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GENERAL ATOMIC

DIVISION OF **GENERAL DYNAMICS**

AEC RESEARCH AND
DEVELOPMENT REPORT

GA-6113

TARGET

A Program for a 1000-Mw(e) High-temperature
Gas-cooled Reactor

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
FEBRUARY 28, 1965

Prepared under
Contract AT(04-3)-167
Project Agreement No. 17
for the
San Francisco Operations Office
U. S. Atomic Energy Commission

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

March 31, 1965

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GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

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INTRODUCTION

This report covers the work performed by General Atomic Division of General Dynamics Corporation under U.S. Atomic Energy Commission Contract AT(04-3)-167, Project Agreement No. 17. This Project Agreement calls for preliminary design and related exploratory development of a 1000-Mw(e); gas-cooled, nuclear power reactor incorporating advanced technology, and a 150-300 Mw(e) prototype. This program, which has been designated TARGET (Thermal Advanced Reactor, Gas-cooled, Exploiting Thorium), is based on the concept of the High-temperature Gas-cooled Reactor (HTGR) system developed by General Atomic.

The related exploratory development effort is aimed at a 1000-Mw(e) gas-cooled reactor plant that could be placed in operation in the 1970's, with operation of the prototype at an earlier date. The plant would demonstrate such new design features as:

1. Breeding or near breeding of fuel in a high-temperature, gas-cooled, thermal reactor using a Th-U²³³ fuel cycle.
2. Advanced reactor fuel elements, which may include new and simpler configurations and new methods of processing and fabricating to extend fuel lifetime and reduce fuel costs.
3. Alternative plant arrangements to reduce plant capital costs, including such possibilities as alternative materials (e. g., concrete for pressure vessels) and the assembly of the reactor and steam-generating equipment in a single containment.
4. A supercritical-pressure steam cycle that is particularly adaptable to a 1000-Mw(e) plant and to the inherent high-temperature capabilities of the HTGR system.

I. OPTIMIZATION DEVELOPMENT

The optimization program development has progressed to a point at which the major parts of the program may be considered completed. Work is now under way to interconnect the parts. When this task is accomplished and a preliminary version of an "Optimization Operator" subroutine has been written, the program will be ready for the first full test problem.

STEAM GENERATOR

The steam generator design and cost programs to be used in the optimization code are ready for production use. The programs will design and establish the cost of a two-pass U-tube reheater and superheater, and a circular economizer-evaporator. The banks of superheater and reheater U-tubes are arranged in a circular pattern with alternate reheater and superheater tubes. The economizer-evaporator is composed of platens of spirally wound tubes. Parallel helium-steam flow is assumed in the reheater-superheater section, and counterflow is employed in the economizer-evaporator. The design parameters are as follows:

1. Reheat pressure drop and specified inlet and outlet steam conditions.
2. Superheater outlet conditions.
3. Economizer inlet conditions.
4. Total helium pressure drop.
5. Total economizer-evaporator-superheater steam pressure drop.
6. In addition, a maximum tube metal temperature constraint is imposed on the superheater reheater section.

A preliminary analysis with the first version of the code indicated a very pronounced sensitivity to the steam and helium film coefficients in the reheater section. A detailed parameter study was conducted with the performance code HEAT to determine the importance of each of the major variables and to establish a more efficient iteration technique. The following paragraphs summarize this study and discuss the effect of the more important variables on the design of the reheater.

The first version of the circular U-tube reheater design code used the following method to compute the dimensions of the reheater.

1. Vary the longitudinal pitch spacing to satisfy the tube temperature constraint.
2. Vary the number of tubes per pass for large variations in the heat-transfer surface area.
3. Adjust the tube length to produce the exact steam conditions.
4. Change the tube diameter to obtain the desired steam pressure drop.

The code was very unstable and often failed to converge. The problem was caused by selecting the wrong parameters to vary the heat-transfer surface and the steam pressure drop. A detailed parameter analysis was conducted with the performance code HEAT to determine the controlling variables. The results are as follows:

1. Tubes per Pass and Tube Length. Varying the number of tubes per pass produces a large change in the mass flow rate and steam heat-transfer coefficient. As a result, the number of tubes per pass are more effective in controlling the steam pressure drop than in varying the heat transfer area. The tube length should be used to vary the heat transfer area. The following example will demonstrate the importance of the variation in the film coefficient (see Table 1.1): Three heat exchangers with four, six, and eight tube rows per pass were designed for the same heat load and maximum metal temperature. The longitudinal pitch spacing was adjusted for each arrangement to maintain the tube temperature at its design point. The tube length was changed to produce the required heat transfer surface. The steam film coefficient was considerably higher in the four-tube arrangement than it was in the eight-tube exchanger. As a result the four-tube reheater required only 63% of the heat transfer area needed in the eight-tube configuration.
2. Tube Diameter. (See Table 1.2). Small variations in the tube diameter do not significantly affect the heat transferred if the longitudinal pitch spacing is adjusted to maintain the design metal temperature. However, the associated change in the helium flow area is large (see Table 1.2). For a constant pitch spacing there is a substantial change in the metal temperature for a small change in diameter. The tube diameter and the longitudinal pitch spacing are both effective parameters for controlling the metal temperature.

3. Performance. The effects of boiler scale and variations in flow rate are given in Tables 1.3 and 1.4 respectively.

The results of the above parameter study were used to select an effective set of independent variables and to establish a stable iteration technique. The current version of the design code employs the following method of analysis:

Independent Variables - Constraints

- a. Outer diameter of the reheater
 - b. Number of tubes per row
 - c. Steam and helium flow rates, lb/hr
 - d. Inlet and outlet steam enthalpies
 - e. Inlet and outlet helium temperatures
 - f. Steam pressure drop
 - g. Maximum metal temperature
1. Compute the upper limit of the tube diameter (maximum tube diameter) as a function of the outer diameter of the reheater and the number of tubes per row. The maximum diameter will produce a closely packed equilateral tube arrangement.

$$DO = \frac{ST}{2 \cdot \sin 60}$$

ST = Transverse pitch spacing (see Fig. 1.1)

DO = Tube diameter

The maximum tube diameter is used as the initial value for the tube diameter. It was found that the number of iterations could be reduced by relating the tube diameter to the number of tubes per row.

2. Determine the value of the longitudinal pitch spacing SL and the tube diameter needed to produce the desired metal temperature.
3. Find the tube length required to transfer the design heat load.
4. Check the steam pressure drop. If it is too high, add another row of tubes to each pass. If it is too low, remove a row. Repeat the above computation until the pressure drop is satisfied. (The tolerance is $\pm$ one row.)

Table 1.1
REHEATER PARAMETER VARIATIONS
(Number of Tubes)

Number of Tubes per Pass	Effective Tube Length (in.)	Change in Tube Length (in.)	Steam Outlet Enthalpy (Btu/lbm)	Steam Outlet Temp. (°F)	Max. Tube Temp. (°F)	Relative Helium Flow Area (%)	Relative Tube Length (%)	Relative Heat-transfer Area (%)
8	113.0	---	1546.8	1050.3	1150.8	100	100	100
6	123.735	+10.735	1546.9	1050.6	1150.5	63.75	109.50	82.3
4	142.38	+29.38	1547.0	1050.8	1150.2	33.62	126.0	63

NOTE: Steam flow rate = 706,600 lbm/hr Helium flow rate = 604,200 lbm/hr
 Steam inlet temperature = 618.1°F Helium inlet temperature = 1470.0°F
 Steam inlet enthalpy = 1308.8 Btu/lbm Steam operating pressure = 510-500 psia

Table 1.2
REHEATER PARAMETER VARIATIONS
(Tube Diameters)

Tube Outside Diameter (in.)	Change in Outside Diameter (in.)	Tube Inside Diameter (in.)	Changes in Inside Diameter (in.)	Tube Wall Thickness (in.)	Maximum Tube Temperature (°F)	Steam Outlet Enthalpy (Btu/lbm)	Change in Outlet Enthalpy (Btu/lbm)
0.8750	---	0.7450	---	0.130	1150.7	1546.8	---
0.9375	+1/16	0.8075	+1/16	0.130	1150.1	1542.25	-4.55
0.8125	+1/16	0.6825	-1/16	0.130	1150.1	1552.3	+5.5

NOTE: Effective tube length = 113 in. Operating press., steam = 510-500 psia
 Helium flow rate = 604,200 lbm/hr Average helium flow area D_o = 0.8750: 100%
 Helium inlet temp. = 1470°F D_o = 0.9375: 140.5%
 Steam flow rate = 706,600 lbm/hr D_o = 0.8125: 65.0%
 Steam inlet enthalpy = 1308.8 Btu/lbm

Table 1.3
REHEATER PARAMETER VARIATIONS
(Scale Resistance)

Scale Resistance (hr-ft ² -°F/Btu)	Steam Outlet Enthalpy (Btu/lbm)	Change in Outlet Enthalpy (Btu/lbm)	Steam Outlet Temp. (°F)	Change in Outlet Temp. (°F)	Maximum Tube Temp. (°F)	Change in Maximum Tube Temp. (°F)
0.0	1546.8	---	1050.3	---	1150.8	---
0.0005	1537.6	-9.2	1033.3	-17.0	1165.5	+14.7

NOTE: Effective tube length = 113 in. Steam operating press. = 510-500 psia
 Steam flow rate = 706,600 lbm/hr Helium flow rate = 604,200 lbm/hr
 Steam inlet enthalpy = 1308.8 Btu/lbm Helium inlet temp. = 1470.0°F
 Steam inlet temp. = 618.1°F

Table 1.4
REHEATER PARAMETER VARIATIONS
(Steam and Helium Flow Rates)

Helium Flow (lbm/hr)	Change in Helium Flow (%)	Steam Flow (lbm/hr)	Change in Steam Flow (%)	Outlet Enthalpy (Btu/lbm)	Change in Outlet Enthalpy (Btu/lbm)	Maximum Tube Temp. (°F)	Change in Maximum Tube Temp. (°F)
604,200	---	706,600	---	1546.8	---	1150.7	---
664,600	+10	606,600	---	1554.1	+7.3	1166.9	+16.2
543,800	-10	706,600	---	1538.5	-8.3	1135.2	-15.5
604,200	---	777,300	+10	1535.7	-11.1	1132.9	-17.8
604,200	---	635,900	-10	1559.0	+12.2	1170.2	+19.5
664,600	+10	777,300	+10	1543.0	-3.8	1149.4	+1.3
543,800	-10	635,900	-10	1550.8	+4.0	1155.1	+4.4

NOTE: Tube OD = 0.875 in. Helium inlet temp. = 1470.0°F
 Tube ID = 0.745 in. Steam inlet enthalpy = 1308.8 Btu/lbm
 Effective tube length = 113 in. Operating pressure = 510-500 psia
 Tube Spacing = Constant

Fig. 1.1--Transverse pitch spacing

5. Compute the performance of the superheater.
6. Design the economizer-evaporator section.

The headers for the various generating surfaces are designed in the subroutine MEDUSA. The inlet and outlet headers for superheater and reheater sections are coaxial. The header for the economizer-evaporator is similarly constructed. The purpose of this type of header design is to minimize the length or space required for the header. Since the cost of the headers is also likely to be a strong function of the total volume of metal, the header dimensions are to be minimized within any given restraints. Owing to the constraints of header diameter and short header length, the use of helical hole patterns is of no advantage; therefore only rectangular hole patterns are considered.

The subroutine MEDUSA is capable of designing to any of the following criteria.

1. A fixed length and fixed number of holes per row (rectangular hole pattern with rows running longitudinally),
2. A fixed length, but optimizing the hole pattern (to yield minimum material volume),
3. A fixed hole pattern, but optimizing the length, and
4. The optimum length and hole pattern.

The superheater-reheater section of the STMGEN code necessarily specifies the hole pattern to be used; therefore either case 1 or 3 above must be used. The evaporator-economizer header is not as restrictive, and any of the four cases may be used.

The current plan is to use case 3 for the superheater-reheater and case 4 for the evaporator-economizer. This can be altered if better design criteria are offered.

REACTOR CORE

Core Performance

The heat-transfer and flow-analysis subroutines for the reactor core have been written and debugged. The design iteration method is being developed. The following paragraphs describe the basic analysis employed and the proposed design procedure.

The fuel element configuration is given in Fig. 1.2. It is composed of an inner plug or spine, an inner flow annulus, an inner graphite ring, a fuel region, an outer graphite ring, and an outer flow channel. The heat-transfer and flow analysis must be performed simultaneously, because the coolant temperatures determine the fraction of the heat that flows to the inner and the outer flow channel. The analysis is performed by setting up a heat balance at each material boundary and iterating on the coolant temperatures. A Peaceman Rackford matrix solution is used to evaluate the temperatures at the boundaries for each iteration. The flow is then adjusted to produce the same pressure drop across the inner and outer flow channel. This procedure comprises the basic heat transfer and flow analysis section of the code. The reactor core will be designed by using the basic computation to satisfy the design criteria, which are as follows:

1. Channel exit temperature ratio

$$\text{CONVDT} = \frac{T_I - T_O}{T_{AV}}$$

T_I = Inner channel exit temperature

T_O = Outer channel exit temperature

T_{AV} = Mixed mean exit temperature

2. Core pressure drop
3. Maximum fuel temperature

Fig. 1.2--Fuel element configuration

The size of the inner plug will be adjusted to satisfy the specified channel exit temperature ratio, CONVDT. The fuel element pitch will be varied to adjust the over-all core pressure drop, and the maximum fuel temperature will be kept within bounds by varying the number of fuel elements. Unfortunately, the design parameters are not independent. Therefore, a stable iterative technique must be developed to find the core dimensions that will simultaneously satisfy all three design criteria.

Core Cost

The core cost will be computed by two existing codes, GAFFEE and WAMPUM. Substantial modifications were made to the input and output routines of WAMPUM to make it compatible with the optimization code. Similar but more complex modifications are being made to GAFFEE.

Flow Network Analysis Code

The development of this portion of the optimization program is essentially concluded. The code has been employed successfully to analyze the performance of the 260-Mw(e) reactor and several other systems selected at random within the power range of 100 to 1000 Mw(e). The results have clearly demonstrated the versatility of the code as well as the expected accuracy. Experience with the code, however short, indicates that extending the code to perform the more detailed standard heat balance calculations is worth considering.

Plans for the near future include the solution of a series of problems in order to accumulate some experience on the behavior of the code when random numbers are used to determine the values of the independent variables. The results are expected to furnish useful information for the development of an optimization scheme. They will also be used to accelerate the debugging procedure of other portions of the optimization program.

Tests are presently under way to evaluate possibilities of improving the computational efficiency. Also, it is planned to gradually increase the system complexity and to add more detail as well as new components.

II. FUEL DEVELOPMENT

Work continued during the past quarter to evaluate variations in the fuel and coolant hole patterns of the nonpurged packed-bed fuel element concept. The objective of the work is to develop a geometrically simple concept with a high degree of structural integrity in which temperature gradients and graphite contraction gradients due to fast neutron damage are minimized.

MECHANICAL DESIGN

Nonpurged Fuel Element Design

During the past quarter, design evaluations have been performed on other suitable arrangements of fuel and coolant hole patterns within the fuel element. The two alternative arrangements were evaluated: (1) a spine and sleeve arrangement with 16 fuel holes in the sleeve and (2) a spine and sleeve arrangement with a fuel annulus in the sleeve. These two arrangements, plus the fuel element design reported during the last quarter, should define the basic combinations of fuel holes and flow channels that are suitable for attaining the fuel element design goals.

One purpose for examining the two alternative design arrangements is to determine whether the effect of graphite irradiation contraction on fuel element performance can be minimized by proper design selection. An analysis of graphite contraction data indicates that the effects can be reduced by reducing the mean temperature of the graphite body and minimizing the temperature differences existing within the fueled graphite body while at operating conditions. Thermal analysis has shown that the two alternative designs will have lower mean temperatures and temperature differences, as was anticipated.

The primary components making up the single-row 16-hole fuel element are a bottom reflector, a center spine, a fuel-containing sleeve, and an upper reflector. A cross section of the fuel element is shown on Fig. 2.1.

The fuel-containing sleeve assembly consists of six axial fueled sleeve sections joined together. Each section is approximately 31 in. long making a total active fuel length of 186 in. The diameter of the sleeve assembly is 4.670 in. Each sleeve section is joined to the adjacent

Fig. 2.1--Cross section of fuel element with a single row of 16 fuel holes

sleeve section by a cemented threaded joint. The top of the sleeve assembly is joined to the top reflector by a cemented threaded joint.

Each sleeve section contains 16 equally spaced fuel holes filled with coated fuel particles. Space is provided at the top of each fuel hole to accommodate differential thermal expansion and graphite irradiation contraction between the sleeve and fuel particles. The fuel particles are contained within the fuel holes by the joint with the adjacent section at the top of each sleeve section.

The center spine, which is located inside the sleeve assembly, controls the cross-sectional flow area of the internal coolant channel. The cross-sectional shape of the spine is hexagonal with rounded corners. The use of the multisided spine provides its own spacing with respect to the fuel sleeve. Sufficient radial clearance is allowed to accommodate differential thermal expansion and graphite irradiation contraction between the spine and sleeve. The top and bottom of the spine assembly is joined to the upper and lower reflectors respectively by cemented threaded joints.

The lower reflector, approximately 26 in. long, is made from a solid piece of graphite and contains flow holes to allow the helium coolant to enter the internal coolant channel of the element. The top reflector, approximately 28 in. long, is fabricated from a solid piece of graphite and contains the exit flow holes for the internal coolant stream. The top reflector is joined directly to both the fuel sleeve assembly and the center spine assembly.

The primary components making up the annular bed fuel element are a bottom reflector, a center spine, an annular fuel bed containing sleeve, and an upper reflector. A cross section of the fuel element is shown on Fig. 2.2.

The fuel-containing sleeve assembly consists of six axial fueled sleeve sections joined together. Each section is approximately 31 in. long, making a total active fuel length of 186 in. The diameter of the sleeve is 4.670 in. Each sleeve section is joined to the next sleeve section as is the top of the sleeve assembly and the top reflector, by a cemented threaded joint.

Each fuel sleeve section contains an annular fuel bed filled with coated particles. The annulus extends from one end of the fuel sleeve section to within about 1/2 in. of the other end. The remaining solid portion of the sleeve holds the inner and outer sleeve walls in position and provides material for the joint. Space is provided at the top of each fuel bed annulus to accommodate differential thermal expansion and

Fig. 2.2--Cross section of fuel element with annular fuel bed

graphite irradiation contraction between the sleeve and fuel particles. The fuel particles are contained in the annular fuel bed by the joint with the adjacent section at the top of each fuel sleeve section.

The center spine, top reflector, and bottom reflector are of the same basic design and serve the same purpose as in the single-row 16-hole fuel element.

Graphite Contraction Analysis

The effect of fast-neutron exposure on the dimensional stability and structural properties of graphite is an important factor in the over-all evaluation of predicted performance of a proposed fuel element. From available experimental data, the major effect of large fast-neutron doses on graphite at HTGR temperature levels is to cause the material to contract. Recent data obtained from graphite irradiation tests performed at Hanford* at temperatures up to 1150°C show that above 800°C the extent of contraction is strongly dependent on the irradiation temperature and fast-neutron exposure level. The data also show that extruded graphite bodies have a higher contraction rate in the parallel direction (i. e., along the axis of extrusion) than in the perpendicular or radial direction.

During reactor operation, the HTGR fuel elements will have temperature gradients present within the graphite bodies. As a result, the temperature dependence of the graphite contraction will cause differential contraction strains to be induced in the fuel bodies. The most severe temperature gradients in the graphite fuel bodies are in the radial direction (i. e., parallel with the primary heat flow paths within the graphite). For extruded graphite bodies, the resulting contraction induced strains are most significant in the axial direction, since contraction is greatest in the direction parallel to the extrusion axis of the graphite body. As exposure accumulates, the graphite contracts more rapidly in the hottest parts of the body resulting in tensile forces in the high-temperature regions of the structure and compressive forces at the cooler outer surfaces. A thermally induced strain can be subtracted from the contraction strain while the graphite bodies are at operating temperature. Thus, even though thermal expansion strains are larger in the higher temperature regions, they generally are small with respect to the contraction gradients; strain in the higher temperature regions therefore becomes tensile quite early in life. An additional reduction in strain will result from creep, which occurs during irradiation. Some of the creep strain,

*J. W. Helm, BMI-Northwest (Hanford), private communication; and J. W. Helm, The H-3 Irradiation Experiment: Irradiation of EGCR Graphite: Interim Report No. 2, HW-71500B

up to the amount of the thermally induced strain, will appear as an additive to the residual contraction strain when the fuel elements cool down.

Local differential contraction strains within the three arrangements of the nonpurged packed bed fuel element concept were evaluated at several reactor core locations, where maximum contraction strains are expected to occur. Graphite contraction was evaluated using irradiation data for EGCR needle coke graphite in the direction parallel to the extrusion axis. The three graphite body geometries and fuel distributions studied are listed below:

1. The fuel body arrangement containing 18 holes in two concentric rings described last quarter (see Fig. 2.3).
2. A 16-hole single-ring arrangement of fuel holes within a thick-walled hollow graphite cylinder with an hexagonal graphite spine (see Fig. 2.1).
3. An annular packed bed of fuel particles between two graphite sleeves with an hexagonal graphite spine within the inner sleeve (see Fig. 2.2).

The cumulative contraction profile through the graphite bodies, for the three internal fuel bed arrangements, were evaluated as of the end of each one-year period of the six-year fuel element life at core locations expected to produce the worst damage conditions. All cases are for reactor operation at 275 Mw(e) over the core lifetime. The approximate maximum cumulative local tensile strain was then computed at the end of each time period without correction for creep relaxation. The predicted cumulative local tensile strain for the three fuel element arrangements analyzed are shown in Fig. 2.4. Dashed tangents from each curve at the point of greatest slope identify the period in life and the magnitude of the maximum tensile strain accumulation rate.

Changes in outside dimensions of the fuel elements within the reactor core are not expected to present any serious design problems. Axial shortening of the fuel elements is not restrained, and radial shrinkage at the top reflector spacer where fuel element pitch spacing is maintained is expected to be negligible.

Effect of Irradiation Induced Creep

Although rather large cumulative strain differences are induced by the strong temperature dependence of graphite contraction, a considerable reduction in net elastic strain can be expected because of irradiation-induced creep deformation of the graphite.

