40-MW(E) PROTOTYPE HIGH-TEMPERATURE GAS-COOLED REACTOR RESEARCH AND DEVELOPMENT PROGRAM

Summary Report for the Period January 1, 1959—
December 31, 1959 and Quarterly Progress Report
for the Period October 1, 1959—December 31, 1959

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General Atomic Division
General Dynamics Corporation
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GENERAL ATOMIC

DIVISION OF GENERAL DYNAMICS CORPORATION

JOHN JAY HOPKINS LABORATORY
FOR PURE AND APPLIED SCIENCE

SAN DIEGO 12, CALIFORNIA

GA-1235

40-MW(E) PROTOTYPE HIGH-TEMPERATURE
GAS-COOLED REACTOR
RESEARCH AND DEVELOPMENT PROGRAM

SUMMARY REPORT
FOR THE PERIOD
JANUARY 1, 1959 — DECEMBER 31, 1959
and
QUARTERLY PROGRESS REPORT
FOR THE PERIOD
OCTOBER 1, 1959 — DECEMBER 31, 1959

Contract AT(04-3)-314
U. S. ATOMIC ENERGY COMMISSION

PREFACE

Under Contract No. AT(04-3)-314, General Dynamics Corporation as Contractor to the U. S. Atomic Energy Commission has agreed to perform the research and development necessary to the design of the high-temperature, gas-cooled, graphite-moderated reactor known as the HTGR. This research and development effort is directed specifically toward the construction of a 40-Mw (electrical) prototype plant, as described in Appendix A of the Contract. Under separate contractual arrangement, the 40-Mw(e) prototype plant will be constructed for the Philadelphia Electric Company. The General Atomic Division of General Dynamics Corporation is responsible for furnishing the nuclear steam supply system for this prototype plant. The Bechtel Corporation is prime contractor for construction of the plant. Completion of the plant is scheduled for the end of 1963. The Philadelphia Electric Company and 52 other utility companies, comprising the High Temperature Reactor Development Associates (HTRDA), are sponsors of the project.

Under the terms of the research and development Contract, the Contractor is required to furnish such progress reports as the Atomic Energy Commission may reasonably require. Since this is the first quarterly report prepared under the Contract, we have endeavored not only to cover work done during this quarter but also to summarize work done between January 1 and September 30, 1959, and to give sufficient background for the report to be understood without reference to other documents. It is to be noted that a brief account of the HTGR design concept has appeared in the open literature.* The conceptual design is described in somewhat more detail in General Atomic Report GA-593,

*P. Fortescue, D. Nicoll, C. Rickard, and D. Rose, "HTGR-- Underlying Principles and Design," Nucleonics, Vol. 18, No. 1, 1960, p. 86.

dated November, 1958. The more important design parameters are presented in the Summary of this report.

The text of this report has been written to follow, in general, the task divisions of the research and development effort as set forth in Appendix B of the Contract. The titles of these tasks and their objectives are outlined here.

Task I. Develop Materials for the Reactor Core

The objectives of this task are to develop materials and generally specify fabrication procedures for the components of the reactor core. The items include metal-clad fuel elements (as a first step), graphite-clad elements (as the ultimate objective), control rods, and other core components. Each core component must satisfy the geometric and operating conditions set by the design of the reactor core, must withstand static and dynamic stresses generated as a result of reactor operation, and must have a satisfactory life with respect to prolonged exposure to radiation and to interaction with coolant impurities and with other parts of the reactor.

Task II. Develop Reactor Core and Reflector

This task is to develop a design of the reactor core and reflector to the extent necessary to provide reasonable assurance of satisfactory operation in the HTGR, in terms of (a) materials and composition; (b) physical dimensions of core and reflector structures and assemblies, including fuel-moderator and control elements; (c) design tolerances; and (d) design operating conditions of temperature, pressure, and burnup. The reactor core must meet certain requirements imposed by the over-all performance specifications. These requirements include provision of heat-transfer passages to raise the specified gas flow to the proper temperature with tolerable pressure and pressure drop, and adequate core burnup lifetime with reasonable control and fuel inventory. The design must meet

the above requirements and must be satisfactory in terms of ease of fabrication, assembly, and maintenance, feasibility of core support and pressure-vessel design, pressure level required for heat transport, and estimated cost.

Task III. Experimental Reactor Physics Measurement to Establish Nuclear Characteristics of the Reactor

This task is to obtain experimental values for the critical mass, control-rod worth at different positions and degrees of insertion, flux variations over the core, and other data necessary to establish the nuclear performance of the reactor core.

Task IV. Develop Reactor Equipment

This task is to develop and specify a conceptual design of the reactor vessel, thermal shield, core supports, flow partitions, vessel closures, control-rod drives, fuel-handling and fuel-transfer mechanisms, and other systems and equipment directly related to the reactor. This task includes determination of the operating requirements; environmental, space, and weight conditions; the conceptual design of the equipment; work leading to specification of materials of construction; and, also, the construction and testing of prototype units where necessary.

Task V. Develop Primary-loop Equipment and Reactor Plant Layout

This task is to develop: (a) The conceptual design of those components other than the reactor which are in the primary loop. Included are the primary-loop piping, steam generator, helium circulators, valves, and subsystems for operation of these primary-loop components. (b) The over-all reactor plant layout, including the primary coolant loop, major components, and auxiliary systems in sufficient detail to define spatial location of components for shielding, installation, servicing, and freedom from excessive thermally induced stresses and reaction levels.

Task VI. Develop Reactor Plant Auxiliary Systems

This task is to establish the preliminary specifications and design criteria and to develop the conceptual design of those systems and components thereof which are necessary to maintain and ensure reactor operation. Included are the helium purification and storage systems, fission-product trapping system, and shutdown and auxiliary cooling system.

Task VII. Develop Conceptual Design of Control Systems and Instrumentation

This task is to solve those special development problems in connection with instrumentation and control of the reactor, primary loop, and related subsystems, and to arrive at a conceptual design of the necessary system control circuit and its instrumentation. The power level of the reactor and steam-generating system must be controlled in such a way as to prevent temperatures and temperature gradients from exceeding maximum allowable values, to limit rates of nuclear power change, and, in general, to assure proper operation of the system and its components.

Task VIII. Over-all Plant Analysis

This task is to carry out the design studies and any necessary supporting experimental work to determine the essential operational features of the reactor plant.

SUMMARY

For reference purposes, there is given below a brief description of the 40-Mw (electrical) prototype HTGR plant design. In general, this description comes from the conceptual design study presented in GA-593, but in a few cases more recent data have been included to bring the description up to date. It is expected, of course, that the research and development program presently under way will result in changes and improvements in this design.

THE 40-MW(E) PROTOTYPE HTGR

The HTGR prototype plant is being designed to produce steam at 1450 psi and 1000°F and to have a net capacity of 40 Mw(e). The reactor data and operating conditions for the graphite-clad core are summarized on the following page.

The plant will be a prototype for larger power plants. The fuel temperatures and gas pressures will be approximately the same as those required for the larger plants. Also, the prototype plant equipment will be sufficiently large so that it can be utilized as the basis for the design of larger equipment needed for commercial plants. These features of the prototype will permit a sound extrapolation of the technical and economic data to larger plants.

Reactor

The general arrangement of the reactor is shown in Fig. 1. The reactor core is made up of 855 cylindrical elements arranged in a closely pitched equilateral triangular array. Approximately 40 of these elements are control rods. The fuel elements are oriented vertically and supported by a bottom grid plate. The coolant gas passes through the tricuspid-shaped passages formed by the elements. The bottom and top reflectors, each

TECHNICAL DATA FOR THE HTGR

Operating Conditions

Coolant	Helium
Pressure, atm	20
Inlet temperature, °F	660
Outlet temperature, °F	1380
Maximum fuel-surface temperature (approximate), °F	2300
Steam temperature, °F	1000
Steam pressure, psia	1450
Net thermal efficiency, %	34.8
Reactor thermal output, Mw	115
Net electrical power, Mw	40

Reactor Parameters

Fissile investment (U^{235}), kg	190
Thorium investment, metric tons	1.19
Core diameter, ft	9.0
Core length, ft	7.5
Core coolant voidage, %	12.8
Fuel-element diameter, in.	3.50
Number of fuel elements (approximate)	810
Reflector thickness, ft	2
Core graphite weight, metric tons	19.5
Reflector graphite weight, metric tons	47.7
Pressure-vessel diameter (inside), ft	14
Pressure-vessel height, ft	34
Core graphite-to-uranium ratio	1900
Initial conversion ratio	0.45
Average conversion ratio	0.46
k_{eff} , cold, clean	1.293
k_{eff} , hot, clean	1.205
Average thermal neutron flux, beginning of life	4.1×10^{13}
Burnup	
Fissions per original fissile atom	0.63
Mw-days (heat) per metric ton of $U^{235} + Th^{232}$	75,000
Core life (at 80% load factor), yr	3

Fig. 1--Isometric of high-temperature, gas-cooled, graphite-moderated reactor

2 ft thick, are made an integral part of the fuel elements. The side reflector, which extends the full height of the fuel elements, is also 2 ft thick and is made up of a row of dummy graphite elements and a concentric ring which encloses the core. The core and reflector are surrounded by a steel thermal shield.

Incoming coolant gas passes through the coolant passages formed by the thermal shield and the pressure-vessel wall and through the side reflector before passing up through the reactor core. A shroud above the core serves as a thermal barrier between the outgoing hot helium and the incoming cooler helium and is connected to the coolant outlet from the reactor.

The fuel element consists of a graphite sleeve containing the fuel compacts and both the top and bottom reflectors. The element is a 12-ft-long, 3.5-in.-diameter cylinder. The fuel compact consists of fuel (U^{235}) and fertile material (Th^{232}) in the form of carbides dispersed in a graphite matrix. The composition of the fuel compacts is approximately 1 atom of U^{235} to 10 atoms of thorium to 700 atoms of carbon.

Elimination of metal cladding for fuel elements eases the cooling problem, improves neutron economy, and allows high-temperature operation, but it introduces the problem of fission-product diffusion. This problem would probably be associated with all really-high-temperature reactor systems, for at high temperature any cladding system--metallic or otherwise--must be regarded as being to some extent permeable to some fission products.

In the HTGR, the approach is not to attempt complete retention of all the fission products within the fuel elements, but rather to minimize the loss by the use of relatively impervious graphite cladding and to control those products which do escape in such a way as to avoid the major undesirable consequences of their release into the primary circuit.

In particular, the fission-product control system will function

1. To limit the activity build-up in primary-loop equipment from

deposited fission-product solids to a level sufficiently low that fairly direct maintenance of the equipment can be performed.

2. To limit the gaseous fission-product activity level in the primary helium coolant (a) so that accidental release of the coolant into the secondary containment will not result in a serious operational difficulty or hazard and (b) so that continuous release of activity to the site surroundings as a result of normal coolant leakage does not exceed permissible concentrations.

Roughly one-half of the fission-product elements formed can be expected to remain fixed in the fuel compacts, either because of their own nonvolatility or because of their formation of stable, nonvolatile carbides. The fuel compacts are contained in highly impermeable graphite to minimize and/or delay the escape of the remaining fission products. Those which do escape are collected in an external fission-product trapping system by means of a helium purge stream flowing between the graphite-contained fuel and the outer graphite sleeve. Connection of the purge stream from the fuel elements to the external trapping system is made at the bottom support point of the elements. From the trap, the purified helium purge stream returns to the primary circuit. The principal advantage of this system over a bypass system operating on the primary coolant stream is that the chemically reactive or condensable fission products are prevented from plating out in the primary circuit prior to reaching the trap. Furthermore, the fission products are relatively undiluted when they reach the trap, and thus trapping difficulties are decreased.

A core consisting of metal-clad graphite fuel elements may be used initially for the start-up and testing of the prototype plant. During the initial operational testing phase there would be a gradual transition to the graphite-clad elements. The reactor plant is designed to accommodate these core changes. The purpose of this approach would be that it might permit earlier operation and testing of the plant and components. Since

metal cladding imposes a temperature limitation on the fuel surface, some sacrifice in steam conditions and thermal output would occur during the transition period. However, a steam temperature of around 850°F and a pressure of around 850 psi, which can be attained with the metal-clad core, are still high. The corresponding plant rating could still be kept in the range of 25,000 to 30,000 kw.

The control rods are operated from beneath the reactor; they are inserted upward into the core to reduce reactivity. The top of the reactor is thus clear for ease of fuel-element handling. To provide fail-safe operation, the control-rod drives are designed so that either pneumatic pressure or hydraulic pressure acting alone will scram a rod. Each rod may be used either as a shim or safety, and all rods scram regardless of their assigned functions.

The control rod consists of B_4C dispersed uniformly in a graphite matrix in the form of hollow cylinders. A number of these cylindrical sections supported on a central rod form the control rod. The support rod is flexibly jointed to provide for any misalignment between the control rod and its actuating mechanism. Each control rod moves within a graphite guide tube similar in appearance to a fuel element except that the lower section extends into a recess in the grid plate.

The fuel is changed with the reactor shut down and with the coolant pressure reduced to slightly below atmospheric. A single fuel-transfer machine, retractable through a port in the top of the reactor vessel, serves the entire core. The fuel-transfer machine and pick-up mechanism may be seen in Fig. 1. This transfer machine removes a single fuel element from the core and transfers it to a charge-discharge tube in the side reflector. The element is then lowered by a fuel unloading machine, located below the reactor, and deposited within a transfer coffin. The element is sealed in a loosely fitting metal can by a mechanism contained within the transfer coffin; the coffin is then transferred to a position over a spent-fuel-element storage pit, where the element is discharged and

moved to a storage position under water.

New fuel elements are loaded into the reactor in much the same manner, though in reverse order.

Primary Coolant System

A flow diagram of the primary-coolant circuit and its auxiliaries is shown in Fig. 2. The duct between the reactor vessel and the steam generator is made up of concentric pipes. The hot gas (1380°F) flows through the inner pipe to the steam generator and the cooled gas (660°F) returns to the reactor via the annular duct. Regenerative heating of the gas is reduced to a small effect by internal insulation of the inner duct. An important advantage of the concentric ducts is that the outer, pressure-bearing pipe is cool. In addition, the thermal stress problem at the penetrations of the reactor vessel and steam generator is considerably reduced by the concentric pipe.

In passing through the steam generator, the hot coolant gives up its heat to make 367,000 lb/hr of 1000°F , 1450-psi superheated steam and leaves the generator at 630°F . The coolant then passes through the helium circulators and an outer shell within the steam generator and returns to the reactor vessel through the outer annulus of the concentric duct. Since concentric flow of the hot and cold helium coolant is used throughout the plant, all pressure-containing surfaces, including the reactor vessel, are exposed only to the low-temperature (660°F) coolant. Hence, carbon steel may be used for most of the structural parts.

Two circulators are used in series to circulate helium through the coolant loop. These are canned-motor, single-stage, centrifugal compressors with overhung impellers. The compressors circulate 80,000 cfm of helium (480,000 lb/hr) at 290 psia, with an inlet temperature of 633°F and a pressure rise of 5 psi in each (a total of 10 psi) when operating at design speed of 3475 rpm. The drive motors are rated at 2500 hp with a 2400-volt, three-phase, 60-cycle power supply. To provide a

Fig. 2--Flow diagram of 40-Mw(e) high-temperature, gas-cooled, graphite-moderated reactor

variable helium flow rate, the compressor drive motors are operated from a variable-frequency power supply.

The compressor-motor assembly is completely enclosed within a sealed can to prevent helium leakage. Cleaned helium is fed to a labyrinth-type shaft seal to prevent contaminated helium from entering the motor compartment.

Isolation of the steam generator from the reactor is provided by a specially designed concentric gate valve installed in the concentric piping run. This valve is required only to stop flow through the concentric ducts by sealing tightly against the outer, cooler duct, since some flow between the inner pipe and the outer annulus on either side of the isolation point is of no concern. Consequently, the valve body service is limited to that of roughly 660°F operation. Ports in a follower plate permit separated flow of inner and outer gas streams when the valve is open.

Approximately 10,000 ft³ of helium at system pressure are contained in the primary coolant system, which consists of the reactor, steam generator, circulators, and associated piping. Helium storage tanks are provided in the containment vessel. A vacuum pump and compressor are provided to transfer helium between the primary and the storage systems.

Continuous bypass purification of the helium in the primary system is provided to maintain minimum equilibrium impurity levels. It is anticipated that the total impurity levels of less than 10 ppm will be necessitated by mass-transfer considerations. Both cold adsorption and hot, chemically reactive traps are being considered for removal of oxygen, nitrogen, water vapor, and other gaseous impurities. A portion of the cleaned helium is utilized for sealing valves, circulator cans, and other buffer-sealed enclosures.

An auxiliary cooling system, sized for the extended period of low-level heat removal following reactor shutdown, is provided in parallel with the primary cooling system. Following normal reactor shutdown, the primary-circuit helium circulators will continue to operate until the

afterheat decays to a level at which the auxiliary system is capable of removing the heat load by itself.

THE RESEARCH AND DEVELOPMENT PROGRAM

The research and development program during this quarterly period is presented briefly here. The various sections of the report cover these topics in detail.

The prospects for development of the graphite-clad fuel element in time for use in the first loading of the reactor have been improved by important advances in methods of fabrication and testing of both fuel compacts and graphite sleeves. The hot-pressing process for making fuel compacts has been used successfully to make full-size compacts with a uniform distribution of $\text{ThC}_2\text{-UC}_2$ particles. Full-diameter, 4-ft-long graphite sleeves with a permeation constant of $10^{-7} \text{ cm}^2/\text{sec}$ have been fabricated by an outside supplier.

Three additional irradiation capsules have been fabricated and inserted in a test reactor to determine fuel compact and sleeve performance under HTGR conditions of irradiation and temperature. Two of these have run satisfactorily for the scheduled time of operation.

A scope design study of the in-pile loop that will be used to evaluate the full-diameter graphite-clad element has been completed.

Experiments to determine the extent of fuel migration within the element have been undertaken. Preliminary results indicate that the central fuel-element temperatures must not exceed 2300°C for routine operation.

An important start has been made in developing an understanding of how to treat the neutron thermalization process in high-temperature graphite reactors.

Analytical techniques for calculating the thermal neutron spectra in poisoned graphite media have been developed and programmed for the IBM 704 computer.

The experimental technique of measuring neutron spectra by using a pulsed linear electron accelerator has been demonstrated by measurements made with boron-loaded graphite.

A mockup of a small portion of the reactor core has been constructed and operated to determine the local heat-transfer coefficients and pressure drop in the tricuspid-shaped coolant passage. Initial results indicate that the variation of the heat-transfer coefficient around the circumference of the element is less than expected.

Studies have been started of the transient temperatures and stresses developed in the pressure vessel as a result of load changes or a scram.

A detailed study of several types of steam generator for use in the nuclear steam supply system has been completed. A design incorporating a steam drum has been selected for further study.

Preliminary flow diagrams have been completed for the helium-purification and fission-product trapping systems. Adsorption isobars for selected fission products in activated carbon have been measured and will be used in the detailed design of the trapping system.

Detailed planning of the experimental reactor physics program has been initiated. A safeguards report has been written for the critical experiments. Construction of the building to house the critical assembly is under way and part of the experimental equipment has been ordered.

Progress has been made in the identification of the principal safeguards problems for this type of reactor, and a preliminary safety analysis of the plant has been completed.

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TASK I. DEVELOPMENT OF MATERIALS FOR REACTOR CORE

The objectives of this task are to develop the materials and to specify the fabrication procedures for components of the reactor core. The materials under consideration include those for fuel and control-rod bodies, the cans and sleeves for these bodies, and moderating materials.

Development of these materials includes tests for their compatibility and physical stability in different environments, especially gas-moderator interactions; temperature effects, both dynamic and static; and irradiation effects in connection with the above parameters.

The HTGR reactor operating conditions are sufficiently advanced to preclude the use of many commonly used reactor fuel and construction materials. Graphite is one of the few acceptable materials because of its strength at elevated temperatures, its availability, its comparatively low cost, and its good nuclear properties. It will be used as a matrix for fuel bodies, as a moderator, and as a fission-product container for fuel bodies. Recently, advances in the production of graphite of low permeability make the use of this material most attractive.

Low-permeability graphites are being developed by industrial concerns, such as Hawker Siddeley Nuclear Power Company Ltd., National Carbon Company, and Great Lakes Carbon Corporation. At present, there are available graphite tubes whose permeability would make them acceptable as fuel-compact containers. General Atomic is also developing techniques for producing low-permeability graphite involving impregnation of small-pore base graphite followed by regraphitization. Also, coatings, binders, and different types of graphite are being tested for their usefulness in producing low-permeability graphite.

Industrial concerns have been requested to submit samples of fuel and control-rod materials for our evaluation. General Atomic also is conducting a program for the development of these materials in which

different pressing techniques, binders, fuel and mixture ratios, and fuel-body sizes are being tried. The best combination of these methods will then be tested in different environments.

The tests to be conducted are primarily divided into two categories--out-of-pile and in-pile. The out-of-pile tests include full-scale mechanical strength, thermal shock, temperature-cycling effects, and fuel migration. The in-pile tests include the irradiation of subsized fuel compacts and samples of graphite container materials, as well as full-scale compacts and full-diameter graphite sleeves and containers. Examinations will be made after irradiation, including tests for materials compatibility, fuel diffusion, physical stability, thermal conductivity, and permeability changes.

As was pointed out in the Summary, it may be necessary to use a graphite-base fuel element clad with metal for the first loading of the HTGR. Consequently, during this quarter, work has continued on the development of a metal-clad element. Previous work on this program showed that Type 430 stainless steel carburizes above 1500°F, whereas Monel will resist carburization in contact with graphite up to 1850°F; as a result, work has been concentrated on Monel. Thermal-cycling tests on full-diameter elements and small-scale irradiation tests have been carried out.

1.1. FUEL COMPACTS

The fuel compacts for the HTGR will consist of thorium-uranium carbide particles dispersed in a graphite matrix. The compacts will be 2.25 in. in diameter and 4.5 in. long, with the fuel dispersed in the outer 3/8 in. of the compact. The central core of unfueled graphite may contain the burnable poison (boron).

The carbides of uranium and thorium are to be employed as the fissile-fertile phase because of the compatibility of these compounds with graphite at the high operating temperatures of the HTGR. Since high

burnups are required, the fissile and fertile phases will be combined, thereby diluting the fuel with fertile material and spreading the radiation damage over a larger volume.

The metal (thorium and uranium) concentration in the annular fueled region will not exceed 30 wt-%, and the average particle size of the ThC_2 - UC_2 will be approximately $150\ \mu$. This concentration leaves $150\ \mu$ of carbon between adjacent fuel particles (assuming uniform distribution), thereby reducing radiation damage to the matrix caused by recoiling fission products.

During the past few months, techniques have been developed for producing fuel compacts in which ThC_2 - UC_2 particles are uniformly dispersed in a graphite matrix, and a large number of such compacts 1 in. in diameter by 2 in. long have been prepared for both out-of-pile and in-pile testing. Full-size fuel compacts of the annular design have recently been prepared as part of the fabrication development program. General Atomic's process for hot-pressing graphite fuel compacts has been improved to the point where temperatures of 4300°F at pressures of 10,000 psi are obtained. This has been accomplished both in the laboratory and on pilot-plant scale with full-size compacts.

The extreme reactivity of ThC_2 - UC_2 with atmospheric humidity necessitates tedious handling procedures in forming the fuel compacts. Studies to alleviate this problem are meeting with success, as silicides of thorium and uranium have been developed which appear to be nonreactive with air. These silicides are readily prepared by arc-melting and crushing to produce particles suitable for incorporation into the fuel compacts. During the final firing of the compact, the silicides are converted to carbides in situ. X-ray diffraction studies have been used to follow the course of these reactions. Apparatus is being constructed to determine quantitative rates of reaction of fuel materials with air and moisture.

Experiments are in progress to determine the diffusion and migration rates of the thorium and uranium carbides in graphite at high

temperatures (2500° to 4000° F), and some preliminary results have been obtained. Metallographic facilities are being set up to permit complete microscopic studies of fuel particles and fuel compacts in a highly purified inert atmosphere.

A technique for measuring the thermal conductivity of fuel compacts at temperatures to 4000° F has been developed, and some measurements have been made.

Fuel-materials studies for the next year will be aimed at developing an understanding of the chemistry involved in the formation of thorium and uranium compounds and in their reactions with graphite. From these data, preliminary process specifications can be established. Continued development of hot-pressing compacting techniques should lead to a process suitable for production.

A large number of full- and half-scale fuel compacts will be prepared for irradiation and other testing. Diffusion and migration studies, combined with thermal-conductivity measurements, will aid in the establishment of an upper operating temperature for the fuel compacts.

Fissile-Fertile Phase Studies

An experimental program for the study and identification of various thorium and/or uranium compounds and their reaction products with graphite has been initiated. X-ray diffraction and metallographic techniques are being employed for identification of the compounds and their reaction products, and arc-melting, annealing, vacuum-sintering, and similar techniques are being employed for materials preparation.

Metal Carbides. A comparison was made between two laboratory techniques for preparing uranium and thorium carbides: (1) sintering UO_2 or ThO_2 with graphite and (2) arc-melting the pure metals plus carbon. X-ray diffraction results are given in Table 1.1.

Carbides from Oxides. A series of compacts of solid-solution Th-U-O_2 ($\text{Th:U} = 5:2$), 75 wt-%, plus graphite and pitch, 25 wt-%, were

sintered for various times and at various temperatures to determine the conditions necessary to convert the oxide compounds to the carbides. The results, as revealed by X-ray data, are given in Table 1.2.

Table 1.1
COMPARISON OF ARC-MELTED AND SINTERED CARBIDES

Carbide	Arc-melted	Sintered
UC	Obtained pure UC	Obtained pure UC (1 hr, 1850°C)
UC ₂	Trace of UC	Considerable UC (1 hr, 2000°C)
ThC	No detectable oxide	Considerable oxide, incomplete conversion (1 hr, 1650°C)
ThC ₂	No detectable oxide, considerable ThC	No detectable oxide, specimen in excellent condition

Table 1.2
CONVERSION OF OXIDE COMPOUNDS TO CARBIDES

Sintering Time (hr)	Sintering Temp. (°C)	Results
2	1800	Oxide present
4	1800	Oxide present
2	2000	Oxide present
4	2000	No oxide
2	2200	No oxide
4	2200	No oxide

Thorium-Uranium Silicides. Uranium silicides containing 7.4% to 18.5% silicon, and thorium-uranium silicides containing 6.4% to 20% silicon and having a Th:U ratio of 5:2 were prepared by arc-melting and were ground with ease in a blender, using a tungsten carbide vial and ball

to minimize contamination. (Uranium-silicon alloys containing 7.4% to 18.5% silicon are easily crushed. However, U_3Si (3.8% silicon) is too malleable to be crushed to powder.) Three mesh sizes of each composition were obtained for fuel-compact studies, and X-ray patterns of -300 mesh powder from each specimen were recorded on film. The X-ray diffraction results are summarized in Table 1.3. These silicides, even though finely ground, are stable in air.

Table 1.3
X-RAY ANALYSES OF THORIUM-URANIUM SILICIDES^a

Silicon Content (%)	Results
6.4 } 7.4 }	Patterns appeared identical, revealing M_3Si_2 -type compound. No oxide
10	Mixture of M_3Si_2 - and MSi -type compounds. No oxide
15	MSi_2 -type compound, plus trace of another compound for which no comparison available (probably M_2Si_3 type)
20	MSi_2 -type compound, trace of unidentified compound (probably MSi_3 -type). No oxide

NOTE: Visual comparisons were made with diffracted patterns of thorium silicides as recorded by Jacobson, Freeman, Tharp, and Searcy, J. Am. Chem. Soc. 78, 4850 (1956).

^aTh:U = 5:2.

Metallographic studies are also in progress on buttons of each of the above compositions. From these studies we hope to learn the phases present at various silicon concentrations. X-ray diffraction and metallographic investigations are also being performed on the powders after they

have been compacted with graphite and converted to carbide, in order to learn the carbide phases formed and the temperatures required for complete conversion to carbide.

Carbides from Silicides. Thorium-uranium silicides prepared by arc-melting procedures and varying in composition from 6.4% to 20% silicon were crushed, sieved to three particle sizes (250 to 500 μ , 110 to 250 μ , and < 50 μ), and then mixed with graphite and pitch and sintered. X-ray patterns have not been recorded for each of these various conditions, but patterns were recorded for a Th-U-Si mix that contained 7.4% silicon, had a mesh size of 110 to 250 μ , and was sintered at 2000°C for 2 hr. X-ray diffraction results on this compact showed (1) that all silicides had been converted to carbides, and (2) that no oxide was present.

A plastic vacuum impregnation unit has been designed and is under construction to "freeze in" the carbide particles in the graphite matrix so that the fuel compacts can be examined under a microscope without disturbing the microstructure. Several preliminary experiments are now being conducted to check the compatibility of the carbides with various nonaqueous liquids, so that the samples can be polished or examined in the laboratory or hot-cell facilities.

Metallographic facilities to permit complete examination of reactive fissile-fertile phase materials in protective inert atmospheres have been designed. Completion is scheduled for early in 1960.

Hot-pressing

The fuel compacts are prepared by mixing the fissile-fertile phase particles with graphite and pitch and then hot-pressing this material to form a dense, strong compact. Two types of hot-pressing procedures have been employed. The first is a pressure baking process utilizing graphite dies heated by an external furnace. Temperatures are limited to about 1000°C by the furnaces, and pressures are limited to about 5000 psi by the strength of the graphite dies. The second is a resistance-

heating procedure wherein large currents are passed through the compact, heating it internally during pressing. This technique permits much higher temperatures (over 2000°C) and higher pressures because the graphite die may be strengthened by an external, water-cooled, metal jacket.

