viscoseal is used to limit the water leakage out of the bearing, and as in the previous design, clean helium buffer gas from an external supply is used to prevent diffusion of water vapor into the primary system gas, and vice versa. The gas-water mixture is ducted from the machine into the accessory system, where a heat exchanger, mechanical separator (bed type), and molecular sieve are used to separate the helium and water and return each to their respective systems. The buffer gas supply, which is furnished from the purified helium system, enters a double labyrinth seal and leaks in both directions. Inboard of the viscoseal, or compressor side of the machine, there is a shutdown seal that is open when the pressure in the lubrication system is the same as the helium pressure but is closed when the water pressure is reduced.

At the beginning of the quarter, the development program for achieving the objectives set forth above had been formulated. The following elements were included:

1. Rotor Dynamics Simulation. A program for designing and developing water-lubricated bearings for this application can only be assessed by the simulation of the operating conditions of pressure, temperature, speed, and load, both statically and dynamically. This requires a full-scale rotor rig to simulate the polar moment of inertia and the transverse radius of gyration. The seals and sealing system, the bearings and lubrication system, the rotor vibration, and the method of disk attachment can all be evaluated, and proof of the satisfactory solution of these mechanical problems can be established. The performance of both hydrostatic and combined hydrostatic-hydrodynamic bearings and their required accessories will be determined so that the choice of the type of bearings can be made.

2. Static Aerodynamics. The compressor inlet and compressor diffuser are components for which proof of performance is necessary early in the program. This development can be accomplished with a scale model if the Reynolds number is simulated. The essential feature of the test equipment is a water model of both components with a means for simulating the boundary layer imposed by the blading and for testing the components over a range of velocity gradients; the absence of flow separation is the index of achievement. A number of different designs will be evaluated, including the use of bound vortices to avoid separation and the feasibility of a radial discharging diffuser.

3. Full-scale Test Facility. The construction of a prototype test facility independent of any particular vendor will require, in addition to the determination of a location, that a test rig be designed that is suitable for prototype development testing, production machine acceptance testing, and reliability testing. This rig would also be useful for evaluating casing seal design.

During the present quarter, work has begun on all three development areas described above.

ROTOR DYNAMICS SIMULATION, LUBRICATION AND SEAL SYSTEM

The design of a simulator and test facility for the simulation of the rotor dynamics, for bearing evaluation, and for accessory system tests has been started. The work is being accomplished by Mechanical Technology, Inc., located in Latham, N.Y. In addition, two different bearing designs are being made by both Mechanical Technology, Inc., and Franklin Institute, Philadelphia, Pennsylvania, so that a total of four designs will be evaluated in a later phase of the program. The basis for the two design concepts that each vendor will do is, first, a hybrid bearing which is a hydrostatic bearing that has hydrodynamic action and, second,

a hydrodynamic bearing with hydrostatic lift for startup and low-speed running.

The problems that are being considered in the journal bearing design are: the prevention of dynamic instabilities such as half-speed whirl; the maintenance of sufficient clearance to ensure operation and assembly; and the provision of sufficient bearing stiffness so that the rotor and bearings do not, in combination, resonate in the running range of the machine. The major problem in the design of the thrust bearing is developing the ability to carry the loads at acceptable minimum film thickness. The thrust loads and their variation with speed are shown in Fig. 47. The curves shown are for three different shaft sizes and for three different regimes, startup, normal operation, and overspeed. The assumed mode of operation for the startup regime results in an acceleration of the machine to 45% speed by means of steam from the auxiliary boiler, and then, through the use of steam from the flash tank (employing nuclear heat), a deceleration to 25% speed at which time the nuclear steam supply is at 25% load. Normal power operation would now begin. Overspeed operation cannot occur with all of the circulators operating since the steam supply is limited. It was therefore assumed that half of the machines were shut down, and the thrust loads shown are those in the operating circulators.

The high startup load results from the difference in the steam and helium pressure at the two ends of the machine which are almost balanced at design speed. Because (1) 95% of the operation of the circulator should be at above 75% speed over the plant life, (2) wear of the bearing surfaces can only occur during boundary lubrication (before the fluid film is of sufficient thickness), and (3) the startup of the plant, and hence the circulator, will take an appreciable time, use is made of hydrostatic action for external lift-off of the rotor from the bearing surfaces. The other fact that is important in this design is that the bearings will be operating in the turbulent regime, which means that the frictional losses and the ability to carry load are both increased substantially.

The accessory system required for the lubrication and seal system is shown in Fig. 48. The simplification inherent in the use of water is apparent when one compares this with the accessory system required for oil lubrication which is shown in Fig. 49. This simplification results from the fact that the buffer requirements on the steam end are no longer present; since the fluid in the bearing and the fluid in the turbine are the same (except for the physical state they are in), limited and direct mixing are now acceptable so that a single pressure lubrication system is feasible. Although the present reference design uses a labyrinth on the steam side and allows both the steam and the water to leak out into a condenser, it is possible to allow the leakage of the water into the down-

Fig. 47--Plot of series-steam-turbine-driven circulator downward axial thrust as a function of speed for normal operation and startup

Fig. 48--Accessory system required for the lubrication and seal system with water-cooled bearings

Fig. 49--Accessory system required for the lubrication and seal oil system without water-cooled bearings

stream steam side of the turbine, thereby eliminating the condenser and its control. This would mean the use of a controlled clearance water seal.

