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GENERAL ATOMIC

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AEC RESEARCH AND
DEVELOPMENT REPORT

GA-7314

Project on

PUBLIC SERVICE COMPANY OF COLORADO
330-MW(E) HIGH-TEMPERATURE GAS-COOLED REACTOR, 6/66
RESEARCH AND DEVELOPMENT PROGRAM

QUARTERLY PROGRESS REPORT
FOR THE PERIOD ENDING
JUNE 30, 1966

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

September 30, 1966

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H.Q. # 3.11, MN. 65

JOHN JAY HOPKINS LABORATORY FOR PURE AND APPLIED SCIENCE

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

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QUARTERLY REPORT SERIES

GA-6830--July, 1965, through September, 1965

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INTRODUCTION

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

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

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PART I

Task I
COMPONENT DEVELOPMENT

REACTOR VESSEL

The objective of this subtask is to acquire information necessary to develop and specify the design of the Prestressed Concrete Reactor Vessel (PCRV) for the plant and to provide a technical basis for obtaining necessary approvals of regulatory bodies and insurance groups by means of conceptual design, analyses, and testing of materials, scale models of the vessel and full-scale parts of the vessel, such as tendons and penetrations, where necessary.

Model testing will include heating internally to produce appropriate temperature differences across the concrete, long-term tests, measurement of leakage through the concrete resulting from a simulated crack in the liner, test to destruction and test to demonstrate the adequacy of the double closures on penetrations. Representatives of interested technical societies, regulatory bodies, and insurance groups will be kept informed of the progress of the program and will be encouraged to witness key tests.

PCRV Model No. 2 Testing

Early in the quarter, PCRV Model No. 2, shown in Fig. 1.1, was tensioned to design loading, and data were obtained for the calculation of the prestressing system characteristics. A photograph of the model taken just after the prestressing was completed is shown in Fig. 1.2.

The model, which was designed for an internal operating pressure of 600 psig, has been pressurized up to its reference pressure of 705 psig several times. Progressive pressure levels were applied to the vessel in 100-psi increments before the 705-psig pressure was reached. From five to ten pressure cycles were made at each increment. The pressure of 705 psig was held for 24 hr; data taken during this period indicated that the vessel was responding as predicted. Permeation tests at gas pressures to 500 psig have produced no unexpected results.

The liner of PCRV Model No. 2 was visually inspected after completion of the pressure testing phase of the program. No evidence of buckling or other indications of yielding were found.

The temporary scaffolding and jack handling structure was removed and a Butler granary was erected around the model. The heating system is being completed in preparation for the temperature tests. Model moisture-level data were reviewed to determine whether it would be possible to start the temperature tests before the scheduled date. The data indicated that the rate of decrease in moisture level was becoming asymptotic with time and

Fig. 1.1--PCRV Model No. 2

Fig. 1.2--PCR Model No. 2 just after the completion of prestressing

that a relatively long period would be required before the moisture level decreased to a significant degree.

At the end of the quarter, testing of PCR Model No. 2 with gaseous nitrogen was under way with a maximum pressure level of 705 psig. The overall structural response of the model has been as expected, and only minor leakage has been observed from instrumentation wiring penetrations and piping connections.

All equipment and material have been received for the heating test. The granary enclosing the model has been insulated. Lighting has been installed, and scaffolding has been erected around the model to allow close inspection of the concrete surface during the nitrogen pressure and heating tests.

PCR Model No. 2 Design and Analysis

Analytical Studies. In order to provide temperature distributions for a creep analysis of PCR Model No. 2, a transient thermal analysis of this structure was performed. It was found that the desired initial temperature difference of 25°F across the wall will be reached after 600 hr. After a period of holding at this condition, heating will be resumed and the ultimate temperature difference of 50°F across the wall will be reached after approximately 1100 hr.

To expedite temperature data preparation for use in stress analysis, a computer program was written to read the RAT-code output tapes and punch temperature data decks for any arbitrary time during the transient.