Fig. 2-3--Cross section of fuel element with
18 fuel holes in two concentric rings

Fig. 2.4--Maximum local tensile strain induced by contraction gradients in three fuel elements having different fuel bed arrangements

At temperatures below about 2000°C, thermal creep in graphite is extremely small; however, data from irradiation experiments has shown that creep is induced, at even relatively low temperatures, by irradiation in a fast neutron flux. Creep experiments have been reported by Kennedy* at ORNL and by workers at Hanford Laboratories† at temperatures up to 600°C and fast neutron exposures up to 1.6×10^{21} nvt. Also, irradiation creep experiments have been reported by British investigators Losty, et al.,** and Perks and Simmons.††

The available data may be summarized as follows:

1. There is little evidence for a creep-rate dependence on either the irradiation temperature or the type of graphite in the data.
2. The creep is characterized by an initial, stress-dependent "transient" creep strain, followed by "steady-state" creep at a rate proportional to some power of the applied stress between 1 and 1.7. If a linear dependence on stress is assumed, the "specific creep constant" for steady-state creep lies between 1 and 10×10^{-7} /psi per 10^{20} nvt ($E > 0.18$ Mev). The specific creep constant for samples cut parallel to extrusion appears to be less by a factor of about two than for samples cut perpendicular to extrusion.
3. Creep strains of about 0.4% in tensile samples and up to 1.7% in compression samples have been observed. There is little evidence yet to indicate that an ultimate strain limit exists in specimens deforming by irradiation creep.

Using the Hanford data for EGCR graphite (NC-8) cut parallel to the extrusion axis and irradiated under a tensile stress of 800 psi at 475°C to 1.6×10^{21} nvt ($E > 0.18$ Mev), an average specific creep constant of 2.5×10^{-7} /psi per 10^{20} nvt was computed for a tensile creep strain of 0.32%.

During the period of maximum tensile strain accumulation rate, shown by the dashed line tangent to the steepest slope of each curve on

*C. R. Kennedy, Gas-Cooled Reactor Program Semiannual Progress Report, periods ending March 31, 1963 (ORNL-3445); and September 30, 1963 (ORNL-3619); Oak Ridge National Laboratory, 1963-64.

†AGR/EGCR Graphite Newsletter, Numbers 8-14 (HW-74041, HW-75115, HW-76446, HW-77896, HW-79638, HW-82563, and HW-84350); Hanford Laboratories, 1963-64.

**H. H. W. Losty, et al., presented at the Sixth Carbon Conference, Pittsburgh (1963).

††A. J. Perks and J. H. W. Simmons, Carbon 1, 1964, p. 441.

Fig. 2.4, the steady-state axial tensile stress will reach a maximum. Using a specific creep constant of 2.5×10^{-7} /psi per 10^{20} nvt, the maximum local tensile stresses for each of the three fuel element arrangements were evaluated at the time of maximum strain accumulation and are as follows:

1. Two concentric rings of 18 fuel holes (Fig. 2.3).

Maximum tensile strain rate = 0.62% $\Delta L/L$ per year
at 1.3×10^{21} nvt/year ($E > 0.18$ Mev).

$$\sigma = \frac{4.76 \times 10^{-4} \text{ in./in.} \cdot 10^{20} \text{ nvt}}{2.5 \times 10^{-7} \text{ in./in.} \cdot \text{psi} \cdot 10^{20} \text{ nvt}} = 1900 \text{ psi.}$$

2. Single ring of 16 fuel holes (Fig. 2.1).

Maximum tensile strain rate = 0.165% $\Delta L/L$ per year
at 7×10^{20} nvt/year.

$$\sigma = 940 \text{ psi}$$

3. Annular fuel bed in sleeve (Fig. 2.2).

Maximum tensile strain rate = 0.11% $\Delta L/L$ per year
at 7×10^{20} nvt/year.

$$\sigma = 630 \text{ psi.}$$

The stress caused by contraction gradients will be lower than the values given above at any time in life when the slope of the tensile strain versus time curve is less than the peak rate.

In most instances the local thermal-gradient strain differences will add to the residual elastic contraction strains when the reactor is reduced in power, because the initial elastic thermal strain will be reduced by irradiation-induced creep and contraction gradients. Peak shutdown tensile thermal stresses in the axial direction are estimated by a first-order calculation to be 1000 psi within the 18-hole element, 350 psi within the 16-hole single-ring element, and 210 psi within the sleeve used in the annular bed element. The shutdown stress would be the sum of the local creep stress and thermal stress distributions for any specific location and fuel element age.

Fuel Element Bowing Analysis

The development work on the fuel element bowing code, ELBOW, has been completed and debugged. The code is now available for solving the combined effect of bowing resulting from temperature and fast-flux gradients in (1) the concentric-ring fuel element cooled on the outer surface (2) the single-dial packed-bed fuel element cooled on the outer surface,

(3) the dual-dial, packed-bed fuel element cooled on both the inner and outer surfaces, and (4) the hexagonal reflector element. In the case of the hexagonal reflector element, temperature gradients along the axial length are obtained from a separate analysis and read in as input data. Also, an option is available to read in temperature gradients along the axial length for the three fuel element types if desired.

A preliminary analysis has been made of the bowing of a fuel element located at the reflector interface. An element in this position would experience the greatest temperature differences from one side of the element to the other and the greatest regional fast-flux gradients of any in the reactor. The analysis was based on the most recent data for fast-flux damage to graphite and creep for graphite exposed to a fast-neutron flux field. Results indicate that after one year of full-power operation, the maximum stress is approximately 290 psi during operation and 465 psi when shut down. The bowing resulting from temperature and fast-flux gradients are in opposite directions. When the reactor is shut down and the temperature bowing relaxed, the net deflections are therefore greater; consequently, the maximum stress is greater than at full-power operation. The accumulated maximum deflection of the fuel element, which occurs at the outlet end of the reactor, is approximately 2.2 in. after one year of full-power operation. The temperature gradients calculated for this case are preliminary and will be checked against more detailed temperature results now being calculated.

A plan is being considered in which the fuel elements at the reflector interface would be rotated 180° after a period of plant operation. These elements could then remain in the reactor for extended times without the maximum accumulated deflection at the outlet end of the reactor exceeding the capabilities of the fuel handling equipment. The analysis of this situation requires that the ELBOW code be revised, and this work is now being planned.

Fuel Element Vibration Test

The fuel element vibration tests have been completed. Three types of tests were conducted on a 19-element cluster: (1) Coolant flow (air flow at 50% to 150% of design Reynolds number) was utilized as the only means of inducing vibration. This is the most significant test, because it demonstrates the vibration characteristics of the elements in reactor normal operation. (2) The elements were subjected to pulsating flow. This simulates a postulated condition in which the coolant flow is surging in the ducting or a beat phenomenon is present as a result of the circulators being at slightly different speeds. (3) The vibration characteristics were further studied by forcing vibrations with a motor and eccentric at two locations within the central element.

The central element was instrumented with seven accelerometers spaced at 35-in. intervals to provide a means of determining the frequency and amplitude of vibrations. The significant results are as follows:

1. It was determined that the sleeve and compact type fuel element vibrates with a maximum amplitude of 0.0001 in. at 26 cps for normal reactor flow conditions. This measurement was taken 29 in. from the base of the fuel element, which is very close to an antinode. Measurements taken at other positions gave lower deflections.
2. The 26 cps frequency observed was present for all flow tests without special vibration forcing devices. The rod thus vibrates in the fourth mode, which has nodal points at or near spacer locations.
3. Higher modes of vibration were superimposed on the fourth mode at higher flow rates, but the resulting deflections were smaller.
4. Higher deflections can be induced in the rod by applying forcing functions to it. One such condition that might be encountered during reactor operation is pulsating coolant flow. With a pulsating flow, the deflections appear to increase linearly with flow rate, the maximum amplitude being 0.0018 in. at 100% of design flow.
5. Forced vibrations by a motor and eccentric were used to further investigate resonant frequencies in a fuel element as assembled in the core with coolant flow. A wide spectrum of apparent resonances were found, but for the many tests conducted, including both motor locations and the pulsed-flow test, the propensity to resonate was greatest at 17, 24, 35, 39, 44, and 54.5 cps; all of these values are within 2 cps of a predicted resonant value.
6. Amplitudes observed during forced vibration tests promptly decreased to the nonforced values when the forcing function was removed.
7. In general, the amplitude forced in one rod is reduced by some fraction ($1/5$ to $1/10$) when transmitted to an adjacent rod. This indicates that unusually large vibrations set up in one rod due to an unforeseen condition would propagate to its neighbor only at greatly reduced amplitudes and therefore would not set up vibration in a large cluster of elements.
8. The sleeve-compact type elements used for vibration testing reflect the early reference design for the large HTGR. However,

it is expected that the vibration frequencies and amplitudes of the packed-bed configuration will be essentially the same as those demonstrated for the sleeve-compact type, since the weight and flexural stiffness of the cross section are similar.

THERMAL DESIGN

Nonpurged Fuel Element

Thermal performance analyses of the various fuel hole and coolant channel arrangements within the nonpurged fuel element were performed, and the results provided basic information for predicting the contraction-induced differential strains within the graphite fuel element bodies.

The effect of adding six holes for coolant flow in a third ring between the fuel holes of the 18-hole design described last quarter was evaluated. The analysis showed that both the fuel and the graphite temperatures were reduced. The maximum temperature difference experienced within the graphite body was also reduced, but it appeared that no large reduction in the maximum thermal gradient induced contraction strain would be achieved by this modification compared with the other internal fuel element arrangements studied.

Temperature profiles within a 16-fuel-hole, single-ring arrangement (see Fig. 2.1) were calculated for various axial locations along the fuel element. These profiles are shown in Fig. 2.5 for a vertical plane through the fuel holes, and in Fig. 2.6 for a plane through the web between fuel holes. The profiles shown are for a design that has 19% coolant channel voidage including both the inner and outer coolant channels and that produces 1.5 times the average element power. Further analysis is continuing to determine the thermal effects of reducing coolant channel voidage and to establish the division of heat flow when more detailed geometry adjustments are made to accommodate mechanical design requirements.

The annular fuel bed concept shown in Fig. 2.2 was analyzed, and the radial temperature profiles at various axial locations along the fuel element were plotted on Fig. 2.7. The profiles shown are for a design to have 20% coolant channel voidage within the core and to produce 1.5 times the average element power. The study of this concept is also continuing.

The thermal performance of a fuel element adjacent to the core side-reflector interface was analyzed. Thermal neutron peaking on the side of the fuel element next to the reflector results in power peaking

Fig. 2.5--Temperature profiles through a fuel hole in a packed-bed 16-hole fuel element having a 19% voidage annular coolant passage and a diameter of 4.67 in. (see Fig. 2.1)

Fig. 2.6--Temperature profiles through the web of a packed-bed 16-hole fuel element having a 19% voidage annular coolant passage and a diameter of 4.67 in. (see Fig. 2.1)

Fig. 2.7--Radial temperature profiles at various axial locations along a packed annular-fuel-bed element having a 20% voidage annular coolant passage and a diameter of 4.67 in. (see Fig. 2.2)

and causes an over-all temperature gradient from one side of the element to the other.

The temperature profiles across an average reflector interface fuel element at several axial locations are shown in Fig. 2.8. These analyses will provide temperature data for (1) bowing analysis of core boundary fuel elements, (2) evaluation of coolant temperature distributions at the core-reflector interface for reflector thermal analysis, and (3) evaluation of the radial fuel zone loading within the core to keep peak fuel bed temperatures within the design limit.

FISSION PRODUCT TRANSPORT

In order to obtain more data on the behavior of condensable fission products in a reactor loop, a series of diffusion-tube experiments has been designed for the General Atomic In-Pile Loop (GAIL) experiment. A diffusion tube is simply a small-diameter tube that bypasses a portion of the coolant stream containing the fission products of interest. If this bypassed flow is small enough and the wall of the tube is sufficiently retentive, the majority of the fission products will have reacted with the inside surface before the flow passes through the tube. Under these conditions, the activity profile along the length of the tube can be theoretically predicted by diffusion theory. Thus, when the flow rate through the tube is known, the measured activity profile along the length of the tube can be used to calculate the diffusion coefficient of the fission product species. From this diffusion coefficient, the mass associated with a diffusing particle can be calculated and, finally, the chemical nature of the fission product can be inferred (i. e., monoatomic, molecular, aerosol, etc.). In addition, the measured activity profile can be used to calculate the fission product concentration at the entrance to the diffusion tube and thereby estimate the amount of condensable fission products in the coolant stream.

The first two diffusion tubes were installed in the GAIL at the non-cooled probe positions located downstream from the regenerative heat exchanger and downstream from the main helium circulators. These two diffusion tubes were simply short sections of 3/16-in. copper tubing through which a small portion of the coolant stream was bypassed. Since the geometry of the probe installation made it very difficult to accurately predict the flow through these tubes, they were intended only to supply some needed data on the procedures and analysis problems that might be encountered in these tests. The data obtained from the two probes exposed during GETR Cycle 59 and a similar set for Cycle 60 are reported below in "Fission-product Deposition and Trapping Studies." In general, it appears that these tests offer a promising line of research and may make

Fig. 2.8--Radial temperature profiles at various axial locations along a 16-hole, 4.75-in. diam, spine-supported fuel element at the core-reflector interface

possible the direct calculation of the release of condensable fission products from the GAIL fuel element.

Starting with GETR Cycle 61, which began in late December 1964, diffusion tubes probe assemblies containing diffusion tubes were installed at cold finger positions 1, 3, and 5; these will be replaced after each reactor cycle. Position 1 is located in the return hot-gas pipe from the fuel element immediately upstream from the regenerative heat exchanger. Position 2 is located immediately downstream from the first section of the main cooler, which is a three-section water-cooled unit, and position 5 is immediately downstream from the third section of the main cooler.

The external appearance of the probes will be similar to the previously used cold finger probes, but they will not be water cooled. Instead, the inside of the probe will contain three 17-in. long copper diffusion tubes mounted in a triangular pattern. Each tube will be orificed so that the flow through the individual types will be approximately 10%, 25%, and 65% of the total flow. After passing through the tubes, the flow will be combined, passed through a flow measuring orifice, and then readmitted to the loop upstream from the circulators. Since each of the probes will have a similar arrangement, each set of three probes will result in nine diffusion tubes for analysis plus the external surfaces of the three containment tubes. The reason for providing three diffusion tubes at each probe location is to obtain a range of velocities, so that the various details of the plateout profiles will be displayed to better advantage.

MATERIALS

Fuel Development

The reference fuel element system for TARGET has retentive coated fuel particles in elements of the telephone-dial geometry. Various tests are being performed on beds of loose coated particles to determine their operating characteristics and guide the selection of suitable coated particles with high fuel loadings. In addition, the compatibility of (Th, U)O₂ fuel pellets with graphite at reactor temperatures is being studied as part of a secondary effort on an alternative fuel concept.

Loose Coated Particle Fuel Systems. Various studies and experiments are being performed on loose beds of coated fuel particles in an effort to aid in the development of practical loose coated particle fuel systems. The objective of these studies is to develop a fuel system with high thermal conductivity and high impact strength (in case of accidental release into the coolant stream) and to determine a fuel particle geometry

that will produce maximum fuel loading while maintaining radiation stability and compatibility with the reactor environment.

The heat-transfer properties of beds of loose coated particles are of particular interest in these studies because of their marked influence on the design and operation of the core. Past tests (see previous quarterly progress reports) have bracketed the range in conductivities to be expected from loose pyrolytic-carbon-coated (Th, U)C₂ particles. It is believed that these investigations have been carried far enough that future work can be concentrated on methods of increasing the conductivity and the determination of the conductivity of beds of PyC-coated (Th, U)O₂ particles and various fuel particles of particular interest.

The conductivity tests are performed on annular beds of fuel particles (see Fig. 2.9) using a comparative test method in which the thermal gradient in the unknown sample is compared with that in a sample of known conductivity. This procedure eliminates the need for heat-flow measurements, if it is assumed that the heat flow from the specimen is dependent only on the cold surface temperature of the specimen. The data are corrected for radiation by assuming that there are n heat shields between the hot and cold surfaces of the specimen, where n is the ratio of the distance between the hot and cold surfaces to the average diameter of the particles. The radiative heat flow, then, is inversely proportional to $n + 1$.

Several tests were conducted in a vacuum to obtain some experimental verification that the method employed to correct the data for radiative heat transfer is valid. The particles tested are described in Table 2.1, and the corrected data are given in Fig. 2.10. Since the particles were in a vacuum and radiative heat flow was subtracted, the remaining mode of heat transfer was by heat conduction through the contact area between the particles. The data on the buffer PyC coated particles (lot 2811-63) show that at 2550°F the measured conductivity was almost all due to radiation. The measured value was 0.789 Btu/hr-ft-°F, and the corrected value was 0.037. The addition of small particles (lot 55, 65 DK-2) to the larger (lot 2576-106) particles caused the corrected conductivity to increase 20%, but also caused a 30% decrease in the uncorrected conductivity. These data make the radiation correction seem reasonable, but it has not yet been experimentally proven.

In an attempt to determine the effect of using a binary system in which the average over-all particle sizes of the two components are separated by a factor of only 5:1, the larger size particles (lot 2576-106) were blended with 13.7 wt-% of the smaller particles (lot 55, 65 DK-2). With the addition of the small particles, the bed density increased from 64.2 to 69.3 vol-% solids. However, the resulting thermal conductivity was 15% lower than that of the larger particles alone before correction

Fig. 2.9--Thermal conductivity apparatus for loose coated particles

Δ BLEND OF 2703-106 WITH 19.2 WT-%
 55; 65DK-2; 74.6 VOL-% SOLIDS IN BED
 $\square$ NO. 2703-106; 66.1 VOL-% SOLIDS IN BED
 $\circ$ NO. 2811-63; 68.0 VOL-% SOLIDS IN BED
 NOTE: GAS PRESSURE $< 2.4 \times 10^{-4}$ MM Hg

Fig. 2.10--Thermal conductivity of particle beds in vacuum

Table 2.1

DESCRIPTION OF COATED PARTICLES USED FOR
THERMAL CONDUCTIVITY AND IMPACT TESTS

Lot No.	Core	Core Diameter (μ)	Coating	Coating Thickness (μ)	Average Particle Diameter (μ)
2576-106	ThC ₂	350-420	Isotropic	205	795
55,65DK-2	UC ₂	88-125	Laminar	25	155
GAIL-24-BLC	(Th, U)C ₂ *	350-420	Triplex	105	595
2703-51	(Th, U)C ₂ *	105-210	Isotropic	215	590

*Th:U ratio of 5:1

for radiative heat transfer. After correction for radiation, the data on the blend was still slightly lower, but the difference was less than experimental error. This is the same result as was obtained previously on a blend containing 20 wt-% small particles (same particles as above) which had a density of 73.8 vol-% solids.

An experiment was performed to determine the relationship between bed conductivity and the conductivity of the solid particles. A sample of 780-S graphite of known conductivity was ground and sieved to a desired size range (417 to 595 μ). The density of the particles was then determined in a helium pycnometer to be 2.10 g/cm³. The packed bed had an apparent density of 48% of the particle density.

The bed conductivity was measured in 45 and 30 psig of helium, and also in a vacuum. The data taken at 30 psig were essentially the same as those taken at 45 psig, indicating that a plateau pressure had been reached. The graphite was then outgassed at 3350°F, and the 45-psig and vacuum tests were rerun. The 45-psig data were essentially unchanged by the outgassing, but the vacuum data were reduced by 10% to 15%. The results are compared to the solid-graphite conductivity in Fig. 2.11.

The increasing conductivity in vacuo with increasing temperature is an indication that the contact resistance between particles and/or the radiation correction errors vary systematically with temperature. In addition, it can be concluded that since the conductivity of coated particle beds in helium (see Fig. 2.28 of GA-5866)* is usually near, or above, that of the graphite particle bed, the "effective conductivity" of the fuel particles is near that of the solid graphite, i. e., between 20 and 30

*TARGET: A Program for a 1000-Mw(e) High-temperature Gas-cooled Reactor, Quarterly Progress Report for the Period Ending November 30, 1964, General Atomic, Report GA-5866, December 31, 1964, p. 42.

Fig. 2.11--Thermal conductivity of solid and granulated 780-S graphite

Btu/hr-ft-°F. This effective conductivity is determined by the individual conductivities of the coating and fuel. Since the direction of heat flow through the coated particle is not defined, and since the individual conductivities of the coating and fuel are not well known at the temperatures in consideration, the effective conductivity is somewhat ambiguous. There are some uncertainties in making the comparison of solid conductivities from heat conductivities, e. g., differences in packing fraction and particle shape. (The graphite particles were very irregular in shape.)

One further conclusion that can be drawn from these tests is that the use of a powdered graphite filler between fuel particles should be of little value, because the bed conductivity of the graphite is no higher than that of the fuel particles. This is in agreement with previously reported tests (see GA 5866), but it is in disagreement with Bildstein, et al.* In the latter work, tests are described on the conductivity of loose pyrocarbon coated fuel particles (~600 μ over-all) mixed with 50 vol-% graphite powder (100 to 200 μ). They report that with the graphite added to the fuel the conductivities were 2.6 and 4.6 Btu/hr-ft-°F at 930° and 2190°F, respectively, and that compared with the results without graphite, these values represented an increase of a factor of 3. However, the particle bed was contained in an annular gap of 1.5 mm, which is only 2 to 3 fuel particle diameters wide. Therefore, bridging is quite likely, and the entire bed volume would be in the region of the "wall effect." The packing density was probably greatly increased with the addition of the graphite powder, but this can only be supposition, since data on packing density was not included in the report. Other effects not discussed in the Bildstein report include: (1) excessive thermal gradients (~2000°C/cm), (2) radiation heat transfer, and (3) gas pressure (tests were at 1 atm helium only).

With the use of loose coated particle fuel systems, the occurrence of a leak in the fuel element could result in particles pouring out into the coolant stream. Experiments are therefore being performed to determine whether coated particles accelerated by the coolant and impacting with the walls of the plenum chamber would break. The effects of particle structure and geometry are being investigated in an attempt to develop fuel particles that will withstand impact at 180 ft/sec.

Various experimental devices are being evaluated for use in measuring the impact strength of coated particles. The present test device consists of a narrow bore particle gun utilizing a high-velocity air stream as the propellant. A phototube-electronic counter device is used to measure the time of flight during the first and last halves of the particle's flight through the bore. The particle then impacts with a steel plate 1/2-in. beyond the bore exit.

*H. Bildstein, et al., "Irradiation Behavior of Coated Fuel Particles," Third United Nations Conference on the Peaceful Uses of Atomic Energy, A/Conf. 28/P/398, Austria, May 1964.

In an attempt to reduce the previously reported scatter in particle velocity, the bore diameter of the particle gun was decreased in size. However, the results with this equipment were similar to those obtained before the modification. Although the impact velocity at which half the particles broke was unchanged, the threshold velocity at which the particles just began to break was higher than before.

The impact failure of Triplex coated particles (from the lot used in GAIL IV) was found to be dependent on over-all particle size (and mass). The coated particles described in Table 2.1 were separated into three size ranges and tested at constant velocity. The size ranges, average impact velocities, and failure rates are given in Table 2.2.