The pressure baking process has been extensively studied during the past eighteen months, and compacts are routinely prepared by this method. After pressing, the compacts are fired to 2000°C or higher in a vacuum furnace to convert the fissile-fertile phase to carbide.

The resistance hot-pressing process permits direct formation of carbides during pressing, thus eliminating the furnace-firing step. However, numerous problems are associated with this newer process, e.g., temperature control, die deformation due to creep at high temperatures, etc. These are actively being investigated.

Temperatures of the compacts during resistance hot-pressing were measured optically by sighting through hollow punches. Centerline temperatures of 2100°C were obtained at 4000 amp. Using temperature-current curves constructed in this manner, it is hoped that compact temperatures during pressing may be controlled by controlling the current passed through the compact, and that actual temperature measurements will not be required in each instance.

An investigation of sintering aids to produce stronger, more dense compacts was undertaken. Metals which form stable carbides and have low neutron-absorption cross sections have been mixed with graphite and pitch and hot-pressed. The formation of the metal carbide at high temperature and pressure may yield graphite compacts of lower porosity and, hence, greater fission-product retentivity.

Compacts containing admixed silicon, niobium, and zirconium (hydride) powders in concentrations up to 20 wt-% have been hot-pressed and partially evaluated. Some increase in density was obtained with silicon, whereas the addition of niobium and zirconium to the mixture gave compacts of lower density. Further evaluation, including

determinations of crushing strength and permeability, is in progress.

Various binders other than pitch were investigated, but to date pitch remains the best binder for hot-pressed fuel compacts. A program has been initiated to re-evaluate the pitches and solvents to be used in making resistance hot-pressed fuel bodies. The objective of the program is the selection of a pitch and solvent combination which will impart the optimum properties to the finished bodies.

An apparatus for resistance hot-pressing in vacuo has been designed and is under construction. With this apparatus it should be possible to remove gases more quickly and thoroughly during pressing and thereby to produce denser compacts.

Fuel Bodies for Irradiation Testing

Cylindrical UC_2 - ThC_2 -graphite fuel bodies for the GA-308-1* and -2 irradiation capsules have been prepared (GAMD-973).†

Annular fuel bodies have been prepared for the GA-308-3 irradiation capsule. The purpose of this irradiation is to evaluate (1) the effect of increased metal concentration in the fuel body brought about by the change to annular design, and (2) the effect of fuel particle size. The capsule contains six fuel bodies--two each with fuel particles of $< 50 \mu$, 110 to 250 μ , and 250 to 500 μ . Solid-solution ThO_2 - UO_2 powder was converted to carbide in situ to yield 25 wt-% metal (thorium and uranium) in a graphite matrix. A total of 25 fuel bodies have been prepared--eight with enriched uranium and the balance with natural uranium. The extras were for process development and control evaluation. Radiography and chemical analyses indicate a substantial improvement in fuel distribution over previous compacts. Uniformity is best in fuel bodies with the smallest fuel particle size, as would be expected.

*The irradiation-capsule numbering system is explained in Section 1.6.

†General Atomic informal reports (GAMD series) are listed in an Appendix to this report.

The next irradiation capsule, GA-308-4, will be used to evaluate similar fuel bodies prepared from silicides rather than from oxides. In other respects (composition, fuel particle size, etc.), the fuel bodies and test conditions are to be the same as for GA-308-3. These two irradiations will therefore furnish a direct comparison between the two types of fuel bodies.

Before the enriched samples were prepared for the GA-308-4 capsule, a large number of trial compacts were made to evaluate silicide concentrations and processing variables. Three warm-pressed and sintered (2 hr at 2000°C) annular fuel bodies were prepared from each of five different Th-U-Si compositions (6.4% to 20% silicon)--one using < 50- μ powder, one using 110- to 250- μ powder, and one using 250- to 500- μ powder. No significant differences were noted in ease of grinding and handling the silicides, or in the final compact dimensions or densities (average, 2.45 g/cm³).

A composition containing 7.4% silicon was chosen for the enriched fuel bodies. This silicon content is high enough for easy grinding, yet not so high as to constitute a major addition to the fuel. Also, this composition had the densest-appearing particles when it was examined metallographically.

The preparation of nine enriched fuel bodies (three each with fuel particles of < 50 μ , 110 to 250 μ , and 250 to 500 μ) has been completed. No significant differences were noted in dimensions or densities among any of the fuel bodies. The average sintered dimensions were as follows:

Outside diameter	1.001 in.
Inside diameter	0.499 in.
Length	1.772 in.
Weight	40 g
Density	2.33 g/cm ³
U ²³⁵ content	2.23 g

The composition of the fuel bodies is 14.4% thorium, 6.0% uranium, and 1.64% silicon, balance carbon.

Thermal expansion coefficients of GA-308-3 fuel bodies were determined between room temperature and 700°C and were found to be as follows:

Transverse	$1.25 \times 10^{-6} / ^\circ\text{F}$
Longitudinal	$4.2 \times 10^{-6} / ^\circ\text{F}$

Fuel particle size appeared to have little, if any, effect on the thermal expansion.

A report on the preparation and properties of fuel bodies for the GA-308-3 and -4 irradiations is currently being written.

Reaction Rates of Carbides with Moisture

The determination of the reaction rates of UC_2 and ThC_2 with moisture is essential to development of a means of inhibiting this hydrolysis reaction.

Both UC_2 and ThC_2 hydrolyze to the oxide with the evolution of hydrocarbon gases. It is expected that the reaction will have to be studied by determining the gaseous products with a gas chromatograph and the uranium products by X-ray diffraction.

Data from the available literature give the reaction-rate constant for the hydrolysis of UC_2 as $0.044 \times 10^{-6} \text{ g/cm}^2\text{-sec}$. However, for the fuel bodies, four competing hydrolysis reactions involving both the mono- and dicarbides of thorium and uranium must be considered to determine a composite rate constant. In order to obtain this rate constant, the fuel body or carbide sample will be placed in a controlled environment of humidity and temperature (e.g., saturated air at 50°C). Gravimetric techniques will be used for determining the reaction rates. A vacuum, recording semi-micro balance which is suitable for the job has been obtained. Detailed design of the rest of the apparatus has been completed, and fabrication is in process.

Fuel Migration Experiments

HTGR fuel compacts consist of solid-solution particles of $\text{ThC}_2\text{-UC}_2$ dispersed in a graphite matrix. These particles are large (150 μ) so that

the majority of fission recoils will be stopped in the particles rather than in the surrounding graphite matrix. For nuclear reasons, the particles should be uniformly distributed within the fueled region.

Under high temperatures, diffusional processes could lead to loss of particle integrity and migration of the fissile-fertile phase from the hotter central regions of the fuel compacts to the cooler edges and even to the graphite can which contains the fuel compacts.

The present experiments have been designed to measure

1. Fuel particle integrity at elevated temperatures.
2. The extent of fuel migration as a function of temperature, temperature gradient, and time.
3. Transport of fuel from fueled to unfueled graphite at elevated temperatures.

Electric (resistance) heating is used to simulate fission heating; i. e., heat is internally generated in the fuel compacts. Radiation from the surface of the compact gives rise to a radial temperature gradient, as would occur in the HTGR. By varying the reflectivity of absorbing surfaces, the extent of the temperature gradient can be controlled. One such apparatus (Fig. 1.1) has been in operation for several weeks.

Extensive fuel migration was observed in the first fuel migration experiment. A fuel body 1 in. in diameter by 2 in. long was resistance-heated in helium to a surface temperature of 2000°C and a centerline temperature of 2300°C and was held for 210 hr (9 days). The temperature gradient remained fairly constant for about 6 days, then started increasing at a rapid rate. At the end of the 9 days, the surface temperature was 1940°C and the centerline temperature 2380°C (ΔT increased from 300° to 440°C).

Visual and radiographic examination revealed a thin, dense metal carbide crust on the compact and a rapidly decreasing metal concentration from edge to center. A 1/4-in.-diameter core taken across the center of the fuel body was cut into five equal pieces and analyzed for metal by

Fig. 1.1--Electric (resistance) heating apparatus

firing to $\text{ThO}_2\text{-U}_3\text{O}_8$. The following rough profile of the thorium-uranium concentration was obtained:

Location	% $\text{ThO}_2\text{-U}_3\text{O}_8$
Edge	13.6
	6.0
Center	4.5
	8.2
Edge	14.1

A duplicate core was sectioned on a microtome by cutting slices about $400\ \mu$ thick from the surface inward for 0.4 in. (the centerline is at 0.5 in.). The slices were spectrographically analyzed for thorium and uranium, and the thorium concentration was found to vary from 12% at the surface to 2.5% at the inner point and the uranium concentration to vary from 10% to 0.1%. These results confirm the occurrence of severe migration and indicate that uranium diffuses more rapidly than thorium (see Fig. 1.2). Further analysis is in progress.

A summary of the thermal-gradient fuel migration experiments to date is given in Table 1.4.

Table 1.4

THERMAL-GRADIENT FUEL MIGRATION EXPERIMENTS

Run No.	Fuel Body No.	Surface Temp. (°C)	Centerline Temp. (°C)	Time (hr)	Remarks
1	NCC-60	2050	2320	210	Severe migration
		1990	2400		
2	NCC-399	1850	2020	208	Some migration
3	JP1-52-1	(2000)		24	Temp. overshoot; severe migration
4	JP1-52-2	1850	1980	202	Being analyzed
5	H1-31A-1	1935	2130	200	Being analyzed
6	H1-31A-3	1750	1870	(100)	Still running

Fig. 1.2--Spectrographic analysis of ThC_2 - UC_2 -graphite mixtures

X-radiography revealed no pronounced migration in run No. 2 at lower temperature, but chemical analysis gave the following profile:

Location	% ThO ₂ -U ₃ O ₈
Edge	11.7
	11.3
Center	11.2
	11.5
Edge	11.6

Hence, at temperatures of 1800° to 2000°C, migration is observed in 200 hr.

Run No. 3 temporarily overheated, causing melting of the carbide (which flowed into the sight hole and plugged it up). The run was therefore discontinued. Radiography showed severe migration. This run indicated that even a few minutes at very high temperatures can have serious consequences.

Analysis of runs No. 4 and No. 5 is in progress. Two additional test units are being constructed so that these important tests can be accelerated. Also, it is planned to study the migration of thorium and uranium into the unfueled graphite surrounding the fuel body.

In another experiment six fuel bodies, sealed in tightly fitting graphite cans, were held under isothermal conditions for 1 month in helium at 1800° to 2000°C. Results of chemical analyses of cores cut from these fuel compacts before and after heating are report in Table 1.5.

The three fuel bodies supplied by National Carbon Company contained very fine particles of ThC₂ and UC₂, and the initial fuel distribution was excellent ("Before" columns in Table 1.5). The uniformity of the fuel distribution did not change during the run, but the amount of Th-U decreased by a significant amount, possibly because of diffusion into the graphite cans. The cans will now be analyzed to see if this is true, and if so, to what extent thorium and uranium have diffused into the can.

Table 1.5
CORE ANALYSES FOR ISOTHERMAL FUEL MIGRATION EXPERIMENT
(% ThO₂-U₃O₈)

Core Section No.	Compact No.											
	NCC-51		NCC-59		NCC-62		GA-128B-18		GA-128B-19		GA-110B-3	
	Before	After	Before	After	Before	After	Before	After	Before	After	Before	After
A	11.9	11.4	11.6	11.4	11.6	11.5	9.9	12.2	11.6	12.1	12.1	11.0
B	12.0	11.4	11.7	11.6	11.6	11.5	10.6	12.5	12.2	12.6	12.0	13.3
C	11.9	11.5	11.6	11.4	11.6	11.6	11.2	11.6	10.6	10.9	10.8	10.7
D	11.9	11.5	11.6	11.4	11.7	11.6	9.8	10.5	11.8	12.0	13.1	10.4
E	11.9	11.5	11.8	11.5	11.8	11.6	8.8	11.0	12.0	11.2	12.8	11.2
F	11.9	11.5	11.5	11.5	11.6	11.5	11.9	11.8	8.6	11.1	11.5	11.6
Average	11.92	11.47	11.63	11.50	11.65	11.55	(10.4)	(11.6)	(11.1)	(11.6)	(12.0)	(11.4)

NOTE: Two 1/4-in.-diameter cores were cut from each fuel body both before and after the run. Each core was cut in three pieces for analysis: Pieces A, B, and C are from the first core; D, E, and F are from the second core. "Before" and "After" cores were taken from adjacent locations.

The General Atomic fuel bodies were some early ones with large ThC_2 - UC_2 particles and poor fuel distribution. It is most interesting to note that the distributions are better after the run than before, indicating homogenization through diffusion. No conclusions can be drawn about the relative amounts of thorium and uranium before and after the run, as the distributions are too poor for accuracy. The cans will be analyzed for thorium and uranium as above.

Thermal Conductivity

A simple experiment was designed to obtain reasonably good thermal-conductivity data on laboratory-scale fuel bodies. The fuel bodies, 1 in. in diameter by 2 in. long, are resistance-heated in vacuo or in helium to a surface temperature of 2000°C , and the central temperature is obtained optically by sighting in a small hole. Since heat losses are equal from sample to sample (for the same surface areas and emissivities), the thermal conductivities are inversely proportional to the temperature gradients.

The fuel-migration apparatus was used to compare the thermal conductivities of fuel compacts with that of AGOT graphite. The preliminary data obtained are given in Table 1.6. The increased thermal conductivity shown for hot-pressed graphite might be attributable to its greater density.

Table 1.6
THERMAL CONDUCTIVITIES OF FUEL COMPACTS

Material	Temp. Difference between Center and Surface ($^\circ\text{C}$)	Thermal Conductivity [Btu/(ft ²)(hr)($^\circ\text{F}$ /ft)]
AGOT graphite	290	15.0 (assumed)
Hot-pressed graphite	250	17.4
NCC fuel compact	295	14.8

With the feasibility of the measuring technique established, an apparatus was designed and built to carry out these measurements. Samples of graphite of known conductivity are being obtained to permit conversion of ΔT values to reasonably accurate k values.

Scale-up of Fuel Bodies

Facilities were set up for making full-sized fuel compacts on a pilot scale, and studies were initiated on production hot-pressing techniques.

Temperature calibration runs were performed on a resistance hot-pressed compact 2 in. in diameter. Red heat was observed in the center of the compact 2 min after the start of heating, and after an additional 5 min a temperature of 1900°C was reached at 80% of power. With full power, a temperature of 2350°C was obtained. This temperature decreased with time, however, falling to 2200°C and then to 2150°C . This decrease was evidently caused by graphitization of the carbon liner of the hot press.

Ten full-size compacts were prepared for an experiment to be made in the NRU reactor (see Section 1.6). These compacts were of the annular design, 2.50 in. in diameter by 3.00 in. long, and contained 5.14 g of boron in the 1/4-in.-thick annular region and 0.18 g of boron in the central core. The core and annular region were cold-compacted separately, slipped together, and resistance hot-pressed to form the composite compact.

A detailed analysis of the mixing problems involved in producing uniform fuel compacts was completed, and a process outline was prepared.

1.2. GRAPHITE CAN AND SLEEVE MATERIALS

The can and sleeve for the HTGR fuel element are concentric graphite tubes about 10 ft long and 3-1/2 in. OD. In order to impede the passage of fission products from a contained fuel body, the tubes must have a permeability equivalent to about $10^{-7} \text{ cm}^2/\text{sec}$ measured with helium at room temperature. The objective of the work reported in

this section is to develop a source of such material and to evaluate such material on the bases of materials properties and quality control.

Small graphite disks have been impregnated at General Atomic with a resulting permeability of $<10^{-7} \text{ cm}^2/\text{sec}$. Techniques for applying pyrolytic carbon coatings are also being developed. Low-permeability SiC coats made by American Lava Corporation have been tested and appear to be unreliable at maximum fuel-element temperature. Full-diameter, 4-ft-long tubes of graphite with a permeability of about $10^{-8} \text{ cm}^2/\text{sec}$ have been received from Hawker Siddeley. Two other companies are also actively developing impermeable graphite tubes which may be suitable for the HTGR fuel element.

Silicon Carbide Coating

Coating of graphite with SiC or some other metal carbide offers another approach to the problem of making graphite impermeable. It also offers a method of protecting graphite from reaction with trace O_2 or water in the reactor coolant stream.

The American Lava Corporation and Central Research Laboratories of Minnesota Mining and Manufacturing Company have produced SiC-coated samples for General Atomic evaluation. Permeabilities of $<10^{-10} \text{ cm}^2/\text{sec}$ have been obtained, but rapid deterioration at 1200°C in helium has been noted (see Section 1.7). A full-diameter tube over 4 ft long will be coated with SiC by American Lava. They will also coat a grade "W" graphite tube from Graphite Specialties Corporation when the latter firm delivers a sound tube.

The Carborundum Company has submitted samples of Si-SiC that have impermeabilities of $<10^{-10} \text{ cm}^2/\text{sec}$. These samples are most difficult to machine and are not now available in any large size.

Both Minnesota Mining and Manufacturing and Carborundum will coat small samples for evaluation as an O_2 - or water-protection barrier.

A coefficient of thermal expansion close to 4.5×10^{-6} cm/cm/ $^{\circ}$ C for the graphite is necessary to match that of the SiC coating.

Stanford Research Institute has offered to do SiC or other metal carbide coating studies on contract. Their possible contributions on materials other than silicon for use as barriers to water damage will be evaluated.

The testing program falls into two phases. In the first, the properties of different graphites are determined from essentially destructive tests of small pieces. The second phase is directed at determining the quality of full-size tubes of graphite. This evaluation will at first utilize exhaustive destructive testing where necessary, and must evolve later into a nondestructive quality-control procedure.

Equipment available includes devices for measuring density, strength, porosity, thermal expansion, and permeability, as well as for metallographic examinations, and for high-temperature tests on small specimens. Permeability-measuring equipment that will handle full-diameter tubing up to 4 ft long at room temperature and 1 atm of helium has been built, as has a bursting-strength tensile-test unit to test a short section of full-diameter tubing.

To date, some 50 graphites have been evaluated for use in the HTGR (GAMD-990). From this study of density, strength, porosity, thermal expansion, permeability, metallography, and temperature effects, five materials were selected for a scale-up study and one was selected as a base stock for our own impregnation studies.

Four 4-ft-long, 3-1/4-in.-OD impervious graphite tubes have been received from Hawker Siddeley. They are currently undergoing tests.

Equipment to determine the permeability of a full-diameter tube at reactor temperature is being designed.

Pyrolytic Carbon Coating

When CH_4 or a similar hydrocarbon gas is decomposed by contact

with a hot surface, the carbon will remain on the surface. Carbon so deposited is called pyrolytic carbon and is highly oriented. The high degree of orientation makes such coatings an effective gas barrier against fission-gas transport.

A rotating-kiln furnace has been designed and built for small-particle coating and for producing uniform coatings on graphite pieces. This furnace will reach over 2000°C with a 30-kva, 10-kc induction power supply. The furnace tube is 20 in. long and the hot susceptor is approximately 4 in. long and 2 in. in diameter. An atmosphere of CH_4 is used at a pressure of a few millimeters. The kiln rotates at ~ 60 rpm, and the furnace can be tilted at any angle between horizontal and vertical.

An experiment was conducted in which gas was decomposed in a 5-mil annulus between two sleeves 3 in. long and 1-1/2 in. in diameter with twenty 1/32-in.-square grooves between the sleeves. In 2-3/4 hr at $1300^{\circ}\text{C} \pm 50^{\circ}$, the annulus was filled without plugging of the grooves. Some of the pieces cracked and appeared to be under stress. The deposited material in the annulus was bonded only to the inner tube. The inner sleeve was UF-4S graphite and the outer was AGOT.

The pyrolytic-carbon layer was further evaluated. The piece had an over-all impermeability coefficient of $1.7 \times 10^{-8} \text{ cm}^2/\text{sec}$, and the permeability of the pyrolytic layer was calculated to be about $4 \times 10^{-9} \text{ cm}^2/\text{sec}$. Photomicrographic studies of the coating in the annulus and in the slots show that a complete, essentially monolithic deposition was made (see Fig. 1.3). The only exception was at the corners of the slots, where some discontinuity in the coating was noted.

In another experiment, an attempt to apply zone heating to the deposition of material in the annulus did not result in sufficient carbon to close the gap. A longer run at higher temperatures will be made.

The initial experimental phase of an investigation of the rate of deposition of pyrolytic carbon in the range 1200° to 1800°C at CH_4 pressures from 2 to 8 cm Hg has been completed. A reasonably consistent set of data has been obtained.

Fig. 1.3--Photomicrograph of pyrolytic coating on graphite

The next phase of this investigation will deal with the effects of pre-heating and retention time on the reaction rate. More precise control equipment is being assembled for this study.

The effects of small amounts of silicon in the CH_4 stream will be studied by means of X-ray diffraction patterns of the deposited graphites. Prior work suggests that a small amount of silicon impurity may have a beneficial effect on the crystal structure.

Purge capsules for irradiation tests will be coated in the rotating furnace. Runs to date have demonstrated that the rotated bodies can be coated so that there is no blemishing of the coating by contact with supports. However, coatings are much thicker at the gas entry point than at the exit end of the kiln. Entry of the gas at both ends of the kiln will be attempted.

Procurement of Materials

At the present time, the specifications of paramount interest in procuring materials for the HTGR sleeve and can are as follows:

Permeability: better than 10^{-7} cm²/sec.

Thermal neutron absorption cross section: $\leq 5 \times 10^{-3}$ barns.

Ash content: < 0.1%.

Dimensions (ft):	<u>Can</u>	<u>Sleeve</u>
Length	10	10
Outside diameter	2.75	3.5
Inside diameter	2.25	2.75

Three graphite companies are actively engaged in developing impermeable graphite to these specifications. Other companies have expressed interest in supplying base stock graphites for impregnation by General Atomic.

Hawker Siddeley has delivered four impermeable graphite tubes about 4 ft long and 3-1/4 in. OD. These tubes have room-temperature permeabilities of 10^{-7} to 10^{-8} cm²/sec by measurements made at General Atomic. As the Hawker Siddeley process puts the bulk of the impermeable material near the outer surface, machining must be done before impregnation. The tubes are also subject to the danger of slight mechanical damage affecting their permeability. The base stock is Morgan EYX-60 graphite, which is extruded as solid cylinders. This is an experimental grade and is not obtainable at present in long lengths.

National Carbon Company has provided small graphite samples approaching 10^{-7} cm²/sec permeability. They state that they are developing a process that will produce tubes to the above specifications, and eventually to 10^{-10} cm²/sec. They are developing a special base graphite which has been fabricated in 10-ft lengths. General Atomic has not, however, received any full-length tubes for evaluation.

Great Lakes Carbon Corporation is engaged in developing a base-stock

graphite tube for HTGR use. They are also investigating methods of making such a tube impermeable. They now have the capability for extruding such tubes, but have yet to achieve the baking step. Pore-spectrum data, of course, are not yet available. The base stock could be made available for General Atomic to impregnate, if desired.

Speer Carbon Company is willing to put some effort into scaling up existing materials if they are suitable as base stocks.

1.3. CONTROL-ROD MATERIALS

Tentative specifications for control-rod materials have been established. B_4C -graphite samples of various compositions were obtained from several vendors, and their physical properties were evaluated. Several promising materials were selected for irradiation testing.

Samples of B_4C -graphite containing 10, 20, and 30 wt-% boron were evaluated. The tests included density and density change on heating to $2000^{\circ}C$, crushing strength, thermal expansion, and porosity, as well as metallographic studies. Table 1.9 contains the results of these tests, and on the basis of these results, the following six materials were selected for irradiation testing in capsule GA-310-1:

Speer 8736--high fired, 10% boron;
Speer 8710--low fired, 20% boron;
Speer 8734--high fired, 20% boron;
Speer 8735--high fired, 30% boron;
Carborundum--20% boron;
Carborundum--30% boron.

1.4. METAL-CLAD FUEL ELEMENT

Experiments designed to elucidate the effect of steam on properties of Monel at elevated temperatures are planned, but the exact experimental conditions have not been determined as yet. Analysis of the cladding

Table 1.9
EVALUATION OF B₄C-GRAPHITE CONTROL-ROD MATERIALS

Sample ^a	Boron Content ^b (%)	Firing Temp. ^c (°C)	Density (g/cm ³)		Weight Loss on Heating to 2000°C in Vacuo (%)	Crushing Strength ^d (psi)		Coefficient of Thermal Expansion (in. / in. / °F)	
			As Received	After Heating to 2000°C in Vacuo		Longitudinal	Transverse	Longitudinal	Transverse
Speer 8712	10	850	1.65	1.58	3.79	8,000	9,000	1.4x10 ⁻⁶	2.4x10 ⁻⁶
Speer 8736	<10	>2,700	1.63	1.56	3.81	3,400	4,700	0.9	1.1
Speer 8710	20	850	1.64	1.62	1.95	12,000	8,100	1.5	2.6
Speer 8734	<20	>2,700	1.69	1.68	0.11	8,100	7,000	1.0	2.0
Speer 8711	30	850	1.74	1.68	4.54	7,800	9,500	1.5	1.3
Speer 8735	<30	>2,700	1.68	1.64	2.88	5,300	3,600	1.3	1.2
Carborundum	20	-----	1.71	1.58	4.90	4,300	3,700	2.1	2.0
Carborundum	30	-----	1.60	1.46	5.39	3,900	4,200	1.2	2.1

^aAll samples weighed 22 g.

^bAs reported by manufacturer. High-fired samples not analyzed after firing.

^cProcessing temperatures reported by Speer Carbon Company.

^dDirections with reference to grain orientation.

problem is currently in progress and will be completed shortly. The present status of this analysis has indicated the desirability of performing tension tests on Monel as a function of continuously decreasing temperature. This is necessary because of the structural changes that occur when a metal strain-hardens.

At this time the Monel cladding is thought to act as follows: Since under conditions of insufficient external pressure, the cladding may not remain in contact with the graphite, it would appear that a transition occurs in which the cladding collapses onto the graphite. This transition depends on such variables as temperature, creep strength, and external pressure. It would be desirable to know the experimental conditions under which this occurs, and to this end the possibility of using weldable metal strain gauges is being explored. If small-scale laboratory tests show that they are practical, these gauges will be employed in cycling experiments in order to determine the strain conditions in the cladding as a function of temperature, which will also be measured. Provisions for measurements of strain and temperature are currently being made on the autoclave.

An analysis of some of the factors involved in the recent failures of the HTGR Monel-clad fuel elements has been made, and a report is in preparation. The difficulties with the Monel cladding are most likely due to the low hoop stresses obtained with 0.015-in. - thick material. A suggested remedy is the use of 0.010-in. cladding, which should increase the hoop stresses by 50%. This should cause the cladding to always remain in contact with the graphite. Results of tension tests on specimens cut from the 0.015-in. Monel tubing presently in use show a yield strength of about 30,000 psi. More tension tests are planned in order to obtain yield-strength data for Monel specimens in various conditions. Hardness measurements indicate that the Monel tubing as received is in the annealed condition ($B_{hn} \sim 114$). After draw-cladding, the hardness of the Monel increases to $B_{hn} \sim 118$. This corresponds to

cold-working Monel less than 1%. Tensile tests are in progress to evaluate more accurately the increase in the strength of the Monel caused by draw-cladding the Monel over graphite. Contact between graphite and cladding may be more readily maintained by the use of thinner cladding and higher pressures.

Collapsing tests have been performed on the dome-shaped end cap contemplated for use on the HTGR metal-clad fuel element. The results of these tests indicate that it is not feasible to attempt to bond the end cap to the graphite by any method, since rupture of the graphite would be inevitable.

During this reporting period, the inner-chamber autoclaves have been put into operation. The inner chambers have an inert atmosphere fed into them, after which the atmosphere is fed into the heating chamber of the autoclave proper. The inner chamber maintains a high-purity atmosphere around the fuel elements, while the outer chamber maintains the pressure and contains the heating elements. There is at most a very slight pressure difference between the inner and outer chambers. The first inner-chamber autoclave was operated by automatic control and has worked very well. Automatic temperature cycling of the autoclave can not be achieved until additional equipment is purchased.

Five fuel elements have been thermal-cycled to failure in the autoclaves, and various end caps that had been seam-welded into the tubing were cycled, including one stepped end cap and one dome end cap. Except for one fuel element, all failed within one cycle. The element with the dome end cap exhibited an early failure. Upon heating this element, the cladding material wrinkled severely around the juncture where the dome meets the straight portion of the cladding. The element failed when a temperature of 1350°F was reached. A seam-welded fuel element failed when a temperature of 1670°F was reached. The seam-welded elements all failed at their top halves, indicating that the top half of the inner chamber was at a higher temperature than the bottom. The five

fuel elements tested were all cycled in a horizontal autoclave.

Upon removal of the elements from the autoclave it was found that most of the thermocouples were loose, but it was not certain exactly when they had become disconnected, and it is probable that a good number of the thermocouples were reading gas temperatures rather than the fuel-element temperature. It is expected that sometime in January, 1960, a short vertical autoclave will be put into operation, and elements will then be tested in the vertical position, which more nearly simulates reactor conditions. An improved method for attaching the thermocouples to the metal cladding is necessary not only for test work but for some of the fuel elements which will be inserted in the reactor. An accelerated program for developing methods for attaching these thermocouples to the fuel elements is being initiated.

In the fabrication of the fuel elements, especially the dome-end elements, the mating fit of the parts is very important. Since the cladding material is so thin, a small misfit results in a poor weld. Patching is very difficult, if not impossible. Since the Monel does absorb some graphite on heating, subsequent remelting for patch welding allows the dissolved graphite to come to the surface and makes a fusing of the seam difficult.