Calculations to determine the unit tendon loads for the as-built Model No. 2 have been completed using the SAFE-PCRS code. The work entailed transforming the prestressing tendon force reactions into axisymmetric forces. The unit load information allows the simulation of any state of stress in the model by superposing the tendon loads, pressure loads, and thermal loading effects.

The as-built Model No. 2 has been analytically simulated by a grid system specially devised for the numerical analysis of creep. The prestressing cables, reinforcing steel, and liner have been included in this simulation. A projected loading history of Model No. 2 has been assembled for evaluating concrete properties.

An ultimate failure analysis for PCR Model No. 2 has been initiated. As a preliminary step in this work, the liner stresses and deformations are being computed using the SAFE-SHELL computer code.

Data Evaluation. Proper identification of the data point samples (DPS) and specific sensors is an important phase of data evaluation processes, especially when a large mass of test data is accumulated. For PCR Model No. 2, a complete record is made of the DPS together with other information including time, storage tape number, test phase, and pertinent remarks such as jacking operations and pressurization levels. A complete list of all

sensors has been prepared incorporating identification of channel and gauge numbers, and sensor type, location, and orientation. A history of the status of sensor operation at the different stages of the test program is being recorded. In addition, drawings showing the as-built sensor locations and orientations with respect to reinforcing steel, prestressing tendons, and penetrations were completed for use in the evaluation of data.

Before the sensor output is interpreted in terms of strain, load, temperature, and other parameters, the reliability of each sensor output during each phase of the test program is investigated. Several methods for establishing sensor output validity are employed:

1. Sensor Status Report. Questionable gauges, those showing a rapid change in measurement with no environment or load change, have been recorded for further evaluation using other available techniques.
2. Computer Out-of-range Voltage Check. A computer means of listing questionable gauges is in operation. The computer program tabulates the cumulative number of out-of-range outputs for specific gauges after a certain number of DPS scans. These limits are large so that high strains are not listed as out of range.
3. Comparison Check. Gauges that are symmetrically located and similarly oriented with respect to the principal direction of the PCRV should exhibit comparable but not identical response under similar conditions. Computer programs are in operation to plot and tabulate sensor outputs for comparable groups of sensors. This facilitates isolation of trend lines.
4. Statistical Analysis. Statistical techniques are being utilized to establish sensor output reliability in accordance with the criteria of linearity and consistency. The linearity check is based on the expected linear strain response of gauges within a range of pressure levels. The consistency check is based on the expected consistent repeatability of a measured parameter (i.e., the strain response under cyclic pressurization) under repeated test conditions.

The statistical analysis programmed in the computer includes:

- a. Linear regression and correlation analyses for the pressure-strain relationship for specific gauges.
- b. Analysis of the distribution of statistical parameters, such as means and standard deviations (consistency check) for groups of sensors.
- c. Limits on dispersion for sensor outputs with a wide range of scatter.
- d. A facility for comparing groups of sensors using the linear equations obtained.

Sensor outputs that do not meet the requirements of the evaluation methods enumerated above, either completely or partially, will be examined carefully and disposed of as follows:

1. Rejection of bad sensor output.
2. Limited application of sensor output on a short-term basis only.
3. Proper qualification of inferences based on doubtful data by stating a level of confidence to reflect the degree of reliability.

After sensor outputs have been validated, interpretation and comparison of the test results can be made with greater confidence.

Sample Results of Sensor Outputs. Figure 1.3 shows the strain response to cyclic pressurization of three resistance-type (SR) gauges attached to the reinforcing steel. The location and orientation of the SR gauges are indicated on the sketch in Fig. 1.3. Initial time reference (zero day) was taken at the start of the job-mixed concrete placement. The prestressing operation was completed 50 days later.