Included in Table 2.2 are the impact data on the thick isotropic PyC-coated (Th, U)C₂ particles also described in Table 2.1. These particles show superior impact resistance compared with the Triplex coated particles. The internal buffer layer in the Triplex particles resulted in bending failure, whereas the isotropic coated particles failed in compression near the impact point. The lightweight isotropic coated particles performed better than is indicated in Table 2.2, since most failures were chips in the coating that did not extend to the fuel core.

Table 2.2
RESULTS OF IMPACT TESTING OF COATED PARTICLES

Lot No.	Coating Type	Over-all Size Range ^a (μ)	Avg. Impact Velocity (ft/sec)	Number of Tests	Failure Rate (%)
GAIL-24-BLC ^b	Triplex	480-510	73.7	50	6
		510-545	73.9	50	14
		545-580	76.2	50	28
2703-51	Isotropic	540-640	206.5	49	26.5
		540-640	237.4	50	54
		540-640	306.5	50	92

^a—Diameter of coated particle.

^b—Used in GAIL IV element.

Packing studies are being performed to determine the effects of particle and container parameters on packed bed density, ease of separation of different particle types, and extent of particle segregation during operation. This includes the development of a practical means of blending loose particles, loading fuel tubes without segregation, and compacting particle beds.

Several packings were made in 30-in. long by 1/2-in. ID glass tubes using particles of glass and coated fuel particles. No segregation due to extended vibration at ~ 30 g occurred in binary blends as long as two prerequisites were met:

1. Sufficient tare weight was allowed to rest on the particles so that they could not move freely, and
2. An optimum blend was used in which all the voids between large particles were completely filled with small particles.

If (1) was not met, the large particles tended to "float" on the small ones. If (2) was not met, the small particles filtered toward the bottom of the tube.

A packing apparatus for a 30-in. tube with an air vibrator was completed and instrumented for g-level measurement. Two batches of fuel particles were blended and packed to determine packing density, blend uniformity, and extent of segregation under prolonged vibration. The particles are described in Table 2.3.

Table 2.3

DESCRIPTION OF PARTICLES USED IN PACKING TESTS

Lot No.	Particle Type	Particle Diameter (μ)	Coating Type	Coating Thickness (μ)	Packed Bed Density (Vol-% Solids)
61, 63DK-2	UC ₂	90-125	Laminar	25	70
2703-51	5:1(Th, U)C ₂	105-210	Isotropic	215	67

Using dry and wet blending methods, only very nonuniform blends have been packed, owing to the small diameter ratio of large to small particles. However, packing densities approaching 80 vol-% solids have been achieved. Uniform blends of these particles should be obtainable if the pores between the large particles are allowed to open under vibration due to dilatancy.

Carbon dust, apparently due to particle attrition, was observed after exposure to prolonged 30-g vibration. The particles were leached, and the leach solutions revealed that $\sim 10\%$ of the particle coatings had broken at these extremely high vibrational load levels. (Subsequent tests are being conducted at the lower (< 1 g) vibrational loads actually expected in the reactor.)

A test was conducted to determine the extent of segregation of a blend of two different coated particle types when exposed to ambient reactor vibration without a tare or restricting force at the top of the particle column. The blend consisted of 50 wt-% 150- μ (over-all diameter) particles (4.75 g/cm³ particle density) and 50 wt-% 590- μ (over-all diameter) particles (1.81 g/cm³ particle density). The 30-in. long by 1/2-in. ID tube was vibrated axially at $\sim 1/2$ g (white-noise spectrum). During the first few minutes of the test, the large particles began to "float" to the top of the column, but this process stopped after a layer of particles formed ~ 3 particle-diameters deep. Vibration was continued for 10 hr with no change in the blend.

An apparently successful method of blending particles of different sizes was developed. This consists of simultaneously metering the two particle batches through individual hoppers with adjustable orifices. The desired quantity of each particle type is loaded into the hoppers. If the orifice sizes are properly adjusted, the two hoppers will empty at the same time. As long as the emptying time was over ~ 30 sec (for 100 g of each particle type), the blend uniformity as visually determined was good. (Quantitative evaluation of the uniformity has been unsuccessful owing to segregation during sampling.)

A test was conducted on a 12-in. long by 1/2-in. diameter column of particles held firmly in place in a graphite tube by a 1/2-in. diameter by 1/2-in. long piece of styrofoam, partially compressed by the lid of the tube. Such a scheme was suggested to restrain the particles during shipping and handling. The styrofoam could then be volatilized during reactor startup. This test was to determine the extent of residue left. The assembly was heated to 800°C (minimum fuel temperature anticipated in the reactor) for one hour in helium, removed, and inspected microscopically. There was no visual evidence of residue from the styrofoam and the particles remained loose and free-flowing.

Oxide Pellet Fuel Systems. Oxide fuel, in the form of cylindrical pellets, is being investigated as an alternative fuel system for HTGR. These studies are aimed at assessing the concept of a fuel element having oxide fuel pellets stacked in tubular (telephone-dial) graphite elements. Current work is concerned with: (1) evaluating the performance of oxide pellets in graphite by a high-temperature irradiation experiment to be conducted as a GA-ORNL cooperative task, and (2) assessing the compatibility of the oxide-graphite system by measuring the rates and consequences of carbide formation and hydrolysis under various conditions of temperature and gas overpressure.

During the past quarter, the design of the planned oxide-graphite irradiation experiment was discussed with ORNL, and most of the

operating techniques and examination methods for the experiment were agreed on (see below "ORNL-GA Cooperative Irradiation Program for TARGET"). Fabrication of the (Th, U)O₂ fuel pellet specimens was begun and is about 50% complete. Two small (50-g) lots of enriched UO₂ were procured and tested for sintering behavior. Although both lots were significantly different (less reactive) from the UO₂ (natural uranium) powder used in initial experiments, one lot was made sufficiently sinterable by chemical reprocessing. This lot has been used to fabricate forty specimens of the high-uranium composition (Th, ⁶⁷U, ³³O₂) to be irradiated in the lower region of the capsule. Fabrication of the low-uranium specimens (Th, ⁹⁰⁸U, ⁰⁹²O₂) will await final confirmation of the desired fuel composition and specimen dimensions being determined in the ORNL computer program.

The compatibility of the ThO₂-UO₂-graphite system is being studied by use of a high-temperature microscope to visually measure the rate of interfacial diffusion in sandwich specimens. An experiment was conducted at 2000°C using ThO₂ and pyrolytic carbon. Emphasis has now shifted to the UO₂-graphite system, since UO₂ is the more reactive material in the (Th, U)O₂ system. Single crystals of ThC₂ grew profusely around the carbide reaction area during a run between ThO₂ and PyC that was cycled. The crystals appeared to have developed on all three available carbon-bearing sources (PyC, carbide, and graphite) from vapor-deposited ThO₂. This result has implications for UO₂. The matter of UO₂ volatility may be of greater concern than at first anticipated, because it is more volatile than ThO₂. The question of its stability in solid solution with ThO₂, as well as the potential of producing UC₂ crystals in undesirable regions, may need study.

Fuel Irradiations

Irradiation studies on the TARGET program are intended to guide the selection of coated particles that have good stability and good fission-product retention under high-temperature, high-burnup, and high-fast-flux reactor environments. The particles should also be economical and contain high fuel loadings. To achieve this goal, a series of controlled-temperature capsule irradiations are being conducted to quickly test many types of advanced coated particles and evaluate factors influencing irradiation stability.

Results from previous capsules in this series have indicated that the irradiation stability of coated particles is greatly increased by the presence of one or more of the following features:

1. A porous pyrolytic carbon (PyC) "buffer zone" between the fuel particle and the coating.

2. Porosity in the fuel particle.
3. An isotropic PyC layer.
4. Circumferential discontinuities in the coating.

Capsule P7. Capsule P7 was a screening test for twelve types of coated particles to evaluate the radiation stability of isotropic, buffer-isotropic, and Triplex PyC coatings of varying thicknesses on oxide and carbide particles of varying size and porosity. The capsule was irradiated at an average fuel temperature of 1250°C for 30 hr and 1000°C for 800 hr to a burnup of 16 to 24% FIMA, (fissions per initial heavy metal atom) and a fast-flux exposure of 3×10^{20} nvt ($E > 0.18$ Mev).

A description of the coated particles tested and the preliminary postirradiation examination results were presented in the last quarterly report, GA-5866. The examination has been completed and the final results are given in Table 2.4.

The results showed a good correlation of coating failure with coating thickness and particle size (i. e., failure increased with decreasing coating thickness and/or increasing particle size). Extrapolation of the data indicated that a minimum coating-thickness to core-diameter ratio of about 0.4 (80- μ coating on 200- μ diameter particles) may be required for irradiation stability. Buffer-isotropic (BISO) coatings appeared to be equivalent in irradiation stability to Triplex coatings, and coated oxides were equivalent to coated carbides.

Capsule P8. Capsule P8 was a high-temperature screening test for twelve types of advanced coated particles containing high fuel loadings (> 25 vol-%), and included various types of BISO (buffer-isotropic) and ISO (isotropic) PyC coated oxides; and buffer-isotropic and Triplex PyC coated carbides. Several of the samples had been thermally pretreated to cause fuel migration such as might be present in the coatings at the end of reactor core life in order to aid in evaluating the effect of diffused uranium on the irradiation stability of coated particles.

The capsule achieved 12% fuel burnup (FIMA) at fuel temperatures of 900° to 1500°C and an estimated fast-flux exposure of 3×10^{20} nvt. Flux-dosimeter analyses are currently in progress.

A description of the coated particles tested and the results of the postirradiation examination are given in Table 2.5. A Triplex coated carbide, which had shown outstanding stability in previous tests in the P6 and P7 capsules and in the ORNL B9-14 and B9-15 capsules, again did not fail. A BISO coated oxide, which had survived 7% burnup at 1370°C

Table 2.4

POSTIRRADIATION EXAMINATION OF COATED PARTICLES TESTED IN CAPSULE P7

Lot Not.	Coating		Particle			Burnup (% FIMA) ^b	Microscopic Examination		Metallographic Examination			
	Type	Thickness (μ)	Type	Size (μ)	Porosity ^a		Total Particles Examined ^c	Broken Coatings (%)	Total Particles Examined	Broken Coatings (%)	Unbroken Coatings (%)	
											Damaged	Undamaged
2340-86E	Triplex	100	UC ₂	200	Dense	23.5	1000	0	601	0	0	100
2576-58E	Triplex	61	UC ₂	200	Dense	16.9	500	38	233	21	15	64
2576-28E	Triplex	42	UC ₂	200	Dense	16.3	400	84	350	87	5	8
2511-85E	BISO	61	UC ₂	200	Dense	15.8	800	21	409	6	3	9
2576-50E	BISO	48	UC ₂	200	Dense	16.7	1000	70	418	62	29	9
2511-95E	BISO	31	UC ₂	200	Dense	16.5	1000	100	193	92	5	3
2511-123E	ISO	65	UC ₂	200	Porous	15.9	1500	25	792	17	81	2
2576-46E	ISO	43	UO ₂	200	Porous	18.7	1000	70	150	78	22	0
2511-121E	ISO	33	UO ₂	200	Porous	22.0	1000	96	295	93	6	1
2511-127E	BISO	279	UC ₂	1500	Dense	(d)	3	100	3	100	0	0
2701-9E	BISO	101	UC ₂	1500	Dense	(d)	3	100	3	100	0	0
2576-60E	ISO	91	UO ₂	1300	Porous	(d)	11	100	11	100	0	0

^aAll dense particles are 95% T.D. (or greater) and spheroidal; all porous particles are 50% T.D. and irregularly shaped.

^bPercent fissions per initial heavy metal atom.

^cEstimated.

^dNot determined.

Table 2.5

POSTIRRADIATION EXAMINATION OF COATED PARTICLES TESTED IN CAPSULE P8

Lot.No.	Coating		Particle			Irradiation Conditions		Microscopic Examination		Metallographic Examination				
	Type	Thickness (μ)	Type	Density (% T. D.)	Size (μ)	Temp. ^b (°C)	Burnup ^c (% FIMA)	Total Particles Examined	Broken Coatings (%)	Total Particles Examined	Broken Coatings (%)	Unbroken Coatings		External Attack (%)
												Damaged ^e (%)	Undamaged (%)	
2703-117E ^d	BISO	104	UO ₂	60	400	900-1400	12.7	127	9	91	12	30	58	18
2703-117EH ^e	BISO	104	UO ₂	60	400	900-1400	12.9	133	11	114	10	13	77	1
2702-39E	BISO	106	UO ₂	60	525	950-1420	12.0	71	96	59	90	8 ^f	2 ^f	27
2702-41E	BISO	97	UO ₂	60	550	950-1420	12.5	60	87	58	91	9 ^f	0 ^f	28
2703-125E	ISO	97	UO ₂	60	400	900-1400	12.6	124	23	88	10	56	34	17
2703-115E	ISO	98	UO ₂	35	400	1020-1520	12.9	79	86	77	69	21	10	50
2703-123E	BISO	110	UO ₂	35	400	1020-1520	12.7	62	71	54	91	4	5	48
2703-95E	BISO	111	UC ₂	95	400	1020-1520	12.2	81	83	55	45	45 ^f	10 ^f	71
2703-95EH ^e	BISO	111	UC ₂	95	400	950-1420	11.1	99	74	56	54	14	32	47
2702-33E	Triplex	111	UC ₂	95	400	1000-1500	11.5	100	99	42	80	18 ^f	2 ^f	74
2702-33EH ^e	Triplex	111	UC ₂	95	400	1000-1500	11.1	87	96	54	94	6 ^f	0 ^f	94
2340-86EA	Triplex	103	UC ₂	95	200	1000-1500	10.9	1125	0	900 ^h	0	0	100	0

^aOxide particles are irregularly shaped; carbide particles are spheroidal.

^bMaximum and minimum temperatures at full reactor power. The maximum observed temperature difference between the four cells was 190°C, and the average ΔT was about 125°C.

^cPercent fissions per initial heavy metal atom.

^dPreviously tested in ORNL B9-23 capsule to 7% burnup at 1370°C without apparent failure.

^eHeat treated before irradiation to simulate amount of fuel migration expected in coatings at end of reactor core life. Sample No. 2703-117EH had 2- μ migration, 2703-95EH had 43- μ , and 2702-33EH had 63- μ .

^fRelative ratio of damaged to undamaged coatings was estimated.

^gPreviously tested in General Atomic P6 and P7 capsules, and ORNL B9-14 and B9-15 capsules, without failure.

^hEstimated.

in the ORNL B9-23 capsule, had about 12%* failure in capsule P8. All other types of particles failed badly (45% to 95%), except for an ISO coated oxide which had 10% failure. (See below, "Effect of Burnup Rate on Coated Particle Temperature," p. 47, for a discussion of a possible reason for the high failure rate of coated particles in this capsule.)

External attack was observed on a high percentage of coatings, both broken and unbroken. In many cases, coating fracture appeared to have originated from the outside in. The cause of this attack is not known as yet.

Some tentative conclusions may be drawn from this experiment:

1. For carbide particles, BISO coatings appeared to be better than Triplex coatings of the same thickness (45% failure versus 80%).
2. Preirradiation high-temperature thermal treatment of coated oxide particles (resulting in 2- μ fuel migration) appeared to have little effect on their radiation stability (10% failure compared with 12% for unheated particles). However, the same thermal treatment appeared to have a slight detrimental effect on coated carbides, apparently because of the higher amount of fuel migration (BISO coatings had 54% failure after heating, which resulted in 43- μ migration, versus 45% unheated; and Triplex coatings had 94% failure after heating, which resulted in 63- μ migration, versus 80% unheated). The pre-irradiation thermal treatment was intended to simulate possible fuel migration in the coatings at the end of reactor core life in order to aid in evaluating the end-of-life stability of coated particles.
3. Oxide particles of 60% density appeared to be superior to 35% dense particles (12% failure versus 91% with equivalent BISO coatings, and 10% versus 69% for ISO coatings). This is the reverse of what might be expected.
4. The high failure of most particles in this experiment was probably due to the low coating-thickness to core-diameter (t/d) ratio (0.25 or less). The Triplex with a t/d ratio of 0.50 did not fail, and results from previous capsules at BMI and ORNL as well as at General Atomic have indicated that a t/d ratio of at least 0.4 may be required for stability in high-burnup, high-temperature irradiations.

*Failure percentages reported are those from the metallographic examination.

Capsule P9. Capsule P9 was a screening test for various types of advanced coated particles to evaluate the effects of over-all coating thickness and porous "buffer" layer thickness on the radiation stability of various types of buffer-isotropic coated oxides and carbides. A description of the coated particles tested is given in Table 2.6.

The capsule was irradiated to an estimated 8% to 10% fuel burnup at fuel temperatures of 1700° to 1350°C. Postirradiation examination is currently in progress.

Capsule P10. Capsule P10 is a test to evaluate the effects of various buffer and isotropic coating structures, and over-all particle size (with constant coating-thickness to particle-diameter ratio) on radiation stability. A description of the coated particles is given in Table 2.7.

The capsule is currently being irradiated in the GETR at fuel temperatures of 1400° to 1550°C.

Capsule P11. The objectives of the P11 capsule are:

1. To determine the effect of burnup rate on coated particle stability at two different irradiation temperatures (1200° and 1500°C).
2. To retest promising coated fuel particle samples from previous capsules which operated at lower temperatures.
3. To make additional comparisons of the radiation stability of buffer-isotropic and Triplex coatings.

Coated UC₂ and (Th, U)C₂ particles with different Th:U ratios (0:1, 0.4:1, 1:1, and 3:1) will be used to achieve the burnup rates of 18%, 13%, 9%, and 5% per month, respectively. Also included in this capsule is a sample of ORNL Sol-Gel oxide particles with a Triplex coating. This will be the first General Atomic irradiation test of Sol-Gel particles. A description of the coated particles and the design irradiation conditions are given in Table 2.8.

Capsule P12. Capsule P12 is scheduled to be a three-cycle irradiation test to evaluate coated particles at lower burnup rates and to achieve a significant fast-flux exposure ($>10^{21}$ nvt). (Th, U)O₂ particles with Th:U ratios of 17:1 (fertile) and 4:1 (fissile) will be used to achieve burnups of 3% and 11%, respectively, in three cycles. Coating types will be selected after postirradiation examination of the P9 and P10 capsules. The design fuel temperature will be 1400° to 1500°C. Capsule P12 is tentatively scheduled to begin irradiation in April 1965.

Table 2.6

DESCRIPTION OF COATED PARTICLES TESTED IN CAPSULE P9

Lot	Coating ^a					Particle		Size (μ)
	Type	Thickness (μ) ^b				Type ^c	Density (% T. D.)	
		B	I	C	Total			
2813-19E	ISO	--	107	--	107	(Th, U)C ₂	86	250-350
2813-21E	BISO	21	82	--	103	(Th, U)C ₂	86	250-350
2813-25E	BISO	33	70	--	103	(Th, U)C ₂	86	250-350
2813-27E	BISO	26	59	--	85	(Th, U)C ₂	86	250-350
2813-29E	BISO	35	92	--	127	(Th, U)C ₂	86	250-350
2813-33E	BISO	35	157	--	192	(Th, U)C ₂	86	250-350
2813-43E	ISO	--	(29-63)	--	92	(Th, U)C ₂	86	250-350
2813-45E	ISO	--	(30-70)	--	100	(Th, U)C ₂	86	250-350
	ISO							
	ISO							
2813-49E-2	BISO	35	79	--	114	(Th, U)O ₂	71	300-420
2813-49E-3	BISO	35	111	--	146	(Th, U)O ₂	71	300-420
2813-49E-4	BISO	35	135	--	170	(Th, U)O ₂	71	300-420
2152-37E ^d	Triplex	21	56	61	138	(Th, U)C ₂ ^e	91	150-250

^a Detailed structural characterizations of the pyrolytic carbon coatings are being obtained from deposits on small graphite disks that were in the fluidized bed with the fuel particles during the coating operation. Structural properties being determined include density, preferred orientation, and crystallite size.

^b B = buffer; I = isotropic; C = columnar.

^c Th:U = 1:1 except where noted.

^d This sample was also irradiated in the ORNL C1-11 capsule.

^e Th:U = 0.6:1.

Table 2.7

DESCRIPTION OF COATED PARTICLES TO BE TESTED IN CAPSULE P10

Lot	Coating ^a					Particle			Remarks
	Type	Thickness (μ) ^b				Type ^c	Density	Size (μ)	
		B	I	C	Total				
2813-95E	ISO	--	109	--	109	(Th, U)C ₂	Porous	250-350	Discontinuity at 32μ
2813-99E	ISO	--	108	--	108	(Th, U)C ₂	Porous	250-350	Discontinuity at 22μ
2813-101E	ISO	--	98	--	98	(Th, U)C ₂	Porous	250-350	Discontinuity at 24μ
2813-93E	ISO	--	92	--	92	(Th, U)C ₂	Porous	250-350	Discontinuity at 26μ
2813-85E	BISO	42	57	--	99	(Th, U)C ₂	Dense	250-350	(e)
2813-79E	BISO	33	74	--	108	(Th, U)C ₂	Dense	250-350	(e)
2813-87E	BISO	39	57	--	96	(Th, U)C ₂	Dense	250-350	(e)
2813-83E	BISO	43	51	--	94	(Th, U)C ₂	Dense	250-350	(e)
2813-81E	BISO	17	61	--	78	(Th, U)O ₂	Porous	175-280	
2813-103E	BISO	47	78	--	125	(Th, U)O ₂	Porous	300-420	
2813-105E	BISO	49	146	--	195	(Th, U)O ₂ ^d	Porous	500-590	
2152-37E	Triplex	25	41	37	103	(Th, U)C ₂ ^d	Dense	150-250	C1-11, P9 Triplex

^a Detailed structural characterizations of the pyrolytic carbon coatings are being obtained from deposits on small graphite disks that were in the fluidized bed with the fuel particles during the coating operation. Structural properties being determined include density, preferred orientation, and crystallite size.

^b B = buffer; I = isotropic; C = columnar.

^c Th:U = 1:1 except where noted.

^d Th:U = 0.6:1.

^e Buffer coatings of these samples vary in porosity.