STATIC AERODYNAMICS, DIFFUSER AND INLET

The importance of the compressor diffuser efficiency is shown by Fig. 50. An increase in diffuser efficiency from 80% to 90% together with the slightly thicker entering boundary layer resulting from increased loading of the compressor blading by designing for a diffusion factor of 0.55 instead of 0.50, allows an increase of pressure rise capability from 14 to 16 psi in a single-stage machine of the same size and stress levels. This increase in capability is highly desirable when one considers higher pumping power in future HTGR plants. Since there is a lack of data on annular diffusers and on the effect of inlet boundary layer on diffuser performance, one must, for a particular design, assume relatively low values in the reference design of the circulator. However, diffuser efficiencies greater than 90% have been achieved in wind tunnels, and the proof of this performance level so that it can be incorporated into the design is of great advantage in the case of the circulator. The organization of work to prove this higher performance is being planned, and the work should start within the next quarter.

FULL-SCALE TEST FACILITY FOR HELIUM CIRCULATOR DEVELOPMENT PROGRAM

A facility for testing elements of the circulator at the operating conditions of speed, temperature, and pressure with helium is required in the development program in order (1) to ensure the solution of problems such as the delineation of the compressor blade vibration characteristics which cannot be accomplished in the simulator or other test rigs; (2) to allow for measurement of performance in the compressor and turbine components; and (3) to ensure the attainment of satisfactory reliability in the machine for power plant use. The steam requirements for operation at full speed, temperature, and pressure are so great that this testing can best be accomplished at a utility power plant.

The steam flow requirement is based on using less than design flow rate and increasing the work done per pound of steam flow; this reduces the flow required in direct proportion to the ratio of test enthalpy drop to plant enthalpy drop. Since the blade speed and the rotor blading angles are the same as for the design case, the variable is the steam velocity. This can be increased with the design nozzles to a value

Fig. 50--Plot of compressor parameters versus pressure rise

corresponding to slightly supersonic flow or, with a special set of test nozzles, to higher Mach numbers. The increased work follows from the change in momentum through the blading with the accompaniment of high losses, both in the incidences that the flow makes with the rotor blades and in the component of velocity in the peripheral direction leaving the rotor blades that cannot be recovered.

Neither of these losses detracts from the work done, since they are compensated for by an increase in the enthalpy of the supply steam. However, flow separation in the blade passages, if it does not reattach before leaving the blading, will reduce the work done. Therefore, it is assumed that there will be no expansion in the turbine wheel. This is a severe restriction, since the design is based on some reaction, and therefore expansion in the buckets would increase the work done. The turbine would operate at full admission by reducing the discharge pressure so that the volume flow is consistent with the design. Table 26 gives the conditions for six different flows. The first four can be accomplished with the actual nozzle, while the remaining two will require a special test nozzle. Figure 51 shows the vector diagram for these conditions as well as for the design case. The steam supply required is approximately 350,000 lb/hr at 350 psia for the full-scale test.

The test facility, shown schematically on Fig. 52, consists of a large tank with the test bed circulator installed in the bottom flange and the compressor diffuser mounted coaxially with the pressure vessel. At the top of the diffuser there is a symmetrical center body, the axial position of which can be varied to change the flow resistance imposed on the compressor so that the compressor can be tested over its entire operating range. The heat exchanger is in the outer annulus formed by the shell of the tank and the diffuser. The steam connections to the turbine drive are at the bottom of the tank. The operation of the system will be varied by throttle and bypass valves in the steam supply system. The tank is supported by a skirt and can be mounted out of doors. The circulator will be installed by means of a fork lift truck or a hydraulic jack from below the tank.

Table 26

TURBINE CONDITIONS FOR SIX DIFFERENT STEAM FLOWS

Mach No. Leaving Nozzle	Steam Velocity Leaving Nozzle (ft/sec)	Enthalpy Drop (Btu/lb)	Efficiency	Steam Flow (lb/hr)	Inlet Enthalpy (Btu/lb)	Inlet Temperature (°F)	Inlet Pressure (psia)
0.8	1610	45.4	0.747	529,000	1411	790	400
0.9	1810	54.4	0.710	441,000	1424	808	328
1.0	2010	63.5	0.676	378,000	1436	830	295
1.1	2220	72.3	0.645	332,000	1445	845	267
1.2	2420	82.1	0.613	292,000	1456	865	260
1.4	2820	104.5	0.566	230,000	1480	910	265

Fig. 51--Vector diagram showing turbine conditions for six different steam flows and for the design case

Fig. 52--Full-scale test facility layout

TASK III

CONTROL ROD DRIVE

Prototype testing of a control rod drive for use in large HTGR's was initiated early in 1963 and has continued since that date. The basic design of the rod drive has remained unchanged, with only minor design changes being required as a result of testing to date. The basic design is the same for both regulating and safety rods. The rods are inserted from the top of the reactor and a pair of rods is operated by a single drive. The control rods are cable suspended from the drive, and scram operation is by gravity fall of the rod. The following paragraphs briefly describe the rod drives and their operation. Following the description will be a brief description of the testing program to mid-1965, and a description of the test results during the reporting quarter.