1.5. FABRICATION DEVELOPMENT

Fabrication development work for this period has been concentrated on

1. The assembly of Monel-clad graphite elements of various kinds, and the attachment of thermocouples.
2. The wetting of graphite with Monel.
3. The outgassing of graphite for the purpose of removing sulfur-bearing contaminants, and the brazing of graphite.
4. The swaging of Monel and niobium alloys over graphite tubes.

Seven pieces of graphite tube 3-1/2 in. in diameter and 3 ft long were clad with 0.015-in. Monel tubing. On the first five pieces, attempts to apply tail pressure on the graphite while drawing the cladding met with little success. Tensile cracking of the graphite occurred to varying degrees because of the difficulty in maintaining a compressive force on the graphite while the cladding was being drawn. The graphite tubes for the last two elements to be draw-clad were put into compression by clamping them with a threaded tie bar and end plates. The assemblies were then drawn by pulling on the Monel cladding tube only. There was no tensile cracking of the graphite in this operation. The diametrical interference obtained in all seven pieces was approximately 0.011 in., although measurements immediately after drawing indicated interference as high as 0.015 in. Two of the above draw-clad tubes were fitted with Monel end caps of the step design, the most successful of the cap designs that had been tested on an element clad with Type 430 stainless steel. Two other 0.015-in. -Monel draw-clad elements were fitted with roller-seam-welded end caps. The roller seam weld was expected to minimize shearing of the end seal weld during cycling. In addition, two of the assembled elements had hemispherical end caps with a 3/16-in. -long straight portion. The straight portion was to facilitate the better fit of parts for welding, as the fit-up of parts is extremely important when welding in contact with graphite is required. Although some graphite is absorbed in the weld, the absorption is not a serious problem since the weld remains ductile and amenable to working. All graphite in the elements described above was outgassed before being clad.

The cycling of an element clad with 0.015 in. of Monel resulted in severe wrinkling at the end caps. Measurement after the first cycle showed that diametrical interference had decreased from 0.010 to 0.008 in. The assembly of elements clad with 0.015-in. Monel is being discontinued because of these failures. It has been suggested that wrinkling problems would be generally minimized if 0.010-in. -wall Monel was used instead

of 0.015-in. Some 0.010-in. -wall Monel tubes 3-1/2 in. in diameter are scheduled for delivery by mid-January. Some 10-mil-wall, 2-1/2-in. -diameter Monel tubing is available now and will be used in the assembly of several elements having Monel-wet graphite ends to which the outer Monel cladding will be pressure-bonded. When the 3-1/2-in., 10-mil-wall tube is received, four elements will be assembled.

Considerable effort is being directed toward producing metal-plated graphite end caps to which Monel cladding tubes may be pressure-bonded or brazed. Wetting graphite with nickel or Monel appears to be the most promising approach. A flame-sprayed Monel-clad graphite sample with grooved surfaces was heated in vacuum to 1410°C and held at that temperature for 15 min. The Monel-to-graphite bond appeared to be excellent, both on metallographic examination and on attempts to remove the Monel from the surface mechanically. Four samples of graphite have also been covered with molten nickel. One of these was clad with a nickel tube and end cap and heated in vacuum to 1500°C. Partial wetting occurred, but it appeared that metal had run off the side before thorough wetting took place. The ends were covered completely, and one section of one end was ground to produce a relatively even layer a few thousandths of an inch. thick. One piece from this section was then cycled in helium ten times between room temperature and 1500°F with no visible damage.

Spinning may be tried to remove the excess nickel and Monel from the surface. Other types of graphites will also be tested. Morgan EYX-60 is a fine-grain graphite very useful for impregnation studies and possibly useful in this type of work. The graphite used to date is characterized by large surface porosity. In connection with these studies, a pressure-bonding autoclave has been designed and may be constructed if the method shows further promise.

Four 3-1/2-in. -diameter and three 2-1/2-in. -diameter graphite plugs were flame-plated with Monel, heated rapidly (20 to 30 min) in vacuum to the melting point of Monel, and cooled as soon as the Monel

had melted. This technique caused very little lumping. Two of the samples had no lumping whatever, whereas in previous samples lumping was a serious fault. This Monel-wetting technique is quite time-sensitive. Bonding of Monel cladding to Monel-wet end caps by diffusion at 1000°C in vacuum may be attempted. No pressure-bonding equipment is available at General Atomic for this purpose, but the possibility of using Battelle Memorial Institute equipment is being explored.

The chemical analysis of NCC-CS graphite outgassed at 2000°C in the vacuum induction furnace shows that tubes that were outgassed for 4 hr contain 0.0085% sulfur and tubes outgassed for 10 hr contain 0.0748% sulfur, while as-received graphite contains only 0.0069% sulfur. All of the samples analyzed were taken from the surface at the ends of the tubes. Among many possible explanations for the increase in sulfur are (1) that the sulfur-bearing gases were removed but were reabsorbed, (2) that the gases were dissociated and sulfur was deposited as an element on the surface, and (3) that the vacuum induction furnace may have been contaminated with sulfur. Since small samples outgassed at 2000°C in another furnace showed nearly complete removal of sulfur, it is surmized that one or more of the above processes must have taken place. Other methods of sampling these tubes are being attempted to correlate the reasons for the increase of sulfur. Samples of insulations from the vacuum induction furnace have been submitted for chemical analysis to determine whether the furnace itself was contaminated with sulfur.

One CS-grade graphite tube has been outgassed at 1800°C in a vacuum-graphite resistance tube furnace. Chemical analysis after outgassing showed no trace of sulfur,

Two graphite tubes, each having a cemented joint, were draw-clad with the graphite under compression. No damage to the joint resulted. A graphite tube having a cemented joint was heated to 2000°C in vacuum with no deterioration of the joint. A cemented graphite tube was cycled ten times between room temperature and 1600°F , and the joint showed no

change. All of the above tubes had been joined with National Carbon C-6 cement.

Chemical analysis showed that one of the cemented joints contained 0.078% sulfur, but since heating to 2000°C is possible, it may be possible to remove the sulfur. These tests indicate that long graphite tubes composed of short pieces joined together may be used in place of single full-length pieces for the metal-clad fuel element.

Two brazing capsule experiments have been made. TSF graphite was used for both capsules. One sample contained zirconium as a braze material; the other contained Monel. Both samples were heated in the horizontal position, and neither braze material formed a satisfactory joint. The zirconium had apparently carburized, possibly because the heating rate was too slow. In most areas, only the center of the zirconium wire had melted. The wetting of the joint was spotty. The Monel braze material had run down to the bottom of the capsule and very little of the Monel was in the joint. It is believed that a larger clearance than the 1/2 mil used is necessary for Monel. New brazing capsules are being made, and a faster heating rate will be used in future attempts. A 1-mil clearance in the joint will be allowed for future Monel brazing. Future brazing will be attempted in the vertical position as well as in the horizontal position.

The Monel- and niobium-sheathed graphite tubes clad by swaging at Torrington Company were received. Swaging of Monel over graphite is quite satisfactory. However, all attempts at Torrington, using the best of die finishes and die design and with many different lubricants, resulted in severe galling of the niobium alloy tube surfaces. Draw-cladding with the graphite under compression seems to be much simpler, quicker, and less expensive. Though the capital equipment involved is more expensive, the production rate should be higher and the product more uniform in quality.

During the next month, ten spacer pads will be flame-plated onto graphite tubes clad with 0.015 in. of Monel. The pads will be 3/8 in. wide by 1 in. long by 0.030 in. thick. Two pads will be aluminum oxide and eight pads will be tungsten carbide, of which four will contain a nickel binder and four will contain a cobalt binder. Flame-plating of pads will also be tried, using General Atomic equipment, and the above pieces will then be thermal-cycled between room temperature and 1600°F.

1.6. IRRADIATION TESTING

Capsule Irradiation

In-pile experiments are being made to study the per-cubic-centimeter measurements of all possible core materials, including fuel, graphite structural material, and control-rod materials. The capsules contain subsize specimens which can be irradiated at design temperature to integrated fluxes equivalent to full core lifetime.

The capsule irradiation program, begun in 1959 and continuing through 1960, is made up of five different series of capsules. The general purposes and designations of these capsules are listed below:

1. GA-308. This capsule contains six individual fuel bodies and is used to test fuel stability and materials compatibility. Five tests with this type are included in the program.
2. GA-309. This capsule contains three separate compartments, in each of which a single type of fuel body and graphite can may be irradiated and individually purged at frequent intervals to determine fission-product release from each compartment. Fuel bodies will be run at approximately 3600°F, the expected hot-spot fuel temperature in the graphite core. Three capsules of this type are planned.

3. GA-310. The purpose of this capsule is to test the stability of control-rod materials in a simulated HTGR environment. Two capsules have been scheduled for irradiation.
4. GA-311. These capsules contain samples of graphite to determine the irradiation effects on thermal conductivity, dimensional stability, permeability, and physical strength. Three capsules of this type have been planned.
5. GA-TR. The purpose of this capsule is to determine the diffusion or migration of uranium carbide in low-permeability graphite at high temperature and under irradiation. Three capsules of this type have been scheduled.

The irradiation of two GA-308 capsules was completed in 1959, and these capsules are in the process of postirradiation examination and evaluation. Two additional GA-308 capsules are presently being irradiated. One GA-311 capsule has been irradiated and awaits postirradiation examination.

In-pile Loop Irradiations for Graphite-clad Fuel Element

An in-pile test loop will be constructed in which a prototype graphite fuel element of full diameter, though shortened in length, will be operated under conditions equivalent to or more severe than those to be encountered in the HTGR core. The general purposes of this loop may be summarized as follows:

1. To test the stability of a prototype fuel assembly under irradiation.
2. To determine the fission-product release from the fuel element.
3. To determine the extent of fission-product deposition in loop components.
4. To test the fission-product trapping system and the gas-purification system.
5. To test decontamination procedures.

To define more closely the specifications for the in-pile test loop, a design-scope study contract was let to the Vallecitos Laboratory of the General Electric Company. This work has been completed, and from it preliminary specifications have been assembled and circulated for review within the HTGR project (GAMD-1124). A cost estimate was prepared by the General Electric Company covering the cost of designing, fabricating, operating, and dismantling the in-pile loop in the General Electric Test Reactor (GETR).

A survey was made in order to make certain that there were no existing or proposed loops which could carry out the HTGR experimental program. It was concluded that there were no existing or proposed in-pile loops that would satisfy the requirements of the HTGR project from the standpoints of flux, schedule, sample size, and contamination. A survey was then made to determine whether any other test reactors might be more suitable than the GETR economically or technically. The Brookhaven reactor, the Battelle Research Reactor, the NRU reactor, and the Oak Ridge reactor were all found to be inadequate from the standpoint of flux requirements. The Materials Testing Reactor (MTR) was eliminated because of space requirements. The Engineering Test Reactor was eliminated because of the large number of loops which are now installed or about to be installed. This has led to a very low use factor for this reactor. This leaves only the Westinghouse Test Reactor (WTR) and the GETR as possibilities in the United States or Canada. The only test position in the WTR that is technically suitable is the central reactor thimble. However, the flux is a factor of four higher than is needed. Westinghouse has been given an inquiry based on the preliminary engineering specifications and has been requested to submit a proposal by January 15, 1960.

Specifications on the gas-bearing circulators for the loop were prepared and sent to a large number of prospective manufacturers in the United States and the United Kingdom. Bids have been received, and evaluation has been nearly completed.

In-pile Loop Irradiations for Metal-clad Fuel Element

It is planned to construct an in-pile test capsule capable of irradiating a full-scale metal-clad fuel element under conditions approximating those of the HTGR metal-clad core. The objectives of this task are as follows:

1. To test the dimensional stability of the metal-clad element under irradiation.
2. To test the materials compatibility in a simulated HTGR environment.
3. To isolate the possible modes of test-element failure under design operating conditions.

A preliminary design of the full-scale metal-clad fuel-element test planned for the NRU reactor at Chalk River was prepared (GAMD-978). Reaction by the Chalk River personnel was favorable, and they proposed only minor changes. The success of the experiment will depend, to a large extent, on the proper setting of the annular gap between the fuel-element cladding and the capsule wall. The setting of this gap in turn depends on knowledge of the thermal neutron flux in the J-1 position of the NRU reactor. An unperturbed-flux monitor experiment has been run in this position, and the results indicate a peak flux of 2.1×10^{13} , which is very close to what had been predicted. A mockup of the proposed experiment has been inserted in the NRU in order to determine the perturbed-flux values. Results of this experiment are expected shortly.

Irradiation Experiment Evaluation

Container and fuel-body materials have been exposed at expected reactor operating temperatures to a burnup approximating that for the HTGR. The properties of concern for the graphite are changes in gas permeability, thermal conductivity, physical stability, and fission-product retention. The effects of radiation and temperature on the fuel bodies are expected to be possible fuel migration, growth or concentration, and

physical degradation. It is hoped that within the year many of these questions will be answered by both capsule and loop irradiations.

At the present time, the capsules of the GA-308 type contain specimens of fuel and graphite which will be evaluated on a cubic-centimeter basis. It is anticipated that the problem of growth and warpage of the long graphite tubes will be evaluated by irradiating full-scale samples of impermeable graphite. These samples will purposely be put into a flux field whose gradient is large; this experiment will be done at a temperature which will approximate the expected operating temperature of the HTGR.

Several capsules that contained cylindrical fuel bodies have been irradiated (Refs. 1 and 2 and GAMD-1124). Figure 1.4 shows the graphite cans and the stainless steel can for a GA-308 capsule. Figure 1.5 shows the uranium-thorium carbide fuel body after irradiation. This irradiation was performed in the MTR, and the graphite container temperatures were approximately 1100°F. The fuel body shown is one of a set of three which after irradiation showed no cracking or warping. Three other samples were apparently not affected by the irradiation, but after decapsulation they disintegrated because of moisture in the air. The samples on which dimensional measurements could be made are listed in Table 1.10. It will be noted that contraction occurred in all cases except the length of specimen No. 6. These samples are now being evaluated for fuel burnup.

Table 1.10
RESULTS OF GA-308-1 CAPSULE IRRADIATION

Sample No.	Preirradiation Dimensions (in.)		Postirradiation (in.)	
	Diameter	Length	Diameter	Length
4 (NCC384)	1.000	1.9415	0.9942	1.9361
5 (NCC388)	1.000	1.9410	0.9958	1.9401
6 (NCC387)	1.000	1.9423	0.9965	1.9571

Fig. 1.5--One end of UC-ThC fuel element

Fig. 1.4--Portion of GA-308 capsule

GA-308 capsules that have been irradiated but not examined contain annular fuel bodies and samples of different types of graphite. These have been given the equivalent of 50% to 70% of the anticipated HTGR reactor exposure. It is expected that within the next quarterly period results will be available.

Changes in the gas permeability and thermal conductivity of graphite after irradiation are being investigated with GA-311 capsules. The first of these capsules has been irradiated in the GETR. The graphite temperatures should approach 1000°F because of the gamma absorption effect. Melting wires are contained within the graphite in order to determine these temperatures. Evaluation of the capsule will take place in January.

Capsules under design and construction include a purge type in which gas will be periodically forced over the fuel-body can to determine the degree of fission-product release. The returning gas will be caught in traps and counted. These data will be useful in determining which parameters are important in trapping fission products in the gas stream.

1.7. CHEMISTRY EXPERIMENTS SUPPORTING MATERIALS DEVELOPMENT

Fission-product Diffusion Measurements

In the past months, a fission-product-diffusion apparatus designed to run up to 1200°C has been used extensively to evaluate various graphite materials and coatings which might be useful for canning graphite fuel bodies or serving as an impervious sleeve for the HTGR fuel element. In this apparatus, helium is swept past a capsule, containing irradiated UC_2 , of the material to be tested, and the effusion of the fission products I^{131} and Xe^{133} is continuously monitored. The accumulated activities effusing from the sample are recorded on a chart as a function of time. A schematic diagram of the apparatus is shown in Fig. 1.6.

In the case of somewhat permeable graphite, the plots of the I^{131} and Xe^{133} counting rates are characterized by a transient, relatively

Fig. 1.6--Apparatus for measurements of fission-product diffusion

rapid increase followed by a less rapid, approximately constant increase which appears to be a steady-state effusion rate. The latter is reported as fractional loss per day, f . The corresponding permeability constant, K (in cm^2/sec), is given by the equation

$$K = 1.16 \times 10^{-5} f LV/A ,$$

where L is the thickness of the capsule or its impermeable zone (coating), in centimeters; A is the corresponding area, in square centimeters; and V is the free interior volume, in cubic centimeters.

In the case of nearly impervious materials (such as certain SiC-Si coated graphites), a steady-state effusion rate may not be observable on the recorder plot. In this case, after a run of 24 hr or more a half-hour count is taken of the contents of the xenon trap (efficiency = 1.5%, background $\approx$ 200 cpm), and a similar count is taken of the contents of the iodine trap (efficiency = 2%, background = 300 cpm). The contents of the traps are then reduced in volume and transferred to a NaI well counter (efficiency = $\sim$ 25%, background $\approx$ 100 cpm). This procedure gives a detection limit (95% certainty level) of the order of 10^{-5} to 10^{-6} fractional loss per day.

Examples of the permeability results obtained at 1200°C are shown in Table 1.11. Only quite low-permeability graphites were found to give a low effusion rate of I^{131} and Xe^{133} , whereas the SiC-Si coatings and the ZrC-Zr barrier show considerable imperviousness to these fission products.

In addition to the study of the permeability characteristics of potential can and sleeve materials, the fission-product-diffusion apparatus has been used to study fuel-body samples at temperatures up to 1200°C . An analysis of the data obtained indicates that a portion of the iodine and xenon fission products effused quite rapidly, while the balance effused rather slowly. Specifically, in the cases of two compacts prepared by National Carbon Company, NCC No. 7 and NCC No. 8, 10% to 20% of the iodine or xenon had a characteristic effusion time of 0.3 to 2.5 hr,

Table 1.11
EFFUSION RATES AND PERMEABILITY CONSTANTS FOR GRAPHITE MATERIALS

Capsule Material	Length of Run (hr)	Fission-product Effusion Rate (fraction/day)		Xe ¹³³ Permeability Constant at 1200°C (cm ² /sec)	He Permeability Constant at Room Temperature (cm ² /sec)
		I ¹³¹	Xe ¹³³		
Unencapsulated UC ₂ particles	2	0.2	1	-----	-----
RH0021 graphite (fine pore structure), 0.125-in. wall thickness	14	~0.2	~0.8	-----	~3 x 10 ⁻² (for a similar graphite at p = 2.7 atm)
RXLP-2 graphite (experimental low-permeability graphite), 0.125-in. wall thickness	21	0.7 x 10 ⁻³	1.1 x 10 ⁻²	~7 x 10 ⁻⁹	5.5 x 10 ⁻⁷ (for a similar RXLP graphite sample)
RXLP-7 graphite (similar to above)	24	1.6 x 10 ⁻³	1.3 x 10 ⁻²	~7 x 10 ⁻⁹	-----
Z-2 double-walled graphite with a ZrC-Zr diffusion barrier (~20 mils thick)	24	(2.5 ± 0.4) x 10 ⁻⁴	(1.6 ± 0.4) x 10 ⁻⁵	~0.7 x 10 ⁻¹²	-----
3M-43 SiC-Si coated graphite (coating ~10 mils thick)	12	(4.4 ± 0.3) x 10 ⁻⁵	(4.8 ± 1.8) x 10 ⁻⁵	~1.1 x 10 ⁻¹²	2 x 10 ⁻¹⁰ (for a similar SiC-Si coated graphite) ^a
3M-11 SiC-Si coated graphite (similar to above)	24	(1.0 ± 0.4) x 10 ⁻⁵	<5.9 x 10 ⁻⁶	<1.4 x 10 ⁻¹³	-----
3M-11 SiC-Si (a rerun)	67	-----	<2.3 x 10 ⁻⁶	<0.5 x 10 ⁻¹³	-----

^aThe room-temperature He permeability constant of quartz glass is about 4 x 10⁻¹⁰ cm²/sec.

while the balance of the activities showed characteristic effusion times of the order of 10^4 hr.

Since the fission-product retention characteristics of fuel bodies are quite important in the design of fuel elements and fission-product traps, it was decided to build a fission-product-diffusion apparatus with which studies could be made at much higher temperatures (up to at least 2200°C). Accordingly, the design and fabrication of a King-type high-temperature graphite tube furnace has been undertaken. When completed, this furnace will accommodate samples up to 1-1/16 in. in diameter by 4 in. long. The actual tube is 18 in. long; however, the hot zone will be much smaller than this (6 to 8 in.). The maximum attainable temperature should be around 3000°C . Arrangements are such that the furnace can be operated either at high vacuum or with an inert atmosphere.

Other work carried out on the 1200°C fission-product-diffusion apparatus has included the experimental determination of xenon release during the simulated excursion of an HTGR critical-assembly fuel compact and the determination of effusion rates of Xe^{133} and I^{131} from UC_2 powders of various particle sizes.

The first investigation was carried out in support of the HTGR critical experiment program. Two samples tested showed a rapid initial effusion of Xe^{133} and I^{131} and a leveling-off to a slow steady-state effusion within two hours. The fractional loss rate of Xe^{133} was found to be from 0.96% to 0.99% per day, with a total loss over the 17-hr run of 6.3% to 7.0%. The fractional loss for the first hour was 5.5%, indicating that 85% of the total quantity of Xe^{133} lost had escaped in the first hour. The fractional loss rate of I^{131} was determined for only one of the samples and was found to be 0.03% per day, with a total loss of 4.2%. The fraction lost for the first hour was about 3.0%.

A beginning was made on the series of experiments to determine the rate of effusion of Xe^{133} and I^{131} from UC_2 powders of various particle-size ranges (-50 to +100 mesh, -100 to +150 mesh, -200 to +250 mesh,

-250 to +325 mesh, and -325 mesh). The experiments have been carried out at a series of temperatures to obtain temperature coefficients (activation energies) for the characteristic effusion times.

Permeability Measurements of Various Graphite Materials

To assist in the evaluation and characterization of graphites and carbides which might be used in the HTGR fuel element as sleeve, can, coating, or fuel-compact materials, helium permeability constants have been determined for various materials. To date, measurements have been carried out at room temperature and at helium pressures of no greater than ~ 1 atm. In November the apparatus used in these measurements was modified so that specimens could be measured on a continuous basis by the use of a helium leak detector as well as by the pressure-rise or pressure-decay techniques. A schematic diagram of the apparatus is given in Fig. 1.7. A comparison of measurements of three samples of interest by the pressure-rise and helium-leak-detector methods gave the results shown in Table 1.12.

Table 1.12
PERMEABILITY CONSTANTS OF GRAPHITE SAMPLES

Specimen	Permeability Constant (cm^2/sec)	
	Pressure-rise Method	He Leak Detector
Hawker Siddeley No. 1	$\sim 8 \times 10^{-9}$	1.9×10^{-7}
General Electric No. 1	$< 7 \times 10^{-9}$	6×10^{-9}
Raytheon pyrolytic carbon	$< 5 \times 10^{-10}$	2.7×10^{-10}

Recent results on some Hawker Siddeley graphite samples obtained with the leak detector are as follows:

Fig. 1.7--Apparatus for permeability measurements

	Permeability Constant (cm ² /sec)
H-S 149-11	1.4×10^{-8}
H-S 149-7	3×10^{-5}
H-S 149-2	$(1.0 \pm 0.5) \times 10^{-7}$
H-S 149-12	1.2×10^{-8}

These permeability constants, in general, are determined with an uncertainty (standard deviation) of 30% to 50%. These specimens, along with an RXLP-2 specimen from National Carbon Company, will be prepared as zirconium-braze-sealed capsules containing UC₂ for fission-product-effusion studies. The results of the latter studies will be compared with the helium permeability results to see if Knudsen's law holds, i.e., if $K \propto \sqrt{T/M}$. This has been assumed to be true for low-permeability graphites but has not been rigorously proven.

In the last three months a number of other helium permeability measurements have been made in support of metallurgical work on the HTGR project. These include measurements of coated and impregnated graphites made at General Atomic and of graphites which are to be irradiation-tested. Permeation constants for specimens used in the GA-311-1 irradiation were found to be as follows (results are given in cm²/sec):

CEY (NCC) (3-in. -long hollow cylinder) . .	3.7×10^{-7}
Hawker Siddeley No. 145-17	3.4×10^{-7}
CEY (small capsule)	2×10^{-6}
RXLP-2 (NCC No. 6) (disk)	9×10^{-9}
RXLP-2 No. 61 (small capsule)	2×10^{-10}
CEY (curved section)	1.6×10^{-6} and 1.3×10^{-6}

Current plans call for redetermining the permeability constants of these specimens in a postirradiation examination.

The American Lava Corporation SiC-Si coated graphite tubes, which had been given a life test at 1200°C, were tested after a total exposure of 1000 hr in an inert gas (argon) atmosphere. The specimens, which

developed a greenish cast, showed noticeable flaking of the outside surface. The permeation results given in Table 1.13 indicate that these specimens deteriorated badly.

Table 1.13
HELIUM PERMEABILITY CONSTANTS FOR ANNEALED
3M SiC-COATED SPECIMENS

Sample No.	Permeability Constant (cm ² /sec)		
	Initial	After 350 hr at 1200°C	After 1000 hr at 1200°C
1	7 x 10 ⁻⁷ 2 x 10 ⁻⁷	8.6 x 10 ^{-10^a}	7.4 x 10 ⁻⁴
2	1.6 x 10 ⁻⁹	1.5 x 10 ^{-9^a}	9.6 x 10 ⁻⁶
3	5 x 10 ⁻⁹	6.1 x 10 ^{-8^a}	1.7 x 10 ⁻⁵
4	1.7 x 10 ⁻⁹	9.2 x 10 ^{-11^a} 3 x 10 ^{-11^a}	1.4 x 10 ^{-6^a}
5	3.9 x 10 ⁻⁹	5 x 10 ^{-7^a} 3.8 x 10 ^{-7^a}	1.4 x 10 ⁻³
6	5.3 x 10 ⁻⁷ 4.9 x 10 ⁻⁷	1.6 x 10 ^{-10^a}	2.5 x 10 ^{-6^a}

NOTE: These measurements were made with a 1-atm pressure differential and a maximum pressure of 1 atm.

^aIndicates specimens measured with a helium leak detector. Maximum reading with this instrument and these specimens is 4 x 10⁻⁶ cm²/sec.

These tests were carried out to simulate fuel-element sleeve conditions, in particular the temperature of the sleeve surface at hot spots. The results are discouraging, since the SiC-Si coating would seem to show great promise as an oxidation-resistant coating for graphite sleeves which would prevent the steam-graphite reaction from occurring in case of a steam leak in the reactor. In view of the importance of finding a stable

oxidation-resistant coating (or impregnation) for graphite, other samples of oxidation-resistant graphite materials will be given similar life tests.

It is desirable to make some measurements of the helium permeability of graphite materials at pressures and temperatures corresponding to those expected in the HTGR, and to compare these measurements with those made more conveniently at room temperature and lower pressures. Furthermore, in connection with the design of the purge system of the fuel element, where helium is used as a sweep gas, it is desirable to know the back-diffusion characteristics of porous carbon or graphite materials which may be used as plugs. If the plugs are to be effective, the helium passing through the plug must have a sufficient velocity so that xenon, krypton, and other gaseous fission products will not flow in the direction opposite to that of the flow of helium (i.e., back-diffuse). Also, in physically characterizing graphite, it is desirable to establish its permeation behavior for gases over a fairly wide molecular-weight range. The members helium, neon, argon, krypton, and xenon of the family of noble gases are particularly useful for this purpose. Accordingly, an apparatus has been designed and is now being constructed to measure the permeation rates of noble gases at temperatures and helium pressures up to 1200°C and 300 psi. The apparatus is arranged so that a small partial pressure of a noble gas such as krypton or xenon may be introduced into helium on one side of the sample and pure helium may be maintained at the same pressure on the other side of the sample or at an increased pressure, so that the helium will tend to flow in the direction through the sample opposite to that of the noble gas. It is planned to use Kr^{85} and Xe^{133} tracers for the determination of krypton and xenon permeation rates. The mass spectrometer may be used for the determination of neon and argon permeation rates. When an extensive study is made of the physical characteristics of a graphite, the permeation measurements will be correlated with density (porosity), BET surface area, and mercury porosimeter measurements, as well as with microphotographs, to get as

complete a picture of the graphite as possible. A schematic diagram of the apparatus is shown in Fig. 1.8.

Diffusion of Uranium and Thorium in Graphite at High Temperatures

Since HTGR fuel elements, which are composed of thorium-uranium-carbide particles dispersed in a graphite matrix, will operate in a pronounced temperature gradient, it might be expected that the uranium and thorium will tend to migrate (because of the decrease in fugacity with temperature of uranium and thorium in the carbide phase), provided that the over-all operating temperature is sufficiently high to cause significant diffusion. Recent thermal-gradient fuel-migration experiments indicate that substantial migration occurs in 200 hr at temperatures of the order of 2000°C . In order to contribute to the understanding of fuel migration in thermal gradients, experiments on the diffusion of uranium in graphite have been under way for some time.

The first diffusion samples consisted of 10-wt-% UC_2 -graphite compacts sandwiched between two pure graphite compacts. These samples were prepared by hot-pressing. The diffusion rate was measured by sectioning the sample after an isothermal diffusion anneal and analyzing each section for uranium.

Techniques have been worked out for the accurate cutting and collection of graphite samples through which uranium has diffused. A microtome is used for sectioning, and each section is conveniently collected by sticking a piece of Scotch tape to the surface of the sample before it is sliced off. The uranium content is determined by using a neutron-activation analysis technique. In this procedure, which is based on the measurement of La^{140} activity, one microgram of uranium can be determined to an over-all accuracy of 5% to 10%.