Table 2.8

DESCRIPTION OF COATED PARTICLES TESTED IN CAPSULE P11

Lot	Coating ^a		Particle			P11 Design Irradiation Conditions		Other Irradiation Tests
	Type	Thickness (μ)	Type ^b	Th:U Ratio	Size (μ)	Temperature (°C)	Burnup (%)	
2813-137E	BISO	106	(Th, U)C ₂	0.4:1	300	120C, 1500	13	---
2813-145E	BISO	99	(Th, U)C ₂	1:1	300	120C, 1500	9	---
2813-143E	BISO	102	(Th, U)C ₂	3:1	300	120C, 1500	4.5	---
2813-113E	BISO	75	UC ₂	---	200	120C, 1500	18	P6
2813-141E	BISO	102	UC ₂	---	200	120C	18	---
2340-86E	Triplex	103	UC ₂	---	200	120C	18	B-14, B9-15, P6, P7, P8
2152-37E	Triplex	140	(Th, U)C ₂	0.6:1	200	150C	11	C1-11, P9, P10
2702-57E	Triplex	99	(Th, U)O ₂ (Sol-Gel)	14:1	275	150C	1	ORNL-ETR

^a Detailed structural characterizations of the pyrolytic carbon coatings are being obtained from deposits on small graphite disks that were in the fluidized bed with the fuel particles during the coating operation. Structural properties being determined include density, preferred orientation, and crystallite size.

^b All particles are dense and spheroidal.

Effect of Burnup Rate on Coated Particle Temperature. The thermal gradient across a coated particle during irradiation is a linear function of the burnup rate. Thus, coated particles tested in P-capsules, which reach 10% burnup in only one month rather than in the scheduled 72-month (6-year) HTGR core life, experience thermal gradients 72 times as large as they would in actual service. Using experimentally determined values of thermal conductivity for the PyC coatings, a 300- μ diameter particle with a 100- μ buffer-isotropic coating is found to have a temperature gradient across the coating of 400°C for a surface temperature of 1500°C. Thus, the inside of the particle might reach a temperature of 1900°C in an accelerated burnup rate P-capsule test, whereas at the expected HTGR burnup rate the central temperature would only be 1515°C.

These accelerated burnup tests, with resulting high fuel temperatures, provide a very severe test of coated particles and may well have been a major contributor to the high failure rates that have been observed in many of the P-capsule experiments. It is therefore planned to reduce the burnup rates in future P-capsule tests. Capsule P12 will be a three-cycle test in order to lower burnup rates to 1% and 3% per month for the two types of fuel particles to be used. Various burnup rates will be evaluated in capsule P11.

Evaluation of Irradiation Coated Particles by Radiography. Radiography is a nondestructive testing tool that can yield valuable data on irradiated coated particles before they are subjected to destructive metallographic examination. Initially, radiographs were produced of irradiated samples from the P6 capsule. More recently, efforts were made to increase the number of "hot" particles that could be examined at one time. Six particles from the P7 capsule have been radiographed, and the films are being evaluated. These particles registered a radiation level of ~ 5 r/hr at 1 ft.

Graphite Development

It is the objective of the graphite development program to develop sources of improved, less expensive fuel element graphites. The major portion of the effort is given to evaluating vendor stocks and to leading the vendors to develop suitable materials. A large lot of 780-S graphite is being examined to determine its usefulness as a solid stock that would be machined to obtain a nonpurged, multihole element. Experimental samples from another vendor are also being examined. This vendor expects to form multihole elements, or tube stock, directly by a new forming method.

780-S Graphite. Evaluation of the physical properties of a 72-log shipment of 5-in. -diameter 780-S graphite is under way. Density, thermal expansion, thermal conductivity, crystal lattice parameters, chemical purity, burst strength, and integrity (as evaluated by ultrasonic inspection) have been determined, and the results are reported in Table 2.9. All the properties measured to date, with the exception of burst strength, are within acceptable limits for fuel element sleeves. Burst strength shows a large scatter from piece-to-piece, and the lower values are not acceptable. These burst strength results indicate that a test would have to be run on each piece considered for reactor use.

Table 2.9

PROPERTIES OF 780-S GRAPHITE

Property	Specimens Tested	Average Value
Density, g/cm ³	5	1.83
Coefficient of thermal expansion at 400°C (x 10 ⁻⁶ °C ⁻¹):		
Axial.	2	2.6
Radial	2	3.6
Circumferential	2	3.5
Coefficient of thermal conductivity at 1205°C, Btu/hr-ft-°F:		
Radial.	5	24
Crystal Lattice Parameters, Å:		
a ₀	5	2.129
c ₀	5	6.730
L _c	5	222
Burst strength (hoop stress at fracture), psi	5	338 ^a 552 903 1803 2400
Chemical purity	5	Acceptable
Integrity (evaluated by ultrasonic inspection)	5	No indicated defects

^aThe spread of values was so great that individual values are given for each specimen.

H-315 Graphite. The thermal expansion characteristics of special sample H-315 were measured in order to evaluate the anisotropy of this graphite. At 400°C, the coefficients of thermal expansion were: $5.8 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ axial, $5.7 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ radial, and $4.7 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ circumferential. These results indicate that graphite H-315, which is formed by a new, undisclosed method, is relatively isotropic when compared to the extruded graphites usually considered for fuel element sleeves.

H-315A Graphite. A sample of grade H-315A graphite 4-5/8-in. OD by 3-3/8-in. ID by 14-1/2-in. long that had been directly formed to these diameters by the new process was received from the vendor, who describes this grade as having been treated beyond the grade H-315 material that was received at General Atomic during the last quarter (see GA-5866, Table 2.4). The nature of the additional treatment was not described, but it was designed to improve the density and helium permeability of the graphite. An ultrasonic inspection disclosed no defects, and a test for hoop strength gave a value of 2310 psi. Both tests show the sample to be acceptable for sleeve graphite.

H-316 Graphite. A sample of H-316 graphite that is 4-1/16-in. OD by 11-7/8-in. long and that has four axial 3/4-in. ID holes through the piece was received from the vendor, who states that this sample is intended primarily to demonstrate their ability to form this type of geometry directly by their new forming method. The sample appears sound, and the holes appear straight and evenly spaced.

Coke Graphites. A chemical analysis was made of a vendor-supplied sample of raw coke and also of a sample of graphite extruded at General Atomic from this same coke. The coke impurities greatly exceeded the maximum specified by General Atomic, but the extruded graphite stock made from the coke met all the purity specifications except for the boron content. The boron content of this stock was even greater than that of the raw coke, which suggests that there may have been boron contamination in the graphitization furnace.

The chemical analysis shows that a relatively impure coke can be used to fabricate a graphite that will meet nuclear purity specifications. However, it must be noted that the graphitization at General Atomic was carried out under vacuum during the initial stages where temperature was less than 2000°C; this is not standard industrial procedure.

Graphite Irradiations

The graphite irradiation program is designed to evaluate various graphites in support of reactor design and materials development. The program includes studies of fuel element graphite, reflector graphite, and control rod graphites.

Fuel Element Graphite. Capsule G9 was designed to irradiate reactor graphites in the temperature range 450° to 1100°C to a maximum exposure of about 2×10^{21} nvt ($E > 0.18$ Mev). Graphites included in the capsule were selected from the leading candidate materials submitted by vendors. The graphite grades included 780-S and 711-TS, H-207-85 (isotropic), and pyrolytic carbon structures representative of various coatings for fuel particles. The capsule has operated successfully for four cycles of a scheduled six-cycle exposure in the GETR (see "Capsule Irradiations" below for operating details).

An effort is being made to encourage the graphite vendors to develop and fabricate prototype samples utilizing isotropic or uncalcined cokes in addition to the conventional petroleum coke-pitch materials. Experimental samples of isotropic graphites have been irradiated by BMI-Northwest (Hanford) and by investigators in the United Kingdom, and these have shown less irradiation contraction than conventional graphites.

Preliminary design has been initiated on an experiment to irradiate graphite with a 200° to 250°C thermal gradient. The objective of this experiment will be to determine whether stresses generated by differential contraction will fracture the graphite when fracture strains are reached, or whether strain limits are enhanced by neutron irradiation. A decision to perform this experiment is being held up until the latest graphite irradiation data from BMI-Northwest have been assessed for the TARGET design.

Preliminary plans for Capsule G10 are under way. Graphite samples will be selected based on the results of current surveys being conducted with the vendors.

Reflector Graphites. No experimental work is being done specifically on reflector graphites. Current efforts on fuel element graphites and BMI-Northwest's extensive data on moderator graphites are being monitored to provide specification data.

Control Rod Materials. Material for the control rod design consists of B_4C dispersed in graphite. Current efforts are being directed toward getting prototype samples made to a tentative specification that has been written based on previous irradiation data.

Battelle-Northwest Cooperative Studies. Close liaison is being maintained with the extensive graphite irradiation program being carried out at Hanford in order to ensure that the results of the program will be used effectively in the TARGET design.

The following experiments in support of TARGET are continuing at BMI-Northwest:

1. Impregnated HLM-85 and NPR (TSX) in the ETR at 600° to 700°C.
2. Tube geometry HLM-85 in a Hanford reactor.
3. Telephone-dial geometry 780-S in a Hanford reactor.

Contraction data for graphites irradiated in the temperature range 800° to 1150°C were received from BMI-Northwest (see "Graphite Contraction Studies" below).

Graphite Samples for ORNL Irradiation Tests. Plans are being formulated to fabricate the graphite crucibles to be used in various coated particle experiments from candidate fuel element graphites. Initially, 780-S graphite will be used. Several of the coated particle experiments are planned for periods of up to one year in the core of the ETR. These experiments will be operated in the range 1000° to 1400°C to high fast-neutron exposures, and thus dimensional changes in the graphite components are of interest.

Graphite Contraction Studies

Recent information from BMI-Northwest (Hanford) has indicated that the rate of graphite contraction in the temperature range 1050° to 1150°C may be considerably higher than the design values (based on earlier results from irradiations at lower temperatures) previously used for HTGR. The new data also showed a rather strong temperature-dependence over the range 1050° to 1150°C. A possible consequence of such behavior on the part of the graphite in a tubular or telephone-dial geometry fuel element would be the generation of high stress levels owing to differential contraction in the regions of high temperature and high thermal gradient.

Coordinated studies were pursued in several areas to evaluate quantitatively the latest graphite irradiation data and their effect on fuel element performance, to consider alternative designs and materials, and to determine the effect of the anticipated stress gradients on fuel element integrity through out-of-pile experiments.

Effect of Irradiation Contraction and Creep on Stresses in Graphite Fuel Elements. The stress calculations for Peach Bottom and advanced design graphite fuel elements have been reevaluated in the light of recent Hanford data on radiation-induced contraction rates at temperatures of 1050° to 1150°C. This review has taken into account the pertinent experimental data on dimensional changes in graphite, the available data on irradiation-induced creep in graphite, and calculations of the stress and strain build-up in Peach Bottom sleeves and three tentative designs for advanced reactor fuel elements. It has been concluded that Peach Bottom

sleeves will maintain their integrity even if creep is not taken into account. Assuming that irradiation creep data below 600°C may be applied at higher temperatures, two of the three tentative designs for advanced HTGR fuel elements (single-ring fuel holes and an annular bed arrangement) would fall within acceptable limits of stress and strain. The third design (double-ring fuel holes) would not be acceptable using data for EGCR graphite. However, the use of recently developed, stable graphites (such as JOZ graphite) would greatly reduce the stresses and strains involved and would make all three designs acceptable.

Effects of Thermal and Mechanical Stresses on Graphite. An experiment is under way to evaluate the effect of high internal stresses on the strength of graphite bodies. High internal stress levels are being generated by imposing a large transient temperature gradient across a graphite sample. An attempt will be made to analyze the stresses set up and the mode of failure and to extrapolate these results to the problem of the high internal stresses generated in graphite fuel element sleeves by differential contraction during irradiation.

The basic experimental procedure is to inductively heat a hollow graphite cylinder, 2-in. OD by 1/2-in. ID by 10-in. long, so that a 4-in. -long test section has reached the desired uniform temperature. When the test temperature is reached, a stream of cold water is pumped through the 1/2-in. annulus at a velocity of 40 ft/sec. The water rapidly cools the ID of the cylinder and, in transient, imposes a large thermal gradient across the sample.

Experimental runs have been made at 1000°, 1200°, 1400°, 1600°, 1800°, and 2000°C. There was no visible failure after the tests in the three lowest temperature specimens. The three highest temperature specimens all failed, and all in the same mode. The macroscopic failures were two radial cracks, diametrically opposite, running parallel to the axis of the cylindrical specimens. The cracks began on the ID and ran radially 70% to 80% of the way towards the outside diameter. The failed specimens will be examined metallographically to determine whether or not the stresses caused any microcracks in the graphite. No analysis has yet been made of the stress levels developed during the test.

Mechanical tests are being conducted in conjunction with other methods of evaluating radiation-induced shrinkage in 780-S graphite in out-of-pile testing. Graphite samples are being mechanically stressed to induce noncatastrophic cracks in the graphite. Then the extent of the degradation of the mechanical integrity will be measured as a function of the prestress.

Several methods of achieving this have been evaluated, from which

a flexure specimen has been chosen as offering the most feasible means of precracking the graphite. The degradation of mechanical integrity is being determined by measuring the changes in tensile strength.

Improved Graphites. A survey of the three major graphite companies was made to determine the status of development of graphites that may be more stable under high temperature irradiation than the "conventional" graphites (i. e., made by extrusion from Texas Lockport or Needle Coke). One vendor who has an isotropic graphite under development, would also make a grade like 780-S with Gilsonite coke at our request. Their experience has been that Gilsonite coke graphites are much more difficult to machine than graphites made from the usual cokes. Graphite grades H-315, H-315A, and H-316 are made by a new forming process and are therefore isotropic. Grade H-207-85 is made by extrusion and is isotropic because of the raw materials used.

BeO Moderator Development

The use of a BeO spine in the HTGR fuel element is being considered as a means of improving neutron moderation. The evaluation of materials problems associated with this design concept is the object of the BeO moderator development program.

BeO Graphite Compatibility. The extent and nature of the reaction between BeO and graphite at reactor conditions is important to understand because it affects the feasibility of using the BeO spine design concept. The original objectives for characterizing this reaction included:

1. Measurement of solid-state kinetics of carbide formation.
2. Determination of possible structural effects on graphite and BeO.
3. Determination of the rate-controlling mechanism.

As reported in the last quarterly report (GA-5866), the formation of the carbide was far slower than was at first anticipated (even at 2000°C in vacuo), and BeO sublimation seemed of greater significance. For this reason and because the graphite and BeO maintained their structural integrity under severe conditions, concern over this reaction has been greatly reduced.

Solid-State BeO Stability. The possible erosion and vapor transport of BeO at reactor temperatures now seems of greater importance than carbide formation. Using the oxide-graphite sandwich technique, a BeO-graphite sandwich was heated to 1950°C (uncorrected through a 1/4-in. quartz window) in a static atmosphere of atomic grade helium at 1 atm pressure. The "sublimation" rate was much greater than anticipated.

The observed rate in helium was about 1×10^{-4} mole/cm²-hr; the rate in vacuo at the same temperature previously reported was 5×10^{-4} mole/cm²-hr (see Fig. 2.12 for a plot of indicated oxide-graphite sandwich parameters).

The action of gaseous impurities is suspected. However, a typical spectroscopic analysis for atomic grade helium indicates a low impurity content (~ 0.2 ppm H₂O; 0.2-2 ppm O₂; 5-10 ppm N₂; 10-15 ppm Ne; and undetectable H₂). More likely, the graphite components (heat shields, etc.) of the furnace were not outgassed sufficiently before the helium was introduced and the specimen heated.

The presence of acetylene was detected as the graphite heat shields were removed from the furnace and exposed to room conditions. This suggested the hydrolysis of the carbide, which apparently formed elsewhere than at the BeO-graphite sandwich interface.

Capsule Irradiations

The capsule irradiation experiments on the TARGET program are intended to provide controlled tests by which coated fuel particles and graphites may be subjected to high temperature, high burnup, and high fast-flux exposures in a reactor environment. The program includes a series of instrumented irradiation capsules in which selected fuel materials are being evaluated in support of materials development and reactor design.

Capsule P7. Capsule P7 which contained a variety of multicoated UC₂ particles, was irradiated for one cycle in the GETR to a fast-neutron exposure of about 3×10^{20} nvt ($E > 0.18$ Mev) and a burnup of about 16% to 24% FIMA (fissions per initial heavy metal atom). Hot cell examination and evaluation of the capsule components have been completed.

Metallographic examination was conducted on the irradiated thermocouples and areas of the Inconel can. The high-temperature tungsten-tungsten-rhenium thermocouples contained cracks in the wires, and the tantalum sheath material was very brittle. The wires in the Chromel-Alumel thermocouples were intact, and the Inconel sheath showed only slight attack where it had been in contact with the graphite. The inner Inconel can contained a high density of what appeared to be bubbles, but showed only slight evidence of reaction with graphite. The Inconel exhaust tube, which operated at a considerably higher temperature than the can, showed a marked reaction where it was in contact with graphite.

Capsule P8. Capsule P8 contained various multicoated UC₂ and UO₂ particles irradiated during cycle No. 59 of the GETR. The estimated

Fig. 2. 12--Dimensional change of a BeO-graphite sandwich at 1950°C and 1 atm helium

fuel burnup was 15%, and the design operating fuel temperature was 1500°C.

The design temperature was attained, although the capsule operated at this temperature for only a few hours because of a leak. Because of reactor safeguard requirements, the rest of the total irradiation period of 463 hr was at a fuel temperature of 1100°C.

During hot cell disassembly and examination of the capsule components, a leak between the lead tube and gas control annulus was located in the area where the high-temperature thermocouples were brazed into the outer can cover. An attempt to locate the leak in the inner containment was unsuccessful, but it appears likely that it occurred in the braze area where the high-temperature thermocouples penetrate the inner can cover.

Capsule P9. Capsule P9 contained a variety of multicoated (Th, U)C₂ and (Th, U)O₂ particles. It was irradiated for 615 hr during GETR cycle No. 60. The basic design of the capsule was the same as that of the P8 capsule. Three of the four cells operated at the thermocouple design temperature of 1450°C; however, cell No. 3 exceeded this temperature by approximately 200°C. This temperature variation, as well as that between the P8 and P9 capsules, prompted an investigation into the conditions that might have caused the difference. After considering a number of possibilities, it appeared that the most likely cause was a difference in loading resulting from the variation of fuel particle size and coating thickness. Calculations showed that owing to variations in particle size, loadings might vary in excess of ± 15% which could, in turn, cause an estimated temperature range of 450°C at design temperature. Methods of improving the fuel sampling and loading were developed to overcome this problem.

Hot cell disassembly and examination of this capsule have been completed, except for metallography on two of the high-temperature thermocouples.

Capsule P10. Capsule P10 contains various multicoated (Th, U)C₂ and (Th, U)O₂ particles, which are expected to reach a fuel burnup of 8% to 10% in one reactor cycle. It is presently under irradiation in cycle No. 61 of the GETR.

After approximately 300 hr of operation, the capsule developed a leak in the inner containment. Results from a recently completed investigation into possible braze alloys and pre-operational heat-treatment brazes, indicate that this is very likely the point of failure. However, the capsule reached design temperatures on 100% helium, so that it continues to operate satisfactorily with the exhaust line closed. There was no major change in temperature as a result of the leak.

The capsule was discharged at the end of January.

Capsule P11. Capsule P11 contains various coated (Th, U)C₂, (Th, U)O₂, UC₂, and UO₂ particles. It was inserted at the end of January for GETR cycle No. 62.

The design of capsule P11 is the same as others in the P-series, but it is intended to have two distinct fuel temperature regions in the capsule. Two of the cells will operate at 1200°C and the other two at 1500°C. The capsule design had the benefit of the brazing studies in that all brazes were made with pure copper.

Capsule P12. The P12 capsule is scheduled for GETR cycle No. 64 beginning in April, 1965. This is a one-cycle delay from the original schedule, in order to allow time for the results of the previous capsules to be more completely analyzed. The P12 capsule will be the first multi-cycle irradiation of the present series, with a scheduled irradiation time of three cycles.

Thermal Mockup of P-Series Capsules. Instrumentation for the thermal mockup has been calibrated and is awaiting completion of the remainder of the experimental assembly.

Difficulties encountered during the assembly of the mockup have resulted in design changes that will permit easier installation. The components necessary to complete the mockup are presently being constructed. The revised schedule calls for the mockup to be operated in the near future.

Capsule G9. The G9 capsule has successfully completed four of a scheduled six cycles in the GETR and is presently in its fifth cycle. Representative graphite samples and pyrolytic carbon coatings are being irradiated at different temperatures ranging from 620° to 1075°C.

The capsule has been essentially free of malfunctions, with only minor repairs required during the period. Between cycles three and four, a small leak in the lead tube was repaired by welding, and the capsule has since operated successfully. During cycle four, an instrumentation malfunction resulted in an erroneous high-temperature alarm. The problem was the result of shorting that began to occur across a set of contacts in a switching relay. An addition of diodes to the circuit has minimized the shorting, and preventive maintenance measures are now part of the operating procedure so that further trouble with this recorder is not anticipated.

General Atomic-ORNL Cooperative Irradiation Program for TARGET

A program of irradiation tests in support of the TARGET program is being conducted at ORNL. This program includes tests for the evaluation

of (1) coated particles to high burnup at high temperature, (2) oxide fuel pellets in graphite sleeves, and (3) BeO pellets in contact with graphite. General Atomic is supplying the samples for the tests and also contributing to the design and analysis of the experiments.

Coated Particle Irradiation Tests. The sweep capsule irradiation facilities at ORNL are being used for additional high-temperature and high-burnup tests of coated particle fuels for the TARGET program.

The GAIL IV Triplex coated particles are being tested at ORNL to furnish high-burnup data for correlation with the fission-product release results being obtained at lower burnup in the GAIL IV element.

The first experiment, Capsule B9-21, was terminated after 96 hr at 1370°C owing to a mechanical failure of the capsule. Postirradiation examination revealed that the thermocouple sheath material had melted and attacked the coatings of about 20 particles. Microscopic examination of approximately 1200 other particles that had not been in contact with the molten metal showed them to be unaffected by the short irradiation.

Testing was begun on a new sample of these particles in Capsule C1-14, but this capsule failed similarly after only 0.9% burnup at 1370°C. Again, there was no evidence of irradiation damage to the coated particles. A third test of the particles is currently in progress in Capsule C1-15.

BISO (buffer-isotropic) coated oxide particles are being tested in a series of tests to determine the upper temperature limit for coated particles under irradiation. These particles are to be tested initially at 1370°C to approximately 8% burnup. If they survive this test, duplicate samples will then be tested at successively higher temperatures (1600°, 1800°C, etc.).

Capsule B9-23, the first test in this series, operated satisfactorily at 1370°C up to about 6.5% burnup. During this period, the Kr^{88} release was about 2×10^{-7} . The release suddenly increased to 3×10^{-3} and the capsule was removed. Postirradiation examination indicated that thermocouple melting with subsequent attack on the coatings by the molten metal was again the cause of failure. An examination of the coated particles revealed no coating failure from irradiation effects, however. The only observed irradiation effect was some densification of the porous oxide particles, which left a gap between the oxide particle and the porous buffer coating.