The control-rod drives are winch-type devices, each of which raises and lowers two control rods simultaneously by winding or unwinding cables from two counter-rotating drums (see Fig. 53). The mechanism is contained in a cylindrical housing that is suspended within the reactor vessel nozzle by means of a top flange resting in a counterbore at the top of the nozzle. The housing is divided into an upper and a lower compartment by a lead-filled shield. Attached to the bottom of the drive housing is a cylindrical shield plug through which pass the guide tubes and control rods. The control rods are suspended from cables that pass over the drums, and the guide tubes are attached to mechanisms by means of which they may be manually engaged with the top reflector blocks in the reactor core, when the drives are assembled in the reactor.

The regulating rod and safety rod drives are similar, except for the scram retarder, scram clutch, and retarder release clutch, which are not used in the regulating rod drives. The upper drive mechanism assembly contains those parts that should be readily accessible for servicing. This includes the drive motor, the primary reduction gearing, the scram clutch, the retarder release clutch, the scram retarder, the rod position transmitter, the switches, and the electrical connectors.

The drive motor is a 1750 rpm three-phase induction motor of approximately 0.6 hp rated output. The motor is fitted with a spring-released magnetically applied brake that is applied whenever the motor is

Fig. 53--Control rod drive (safety)

deenergized. The motor connects through reduction gearing to the vertical main drive shaft assembly.

The first unit in the main drive shaft train of the safety rod drive is the magnetically engaged, spring-released scram clutch, which is d-c powered. Next is the scram retarder clutch--an overrunning sprag-type unit. The inner member of the scram retarder clutch is mounted on the output shaft of the scram clutch; the outer member is mounted on the scram retarder unit.

The scram retarder unit is a sealed device of circular configuration consisting of a cylindrical central reservoir with a through tube for the main shaft, a toroidal outer chamber supported from the central reservoir, and two pipes connecting the reservoir and outer chamber. The reservoir is filled with mercury.

The main drive shaft train of the regulating rod drive consists only of a coupling and a main shaft between the reduction gearing and the lower drive. The coupling is a bolted-flange type that is not disengaged in service. In both kinds of drive the main drive shaft is coupled to the bevel pinion shaft, which is part of the lower drive mechanism assembly. The latter consists of two cable drums, two cable guide assemblies, and bevel drive gearing. The bevel pinion shaft drives two bevel ring gears mounted on cable drums. The drums, being mounted on the opposite sides of the pinion, are counter-rotating.

The cable drums are of the "pancake" type, i. e., the cable is wound upon the drums spirally in a narrow groove. Below each drum is a pair of guide pulleys mounted on a rocker arm. The guide pulley and rocker arm assembly is designed so that its position will indicate when the control rod is at the bottom position or the cable has become slack. Rods attached to the rocker arm pass through the shield into the upper chamber to actuate limit switches indicating rocker arm position. The lower drive mechanism assembly is bolted to the underside of the cylindrical shield separating the upper and lower mechanisms and is removable with the shield through the top of the housing.

In the drive housing below the lower drive mechanism assembly, there is a thick shield through which the control-rod cables pass. Surrounding the cables in this shield are tubes from which the control-rod guide tubes are suspended. The suspension tubes are connected by reach rods to the upper mechanism compartment, where they are screw-operated to lower the guide tubes into engagement with the top reflector blocks in the reactor core after the control-rod drive assembly has been mounted in the reactor vessel nozzle.

The electrical and gas connections are made to the control-rod drive through sealed connectors in the side of the vessel nozzle near the top.

During normal operation of the control-rod drives, the control rods are moved out of or into the core by rotation of the drums driven by the motor. Rod speed varies between approximately 2 and 3 in./sec in the range of travel, the variation resulting from the spiral winding of the cable. In the safety rod drives, the scram retarder rotates, but at a velocity too slow to have any appreciable retarding effect.

When the control rods are at the fully inserted position, the cables have been completely unwound from the drums, and the guide pulley rocker arms are rotated to a position in which the "rod bottom" limit switches are closed. If a cable goes slack, the rocker arm is rotated to a position in which the "slack cable" switch is closed. If cable slack occurs as a result of a control-rod sticking during insertion, reversal of the drive by the operator will cause the switch to open again, thus differentiating between a stuck rod and a broken cable.