In some preliminary experiments, samples were annealed at 2000°C for 50 to 100 hr. From the results to date, it appears that two diffusion mechanisms are at work: a shallow penetration having a steep concentration

Fig. 1. 8--Apparatus for permeability measurements at high temperature and pressure

profile, and a deep penetration in which the uranium concentration is quite low. These two phenomena could be interpreted as being representative of volume and surface diffusion mechanisms, respectively. A similar result was obtained at Battelle Memorial Institute.⁽³⁾ We have found a volume diffusion coefficient of 5×10^{-11} to 5×10^{-10} cm^2/sec at 2000°C , which is essentially in agreement with the BMI work. The diffusion coefficient for the surface mechanism is orders of magnitude greater and appears to be approximately 10^{-5} cm^2/sec . This number is typical of a grain boundary or surface mechanism.

Another more empirical approach has been made to this problem. Discrete particles of UC_2 were hot-pressed into a plane in a graphite rod. This sample was annealed at 2000°C for five days and sectioned up to the original interface. Analysis of the sections showed a concentration profile that was in good agreement with that of the surface mechanism reported above. An X-radiograph of the remaining sample showed the original UC_2 particles to be intact, although possibly slightly diminished in size.

In order to increase the understanding of the phenomena at work, it is planned to use a diffusion sample having a more well-defined interface. An arc-melted button of UC_2 will be polished and held mechanically in contact with graphite. It is also planned to study the diffusion coefficient as a function of temperature, thereby obtaining an activation energy for the process. In order to check the hypothesis of surface migration, it is planned to investigate the effect of the internal surface area of the graphite on the observed diffusion characteristics.

Migration of Uranium from Fuel Compacts

The impermeability of graphite to through-pore migration of the fissile material will be one measure of the stability of the fuel compact. Evaporation of uranium from the surface of the compact might lead to a substantial migration to the can or sleeve material, or to loss into the

coolant stream. It is impossible to predict the evaporation rate^{*} for two reasons. First, the vapor pressure of uranium over uranium carbide is not well known; the heat of formation of UC_2 or UC is in doubt by several kilocalories per mole. Second, the evaporation coefficient, which gives the ratio of the evaporation rate predicted from vapor-pressure considerations to that actually occurring, can only be determined experimentally. Furthermore, the evaporation rate, dm/dt , given in the footnote applies to evaporation from a well-defined free surface, A, of the material of interest. In the case of the evaporation of uranium from a fuel compact made of UC_2 particles in a graphite matrix, the rate of evaporation may be determined by the area of the pores on the surface which communicate through passages in the matrix to the UC_2 in the interior of the compact. Accordingly, a rather empirical approach has to be made in the study and prediction of uranium loss (migration) from fuel compacts.

As a start, a few preliminary experiments were carried out. A closed crucible of AGOT graphite was observed to lose about $0.1 \text{ mg/cm}^2\text{-day}$ when heated in vacuo to 2100°C . In order to evaporate, the uranium had to first migrate through the 1/8-in. wall of the graphite crucible.

In the Langmuir method of measuring vapor pressures, the rate of evaporation of a material through a known area is measured. Experiments of this kind have yielded evaporation rates of uranium from UC_2 of

* The equation giving the evaporation rate in vacuo from a well-defined surface (the Langmuir equation) may be written as follows:

$$\frac{dm}{dt} = \frac{44.3 \alpha P A}{\sqrt{T/M}},$$

where dm/dt = evaporation rate (g/sec),

α = evaporation coefficient,

P = vapor pressure in atm of evaporating species at temperature T ($^\circ\text{K}$),

t = time (sec),

A = area (cm^2),

M = molecular weight of evaporating species.

3 mg/cm²-day at 2000°C and 5 mg/cm²-day at 2100°C. These values are consistent with a vapor-pressure relationship calculated from data by Brewer⁽⁴⁾ and an evaporation coefficient of approximately 0.1. Thus, the 1/8-in. AGOT graphite wall reduced the loss of uranium considerably.

In continuing this work it is planned to study uranium evaporation rates from closed, relatively impervious graphite crucibles and from] pyrolytically coated graphite crucibles containing UC₂.] More sophisticated measurements of evaporation rates and vapor pressures of uranium and thorium carbides will be attempted in the future if these seem warranted for the understanding of the behavior of HTGR fuel elements at high temperature.

Phase Behavior of Uranium and Thorium Carbides with Fission Products

The purpose of this series of investigations is to try to predict the physico-chemical properties of the fuel element after high burnups. Brewer (GAMD-901), Searcy (GAMD-724), and Zumwalt (GAMD-891) have already given consideration to the number of moles of certain important fission products which will be produced, their relative volatility, and the possible importance of compound formation within the fuel element. It is certain that a series of carbides and other compounds will be formed, many of whose physical properties are either unknown or are poorly defined. It is predicted that many other compounds will form, and there will probably be some which are as yet unpredicted. The mutual solubility of these compounds with thorium-uranium carbide and graphite, as well as the phase behavior in the system Th-U-C, will determine many of the physical properties of the fuel element at high burnups. The temperature of incipient melting of the several phases is of prime importance, as it would establish the upper limit of safe excursions in fuel-element temperature.

To the present, only a little work has been carried out on this study. An induction furnace apparatus has been assembled, and samples of uranium, thorium, and some of the fission-product carbides have been

prepared and their X-ray spectra taken in preparation for future work.

A preliminary investigation has been made into the solubility of uranium in graphite, a parameter of importance in all further considerations. An X-ray examination of a UC_2 -graphite mixture after heating to $2000^{\circ}C$ indicates a solubility of less than 1 wt-%. The uranium-graphite diffusion experiments discussed above indicate a value closer to 0.1 to 0.3 wt-%. This may not represent a true solubility of uranium in the graphite phase but may rather represent adsorption of uranium on the porous graphite structure. Thus, for a graphite with an internal surface area of about $1 \text{ m}^2/\text{g}$ (the order of magnitude of the area determined by the BET method), and assuming one atom of uranium occupies $20 (\text{\AA})^2$, a monoatomic surface layer corresponds to 2 mg U/g C, or 0.2 wt-%.

1.8. THERMAL ANALYSIS AND EXPERIMENTS

Thermal Conductivity Measurements

Calculations of maximum temperatures in the HTGR core depend greatly on the thermal conductivities of the fuel-element materials. Consequently, it is necessary for the engineering design personnel to have accurate values of this property.

The thermal conductivity of graphite is considerably affected by the fabrication method, the grain structure, the type of graphite, and the temperature, as well as the amount of radiation damage. Since there is not sufficient information available at present to supply the required thermal conductivity values, they must be obtained experimentally.

Three separate apparatus setups are required to obtain thermal conductivity measurements over the required temperature range. The low-temperature apparatus will be used in the 200° to $1000^{\circ}F$ range, and two high-temperature setups will cover temperatures from 1500° to $4500^{\circ}F$.

Low-temperature Measurements. A test apparatus, previously designed and constructed, is now operating to measure thermal conductivities

of fuel-element materials in the low-temperature range (200° to 1000°F). The material being tested now is graphite.

The present arrangement of the apparatus permits conductivity measurements on cylinders in the axial direction by the use of a heat source and sink in conjunction with a standard material of known thermal conductivity. Modification of the equipment will permit conductivity measurements to be made on sleeves in the radial direction.

Results of the data obtained thus far are not yet available.

High- temperature Measurements. One apparatus to determine thermal conductivities of HTGR fuel-rod samples at high temperatures is about to be fabricated. The system will employ a central heating rod to heat tubular specimens of full-size fuel-element components, and external cooling by a series of radiation, conduction, and convection heat-transfer processes. A molten salt will be the heat-removal medium and will be used to determine heat rates through the samples. By this means, absolute measurements of thermal conductivity will be made on unirradiated samples.

Since irradiation of full-sized specimens would be excessively costly, an attempt will be made in a second apparatus to obtain comparative thermal conductivity measurements on small disks before and after irradiation. The disks will be about 1 in. in diameter and 1/4 in. thick. Heating and cooling will be by radiation.

The apparatus for making these comparative measurements is in the design stage now, and is expected to be constructed within the next few months.

To ensure proper design of the high-temperature thermal conductivity apparatus, minor tests are being performed with a molten salt mixture, known as HTS, that is to be the heat-removal medium. The tests consist of determining corrosion rates and reactions of the HTS with any material to be used in the thermal conductivity apparatus.

The corrosion test now in progress is conducted by turning a 1-in. - diameter carbon steel paddle in 1000°F HTS at about 300 rpm. The paddle

has thus far been run about 60 hr and shows no measurable corrosion.

Compatibility tests have been made with zirconia, alumina, graphite, glass, and zircon, with no evidence of reaction.

The designs of the high-temperature thermal conductivity measurement apparatus require the use of graphite in contact with other materials at high temperatures. In many cases, the compatibility of these materials with graphite is uncertain. Consequently, a few simple tests were performed to supply information for proper apparatus design.

The materials tested thus far include boron nitride, zirconia, and alumina, at maximum temperatures of 4200° , 2800° , and 3700°F , respectively. None of these materials showed any reaction with graphite. The method of testing was to place a small cylinder of material inside an electrically heated graphite helix and evacuated bell jar. All the tests lasted for about an hour. The reactions between these materials and graphite were studied by inspecting them with a microscope after each test. Similar studies will continue on other materials as necessity demands.

Thermal Mockups and Analyses of Irradiation Capsules

A fuel-element irradiation capsule has been designed for in-pile testing in the NRU reactor. A special feature of this capsule is that in-pile control of the fuel-element temperature is possible by altering the thermal conductivity of the gas in an annular gap between the fuel element and the outer containment. Fabrication of a thermal mockup of the capsule was initiated, and an experimental check of the heat-transfer calculation is planned.

Thermal mockups of the irradiation capsules employ graphite electrical heaters as internal sources of heat. The success of most of these experiments depends on knowing the approximate value of the resistance of the graphite heater at high temperatures. The resistivity varies greatly with the grade, manufacturer, grain direction, and temperature of the graphite. Since no adequate information has been available that describes

the dependence of the resistivity of graphite on these factors, some simple experimental determinations of the high-temperature resistivity of three graphites have been made.

Both TS-146 and EYX-60 graphites showed evidence of incomplete graphitization, though HLM and MHLM graphites did not. The resistivity of EYX-60 graphite at 2000°C was found to be about twice that of either of the other graphites at the same temperature. Furthermore, its resistivity was a decreasing function of temperature.

The data were obtained by electrically heating graphite rods and helices in an evacuated bell jar. All the helices were about 1/2 in. OD, 1 in. long, and 1/16 in. square in cross section. Final results were determined from the power supplied and the physical dimensions of the test pieces.

Another problem encountered in connection with thermal mockups of irradiation capsules is that the temperature range of interest in both out-of-pile and in-pile experiments is sufficiently high that a particular thermocouple will probably indicate erroneous readings if it is not calibrated. In addition, sheathed thermocouples received from manufacturers often fail under the temperature and environmental conditions which exist in HTGR experiments. A testing and calibration unit has been constructed, and a calibration procedure is being established. Several thermocouples have been examined and their calibrations determined. It is hoped that information gained from further studies will lead to the development of a suitable method of measuring the surface temperature of graphite at high temperatures.

Analyses of the expected thermal behavior of irradiation capsules GA-308-2, GA-308-3, and GA-309, and also of the NRU in-pile loop irradiation, have been performed.

Comparisons of calculated temperatures with actual temperatures subsequently measured in-pile in the case of capsule GA-308-2 indicated that fission and gamma heat generations in the irradiation position were

only of the order of 50% of those anticipated.

A comparison of one-dimensional and two-dimensional temperature-distribution calculations was made in order to determine the need for refined methods in capsule temperature calculations. It was found that one-dimensional calculations, even if supplemented by much interpretive judgement, were more or less valueless for the particular capsule designs employed. This emphasized the need for the RAT code reported in Section 2.2.

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TASK II. DEVELOPMENT OF REACTOR CORE AND REFLECTOR

2.1. NUCLEAR DESIGN AND THEORETICAL PHYSICS

The major emphasis has been on theoretical reactor physics. Survey calculations have already been done for the HTGR and the reference design is based on these results. In order to obtain a better understanding of the reactor performance, it will be necessary to have improved computational techniques and a better understanding of the neutron thermalization process than existed at the time the survey calculations were done. Considerable effort has been expended on both of these subjects during this quarter and the results are reported. The nuclear engineering calculations are reported only very briefly; it is planned to treat this subject much more fully in the next report.

The effort in theoretical reactor physics has been in three major areas: (1) solution of problems in neutron and reactor physics, (2) assistance to the experimental program and interpretation of experimental results, and (3) development of computational methods and the evolution of nuclear data for use in nuclear design. The specific topics are as follows:

1. Neutron thermalization studies. The central problem is the calculation of a realistic scattering kernel, with which thermal neutron spectra in poisoned graphite can be calculated as a function of position, temperature, and poison concentration.
2. Thermalization codes. Calculational techniques and digital computer programs have been developed for solution of thermalization problems and for their application to specific reactor physics problems.
3. Specific HTGR design problems. Kinetics studies have been carried out, and estimates of biological shielding have been made.
4. Nuclear data compilation.

5. Nuclear engineering codes. Computational techniques and computer codes have been developed and adapted for use in calculating specific performance characteristics of the HTGR.

These topics are discussed in turn, and where relevant, a summary of the work done over the past year is included.

Neutron Thermalization Studies

A major effort of the Physics Section of the HTGR Project has been directed toward the calculation of thermal neutron spectra in poisoned graphite media.* The central problem is the evaluation of the scattering kernel; the methods employed are summarized below. In addition, suitable digital computer codes are required to solve any problem of interest; these are summarized in the section entitled "Thermalization Codes."

When this program was initiated some 16 months ago, preliminary data were available from the LRL Hot Box experiments on the variation of critical mass as a function of temperature for highly enriched U^{235} -graphite systems. The results indicated some reduction in the temperature defect from the values predicted by use of a free-gas scattering kernel for the description of the thermal neutron scattering. In addition to these data, it is obvious that in the region of a temperature discontinuity, like that at the core-reflector interface of the HTGR, there will be a strong spatial dependence of the thermal neutron spectrum. The calculation of temperature coefficients at any temperature will then require an evaluation of the changes in the spatially dependent thermal spectrum with temperature. Likewise, the worth of control rods near the core-reflector interface will be affected by the current of cold neutrons from the reflector. In short, all aspects of reactor performance will be influenced by these thermalization effects. Consequently, a program for studying neutron thermalization in graphite was initiated, and our progress to date is summarized in the

* This work was originally being carried out as a program sponsored by General Atomic. It was subsequently absorbed into the HTGR Research and Development contract.

remainder of this section and in the following section. Some applications of this work are given in the section on specific HTGR design problems.

The early part of the following discussion is limited to those aspects of slow neutron scattering theory which are most relevant for thermalization studies. In particular, we shall be concerned with the use of the Schofield approximation⁽¹⁾ as a method for computing the scattering kernel. Most spectrum calculations performed so far have used kernels obtained with this formalism. It is known, however, that certain deficiencies are inherent in the use of the Schofield approximation. These deficiencies are discussed below, and methods for correcting them are given.

The Scattering Kernel. The effects of thermal agitation and chemical binding on slow scattering have been investigated by many authors.⁽²⁻⁵⁾ The basis for most of the work is the Fermi pseudopotential approximation,^{(6)*}

$$V = \left[\frac{2\pi a}{m} \delta(\vec{r} - \vec{r}_n) \right]. \quad (2.1)$$

Here, a is the bound scattering length for the neutron nucleus system, m is the neutron mass, and $\vec{r}$ and $\vec{r}_n$ are the position vectors for the nucleus and neutron, respectively. In this approximation, the short-range potential between the neutron and nucleus is replaced by a point interaction whose magnitude is adjusted so that scattering by an isolated nucleus is represented correctly in the Born approximation. Corrections to the scattering cross sections obtained from the Fermi pseudopotential approximation tend to be of the order of a few tenths of a per cent.

In addition to the Fermi pseudopotential, use is made of the incoherent approximation, i. e., interference effects from different scattering centers will be neglected. This is accomplished by regarding the scattering from a system of N atoms as N times that from a single atom. This approximation is inappropriate for single crystals and for neutron wavelengths much greater than interatomic separations. Since we are primarily

* We shall work in a system of units where $\hbar = k$ (Boltzmann's constant) = 1.

interested in polycrystalline substances, and since there are relatively few neutrons with energies below 0.01 eV, the incoherent approximation should be suitable for all ranges of energy which are of interest for neutron thermalization studies.

The differential scattering cross section for scattering of slow neutrons of initial momentum $\vec{k}_0$ to final momentum $\vec{k}$ by an atomic system originally in state $|a\rangle$ with energy E_a is given by

$$\frac{d\sigma_a}{d\vec{k}} = \frac{(2\pi)^4}{k_0} \sum_b \left| V_{b\vec{k}, a\vec{k}_0} \right|^2 \delta(E_b + E - E_a - E_0), \quad (2.2)$$

where E_0 and E are, respectively, the initial and final neutron energies. The scattering kernel $\sigma(E_0 \rightarrow E)$ is obtained by multiplying Eq. (2.2) by the density of final states factor $k^2 (dk/dE) d\Omega$, averaging the result over the Boltzmann distribution of initial states and integrating over solid angle Ω . By using the Fourier integral representation of the energy-conserving delta function, the sum over final states $|b\rangle$ can be easily carried out, and one obtains

$$\sigma(E_0 \rightarrow E) = \frac{\sigma_b}{8\pi^2} \left(\frac{E}{E_0} \right)^{1/2} \int d\Omega \int_{-\infty}^{\infty} dt e^{i\epsilon t} X(t), \quad (2.3)$$

where $\epsilon = E - E_0$ is the energy transferred to the neutron and σ_b is the bound-atom cross section. The dependence of the scattering on the atomic motions is contained in the function

$$X(t) = \left\langle a \left| e^{i\vec{k} \cdot \vec{r}(0)} e^{-i\vec{k} \cdot \vec{r}(t)} \right| a \right\rangle_T, \quad (2.4)$$

where $\vec{k} = \vec{k} - \vec{k}_0$, and $\langle \rangle_T$ denotes an average over the thermal distribution of states $|a\rangle$ of the atomic system. It is clear that the energy-angle distribution of scattered neutrons depends on the atomic motion in a very complicated manner. For many systems, and in particular for harmonic systems, Eq. (2.4) may be reduced to a somewhat more tractable form.

Crystalline Moderators. The normal modes of crystal lattices bound

by Hooke's-law forces are plane vibrational waves with wave vector $\vec{k}$, polarization $\vec{e}^{(p)}$ ($p = 1, 2, 3$), and frequency $\omega^{(p)}(\vec{k})$. The motion of any atom in the lattice is a superposition of the normal vibrations. In the following calculations, we make the simplification that the frequencies ω of the normal modes are independent of their direction of propagation, that $\vec{e}^{(1)}$ is orthogonal to $\vec{e}^{(2)}$ and $\vec{e}^{(3)}$, and that $\omega^{(2)}(k) = \omega^{(3)}(k)$. Isotropic crystals and crystals which have highly anisotropic layer structures of the type exhibited by graphite satisfy these assumptions.

Using these assumptions and a theorem due to Bloch for performing averages of harmonic oscillator coordinates over equilibrium distributions,⁽⁷⁾ one can reduce Eq. (2.4) to the form

$$X(t) = X_1(t) X_2(t), \quad (2.5)$$

where

$$X_i(t) = \exp \left\{ \frac{\kappa_i^2}{2M} \int_0^\infty \frac{\rho_i(\omega)}{\omega} \left[(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{-i\omega t} - (2\bar{n} + 1) \right] d\omega \right\}, \quad (2.6)$$

and κ_1 and κ_2 are, respectively, the components of momentum transfer parallel and perpendicular to $\vec{e}^{(1)}$. The corresponding frequency distribution $\rho_i(\omega)$ ($i = 1, 2$) multiplied by $d\omega$ gives the fraction of normal modes with polarization index i which lie in the frequency range ω to $\omega + d\omega$. The quantity

$$\bar{n} = \frac{1}{e^{\omega/T} - 1} \quad (2.7)$$

is the equilibrium population of each oscillator with frequency ω . For graphite, we can take $\vec{e}^{(1)}$ to be perpendicular to the principal planes of the graphite lattice, so that $\vec{e}^{(2)}$ and $\vec{e}^{(3)}$ lie in the planes. The X function may thus be written

$$X(t) = \exp \left\{ \frac{\kappa^2}{2M} \int_0^\infty \frac{\rho(\omega, l)}{\omega} \left[(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{-i\omega t} - (2\bar{n} + 1) \right] d\omega \right\}, \quad (2.8)$$

where

$$\rho(\omega, \ell) = \ell^2 \rho_1(\omega) + (1 - \ell^2) \rho_2(\omega), \quad (2.9)$$

and ℓ is cosine of the angle between $\vec{\kappa}$ and $\vec{e}^{(1)}$. Equation (2.8) is a convenient point from which to begin an analysis of slow neutron scattering in graphite. It is, however, not in a convenient form for the direct numerical calculation of energy-transfer cross sections. In order to evaluate $\sigma(E_0 \rightarrow E)$, it is necessary to find an approximate expression for X , in which the integrations over the variable t may be performed analytically.

The choice of an approximate method for computing the scattering kernel depends primarily on the energy E_0 of the neutron before scattering and on the temperature T of the moderator. The simplest approximation that may be used occurs for temperatures sufficiently high for the excitation of a large number of degrees of freedom in the solid. Here a neutron behaves as though it were being scattered from a free gas at the temperature T of the moderator. In graphite, the free-gas approximation is applicable for T greater than $\sim 1200^\circ\text{K}$.

For temperatures which are low compared to the high-frequency cutoff of the frequency distribution, the calculation of $\sigma(E_0 \rightarrow E)$ is still simple, provided we deal with sufficiently large neutron energies (say, E_0 greater than $\sim 3\theta$). In this region, the scattering may be considered to be from free atoms at a temperature $\bar{T} = (2/3)K_{av}$, where K_{av} is the mean kinetic energy per atom in the crystal. Except for high moderator temperatures T , $\bar{T}$ is generally quite different from T (in graphite, $\bar{T} \approx 700^\circ\text{K}$ at $T = 300^\circ\text{K}$).

For neutron energies E_0 less than $\sim 3\theta$, the kernel is calculated by using the phonon expansion. This consists in extracting the factor

$$\exp \left[- \frac{\kappa^2}{2M} \int_0^\infty \frac{(2\bar{n} + 1) \rho(\omega)}{\omega} d\omega \right]$$

from the X function and expanding the rest of the exponent in powers of

its argument. The n^{th} term in this expansion is

$$X_n(t) = \frac{e^{-\kappa^2 \alpha/2}}{n!} \left(\frac{\kappa^2}{2M} \right)^n \left\{ \int_0^\infty \frac{\rho(\omega, \ell)}{\omega} [(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{-i\omega t}] d\omega \right\}^n, \quad (2.10a)$$

$$X_n(t) = \frac{e^{-\kappa^2 \alpha/2}}{n!} \left(\frac{\kappa^2}{2M} \right)^n \sum_{r=0}^n \frac{n!}{r!(n-r)!} \ell^{2r} (1 - \ell^2)^{n-r} \\ \times \left(\int_0^\infty \frac{\rho_1(\omega)}{\omega} [(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{-i\omega t}] d\omega \right)^r \\ \times \left(\int_0^\infty \frac{\rho_2(\omega)}{\omega} [(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{-i\omega t}] d\omega \right)^{n-r}, \quad (2.10b)$$

where

$$\alpha = \frac{1}{M} \int_0^\infty (2\bar{n} + 1) \frac{\rho(\omega)}{\omega} d\omega.$$

This describes a process in which n phonons, or vibrational quanta, are exchanged between the neutron and the lattice. The principal problem involved in the use of the phonon expansion is associated with the exceedingly complicated form of the higher phonon terms ($n > 2$).

In resolving this problem, we first proceed in the manner described by Schofield and Hassitt at the 1958 Geneva Conference.^{(1)*} For this purpose, it is first convenient to introduce the variable

$$z = t - \frac{i}{2T}.$$

* The approximation for multiphonon processes that was presented by Schofield and Hassitt was considered at an earlier time by Alf Sjölander at the University of Uppsala.⁽⁸⁾

Specifically, for the n^{th} power of the integral appearing in Eq. (2.10a), we obtain approximately

$$\begin{aligned} & \left\{ \int_0^\infty \frac{\rho(\omega, \ell)}{\omega} \left[(\bar{n} + 1) e^{i\omega t} + \bar{n} e^{i\omega t} \right] d\omega \right\}^n \\ &= (M\alpha)^n \left[\int_0^\infty g(\omega) e^{i\omega t} d\omega \right]^n \\ &= (M\alpha)^n \exp \left[i n \bar{\omega} t - (n \sigma^2 t^2 / 2) \right] \left[1 + \frac{\overline{n\omega^3}}{3!} (it)^3 + \dots \right], \end{aligned}$$

where

$$g(\omega) = \frac{1}{M\alpha} \frac{\rho(\omega, \ell)}{\omega} \frac{e^{\omega/2T}}{2 \sinh(\omega/2T)},$$

$$\overline{\omega^n} = \int_0^\infty \omega^n g(\omega) d\omega,$$

$$\sigma^2 = \overline{\omega^2} - (\bar{\omega})^2.$$

The integration over t that must be performed to give the scattering kernel can be done analytically and yields a result that is feasible for machine computations. Programs for computing scattering kernels on the basis of the Sjölander approximation are currently being developed for use in the energy region from 0 to about 0.8 eV.

For neutron energies less than ~ 0.5 eV, many terms in the phonon expansion are required to obtain the scattering kernel. In this region, the principal contribution to the energy-exchange moments comes from processes in which the neutron transfers a large momentum $\vec{K} = (\vec{k} - \vec{k}_0)$ to the colliding nucleus. An asymptotic expansion of $\sigma(E_0 \rightarrow E, \Omega)$ in inverse powers of $(K^2/2M)^{-1/2}$ has been developed by Nelkin and Parks.⁽¹²⁾ This expansion consists in applying the central-limit theorem directly to the X function in Eq. (2.8). The leading term in this expansion is the kernel

for scattering from a free gas at a temperature $\bar{T}$. Higher terms in the expansion may be calculated analytically, and they will give directly the influence of the crystalline binding on the slowing-down process. It is intended eventually to apply this formalism to the computation of scattering kernels in the region E_0 greater than ~ 0.5 ev.

Application to Reactors. It is now possible to make more definite comments on the effects of crystalline binding on the temperature derivative of thermal reaction rates in high-temperature graphite reactors. When operating, these reactors have very hot cores and cold reflectors. We can anticipate that the estimated temperature defect determined by using a crystal kernel will be reduced from that computed by using a free-gas representation of the scattering kernel, for two reasons:

1. At the center of the core, changes in spectrum-averaged cross sections will be smaller.
2. The presence of a reflector softens the spectrum in the core near the reflector-core interface, and since the temperature of the reflector does not change appreciably in going from room temperature to operating conditions, the value of spectrum-averaged cross sections in this volume of the core will change even less than those at the center of the reactor. The volume of the core affected by the reflector is expected to be significantly larger than that estimated from free-gas considerations. The characteristic distance for the relaxation of the thermal neutron spectrum from the reflector to the core is approximately two times larger, because of chemical binding effects.

The temperature derivative of any thermal reaction rate at operating temperatures will be space dependent. This is illustrated in Figs. 2.2 and 2.3, in which characteristic thermal spectra of the core and reflector are shown, respectively. For example, from Fig. 2.2, it is clear that the peak of the thermal spectrum at the center of the core occurs at an energy above the peak of the xenon absorption resonance at 0.09 ev.

Equation (2.10b) then becomes

$$X_n(t) = \phi_n(z)$$

$$= \frac{e^{-K^2 \alpha/2}}{n!} \left(\frac{K^2}{2M} \right)^n \sum_{r=0}^n \frac{n!}{r!(n-r)!} \ell^{2r} (1 - \ell^2)^{n-r} \\ \times \left[\int_{-\infty}^{\infty} \frac{\rho_1(\omega) e^{i\omega z} d\omega}{2\omega \sinh(\omega/2T)} \right]^r \left[\int_{-\infty}^{\infty} \frac{\rho_2(\omega) e^{i\omega z} d\omega}{2\omega \sinh(\omega/2T)} \right]^{n-r}, \quad (2.11)$$

where $\rho_i(\omega)$ is now understood to be an even function of ω . Schofield and Hassitt have noted that each of the integrals appearing in the r^{th} term of Eq. (2.11) may be considered to be the Fourier transform of a probability distribution function (unnormalized). According to the central limit theorem of statistics,⁽⁹⁾ such integrals raised to large powers asymptotically approach a Gaussian in the Fourier transform variable. Specifically, we have

$$\left[\int_{-\infty}^{\infty} \frac{\rho_i(\omega)}{2\omega \sinh(\omega/2T)} e^{i\omega z} d\omega \right]^k \xrightarrow{k \rightarrow \infty} \left[\int_{-\infty}^{\infty} \frac{\rho_i(\omega)}{2\omega \sinh(\omega/2T)} d\omega \right]^k e^{-\frac{1}{2} k s_i^2 z^2}, \quad (2.12)$$

where s_i^2 is the variance of the distribution $\rho_i(\omega)/[2\omega \sinh(\omega/2T)]$. With this result, the integrations over t (and subsequently, over solid angle) can be easily done analytically. The resulting expressions have been programmed for computation on the IBM 704 (KRYOS-II; see section on thermalization codes).