The experiment was repeated in Capsule B9-24, but this capsule failed after only 12 hr at 1370°C. Postirradiation examination has not yet been conducted on this capsule, and plans for retesting are not yet firm.

In the partial fuel recycle scheme for TARGET, coated UC_2 or UO_2 particles might be subjected to very high burnups. At present, no irradi-

ation experiments have been carried beyond 24% burnup. ORNL will therefore test the coated particles to 50% to 70% burnup in capsules in the ETR and ORR as a first investigation of the high-burnup performance of coated fuel.

Various types of advanced coated particles were selected for use in these high-burnup tests, and the samples were sent to ORNL. Since the graphite components of these capsules will receive a significant fast-flux exposure at a high temperature, samples of candidate fuel element graphites were also sent to ORNL for use in fabricating the capsule components. This test is scheduled to begin this Spring.

Oxide Pellet — Graphite Irradiation Experiment. The General Atomic-ORNL cooperative irradiation experiment is intended to assess the performance, under HTGR conditions, of oxide-graphite fuel elements made from sintered thorium-uranium oxide pellets in contact with commercial graphite.

During the past quarter, plans were finalized for the experiment, and most of the operating techniques and examination methods for the experiment were agreed upon. Only the final computation of the uranium specimen sizes and temperature gradients remains to be completed.

BeO-Graphite Irradiation Experiment. ORNL is performing a BeO-graphite irradiation experiment in support of the TARGET program. The in-core capsule is being designed to operate at 1500°C in a sweep gas of He or Ne containing 300 ppm CO. Samples of BeO and 780-S graphite were furnished by General Atomic for the experiment.

A thermal mockup has been prepared to obtain and confirm capsule design data. This experiment, designated Mark I, has been installed in the ORR core. The actual irradiation experiment, designated Mark II, will soon begin.

CHEMISTRY

Fission-product-release Studies

Kr and Xe Release of Irradiated Pyrolytic-carbon-coated UO₂ Particles from the P6 Capsule and Comparison with Unirradiated Control Particles. Steady-state fission-gas-release experiments have been completed on both the high burnup and control P6 particles. These particles consist of 150- to 250-μ diameter UO₂ particles coated with a layer of about 95 μ of isotropic pyrolytic carbon (lot 2358-117E).

The high-burnup particles were run in a new TRIGA King furnace

(No. III) because of the contamination levels in furnaces I and II. Runs were made at 1100°, 1300°, 1500°, and 1650°C, and the release data obtained are given in Table 2.10. Runs were also completed on unirradiated control particles from the P6 capsule at room temperature (recoil release) and over the range 1000° to 1700°C. These data are also shown in Table 2.10. The release rates are similar to those found for pyrolytic-carbon-coated carbide particles. A comparison of the before and after release data in Table 2.10 reveals no significant deterioration of the particles owing to irradiation.

The preirradiation data given in Table 2.10 show that the fractional release values vary little between room temperature and 1400°C. Apparently recoil release predominates in this temperature range. However, there appears to be a sharp break at 1400°C, and the release rate increases rapidly above this temperature. The effect of temperature on release data for Kr^{85m} is shown in Fig. 2.13. Apparent energies of activation for release from the particles above 1400°C, obtained from Arrhenius plots like that of Fig. 2.13, are shown in Table 2.11.

Release of Ba¹⁴⁰, Ce¹⁴⁴, and Cs¹³⁷ From Pyrolytic-carbon-coated Particles. In previous postirradiation annealing studies to determine metallic fractional release values (see GA-5866, pp.68-69), one sample of pyrolytic-carbon-coated UO₂ particles from the P6 capsule was successively annealed at 1300°, 1500°, and 1700°C to see whether lower temperature annealing would affect fission-product release at higher temperatures.

In recent fission-product release experiments on two additional samples of PyC-coated UO₂ particles from the P6 capsule, one sample was annealed at 1500° and the other at 1700°C, and the release fractions for Cs¹³⁷ and Ce¹⁴⁴ were measured. The data which are given in Table 2.12, correspond well with data found in the previous experiments, indicating that low-temperature annealing has little effect on high-temperature fractional release values.

Samples of pyrolytic-carbon-coated UC₂ particles (150 to 250μ dense UC₂, 103-μ Triplex coating, lot 2340-86E) irradiated in the P7 capsule are being studied at present. One sample has been annealed at 1700°C for approximately three days. The fractional release data for Ba¹⁴⁰, Ce¹⁴⁴, Cs¹³⁷, and Kr⁸⁵, determined by gamma spectrometry, are shown in Fig. 2.14. These data are tentative and may need correction after radiochemical analyses are completed. The data are based on total fissions calculated from radiochemical Zr⁹⁵ determinations on nonannealed samples.

It may be noted in Fig. 2.14 that the Ce¹⁴⁴ fractional release did

Table 2.10

STEADY-STATE RELEASE OF FISSION GASES FROM COATED UO_2
 PARTICLES BEFORE AND AFTER IRRADIATION IN P6 CAPSULE
 (200- μ UO_2 Particles with 95- μ Layer of Pyrolytic Carbon)

Temp. (°C)	Steady-state Fractional Release					
	Kr ^{85m}	Kr ⁸⁸	Kr ⁸⁷	Kr ⁸⁹	Xe ¹³⁸	Xe ¹³⁹
After Irradiation						
1100	1.4×10^{-5}	1.3×10^{-5}	1.2×10^{-5}	6.0×10^{-6}	5.8×10^{-6}	---
1300	1.1×10^{-5}	1.4×10^{-5}	1.2×10^{-5}	6.0×10^{-6}	3.4×10^{-6}	3.0×10^{-6}
1500	1.4×10^{-5}	1.2×10^{-5}	1.5×10^{-5}	2.2×10^{-5}	7.3×10^{-6}	1.1×10^{-5}
1650	1.7×10^{-5}	2.1×10^{-5}	2.3×10^{-5}	2.3×10^{-5}	1.5×10^{-5}	2.1×10^{-5}
Before Irradiation						
RT	2.1×10^{-6}	1.7×10^{-6}	1.5×10^{-6}	2.2×10^{-6}	1.9×10^{-6}	1.1×10^{-6}
RT	3.7×10^{-6}	6.5×10^{-7}	5.5×10^{-7}	1.4×10^{-6}	1.6×10^{-6}	5.1×10^{-6}
1000	9.1×10^{-6}	5.6×10^{-6}	6.4×10^{-6}	7.0×10^{-6}	7.5×10^{-6}	2.0×10^{-6}
1200	9.4×10^{-6}	1.0×10^{-5}	9.7×10^{-6}	5.2×10^{-6}	3.9×10^{-6}	4.8×10^{-6}
1400	8.4×10^{-6}	6.0×10^{-6}	7.2×10^{-6}	1.0×10^{-5}	6.0×10^{-6}	4.0×10^{-6}
1400	1.1×10^{-5}	1.0×10^{-5}	1.1×10^{-5}	7.0×10^{-6}	8.8×10^{-6}	$< 8 \times 10^{-6}$
1600	1.8×10^{-4}	1.5×10^{-4}	1.4×10^{-4}	1.2×10^{-4}	5.4×10^{-5}	3.7×10^{-5}
1700	6.2×10^{-4}	6.0×10^{-4}	5.5×10^{-4}	5.2×10^{-4}	3.4×10^{-4}	1.9×10^{-4}

not change after 20 hr. This behavior is unusual and unexpected, and an explanation is not at hand. It is hoped that further studies will provide an explanation for this behavior.

Fission-product Sorption and Diffusion in Graphite

Vaporization of Cesium from Graphite. During the past three months, efforts to measure the vapor pressure of cesium in equilibrium with relatively low-surface-area graphite granules have been delayed in order to study the influence of the presence of other species on the vapor pressure of cesium over finely divided (-200 mesh) HLM-85 graphite.

If the proposed model of chemisorption is correct, we would expect, in the presence of cosorbed species, a competition for sorption sites so that the graphite interaction of each species at a given loading would be reduced and its vapor pressure increased. In such a situation, the most strongly chemisorbed species would tend to be retained by the graphite at the expense of those species that do not chemisorb as strongly. In the HTGR sleeve, a host of fission-product species will be present at various concentrations, so that a measure of the vaporization of cesium

Table 2.11

APPARENT ACTIVATION ENERGIES FOR NOBLE GAS RELEASE
FROM P6 PARTICLES BEFORE IRRADIATION
(UO₂ Coated with Pyrolytic Carbon)

Isotope	Temp. Range, 1400°-1700°C (kcal/mole)
Kr ^{85m}	84
Kr ⁸⁸	98
Kr ⁸⁷	88
Kr ⁸⁹	109
Average	95 ± 8
Xe ¹³⁸	130
Xe ¹³⁹	116
Average	123 ± 7

Table 2.12

FRACTIONAL RELEASE -- UO₂-PyC

Sample	Temp. (°C)	Annealing Time (hr)	Cs ¹³⁷	Ce ¹⁴⁴
2359-117E-4	1500	24.3	0.026	0.029
2359-117E-4	1500	45.2	0.11	0.034
2359-117E-5	1700	26.5	0.30	0.080
2359-117E-5	1700	46.5	0.60	0.12

Fig. 2. 13--Preirradiation steady-state release of $\text{Kr}^{85\text{m}}$ from coated UO_2 particles (P6 capsule)

Fig. 2.14--Postirradiation annealing study of Cs, Ce, and Ba release

from graphite does not give a realistic description of what is to be expected during reactor operation when other cosorbed species are present.

In order to better define this condition, experiments have been undertaken to measure the vaporization of cesium from a graphite powder that also contains other metallic fission-product species that might be expected in the graphite sleeve of a reactor. These fission products are the rare earths Sr, Ba, and Rb, in addition to Cs. The choice of loading for the present experiments was based on estimates of the end-of-life (EOL) fission-product inventory for a fuel compact of a large HTGR. The fission-product mixture prepared contained CeO_2 to represent Ce, Y, La, Pr, and Nd; Sm_2O_3 to represent Pm, Sm, and Eu; $\text{Ba}(\text{NO}_3)_2$; and $\text{Sr}(\text{NO}_3)_2$. A mixture of about 4 mg of these materials (the approximate EOL loading of the metallic fission products which might reach the sleeves if one uses the ratio graphite in fuel compacts/total graphite in reactor = 0.36) in the end-of-life relative composition was mixed with 1 g of -200 mesh HLM-85 graphite powder, pressed into a pellet, and heated at approximately 900° to 1000°C for several hours in a vacuum furnace in order to reduce the oxides and nitrates of this modified mixture to the chemisorbed elements. The reaction was assumed to be complete when the vacuum in the system returned to normal, 2×10^{-6} mm Hg. This pellet was then broken up and aqueous CsNO_3 containing Cs^{137} was added and the sample was dried, weighed, and γ -counted.

A total of three series of Cs vapor pressure measurements have been carried out with mixtures of this type. In the first series, the initial Cs loading was 2 mg Cs/g graphite and the system was heated as high as 1000°C during the run. (This loading was well in excess of the EOL loading, but it was expected that a large fraction of the cesium would vaporize from the Knudsen cell during the reduction or conversion step.) No meaningful pressure data were obtained, however, because nearly all the cesium was lost from the cell by vaporization during the first few minutes of the run. In the next measurement an initial loading of 4 mg Cs/g graphite was used, and the cell was heated to a maximum of 800°C during the run. Even at these relatively low temperatures, about 3/4 of the cesium was vaporized from the cell, which had an orifice area of only $1.3 \times 10^{-4} \text{ cm}^2$ (diameter ≈ 6 mils).

In the most recent series of measurements, a cesium loading of 4 mg Cs/g graphite and a fission-product mixture (fissium) loading of only 1 mg fissium/g graphite was used. In addition, a Knudsen cell with an orifice of only $3 \times 10^{-5} \text{ cm}^2$ (diameter 2.5 mils) was used. Under these conditions, a cesium-fissium-graphite mixture has been produced in which the cesium pressure can be measured in the temperature range of interest (800° to 1200°C) without excessive cesium loss from the cell during the measurement. In the first three vapor pressure runs with

this mixture, the observed cesium pressures have been well in excess of the pressures observed previously in the absence of fission. It is believed that these high observed pressures are due, at least in part, to some poorly designed portions of the apparatus, and these features are presently being improved. The defects arise from the fact that cesium vapor is difficult to condense and is subject to considerable bouncing within our system, so that all the cesium found on the targets does not always arrive directly from the effusion orifice.

In order to demonstrate that the apparatus can yield correct vapor pressure results (for elements that are not as difficult to condense as cesium), a calibration run was made with silver using $\text{Ag}^{110\text{m}}$ tracer. The observed pressures compared quite well with the known vapor pressure of silver and the heat of vaporization was in acceptable agreement with the value found in the literature, indicating that temperature-dependent errors are not a serious problem in this apparatus.

Cesium Sorption in Graphite. Experimental work is continuing to obtain cesium-graphite sorption isotherms in the temperature range 800° to 1100°C and cesium partial pressures of 1×10^{-8} to 7×10^{-3} atm. All measurements have been made on HLM-85 graphite using the system described in GA-5618. A summary of the equilibrium sorption data is shown in Table 2.13. Cesium loadings expressed in mg Cs/m^2 graphite agree very well with sorption values obtained from a Knudsen cell method, as shown in Fig. 2.15.

The data obtained indicate chemisorption at concentrations below that corresponding to a monolayer of coverage of internal (BET) surface area ($\sim 1 \text{ mg Cs/m}^2 \text{ C}$). Under these conditions the cesium pressure has been found to be proportional to the cesium concentration ($\text{mg Cs/m}^2 \text{ C}$) to the n th power, where n is 3 to 6, depending on temperature. Isosteric heats of vaporization are ~ 40 to 90 kcal/mole , depending on concentration, as shown in Fig. 2.16. More complex isotherms and less strong sorption are found for concentrations corresponding to greater than a monolayer of coverage.

One run has been completed to study the effect of barium-impregnated graphite on equilibrium cesium pressure at 900°C . The results of this run indicate an increase of the cesium pressure by a factor of 3 with a barium loading of 0.55 mg Ba/g C ($0.79 \text{ mg Ba/m}^2 \text{ C}$). It is also noted that the combined barium and cesium loadings are far greater than for cesium alone in a barium-free graphite at the same cesium pressure. A similar run is now in progress at 1000°C over the same cesium pressure range.

Table 2.13
SUMMARY OF CESIUM-GRAPHITE EQUILIBRIUM-SORPTION DATA

Temperature (°C)	P _{Cs} (atm)	BET Surface Area (m ² C/g C)	Loading (mg Cs/g C)	Loading (mg Cs/m ² C)
1100	5x10 ⁻⁴	0.7	0.84	1.20
1100	9x10 ⁻⁵	0.7	0.54	0.78
1100	1.2x10 ⁻⁵	0.7	0.25	0.36
1100	8.5x10 ⁻⁷	0.7	0.093	0.13
1000	5x10 ⁻⁴	0.57	1.26	2.21
1000	9x10 ⁻⁵	0.7	0.73	1.04
1000	1x10 ⁻⁵	0.7	0.43	0.61
1000	1x10 ⁻⁶	0.7	0.24	0.34
1000	1x10 ⁻⁷	0.7	0.13	0.19
1000	1x10 ⁻⁸	0.7	0.062	0.089
900	6.5x10 ⁻³	0.5	111	222
900	2.7x10 ⁻³	0.5	51.4	103
900	1x10 ⁻³	0.57	21.1	37
900	1x10 ⁻⁴	0.7	1.01	1.44
900	1x10 ⁻⁵	0.85	0.76	0.89
900	1x10 ⁻⁶	0.85	0.50	0.59
900	1x10 ⁻⁷	0.85	0.31	0.37
900	1x10 ⁻⁸	0.85	0.23	0.27
800	6.5x10 ⁻³	0.5	188	376
800	5x10 ⁻⁴	0.57	46.3	81
800	1x10 ⁻⁴	0.57	1.13	1.98
800	9x10 ⁻⁵	0.7	1.52	2.17
800	1.2x10 ⁻⁵	0.7	1.01	1.44
800	1.10x10 ⁻⁶	0.7	0.75	1.08

Diffusion of Metallic Fission Products in Graphite Sleeves. Studies of the diffusion of metallic fission products in graphite have continued with the purpose of obtaining additional basic data that can be used in the prediction of fission-product release from advanced HTGR fuel elements.

A final coring of the sleeve for cesium permeation experiment No. 35 (848 hr at 500°C) was made in order to establish whether a diffusion-front type of concentration gradient for cesium existed at the end of the experiment or whether the concentration profile was relatively flat. The former would indicate a transient condition, while the latter would indicate that a steady-state condition had been reached. The profile obtained is somewhat inconclusive in that the two outside corings

Fig. 2.15--Cesium sorption isotherms in HLM-85 graphite

Fig. 2.16--Heats of vaporization of cesium in HLM-85 graphite

show quite high cesium concentration, while the remainder of the graphite samples indicate a flat profile. It is felt that contamination is one reasonable explanation for this, since the compacts and sleeve are assembled and disassembled quite often, which could lead to contamination of the two surfaces.

Thus it is concluded that a steady-state condition was reached. The experiment has also shown that a backflow purge of helium through the sleeve has little effect on the movement of cesium through the protective graphite sleeve.

Barium permeation experiment No. 36 has run 8-hr at 1200°C. A coring was made of the sleeve and a diffusion coefficient $D \approx 8.3 \times 10^{-8} \text{ cm}^2/\text{sec}$ was obtained. An activation energy of 69 kcal/mole for barium diffusion was calculated on the basis that past experiments carried out at 1000°C gave values of $D \approx 2 \times 10^{-9} \text{ cm}^2/\text{sec}$. This value agrees reasonably well with the value given by Bryant, *et al.*,* who reported a 70 to 75 kcal/mole activation energy for the diffusion of Ba^{140} in graphite.

Cerium permeation experiment No. 38 is being carried out to determine the movement of Ce^{144} through graphite. Compact loading is 1.91 mg Ce/g C. This experiment was initiated primarily to determine if it is possible for cerium to escape the compact by simple diffusion in quantities large enough to account for the cerium found in the GAIL loop (for GAIL III-B end of Cycle 52, releases were Ce^{141} , 0.03%, and Ce^{144} , 0.12%).

Data on the four runs completed to date are shown in Table 2.14. The water contamination has been held at <1 ppm, and CO contamination has been held at 500 ppb.

Corings have indicated movement of cerium to the inside surface of the sleeve, but the depth of penetration is not yet sufficient to obtain accurate measurements of a diffusion coefficient. Longer runs are planned. However, it can be estimated as a limit $\sim D = <1 \times 10^{-9} \text{ cm}^2/\text{sec}$ at 1500°C.

Assuming cerium has an activation energy of ~ 70 kcal/mole, the diffusion coefficient at 1000°C would be $< 5 \times 10^{-13} \text{ cm}^2/\text{sec}$. Tentatively then, it can be said that the quantity of cerium found in the GAIL loop measurements is considerably more than could have gotten there by diffusion.

*E. A. Bryant, *et al.*, "Rates and Mechanisms of the Loss of Fission Products from Uranium-Graphite Fuel Materials," Nucl. Sci. and Eng., Vol. 15, 1963, pp. 288-95.

Table 2.14

CERIUM DIFFUSION THROUGH GRAPHITE

Run No.	ΔT Hours	ΣT Hours	Total Ce in Cold Finger (μg)	Total Ce in Sleeve (μg)
1	64.6	64.6	0	0
2	94.2	158.8	0	161
3	47.2	206.	0	171
4	48.5	254.7	1.1×10^{-1}	228.5

The most likely mechanism for cerium escape in the quantity found would be by the mechanical attrition of the broken fuel particles on the outer surface of the compacts. Another possibility is that the quantity of cerium is due to fuel material that has escaped the compacts on previous experiments and moves around the loop as dust.

Release of Metallic Fission Products from HTGR Fuel Elements

Up to the present, estimates of fission-product release from HTGR fuel elements have been made assuming noble-gas-like behavior of the fission products. A noble gas model should be satisfactory for nonmetallic fission products as well as for Kr and Xe nuclides. On the other hand, metallic fission products are strongly sorbed by graphite and do not diffuse appreciably through graphite pores as gas molecules. Instead, they diffuse through the graphite by surface (along internal pore surfaces) and by volume (along grain boundaries) diffusion. Accordingly, equations and input data for calculating the release of metallic fission products must be different than those used for fission products behaving like noble gases.

A start has been made on developing the mathematical treatment of two cases:

1. Release limited by the decay of metallic fission products during their diffusion through the unfueled portion of graphite fuel elements.
2. Release limited by evaporation from the fuel elements.

The first case applies to fission products with a half life that is small compared to the time required for diffusion through the unfueled portion of the fuel element. Then with ($\lambda \tau \gg 1$) the fractional release (taking the geometry to approximate that of an infinite slab and the concentration at the boundary to be zero) is given by

$$F(t) \lesssim \frac{A}{V_F} \left(\frac{D}{L} \right) \frac{1}{\lambda(R + \lambda)} \left[R \frac{\pi \sqrt{\lambda \tau}}{\sinh(\pi \sqrt{\lambda \tau})} - \frac{2}{\tau} \sum_{n=1}^{\infty} (-1)^{n+1} n^2 \right. \\ \left. \left\{ \frac{R}{(n^2/\tau + \lambda)} \exp[-(n^2/\tau + \lambda)t] + \frac{(R + \lambda) \exp(-\lambda t)}{(n^2/\tau)} \left[1 - \exp[-(n^2/\tau)t] \right] \right. \right. \\ \left. \left. - \frac{\lambda \exp[-(R + \lambda)t]}{(n^2/\tau - R)} \left[1 - \exp[-(n^2/\tau - R)t] \right] \right\} \right],$$

where t = time, sec,

A = external surface area of fuel element, cm^2 ,

V_F = volume of fueled region of fuel element, cm^3 ,

L = thickness of unfueled (graphite) region of fuel element, cm,

D = diffusion coefficient of given fission-product element, cm^2/sec ,

λ = decay constant of the isotope of interest of the given fission-product element, sec^{-1} ,

τ = characteristic diffusion time, $\tau = L^2/\pi^2 D$, sec, and

R = the release constant of the given fission-product element characteristic of coated particle fuel, sec^{-1} .

R is determined by postirradiation annealing experiments and is given by

$$R = \frac{\ln\left(\frac{1}{1-\mathcal{J}}\right)}{\Delta t} \sim \frac{\mathcal{J}}{\Delta t},$$

where $\mathcal{J}$ is the fractional release upon annealing the particles for a time, Δt , at a given temperature. Empirically, it is found that

$$\ln R = \ln R_0 - \frac{Q}{R} \left(\frac{10^3}{T} \right),$$

where R_0 = a constant,

Q = activation energy for release, kcal/mole,

R = gas constant, cal/mole- $^{\circ}\text{K}$, and

T = temperature, $^{\circ}\text{K}$.