Rod position indication is obtained from an electrical position transmitter geared to the main drive shaft. The transmitter is connected to a position readout in the control room.

When the reactor is scrammed by a signal from the plant protective system or the operator, the magnetic scram clutches in the safety rod drives are deenergized, disconnecting the drum and scram retarder systems from the drive motors. The control rods drop into the core by the action of gravity, rotating the drums and retarders. The scram retarder limits the rod-drop velocity, so that the rods can be stopped at the bottom position with a reasonable decelerating force. As the velocity increases at the beginning of the drop, the mercury in the central reservoir of the retarder moves out under centrifugal force through the pipes to the outer toroidal chamber, where it is discharged in a direction tangentially forward from the jets. The reaction to the jets produces a retarding torque on the main drive shaft, which regulates the drum speed. Near the end of the rod drop, the cable is completely unwrapped from the drum, and during the last fraction of drum turn, the rod is decelerated by a combination of forces produced by the jets and the changing moment arm of the cable attachment point. When the rods reach bottom, the retarder freewheeling clutch permits the retarder to continue rotating until it coasts to rest. Reactor shutdown is completed by releasing the brakes on the shim rod drives and allowing the shim rods to coast down to the fully inserted position. The coasting speed of the shim rods is retarded by self-excited generator mode operation of the induction motors, which allows the rods to coast in at a speed only slightly greater than their driven speed.

The testing program was divided into two main phases with the first phase divided into two parts. In Phase I the drive assembly was tested with short dummy weights simulating the control rods. Part A of Phase I consisted of functional tests of the drive in air at room temperature. Part B of Phase I consisted of an environmental test in high-purity helium at drive operating temperature. The Phase II testing program (currently in progress) consists of functional tests similar to part B of Phase I, with the addition that the complete drive, guide tubes, and control rod assemblies are being tested. Furthermore, the test apparatus was modified to simulate the design temperature expected at the top of the reactor core over a section of the guide tubes and cables approximately 2 ft in length. The test continues to simulate design temperature in the entire drive area.

Early testing determined the orifice sizes and mercury inventory in the scram retarder necessary to provide the desired deceleration and confirmed that other operating parameters, such as speed and torque, were as specified. Life testing disclosed some difficulties in the bearings and lubricants. The bearing problem was subsequently overcome, but it remains to be determined whether or not a satisfactory high-temperature lubricant has been found.

Side deflection of the cable during a scram was determined to be a function of the final deceleration of the rod. Satisfactory operation was obtained by proper adjustment of the retarder jet size and mercury content and the proper adjustment of the distance between the cable drum and the cable guide pulley.

The original drive motor used in the test was a 220-v three-phase motor of 0.6 hp. As a result of proposed project standardization on 440-v supplies for motors of this size, an attempt was made to use a 440-v motor. Difficulty was encountered in maintaining insulation integrity in the low dielectric strength helium atmosphere. As a result, the motor has been changed to a 110-v three-phase unit.

Phase II testing began in early July. The test is divided into an as yet undetermined number of equal periods, each period consisting of 40 hr of operating time and 128 hr of idle time, environmental conditions being maintained during the idle time. The test was interrupted early in August as a result of difficulties in maintaining the desired purity level in the helium gas. After repairing the vacuum pump and conducting a number of evacuations and purges of the test autoclave, an acceptable helium purity level was achieved, and testing was resumed early in September.

During the latter part of September, difficulties were encountered with apparent drive motor stall and decrease in motor brake torque. The

difficulty with the motor brake was traced to a decrease in stiffness of the springs in the ball plungers, which transmit the solenoid force to the pressure plate. Replacement of the springs with higher capacity units restored brake torque to the proper level. So far, the cause of the motor stalling has not been determined.

TASK IV

STEAM GENERATOR

MATERIALS

Metal-Coolant Compatibility Test

In the HTGR, structural materials may have to operate at temperatures of 1100° to 1375°F for extended periods of time in helium that may contain CO and H₂. These impurities would result from water vapor entering the core from steam generator leakage. To evaluate the effects of small partial pressures of CO on materials and to attempt the establishment of reaction kinetics, an 18,000-hr experiment was initiated in 1962. Eight metallic materials were exposed at 1100° and 1375°F in purified helium and in helium containing 300 and 3000 ppm CO and H₂.

Mechanical properties and weights per unit area were measured after 3,000, 6,000, 9,000, and 18,000 hr of exposure. The 18,000-hr exposure was completed during the quarter, and the furnaces have been shut down. Room-temperature mechanical testing and metallography of the various specimens have been completed. Elevated temperature tests and chemical analysis for these specimens have also been completed, and evaluation of the data is under way. Further testing is in process on Incoloy 800 in order to determine the effects of heat treatment on the properties of this material.