Figure 1.3 graphically illustrates the strain response to the different pressure levels. The high points indicate strain output at test pressure; the low points indicate strain output at zero pressure. It should be noted that there is a variation in strain outputs at zero pressure. At the maximum applied pressure of 705 psi, the measured strains for the three SR gauges have not exceeded the strains due to prestressing. At the start of prestressing, SR823 read +75 $\mu\text{in./in.}$

No strain outputs were monitored during the permeation tests. At the completion of the permeation tests, a continuous pressurization test at 100-psi increments from 0 to 700 psi was conducted with sensor outputs being monitored at each pressure increment.

PCRv Response to Vertical Prestressing. The behavior of the PCRv as a whole unit at prestressing was investigated using information obtained from sensor outputs and jack forces during the tensioning of the vertical tendons. A comparison of the applied total force from tendon prestressing with the forces computed from the average strain in the concrete and reinforcing steel indicates that the PCRv responded as expected.

The forces in the concrete vessel were calculated using the following approximations:

1. An average vertical strain from 37 sensors (24 electric resistance gauges and 13 frequency gauges) located within the middle 7-ft. section.
2. A gross concrete area of the cylindrical portion of the vessel of 85.8 ft^2 .
3. Average modulus of elasticity (E_{conc}) of 4,100,000 psi.
4. Force in concrete = (Strain)(E_{conc})(Area).

The comparison of results is as follows:

1. After the tensioning of 24 vertical tendons, the total jack force was 6,179,000 lb; the total calculated force in the concrete was 6,180,000 lb (Average strain = $121 \times 10^{-6} \text{ in./in.}$).

Fig. 1.3--Strain response to cyclic pressurization of three SR gauges

2. After the tensioning of 36 vertical tendons, the total jack force was 9,257,000 lb; the total calculated force in the concrete was 9,920,000 lb (average strain = 194×10^{-6} in./in.).

PCRV Liner

Preliminary designs of a liner buckling test specimen and tension anchor shear and pullout specimens have been developed.

Prestressing System

Plans are being made for a series of full-scale tests of the prestressing system to determine relaxation and anchor efficiency, installation problems related to curved tendons, and methods of corrosion protection. Preliminary design of the prestressing test beds was completed.

Penetration Closures

Preparations are being made for the fabrication of a test rig that will be capable of testing several different types of penetration gaskets. The rig will duplicate the steam generator primary flange and gasket geometry. The primary purpose of the test is to determine the effect of thermal cycling on the leakage characteristics of different gaskets.

Concrete Properties

Preparations are being made for the measurement of various physical properties of concrete needed for the PCRV design. Of principal interest is creep under multiaxial loading. Other properties that will be determined are compressive strength, modulus of elasticity, Poisson's ratio (elastic), thermal expansion, and thermal conductivity.

The specimens for the experiment will be modified cruciforms, cylinders, and cubes that will be sealed to assure a condition of hygral equilibrium. Strain measurements will be made with vibrating wire strain gauges (developed by General Atomic) embedded in the specimens.

Approximately 60% of the specimens for the first phase of the program have been cast and sealed and are curing in the 70°F curing tank.

All components for the hydraulic equipment have been procured and installation is under way. The fabrication of the test load racks is 90% complete.

The facilities for coupling to the automatic data acquisition system (DAS) are being fabricated. Because they will not be completed in time for use on the first set of test specimens, testing will start using manual facilities.

Data concerning autogenous shrinkage have already been acquired. Four cruciform specimens are being monitored in detail. The sensors were read just before casting, immediately after casting, and once each day thereafter.

No data have yet been completely reduced and analyzed. Data reduction mechanisms are being considered.

THERMAL BARRIER

The objective of this subtask is to establish the suitability and reliability and predict the maintainability of thermal barrier materials and designs for predicted operating conditions of the PCRV by means of analyses, conceptual designs, fabrication development, materials testing, thermal testing and accelerated life testing, where meaningful, on thermal barriers under simulated environmental conditions (in helium, at temperatures up to or greater than those predicted in the reactor and, where analyses indicate necessary, at reactor pressure).