Most of the calculations of thermal neutron spectra so far have been made by using scattering kernels in which all terms in the phonon expansion were approximated in the manner described above. The justification for this procedure was that the spectrum of neutrons slowing down from the region of a few electron volts to energies of the order of kT would depend

principally on multiphonon processes. In our calculations, we evaluated s_i^2 and

$$\int_0^\infty \frac{\rho_i(\omega)}{2\omega \sinh(\omega/2T)} d\omega ,$$

with the ρ 's that were used by Krumhansl and Brooks in their calculation of specific heats.⁽¹⁰⁾ The averages over orientations ℓ were performed by the computer. The results of our calculations are described in detail in a paper presented at the June, 1959, meeting of the American Nuclear Society.⁽¹¹⁾ As shown there, the neutron spectra for absorber concentrations of interest to the HTGR are significantly harder than in the corresponding cases for a gaseous moderator.

It has recently been demonstrated, however, that the use of the Schofield approximation still underestimates the effects of chemical binding on the neutron spectrum by about 50%. To illustrate the defects of the Schofield approximation, we have calculated a neutron spectrum using a kernel in which the first phonon term was correctly calculated from the assumed phonon distributions. All other terms in the phonon expansion were calculated as before. The calculation was then repeated using a kernel in which the first two phonon terms were correctly computed. The results of these calculations are shown in Fig. 2, 1. These results strongly suggested that the use of the Schofield approximation seriously overestimates the slowing down by the moderator in the thermal energy range (0 to 2 ev).

The reason for the inadequacy of the Schofield approximation can be seen by considering the inelastic cross section and mean energy loss per collision in the limit of large neutron energies. As first shown by Placzek⁽³⁾ and later by Wick,⁽⁴⁾ the cross section and mean energy loss per collision $\langle E_t \rangle_{av}$ are given by

$$\sigma(E_0) = \sigma_f \left[1 + \frac{1}{2} \frac{\bar{T}}{ME_0} + O(m^{-1}E_0^{-3}) \right] , \quad (2.13a)$$

Fig. 2.1--Neutron flux spectra for infinite medium at 300°K with 0.5 barn per carbon atom at 2200 m/sec

and

$$\langle E_t \rangle_{av} = \frac{2ME_0}{(M+1)^2} \left[1 - 2 \frac{\bar{T}}{E_0} \left(1 + \frac{1}{4M} \right) + O(E_0^{-2}) \right], \quad (2.13b)$$

where σ_f is the free-atom cross section and

$$\bar{T} = \frac{1}{2} \left[\frac{1}{3} \int_0^{\theta_1} \omega \rho_1(\omega) \coth \frac{\omega}{2\bar{T}} d\omega + \frac{2}{3} \int_0^{\theta_2} \omega \rho_2(\omega) \coth \frac{\omega}{2\bar{T}} d\omega \right].$$

In order to describe the thermalization process correctly, any approximate scheme for computing kernels must yield the same results for $\sigma(E_0)$ and $\langle E_t \rangle_{av}$. It can be shown, however, that the approximation of Schofield and Hassitt leads to results which differ from those of Eq. (2.13) by the replacement of $\bar{T}$ by the effective temperature T'

$$T' = T \left\{ 1 + \frac{1}{2T} \left[\frac{2}{3} \int_0^{\theta_1} \frac{\rho_1(\omega)}{\omega} \coth \frac{\omega}{2T} d\omega + \frac{1}{3} \int_0^{\theta_2} \frac{\rho_2(\omega)}{\omega} \coth \frac{\omega}{2T} d\omega \right]^{-1} \right\}.$$

For the ρ 's used in our spectral calculations, we find that at $T = 300^\circ K$

$$\bar{T} = 709^\circ K,$$

$$T' = 436^\circ K.$$

Thus, the reduction in the mean energy loss due to binding has been underestimated by a factor of 1.6 with the Schofield model.

The contribution of multiphonon processes to the neutron scattering has been approximated by Sjölander⁽⁸⁾ in a manner that gives correctly the effects of chemical binding on the energy-change moments of the scattering kernel. As before, the approximate result for the contribution of the multiphonon processes is obtained by an application of the central limit theorem. The essential difference is that the application of the central limit theorem is made in Eq. (2.10a) with t as the Fourier transform variable, rather than in Eq. (2.11) with the variable $z = t - (i/2T)$.

Fig. 2.2--Infinite-medium spectra characteristic of the core

Fig. 2.3--Infinite-medium spectra characteristic of the reflector

Therefore, increases in the core temperature will result in fewer absorptions in xenon, and, consequently, this poison gives positive contributions to reactivity, the temperature coefficient being of the order of $+2 \times 10^{-5}$ per $^{\circ}\text{C}$. However, near the core-reflector interface, a comparison of Fig. 2.3 with Fig. 2.2 leads one to conclude that the peak of the thermal spectrum in core regions near the reflector is below the xenon resonance and, therefore, that increases in core temperature will result in negative contributions to reactivity from xenon in this region. In addition to this effect, however, the spectrum in this region of the core is softer, and we know that any reaction rates--for example, absorptions in U^{235} and U^{233} --are then more sensitive to temperature changes. Their contributions to the temperature coefficient of reactivity will thus be more negative for U^{235} and more positive for U^{233} . These competing effects make $(1/k)(\partial k/\partial T)$ more positive or more negative, depending on the particular geometry and composition of the reactor system. Consequently, no generalizations can be made without a large number of specific calculations. A wide variety of such space-dependent calculations has been started and will be reported later.

A number of theoretical investigations relating to the program of experimental reactor physics have been started. The results to date are described in GAMD-805. A general description of these experiments is given in Section III.

Thermalization Codes

Calculational methods for neutron thermalization studies and their application to specific problems are conveniently divided into two groups: infinite-medium and space-dependent methods.

The infinite-medium problems we have considered include the evaluation of scattering kernels, the generation of infinite-medium spectra, and the preparation of properly averaged cross sections and related nuclear parameters. Among the space-dependent problems important to

the HTGR are the determination of fluxes within fuel cells and control-rod cells as a function of position and energy, the calculation of spectral variations across a temperature and absorption discontinuity, and the detailed determination of the effects of changing temperature and other physical conditions upon critical reactor systems.

Infinite-medium Methods. The majority of the methods for performing infinite-medium calculations were developed before October, 1959. They are outlined in GAMD-754(Rev.). The relevant codes described in that report are: SIGMA-GAS, for generating scattering kernels according to the free-gas model; KRYOS-II, for generating kernels for crystalline materials using the Schofield approximation for all phonon terms; SPECTRUM, for generating the infinite-medium thermal spectrum in a poisoned moderator, with a slowing-down source; and CRISSCROSS, which averages pointwise cross sections over given spectra and gives microscopic and macroscopic quantities for designated subdivisions of the thermal energy range.

For the evaluation of graphite kernels, two other codes have been developed. PHONON computes the first and second phonon terms correctly, integrates over an assumed phonon distribution, and thus gives data for use in correcting the KRYOS-II kernels (see the preceding section). KRYOS-II evaluates the kernel for all phonon terms, using the Sjölander (rather than the Schofield) approximation.

Two variants of SPECTRUM exist, and another has been proposed and is currently being written. In addition to performing all the functions of SPECTRUM itself, B-SPECTRUM (sometimes called THERMALIZATION) computes the integral properties of the scattering kernel: total inelastic cross section, mean and mean-square energy change, mean lethargy change per collision, etc., as a function of energy. Also, it can linearly combine two kernel matrices, and for certain space-dependent problems it has options for using a converged spectrum as a source spectrum for the next case. C-SPECTRUM performs the functions of SPECTRUM and also allows any

number of kernel matrices to be combined. These may be as large as 150×150 , and at different temperatures; detailed balance is applied to each kernel individually.

Two codes exist for obtaining group-transfer cross sections (or transfer coefficients, when expressed as probabilities): KITTY, which uses the free-gas kernel model, and GRAYMALKIN, which uses any read-in kernel matrix and spectrum. The groups here are subdivisions of the thermal energy range. GRAYMALKIN provides both cross sections and transfer coefficients, and arranges the former in the format required by DSN, which is discussed in the following section. The integration scheme in KITTY is a two-dimensional Simpson's rule, with successive subdividing of intervals as required for convergence. The trapezoidal scheme only is used in GRAYMALKIN.

All the codes mentioned above have been developed at General Atomic.

Space-dependent Problems. To perform one-dimensional space-dependent calculations that incorporate thermalization effects, a space-dependent code is required which allows at least a few "upscattering" groups. The available codes which embody this provision are DSN⁽¹³⁾ (transport theory approximation), FIRE,⁽¹⁴⁾ and ZOOM.⁽¹⁵⁾ Our major experience has been with DSN, since it incorporates a wide variety of options that enable it to perform cell calculations in three geometries and to search for critical size, axial buckling, concentration, etc., as well as k_{eff} .

The DSN code, developed by Bengt Carlson and his associates at the Los Alamos Scientific Laboratory, is a successor to SNG; SNG cross-section cards are compatible with DSN. We had obtained some previous experience with SNG in setting up and running fuel-cell and control-rod cell problems, as well as reactor criticality problems, using several thermal groups (with upscattering). In September, 1959, we obtained an interim version of DSN and put it into operation, with some changes. One major advantage of DSN is that it is written in FLOCO-II (successor to

FLOCO-I), which allows nearly all of the output, and some of the input, to be on magnetic tapes rather than on-line. Furthermore, DSN embodies an improved convergence scheme. The principal change we made to the interim DSN was the introduction of a second convergence criterion; the first criterion applies to the flux iterations for a given guess at the eigenvalue, and the second applies to the convergence of the eigenvalue guesses themselves. This enables the machine to spend most of its time improving its eigenvalue guess, rather than improving the flux shapes when there is a poor guess at the eigenvalue. Other changes were directed towards ease of manipulation, running of successive problems, etc. The temperature-coefficient studies of the HTGR critical assembly were performed using the interim version; they are described in a later section.

In December, we received the "final" version of DSN and visited LASL for talks and conferences relating to DSN and its two-dimensional version, TDC. The "final" version incorporates several convenient improvements over the interim version, particularly as regards output. As before, it was necessary to make certain changes in this version. These were a modification of the iteration scheme to allow two convergence criteria, and a modification of certain details to facilitate running sets of problems. The new version has been used for cell studies, as well as reactor problems, and will be a primary tool in studying space-dependent thermalization effects and in performing special HTGR calculations. An operating manual for DSN will be issued after completion and testing of all the modifications.

Copies of FIRE, a rapid diffusion-theory code, have been obtained from LASL.

A version of ZOOM has been put into operation by another project at this Laboratory. For the HTGR, it will be used mostly in connection with the nuclear analysis work. Our plans regarding ZOOM are discussed further in the section on nuclear engineering codes.

Some of the hazards calculations for the HTGR critical assembly

have been done with PDQ,⁽¹⁶⁾ using one thermal group; we have obtained copies of TDC and of ANGIE,⁽¹⁷⁾ the two-dimensional version of ZOOM, but we have not yet put either of these into operation.

Specific HTGR Design Problems

During the past six months, calculations of interest to the design of the HTGR and of the HTGR critical assembly have been performed. The former include several studies of the kinetics of the HTGR and preliminary estimates of the shielding requirements around the core and compressor. For the HTGR critical assembly, we have considered (1) the calculation of the prompt and slow temperature coefficients and (2) the determination of kinetic properties of these assemblies. These calculations are summarized in the following discussion.

Accidental Loss of Xe¹³⁵ and Related Fuel-compact Temperature Excursion. During the past three months, work supplementing our studies on nuclear hazards was undertaken to estimate the temperature excursions in a fuel compact in the case of a rupture of the fuel can and subsequent loss of Xe¹³⁵.

Two different types of fuel design were considered:

1. A vented element in which a stream of helium is maintained between the can and the fuel compact, thus purging out of the reactor a certain amount of gaseous Xe¹³⁵.
2. A closed element, in which an impervious can contains all the fission products within the core and bottom reflector regions. For this design, it was supposed that the rupture of the can resulted from:
 - a. An increase in differential pressure across the sleeve and can due to the loss of helium coolant pressure;
 - b. An increased internal pressure in the fuel due to an accidental rod withdrawal at steady-state conditions [115 Mw(thermal) and an average temperature of 1700°C in the fuel compact].

The two postulated accidents for the closed element give different temperature excursions and were considered separately. Numerical answers were obtained with the IBM 704 code BLUP (described in GAMD-1072). The delayed-neutron characteristics for the end-of-life graphite-clad core were taken from GAMD-943.

In the vented fuel can, the steady-state concentration of Xe^{135} was derived under the assumption that the gaseous Xe^{135} would diffuse toward the purging helium stream in the helium that fills the voids in the fuel compacts. The concentration of Xe^{135} in this stream was assumed to be negligible. The average concentration of Xe^{135} in the core was found to be:

$$C_{av} = 7.56 \times 10^{10} \text{ atoms/cm}^3 \text{ at steady-state conditions,}$$

$$C_{av} = 9.80 \times 10^{10} \text{ atoms/cm}^3 \text{ at } \text{Xe}^{135} \text{ peak after shutdown.}$$

These concentrations correspond, respectively, to reactivities of 0.17 and 0.22 cents, which, if introduced stepwise, would result in a temperature increase of 10° to 20°C in the fuel compact during an excursion.

With the unvented can, the worst possible hazard occurs if the can ruptures immediately after a break occurs in the helium coolant line. The initial pressures used were 30 atm for the gases in the compact voids and 20 atm for the helium coolant system. It was further assumed that all the fuel cans would break with a crack equivalent to a 3/8-in.-diameter hole, and that the controlling factor was the sonic velocity of the gases flowing through the cracks. These considerations lead to an exponential reactivity insertion

$$t \leq 0.0276 \text{ sec; } \rho = \rho_0 (1 - e^{-17.3t}),$$

$$t > 0.0276 \text{ sec; } \rho = \rho_0 (1 - 0.67 e^{-2.61t}).$$

The first fast exponential corresponds to the rapid decrease in pressure of the gases in the compact voids. After 0.0276 sec, the pressures inside and outside the can will have practically equalized. At later times, the

decrease in gas pressure is given by the slower second exponential, which is dictated by the rate of helium coolant loss out of the piping system. With our present knowledge of fuel-compact retention and with a consideration of the effectiveness of the voids in the bottom reflector, we estimated the value of ρ_0 at the Xe^{135} peak after shutdown to be

$$\begin{aligned}\rho_0 &= 0.0160 \text{ in units of reactivity} \\ &= 3.20 \text{ dollars.}\end{aligned}$$

With a temperature coefficient $\alpha = -1 \times 10^{-5}$ per $^{\circ}\text{C}$, this rate of reactivity insertion gives a maximum fuel-compact temperature excursion of 4200°C , which is close to the 4500°C danger point. However, the assumptions and the computational approximations tend to give a conservative answer. Further details on the analysis and numerical results are given in GAMD-1146.

Preliminary Estimates of Biological Shielding Requirements. A series of shielding studies was started during the last quarter of 1959. The first two estimates to be made were (1) the thickness of concrete required for the main biological shield of the reactor and (2) the shielding required at the compressor to attenuate gamma rays from gaseous fission products.

The methods used are those described by Etherington in Chapter 7 of the Nuclear Engineering Handbook.⁽¹⁸⁾ Essentially, the method consists of integrating a point-source kernel over some assumed source distribution, including build-up factors for gamma rays, and applying tabulated removal cross sections for neutrons of various energies.

Using a value of 100 mrad/week as the permissible radiation tolerance, and selecting typical values, we estimate the maximum working time for a man adjacent to the compressor wall to be 0.3 hr/week for a 3-ft iron shield and 1.5 hr/week for a lead shield of the same thickness.

It is anticipated that the estimates of the main biological shield will be made early in 1960.

Temperature-coefficient Calculations for HTGR Critical Assembly.

The temperature coefficients of a bare and a reflected configuration of the HTGR critical assembly have been calculated over the temperature range from 300° to 900° K. The core was taken to be a homogeneous slab, with a carbon-to- U^{235} atom ratio of 1900 and a carbon-to-boron ratio of 3×10^4 . This boron concentration is equivalent to the thorium concentration in the HTGR (carbon-to-thorium ratio of 300), which corresponds to 0.0076 barn per carbon atom at 2200 m/sec. The carbon density was taken as 1.645 g/cm^3 .

In carbon-moderated U^{235} systems with $1/v$ poisons, the temperature coefficient of reactivity is primarily due to changes in the thermal neutron distribution. Consequently, realistic thermal neutron spectra are required, both in bare systems and reflected systems, in order to calculate the temperature dependence of any reaction rate. The leakage term dominates the temperature coefficient in both systems. In calculating thermal spectra, we have made use of the best available techniques that embody the results of recent theoretical work in neutron thermalization in crystalline materials.

The calculational procedure is outlined and the results are presented.

The bare reactor chosen was a 3-ft slab; the reflected reactor was reflected only at the ends of the 3-ft core by 1-ft slabs of graphite. In each case, the total transverse buckling (corresponding to the other two dimensions) which made the reactor critical at room temperature (300° K) was found.

Six thermal groups and three fast groups were used in all the calculations. The energy ranges of the groups are shown in Table 2.1.

Thermal neutron spectra that are characteristic of an infinite medium were calculated for the core material compositions mentioned above and for the reflector, at four temperatures: 300° , 450° , 600° , and 900° K. Scattering kernels for crystalline graphite, calculated according to the Schofield model, were used in generating the spectra.⁽¹⁾ The spectra so obtained were used to calculate group-averaged cross sections, including

Table 2.1
ENERGY GROUPS

Group No.	E_{max} (ev)	E_{min} (ev)
1	10^7	9.8×10^4
2	9.8×10^4	243
3	243	2.154
4	2.154	0.6813
5	0.6813	0.2154
6	0.2154	0.06813
7	0.06813	0.02154
8	0.02154	0.006813
9	0.006813	0.002154

intergroup transfer cross sections and transport mean free paths. These calculations were done on the IBM 704, using most of the family of thermalization codes developed at General Atomic and described in GAMD-754(Rev.): scattering kernels from KRYOS-II and SIGMA-GAS, infinite-medium spectra from SPECTRUM, group-averaged cross sections from CRISSCROSS, and intergroup transfer cross sections from GRAYMALKIN.

Fast spectra for the core, interface region, and reflector were obtained from the output of a series of criticality calculations for spherical, reflected cores, using the MAGNUM system.⁽¹⁹⁾ Average cross sections for the three fast groups were obtained by averaging the cross sections of Garrison (from GAMD-603) with these spectra, using CRISSCROSS. Slowing-down cross sections from Groups 2 and 3 were calculated by assuming elastic scattering by free atoms in a $1/E^n$ spectrum in each group. The value of n was found from the MAGNUM calculations. For Group 1, the slowing-down cross section was normalized to the Fermi age: $\Sigma_{s1} = D/\tau$. The value of the Fermi age for Group 1 was obtained by standard decomposition, using experimental determinations of the Fermi age to indium resonance in graphite.⁽²⁰⁾ The diffusion coefficient was spectrum averaged.

Criticality was calculated for each temperature, using the LASL multigroup transport-theory code DSN⁽¹³⁾ on the IBM 704.

The multigroup calculations were done for bare and reflected assemblies. The thermal spectra in core and reflector used in the calculations are shown in Figs. 2.2 and 2.3, respectively.

For the reflected assembly, we considered three situations:

1. The core and reflector were heated together.
2. The core only was heated, with the reflector held at 300°K.
3. The reflector only was heated, with the core held at 300°K.

For these three cases and for the bare system, k_{eff} is plotted versus temperature in Fig. 2.4.

The temperature coefficient of reactivity, $(1/k)(\partial k/\partial T)$, is seen to vary only slowly with temperature throughout the range, the maximum variation amounting to about 30%. The average temperature coefficient over the range from 300° to 900°K is found to be -5.1×10^{-5} per °C when only the core is heated, -1.1×10^{-5} per °C when only the reflector is heated, and -6.2×10^{-5} per °C when both core and reflector are heated. It may be noted that, to this order of precision, the effects of core and reflector heating are additive. For the bare assembly, the average coefficient was -4.6×10^{-5} per °C; that is, less negative than in the reflected case. In a highly enriched graphite reactor, the temperature coefficient is due to thermal base effects, and it becomes more negative as the carbon-to- U^{235} ratio increases. For small reflected systems of this type, the average spectrum in the core is softened, and the resulting temperature coefficient is more negative by 10% to 20% for this configuration.

These values refer to the "slow" temperature coefficient, i.e., that appropriate to a uniform heating of all the moderating material. During a prompt excursion, these numbers will be reduced, to first order, by a factor corresponding to the volume fraction of the moderator in immediate contact with the fuel. For the HTGR critical assembly, this

Fig. 2.4--Plot of k_{eff} versus temperature

volume fraction is about 50% for one experimental situation, and about 25% for another. Subsequent cell calculations have shown that the prompt temperature coefficient is $2/3$ the slow coefficient in the 50%-volume case, so the use of the slow temperature coefficient reduced by the volume fraction of the moderator represents a conservative estimate of the prompt temperature coefficient.

Nuclear Data

Although several compilations of nuclear data are available, none of them is sufficiently complete, self-consistent, or well documented to be used in its entirety. Furthermore, such compilations are rapidly outdated as new data become available. For our work, recent data have been obtained from the Sigma Center at Brookhaven on the total and fission cross sections of U^{235} at the energies considered in the experiments, from 8 to 1000 ev. These data are being tabulated in the same manner as the data previously obtained from Shore and Sailor,⁽²¹⁾ which are recorded in GAMD-828. The construction of group cross sections for homogeneous and heterogeneous reactor systems is facilitated by using such data rather than BNL-325.⁽²²⁾

Theoretical estimates of the capture cross sections for the stable and long-lived fission products and for Pa^{233} and Np^{239} have been made by the General Atomic reactor physics group; preliminary results have been reported in GAMD-730 and GAMD-876. Improvements are being made by using more recent data. The theoretical methods used are similar to those employed by Greebler and Hurwitz.⁽²³⁾

The thermal diffusion length in graphite has recently been measured as a function of temperature by Lloyd, et al., at Hanford.⁽²⁴⁾ Attempts to fit these data by using spectrum-averaged absorption cross sections and diffusion lengths were not successful when BNL-325 cross sections were used. In the calculations, the following assumptions were made:

$$L^2 = \bar{D} / \bar{\Sigma}_a, \quad (2.14)$$

$$\bar{\Sigma}_a = \frac{\int_0^\infty N_0 \sigma_a^C(E) M_T(E) dE}{\int_0^\infty M_T(E) dE},$$

$$\bar{D} = \frac{\int_0^\infty [1/3 N_0 \sigma_t^C(E)] M_T(E) dE}{\int_0^\infty M_T(E) dE}.$$

Here $M_T(E)$ is the Maxwellian at temperature T ; $\sigma_t^C(E)$ is the total cross section of graphite; $N_0(C)$ is the atom density of graphite; and σ_a^C equals 3.4 mb at 2200 m/sec and varies as $1/v$. In the expression for $\bar{D}$, one usually uses the transport cross section; however, for all elements except hydrogen, in the energy range below a few tenths of an electron volt it is probable that chemical binding effects reduce the average value of the neutron scattering angle in the laboratory system to zero.

The data in BNL-325 for $\sigma_t^C(E)$ gave consistently high values of L , as shown in Table 2.2. In addition, the value of $\bar{D}$ did not agree with results of pulsed neutron experiments.⁽²⁵⁾ Consequently, the total cross section of graphite was remeasured at General Atomic using the electron linear accelerator. The results are shown in Fig. 2.5. With these data, average diffusion coefficients which agree with the results of the pulsed experiments were calculated. By using these computed values of $\bar{D}$ and the experimental values of the thermal diffusion length, the 2200-m/sec value of the absorption cross section of graphite was calculated to be 4.03 mb. This is in substantial agreement with the value reported by Hughes at the 1958 Geneva Conference.⁽²⁶⁾ This is not surprising because σ_a is a derived quantity; it is obtained with Eq. (2.14), where the value of $\bar{D}$ is taken from pulsed neutron experiments.

Using the remeasured total cross section of graphite and the rederived value of its absorption cross section, we have calculated values of the

Fig. 2.5--Total cross section of graphite

thermal diffusion length for graphite as a function of temperature, which are shown in Table 2.2.

Table 2.2
VARIATION OF GRAPHITE DIFFUSION LENGTH WITH TEMPERATURE

Temp. (°K)	Experimental Value of L (cm)	Values of L Calculated from Three $\sigma_t^C(E)$ Values* (cm)		
		BNL-325	Ref. 26	Rederived
300	53.76	59.95	56.10	53.76
450	59.25	65.52	61.97	59.39
600	63.49	70.04	66.63	63.86
900	69.98	77.21	73.83	70.76

*The σ_a^C values (for 2200 m/sec) were: BNL-325, 3.4 mb; Ref. 26, 3.7 mb; and rederived, 4.03 mb.

The agreement with experiment is reasonably good. The evaluation of the thermal diffusion length is essential to our reactor analysis, since measurements of $L(T)$ exist only for pure graphite and, consequently, are not suited for analysis of poisoned graphite systems.

We are keeping in close contact with cross-section groups at the MTR and at ORNL that measure η values in U^{233} and the resonance parameters of Th^{232} . A remeasurement of these resonance parameters has been under way by W. W. Havens, Jr., and his group at Columbia University for some time. We are awaiting his results with interest because of existing discrepancies in the published values of these parameters.⁽²⁷⁾ We are entirely dependent on calculations for the determination of the resonance integral of thorium and its temperature derivative. Published experimental data on these quantities are inconsistent and, furthermore, do not cover our range of dilution.

The general problem of the calculation of resonance absorption is a continuing program by L. W. Nordheim of this Laboratory. His theories

are used in all our computations; these methods have recently been described in GA-638(Rev.).⁽²⁷⁾ New and improved digital computer programs are being written under his supervision and will be available to us in the near future.

Nuclear Engineering Codes

Reactor analysis now requires high-speed digital computers. Their use enables the reactor engineer to design a reactor in considerable geometrical and material detail.

A number of National Laboratories have written criticality and life-time codes in one and two dimensions.⁽²⁸⁾ These codes form a consistent set of nuclear design tools. They have all been reviewed recently by a committee at General Atomic, and none has been found to be completely suitable for our nuclear design effort.

Some of the shortcomings are:

1. Most of the nuclear codes have been written for the IBM 704 and would require extensive revision for use on the IBM 7090. We expect to have this type of machine available for our use by the summer of 1960.
2. They do not provide for an arbitrary energy-transfer matrix.
3. The Naval Reactor groups at Bettis Atomic Power Laboratory and Knolls Atomic Power Laboratory are largely concerned with water systems. In their codes, cross-section subroutines are frequently included and are hard to alter. Cross sections which are generated for H_2O systems cannot be used for the HTGR.
4. The thorium cycle is seldom included in existing burnup routines.

Consequently, the following recommendations were made:

1. We should undertake the writing of a compatible system of nuclear codes that will allow the calculation of k_{eff} and reactor lifetime in one and two dimensions.
2. The new codes should be written for the IBM 7090 (or 709).

3. The codes should be written so that they may be changed easily, particularly with respect to input preparation, and the cross-section preparation routines should be kept separate from the space-dependent codes.
4. Provision should be made for the use of upscattering in the thermal energy region, if desired.

This system of nuclear codes would resemble quite closely that developed by Bettis. The major code types are outlined in Table 2.3.

Table 2.3
MAJOR CODE TYPES

Input Preparation Routines		Space-dependent Codes *	
Code Type	Examples	One Dimension	Two Dimension
Fast group cross sections	MUFT-IV	Criticality	
Slow group cross sections	CRISSCROSS	ZOOM (18 groups; arbitrary energy- transfer matrix)	ANGIE (18 groups; limited energy- transfer matrix)
Resonance integrals	Nordheim's codes	Lifetime	
Scattering kernels	KERNEL, KRYOS	GROM (18 groups; thorium cycle included; 70 fission products treated separately)	To be written at General Atomic
Thermal spectrum	SPECTRUM		
Group transfer coefficients	KITTY, GRAYMALKIN		

* Written for the IBM 709.

Of the input-preparation codes, all but the fast-group cross-section preparation code exist in workable form, and all were developed at General Atomic. They are distinct codes and separate from the reactor codes ZOOM, ANGIE, and GROM, all of which are products of Livermore. It was recommended that ZOOM, ANGIE, and GROM be obtained by

General Atomic and put into operation, thereby satisfying in large part items 1 through 4 listed above.

It was also recommended that we at General Atomic assist in writing the burnup routine to accompany ANGIE, using ANGIE itself as the basic criticality routine, and that we make whatever changes we feel are required in the other reactor codes. At this time, these recommendations have been approved and work has been initiated.

2.2. REACTOR DESIGN AND ENGINEERING ANALYSIS

Two basic types of fuel elements are being considered for the HTGR: a graphite-clad element and a metal-clad element. The metal-clad element is being considered only for the first core loading. Fuel elements are generally cylindrical in shape, 3-1/2 in. in diameter, and 12 ft long. The active section of the fuel element is 7-1/2 ft long and contains highly enriched uranium and thorium in the form of carbides. For the graphite-clad element, release of fission products into the main coolant stream is controlled by a combination of the use of impervious graphite and a venting system on each fuel element. A core loading consists of approximately 800 fuel elements arranged on a closely-spaced triangular pitch. Fuel elements are supported by a standoff on a grid plate at the bottom and receive lateral support at the top from the reflector and from their neighbors.

Fuel-element Design

Three fuel-element designs have been under development during the quarter. These are the metal-clad closed can, the all-graphite closed can, and the all-graphite vented can. A number of variations on these three designs may be used, such as changing the flow paths of sweep gas or varying the method of assembling wall section. .