A program, FREVAL, has been written for computing the fractional release values and is now being debugged. The terms of the infinite series become negligible when $t \gg 1/\lambda$; then fractional release is given by

$$F = \frac{A}{V_F} \left(\frac{D}{L} \right) \frac{R}{\lambda(R + \lambda)} \frac{\pi \sqrt{\lambda \tau}}{\sinh \sqrt{\lambda \tau}}.$$

Application of this equation to Ba^{140} and Ce^{144} indicates $F \lesssim 1 \times 10^{-5}$ and $F \lesssim 3 \times 10^{-5}$, respectively, at 900°C ($D = 1 \times 10^{-8}$ and 4×10^{-10} , respectively, with $R = 10^{-2}$).

Release may be evaporation limited in the case of long-lived metallic fission products where $\lambda\tau \ll 1$, and also where the diffusion time, τ , is small compared to the time of interest, t . If evaporation does not limit the release of a long-lived fission product, then the release constant R and the decay constant λ are the only determining factors. The release fraction, $F(t)$, for long-lived isotopes is given by

$$F(t) \left[B_1 t + \sum_{n=2}^N \frac{B_n}{\lambda_n} (1 - e^{-\lambda_n t}) \right] = B_1 \left[t - \frac{1}{R} (1 - e^{-Rt}) \right] + \sum_{n=2}^N B_n \left(\frac{R}{R + \lambda_n} \right) \left[\frac{1}{\lambda_n} (1 - e^{-\lambda_n t}) - \frac{e^{-\lambda_n t}}{R} (1 - e^{-Rt}) \right] - M_c C(t),$$

where

t = time, sec,

B_1 = production rate of the stable isotopes of a given fission-product element, moles/sec,

$B_n \dots, N$ = production rates of radioactive isotopes, moles/sec,

$\lambda_n \dots, N$ = decay constants of radioactive isotopes, sec^{-1} ,

R = release constant, given fission-product element, sec^{-1} ,

M_c = mass of graphite in a fuel element, g, and

$C(t)$ = effective average concentration, moles/g, of a given fission-product element in the graphite at time, t .

$C(t)$ is determined by iteration of a nonlinear differential equation that incorporates isotherm (partial pressure versus concentration) data for the given fission-product element sorbed in graphite. This is computed by the FREVAP program. Preliminary data indicate that Cs^{137} release is not strongly affected by sorption (i. e., evaporation control is weak, especially for $t > 1$ to 2 years), whereas Sr^{90} release appears to be substantially affected.

Reaction Between Graphite and Oxides

Since in large HTGR's BeO may be used as a moderator material in contact with graphite, the chemical compatibility of BeO and graphite at high temperatures is of interest. The rate of reaction between BeO and graphite to form CO and Be_2C is presently being measured by determining the CO evolution rate from mixed BeO and graphite powders held at temperatures from 1500° to 1900°C . The apparatus is composed of a mini-King (graphite tube) furnace and a gas chromatograph. Some preliminary experiments have yielded interesting data. A

mixture of 0.127 g of BeO and 0.50 g of graphite was heated in a closed graphite crucible to 1500°C for ten days in helium flowing at 130 std cm³/min at 28 psig. The CO evolved from this system was measured frequently. The initial CO evolution was predictably high owing to the outgassing of the crucible, but within an hour the true reaction rate was observable. The CO evolution rate decreased steadily from an initial 3×10^{-7} moles CO/min to a level so low (in four days) as to be practically indistinguishable from the background CO (5×10^{-8} moles CO/min) that continuously evolves from the graphite furnace tube.

Much time and effort has been spent to reduce the CO background in the graphite tube furnace apparatus to lower levels, but without success. Apparently CO will continue to evolve for an indefinite time because of the outgassing of parts of the graphite resistance tube at intermediate temperatures. It is therefore felt that revisions in experimental apparatus are necessary. The first step will be to utilize a tantalum heater tube with a ceramic or TaC liner to limit CO uptake. It is felt that this simple and inexpensive revision of existing equipment will solve the problem.

In the above experiment, the total amount of BeO that reacted in ten days was about 0.012 g, or only 10% of the initial amount. It is believed that the reaction rate at the end of ten days was below 2×10^{-8} moles of CO/min. Steps are presently being taken to determine the reaction rate accurately. The low reaction rate may be associated with a buildup of Be₂C around each BeO particle, which acts as a diffusion barrier for the reacting species. Because this occurs in a system that affords fairly intimate contact (i. e., mixed powders), one speculates that for the case of solid BeO contacting solid graphite (as in a BeO spine-graphite fuel element configuration) when exposed surfaces are minimized, the amount of BeO reacted may be quite small.

HELIUM PURIFICATION SYSTEM

Chemistry of the Helium Purification System

Krypton-Xenon Adsorption on Molecular Sieves. The determination of xenon and krypton isotherms on molecular sieves at room temperature and at a total helium pressure of 450 psig is about complete. The experimental procedure (see GA-5366, p. 118) was to circulate known concentrations of tagged Xe and Kr in helium through a molecular sieve bed held at a desired temperature. The work was done to obtain information for use in predicting before regeneration the gaseous activity levels in dehydrator beds in the TARGET reactor. It is expected that the beds will become contaminated with sorbed Xe and Kr fission products.

The experimental results include four isotherms, Xe and Kr on 4A and 5A molecular sieves, along with adsorption-rate curves for each system. Figure 2.17 shows the isothermal data and illustrates quite obviously the differences in adsorptive capacity for noble gases on 4A and 5A sieves. Figure 2.18 illustrates the rates of adsorption for these systems.

It is seen that whereas Kr and Xe on 5A molecular sieves equilibrate within an hour or two, the 4A variety, having a much smaller pore structure, offers sufficient diffusional resistance to delay equilibrium for 30 or 40 hr. Indeed, some of the experiments with 4A sieves ran for 100 hr, at which point the Xe or Kr pressure was still decreasing (though almost negligibly), indicating further pickup by the adsorbent.

It is interesting to observe that the adsorption behavior for Xe and Kr on the 5A variety is exactly what one might expect; that is, Xe is adsorbed the most because of its higher adsorption coefficient. This phenomenon is quite predictable owing to the larger size, the greater electron cloud, and thus the greater polarizability of the xenon atom as compared to the krypton atom.

The situation is quite the reverse, however, for the 4A material. Apparently, the smaller-pore-size 4A molecular sieve allows more of the smaller Kr atoms ($\sim 3.7 \text{ \AA}$) to enter than the larger Xe species ($\sim 4.0 \text{ \AA}$).

It is concluded from this work that the 4A molecular sieve variety has a much lower capacity for Kr and Xe than the 5A variety. The 4A variety is therefore preferable for use in dehydrator beds.

Outgassing of Sleeve Graphite. A Cahn recording micro-electro-balance recently set up has proved to be useful in measuring minute weight changes in gram-size samples. Weight losses in selected graphites due to outgassing are presently being studied.

Two specimens of GLIS-12 graphite (He permeability = $\sim 10^{-3}$) were heated in vacuo (1 to 2 μ pressure) in a quartz vessel for 2 to 3 days. The maximum temperature (950°C) was attained by heating in discrete stages. The weight loss at each temperature increment was continuously recorded. Table 2.15 lists the conditions and results of these preliminary experiments.

These results show that more than half of the graphite outgassing can be accomplished at comparatively low temperatures (100° to 300°C), which indicates that adsorbed water may be a major contributor. Another interesting feature of these data is that essentially 99% of the weight loss occurs at as low a temperature as 700°C.

Fig. 2.17--Xenon and krypton adsorption isotherms

Fig. 2.18--Xenon or krypton adsorption versus time for 4A and 5A molecular sieves and 1/16-in. pellets

Table 2.15
WEIGHT LOSSES DUE TO OUTGASSING

Temp. (°C)	Time	Weight Loss (mg)
Experiment No. 1 (1.1306 g GLIS-12)		
100	90 min	0.34
180	200 min	0.46
300	15 hr	0.54
410	80 min	0.56
510	300 min	0.64
605	120 min	0.66
700	200 min	0.70
950	15 hr	0.70
Experiment No. 2 (0.6694 g GLIS-12)		
250	18 hr	0.30
360	8 hr	0.40
435	62 hr	0.54
550	8 hr	0.54
950	18 hr	0.59

Evaluation of GAIL No. 1 Delay Bed. Reported in the last quarterly progress report, GA-5866, were the results of an experiment designed to measure the axial distribution of short-lived xenon and krypton in the GAIL No. 1 delay bed. The experiment involved purging fission product Xe and Kr generated in the TRIGA reactor through the activated-carbon-filled GAIL No. 1 trap under flow and temperature conditions similar to those in the GAIL (Vallecitos) facility. Later sectioning and radiochemical examination of the activated carbon was performed.

The results of the preliminary analysis (see GA-5866) showed no spurious distribution of Cs^{137} arising from 3.9-min Xe^{137} or of La^{140} arising from 16-sec Xe^{140} . These specimens were subjected to radiochemical separation and then radiocounting of Sr^{90} , Sr^{89} , and La^{140} . The results of the procedure are shown in Fig. 2.19.

It is seen that the analytical results of the Kr^{89} daughter agree quite well with the theoretical curve (diagonal line). The results for Sr^{90} , however, seem to agree with the theoretical line only for about the first

Fig. 2. 19--Distribution of short-lived noble gases in the
 GAIL No. 1 trap TRIGA experiment obtained by
 radiochemical analysis

400 g of carbon, and for La^{140} the agreement holds for the first 100 g of the delay bed, after which, these two isotope concentrations appear to flatten out considerably. The theoretical curves were obtained as described in GA-5866.

The results obtained thus far only add to the uncertainty, because it is not certain that they represent the true physical distribution in the trap. There are several reasons why the analytical results may be in error.

1. The La^{140} distribution derived from radiochemical separation does not agree with that derived from the direct gamma-counting procedure (see GA-5866).
2. There is the possibility of cross contamination of carbon samples, such as spray or dust. First, the radiochemical technique involved several problems which resulted in the utilization of complicated, lengthy, and untried procedures. Second, an extremely minute contamination of the low-level carbon samples could account for these spurious results (i. e., only 38 cpm La^{140} were found in the 400-g bucket No. 6 sample, whereas the 70-g bucket No. 1 sample contained 19,000 cpm).
3. The Sr, Y, and Ba carriers used in the separation procedure may have been contaminated. The Y and Ba carriers were recently checked and found to be relatively free of contamination, but the Sr carrier had been used up and therefore could not be checked.
4. The coconut charcoal may have been contaminated by fission products. This is a distinct possibility, especially if the coconut shells used in the charcoal manufacture were obtained from the Pacific area, where fission-product contamination (Sr^{90}) of vegetation from nuclear testing is a possibility. A large sample of the identical carbon is presently being ashed and prepared for beta counting to check this.

It is possible, however, that the observed distributions are real. One explanation for such an axial distribution is aerosol formation or carbon dusting in the trap downstream from the millipore filter.

The only way to eliminate all of these doubts and possibilities of discrepancy is to repeat the experiment, making sure that adequate precautions and controls have been taken.

Fission-product Deposition and Trapping Studies

Cesium Plateout on Steel. Additional data have been obtained on the plateout of cesium on Type 304 stainless and carbon steel with the termination of five runs during the quarter. The measurements were made using an isopiestic (static) system similar to that shown in Fig. E. 8 of GA-5366, p. 126. Metallic cesium tagged with Cs^{137} was used as the source of cesium vapor. A summary of the data and the conditions for each run are shown in Table 2. 16.

The level of sorption of cesium is quite dependent on the degree of surface oxidation of the steel as shown by the values for two different bakeout periods at 1020°C . Exposure of carbon steel to 1% H_2 in He at 400°C (750°F) was not effective in reducing surface oxides on the steel. In fact, the plateout level was higher than would be expected for an untreated steel specimen. Obtaining equilibrium Cs plateout data on carbon steel is difficult owing to the long times necessitated by the low specimen temperatures.

An attempt was made to correlate the oxide film thickness on steel with the degree of cesium plateout. Five 1-in. diameter by 0.002-in. - thick foils (both stainless steel and carbon steel) that had been outgassed to varying degrees were subjected to oxygen analysis. This analysis was made by bombarding the foils with He^3 ions and then measuring the amount of F^{18} produced from the reaction $0^{16}(\text{He}^3, \text{H}^1)\text{F}^{18}$. All samples showed ~ 100 μg oxygen, which approximates one-half of the total oxygen present in the steel, since the He^3 ions would penetrate ~ 0.001 in. into the 0.002-in. -thick foil. If all oxygen were assumed to be present as surface oxygen film, this would correspond to ~ 1800 monolayers of oxygen. This value appears unreasonable for two reasons: (1) one would expect variation in the oxide film thickness with the type of steel, and (2) the value is too high to account for observed plateout behavior. A further investigation of this technique of analysis is being made.

A loop has been assembled to attempt to measure the cesium plateout on carbon steel from a helium stream. In this experiment cesium will be furnished by a $\text{Cs}_2\text{O-C}$ source, rather than by metallic cesium which proved unsuccessful in previous loop experiments. Two 10-in. - long, 1/4-in. ID A-126B carbon-steel tubes are included in the loop. The first tube was vacuum baked at 1000°C for 24 hr to provide an oxide-free surface. The second tube was only degreased and the surface will be considered "as received". It is hoped that the cesium plateout on the two different steel surfaces can thus be evaluated.

The loop also contains a "graphite getter" forward of the cesium source, held at 900°C to reduce O_2 and H_2O impurities in the helium

Table 2.16

PLATEOUT OF CESIUM ON TYPE 304 STAINLESS AND CARBON STEELS

Specimen	Temp. (°F)	Mono- layers Cs*	Atoms Cs/cm ²	Mono- layers Cs*	Atoms Cs/cm ²	Mono- layers Cs*	Atoms Cs/cm ²	Mono- layers Cs*	Atoms Cs/cm ²	Mono- layers Cs*	Atoms Cs/cm ²
Type SS vacuum baked 90 hr at 1020°C	1350	0.83	3.3x10 ¹⁴	0.70	2.7x10 ¹⁴	---	---	0.49	2.0x10 ¹⁴	---	---
Type SS vacuum baked 22 hr at 1020°C	1350 1100 840	1.93 --- ---	7.7x10 ¹⁴ --- ---	1.75 1.89 2.10	7.0x10 ¹⁴ 7.6x10 ¹⁴ 8.4x10 ¹⁴	1.63 1.77 1.95	6.5x10 ¹⁴ 7.1x10 ¹⁴ 7.8x10 ¹⁴	1.47 1.65 1.85	5.9x10 ¹⁴ 6.6x10 ¹⁴ 7.4x10 ¹⁴	1.38 --- ---	5.5x10 ¹⁴ --- ---
Carbon steel vacuum baked 24 hr at 800°C	800 500	--- ---	--- ---	22 ---	8.8x10 ¹⁵ ---	18 35	7.2x10 ¹⁵ 1.4x10 ¹⁶	14 24	5.6x10 ¹⁵ 9.6x10 ¹⁵	11 ---	4.4x10 ¹⁵ ---
Carbon steel vacuum baked 16 hr at 400°C	800	---	---	---	---	---	---	35	1.4x10 ¹⁶	---	---
Carbon steel vacuum baked for 16 hr at 400°C. Reac- ted with 1% H ₂ in He for 24 hr at 400°C	650	---	---	420	1.7x10 ¹⁷	285	1.1x10 ¹⁷	---	---	---	---

*A monolayer of cesium is arbitrarily defined as 4×10^{14} atoms/cm² of surface.

stream. Also, it will be of interest to see whether graphite dust is carried throughout the loop by the gas recirculating at 4 liters/min. The loop experiment was terminated after 30 days running time when it became apparent that CO was building up in the gas stream. The CO from the reaction $\text{Cs}_2\text{O} + \text{C} \rightarrow 2\text{Cs} + \text{CO}$ depresses the partial pressure of cesium in the system and eliminates the possibility of maintaining a constant cesium pressure. The loop is being modified to remove CO generated in this manner.

GAIL Plateout Analyses. Fission-product plateout in the HTGR primary and purge systems is being studied by analyzing fission-product deposits in the General Atomic In-Pile Loop (GAIL).

Specimens, principally cold fingers, are periodically removed from the GAIL. Cold fingers are 0.5-in. diameter stainless steel probes that protrude approximately 15 in. into the main coolant piping at various positions in the loop. The first half of each cold finger operates at the local gas temperature. The latter half is water cooled, except when it contains a diffusion tube (described later in this section). Two graphite sleeves were built into the Cycle 58 cold fingers. Later cold fingers have been constructed entirely of 321 stainless steel. During the GAIL III-B irradiation, there were rarely differences in plateout levels on graphite and stainless steel.

Upon receipt at General Atomic, each probe is sectioned and the sections counted at reduced geometry. A 3-in. by 3-in. NaI(Tl) canned crystal and multichannel analyzer are used. Selected sections are then dissolved or etched, and the resultant solutions are subjected to radiochemical analysis. Surface concentrations of various nuclides are reported in $\mu\text{c}/\text{cm}^2$ and atoms/cm^2 .

Cold fingers have been regularly exposed during the GAIL IV irradiation. Locations of the various cold fingers are given in Table 2.17. Cold fingers from positions 1, 3, and 5 for Cycle 58 and cold fingers for all positions for Cycles 59 and 60 were received for analysis.

Plateout activity data for Cs^{134} and Cs^{137} have been determined for positions 1, 3, and 5 for all three cycles. Cycle 58 cold fingers, from positions 1, 3, and 5, have been analyzed radiochemically for Sr^{89} , Sr^{90} , $\text{Ag}^{110\text{m}}$, Ag^{111} , and Ba^{140} . The plateout activities were corrected to the end of the cycle. Values for Cs^{134} and Cs^{137} determined radiochemically now agree well with those determined by gamma spectrometry. Earlier disagreements were due to errors in standards. Analyses for nuclides other than Cs were difficult because of low levels of activity. Cold fingers at positions 2 and 7 were utilized for diffusion-tube experiments and the results are discussed in another section.

Table 2.17

POSITION OF COLD FINGERS

<u>Position No.</u>	<u>Position</u>
1	Upstream from regenerative heat exchanger (not used as such) and main-loop cooler.
2	Between regenerative heat exchanger and main loop cooler. Contained diffusion tube for Cycles 59 and 60.
3	Between first and second banks of main loop cooler.
5	Downstream from main loop cooler
7	Upstream from main loop heater. Downstream from main loop filter and circulator. Contained diffusion tubes for Cycles 59 and 60.

Plateout activities for Cs^{134} were around 0.1 to $0.4 \mu\text{c}/\text{cm}^2$ (10^{11} to 10^{12} atoms/ cm^2) and those for Cs^{137} were around 1 to $5 \mu\text{c}/\text{cm}^2$ (10^{13} to 10^{14} atoms/ cm^2). Ratios of Cs^{134} and Cs^{137} deposited downstream from the main loop cooler to Cs^{134} and Cs^{137} deposited upstream are compared in Table 2.18. The ratios appear to be increasing slowly with time, indicating that cesium is being gradually spread through the loop.

Radiochemical analyses of Cycle 58 cold fingers yielded the following results. The nuclides I^{131} and $\text{Te}^{129\text{m}}$ were not detected. Limits of detection for $\text{Te}^{129\text{m}}$ ranged from $3 \times 10^{-4} \mu\text{c}/\text{cm}^2$ (4×10^7 atoms/ cm^2) to $5 \times 10^{-4} \mu\text{c}/\text{cm}^2$ (7×10^7 atoms/ cm^2). Plateout activities for Sr^{89} were around 10^{-2} to $10^{-3} \mu\text{c}/\text{cm}^2$ (10^8 to 10^9 atoms/ cm^2), and those for Sr^{90} were around 10^{-2} to $10^{-3} \mu\text{c}/\text{cm}^2$ (10^{11} to 10^{12} atoms/ cm^2). The activity values for Sr^{89} are somewhat uncertain, since they were determined by subtracting relatively large $\text{Sr}^{90}(\text{Y}^{90})$ activities from total $\text{Sr}^{89} + \text{Sr}^{90}$ activities. Results for Ce^{141} and Ce^{144} are too questionable to be reported, and a new procedure is being developed for these isotopes. The nuclide Ba^{140} was barely detected; $\text{Ag}^{110\text{m}}$ was present in small amounts; and the nuclides Ru^{103} and Ru^{106} were not determined because of radiochemical procedural problems.

The nuclides $\text{Ag}^{110\text{m}}$ and Ag^{111} are being dropped as nuclides of interest (as was Zr^{95}) and will only be spot checked in the future. $\text{Ag}^{110\text{m}}$ always appears in small amounts in GAIL and usually behaves like Ta^{182} . This suggests that stray silver in the loop is being irradiated and circulated

Table 2.18

CESIUM DOWNSTREAM RELATIVE TO UPSTREAM OF GAIL MAIN-LOOP
COOLER AS A FUNCTION OF TIME

Fuel Element	GETR Cycle	Purge Conditions	Average Ratios ^a (atoms/cm ²)					
			Gas Temp. Region (Sections -01 thru -03)		Water-cooled Region (Sections -05 thru -08)		Over-all Average (Sections -01 thru -08)	
			Cs ¹³⁴	Cs ¹³⁷	Cs ¹³⁴	Cs ¹³⁷	Cs ¹³⁴	Cs ¹³⁷
GAIL III-B	49-51	Normal and bypass	0.036	0.038	0.015	0.018	0.028	0.030
	52	Bypass	0.075	0.081	0.015	0.014	0.045	0.048
GAIL IV	58	Normal	0.62	0.72	0.26	0.28	0.36	0.39
	58	Normal	0.71	0.70	0.29	0.29	0.47	0.47
	59	Bypass	0.69	0.70	0.41	0.37	0.49	0.51
	60	No purge	1.0 ^c	1.5 ^c	0.39	0.46	0.46	0.55

^aAverage ratios in atoms/cm² are of Ce on downstream cold finger (position 5) to Ce on upstream cold finger (position 1).

^bAs determined from radiochemical data; other ratios calculated from gamma-spectrometry data.

^cShould be confirmed by radiochemical analysis.

as dust. Analyses of dust in the GAIL "mud-leg" verified this contention. Silver was positively identified by emission spectroscopy.

GAIL Dust Analyses. Fission products in the GAIL primary and purge systems are associated to some extent with particulate matter. Work is under way to characterize these particulates, and the results are promising despite difficulties owing to operating temperatures, pressures, and safety requirements of the loop.