Work has commenced on the single-gas-layer conductance measurements, which constitute the first stage of the test program for the Class A thermal barrier presented in the previous quarterly report. The objective of this portion of the program is to determine the effects of spacers and leakage convection flows on the conductivity of a gas layer. The test will be carried out with air, using thermocouples for temperature measurement and flow visualization techniques to observe convection effects. Design and construction of the test equipment is in progress.

PRIMARY COOLANT SYSTEM FLOW

The objective of this subtask is to determine the pressure drop coefficients in the primary coolant system and to verify the distribution, mixing, and stability of the coolant flow in the primary coolant system under normal and emergency cooling conditions of the Plant by means of analyses and model test(s) simulating the PCRV cavity and/or pertinent internal structures thereof.

Checkout of the pumping facilities and the instrumentation for the primary coolant system flow model testing was completed.

Most of the tests on the flow characteristics of the region from the bottom of the reflector blocks to the steam generator entrances (Region I) have now been completed. The results, which are now being analyzed, will provide information on the flow distribution to the steam generators, mixing of flow from core regions with different outlet temperatures, effects of varying the plenum height, and some details of the flow passages.

COMPRESSORS

The objective of this subtask is to establish the suitability and reliability of the compressors and their related operating controls and instrumentation for expected operating conditions in the Plant, except radiation, by means of testing and analysis of such items as over-all equipment performance, operational limitations, and maintenance requirements.

The inquiry specification for the purified helium compressor (a low-flow, high-head machine) has been completed and sent to potential vendors. Plans for the test program on this compressor are being reviewed.

VALVES

The objective of this subtask is to determine the operating characteristics of the steam/water dump valve(s) and associated system, including related instrumentation, and critical-service helium valves, and their suitability, reliability, and maintainability for service in the plant by leak, performance, and accelerated life testing of full-size valves and analyses.

Steam/Water Dump Valve

Additional runs were made using the computer program WOOSH with a piping and valve configuration more closely approximating the piping layout for the plant. These confirmed the results reported in the previous quarterly report.

Helium Service Valves

An investigation of helium valves being tested or used on other reactor plants has been initiated. This investigation will look into problems encountered in the design, fabrication, field installation, and use of the valves. From this study a detailed program for the PSC valves can be developed.

INSTRUMENTATION AND ELECTRICAL

The objective of this subtask is to develop thermometers for measuring fuel element temperatures, and moisture detectors and electrical penetrations, and to establish their operating characteristics, reliability, and maintainability in the Plant by means of analysis and testing in helium, at temperature, and where analyses and/or preliminary tests indicate a necessity, at pressure and other environmental conditions.

Moisture Detectors

During this quarter, the moist helium test source and the detailed design of the optical-dewpoint moisture detector head were completed. All mechanical parts for the first prototype detector head assembly have been fabricated and are being welded and brazed. Before assembly, the light sources, the photocells, and the windows were subjected to pressure to test their integrity. These tests indicate that a mechanically stronger photocell is needed. Attempts are being made to obtain reinforced photocells from various sources. The moisture detector development has not been hampered by the deficiency of these sensors. Tests on light sources and windows were carried to over 2000 psi without failure.

A pressure test on the heater assembly is contemplated before final assembly of the first prototype.

Electronic circuits have been developed for light sources and photocells. The breadboard stage of the development is complete and prototype circuit fabrication has begun. Life testing of six light sources is in progress.

Electrical Penetrations

A preliminary survey was made to determine the number of containment penetrations and the types of wiring required, and a literature search concerning containment penetrations at other gas-cooled nuclear facilities was conducted.

Inquiries were sent to manufacturers of hermitically sealed electrical connectors to determine their specific products and capabilities, and the specific requirements were reviewed with those vendors considered best qualified. From these reviews, general agreement was reached concerning fabrication of prototype connectors to meet outlined requirements.