HEAT TRANSFER AND FLUID FLOW

Steam/Water Flow Stability

Before this quarter, an analog computer study of once-through boiling flow stability was started. Solutions of a preliminary model were carried out using eight lumped, spatial nodes. The nonlinear conservation equations representing the system were based upon a homogeneous density and single mass-velocity model (i. e., slip flow was not considered and the mass velocity was uniform along the channel during integration). Den-

sity variations were functions of time and space, whereas velocity variations occurred in time only. This relatively simple model resulted in fair agreement with experimental data at high pressure and low values of inlet subcooling. Attempts to duplicate experimental results at lower pressure and increased values of inlet subcooling yielded poor results. Modifications that involve a more detailed treatment of the conservation equations were started.

Analog studies of boiling stability continued during this quarter with an improved model, which considers spatial variations in mass velocity and inertia in the momentum and continuity equations. Results from the current model demonstrate a considerable improvement over those from the previous model in predicting system response at conditions corresponding to the inception of oscillations for existing experimental data. In one comparative case, the current model requires an inlet damping ratio of 0.12 and the previous model requires a ratio of 0.22 to yield neutral stability (incipient oscillations). The inlet damping ratio is defined as the ratio of pressure drop at the channel inlet to the over-all channel pressure drop. The improved model is still somewhat conservative, however, since excess damping is required to achieve neutral stability in the above case and in other cases as well.

The current model has recently been modified further to include energy storage effects and a spatial variation in mass velocity in the energy equation. This latest version represents the finalized model wherein the development is based upon consistent assumptions and coolant models in all the conservation equations. Testing of the new model is awaiting machine availability.

The digital computer boiling stability code developed at Knolls Atomic Power Laboratory has been converted to FORTRAN language and minor debugging has been completed. In its present form, the code analyzes the stability of a once-through boiling channel with partial evaporation at the channel exit. The code describes the dynamic aspects of boiling flow considering such effects as two-phase slip flow, subcooled boiling bubble formation, nonequilibrium thermodynamic conditions, transient heat conduction at the wall, spatial variation of mass flow rate, and a nonuniform axial heating distribution. A parameter study was carried out to describe the performance of the code in terms of the system response to changes in various system variables and parameters employed in the model.

A number of comparative cases were run at conditions duplicating experimental instability. Results of a few of these code predictions are listed in Table 27 in terms of the system frequency response. The frequency response of a well-behaved system can be examined using the

Nyquist criterion. It suffices for these cases to say that if the magnitude of the gain at 180° phase shift exceeds unity the system is unstable. The margin of stability expressed in degrees is a measure of the extent to which the system operation falls on either side of neutral stability.

Table 27

COMPARISON OF CALCULATED AND EXPERIMENTAL RESULTS

Experimental Conditions at Oscillation Inception				Predicted		
Pressure (psia)	Inlet Subcooling ($^\circ\text{F}$)	Exit Quality	Frequency (cps)	Frequency (cps)	Gain	Phase Margin Degrees
600	36	0.29	2.0	2.2	1.46	-53
800	10	0.53	2.1	2.3	1.77	-40
800	55	0.27	1.2	1.25	1.32	-50
1200	61	0.47	1.6	1.5	1.31	-40
1600	145	0.37	---	---	---	140

There is good agreement with experiment in most cases. Predicted oscillatory frequency is in excellent agreement with that observed. A more useful indication of the stability margin is developed by inserting various degrees of inlet orificing in a given case until neutral stability is achieved. This procedure was carried out in the parameter study mentioned previously. The digital computer solution tends to underestimate the destabilizing effect of high values of inlet subcooling, as opposed to the analog study, which overestimated this effect.

An analysis is under way that is intended to extend the digital computer program into the once-through regime with superheat. Transfer functions are to be developed for the superheat regime and for the transition between vaporizing and superheating regimes. The spatial variations will be considered in a lumped model analysis.

Tube Vibration Studies (Including Flow Pulsation and Impedance)

A test rig that can be used to maintain dynamic similarity by matching the Reynolds number of the full-scale equipment with that of the test equipment and the nondimensional frequencies is being developed. Since it has been established that reliable data can be determined through tests, a test program consisting of two phases has been formulated. The first phase will obtain general, basic information, and the second phase will be concerned with testing proposed steam generator configurations.

Prior to this quarter, the detailed test specification and the sequence of testing for the first phase were developed. With the planned test rig, it will be possible to test the effect of a large number of variables, such as changes in plenum volume, tube bundle arrangements (triangular and rectangular pitched), changes in tube support spacing, gas flow rate as well as flow pulsations, and external, mechanically induced vibrations.

The fabrication and installation of the tube vibration test rig has essentially been completed. Work is progressing on the preparation and installation of the instrumentation on the tube bundle and test rig. The testing program for Phase I of the study should be initiated during the coming period. Figure 54 is a simplified loop schematic of the vibration test facility, and Fig. 55 is a photograph of the test rig itself.