A related question, that of the amount and nature of radioactive particulate matter in the GAIL containment (cubicle) was more easily answered. The particulates were collected by drawing GAIL cubicle air through an impaction device and various filters. Five samples were taken, four during the GETR Cycle 59 shutdown and one during operation of the loop. All showed the cubicle air to be nearly free from radioactive particles. Activity levels were too low for gamma spectrometry, autoradiography, or particle analysis. The levels were comparable to those found in the general environment due to fallout.

Details and results of the first four samples are given in Table 2.19. During the Cycle 59 shutdown, while the samples were being taken, workmen were entering and leaving the cubicle, hammering on pipes, welding, and performing other activities that tended to stir up dust. This may account for the slightly higher particulate activity levels than were detected during the operation of the loop with the cubicle closed. Microscopic examination of the glass fiber filters showed that considerable dust was picked up, but very little of the dust collected was radioactive.

A final sample of GAIL cubicle air was taken during the last eight hours of Cycle 59. The cubicle was closed and the reactor and loop were operating. Particles were collected with a UNICO Cascade Impactor, and the impactor slides and filter were β -counted. Results are reported in Table 2.20.

Laboratory Dust Analyses. Plateout analyses have indicated that fission products in the GAIL system are associated to some extent with particulate matter. Accordingly, a laboratory program was instituted to evaluate methods for collecting and analyzing radioactive dust particles.

In the laboratory work, the air in a glove box employed for sectioning GAIL plateout specimens was used as a source of airborne particulates. Three samplings have been taken of the glove-box air. Two of the samplings were collected on filters and the results are described in GA-5866. A UNICO Cascade Impactor, which fractionates collected particles into five groups according to mass, was used in the third sampling. Air was drawn from the glove box through the impactor at a rate of 16.8 liters/min

Table 2.19
PARTICULATE ACTIVITY IN GAIL CUBICLE AIR
(Samples Taken During GETR Shutdown Preceding Cycle 59)

Sample No.	Type of Particle Collector	Collection Time	Rate of Sampling (ft ³ /hr air, 1 atm, 70°F)	Location of Sampling Head	Appearance of Filter or Impactor Slide	Net Count Rate in Low Background β -counter (cpm)	Estimated Particulate β -Activity in Cubicle Air ($\mu\text{mc}/\text{m}^3$) ^a
GC-1	UNICO Cascade Impactor with Gelman Type E glass fiber filter as final stage	20 min	35	Hanging near ceiling in center of lower section of cubicle	Very little visible dust on any slide or on filter except some orange agglomerates on first slide (perhaps rust or orange primer)	low	---
GC-2	1.2- μ Metrical filter	20 min	50	Hanging near ceiling in center of lower section of cubicle	No visible dust (completely white)	0 $\pm$ 0.8	---
GC-3	Gelman Type E glass fiber filter (claimed to be 98+% efficient for particles as small as 0.05 μ in diam)	6 hr	50	Hanging 9 in. from floor	Discolored (light brownish-gray)	66.0 $\pm$ 4.3	5.0 $\pm$ 0.3
GC-4	1.2- μ Metrical filter	6 hr	50	On floor	Discolored (light brownish-gray).	28.7 $\pm$ 1.7	2.2 $\pm$ 0.1

^aBased on efficiencies for the Cl^{36} 0.71 mev β^- .

(25°C, 1 atm) for 2.5 hr. During part of that period, a diamond wheel was used to make two cuts in a GAIL specimen, and the glove box was cleaned and brushed to raise dust.

Table 2.20

PARTICULATE ACTIVITY IN GAIL CUBICLE AIR
8-HOUR SAMPLE WHILE LOOP OPERATING
CYCLE 59

Impactor Stage ^a	Average Particle Size, MMD ^b (μ)	Net β ⁻ Count Rate, 49% Geometry (cpm)	Estimated Particulate β ⁻ Activity Cubicle Air ^c (μμc/m ³)
1	Large particles	2.6±1.2	0.30±0.14
2	4.1	2.6±1.2	0.30±0.14
3	2.1	2.6±1.2	0.30±0.14
4	1.1	0.3±0.9	0.03±0.09
5	Fines down to 0.05μ diam	2.3±1.2	0.26±0.14

^aParticulates were collected with a UNICO Cascade Impactor through which cubicle air was drawn at 35 ft³/hr. All stages were lightly greased microscopic slides except stage 5, which was a Gelman Type E glass fiber filter.

^bAssumes particles of specific gravity = 5.24 (Fe₂O₃). The mass median diameter, MMD, must be related to average particle diameter experimentally.

^cThese levels are comparable to those found in the general environment due to fallout.

The impactor contained five collection stages, the first four of which were lightly greased microscope slides. A Gelman Type E glass fiber filter was chosen for the final stage. After the exposure, each stage was examined microscopically at 40X and then gamma counted (integrally above 0.020 mev). The distribution of activity as a function of mass median diameter (MMD) is given in Table 2.21. There may have been some losses from Stage 2. A particle specific gravity of 7.86 (Fe) was assumed in determining the MMD for each stage. Attempts to calculate average particle diameters from the MMD's produced unreasonable values, apparently because the activity distribution did not serve as a guide to the mass distribution, which had not been measured directly.

The impactor was sent to GETR for sampling GAIL cubicle air, and other particle counting and sizing instruments are now being examined.

Table 2. 21

ACTIVITY DISTRIBUTION FOR VARIOUS DUST PARTICLE SIZES

<u>Particle Size</u>	<u>Activity (%)</u>
Stage 1, large particles	79.1
Stage 2, 3.5 μ MMD.	11.3
Stage 3, 1.7 μ MMD.	8.1
Stage 4, 0.9 μ MMD.	0.4
Stage 5, fines down to 0.05 μ diameter	1.1
Total	100.0

GAIL Diffusion-tube Experiments. In high-temperature, gas-cooled reactors, the amount and chemical nature of the condensable fission products in the coolant stream must be predicted in order to correctly design the reactor shielding. To obtain some of the needed information, diffusion-tube experiments have been undertaken in the GAIL system and in the laboratory. A diffusion tube is simply a small diameter tube that bypasses a portion of the coolant stream containing the fission products of interest. If the bypassed flow is small enough and the tube wall sufficiently reactive, most of the fission products will have diffused to and reacted with the inside surface before the flow passes completely through.

The diffusion and deposition of a species from a gas stream in a cylinder is described by the following equation used by Browning and Ackley:*

$$n(Z) = \left(\frac{N_o D}{Q} \right) (9.4106 e^{-11.489 DZ/Q} + 6.8309 e^{-70.06 DZ/Q} + 5.819 e^{-179.07 DZ/Q})$$

*W. E. Browning, Jr., and R. D. Ackley, "Diffusional Characterization of Millimicron Radioactive Aerosols and their Removal from Reactor Gases," in Nuclear Safety Program Semiannual Progress Report for Period Ending December 31, 1962, ORNL-3401, Oak Ridge National Laboratory, p. 44.

where $n(Z)$ = number of particles per unit length, cm^{-1} ,
 N_o = integrated number of particles entering the tube,
 D = diffusion coefficient, cm^2/sec ,
 Q = volumetric flow rate, cm^3/sec ,
 Z = distance from tube entrance, cm .

The equation assumes that laminar flow exists, that diffusion in the gas phase is the dominant mechanism for transport of the particles to the wall from the gas stream, and that the particles stick on hitting the walls of the cylinder.

Normalizing $n(Z) = n(0)$ at $Z = 0$, one obtains:

$$\frac{n(Z)}{n(0)} = 0.4266 e^{-11.489 DZ/Q} + 0.3096 e^{-70.06 DZ/Q} + 0.2638 e^{-179.07 DZ/Q}$$

The normalized equation is plotted for various Q/D values in Fig. 2.20. Thus, the activity profile along the length of a diffusion tube can be predicted.

By using the above equation, diffusion coefficients can be calculated from a knowledge of the flow rate through the tube and the measured activity profile along the length of the tube. This can be done easily by comparing the activity profile with the curves of Fig. 2.20. From the diffusion coefficient, the mass associated with a diffusing particle can be estimated, and the chemical nature (i. e., monatomic, molecular, aerosol, etc.) of the fission-product species can be inferred.

Two preliminary diffusion-tube experiments were run in GAIL during Cycles 59 and 60 and the results are encouraging. The diffusion tubes were contained in probe assemblies installed at cold finger positions 2 and 7. Position 2 is between the regenerative heat exchanger and cooler, position 7 is immediately upstream from the heater and downstream from the filter and circulator. Each diffusion tube was a 3/16-in. -diameter copper tube, slightly more than 9 in. in length, the latter half of which had been coated with a thin layer of activated charcoal.

The diffusion tubes were removed from their respective probes when received at General Atomic and sectioned into 1-cm lengths. The sections were gamma counted and a few dissolved for radiochemical analysis. The data were plotted as $\log \mu\text{c}/\text{cm}$ versus length. Plots

Fig. 2.20--Plot of normalized deposition equation (see text)

typical of those obtained are shown in Figs. 2.21 and 2.22. The resulting diffusion coefficients are given in Table 2.22. The magnitude of the diffusion coefficient values indicates that the diffusing species were in the atomic or molecular form.

In continuance of the work, diffusion tubes will be contained in diffusion probe assemblies mounted at cold finger position 1 (upstream from main loop cooler), position 3 (between first and second banks of cooler), and position 5 (downstream from cooler). One set will be removed for analysis following Cycle 61.

Laboratory Diffusion-tube Experiments. Laboratory work has been undertaken to determine fission-product diffusion coefficients for use in interpreting the data from GAIL diffusion-tube experiments. In the laboratory apparatus that has been set up, a helium stream passes through fission-product-vapor generators and then into diffusion tubes similar to those used in the GAIL experiments. The laboratory work so far has been concerned with the determination of diffusion coefficients for Cs, I, and CsI in helium.

The diffusion tubes are 1/4-in. hard-drawn copper tubes nickel-plated on the outside and with either bare copper or silver-plate on the inside. The downstream half of each tube is coated on the inside with activated charcoal so that the gas stream first sees a metal surface, either silver or copper, and then a charcoal surface. Stainless-steel lines connect the diffusion tubes to activated charcoal traps that collect any Cs or CsI that is not deposited in the diffusion tubes. The flow is controlled by needle valves and measured with wet test meters. The furnace was built in two sections so the generators can be maintained at different temperatures. The temperatures along the length of each diffusion tube can be held constant within $\pm 5^\circ\text{C}$. Temperatures are measured by four thermocouples spot-welded to the generators and bypass line.

AEC-grade helium is used, and N_2 and O_2 concentrations are reduced to less than 1 ppm (below detectable levels) by the liquid-nitrogen-cooled activated-charcoal trap. A millipore filter is used to ensure that no charcoal fines enter the experiment. Cs is tagged with Cs^{137} , CsI with Cs^{137} and I^{131} . Care is taken to match the Cs^{137} specific activities.

The experimental data are interpreted by use of the following equation described in the previous section:

Fig. 2.21-- ^{131}I activity profile in diffusion tube, position No. 2, Cycle 60

Fig. 2.22-- Cs^{137} activity profile in diffusion tube,
position No. 2, Cycle 60

Table 2. 22
GAIL DIFFUSION-TUBE EXPERIMENTS

GETR Cycle No.	Position No.	Avg. Temp. (°F)	Est Flow Through Diff. Tube at Local Conditions (cm ³ /sec)	Diffusion Coefficient, D, Calculated from Activity Profile Through Diffusion Tube ^a (cm ² /sec)													
				Charcoal Region							Copper Region						
				Sr ⁸⁹	I ¹³¹	Cs ¹³⁴	Cs ¹³⁷	Ce ¹⁴¹	Ce ¹⁴⁴	Ba ¹⁴⁰	Sr ⁸⁹	I ¹³¹	Cs ¹³⁴	Cs ¹³⁷	Ce ¹⁴¹	Ce ¹⁴⁴	Ba ¹⁴⁰
59	2	880	0.8	<0.005	0.01+	<0.005	<0.005	---	---	<0.005	---	0.08	0.06	0.08	<0.005 ^b	<0.005 ^b	---
60	2	870	2.0	TD ^c	0.02	ND ^c	<0.01	TD	TD	ND	TD	0.08, 0.03 ^d	0.1	0.1+, 0.08 ^d	TD	TD	ND
59	7	250	0.8	ND	0.003	ND	ND	ND	ND	ND	ND	0.03	ND	ND	ND	ND	ND
60	7	270	2.0	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

^aValues are tentative; dependent on the accuracy of estimated flow rates.

^bData show considerable scattering.

^cTD = to be determined; ND = not detected.

^dTwo component profile--may indicate two gaseous species.

$$n(Z) = \left(\frac{N_o D}{Q} \right) (9.4106 e^{-11.489 DZ/Q} + 6.8309 e^{-70.06 DZ/Q} + 5.819 e^{-179.07 DZ/Q})$$

For the accuracy required in this work only the first term of the equation is needed. Thus, the equation reduces to the linear relationship:

$$\ln n(Z) = -11.5 \frac{D}{Q} Z + \ln \left(9.41 \frac{N_o D}{Q} \right).$$

The equation is used in this form to calculate diffusion coefficients from plots of $\log n(Z)$ versus Z .

Five runs have been completed. Experimental data for the completed runs 3, 4, and 5 are summarized in Table 2.23, and the data are plotted in Figs. 2.23, 2.24, and 2.25.

In these experiments it was intended that both the Cs and CsI partial pressures be of measurable magnitude. However, it appears that the Cs generator was depleted prior to run No. 3. This is indicated by the observation that the total Cs and I condensed in each of the last three runs was in the proper ratio for CsI. In run No. 3 both the silver and charcoal were apparently saturated with CsI as a result of the long running time. In runs 4 and 5, Cs and I ratios changed along the length of the diffusion tube, indicating CsI dissociation. It is believed that in run No. 5 the CsI generator was becoming depleted because (1) the total amount of CsI generated in this and previous runs approached the initial generator loading, (2) the CsI transpiration vapor pressure measurement was slightly low (see Table 2.22), and (3) the shape of the Cs profile on the copper was odd (see Fig. 2.25).

From the slopes of the curves in Fig. 2.24 are obtained the diffusion coefficient values 0.076 (Cs on charcoal), 0.050 (I on charcoal), and 0.042 (Cs on copper). From Fig. 2.25 the value 0.034 is obtained for Cs on charcoal. These values apparently apply to CsI in helium, since Cs was not an important gaseous species in the runs owing to depletion of the Cs generator. The diffusion coefficient values are reasonable and are in the range of values determined from GAIL diffusion-tube experiments.

The diffusion coefficient values show considerable spread; however, it is believed that as experience with the apparatus is gained, the degree of accuracy will increase.

Table 2.23
DIFFUSION-TUBE EXPERIMENTS

Exp. No.	Cs Gen. Temp. ^a (°C)	CsI Gen. Temp. (°C)	Diff. Tube Temp. (°C)	Total Time (min)	Flow Rate (ml/min)	CsI Partial Pressure (atm)		Type of Diffusion Tube
						Calculated ^b	PV = nRT ^c	
3	530	515	540	9630	75.6	4.1×10^{-6}	5.7×10^{-6}	Ag-Char
4	525	510	500	1260	31.8	3.3×10^{-6}	3.1×10^{-6}	Cu-Char
5	525	510	500	2700	30.9	3.3×10^{-6}	2.4×10^{-6}	Cu-Char

^aThe Cs generator was apparently not working (see text).

^bCalculated from CsI inlet concentrations.

^cCalculated from generator temperature using: $\log P(\text{atm}) = 5.32 - (8.44 \times 10^3)/T(^{\circ}\text{K})$

Fig. 2.23--Laboratory diffusion tube experiment No. 3 (silver-charcoal)

Fig. 2.24--Laboratory diffusion tube experiment No. 4 (copper-charcoal)

Fig. 2.25--Laboratory diffusion tube experiment No. 5 (copper-charcoal)

III. GAIL LOOP OPERATION

GENERAL PERFORMANCE

The GAIL IV fuel element operated at full power during the entire quarter except for the scheduled reactor refueling periods. The element was operated in three modes: the normal-purge mode, bypass-purge mode, and no-purge mode. During all three modes of operation, the fission product activity in the loop coolant gas remained at a very low level. Operating data for the quarter are presented in Table 3.1.

Shortly after the GETR reached full power (32.5 Mw) for Cycle 61 operation, the GAIL IV experiment was adjusted closer to the reactor core to increase the power in the fuel element to the normal operating value of 73 Kw. (Power in the element is adjusted by a remote positioning device that moves the experiment with respect to the reactor core.) The element was inadvertently moved too close to the reactor, and a reactor scram was initiated by high exit gas temperature from the GAIL IV fuel element. It was subsequently estimated from loop data charts that the fuel element reached a power of 84.4 Kw. Operating data and gas activity samples taken since the incident do not indicate that any fuel element damage resulted from the power excursion.

Table 3.1

LOOP OPERATING PARAMETERS FOR THE QUARTER

<u>Parameter</u>	<u>Data</u>
Integrated power generated, Mw-hr	116.1
GAIL IV total cumulative power, Mw-hr	238.0
Operational full power	1604.9
GAIL IV cumulative full power hr	3312.4
Average thermal power, kw	72.4
Thermal power range, kw	43.5 to 76.1
GAIL IV lifetime average power	71.9

During the quarter the loop was operated for one GETR cycle in the bypass-cleanup mode. For the remainder of the period the GAIL IV fuel element was operated in the purged mode for the first few days of

each operating reactor cycle then operated in the no-purge mode. The purge stream is sampled after fission product activity equilibrium is reached in the loop coolant to obtain greater activity concentrations for the accurate determination of fission product release from the fuel. After sampling, the loop is operated in the unpurged mode to provide a greater concentration of fission products in the main loop helium stream for plateout and diffusion tube studies.

Electrical circuitry in the No. 2 loop circulator was checked and found to be malfunctioning. The power supply was repaired and the circulator was subsequently restarted and operated normally. During cycle 61 operations, reactor scram caused by a spurious instrumentation signal tripped out the No. 2 circulator, and again it could not be restarted. Operations continued using the No. 1 circulator; it is planned to repair the electrical system for the No. 2 circulator during the next reactor outage.

FISSION-PRODUCT RELEASE

Diffusion tubes were installed at the probe access ports in the loop at the beginning of each reactor operating cycle. These tubes are described in Section II.

Periodic gas samples have been collected and analyzed for the noble gas fission products released from the fuel material. The steady-state release fraction values of these isotopes ($\text{Kr}^{85\text{m}}$, Kr^{87} , Kr^{88} , Kr^{89} , Xe^{133} , Xe^{135} , and Xe^{138}) were calculated and the results plotted in Figs. 3.1 and 3.2 as a function of accumulated burnup.

Since the loop has the capability of operation in three different purge modes, samples have been taken from the loop during each of these modes of operation. In the first mode, normal purge operation, a fraction of the loop helium gas is passed through pores in the graphite over the fuel material to carry the freshly released fission gases into a delay bed-cold trap cleanup system. Normal purge results are indicated in Figs. 3.1 and 3.2 as open data points.

In the second mode, bypass-purge operation, the fuel element is not purged, but a fraction of the loop gas is bypassed directly into the cleanup system. The results for bypass purge are indicated in Figs. 3.1 and 3.2 by shaded $\text{Kr}^{85\text{m}}$ and Xe^{135} data points.

In the third mode, no-purge operation, there is no cleanup purge and the fission gases are allowed to build up to their equilibrium values in the main coolant gas. Results for no-purge operation are indicated in Figs. 3.1 and 3.2 by solid data points.

Fig. 3.1--Steady-state release fraction values for krypton as a function of accumulated burnup

Fig. 3.2--Steady-state release fraction values for xenon as a function of accumulated burnup

In order to have a least one readily correlated data point from each cycle, the loop is started in the normal purge mode of operation, sampled, and then switched to another mode. Cycles 60 and 61 were run in this way, switching from normal purge to no-purge operation. The related data points (normal purge) are indicated at ~130 Mw-hr and ~175 Mw-hr. On first glance it appears that both of these data points are low; however, these samples were taken at a time when the element thermocouples were indicating lower-than-average temperatures. Accordingly, these data points would be expected to be lower and, thus, there appears to be reasonably good correlation of data obtained by the three different modes of operation.

The isotopes analyzed (with two exceptions) appear to be leveling out with release fraction values between 3×10^{-6} and 2×10^{-5} . The exceptions noted are the shorter-lived Kr^{89} and Xe^{138} . These two isotopes are determined by way of their daughter products (Rb^{89} and Cs^{138}), and it appears that the analyses are probably near the limits of sensitivity of determination and, therefore, show much more scatter.

The fractional release values appear to be proportional to the square root of half life, indicating that the major portion of the fission-gas release is diffusion controlled.

At the beginning of cycle 61 (just before the last three data points presented in this report) there was an elevation of power above the normal power level. The data presented here do not show any evidence of fuel damage. These data points appear a bit higher than normal; however, it is possible that this could be simply due to elevated operating temperatures.

At the conclusion of cycle 58, purge stream and main-coolant circulation were maintained and periodic purge stream samples were taken for Xe^{133} and Xe^{135} analysis. The results of these analyses were then used to calculate the release fraction of the $\text{I}^{133, 135}$ precursor. The iodine release fractions were determined to be:

$$\text{I}^{133} (20.9 \text{ hr}) = 1.65 \times 10^{-6}$$

$$\text{I}^{135} (6.8 \text{ hr}) = 1.30 \times 10^{-6}$$

Figure 3.3 shows these iodine release results along with the noble gas release results taken just before shutdown. A line showing the ideal square-root-of-half-life relationship (expected if release is diffusion controlled) is included for comparison. The iodine and xenon results show very good agreement considering the problems involved in the iodine analysis.

Fig. 3.3--Release fraction values for iodine and the noble gases as a function of half life at the end of Cycle 58

Because of the very low specific activity of the main coolant gas associated with the normal purge mode of operation, a more sensitive method of analysis had to be devised. An alternative method of sampling has now been tested that employs the use of a liquid-nitrogen-cooled charcoal trap. The higher main coolant specific activity present in the cycle 60 no-purge operation provided a means of cross-calibrating activity determinations by means of sampling by cold traps with determinations carried out using 125 cm³ sample bottles. The cold trapping method is limited in that it does not permit the analysis of the shorter-lived Kr⁸⁹ (3.2 min) and Xe¹³⁸ (17 min).

ANALYSIS FOR IMPURITIES IN HELIUM COOLANT

Analysis of GAIL gas samples for N₂, O₂, CO, CO₂, CH₄ showed no detectable impurities (<1 to 3 ppm) until no-purge operation (which started in cycle 60) had been under way for a while. Gas chromatograph samples taken during the cycle 61 no-purge operation show the coolant gas to contain between 5 and 10 ppm of CO. This concentration is fairly near the minimum detectable and appear to be remaining at a fairly constant value.