After this preliminary work, the test program and test specification were prepared. At the same time negotiations were conducted with vendors and purchase orders were issued for the fabrication of prototype connectors for use in the test program.

AUXILIARY EQUIPMENT AND SPECIAL TOOLS

The objective of this subtask is to establish the suitability of the emergency tools and the reactor viewing device for use in the plant by means of analysis and scale model or full-scale test.

A test program for the auxiliary manipulator and tools was outlined. The program consists of three phases: postconstruction calibration, functional testing in ambient air, and final functional testing in helium at shutdown temperatures.

Functional testing will be based on the types of manipulation required for predicted types of failure, such as dropped element handling and control rod servicing.

The radiation dosage which a viewing device will be subjected to during refueling was determined and work was begun to determine the cooling requirements for a television camera. It appears that an available radiation-hardened camera would require little, if any, shielding for the life expectancy required for this application. Indications are that electrical cooling by the Peltier effect is impractical because of its low efficiency.

Task II
NUCLEAR ANALYSIS

FUEL CYCLE ANALYSIS

The objective of this subtask is to establish the fuel management program for the reactor based on analyses of the effects of the loadings in the initial and replacement segments, including a recycle segment in an equilibrium core, on local power generation, reactor safety, and economics.

Maximum Fuel Burnup

Recent burnup calculations indicated that for the reference fuel cycle the fractional burnup (fissions per metal atom) in the fuel particles may be higher than originally assumed. The fractional burnup in the $(\text{Th,U})\text{C}_2$ particles can easily be controlled by an appropriate choice of the dilution factor. It appears that a Th/U ratio of 4.25 to 4.5 will be required to limit the maximum fractional burnup to 20%. However, the maximum burnup of the ThC_2 particles cannot be precisely controlled for a given fuel cycle and a maximum burnup of 7% has been specified instead of the current reference value of 6%. This may necessitate changes in the ratio of coating thickness to particle diameter for the ThC_2 particle, but these changes are expected to be neither large nor troublesome.

POWER SHAPING

The objective of this subtask is to establish the fuel and burnable poison distribution and the control rod programming based on analyses of the transverse and axial power distribution as a function of fuel and poison loading, burnup, and control rod positions and considerations of fuel temperatures and economics.

A region peaking factor of 1.83 has been assumed in the design of the primary coolant system. The fuel and poison zoning and the rod programming can be adjusted so that this factor is never exceeded in the operating reactor.

The gross power distribution with the control rods inserted to achieve criticality appears to be no worse than in the unrodded core. Not all rod-programming schemes are acceptable in this regard, however.

Maximum Region Peaking

As a consequence of the regional refueling scheme chosen, the power density in the core varies strongly from region to region. Without orificing, the power peaking limit is set by the maximum permissible fuel

temperature, and a considerable amount of axial zoning would be required to obtain optimum thermal performance in the core. The variations in region power density also result in gas exiting from some regions at temperatures that are considerably above the mixed mean outlet temperature.

With orificing, it is in principle possible to keep the gas outlet temperatures uniform across the core. Orificing, however, increases the pressure drop in the primary coolant system, and the power peaking limit in a region is thus determined by the pressure drop that is economically acceptable. Since the equipment of the primary coolant system, including steam generators and coolant circulators, has to be fixed and ordered long before the exact fuel loadings have been specified, it is important that a reasonable limit on the maximum region power density be set comparatively early. Taking into consideration the fuel cycle chosen for the plant and the power peaking inherently associated with this cycle, a conservative (but not overly so) design limit of 1.83 has been set for the region power density factor.