DESIGN AND ANALYSIS

Design Study

Conceptual design work on steam generators for use in a PCRV was initiated by Combustion Engineering, Foster-Wheeler, Babcock & Wilcox, and Sulzer Brothers just before this quarter started. This work was started on the basis of a functional design specification similar to the one used for the steel shell units for previous study work. At the start of the study, two possible steam generator locations were considered, beside the core and below the core. Space envelope drawings were prepared to show the general configurations required for each of these locations. The beside-the-core space envelope was in the form of a cylinder with a large L/D ratio (5:1), whereas the below-the-core location utilized a cylinder with a much smaller L/D ratio (2:1). The following basic design ideas were developed:

1. U-tube reheater and superheater, spiral coil economizer-evaporator with the gas flowing in the radial direction.
2. Serpentine panel rh, sh, and ev/ec with the gas flowing vertically downward. Serpentine panels lie in a vertical plane with the steam/water flow predominantly in the horizontal direction.
3. Serpentine rh, sh, and ev/ec with the gas flowing horizontally. The serpentine panels lie in the horizontal plane with the steam water flow horizontal.
4. Serpentine rh, sh, and ev/ec with the gas flowing vertically downward. Steam water flow is alternately up and down.
5. Helical coil rh, sh, and ev/ec with the gas flowing vertically downward.

Fig. 54--Tube vibration test loop schematic

Fig. 55--Tube vibration test rig

6. Involute panel rh, sh, and ev/ec with the gas flowing vertically downward.
7. Spiral coil rh, sh, and ev/ec with the gas flowing in a radial direction.
8. Straight tube rh, sh, and ev/ec with the gas flowing vertically downward axially along the tubes. Tube bundles would be made up of a large number (19 or 37) tube modules.
9. Helical coil rh, sh, and ev/ec with the gas flowing vertically downward through many (~16) modules.

All of the basic heat exchanger designs have variations in the arrangement of elements with respect to the gas flow and may come in pairs (one unit containing reheater and other superheater). There are also several types of tube terminations with two basic geometric variations considered: (1) where the tubes are collected into headers inside the PCRVR with headers penetrating the PCRVR, and (2) where the tubes, either individually or in small groups, penetrate the PCRVR and are then collected in the headers. Most of the above design concepts are adaptable to both beside-the-core and under-the-core locations.

Most of the initial designs were based upon using two modules per steam generator loop with each loop equivalent to 165 Mw(e) plant output. Designs actively in progress for this arrangement are Nos. 1, 2, 5, and 6. These designs were pursued for the below-core location that was established in early August from a plant arrangement and steam generator standpoint. The current work is based upon utilizing a larger number of modules (8 per loop) with the plant arrangement modified to accept this concept.

High-temperature Design Basis

The service conditions in a power plant include periods of steady-state operation alternating with short transient periods when the load, the temperature, or both, change rapidly with time. Since in the high-temperature range the material strength is highly time dependent (creep), the effect of loading changes on the creep characteristics of the material is being investigated in detail. It was found that the effects of simultaneous or independent variation of the load and temperature can be estimated in terms of creep and fatigue life fractions.

To estimate the creep life fraction for varying load and temperature, stress-time-temperature relationships are required. It was found that the relationship between rupture time, temperature, and stress can be described with reasonable accuracy by parameter techniques based on the

assumption that complete failure is the result of a single, activated rate process expressed in the form

$$r = \text{rate} = A_1 e^{-(Q/T)}, \quad (1)$$

where A_1 is a stress dependent constant, Q is the temperature-dependent activation energy, and T is the absolute temperature. Integrating Eq. (1), the time can be expressed as

$$t = A_2 e^{(Q/T)}. \quad (2)$$

Several methods employing various assumptions with regard to the variation of the functionals A_1 , A_2 , and Q with stress were evaluated for cyclic application. The parameters associated with Larson-Miller, Dorn, and Manson-Haford were analyzed in detail.

As the first step, a large number of isothermal creep data were collected and, using these parameters, a detailed analysis was performed for 2-1/4 Cr-1 Mo, Type 304, and Type 316 stainless steel materials. The best correlation was found with the Manson-Haford parameter.

As the second step, the available cyclic temperature and cyclic stress creep data were analyzed using a simplified Manson-Haford parameter. The following procedure was used: The loading history of the test was divided into time elements of constant conditions (stress or temperature), and the corresponding rupture life was computed using the stress-temperature master curve. The life fraction was computed as the ratio of the time at load and the rupture time for constant load. For two-step test data, it was convenient to express the reduced rupture life in the following form:

$$t' = \frac{t_1}{t_2} \frac{R + 1}{R + t_1},$$

where t_1 and t_2 are the rupture times for the higher and lower load levels, and R is the ratio of the durations of the extreme values of stress or temperature. The results show that the analysis provides a fair estimate of the effective rupture life for simple cyclic conditions.