IV. RECYCLE DEVELOPMENT

A preliminary design study was initiated on a head-end system for graphite fuel element reprocessing that could be incorporated into the service facilities of a large HTGR. The process under consideration is oxidation of the graphite and the fuel particles. A capacity of two elements per day was selected as a design basis for the oxidizer and the necessary auxiliary equipment such as the off-gas cooler and filter. The oxidizer will be a fluidized bed type with Alundum particles used as a carrier. The bed will be operated at an average temperature of about 750°C.

A major problem associated with burning the fuel elements is that some fission products may become volatile and may, therefore, plate out on the inner surfaces, especially the cooler ones, of the bed, coolers, and filters. A rough check of the possible surface activities after a long period of operation shows fairly high values. This could complicate maintenance considerably, since both decontamination and additional shielding of the burning system would be required. To reduce this additional work as far as possible, all the units of the system that may become highly radioactive are incorporated into one cylindrical container. This is then located directly under the operating floor of the service building, so that it is in the operating range of the shielded auxiliary transfer cask. With the cask, it is possible to remove the radioactive units from the cylindrical container and transfer them to repair or disposal facilities.

Studies have been done to determine the feasibility, from the view-point of process design, of locating the major components of the oxidation system in one compact container.

V. PHYSICS

NUCLEAR ANALYSIS OF 1000-Mw(e) REACTOR CORE

The ability of the HTGR to utilize nuclear resources efficiently has been studied using a symbiotic reactor system. In this system there are two types of HTGR which differ principally in their fuel management. One of the reactors (feeder) is fueled with fully enriched uranium with no re-cycle of the bred uranium (approximate composition 87% U^{233} , 11% U^{234} , 2% U^{235}); the second reactor (advanced) is fueled exclusively with this re-cycle material. Advanced reactors are started only when their inventory requirements are available and the nuclear capacity demands more output. If additional nuclear capacity is required and the advanced reactor inventory is not available, a feeder reactor is installed. As the rapid growth of nuclear power subsides, the symbiotic complex requires fewer feeder reactors to supply inventory and makeup fuel and are thus supplanted by advanced reactors. This is easily accomplished in the HTGR complex, because the reactors are quite similar physically and require only a modification in the fuel management.

Some fuel cycle characteristics of three HTGR complexes are shown in Table 5.1; in all cases the reactors have an output of 1000 Mw(e) and are refueled semiannually. The advanced reactors all utilize thorium blankets, whereas the feeder reactors do not. System 1 in the table represents a large HTGR of fairly standard design using all-graphite fuel elements and complete retention of fission products. System 2 represents a slightly more advanced system in that BeO spines are used in the fuel elements of both the feeder and advanced reactors. System 3 uses the same feeder as system 2, but the advanced reactor releases the volatile fission products. The projected nuclear capacity together with the uranium ore requirements of the three symbiotic reactor systems are shown in Fig. 5.1. The ratio of advanced reactors to feeder reactors as a function of time is shown in Fig. 5.2. It is assumed that the feeder reactors would begin operation in 1970 and that advanced reactors would begin as fuel became available. In reality, the advanced reactors would lag by several years while the handling problems associated with U^{233} were being resolved, but this merely changes the behavior during the early years when quite small capacities are considered. The curious behavior of the ratio of advanced reactors to feeder reactors around the year 2000 results from the transition of a six-year capacity doubling time to a ten-year doubling time. The longer doubling time requires fewer feeder reactors and, indeed, some feeder reactors can be converted to advanced reactors, particularly in system 3.

Table 5.1

SOME CHARACTERISTICS OF SYMBIOTIC HTGR COMPLEXES

System	C/Th(Be/Th)	Fuel Residence Time (years)	Conversion Ratio	Initial Specific Power (kw/kg)
Feeder-Reactor				
1	200	5	0.72	1080
2	(28)	5	0.83	840
3	(28)	5	0.83	840
Advanced Reactor				
1	200	4	0.90	1620
2	(40)	4	0.99	1540
3	(40)	4	1.06	1960

METHODS DEVELOPMENTNew Methods

Work is in progress to develop a new approach to the general problem of calculating space-energy distributions. The basic idea of the method is to expand the spatial dependence of the solution of the appropriate equation (transport, diffusion, etc.) in known functions and solve the resulting energy equations for the expansion coefficients numerically. Both fixed source and eigenvalue problems can be treated by this method. For example, in the fixed source problem,

$$H\phi(\vec{r}, E) = S(\vec{r}, E)$$

is the equation of interest. Here $\vec{r}$ is the spatial coordinate, E is the energy coordinate, $S(\vec{r}, E)$ is the external source, $\phi(\vec{r}, E)$ is the flux distribution, and $H(\vec{r}, E)$ is the operator appropriate to the problem under consideration. It is now assumed that the flux in each region (denoted by i) can be approximated by

$$\phi_i(\vec{r}, E) = \sum_{n=1}^N f_{ni}(\vec{r}) g_{ni}(E),$$

Fig. 5.1--Resource requirements of several symbiotic HTGR systems

Fig. 5.2--Ratio of advanced to feeder reactors for several symbiotic HTGR systems

where the $f_{ni}(\vec{r})$ are taken as known functions (determined from the physics of the problem) and the $g_{ni}(E)$ are to be determined. All of the $g_{ni}(E)$ are not independent, since it is required that the continuity conditions at interfaces and the boundary conditions at the outer surfaces of the system be satisfied by the assumed expansion. For example, in diffusion theory, only $N-2$ of the $g_{ni}(E)$ in each region are independent quantities, since continuity of current and flux imposes two additional conditions. Now substitute Eq. (2) into Eq. (1), multiply by $W_{mi}(\vec{r})$, $1 < m < M$, and integrate over the region. The result is M equations for each region of the form

$$\int dV \left[H \sum_{n=1}^N f_{ni}(\vec{r}) g_{ni}(E) - S(\vec{r}, E) \right] W_{mi}(\vec{r}) = 0 \quad (3)$$

to be solved for the $g_{ni}(E)$. M corresponds to the number of independent $g_{ni}(E)$ in each region ($M = N-2$ in diffusion theory). In general, this coupled set of energy equations will be solved numerically. A more detailed description of the method can be found in GA-5948.*

To obtain some numerical comparisons with exact calculations, this formalism is being coded for a two-region, slab, diffusion-theory thermal problem. The flux within each region is assumed representable by a parabola. Hence one independent energy function is to be determined in each region, and one function, $W_i(\vec{r})$, is required for each region. Two choices are being investigated. The first takes $W(\vec{r}) = 1$, and the resulting equations allow the interpretation that neutrons are conserved in each region. The second choice corresponds to a variational procedure in that $W(\vec{r})$ is taken as the spatial coefficient of the single independent energy function in a parabolic fit to the adjoint flux. These numerical results should be available shortly and will point the way to the use of the formalism for more realistic problems. In particular, it is planned to use this method to develop a new set of different equations for the multigroup diffusion equation that may be useful for multidimensional calculations.

The treatment of the resonance reactions in fissile nuclides generally assumes infinite dilution and no interaction with other nuclides. A method has been formulated that treats all resonance absorption concurrently and is being programmed for the computer.

*G. C. Pomraning, An Approach to Space-Energy Problems, General Atomic, Report GA-5948, January 5, 1965.

Code Development

GAMBLE-IV. The GAMBLE-IV code is a two-dimensional multi-group diffusion theory code that allows an arbitrary scattering matrix and up to 20,000 mesh points with 10 energy groups. The code, which was completed during this quarter and released for initial use at General Atomic, employs the iteration and convergence technique developed at ORNL in the EXTERMINATOR code. Further corrections and revisions in the GAMBLE-IV code are expected to be required as more extensive use of the code tests all of its features.

PIFFIN. A program was written to calculate and plot activity traverses for reactor cells, reactor cores, and critical assemblies, using results from the GAZE-2 code. This code, called PIFFIN, was written in the 7040/7044 FORTRAN-IV language; it computes the relative activity of a detector nuclide as a function of position in the system for which a GAZE-2 calculation was run to obtain fluxes.

Cross Section Methods. Development and checkout of a code (UNPACK) that retrieves cross section data from the BNL data file tape has been completed. The Sigma Center at BNL has developed a system for storage (SCISRS) that provides an efficient method of storage of experimental results from differential cross-section measurements and resonance parameters obtained from these measurements. In order to extract data from the BNL data tape, the UNPACK code was written. This code is able to retrieve either all or any selected types of data for a particular nuclide. The data are printed and punched on cards in a form that is usable for cross-section evaluation.

Cross-section Evaluation

U²³³. The total and partial cross sections of U²³³ have been reevaluated for the energy range from 0.001 kev to 1 kev. This evaluation was based upon the differential total and fission cross section reported in BNL-325* and Supplement 1† along with results from recent measurements. Table 5.2 shows the evaluated 2200 m/sec cross sections. Table 5.3 gives the comparison between the resonance integrals calculated from these cross sections and the results from recent experimental measurements.

*D. J. Huges and R. B. Schwartz, Neutron Cross Sections, BNL-325, 2d ed., July, 1958.

†D. J. Hughes, B. A. Magurno, and M. K. Brussel, Neutron Cross Sections, BNL-325, 2d ed., Supplement 1, January, 1960.

Table 5.2

2200 M/SEC CROSS SECTIONS

<u>Cross Sections</u>	<u>Value</u>
Total	585.1 b
Elastic scattering	11.9 b
Absorption	573.2 b
Fission	525.0 b
Capture	48.2 b
Alpha, σ_c/σ_f	0.0918
Eta, $\nu\sigma_f/\sigma_a$	2.2925
Nu, ν	2.5030

Table 5.3

EPITHERMAL INTEGRALS FOR U²³³

	Cutoff Energy (ev)	I _{abs} (barns)	I _{fission} (barns)	I _{n,γ}	$\bar{\alpha}$	$\bar{\eta}$
Feiner (KAPL) ^a	0.500	958	820	138	0.165	2.148
Halperin (ORNL) ^b	0.540	1012	865	147	0.170	2.139
GA (evaluated)	0.414	944	806	138	0.171	2.137
GA (evaluated)	0.500	916	780	136	0.176	2.131

^aL. J. Esch and F. Feiner, "A Measurement for Alpha for U²³³ and U²³⁵ in the Epithermal Region," Trans. Am. Nucl. Soc., Vol. 7, No. 2, November, 1964.

^bJ. Halperin, et al., "The Average Capture-to-Fission Ratio of U²³³ for Epithermal Neutrons," Nucl. Sci. and Eng., Vol. 16, No. 2, June, 1963.

Other Thorium Cycle Data. A reevaluation has been initiated for the cross sections of nuclides that are of interest to thorium cycle systems, other than for U²³³ discussed above. These nuclides include Th²³², Pa²³¹, Pa²³³, and U²³². These reevaluated cross sections will reflect results from differential and integral cross-section measurements that have become available since late 1962.

CRITICAL EXPERIMENT

The first draft of the hazards report for the TARGET critical facility is about 25% completed. Additional investigations of the reactivity addition that would result in the event of melting and subsequent loss of the aluminum honeycomb and cladding material are being made. Analyses of the consequences of flooding the assembly were initiated. The facility and the assembly are being studied to determine whether either of these postulated accidents is credible.

A further reduction of the requirements for new fuel columns has been achieved. The number of 3-ft columns of the C/U = 2500 material has been reduced by 308. This reduction must be accompanied by an increase of ~50 columns of the C/U = 432 material. An additional cost savings will be obtained by using machined graphite columns in both core and reflector rather than in the reflector alone.

A tentative fuel storage array has been selected on the basis of the results of survey calculations. The survey calculations were done for various geometries both flooded and unflooded, and both with and without permanently installed nuclear poison. Detailed calculations of k_{eff} for the selected array are in progress in order to provide the justification for this selection.

Temperature coefficient calculations are being performed for the critical assembly. Reactivity deficits associated with a change in temperature from a uniform distribution at 27°C to a distribution with a 650°C hot point have been calculated for each array. The 650°C limit is imposed by the melting point of aluminum. It will be necessary to restrict the maximum available reactivity to some fraction of this calculated reactivity deficit.

Kinetics calculations that will be used to help specify maximum allowable reactivity ramps are under way.

The first model of the safety rod proved to be unsatisfactory because sufficiently precise control could not be exercised over the withdrawal rate by control of pneumatic pressure alone. The rod has been redesigned with a mechanical limiting device, and a new prototype is being built. Detailed design of the control rod is about 75% complete.

The original safety circuit design used for the experiments in support of the Peach Bottom reactor is being modified to incorporate an additional independent scram channel. The additional scram will be activated by a temperature sensor set to prevent an aluminum meltdown.

VI. PRESTRESSED CONCRETE REACTOR STRUCTURE

ANALYTICAL METHODS DEVELOPMENT

Elastic Stress Analysis of Axisymmetric Composite Structures. A large-capacity finite element computer program (2400 degrees of freedom) has been developed during the past three months. This program is non-iterative and therefore can be applied to thin as well as thick structures involving axisymmetric stress distribution. Nonhomogeneous structures, such as prestressed and reinforced concrete structures, can be treated as elements that have biaxial and/or uniaxial structural properties. The steel liner is assumed to act as a shell with membrane stiffness only. Prestressing can be included as fully bonded (grouted) or friction-free tendons of arbitrary curvature. Conventional reinforcement is introduced as fully bonded uniaxial elements. The program, called SAFE, is now available for production use. A typical running time for a problem with 1800 degrees of freedom is 30 centihours.

Stresses generated by SAFE are written on tape and can be used as input by another program that plots coordinate and principal stresses and principal stress directions for all or part of the structure being analyzed. The plotting of stress contours will reduce the man-hours required for the interpretation of SAFE output to a very small fraction of the time required previously for this arduous task. Another computer program, SAFE-SHELL, for the analysis of thin shells of revolution is under development. This program is applicable to axisymmetric shells with arbitrary surface of revolution and variable thickness. Thermal as well as mechanical loads can be handled. Several problems have been solved by this program, and it is almost ready for production use.

Creep Analysis. A method of analysis of structures under steady-state creep has been formulated. The method is being incorporated in the stress analysis programs now available. Creep analysis of axisymmetric solids will be attempted first. The procedure is essentially a reformulation of the finite element equations to incorporate time-dependent constitutive equations. Programming of the problem is well under way and progressing rapidly. Some preliminary results have already been obtained. These are still under study; they seem to indicate that the method of solution is basically workable.

DESIGN DEVELOPMENT

The last concrete for the first PCRS concrete model has been placed, and moist curing is under way. During the total curing period, preparations for the next phase, prestressing and testing, are being made.

Test reports on concrete used for the PCRS model show that the strengths exceed the allowable minimum of 5500 psi and that the highest strengths exceed the lowest by approximately 4% of the lowest based on standard ASTM sampling and testing methods for 28-day compressive strength of test cylinders.

Placement, consolidation, and field curing will determine the adequacy of the in-place concrete. The final product will be checked in place by using Schmidt rebound hammers and by radiographic examination in areas where consolidation is more difficult. The most significant tests will be the prestressing operation, in which the concrete will be loaded in compression to a higher strain state than would be required by design pressure loadings. Such a test will always be required on a PCRS, because the prestressing methods require the application of initial loads that exceed the desired residual prestressing loads.

DEVELOPMENT TESTING

Portions of the data acquisition system have been received and are being installed. Sensors are being connected to the system. All embedded sensors have been installed. Plans have been made for installing surface-mounted sensors when the moist-cure period for the model is complete.

CONTROL OF VESSEL TEMPERATURE

A thermal barrier is required on a PCRS to limit heat loss and to protect the concrete from excessive temperature. The thermal barrier is actually a system composed of an insulating material, a sealed liner, and a cooling system. This system must maintain the concrete temperature at or below 150°F. The insulation reduces the heat flow, the sealed liner prevents loss of the reactor coolant, and the coolant passages remove heat that escapes through the insulation and radiation heating.

During the previous quarter (see GA-5866), work included the collection of cost and performance data on alternative insulations, investigations of installation problems, preliminary economic optimization, and analysis of the temperature dependence of conductivity of metal-foil reflective insulations. Work has continued in the present quarter on development

of cost data and economic optimization. Work has been initiated on simplified low-cost metal insulation design and installation concepts, and on analysis of heat flow in the liner and to the coolant channels.

Economic Optimization Analysis

The thermal barrier cost analysis is a computation of the sum of construction costs plus capitalized operating cost from which the cost minimum with respect to a selected parameter can be determined. Construction and installation costs are computed from unit costs for the thermal insulation, the coolant passages, the coolant heat exchanger, the pump and secondary coolant, and the space within the PCRS occupied by the insulation. Costs of operating the heat removal system and the cost of heat lost from the PCRS are computed. The analysis formulation, which includes simple sizing computations for such components as heat exchangers and pumps, has been planned for execution on a computer.

Preliminary hand calculations were performed using this analysis and preliminary cost data. Charges for the heat exchanger cooling system were not included. All costs were capitalized over 30 years, assuming 14.5% interest charges. This value was recommended by the AEC for use by private utilities.* The PCRS considered in this evaluation is 40 ft ID and 32 ft high, and only the portion of the PCRS insulated from the cold gas (750°F) was considered. This portion covers the largest percentage of the surface area. Results obtained from this study are given in Table 6.1. The cost of heat lost included the fuel cycle cost plus fixed costs of the reactor and the portions of the primary loop and PCRS whose cost depend upon reactor thermal power. A breakdown is given in Table 6.2 of the contributions to total cost from each of the major equipment or cost areas considered in the analysis.

Calculation of Heat Transfer Through the Thermal Barrier

An analysis of the heat transfer through the thermal barrier insulation and metal liner to the cooling passages was developed. This analysis will permit the determination of the cooling pipe spacing required to keep the concrete below a specified maximum temperature. The prediction is based on a one-dimensional fin model of the cooling tube and liner system. The study to establish the equation was general in scope and included effects of nonuniform heating and cooling of the fin. The analysis assumes that the concrete is a perfect insulator, that all heat flux in the insulation is uniform, and that the heat flows laterally through the steel liner to the cooling pipes and thence to the cooling fluid. The gamma heat source has been omitted, because it is negligible compared to the conduction heat load from the insulation.

*Kaiser Engineers, Guide to Nuclear Power Cost Evaluation, Supplement No. 1, TID-7025, September 30, 1962.

Table 6.1
COST COMPARISON OF ALTERNATIVE INSULATIONS

Insulation	Optimum Thickness ^a (in.)	Relative Minimum Total Cost	Heat Loss (Mw(t))
Solami ^b	1	215	3.5
Pumice concrete	4	210	2.9
Castable refractory	3.5	210	2.6
Ceramic fiber	2	160	1.5
Ceramic block	1	110	1.4
Marinite ^c	1	125	1.8
Ceramic blanket	1	100	1.2
Metal sheet	2	140	1.7
Min-K ^c	0.5	100	0.9

^aThickness corresponds to minimum total cost.

^bTrade mark of Solar Aircraft Company for metal foil insulation.

^cTrade marks of Johns Manville.

Table 6.2
BREAKDOWN OF MINIMUM TOTAL COSTS FROM TABLE^a

Insulation	Insulation Cost	Space Within PCRS	Heat Lost	Coolant Piping on Liner	Heat Exchanger	Pump
Solami ^b	30	4	32	26	4	4
Pumice concrete	25	15	26	24	5	5
Castable refractory	33	13	22	22	5	5
Ceramic fiber	40	10	18	23	4	5
Ceramic block	30	7	23	31	4	5
Marinite ^c	26	6	28	31	4	5
Ceramic blanket	27	8	24	32	4	5
Metal sheet	28	11	25	27	4	5
Min-K ^c	45	4	20	25	3	3

^aGiven as a percentage for major items.

^bTrade mark of Solar Aircraft Company for metal foil insulation.

^cTrade marks of Johns Manville.

A sample calculation was performed for a PCRS with liner area of 6000 ft² in which cooling-tube spacing was determined as a function of heat lost from the core through insulation. Water at 100°F flowing in a 3/4-in. steel tube at 5 ft/sec was assumed. The results show a spacing of 10-1/2 in. is required for a 1 Mw heat loss (565 Btu/hr-ft²) to limit the peak concrete temperature to 140°F. Other results are shown in Fig. 6.1.

The results in Fig. 6.1 show the effects of pipe wall thickness and attachment-weld cross section upon required pipe spacing. For a 1 Mw(t) heat loss to the cooling system, there is no significant difference in spacing between a schedule 80 pipe (curve A) and a schedule 40 pipe (curve B). The effect of weld cross section becomes significant at higher heat flux as may be illustrated by a comparison of curve (B) and curve (C).

Insulation Concepts Under Study

After the preliminary evaluation of the various types of insulation systems, it appeared that the insulation materials fell in a high-cost and a low-cost group. The high-cost group included Solami,* pumice concrete, and castable refractory. The low cost group included ceramic fiber, ceramic blanket, fiber block, metal sheet insulation, and Min-K. The ceramic fiber block or blanket material are manufactured by many vendors and under many trade names. Min-K is a proprietary item manufactured for Johns Manville. The metal sheet insulation is a metal insulation using carbon steel sheet with metal spacers. The ceramic fiber block, board, and Min-K are usually fabricated in flat sheets. The pieces must thus be trimmed to fit curved surfaces, which increases the installation costs. In addition, these materials must be wrapped in some type of sealed liner to control the release of dust.

To reduce the cost of installation, fabrication of the liner as a polygon rather than a cylinder will be investigated. The metal sheet insulation appears to be a solution whose details could be worked out in a minimum amount of time. Because of this, work was started on design concepts of this type of insulation.

Work was completed on a design and layout study for a metal sheet type of insulation. This concept consisted of alternately stacked layers of 28-gauge corrugated steel sheet and 25-gauge flat steel sheet between a hot- and a cold-side facing sheet. The insulation would be fabricated into panels nominally 30 x 39 x 2 in. thick using 11 corrugated and 10 flat sheets. The panels would be held together by four pins near the corners, and the entire panel would be held in position and supported by one 1/2-in. diameter stud located at the center of the panel and stud welded to the PCRS liner.

*Trademark of Solar Aircraft Company for metal foil insulation.

Fig. 6.1--Cooling tube spacing

The costs and fabrication of this panel design were discussed with a potential vendor. The vendor suggested that the price of the panel might be reduced by redesigning to use existing tooling, thinner gauge material, and a flat-sided liner. These possibilities will be investigated.

The conductivity of this insulation was calculated to be 0.37 Btu/ft-hr-°F, which compares with 0.25 Btu/ft-hr-°F for Solami. Comparable heat-transfer analysis models and data were used in both cases.

The use of dimpled sheets is also being considered in the development of a low-cost metal insulation. This concept would reduce the thermal conductivity by reducing the metal contact area and could reduce the cost of fabrication by reducing both the amount of material and the labor required for installation.

Future investigations of this concept will include dimple spacing, dimple size, thermal expansion characteristics, optimum material thickness, support method, and cost analysis.