Power Distribution in Rodded Configurations

During the first two years of operation, and during any of the following yearly cycles, the maximum excess reactivity at operating temperature, after xenon and Pa^{233} have built up to their equilibrium values, will be about $0.03 \Delta k/k$. Burnable poison will be used to achieve this design goal. As many as 7 to 10 control rod pairs may be required to control the reactor during each cycle. In the previous quarterly report the power distribution in the unrodded initial core was described. To date, several rod patterns that yield both sufficient strength (i.e., up to $0.03 \Delta k$) and an acceptable power distribution (i.e., power density factor less than 1.83) have been investigated. Figures 2.1 and 2.2 show schematic core layouts of possible rodded configurations and the resulting transverse power distributions, i.e., the region peaking factors. In no case does the maximum region factor exceed 1.83. Figure 2.3 gives a summary of this data by showing the fraction of the initial core operating above any relative power density using the different rod patterns. These data are similar to data obtained earlier for the unrodded core.

Several rod patterns have also been investigated for the equilibrium core with results that are comparable to those presented above. These data will be refined and updated in the coming months.

KINETIC ANALYSIS

The objective of this subtask is to predict the required strength of the control system, the control rod worth, the magnitude of the temperature coefficients, and the kinetic behavior of the core including insertions of reactivity and spatial xenon stability, by means of analyses under normal operating and accident conditions.

Fig. 2.1--Core layouts of possible rod configurations and resulting transverse power distributions

SEVEN RODS INSERTED

$$\Delta k_{eff} = 0.027$$

PATTERN C

MAX/AVG POWER

REGION NO.

SEVEN RODS INSERTED

$$\Delta k_{eff} = 0.034$$

PATTERN D

Fig. 2.2--Core layouts of possible rod configurations and resulting transverse power distributions

Fig. 2.3--Fraction of core with power density greater than any specified value; initial core, rodged configurations

Control System

The worths of the rod patterns previously described are summarized in Table 2.1 for the initial core. Since the worth of the control system increases with reactor lifetime (see the previous quarterly report, GA-7086), these preliminary data indicate that seven control rod pairs provide sufficient strength to cover the expected reactivity swing of 3% during any refueling cycle.

As an alternative to the reference control rod system, the use of single large control rods has been investigated. Thirty-seven hollow control rods, each having a 7 in. OD and a 6 in. ID and containing an annular graphite spine, would provide the same total control rod worth as the presently used rod pairs. However, the increase in rod weight together with the increase in core leakage due to neutron streaming ($\approx 0.5\% \Delta k/k$), and the associated penalties in fuel cycle cost, make the single rod design less attractive than the reference system.

Table 2.1
CONTROL SYSTEM WORTH IN INITIAL CORE

Pattern	Regions Rodded	Worth (Δk)
A	Central region	0.005
B	Central region 3 regions in Ring 4	0.017
C	Central region 6 regions in Ring 4	0.027
D	Central region 3 regions in Ring 3 3 regions in Ring 4	0.034

Transient Analysis of Rod Withdrawal Accidents

The greatest amount of reactivity insertion and the largest reactivity insertion rate result from the accidental withdrawal of one control rod pair. Any transient initiated by an accidental rod runout will either be arrested by a rod withdrawal prohibit at 120% rated power or be terminated by an over-power scram at 140% rated power. It appears possible to increase the rod withdrawal rate to yield a rod withdrawal time of about 57 sec. A scram time of 48 sec is compatible with this withdrawal time.

Each control rod pair has a separate control rod drive mechanism. Gang withdrawal is prevented, and it is not possible to accidentally withdraw more than one control rod pair at a time. Gang insertion of the control rods is allowed for scram and reactor runback.

The maximum worth of a control rod pair for the operating core is calculated to be 0.015 Δk for any configuration. The rate of reactivity addition for a single control rod pair is shown in Fig. 2.4.

The minimum negative reactivity insertion during scram following a rod withdrawal accident is at least 0.09 Δk ; following a startup accident, the scram worth is at least 0.04 Δk . This takes into account the possibility of a stuck rod, the possibility of some rods being (initially) partially inserted, and the possible absence of xenon and Pa^{233} . Scram reactivity insertion versus time is shown in Fig. 2.5.