Similar equations for the creep-strain accumulation under fatigue conditions lead to unacceptable scatter in the results. The fundamental reason for this discrepancy is that partial damage--as opposed to complete failure--due to either fatigue or creep is not a directly measurable or observable quantity. Even in a simple, single-stress-level fatigue test, the damage in the specimen occurs in some undertermined fashion as more and more cycles are applied. The accumulation of partial damage

continues until the specimen breaks, when the damage is complete, 100%. Similarly, in a constant-stress-level creep test, partial creep damage accumulates until rupture occurs, and the rupture time defines 100% damage. In both cases the conditions at failure are well determined: The stress level, number of cycles, strain rate, total elapsed time, etc., can be measured and controlled in the experiments. The relative success of the analytical prediction of total life can be attributed in part to the accurate knowledge of the conditions at failure and in part to the fact that in addition to the damage processes, recovery effects, such as strain hardening, creep recovery, stress relaxation, etc., are also activated. These effects tend to compensate for damage, and as a result, the single activated rate equation describes adequately the average conditions leading to failure. However, in estimating the instantaneous creep rate and accumulated creep strain before complete failure, the constant rate equation is applicable only if the variation in stress and temperature is such that the steady-state region dominates the creep deformation. If the changes in the environment are rapid, the steady-state equations become very inaccurate. For this type of load and temperature variation, the single activation model is not applicable. A new approach based on an activation series is therefore being investigated. Using the general Arrhenius rate equation, the multiple activated process can be expressed as

$$r = A_1 e^{-(Q_1/T)} + A_2 e^{-(Q_2/T)} + A_3 e^{-(Q_3/T)} + \dots,$$

representing the sum of the individual processes, activated simultaneously but reaching maximum intensity at different times within the loading cycle. This approach is adaptable to describe both the primary and secondary creep, and currently a convenient method for evaluating the constants A_n and Q_n is being studied.

TASK V

COATED PARTICLE DEVELOPMENT

Coating development studies in this task are directed primarily toward effecting further economic and structural improvements in the reference BISO coating. These studies are intended to determine the optimum materials specifications and process parameters for preparing each of the coating layers.

Emphasis is currently being placed on the study of high-density isotropic coatings. Buffer coatings are also being studied, including development of additional methods for characterizing this layer; these techniques will complement those previously developed for isotropic coatings. Coated particle samples are also being prepared and evaluated for out-of-pile and irradiation testing under the HTGR Public Service Reactor program.

HIGH-DENSITY ISOTROPIC COATINGS

An intensive study of the high-density, isotropic PyC coating is being conducted. The results of earlier irradiation tests of various pyrolytic carbon structures indicate that this structure should have the greatest radiation stability of the structures tested to date. The object of the study is an understanding of the high-density isotropic coating region and an optimization of the coating process necessary to obtain the high-density structure.

Part of the study includes an investigation of the effects of variations in coating parameters on the PyC structure. A systematic series of coating runs has been carried out at 2000° and 2200°C with variations in methane concentrations from 5% to 20%, total gas flows from 6000 to 10,000 cm³/min, and bed surface areas from 800 to 2500 cm². Both temperature series have now been completed.

BUFFER COATING STUDIES

The flash method is being adapted to measuring the thermal diffusivity of low-density buffer coatings. In this technique, one surface of a

sample with slab geometry is irradiated with a pulse of energy from an optical source and the subsequent temperature history is monitored at the opposite surface. From the shape of this transient, the thermal diffusivity of the sample material may be deduced provided the heat-conduction equation has been solved for the boundary conditions of the experiment.

The computer program that has been developed for the reduction of data from the flash method for composite samples has been completely debugged and successfully tested. The method permits the computation of thermal diffusivity and conductivity in one of the layers of a two-layer sample if the corresponding values are known for the second layer. If the thermal diffusivities of both layers are known beforehand, the method affords qualitative information on the magnitude of the thermal contact resistance at the interface between the two layers. Through the introduction of an empirical function accurately describing the actual pulse shape of the xenon flash tube employed, the effect of finite pulse length is automatically taken into account, both for the two-layer and the single-layer computations. This feature permits the use of thin samples, thus making it possible to employ the flash method at elevated temperatures for measurements on both one- and two-layer samples without the necessity of correcting for heat losses.

The computational method may be briefly described as follows. Points from the experimental oscillograph trace are read into the computer, and by means of an iterative procedure, the theoretical transient is passed through the points in a least-squares sense. After certain convergence criteria are met, the adjusted parameters are converted to values of thermal diffusivity and conductivity which are then read out onto a specially-devised output format that serves to record not only the computed results, but also most of the pertinent experimental information as well.

Flash-method oscilloscope traces have been obtained for a series of buffers deposited on ATJ graphite substrates. These buffers were deposited at 1100°C from acetylene-carrier gas mixtures varying in concentration from 50% to 90% acetylene. Traces have been obtained for samples with varying thickness of the buffer layer on the graphite substrate, and comparison of the computed thermal diffusivities of the buffers with the values obtained using samples with the graphite layer removed will provide a qualitative indication of the magnitude of the thermal contact resistance between two layers. An attempt will be made to correlate the information so gained with the results of a metallographic examination of the interfaces.