The two major problems involved in the engineering development of the fuel elements are to keep fission products from migrating into the primary coolant stream and to maintain low stresses in materials,

particularly in the graphite. The fission products must be kept from the coolant stream so that components in the primary loop can be serviced. Stresses in the fuel element must be kept low to prevent the core from becoming so distorted that removal of fuel elements is difficult and to prevent failure of parts of the fuel element that would result in fission-product leakage.

Fission-product Release. Analyses were made on five types of all-graphite fuel elements to determine permeation requirements of the graphite wall cylinders:

1. A single-wall design with purge gas flowing around the fuel compacts;
2. A double-wall design with purge gas flowing in parallel around the fuel compacts and in channels between the double walls;
3. A counter-flow double-wall design in which the purge gas flows in series between the double walls and then around the fuel compacts;
4. A semiclosed can with purge gas flowing around the low-pressure inner can;
5. A closed-can design in which fission-gas pressure may build up in the inner can.

A good means for comparing leak characteristics of the different fuel-element designs is to specify the fraction of fission products that may leak out and then to calculate the permeation coefficient necessary for meeting this leak fraction. If a leak fraction of one part in 10^6 for I^{131} is assumed, the permeation coefficients established for each fuel-element design are those given in Table 2.4.

Since the counter-flow vented design imposes the least stringent graphite permeation requirements, more detailed leakage calculations were made for this design. Nuclides that are significant in yield, emission energy, and lifetime have been investigated as contributors to activity in the primary loop. Activity build-up in the primary loop at the end of

Table 2.4

PERMEATION COEFFICIENTS REQUIRED FOR FUEL ELEMENTS

Fuel Element	Permeation Coefficient (He at room temperature) (cm ² /sec)
Single wall	5.0 x 10 ⁻⁸
Closed can	1.7 x 10 ⁻⁷
Semiclosed	3.2 x 10 ⁻⁷
Parallel flow	1.4 x 10 ⁻⁵
Counter flow	2.0 x 10 ⁻⁵

given periods of operation was calculated for different values of permeation coefficient. Figure 2.6 shows activities of significant isotopes after 3 yr of operation, assuming that all nuclides leaking into the primary loop stay there until they decay. Figure 2.7 shows the activity as a function of time for a permeation coefficient of 10^{-5} cm²/sec in the case of the counter-flow vented element.

In the closed-can design, total pressure inside the can will build up from two sources. First, volatile fission products born in the element will contribute to the total pressure. Second, helium coolant will permeate through the graphite walls and reach an equilibrium pressure within the can. The original reference-design fuel element contained inner cans 9 in. long, stacked within an outer sleeve. Thus, a fuel can with a low permeability coefficient located in the hottest region of the core would build up high pressures because of the high local temperature and burnup. Pressure can be reduced somewhat by lengthening the inner can so that volatile fission products can diffuse to the cooler region of the lower reflector portion of the fuel element. The pressure build-up in the closed cans is shown in Fig. 2.8 as a function of permeation coefficient.

Pressure loads on the walls of long cans are not excessively high during reactor operation. However, when external helium pressure is dropped, the pressure differential across the can wall rises greatly.

Fig. 2.6--Activities of significant isotopes in primary loop after operation of counter-flow fuel element for 3 yr.
Complete plate-out is assumed

Fig. 2.7--Activity in primary loop as a function of time for a permeation coefficient of 10^{-5} cm²/sec in the counter-flow fuel element. Complete plate-out is assumed

Fig. 2.8--Pressure in closed can at end of 3 yr as a function of permeation coefficient for helium at room temperature

Stresses in Fuel Elements. Maximum stresses from pressure build-up in the inner can occur where the end cap joins the cylindrical wall. At this point, the end cap exerts a moment and a radial force on the wall. The stress from this pressure load is directly proportional to the pressure in the can. Therefore, the long can is more advantageous than the short can from a stress standpoint since the pressures are lower. Maximum stress values at the end cap for representative end-cap designs are given in Fig. 2.9.

Thermal stresses near the end caps, where pressure stresses are at a maximum, cannot be readily calculated. Calculations can be made for a location far from the end caps by using the theory for thermal stresses in a long hollow cylinder. For a temperature drop of 400°F through the total wall thickness of a fuel element, the tangential thermal stress is 1290 psi compressive on the inside surface of the cylinder and 960 psi tensile on the outside surface. At these surface points, axial thermal stresses will equal tangential stresses.

Steady-state Core Temperature Calculations

Three steady-state temperature studies were made using the CROFLO program (described in GAMD-682) to calculate local temperatures and flow rates. In each study, a graphite thermal conductivity of $15 \text{ Btu}/(\text{hr})(\text{ft})(^{\circ}\text{F})$ was used.

Surface Hot-spot Temperature of the Metal-clad Fuel Element. In these calculations, the surface hot spot of the metal-clad fuel element was studied as a function of power, heat-transfer correlation, and control-rod configuration. The inlet temperature of the coolant was 572°F in each case, and the average outlet temperature was 1015°F . The flow rate was adjusted for each power level to give the desired outlet temperature.

The following cases were studied, each as a function of power level:

Case I. Standard hot-spot allowances that were consistent with the use of a total of 49 control rods were made. Of these 49 control rods, it

Fig. 2.9--Pressure stress at end cap for representative end-cap designs

was assumed that 27 were needed to control excess reactivity at full power. The heat-transfer coefficient and pressure-drop correlations used were derived from a paper by Deissler and Taylor.⁽²⁹⁾

Case II. The standard hot-spot allowances were made except for the control factor. In this case, the total number of control rods was 25, of which seven were needed to control excess reactivity at full power. Burnable poisons controlled the remaining excess reactivity.

Case III. This case used the standard hot-spot allowances, with the control factor as in Case II. However, a less conservative heat-transfer correlation that was based on the results reported in Section 2.3 was used. The results are presented in Fig. 2.10.

Internal Hot Spot of the Graphite-clad Fuel Element. In these calculations, the internal hot spot of the graphite-clad fuel element was studied as a function of power, fuel-element diameter, and graphite thermal conductivity. In all cases, the coolant inlet temperature was 660°F and the average outlet temperature was 1380°F. The flow rate was adjusted for each power level to give the desired outlet temperature. Three fuel-element diameters and three graphite thermal conductivities were used in the study. Reactor power varied from 80 to 200 Mw (thermal). For each combination, two cases were considered; one in which the inner fuel can was present and one in which it was left out. The results are presented in Figs. 2.11 and 2.12.

Effect of Spacing Pads on the Internal Hot Spot of the Graphite-clad Fuel Element. A study was made of the effect of the presence of spacing pads on the internal hot spot of the graphite-clad fuel element for three slightly different fuel-element diameters.

The three diameters studied were 3.50, 3.52, and 3.54 in. The last case had no pads because there is no room for them with the pitch of 3.55 in. and the pad spacing of 0.01 in. For each case, the hot spot was calculated with and without pads. The hot-spot allowances with pads were standard. The hot-spot allowances without pads were standard except for

Fig. 2. 10--Surface hot-spot temperature for metal-clad fuel element as a function of power level

Fig. 2. 11--Internal hot-spot temperature of graphite-clad fuel element with no inner can

Fig. 2. 12--Internal hot-spot temperature of graphite-clad fuel element with inner can

the flow reduction. When no pads were present, it was assumed that adjacent fuel elements would bend and touch at the hot spot. It was assumed that the ends were rigidly restrained and that the flow area decreased linearly from the ends to the hot spot. Results of the calculations are given in Table 2.5.

Table 2.5
HOT-SPOT TEMPERATURE FOR THREE FUEL-ELEMENT
DIAMETERS WITH 3.55-IN. PITCH CONSTANT

Diameter (in.)	Temperature without Pads (°F)	Temperature with Pads (°F)
3.50	3590	3504
3.52	3513	3469
3.54	3426	----

Time-dependent Core Temperature Calculations

Three transient temperature studies were performed. In each case the time history of the hot spot or the average gas outlet temperature was desired.

In each of these three studies, the CROFLO code was used to calculate the initial conditions, and the RZ TEMP code (GAMD-1166) was used to calculate the time-temperature behavior after shutdown or power change.

Two approximate calculations of core temperature following shutdown without coolant flow have been carried out to examine the consequences of such a shutdown on the integrity of various core components.

Average Gas Outlet Temperature as a Function of Time and Flow Rate after Shutdown in the Graphite-clad Core. For these studies, the power at shutdown was considered to be reduced from full power to the afterheat only, and the flow rate to be reduced from full flow to a lower

value. The inlet temperature of the coolant was kept constant at 660°F during each study. Four cases were studied:

1. Full flow after shutdown (440,000 lb/hr);
2. 50% flow after shutdown (220,000 lb/hr);
3. 10% flow after shutdown (44,000 lb/hr);
4. 2% flow after shutdown (8800 lb/hr).

It was assumed that the average behavior of the reactor core would be close to the behavior of the average fuel element. The results are shown in Fig. 2.13.

Surface Hot-spot Temperature as a Function of Time and Flow Rate in the Metal-clad Core. In this calculation, the hot-spot fuel element was studied by following the temperatures as a function of time after shutdown. At shutdown, the power was reduced from full power to the afterheat only and the flow rate was reduced from full flow to a lower value. The inlet temperature was kept constant at 580°F during each study. No hot-spot allowances were made.

Two cases were studied, one in which the flow after shutdown was 300 lb/hr per element (50% of full flow) and one in which the flow after shutdown was 60 lb/hr per element (10% of full flow). The results are shown in Fig. 2.14.

Average Gas Outlet Temperature as a Function of Time after a Sudden Change in Power Level. In this calculation, an average fuel element was studied by following the coolant outlet temperature as a function of time after the power change. At time zero, the power was suddenly increased by 22%, while the flow rate and coolant inlet temperature remained unchanged at 516 lb/hr per element and 662°F , respectively.

The average gas outlet temperature was calculated for 3 min after the change in power. During this period, the average rate of gas temperature increase was about $11^{\circ}\text{F}/\text{min}$. To reach the new steady state, the outlet gas temperature would have to rise by 187°F .

Fig. 2.13--Average gas outlet temperature after shutdown for four coolant flow rates

Fig. 2.14--Surface hot-spot temperature of metal-clad fuel element after shutdown for two coolant flow rates

Temperature-time History of a Graphite-clad Fuel Element after Shutdown without Cooling. In this calculation, the generation of afterheat and the transfer of heat in the axial direction by conduction and radiation were taken into account. It was assumed that stoppage of the coolant flow and reactor scram occurred simultaneously. The conductivity of graphite at high temperatures was assumed to be $15 \text{ Btu}/(\text{hr})(\text{ft})(^{\circ}\text{F})$.

It was concluded that metallic parts (stainless steel) at the bottom end of the fuel element would reach failing temperatures after a period of the order of 2 hr after shutdown. This study is reported in GAMD-1064.

Temperature-time History of Control Rods after Shutdown without Cooling. This calculation was based on the same conditions and assumptions as the previous one except that axial heat flow was ignored. It was assumed instead that the control-rod guide tube received heat only by radiation from the fuel elements, and that this heat was then conducted by the graphite guide tube to the control rod within it.

It was concluded that if the portion of the control rod that would be located deep within the core on shutdown contained any stainless steel parts, these parts would reach failing temperatures within 2 to 3 min after shutdown.

Development of Codes

RAT, a rather versatile code for the IBM 704, has been developed to calculate steady-state temperatures in a cylindrical geometry. The versatility of the code is obtained by utilizing the FORTRAN II subroutine system. The basic code accepts its data only in a standardized fashion, but the subroutines (at least one for each material) translate the data of each special case into the standardized form.

The code will calculate heat transfer by conduction and radiation across a large number of gaps, both perpendicular to the axis and circular cylindrical. The thickness of a gap can be made temperature dependent to allow for thermal expansion. Heat transfer may take place in any desired

manner across the top and bottom faces and across the inside and outside cylindrical surfaces.

All physical properties and heat generation may be made space dependent and temperature dependent. The radial and axial space increments may be variable to suit the geometry and temperature gradients.

Temperatures are calculated in a two-dimensional (radial-axial) finite-difference mesh. The solution is obtained by successive iterations, using the Peaceman-Rachford method.⁽³¹⁾

The code is now in the trouble-shooting stage. It is expected to be very useful in predicting internal temperatures for capsule experiments and fuel elements. A time-dependent version is being developed to study fuel-element behavior.

2.3. EXPERIMENTAL ENGINEERING

Reactor Core Heat-transfer Experiment Studies

The coolant-flow passages between the fuel rods are of a tricuspid shape. The effect of this shape is to cause the local rate of heat transfer at the surface of a rod to be a strong function of peripheral position. The nonuniformity of the local heat-transfer rates gives rise to somewhat different average values than those predicted by conventional empirical relationships. Therefore, the local and average heat-transfer film coefficients were measured under flow conditions similar to those in the HTGR core, using air as working fluid.

The experimental evaluation of a particular variable requires that the values be cross checked by different techniques and by the measurement of other related variables. The average and local film coefficients were determined by different techniques; in addition, two related variables, coolant velocity profiles and friction factors, were measured as further cross checks.

The initial measurements of the average film coefficients are shown

in Fig. 2.15 as a function of Reynolds number. The film coefficients were measured at length-to-diameter ratios (L/D_e) of 39, 59, and 75.5 from the entrance of the coolant passage without any significant difference in values. This indicates that coolant turbulence has largely become established between the entrance to the passage and an $L/D_e = 39$. The measured values are closer to those for a round pipe than those predicted by Deissler and Taylor.⁽²⁹⁾

The peripheral variation of the local film coefficient as a function of angular position is shown in Fig. 2.16 for a Reynolds number of 20,000 and an L/D_e of 39. There is little change in the values at angles greater than 15° from the center of the smallest gap between the rods. The ratio of the highest to the lowest local film coefficients measured is approximately one-seventh of that predicted by Deissler and Taylor.

The friction factors, calculated from measured axial static pressure gradients, are shown in Fig. 2.17 as a function of Reynolds number. Six static pressure taps located at an L/D_e of 40 and 51 were used to measure the pressure drops. Two sets of data were taken, one with a rounded entrance and one with a screen (30 mesh) across the entrance. As expected, there is a difference in the friction factors at the lower flow rates; however, no change is evident above a Reynolds number of about 10,000, which indicates that the undisturbed hydrodynamic entrance length at this Reynolds number is somewhat less than $40 L/D_e$. The shape of a curve drawn through the data taken with the rounded entrance is interesting in that it appears that the coolant does not become fully turbulent until a Reynolds number between 7000 and 9000 is attained. The measured friction factor at the design Reynolds number for the HTGR core is closer to that for a smooth pipe than that calculated by Deissler and Taylor.

Preliminary measured isovelocity lines are shown in Fig. 2.18 for $R_e = 20,000$. The measurements were made at an L/D_e of 51. The influence of the passage wall (i. e., fuel rod) upon the shape of the isovelocity lines is consistent with the data reported by Nikuradse,⁽³⁰⁾ in contrast

Fig. 2.15--Comparison of measured and predicted heat-transfer coefficients for pitch-to-diameter ratio of 1.01

Fig. 2.16--Peripheral variation in heat transfer

Fig. 2.17--Friction factors as a function of Reynolds number

Fig. 2.18--Comparison of measured and calculated isovelocity lines for Reynolds number of 20,000 and pitch-to-diameter ratio of 1.015

to the results predicted by Deissler and Taylor. The rate of change of velocity gradients with respect to perpendicular distance from the fuel rod appears to be rather low until a position approximately 10° to 15° from the center of the smallest gap between the rods is reached. In addition, a plot of the velocities near the rod surface as a function of distance from the surface revealed that in the center section ($20^\circ < \theta < 40^\circ$) the velocity profile approximates the conventional $n^{1/7}$ shape, where n is the dimensionless distance from the wall.

The measured average and peripheral film coefficients appear to be satisfactorily cross checked by the coolant velocity profile and pressure drops thus far measured. Additional measurements and analysis are necessary in order to ensure the accuracy of the values reported here. Confirmation of the hydrodynamic and thermal entrance lengths under these conditions would be desirable if the data reported here are to be used with confidence in future heat-transfer calculations for the core. In addition, another variable to be considered is the effect of rod spacing and misalignment upon these measurements. Convection studies will continue and will be reported in more detail at a later date.

Measurements of Leakage between Standoff and Fuel Element

One portion of the HTGR fission-product trapping system is the standoff support for the fuel element which provides for fission-product discharge to the trapping system. In order to ensure a suitably low rate of leakage between the standoff and the fuel element, an experimental investigation was initiated to develop an applicable seal. Several standoff seal designs and their modifications have been tested under HTGR conditions.

The seal showing the lowest leak rate of those investigated to date is a conical plug type seal. Results of the tests of this design are shown in Fig. 2.19. Other favorable characteristics of this design are its relative simplicity, machinability, and its moderate parting force (dead-load force required to break the seal).

Fig. 2.19--Leak rate between standoff and fuel element as a function of helium pressure

2.4. NONNUCLEAR MOCKUP OF CORE

Design calculations have been made on scale models of the reactor core (GAMD-1135). These calculations will lead to the selection of the model scale and to detailed design work on the model in 1960.

The selection of the model scale is a compromise between flow considerations and mechanical considerations. Calculations indicate that both the design Reynolds number and the pressure drop for the reactor core can be achieved in an air-flow model if the model scale is approximately 0.4. However, for use in solving mechanical problems, a full-scale model is most desirable.

Inquiries as to the price and availability of blowers suitable for use on the model have been made.

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TASK III. EXPERIMENTAL REACTOR PHYSICS

3. 1. NEUTRON THERMALIZATION

To complement the theoretical physics effort on neutron thermalization in graphite, a series of experiments has been undertaken using the General Atomic linear accelerator. Some of these experiments will serve as a check on theoretical calculations of thermal neutron spectra in infinite graphite assemblies (poisoned with $1/v$ and non- $1/v$ absorbers) as a function of temperature. Other experiments will measure the spatial dependence of the thermal neutron distribution across a temperature and absorption discontinuity similar to that encountered in the HTGR at the core-reflector interface.

In particular, the current experimental program includes the following:

1. The measurement of thermal neutron spectra in a graphite block that is poisoned with boron and heated to temperatures up to 600°F .
2. The determination of the influence of a non- $1/v$ poison on the thermal neutron distribution in the graphite block at room temperature.
3. The measurement of the spatial dependence of the thermal neutron spectrum across an absorption and temperature discontinuity.

The heated medium will be poisoned.

The poison concentrations in these experiments will correspond to the number of barns per carbon atom at 2200 m/sec expected in the HTGR.

The physical geometry for these experiments is shown in Fig. 3. 1. The linear-accelerator electron beam produces a very intense burst of neutrons at the center of a 4-ft cube of graphite which is surrounded by heater panels. Sheets of boral in the heater panels absorb stray neutrons from the room which might scatter into the assembly. An independent collimator system holds the neutron chopper, shielded flight path, and detector bank. This collimator setup can be positioned with respect to the

Fig. 3.1--Chopper facility for spectrum measurements

assembly so that the spectra can be measured as a function of distance from the neutron source--an important experiment to establish the validity of the one-dimensional space-energy calculation. With this geometry, there is the capability of measuring spectra to 5-ev neutron energy.

A block diagram of the electronic system constructed for this experiment is shown in Fig. 3.2. Basically, the experiment proceeds as follows: A light impulse from the chopper provides a trigger signal, which, after suitable delays, triggers the linear accelerator and thus produces neutrons. The light signal also gates on the time-of-flight analyzer so that the time spectrum of neutron counts at the detector bank can be registered. The signal to the accelerator is delayed by a fixed delay time in the Electro-pulse Company double pulser and by a programmed variable delay in the "wobulator." With this combination of delays, one is able to pulse the accelerator in exactly the right time region after the chopper light pulse to permit the appropriate neutrons to escape the assembly, pass through the chopper, and be detected. Herein lies a big advantage of this experimental technique over that normally encountered with a steady-state drive source, in that one is not producing background with the neutron source while trying to detect the chopped neutron beam. Another advantage is the simplicity, cleanness, and flexibility of the experimental setup made possible by having essentially a point neutron source in space.

A test program of measuring spectra in known assemblies has been carried out with this experimental arrangement. Spectra in borated polyethylene and pure polyethylene have been studied intensively, and the results obtained with the chopper technique appear to be close to those measured at General Atomic by a pulsed-neutron technique which does not require a chopper.

The neutron spectrum in a 4-ft cube of graphite poisoned with borated stainless steel to 0.4 barn per carbon atom (at 2200 m/sec) has been measured at room temperature. This poison concentration is close to that expected in the operating HTGR. The hardening parameter $\Sigma_A / \xi \Sigma_S$

Fig. 3. 2-- Block diagram of electronic system for spectrum measurements

for the spectrum is about 0.5. Experimental data have not been reduced as yet, but it is clear from the raw data that the neutron spectrum is considerably harder than that for borated polyethylene ($\Sigma_A/\xi\Sigma_S = 0.30$), peaking between 0.05- and 0.1-ev neutron energy with no very pronounced transition to the $1/E$ neutron spectrum.

Improvements in the experimental setup will continue to be made during the next few months, while more of the vital experimental data are being obtained.

3.2. DESIGN AND CONSTRUCTION OF THE CRITICAL FACILITY

Design and Construction Program

The design of a critical facility for the HTGR has been completed and the majority of the items have been ordered. A building to house the assembly is being erected by the General Atomic Division (not under this contract), and completion is scheduled for April 1, 1960. The fuel elements have been ordered, with delivery expected by the time the building is completed. Mechanical and electrical components will begin arriving in February, 1960. The first criticality is planned during April, 1960.

The critical assembly is designed as a graphite-moderated stack which separates along the vertical midplane to provide a large safety-shutdown factor during work on the assembly (see Fig. 3.3). The fuel is in the form of enriched uranium oxide. It is mixed with graphite and compressed into 1.5-in. -diameter compacts which are placed into holes in graphite blocks to form fuel elements, as shown in Fig. 3.4. In this way, the uranium is in intimate contact with a part of the moderator, making possible a prompt negative temperature coefficient. In addition to uniformly loaded clean critical experiments, multiregion experiments are also planned. For these experiments, a relatively small region of an exact mockup of an HTGR lattice can be surrounded by a flux-matching (buffer) region, which is, in turn, surrounded by a driver region to provide a critical system.

Fig. 3.3--The HTGR critical assembly

Fig. 3.4--Graphite fuel-container block

The floor plan of the entire facility is shown in Fig. 3.5.

Critical Facility Safeguards Analysis

The following problems have been studied in order to analyze hazards and to design a safe critical assembly:

1. Determination of the dependence of reactivity on table separation and specification of acceptable table closure speeds.
2. Calculation of energy release and temperature increase associated with the maximum credible accident.

Calculations were done by the method of Tamor,⁽¹⁾ which has been semiquantitatively confirmed by experiment.⁽²⁾ For small table separations, it was found that the HTGR critical assembly of greatest interest shows a reactivity dependence of about 3 dollars/in. for small separations. By using three different table speeds, with the slowest for the smallest gaps, it was ascertained that closure from 3-ft separation can be achieved in about 6 min, without violating the condition that the rate of reactivity increase for reactivities within about 1 dollar of criticality be limited to 0.02 dollar/sec.

Several computer codes have been used to make detailed computations of the start-up accident that could occur if ramp insertion of reactivity is continued after criticality has been reached. The incidents were assumed to be limited only by the negative temperature coefficient, for which the conservative value -3×10^{-5} per $^{\circ}\text{C}$ was assumed. These calculations predict a power surge within several seconds after prompt criticality has been reached, followed by a slow decline continuing over a number of seconds. The maximum power reached and the temperature increase at a given time show considerable sensitivity to the rate of ramp insertion and to whether or not the reactivity insertion is terminated at 1 dollar. However, these two characteristics of an incident show little sensitivity to the neutron source strength present at start-up.

As a result of the calculations, it has been determined that:

Fig. 3.5--Floor plan of the critical assembly building

1. The rate of table closure within about 1 dollar of criticality should be limited so that the rate of reactivity increase is 0.02 dollar/sec.
2. The slow increase of temperature after the power surge allows ample time for manual termination of the incident by interruption of a circuit which opens a valve and permits the energy stored in the hydraulic system to drive the tables apart.
3. On the basis of the foregoing considerations, the maximum credible accident is a postulated accident resulting from a gross loading error, together with failure of safety systems, which causes reactivity to be added at a rate of 0.02 dollar/sec, with the reactivity being inserted beyond prompt criticality. The accident is terminated by a manually activated hydraulic separation of the reactor halves within a conservative time of 4 sec after the 1-Mw level is reached. This accident results in a peak power of 750 Mw and a temperature rise in the ceramic fuel elements of 800°C , at most. The energy release is about 1800 Mw-sec. Because of the compounded conservatism of assumptions of both the conditions and numerical values used in this analysis, the above figures represent upper bounds for the energy released in any real incident.

Control-rod and Safety-rod Worths for Critical Assembly

The worths of control and safety rods in the HTGR critical assemblies have been estimated using PDQ⁽³⁾ and the methods developed in WAPD-218.⁽⁴⁾ These calculations used the same three fast groups as the temperature-coefficient analysis, but they used only one thermal group, in place of the six used for the temperature coefficient calculations in Section 2.1. The safety and control rods were assumed black in the thermal group. The location of the safety rods is shown in Fig. 3.6. Results are given in Table 3.1 for safety-rod widths of 14 and 7 in.

The two control rods, 1.2 in. wide, were located at $X = 13$ in. and were symmetrically placed on either side of centerline $X_0 - X_0$. The calculated $\Delta k/k$ was 2.2%.

Fig. 3.6--Schematic representation of the critical assembly used in safety-rod and control-rod estimates

Table 3.1
 WORTHS OF SAFETY RODS

Coordinates (in.)		Rod Size (in.)		$\Delta k/k$ (%)	
X	Y	Thickness	Width	4 rods	1 rod
13	6.5	0.25	14	9.2	2.3
13	6.5	0.25	7	7.8	1.95

Changes in Reactivity Associated with Fuel-element Loading and Storage

To aid in the planning of fuel-loading schemes for the HTGR critical assembly, k_{eff} was calculated as a function of height for an assembly 4 ft long and 4 ft wide. These results are shown in Table 3.2.

Table 3.2
 k_{eff} FOR VARIOUS HEIGHTS

Height (in.)	k_{eff}
42	0.93024
45	0.95706
48	0.98043
50	0.99343

An analysis was also made to determine if a critical configuration could be obtained in the proposed fuel-storage facility. In this facility, the fuel-bearing graphite blocks will be stacked 8 ft high against a concrete wall. Six cadmium sheets, each 20 mils thick, will be included in the stack at 1-ft vertical intervals, and they will run the entire 18-ft length of the stack. For the criticality calculations, an impossible configuration was chosen: the two stacks, which will be located against each of the longer walls of the storage facility, were assumed to come together intact, resulting in a stack 2 ft by 8 ft by 18 ft. It was further assumed that the

room would be flooded with water so that the stack would be water reflected. For this configuration, k_{eff} was calculated to be 1.08. Should the water subside, then k_{eff} would be reduced to 0.82. Another calculation showed that a critical configuration could result if the thickness of the 8-ft by 18-ft bare slab were increased from 24 in. to 29 in., with no cadmium in the system.

The following parameters are useful for order-of-magnitude estimates of critical configurations of the HTGR critical assembly:

$$k_{\infty} = 1.882,$$

$$L^2 = 102.03 \text{ cm}^2,$$

$$\gamma = 281.4 \text{ cm}^2.$$

Here the η and f components of k_{∞} and L^2 are obtained from spectrum-averaged cross sections. These averages were taken over the energy range from 0 to 20 ev.

Experiments for the Critical Assembly

Planning of HTGR critical experiments is well under way, and certain features of the early experiments have been determined.

The first critical assembly will be a small, flexible, multiregion reactor for studying control-rod effectiveness, phenomena associated with resonance capture in thorium, and reactivity coefficients. The assembly consists of a central region, a buffer, a driver, and a partial reflector. The over-all dimensions of the assembly are about 6 ft. The assembly or parts of it can be heated to about 500°F. Provision will be made for measuring reactivity to within about 0.01 cent. Development of an oscillation technique for the precise measurement of small reactivity changes has been started. This method will be particularly useful for measuring the Doppler coefficient of thorium in graphite.

The central or "exact" region, about 1 ft in diameter, will have fuel elements resembling those of the actual HTGR. The buffer and driver

regions are composed of blocks of graphite containing cylindrical holes which can be loaded with pure graphite or with graphite containing either fissile or fertile material. Fuel compacts of two concentrations have been ordered so that the over-all carbon-to-uranium ratio can be varied from 1900:1 upwards.

The buffer region, which will be several feet across, will have blocks containing thorium in one hole and U^{235} in the other, with concentrations that match those in the exact region. It has been ascertained that thermal self-shielding effects are comparable in these two regions and are less than 10%.

The worth of a single control rod will be determined in several ways. All these experiments will be performed in the exact region. First, an HTGR control rod will be inserted and its worth determined by inverse multiplication measurements. Second, the rod will be removed, and the reactivity change will be reproduced by distributing boron uniformly throughout the region. Finally, the control-rod worth will be measured in the original configuration, using pulsed neutron techniques. The General Atomic electron linear accelerator will be used as the pulsed neutron source.

The thorium resonance integral and other absorption effects will be measured in the central region. These experiments will encompass a variety of fuel and fertile loadings, under various conditions of temperature and geometry. The resonance integral of thorium will be determined by both reactivity and activation experiments.

In the reactivity experiments, reactivity will be measured as a function of thorium concentration. The reactivity changes in these substitution experiments are expected to be several cents; they will not require oscillator techniques. The Doppler coefficient of thorium, however, will be measured by oscillating a hot and a cold fuel element in the central region. The thorium detector foils used in the activation experiments will be thin with respect to an absorption mean free path at resonance peaks.