COATED PARTICLE PREPARATION

Irradiation testing of promising types of reference coated particles is being carried out in connection with the Public Service Reactor program. Preparation of coated particles for these irradiations is being done as part of this task. These irradiation tests are intended to guide the process development of reference BISO coatings with optimum radiation stability and retentivity, and lowest possible fabrication costs. In addition, particles are prepared for various types of out-of-pile tests.

Dounreay Fast-flux Experiment

A series of 15 PyC structures have been selected for irradiation in the Dounreay Fast Reactor (DFR). Included are nine high-density structures obtained in the recent systematic studies (see "High-density Isotropic Coatings" above), two additional high-density structures which have been deposited over a reference buffer layer, and four control structures that have been previously irradiated. For each structure, unrestrained PyC strips, PyC-coated graphite disks, and coated UC_2 (depleted) particles will be irradiated.

Two capsules will be used in the DFR irradiation. one in a core position and the other in a blanket position. The core capsule will operate at 1000° to $1200^{\circ}C$ and experience an integrated flux of $\sim 1 \times 10^{22}$ nvt. The blanket capsule will operate at 1000° to $1200^{\circ}C$ and experience an integrated flux of $\sim 4 \times 10^{21}$ nvt. Both capsules will contain all of the structures, however, the blanket capsule will not contain any of the PyC-coated graphite disks owing to the limited number available.

All the structures for the DFR irradiation have been prepared and are now being characterized and prepared for inclusion in the capsules.

Coated Particles for Packing Studies

A number of coating runs were made to produce some reference coated particles for loose particle packing studies (conducted on the Public Service Reactor program).

COATING EVALUATION TECHNIQUES

Characterization of carbon coatings has been proved to yield essential parameters for predicting and understanding the performance of coated particles under irradiation and thermal testing. This experimental work is intended to improve and add to the existing techniques used in determining those characteristics.

Coating Density Determination by X-Ray Absorption

A new technique for determining densities of PyC coatings on fuel particles has been developed. This technique measures density variations by the difference in X-ray absorption of the material. A large cross section (about 150 μ thick) is removed from a coating disk, placed on a photographic plate, and exposed to an X-ray beam (5 kv and 2 ma) for about 1 hr. The resultant plate is then scanned with a densitometer.

The densitometer readings show a good linear correlation with the logarithms of densities determined by the standard sink-float technique. However, some variation was observed, and much of this is believed due to nonhomogeneity in the sample (density variations up to $\pm 10\%$ have been observed across a single buffer coating). Typical data (for coating thicknesses $> 60\mu$) are shown in Fig. 56.

This process was limited, however, in that coatings less than 50 μ thick could not be used because of densitometer inadequacy. In order to increase the sensitivity of the densitometer to thinner coatings, enlargements of the original radiographs were made. These enlargements could then be read by the densitometer. The densitometer readings of the enlargements also showed a good linear correlation with the logarithms of densities determined by the sink-float technique (see Fig. 57). Using this enlargement technique, densities of coatings of nearly any thickness can be determined by the X-ray absorption method.

Current work is directed toward further understanding of the inverse density-coating thickness relationship in this method. The penetration characteristics of the X-ray beam are being investigated with a variable-thickness graphite wedge. The wedge is made of PyC slivers about 10 μ thick that can be stacked to give various thickness layers of a known density material.

In summary, the determination of coating density by X-ray absorption appears promising. The following observations should be made:

1. The X-ray method is currently the only method that can be used for buffer coatings. These fragile coatings need not be removed from the disks, and the presence of barrier layers over or under the buffer does not interfere with the measurement.
2. The X-ray method is faster (and thus cheaper) than the sink-float method for nonbuffer coatings.
3. The X-ray shows density variations in the coatings.

Fig. 56--Densitometer readings of X-ray films of PyC-coatings compared with densities previously determined by sink-float technique

Fig. 57--Densities determined by X-ray absorption compared with those determined by standard sink-float technique: (a) original radiographic plate, (b) enlargement of original plate

Coating Density Determination by the Electron Microprobe

The electron microprobe is also being investigated as a method for determining coating densities directly on coated particles rather than on coatings applied to graphite disks. In this method, the coating density is determined by relating the voltage required to penetrate the PyC and excite the substrate to the resultant measured emission of gamma rays. This method requires that the coating be fairly uniform and the particle near spherical.

An initial attempt to utilize the microprobe failed because the PyC coatings were too thick. A new batch of particles that have thinner coatings and are rounder are being examined. The results of this trial should determine the feasibility of the proposed method.

Coating Thickness Determination

The use of fast-developing film is being investigated as a coating control tool. The usual technique involves a 2-min exposure to 20 kv X-rays and 7-min developing, which produces a permanent high-quality plate. The first test using fast-developing film involved a 10-sec exposure and 2-min developing and washing, from which an acceptable coating thickness number could be determined. This technique can yield a coating thickness result in about half the present time. The film will now be tested at other voltages.