We anticipate that the accuracy of the resonance-integral measurement will be about 1%, and that of its temperature derivative, about 10%.

At a later date, it is planned to assemble a single-region core with a reflector that will have dimensions somewhat similar to the HTGR design. The use of all 80 kg of fuel in such an assembly will provide a good approximation to the actual HTGR, at least at the end of life. Studies of gross criticality, control-rod array phenomena, temperature effects, and neutron spectra are planned in this assembly.

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TASK IV. DEVELOPMENT OF REACTOR EQUIPMENT

4.1. PRESSURE VESSEL AND INTERNALS

The pressure vessel of the HTGR is approximately 14 ft ID and 37 ft tall. It is a low-pressure vessel (300 to 350 psi pressure) and will probably be constructed from carbon or low alloy steel. There are a number of penetrations in the bottom of the vessel for control rods and for fuel handling, and in the top of the vessel for fuel handling. The main coolant nozzles will be located on the sides of the vessel. Fuel elements are supported by a circular grid plate located near the bottom of the pressure vessel. Lining the inside of the pressure vessel near the core region is a graphite reflector approximately 12 ft tall and 2 ft thick. The reflector is segmented, and some of the blocks are pivoted so that they can provide lateral support for the fuel elements. Steel thermal shields and shrouds adjacent to the pressure vessel reduce the nuclear heating in the vessel to an acceptable value and direct the flow of gas within the vessel.

Cooling of Vessel

1. A study of cooling of the vessel by means of bypassing coolant around core and into upper plenum showed that this method was satisfactory to cool the vessel. However, the bypassed gas dropped the reactor outlet gas temperature and reduced the efficiency of the plant.

An investigation of two-pass cooling of the vessel by introducing the coolant through a bottom inlet showed that this method could keep the vessel cool under steady-state conditions.

A study of the use of internal insulation and external vessel cooling showed that this scheme was feasible. However, it was necessary to remove the external insulation, and this resulted in a high loss in usable heat.

2. The computation of the temperature of gas in the conceptual

design showed that the gas temperature in the plenum rose above 700°F when the flow rate was reduced. This results in the inability of the gas to keep the vessel cooler than 700°F . This effect can be eliminated by modifying the original concept so that the entering gas passes directly along the wall of the pressure vessel.

3. Temperatures in the vessel during emergency cooling are being determined.

Pressure Drop

1. The pressure drop from the vessel inlet to outlet (exclusive of the pressure drop within the core) for the two-pass bottom-inlet concept was computed to be 0.75 psi. This study included a preliminary investigation of the cooling requirements of the graphite reflector.

2. The pressure drop of the modified conceptual design is now being computed.

Mechanical Design

1. The vessel wall, nozzles, and flanges have been sized for pressure loads and were designed to meet the requirements of Section I of the ASME Code. The thickness required for the main vessel wall is 2-5/8 in. for a design pressure of 420 psi and a design temperature of 725°F .

2. The computed thermal stresses are less than 9000 psi with no thermal shields, but the vessel temperature is 777°F . With three shields, the thermal stresses are 652 psi and the temperature of the metal is below 700°F .

3. A preliminary deflection analysis of the core support was made to establish the thickness of the web plates and the depth of the supports. A 3-in. core support plate requires 1-in. web plates, 24 in. deep.

4. A preliminary design of vessel support required for the dead weight and the thermal expansion of vessel was made to assure that the stress in the vessel was below the allowable limit given in the ASME Code. A total of 24 supports is required to keep the stress at an acceptable value.

5. A preliminary evaluation of the stresses in the vessel from transients occurring with small load variation showed step changes of 25°F could be made without using a lower steady-state design stress. This means close control of these variations is not as necessary as with other reactor coolants, such as water or liquid metals.

A literature survey concerning irradiation effects on structural materials is being made. A preliminary specification to be used for identifying design problems has been sent to several pressure-vessel fabricators.

One set of preliminary drawings was made for the purpose of evaluating alternate coolant inlets. One set of drawings was made to show details of the manufacture of vessels and internals, flow path and orifices for coolants, support of internals, and vessel supports.

4.2. CONTROL RODS AND DRIVES

The control-rod drives on the HTGR are located below the reactor. Reactivity is increased by removing a poison section down and out of the core; reactivity is decreased by inserting the poison section up into the core. The rods are scrambled upward against gravity. Control rods operate in graphite guide tubes which have essentially the same external dimensions as the fuel elements; thus, the control rods do not destroy the symmetry of the core. The poison section of the control rods will probably be boron carbide in graphite.

Control-rod Drives

The major effort on the control-rod drives during the last three months has been concentrated on an investigation and evaluation of possible approaches to a control-rod drive system.

To date, the investigation has been concerned with three types of drives: the original ball-screw drive with modification, a rack-and-pinion drive with pneumatic scram, and an all-pneumatic stepping drive.

The selection of a drive system will be based on the general requirements listed below.

1. High mechanical reliability:
 - a. A minimum of mechanical complexity;
 - b. Rugged components;
 - c. Low functional sensitivity to wear and deformation.
2. High fail-safe reliability:
 - a. Continuous standby power for scrambling, with at least two independent energy sources available;
 - b. A positive means of locking the drive to prevent its falling out of the core from any position;
 - c. Positive fail-safe interdependence between control and power circuits.
3. High control reliability:
 - a. Minimum control-circuit complexity;
 - b. Continuous rod-position monitoring;
 - c. Short signal-response time.
4. Servicing accessibility:
 - a. Ability to remove individual drives without disturbing adjacent drives;
 - b. Attachment features that will minimize the time that maintenance personnel must be present in the room below the reactor;
 - c. Mechanical interchangeability to allow complete drive replacement at the reactor, with drive maintenance performed in a normal service area.
5. Low cost:

Cost considerations include the development and prototype cost, production cost, and procurement cost (including availability of materials and of manufacturing or fabricating ability).

Control Rods

Calculations of the control-rod temperatures indicate that with no coolant flow after a full-power scram, the center of the control rod will reach approximately 3000°F within a few minutes. With the stainless steel control-rod joints proposed in GA-593,^{*} at least some of the control rods would be damaged if cooling was not initiated within 2.5 min after shutdown.

In addition to the control rod with stainless steel joints, a control rod with a flexible graphite center support rod has also been designed. Both designs utilize boron carbide-graphite sleeves as an absorber material. A test model of the stainless steel design is now being fabricated.

Plans are being made to irradiate several boron carbide-graphite samples. Results of these tests, the first of which should become available in the third quarter of 1960, are expected to answer questions concerning gas formation in the control rod.

4.3. FUEL-HANDLING EQUIPMENT

Fuel handling in the HTGR is accomplished only after the reactor is shut down, the core cooled down, and the pressure in the vessel reduced to slightly less than atmospheric. A top transfer machine, which is inserted into the central port on the top of the vessel, is used to remove the fuel elements from the core one by one. The machine is positioned remotely over the fuel element to be removed, and it lifts the fuel element vertically until the element is free of the core. The machine rotates and then lowers the fuel element out of the reactor through a hole in the graphite reflector and the bottom of the pressure vessel. After leaving the bottom of the pressure vessel, the fuel element is canned in a metal container to prevent

^{*} High-temperature Gas-cooled Graphite-moderated Power Reactor: A Prototype Nuclear Steam-generating System, General Atomic Report GA-593, November, 1958.

fission-product dispersion and is transferred to a storage pool. Loading of a fuel element is accomplished in essentially the reverse manner of fuel-element removal.

During fuel transfer, the maximum discharge-gas temperature has been tentatively established at 400°F . This limit is set by the use of standard electrical equipment for the grapple head. Preliminary calculations indicate that fuel transfer can be accomplished without special cooling of the element being transferred. A temperature rise of less than 60°F is expected during the transfer operation.

Preliminary calculations indicate that 1 day after shutdown the dose rate on the grapple head of the top transfer machine will be 10^4 to 10^5 r/hr. Therefore, a reasonable service life can be expected from the electrical components of the grapple.

A preliminary calculation of the required lead thickness on the transfer coffin indicates that approximately 9 in. of lead is required to reduce the dose rate in the room below the reactor to an acceptable value during fuel changing.

Top Fuel-transfer Machine

During the last quarter of 1959, several designs for the grapple head of the top fuel-transfer machine were studied. The initial design featured mechanisms that sensed the position of the grapple head relative to the fuel element. Misalignment of the grapple-head forward section caused a wobble plate to change its position. When transmitted through a system of linear variable differential transformers and a control circuit, this motion was used to move the radial index and azimuth mechanisms to the correct alignment over the fuel element.

Later designs featured self-aligning mechanisms that adjusted any misalignment of the grapple head relative to the fuel element.

Transfer Coffin

An experimental program to determine the shape and form of the end closure for the canisters containing spent fuel elements was initiated during this period. The dies were made from plastic blocks to facilitate modification of the die contour. End closures on several aluminum tubes with an outside diameter of 4-1/2 in. and a wall thickness of 0.035 in. have been successfully formed. Satisfactory closure of the metal canister is achieved by flattening the tube into a shallow circular arc with a tapered section extending into the tube.

The experience and technical know-how obtained during the forming of the metal tube in the plastic die are now being used to design a set of metal dies. A prototype of the operating mechanism required in the transfer coffin is also being designed and will be used to operate the metal dies. Upon the completion of the design and manufacture of the equipment, tests will be undertaken to develop the technique required to seal a metal canister for spent fuel elements.

Top Fuel-transfer Closure Valve

Two designs for the valve at the top of the reactor have been started during this quarter. Both valves are designed to provide sufficient shielding against gamma radiation to protect personnel during the operation of the fuel-handling equipment.

TASK V. DEVELOPMENT OF PRIMARY-LOOP EQUIPMENT AND REACTOR PLANT LAYOUT

5.1. STEAM GENERATOR

A survey was made of the literature on conventional boiler design and operating experience in this country. A report summarizing this survey is being written on the following principal subjects: (1) steam generator types, (2) tube-failure problems, (3) cost of steam generators, and (4) control of steam generators. As a result of this survey, it was concluded that a drum-type boiler represented the best choice for the HTGR at the present time. The volatile type of feedwater treatment was selected for a design bases.

Preliminary design criteria for the steam generator were prepared and distributed to seven vendors. Evaluation of the returns has indicated four potential suppliers.

An IBM 704 code was prepared to analyze several designs of interest under part-load conditions. Studies have been made of once-through steam generators and of steam generators with low recirculation rates. Boilers employing forced recirculation with high recycle ratios are now under study.

For steam generators that employ low circulation rates through the boiler section, the following results have been obtained for the reactor plant that uses graphite fuel elements. If the steam is to be held at the design value of 1000°F and 1500 psig, it will be necessary to decrease the hot helium temperature from 1367° to 1167°F when the steam flow rate is reduced from 100% to 30% of design value. These data are being used to analyze the transient response of the reactor-steam generator combination, as based on a required 3%/min change in steam flow. At the part-load condition described, the temperature of the cold gas entering the reactor will be reduced to about 500°F .

5.2. MAIN HELIUM COMPRESSORS

During this quarter, studies were initiated to establish major design parameters for the main helium circulators. Design possibilities under consideration include

1. Various methods of flow control.
2. Canned or shaft-seal construction.
3. Vertical or horizontal mounting.

A comparison was made of the cost and advantages of various methods of flow control. Design arrangements of adjustable inlet guide vanes were studied to make certain that this type of device could be properly serviced. It was concluded that adjustable inlet guide vanes represent an acceptable method of flow control.

During December, a meeting was held at Oak Ridge National Laboratory on the sealing of compressor and gas turbines for gas-cooled reactor systems. The information obtained at this meeting will implement our study of canned versus shaft-seal construction.

5.3. PRIMARY DUCTS AND VALVES

The problem of internally insulating and cooling the reactor coolant outlet pipe was investigated for a concentric-pipe system and for several single-pipe arrangements. The design problems associated with isolation valves for the primary helium loop were also studied. Inquiries were sent to suppliers of valves, internal pipe insulation, and bellows expansion joints. Proposals from manufacturers of bellows expansion joints indicate the availability of reliable joints that would have adequate lifetimes under the pressure, temperature, and cycling conditions required at the HTGR compressor suction and discharge areas. In addition to reducing the pressure-drop and space requirements, these joints would relieve the force and moment loadings on the compressor nozzles.

It has been established that acceptable pipe heat losses for either a

concentric-pipe system or a cooled single reactor outlet pipe can be maintained with moderate thicknesses of internal insulation made with layers of corrugated or dimpled stainless steel. Because of its ruggedness, this type of insulation is preferred to the fibrous or wool types. Experimentally determined conductivity values have been obtained to verify and supplement calculated values. Preliminary designs have been made for a concentric pipe, a water-coil-cooled single pipe, and a forced-convection air-cooled single pipe. At the present time, the air-cooled single pipe appears to have the fewest design problems.

Isolation valves for the reactor outlet pipe will be a developmental item. Studies based on a report prepared for the MGCR Project by the Crane Company and on proposals from other manufacturers indicate that a valve with a single seat would not pose as great a developmental problem as a valve with a concentric seat. Isolation valves for the cooler pipes between the steam generator and compressor and at the outlet of the compressor can be ASA standard 300-lb gate valves, except for the special requirements that they must be capable of remote operation and that they must be leaktight to the atmosphere.

It is hoped that during the first quarter of 1960 an evaluation of the types of pipes and valves to be used will be completed and the preliminary design criteria for a prototype pipe section and valve will be established.

5.4. REACTOR PLANT LAYOUT

A series of layout sketches was made to determine the effect that various solutions to several of the fluid-system design problems would have on the helium-loop and containment layout. Several methods of absorbing the thermal expansion of the piping were investigated. Studies of neutron streaming and shielding requirements were initiated.

The layout studies indicated that from the layout standpoint the following choices are preferable: (1) a two-boiler system, rather than a one-boiler system; (2) a cooled single-pipe system, rather than a concentric-

pipe system; and (3) the use of expansion joints or roller mounts for the compressors, because of the significant savings in the system pressure drop and containment space that could result. Layout sketches are shown in Figs. 5.1 and 5.2 for the one-boiler and two-boiler systems, respectively,

A study of the neutron-streaming problem in the reactor pipe connections is under way at the Electric Boat Division of General Dynamics. Preliminary results indicate that at least one, but not more than two, 90° changes of direction in the shield penetration will be required to keep the activation of boiler material to an acceptable level. Estimates of the required shield thickness and of the radiation heating of the concrete are being made by the HTGR physics section.

Studies directed toward defining the main coolant system and its components have been made. To facilitate use of the information, it has been put into the form of a process flow diagram that shows the primary pieces of equipment, the direction of flow, type of flow medium, and the quantities, temperatures, and pressures. When these data have been determined, the system will be defined sufficiently to permit a more specific mechanical definition, which will be in the form of a piping and instrument diagram. This diagram will define the size and type of piping, the location and type of instruments, and the size and pressure-temperature rating of vessels, exchangers, pumps, and other components. This mechanical definition will permit the preparation of specifications for bids. The piping and instrument diagram also will aid the preparation of final working drawings for the piping.

The first draft of the process flow diagram for the main coolant system has been completed on the basis of the single-boiler concept (see Fig. 5.3). In addition, an alternative design is being prepared for the concept that uses two steam generators. It is planned to prepare the first draft of the piping and instrument diagram during the first quarter of 1960.

Fig. 5.1--Layout study for one-boiler system

PLAN BELOW DECK

Fig. 5.2--Layout study for two-boiler system

Fig. 5.3--Process flow diagram for main coolant system

TASK VI. DEVELOPMENT OF PLANT AUXILIARY SYSTEMS

6. 1. HELIUM PURIFICATION SYSTEM

A study has been completed of various methods of purifying helium, and a process flow diagram of the optimum helium-purification system has been prepared (Fig. 6. 1). A study has been completed which relates the purification flow rate, the steam leakage rate, and the carbon transport rate (GAMD-1139). With present estimates of the boiler leaktightness, a helium-purification-system flow rate of 200 lb/hr and a fission-product trapping-system flow rate of 200 lb/hr are adequate to maintain helium purity below 10 ppm.

A piping and instrument diagram of the purification system will be prepared in January, 1960.

At the present time, a preliminary study is being carried out to help determine the value of an out-of-pile loop to the research and development program. A decision on whether or not to construct the loop will be made at the conclusion of the study.

The following drawings have been completed as part of the preliminary design of the out-of-pile loop: (1) process flow sheet (Fig. 6. 2); (2) piping and instrument diagram (Fig. 6. 3); (3) preliminary equipment layout; and (4) heater test-section design.

A summary report is in preparation which discusses the advantages of constructing a loop such as that shown in these drawings, and also the information to be obtained from experiments performed with this loop.

In brief, the out-of-pile loop would be used primarily for obtaining engineering data and determining operating procedures pertinent to the helium coolant system in the HTGR. The main objectives of the tests to be conducted with this facility would be as follows:

1. Determination of the operational procedures for minimizing the attack on graphite following a steam leak.

		STREAM No.											
COMPOSITION		1	2	3	4	5	6	7	8	9	10	11	12
H ₂ C	PPM	83.7	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4
	LB/MOL	25.3	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7
CO	PPM	25.3	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7	4.7
	LB/MOL	4.7	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845
H ₂	PPM	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4	15.4
	LB/MOL	4.7	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845
CH ₄	PPM	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2
	LB/MOL	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845
CCl ₄	PPM	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2	5.2
	LB/MOL	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845	0.845
PRESS - PSIA		310	309	308	307	306	305	304	303	302	301	300	299
TEMP - °F		100	100	680	800	800	200	80	100	90	-260	-310	40
TOTAL #/HR		200	200	200	200	200	200	200	200	200	200	200	200

Fig. 6.1 - Preliminary process flow sheet for helium purification system

STREAM No	1			2			3			4			5			6			7			8			9		
	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX	MIN	NORMAL	MAX
LB/HR	500	1000	1500	250	500	1125	125	500	750	500	1000	1500	0	633	633				500	1000	1500	500	1000	1500	500	1000	1500
ACFM																											
SCFM	791	1582	2372	395	791	1780	198	791	1186	791	1582	2372	0	1000	1000	867	1000	1500	791	1582	2372	791	1582	2372	791	1582	2372
PSIA	305	305	305	305	305	305	305	305	305	305	304	303	305	304	303	304	3035	302	304	3035	302	304	303	301	302	301	296
TEMP. °F	600	600	600	600	600	600	1000	1000	1000	800	1000	1000	800	1000	1000	1400	1400		1400	1400	1400	1200	1200	1200	600	600	600

Fig. 6.2--Preliminary process flow sheet for out-of-pile loop

Fig. 6.3--Preliminary piping and instrumentation flow sheet for out-of-pile loop

2. Determination of the amount of attack on the hot graphite if it is cooled by an air-helium mixture.
3. Determination of the effectiveness of various helium-purification systems.
4. Testing of the instrumentation developed for analyzing helium purity and for impurity-concentrating techniques.

6.2. FISSION-PRODUCT TRAPPING SYSTEM

As currently envisioned, the fission-product trapping system will be made up of three sets of charcoal beds, operating at different temperature levels, as follows: (1) a water-cooled trap operating at about 100°F ; (2) a refrigerated trap operating at about -40°F ; and (3) a low-temperature trap operating at about -300°F . These traps will be installed external to the reactor vessel on the fuel-element purge line. In addition, traps operating at about 700°F will be inserted in the bottom reflector extension of each fuel element; the Chemistry Section will test suitable trapping agents for this application.

A preliminary design of the external fission-product trapping system has been made. This design was based on the assumption that all fission products which are volatile at core temperatures will escape from the fuel and will be carried to the external traps within 10 sec of being produced, thus giving rise to about 3 Mw of decay heat generation in the external traps. This assumption is very pessimistic and represents the worst possible case. Studies are also being made of the design requirements for fission-product traps to be used with fuel elements which hold up fission products for one day after their production. Preliminary calculations indicate that the heat release could be reduced by a factor of ten under this condition. The design of fuel elements having holdups of this magnitude is under study.

The 10-sec holdup system requires a total of about 7500 lb of charcoal, the charcoal being enclosed in 10-in. -diameter stainless steel

pipes with internal copper fins to improve heat removal. The 100°F traps are submerged in a water pond for cooling and shielding, the -40°F traps are cooled by a CO_2 refrigeration system, and the -300°F traps are submerged in liquid nitrogen. The preliminary process flow diagram for the fission-product trapping system (Fig. 6. 4) has been completed, and the piping and instrument diagram is being prepared.

A trapping system is being designed for the in-pile-loop test. This system will provide data for the detailed design of the fission-product trapping system.

6. 3. SHUTDOWN COOLING SYSTEM

Studies were made of the advantages and disadvantages of decay heat removal by (1) an auxiliary heat exchanger and compressor of 1-1/2% of design capacity at atmospheric pressure and (2) one of two half-size steam generators. The principal conclusions reached are:

1. The heat-transfer surface area of the auxiliary exchanger would have to be quite large because of the need for operation at sub-atmospheric pressure and reduced temperature when changing fuel elements. The volumetric suction capacity of the compressor at the reduced pressure also would be large.
2. Preliminary calculations indicate that the cooling capabilities of a half-size boiler would be required immediately following a scram to prevent the temperature of the hot helium leaving the reactor from exceeding 1500°F . Further reduction in cooling capability would produce greater temperature rises. For example, using the auxiliary heat exchanger, these temperatures could rise to 2000°F . The half-size boiler would thus afford much greater protection against overheating and failure of tubes, tube sheets, pipes, metal cladding, and control-rod joints (if made of metal).

POINT NO.	1	2	3	4	5	6	7	8	9	10	11
Kr PPM	—	.08	.08	.08	.08	.08	.04	.04	<.01	<.01	<.01
Xe PPM	—	.27	.27	.27	.27	.27	.20	.20	<.01	<.01	<.01
TOTAL F.P. PPM	.73	.35	.35	.35	.35	.35	.24	.24	<.01	<.01	<.01
H ₂ PPM	10.00	10.00	10.00	<.01	<.01	<.01	<.01	<.01	<.01	<.01	<.01
H ₂ O PPM	1.00	1.00	1.00	11.00	11.00	11.00	1.70	<.01	<.01	<.01	<.01
CO PPM	10.00	10.00	10.00	<.01	<.01	<.01	<.01	<.01	<.01	<.01	<.01
CO ₂ PPM	1.00	1.00	1.00	11.00	11.00	11.00	11.00	<.01	<.01	<.01	<.01
He Lb/HR	200	200	200	200	200	200	200	200	200	200	200
ACFM	31.8	17.0	41.2	42.8	18.6	12.2	12.2	4.9	4.3	11.7	18.2
PSIA	289	286.5	286	285	284.5	284	283.5	283	282	281.5	281
TEMP °F	600	100	900	950	150	-60	-60	300	-320	-80	130

Fig. 6.4--Preliminary flow sheet for fission-product trapping system

An analysis was made of the effect of reducing the pressure level of steam generation in the event of an emergency scram necessitated by a tube leak. Calculations indicate that the heat-removal capacity of a boiler can be increased by 33% by lowering the steam pressure and feedwater temperature from their normal values of 1500 psia and 426°F to about 450 psia and 100°F.

6.4. CHEMISTRY EXPERIMENTS SUPPORTING PLANT AUXILIARY SYSTEMS

Permeation of Gases through Silica Glass

The possible use of silica glass to purify the HTGR helium purge stream has been suggested by the commercial application of silica glass as a means of recovering helium from natural gas. A literature survey on the permeation of gases through silica glass indicates that the use of this material should be a very effective means of removing fission products from the helium stream. Such a system would remove the noble gases (krypton and xenon) as well as those which can be removed by chemically reactive traps. Also, the effectiveness of separation appears to improve with a decrease in temperature. The ratio of helium permeability to argon permeability at 800°C is 10^4 and when extrapolated to 400°C is 10^8 . No data are available for permeability constants for gases heavier than argon; however, the permeability appears to decrease rapidly with the molecular size (or molecular weight) of the gas. It is therefore estimated that the ratio of helium permeation to that of krypton at 400°C would be at least 10^{10} . Small-diameter quartz tubes are commercially available, but no design calculations of such a system have been carried out. Two of the major design problems are (1) finding a way of making a radiation-resistant, high-temperature seal to the quartz tubes and (2) removal of the fission-product gases to prevent overheating.

Experimental work has been carried out which verifies the literature data on the rate of permeation of helium through quartz. The

experimental data obtained and also pertinent data from the literature are given in Fig. 6.5.

Fission-product Adsorption Isotherms of Fission-product Trap Materials

Adsorbing materials with high surface areas, such as activated charcoals, are promising reagents for trapping fission-product gases or vapors from a helium gas stream. The adsorption of krypton, xenon, and iodine vapor on activated charcoal and the use of charcoal beds to hold up these fission products under flow conditions has been rather extensively studied by Browning, *et al.* ^(1,2) Comprehensive quantitative data on the adsorption of iodine by activated charcoal at low partial pressures, however, do not appear to be available. Furthermore, the HTGR fuel-element purge and fission-product trapping system requires the reduction of fission-product gas or vapor partial pressures from the order of 10^{-6} atm to the order of 10^{-12} atm. Accordingly, experiments were undertaken to obtain adsorption isotherms or isobars at very low specific adsorptions (grams of adsorbate per gram of adsorbent).

The method involved sweeping I^{131} -tagged iodine vapor of known specific activity through a small charcoal trap and monitoring the build-up of I^{131} in the trap with a calibrated scintillation detector. The iodine vapor is generated by slowly passing helium over tagged iodine crystals held at a constant temperature. An iodine pressure approximately equal to its equilibrium vapor pressure is attained; thus, by choosing the temperature of the I_2 generation, the iodine partial pressure may be selected. In checking out the apparatus, which is shown schematically in Fig. 6.6, iodine vapor pressures were determined and found to agree fairly well with literature values. Thus, for iodine held at 0°C the experimental value determined for the vapor pressure was 3.5×10^{-5} atm, whereas a value obtained from the extrapolation of data from the literature was 3.9×10^{-5} atm.

In a series of experiments, isobars have been obtained for iodine

Fig. 6.5--Rate of permeation of helium through quartz. Points are $\circ$, R. M. Barrer, Diffusion In and Through Solids, The University Press, Cambridge, 1941; $\square$, Francis Norton, J. Am. Ceram. Soc., Vol. 36, 1953, p. 90; Δ , experimental

Fig. 6.6-- I_2 adsorption apparatus

pressures ranging from 3×10^{-3} to 3×10^{-6} atm using Fisher 6- to 14-mesh activated charcoal as the adsorbate. The charcoal was determined by BET measurements to have a surface area of approximately $1 \times 10^3 \text{ m}^2/\text{g}$. Since the method for measuring the amount of iodine adsorbed was very sensitive, and since a point source is convenient for calibration purposes, only a small amount of charcoal was used in the experiment (25 to 100 mg).

To determine a point on the adsorption isobar, helium containing the tagged iodine at a fixed partial pressure is passed at a constant rate over the charcoal, which is held at a fixed temperature. The activity of the charcoal, which is monitored by a scintillation counter, gamma spectrometer ratemeter, scaler, and recorder system, is noted until saturation is indicated on the recorder tracer. Then a series of counts is taken to accurately determine the activity level, which can then be related to the amount of iodine adsorbed by means of the known specific activity of the tagged iodine. The time required to reach saturation essentially determines the time required to obtain one point on an isobar. It was found that for vapor pressures of less than 10^{-5} atm, this time became rather long (of the order of days) because of the low rate of transport at low partial pressures.

It is to be noted that even at pressures of 3×10^{-6} atm, substantial adsorptions of the order of 10 mg of iodine per gram of charcoal were obtained at trap temperatures of the order of 300°C . The saturation value of the charcoal used ranges from 1 to 2 g of iodine per gram of charcoal, depending on the temperature of the trap. A plot of three isobars for pressures ranging from 3.1×10^{-6} to 4.1×10^{-4} atm obtained with Fisher charcoal is shown in Fig. 6.7. Work is continuing on obtaining isobars at somewhat higher trap temperatures and somewhat higher iodine partial pressures. When the work on Fisher charcoal is completed, a few runs will be made with Columbia Grade-G activated charcoal, which has a somewhat higher BET surface area according to our measurements ($1.5 \times 10^{-3} \text{ m}^2/\text{g}$) and which has been tentatively specified in the design

Fig. 6.7--Adsorption isobars for iodine, using Fisher activated charcoal

of the external fission-product traps.

An apparatus has been constructed for studying the adsorption of the fission-product element cesium on activated charcoal and other adsorbents (such as activated alumina) at low partial pressures. In this study a static rather than a flow system will be used; however, a tracer technique will be used in a manner similar to that of the iodine experiment to determine the amount of cesium adsorbed. A technique has been developed for preparing pure cesium metal tagged with Cs^{133} . In this procedure care must be taken to avoid contamination of the cesium metal with cesium oxide.

Internal Fission-product Trapping Systems and Reagents

In the design of the HTGR fuel elements, much consideration has been given to the alternative use of chemical or adsorption fission-product traps within the fuel element. With either of these methods, the more volatile fission products would be retained within the fuel element, in general at the cold end, or at least delayed in their passage out of the fuel element (GAMD-304, GAMD-902).

Recent consideration has favored a vented-can, graphite-clad fuel element in which the helium purge gas is first passed between the outer sleeve and the can and then through the can. Thus, fission products effusing from the fuel compacts are swept out through the bottom of the fuel element through a piping system to external fission-product traps. With this system it would seem particularly desirable to incorporate an internal fission-product trap near the bottom of the fuel element. A chemical trap seems desirable, so that chemically reactive fission products such as iodine and bromine will have an opportunity to react in the fission-product trap instead of attacking the piping system and causing corrosion and over-heating of the piping due to the decay heat of deposited short-lived fission products.

A study has begun of internal trapping reagents which will be effective

by means of both physical adsorption and chemical reaction. Activated charcoal appears to be a promising adsorber and also can serve as a matrix for the incorporation of a chemical reagent. Copper appears to be a suitable chemical reagent for reacting with the corrosive fission products iodine and bromine. Accordingly, activated charcoal-copper reagents have been prepared and are in the process of being tested. They were made by impregnating 6- to 14-mesh activated charcoal with an alcoholic copper nitrate solution. The resulting material was dried at 100°C , baked at 300°C to decompose the copper nitrate to oxide, and finally treated at 500°C with hydrogen to reduce the copper oxide to metallic copper. Samples prepared in this way assay from 20 to 30 wt-% copper. BET measurement of the surface area showed no apparent decrease in the surface area of the charcoal ($\sim 1 \times 10^3 \text{ m}^2/\text{g}$), while microscopic examination indicated the copper to be quite well dispersed through the material. The activated charcoal used was supplied by Fisher. Copper-impregnated reagents are also being made from several grades of Columbia G activated charcoal recently received and also from two grades of higher-surface-area activated alumina from Aluminum Company of America. It is believed that activated alumina may also serve as an effective reagent, as well as providing support for the copper.

A series of experiments have been designed for testing these and any other promising internal-trap materials. In these tests reagents will be exposed to a helium gas stream carrying tagged fission-product elements such as iodine, cesium, tellurium, and strontium at the concentration levels (of the order of 10^{-6} atm) to which the reagent will be exposed in an actual fuel element. Testing will be for effectiveness and capacity under the temperature conditions which might prevail in the fuel element. Small-scale laboratory test experiments using tagged iodine in a helium stream have been started. Parts and instruments for a larger-scale test apparatus have been ordered.

An internal fission-product trap for the in-pile test of the HTGR

graphite fuel element has been designed. The trap comprises a cartridge constructed of carbon materials and is made to maximize heat conduction from the fission-product-trapping reagent. The reagent is contained in six concentrically placed holes in the graphite body, which has a porous carbon disk at each end.

Assistance is also being given on the design of the external fission-product traps for the in-pile loop experiment. Here the concern is with designing a facility where candidate materials for use in traps may be tested using helium sweep gas which comes directly from the in-pile fuel element and which has been allowed to bypass the internal trap within the fuel element.

CO Disproportionation Reaction

It appears that even at quite low levels of oxygen-containing gaseous impurities in the helium coolant, an appreciable amount of carbon transport from the HTGR graphite-fuel-element core to heat-transfer surfaces may occur, principally through the reaction $2\text{CO} = \text{C} + \text{CO}_2$. The rate of transport has been estimated (GAMD-690, GAMD-1139), and it is concluded that oxygen impurities at the level of 10 ppm may lead to appreciable carbon transport. It is believed possible that at this or higher oxygen-impurity levels, carbon transport would be serious in that it would lead to a substantial erosion of graphite fuel elements and to the fouling of heat-transfer surfaces by carbon deposition. Also, there is a possibility of carburization of the metal heat-transfer surfaces, which would have a deleterious effect on structural properties. It is believed that the forward reaction involving the disproportionation of CO, which is catalyzed by metals and which will probably occur on the surface of the boiler superheater, will be the controlling rate process rather than the reverse reaction involving the attack of the hot graphite surfaces of fuel elements by CO_2 . Depending on the temperature, the disproportionation reaction at the metal surface will be controlled either by the rate of adsorption and

chemical reaction at the surface or by diffusion of the reactant to the surface.

Calculations to date have been made on the assumption that the reaction is diffusion-controlled. If the rate of the surface reaction of CO with the metal should prove to be the controlling factor, carbon transport rates will be lower than those estimated. Accordingly, the study has been undertaken of the reaction of CO at low partial pressures (in helium) with the metal surfaces in the temperature range from 350° to 750°C. The reaction takes place in a quartz furnace tube which contains the metal in the form of a bundle of fine wire of known surface area. Metals being studied are pure iron and various stainless steels or other alloys which may be used as heat-exchanger materials. The range of partial pressure of the CO reactant being studied is 10^{-2} to 10^{-4} atm. In studying the rate or extent of surface reaction, a major problem is the accurate measurement of small quantities of carbon deposited on the metal surfaces and of the small quantities of CO₂ formed. In order to obtain the required sensitivity, C¹⁴-tagged CO is being used in the experiment. The reaction products are assayed in the gaseous form by means of an ionization counter and vibrating electrometer designed for the determination of C¹⁴. In analyzing for the carbon deposited on the metal, it is planned to react the metal with oxygen following an experimental run; by this means carbon will be converted to CO or CO₂ and thus will be in the form in which it can be assayed in the ionization chamber.

The apparatus for this study has been assembled and tested. Some time has been spent on devising a trap for quantitatively accumulating CO₂ formed in the reaction. A silica gel adsorbent cooled to liquid-nitrogen temperatures has been found to be effective. This trap collects all of the CO₂ and some of the CO in the helium stream that leaves the furnace tube. A scheme has been devised for quantitatively separating the CO and CO₂ collected on the silica gel trap and for transferring the CO₂ to the ionization chamber for assay. The apparatus for this purpose makes use of the

principle of the gas chromatograph and is shown in Fig. 6.8. A diagram of the apparatus for carrying out the disproportionation reaction is shown in Fig. 6.9. It has been found that it is not a simple matter to prepare and maintain the He-CO gas mixtures at very low levels of H_2O , CO_2 , and O_2 impurities (of the order of 1 ppm of each). Accordingly, some time has been spent in working out purification and analysis techniques. The Nuclear-Chicago Dynacon electrometer for C^{14} assays has been found to work quite satisfactorily and with ample sensitivity. A method for preparing C^{14}O directly from solid $\text{BaC}^{14}\text{O}_3$ has been worked out. The BaCO_3 is fused with PbCl_2 to evolve CO_2 , which is then reduced with hot zinc. One hundred per cent yields of CO were obtained with no CO_2 or O_2 contamination. However, it was found that care had to be taken to remove water by suitable traps in order to avoid contamination of the CO_2 with hydrogen produced by reaction with the zinc.

It is expected that considerable experimental data on the disproportionation reaction will be forthcoming in the next few months. The reaction will be studied both with and without hydrogen being present, and it is planned to study this reaction in a gamma radiation field generated by the General Atomic electron linear accelerator.

It is also planned to study the kinetics of the disproportionation reaction in the diffusion-controlled region, using an apparatus with a fixed bed packed with metal spheres.

Steam-Graphite Reaction

In the event of a small or large steam leak into the HTGR coolant, a reaction with the hot graphite of the reactor core may be expected, whereby CO, CO_2 , and H_2 will be formed. It is likely that some coating or impregnation treatment will be found which will protect the graphite. It is even possible that some substance may be introduced in the gas phase which will inhibit or compete with the steam-graphite reaction. However, until foolproof protective measures are worked out, the occurrence of the H_2O -graphite reaction must be anticipated.

Fig. 6.8--Apparatus for CO-CO₂ separation

Fig. 6.9--CO-CO₂ disproportionation apparatus

Even though the reaction may not be extensive, because of rapid reactor shutdown and cooling, considerable H_2O , CO , CO_2 , and H_2 will be introduced and will be adsorbed by the reactor graphite. Furthermore, the graphite which reacts directly with the steam will probably collect of the order of a monolayer of strongly adsorbed (chemisorbed) oxygen. The question then is how difficult will it be, or how long will it take, to degas the reactor graphites and reduce the oxygen impurity content of the coolant to an acceptable level. Accordingly, some small-scale experiments are being undertaken whereby samples of candidate reactor graphites can be reacted with steam-helium mixtures and the rate and extent of the reaction and, subsequently, the degassing behavior of the graphite can be determined.

More particularly, the apparatus shown in Fig. 6.10 is being set up. With this apparatus, a specimen of HTGR graphite can be degassed, the rate of reaction with steam can be measured, and the specimen can again be degassed. The specimen will be heated in a quartz tube in a muffle furnace, and the tube will be evacuated. A helium stream containing water vapor will then be passed over the specimen. The concentration of water will be controlled by bubbling the helium through water at a fixed temperature. The unreacted water will be removed from the helium stream with drying columns, and the helium stream will be analyzed for the reaction products CO , CO_2 , and H_2 with a mass spectrometer. The amount of unreacted water accumulated in the drying columns will be measured by weighing these columns. After the reaction-rate run is completed, the flow of "wet" helium will be stopped, and the specimen will be cooled in the presence of the reaction products. The gases will then be pumped out through the degasification portion of the system, where the amount of each reaction product will be measured. Two types of degasification measurements will be made: the rate of degassing of each gas at constant temperature (600°F and/or 1800°F) and the total amount of gas given off at each of sequentially higher temperatures. During the degassing measurements, the pressure over the specimen will be kept low by pumping the gas out with a Toepler pump.

Fig. 6.10--Apparatus for measuring the rate of the steam-graphite reaction

Additional measurements which may be made include:

1. The weight of the graphite, for a crude estimate of the amount of reaction.
2. The dimensions of the specimen, to indicate the gross localized attack.
3. The surface area (BET). The rate of reaction should be proportional to the surface area contacted by steam.
4. The permeability of the graphite, to indicate whether the steam-graphite reaction has destroyed a protective layer or has enlarged the pores.
5. The porosity of the graphite (by means of a mercury porosimeter) to determine if the pores have been enlarged.

The parts for this apparatus have been accumulated and are being assembled.

Activation Analysis for Oxygen in Graphite

According to both theory and experimental evidence, the activation energy and rate of desorption are related to the amount (surface coverage) of the species adsorbed. However, the final residual quantity of oxygen adsorbed on graphite is difficult to remove and assay. An accurate, non-destructive analysis for the amount of oxygen adsorbed on graphite would therefore be helpful in studying the desorption process. Accordingly, an activation-analysis technique for determining the amount of oxygen in graphite has been investigated.

In this procedure, 7.4-sec N^{16} was produced from O^{16} by means of a pulse of fast neutrons (>67 kev) from the TRIGA reactor. The pulse ($nvt \sim 6 \times 10^{13}$) lasted approximately 10 ms and resulted from a 20 Mw-sec "flash" of the reactor. The specimen was irradiated near the edge of the core. A pulsed neutron source was used because it favors the production of short-lived activities, thereby minimizing interference by longer-lived activities. A 3 by 3 in. sodium iodide scintillation-counter crystal was

used as a detector in conjunction with a single-channel gamma-ray spectrometer circuit. After making certain that N^{16} activity was present, the discriminator was set to count pulses corresponding to gamma rays of energy greater than 4 Mev, and the total number of counts was taken for a period of 60 sec after insertion of the sample into the detector. The detection system was approximately 40 ft from the reactor, in order to minimize background radiation from the reactor. The time interval from the flash to the start of counting averaged about 8 sec.

The detector response corresponded to about 3×10^3 counts per milligram of oxygen (an increase of a factor of 10 to 20 might be expected for samples irradiated at the center of the core of TRIGA). The response was constant for samples containing >20 mg of oxygen, but the ratio of total counts to milligrams of oxygen increased for a total oxygen content of <20 mg and rose sharply for a total oxygen content of <10 mg.

It was concluded that graphite samples containing more than 10 mg of oxygen (0.1% oxygen in the case of the customary 10-g sample) can be accurately analyzed (to 10% or better).

Interaction of Fission Products with Metal Surfaces

In connection with the evaluation of the contamination of metal surfaces by fission products in the primary coolant loop and the effect of fission products on the metal piping of the fuel-element purge system, there is a need for the study of the interactions of certain volatile fission-product elements with metal surfaces. In particular, in regard to the contamination of surfaces, there is an uncertainty as to the extent to which cesium will interact with steel surfaces when it is in helium at low partial pressures. Accordingly, the apparatus shown in Fig. 6.11 has been set up for the study of the adsorption of cesium on metals.

The apparatus is designed so that metallic cesium tagged with Cs^{137} can be produced directly in the apparatus under vacuum conditions and helium can be subsequently passed over the cesium, which is held at a

Fig. 6.11--Cesium adsorption apparatus

constant temperature. The cesium may thus be introduced into the helium sweep gas at a known partial pressure (10^{-3} to 10^{-9} atm). The helium gas carrying the cesium is then swept over a metal test sample, which may be in the form of a bundle of fine wire of known surface area. The sample is held in a section of 1-in.-OD stainless steel tubing, which is maintained at the temperature chosen for a given run. The helium sweep gas exiting from the apparatus is passed through an alcohol trap to dispose of the cesium. After completion of the run, the reaction tube is cooled and an analysis is made to determine the quantity of cesium collected on the metal wire and the furnace tube.

Preliminary studies have been made on the deposition of cesium on the steel wire samples, the furnace tube, and the connecting tubing in preparation for quantitative studies. It is planned to monitor the test sample with a scintillation counter, gamma-ray spectrometer, and ratemeter during the course of a run to obtain data on the rate and total amount of adsorption as a function of time.

As a need exists for information on the interactions of fission-product elements with the piping of the purge system, samples of stainless steel and other pertinent alloys will be reacted with fission-product elements produced in connection with the experiments on internal fission-product traps discussed above. In particular, there is interest in determining the nature and extent of the action of fission-product elements such as iodine and tellurium.

Effect of Conditions of Formation and Nature of Fission-product Aerosols

A preliminary experimental study has been made of strontium vaporization, condensation, and aerosol formation to assist in answering some questions raised in the safety analysis. Strontium vapor at a partial pressure of the order of 10^{-2} atm in helium at 1000°C was allowed to mix with cool helium or air and to pass through a stainless steel tube with a rather steep temperature gradient and then through a glass-wool plug

followed by a series of micropore filters. Under these conditions, there was practically no aerosol formation (with particles $5\ \mu$ in diameter or smaller). When no air was admitted, most of the strontium condensed on the walls of the tubing. When air was admitted, a substantial fraction of the strontium was collected as SrO particles on the glass-wool filter.

It is planned to look for fission-product aerosols (using millipore filters as one means of segregating activities by particle size) in the course of studying the characteristics of the helium purge stream of the in-pile loop experiment. The arrangement described above will be used.

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2. Browning, W. E., and C. C. Bolta, Measurements and Analysis of the Holdup of Gas Mixtures by Charcoal Adsorption Traps, Oak Ridge National Laboratory Report ORNL-2116, August 10, 1958.

TASK VII. DEVELOPMENT OF CONTROL SYSTEMS AND INSTRUMENTATION

An electronic simulation of the HTGR system has been undertaken to facilitate studies of transient performance of the system, to evaluate various modes of control, and to optimize control settings. Methods of simulating the dynamic behavior of the reactor core, coolant, steam generator, and controls have been studied, and an analog computer program of the simulated system has been prepared. At this time, the electronic model of the system is only preliminary, and it will remain so until all components of the system are finalized, but useful information on orders of magnitude of transient performance is now obtainable, and accuracy will improve as the system evolves to its final state.

The neutron kinetics and core thermal capacity are fairly well known, and these portions of the simulator have been used to study the response of fuel-compact temperature and reactor power to step and ramp increases in reactivity, with various scram conditions, to help establish safe limits for allowable reactivity changes. For the most severe disturbance (a ramp increase from source level with end-of-life fuel conditions and a 0.01 δk per second scram starting 0.2 sec after reaching 150% of rated power), a 0.0001 δk /sec ramp caused a 230°C rise in fuel compact temperature and a 0.001 δk /sec ramp caused a 690°C rise. In this work a conservative negative temperature coefficient of 1×10^{-5} $\delta k/k$ per °C was assumed.

Reactor power variations required to effect the specified rates of change in load have been studied to ensure that the specifications can be met. Preliminary results indicate the specified 3%/min load change can be obtained by cutting back reactor power approximately 20 Mw below the load (during a decrease in load), the balance of the power being obtained from stored thermal energy (primarily in the reactor core graphite).

This reactor power cut-back (or increase, in the case of increasing load) will be accomplished with feedback controls rather than by scheduling alone, although a scheduled signal may be introduced in parallel with an error signal to improve dynamic performance.

A bibliography on steam-generator dynamic analysis and control has been prepared during the quarter.

Preliminary studies of the size, number, type, and location of the start-up neutron sources have been initiated. Type and location will be fixed shortly. Further information is forthcoming on reactor flux distribution, which will aid the progress of this study during the next quarter.

A literature survey on the burst-fuel-element detection problem was initiated. A new system concept has been evolved for use with a vented graphite fuel element. Further investigations will continue in the next quarter, and a design basis will be fixed when the fuel-element design is firm.

Preliminary investigations were made of the status of the fuel-element temperature measurement problem. Several thermocouple combinations show some promise, but further study is required. Other types of detectors will be reviewed during the next quarter.

TASK VIII. OVER-ALL PLANT ANALYSIS

General studies of reactor plant behavior have been initiated during this quarter. Preliminary work has been initiated to survey the problems of part-load and transient behavior of the reactor plant, control-rod programming, power output from the metal- and graphite-clad cores, thermal shock of components of the primary loop associated with changes in reactor output, and general criteria for the plant control system. More detailed work on all these subjects will be required, and it is anticipated that a larger effort will be devoted to them during 1960. Progress on these topics will therefore be reported in subsequent documents when additional information has been developed.

During the last quarter of 1959, an intensive analysis of the problems of reactor safety for the conceptual design of the HTGR was made. This analysis included an examination of the following features of the reactor: radioactive effluents under normal operating conditions, inherent safety characteristics of the HTGR, thermal design and control, transient behavior of the reactor system, problems of control and instrumentation of the primary coolant loop, normal operation and malfunction of the steam generator, problems of afterheat removal, loading and storage of fuel elements, handling of spent fuel, generation of A^{41} external to the primary-loop equipment, activation of the steam generator and other components of the primary loop, generation of solid and liquid wastes in the plant, characteristics and safeguards problems of the control rods and control-rod drive mechanisms, overpressure-relief systems, and various auxiliary systems connected with plant safety. In addition, a preliminary survey was made of the type of accidents to which the plant might be subject, and a preliminary definition was made of the maximum credible accident. On the basis of this preliminary analysis, the maximum credible accident appears to be the sudden loss of primary helium coolant due to a failure of the primary loop.

The results of this safety analysis have been made available to the design engineers, and appropriate action is being taken where possible to mitigate the potential hazards of the system by changes in design. It appears at the present time that the principal problems associated with the plant are the provision of an adequate emergency heat-removal system and the prevention of entry of air into the primary system in the event of a major rupture of the primary system.

Appendix

GENERAL ATOMIC INFORMAL REPORTS ISSUED ON THE HTGR
PROJECT FROM JANUARY 1, 1959, THROUGH DECEMBER 31, 1959

GAMD-63 (Rev.) Addendum No. 3	BUNNY. A TWO-GROUP, TWO-REGION REACTOR CODE FOR SPHERICAL SYSTEMS. ADDENDUM. "FURTHER MODIFICATION OF THE GENERAL ATOMIC BUNNY CODE" BALANCED BUNNY. E. K. Booth.
GAMD-497	A HIGH-TEMPERATURE THERMAL CYCLING DEVICE. A. Nordby and L. D. Palmer.
GAMD-603	GROUP NEUTRON CROSS SECTIONS. J. D. Garrison.
GAMD-623 (Rev.)	REVISION OF GENERAL ATOMIC IRRADIATION PROPOSAL TO MTR. W. W. Godsin.
GAMD-650	PRELIMINARY ESTIMATES OF EFFECTS OF CONJECTURED ACCIDENTS TO HTGR. T. H. Pigford.
GAMD-657	EROSION OF GRAPHITE BY HELIUM RECOILS FROM NEUTRON SCATTERING AND BY FORMATION OF HeC. E. Creutz and S. P. Cunningham.
GAMD-659	TARGET DATES FOR THE POWER REACTOR CRITICAL EXPERIMENTS. J. B. Dee.
GAMD-682	BASIC HEAT TRANSFER FORMULAS AND CORRELATIONS USED FOR THE HTGR HEAT TRANSFER CALCULATIONS. M. Troost.
GAMD-689	ANALYSIS OF TEMPERATURE/TIME HISTORY OF HTGR FUEL ELEMENTS AND GRID PLATE FOLLOWING SHUTDOWN WITHOUT COOLING. A. W. Harris.
GAMD-690	TENTATIVE PROGRAM FOR THERMAL CYCLING METAL CLAD HTGR TEST ELEMENTS IN PROCESS METALLURGY. J. R. Lindgren.
GAMD-693	VERY PRELIMINARY ESTIMATE OF THE HTGR TEMPERATURE COEFFICIENT (40 EMW). N. F. Wikner and D. H. Perkel.

- GAMD-702 HTGR FISSION-PRODUCT TRAPPING SYSTEM-
PRELIMINARY SYSTEM ANALYSIS AND SUMMARY
OF FACTORS. T. H. Pigford.
- GAMD-702 (Suppl.) HTGR FISSION-PRODUCT TRAPPING SYSTEM-
Sections III, IV PRELIMINARY SYSTEM ANALYSIS AND SUMMARY
OF FACTORS. T. H. Pigford.
- GAMD-709 HTGR FUEL ELEMENT, CONTROL ROD, AND
CHEMISTRY DEVELOPMENT PROGRAMS.
W. V. Goeddel.
- GAMD-713 STRAIN IN METAL-CLAD ELEMENT DURING
THERMAL CYCLING. R. F. Turner.
- GAMD-717 EFFECT OF ANNULAR FUEL ZONING ON TEMPER-
ATURES IN CYLINDRICAL COMPACTS.
P. Fortescue and M. Troost.
- GAMD-718 RESONANCE INTEGRALS FOR THE HIGH-TEMPER-
ATURE GRAPHITE REACTOR. L. W. Nordheim.
- GAMD-726 EFFECT OF GRAIN SIZE ON REACTIVITY IN HTGR.
L. W. Nordheim and J. B. Sampson.
- GAMD-731 SELECTOR COST ANALYSIS-CRITERIA AND
ASSUMPTIONS. E. H. Buell.
- GAMD-736 TRIPLE CONTAINMENT FOR HTGR. P. Fortescue.
- GAMD-736 TRIPLE CONTAINMENT FOR HTGR. P. Fortescue.
Addendum
- GAMD-738 A FORTRAN PROGRAM FOR CALCULATING RADIAL
TEMPERATURES IN THE HTGR FUEL ELEMENT.
M. Troost.
- GAMD-753 IMPROVED ESTIMATE OF THE THERMAL COM-
PONENTS OF THE HTGR TEMPERATURE COEF-
FICIENT. 40-EMW GRAPHITE-CAN VERSION.
N. F. Wikner and D. H. Perkel.
- GAMD-756 HTGR FUEL HANDLING SYSTEM. PRELIMINARY.
D. Nicoll.
- GAMD-766 HTGR IRRADIATION EXPERIMENT GA-306.
W. W. Godsin.

GAMD-775	ONE-GROUP SELECTOR CODE. E. K. Booth and H. D. Brown, Jr.
GAMD-775 (Rev.)	ONE-GROUP SELECTOR CODE. E. K. Booth and H. D. Brown, Jr.
GAMD-780	CONTROL ROD WORTH IN THE HTGR. L. W. Nordheim.
GAMD-785	TENTATIVE SPECIFICATIONS, HTGR FUEL ELEMENTS AND CONTROL RODS. D. H. Gurinsky and W. V. Goeddel.
GAMD-785 (Rev.)	TENTATIVE SPECIFICATIONS, HTGR FUEL ELEMENTS AND CONTROL RODS. D. H. Gurinsky and W. V. Goeddel.
GAMD-788	A THERMAL MOCK-UP OF AN IRRADIATION CAPSULE. L. D. Palmer.
GAMD-790	FISSION PRODUCT PRESSURE BUILDUP IN UNCLAD HTGR FUEL ELEMENTS. D. C. Morse.
GAMD-797	STRESS ANALYSIS OF INNER GRAPHITE REFLECTOR. D. Nicoll and R. F. Turner.
GAMD-798	THE FORMATION AND PROPERTIES OF PYROLYTIC CARBON. G. L. Allen.
GAMD-820	FUEL ELEMENT FAULT DETECTION SYSTEM. D. Nicoll and D. Rose.
GAMD-824	INITIAL ANALYSIS-HTGR HELIUM PURIFICATION SYSTEM DESIGN. R. J. Mulvihill.
GAMD-834	CREEP IN GRAPHITE. R. F. Turner.
GAMD-855	HOT SPOT CALCULATIONS FOR THE HTGR. M. Troost.
GAMD-860	STRESS IN INNER GRAPHITE CAN. R. F. Turner.
GAMD-863	HTGR NEUTRON KINETICS. H. A. Long.
GAMD-867	FISSION PRODUCT ESCAPE FROM DEFECTIVE METAL-CLAD FUEL ELEMENT. D. C. Morse.

- GAMD-884 ACTIVATION OF THE HEAT EXCHANGER OF THE HTGR DUE TO NEUTRON STREAMING. L. W. Nordheim.
- GAMD-891 FISSION PRODUCT ELEMENT YIELDS AND CHARACTERISTICS. L. R. Zumwalt.
- GAMD-901 MELTING POINT OF Th-U-C FUEL ELEMENTS. L. Brewer.
- GAMD-902 TRAP FOR ALKALI AND ALKALINE EARTH METAL FISSION PRODUCTS. L. Brewer.
- GAMD-903 FATE OF FISSION PRODUCT GASES IN THE COOLANT STREAM. L. Brewer.
- GAMD-904 HEAT TRANSFER FLUIDS FOR FUEL ELEMENT CANS. L. Brewer.
- GAMD-905 THE EFFECT OF A MAJOR STEAM LEAK IN THE HTGR POWER PLANT. H. N. Woebcke.
- GAMD-919 VAPORIZATION PROCESSES IN A RUNAWAY REACTOR. L. Brewer.
- GAMD-925 TWO WOLONTIS PROGRAMS USEFUL IN REACTOR KINETICS STUDIES. R. Engelmores.
- GAMD-926 A PRELIMINARY EVALUATION OF THE PROBLEM OF PROVIDING A METHOD OF MAKING GRAPHITE IMPERVIOUS FOR HTGR REQUIREMENTS. G. R. Tully and G. L. Allen.
- GAMD-943 NUCLEAR EXCURSIONS IN THE HTGR. J. Devooght, H. Fenech, and G. Hinman.
- GAMD-946 FLUOROCARBONS FOR REACTOR COOLANTS. A. W. Searcy.
- GAMD-947 HTGR FUEL HANDLING SYSTEM. GRAPPLER HEAD. PRELIMINARY TYPE 32-KS-322. D. Nicoll and J. A. Paget.
- GAMD-949 USE OF TIN IN HTGR FUEL ELEMENTS. I. Bornstein.
- GAMD-951 THERMAL STRESSES IN METAL FUEL CANS AND THE PROBLEM OF END CAPPING. D. W. Ver Planck.
- GAMD-962 EVALUATION OF MORGAN EYX60 GRAPHITE. W. V. Goeddel.
- GAMD-966 TRANSIENT THERMAL STRESSES IN THE PRESSURE VESSEL AND EXIT NOZZLE DURING HYPOTHETICAL TRANSIENTS IN THE HELIUM TEMPERATURE. G. B. Donaldson.

- GAMD-969 CONTROLS FOR FOSSIL FUEL FIRED STEAM POWER STATIONS. J. F. Dietz.
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- GAMD-984 SHUTDOWN HEAT GENERATION IN HTGR. D. C. Morse.
- GAMD-990 EVALUATIONS OF GRAPHITES FOR HTGR FUEL ELEMENTS. W. V. Goeddel.
- GAMD-995 HISTORY AND STATUS TO DATE OF THE CLADDING AND CYCLING OF HTGR METAL CLAD ELEMENTS. J. R. Lindgren.
- GAMD-1013 INCURRENT ON A BLACK BODY. J. Devooght.
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- GAMD-1032 PROGRAMS FOR FUEL AND CONTROL ROD MATERIALS DEVELOPMENT FOR 1960. R. B. Duffield.
- GAMD-1033 HELIUM PURITY REQUIREMENTS FOR HTGR. R. J. Mulvihill.
- GAMD-1035 ANALYSIS OF TEMPERATURE/TIME HISTORY OF HTGR CONTROL RODS FOLLOWING SHUTDOWN WITHOUT COOLING. F. E. Vanslager.
- GAMD-1048 HTGR CONTROL ROD WORTH. J. Devooght.
- GAMD-1050 EPITHERMAL ABSORPTION BY CONTROL RODS OF VARIABLE BLACKNESS. J. Devooght.
- GAMD-1064 TEMPERATURE/TIME HISTORY OF GRAPHITE CORE AFTER REACTOR SHUTDOWN. R. F. Turner and F. E. Vanslager.
- GAMD-1072 BLUP - A REACTOR KINETICS CODE. C. W. Bell.
- GAMD-1096 DECAY HEAT GENERATION IN BOTTOM REFLECTOR EXTENSION OF HTGR FUEL ELEMENTS. D. C. Morse.

- GAMD-1103 GENERAL ATOMIC CONTROL ROD IRRADIATION PROPOSAL. W. W. Godsin.
- GAMD-1110 HOT SPOT TEMPERATURE IN 115 MW REACTOR WITHOUT ELEMENT SPACING RODS. R. F. Hinz.
- GAMD-1115 DUPLICATION OF MASS TRANSFER COEFFICIENT PRESENT IN HTGR BOILER BY FLOW THROUGH A FIXED BED PACKED WITH SPHERES. R. J. Mulvihill.
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- GAMD-1139 HTGR CARBON TRANSPORT AS A FUNCTION OF PURIFICATION FLOW RATE AND STEAM LEAKAGE. R. J. Mulvihill.
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