The severity of a control rod pair withdrawal accident is greatest in the equilibrium core, at the end of the yearly reactor fuel cycle, for two reasons:

1. The delayed neutron fraction is least, resulting in shorter reactor periods for the same reactivity insertion.
2. The temperature coefficient is least negative, because of the presence of equilibrium xenon, the buildup of U^{233} , and the decrease in the Th^{232} load.

Hence, the rod withdrawal transients have been studied in detail for the equilibrium cycle. Transient analyses were made for the case of one control rod pair withdrawn from the hot operating core and one control rod pair withdrawn from the core when at source power.

The initial core conditions for the transients, together with pertinent assumptions, are given in Table 2.2. The results of the specified reactivity accidents are described below.

If the transient caused by the withdrawal of the maximum-worth control rod pair during full power operation is terminated by the rod withdrawal prohibit at 120% power, the maximum fuel temperature increases by less than 70°C and the maximum coolant outlet temperature increases by less than 40°F. If the transient is terminated by the scram at 140% of full power, the maximum fuel temperature increases by about 150°C. The gas outlet temperature rises slowly during the transient, increasing by approximately 40°F. Neither of these transients produce conditions within the reactor that would present a hazard, either to the plant or the surroundings. The transients are shown in Figs. 2.6 and 2.7, and the pertinent results are given in Table 2.3.

Because of the very large negative temperature coefficient that exists at reduced temperatures, and because of the large heat capacity of the core, the transient behavior of the reactor to reactivity insertions at source-power level is less severe than it is at operating conditions. In the conservative case studied, the reactor is assumed to be critical at a very low power level, on the order of a watt. Pertinent results are summarized in Table 2.3 and in Figs. 2.6 and 2.7. In addition to the normal power level prohibit at 120% and scram at 140% of reactor operating power, intermediate level instruments are capable of initiating a scram at a much lower power on the basis of excessively short periods. If the intermediate range instruments initiate the scram, the transients are even less severe. The source range instruments have a period alarm only.

Fig. 2.4--Reactivity addition during withdrawal of maximum worth rod pair

Fig. 2.5--Reactivity insertion of scram system; fractional worth versus time after start of scram

Table 2.2

INITIAL CONDITIONS AND GROUND RULES FOR ROD WITHDRAWAL TRANSIENTS

	Source Power	Full Power
Average fuel temp., °C	200	804
Maximum fuel temp., °C	200	1128
Average mod. temp., °C	200	728
Maximum mod. temp., °C	200	767
Gas outlet temp., °C(°F)	200(392)	785(1445)
Power level, Mw(t)	10 ⁻⁶	833
Flow rate, % of full flow	10	100
Radial peak-to-average power factor	1.83	1.83
Worth of control rod pair, Δk	0.015	0.015
Time for withdrawal of rod, sec	57	57
Reactivity insertion rate	(a)	(a)
Minimum scram reactivity, Δk	0.04	0.09
Time for scram to fully inserted position, sec	48	48
Reactivity insertion rate	(b)	(b)
Scram set points:		
Overpower scram, % rated	140	140
Overpower rod withdrawal prohibit, % rated	120	120
Delay time, sec	0.2	0.2

^a See Fig. 2.4.

^b See Fig. 2.5.

Table 2.3
SUMMARY OF ROD WITHDRAWAL ACCIDENTS
(Equilibrium Core, End of Cycle)

	Rod Withdrawal Prohibit (120% Power)			Power Level Scram (140% Power)		
	Max. Fuel Temp. in Hottest Element (°C)	Avg. Mod. Temp. in Hottest Element (°C)	Max. Gas Outlet Temp. From Hottest Region (°F)	Max. Fuel Temp. in Hottest Element (°C)	Avg. Mod. Temp. in Hottest Element (°C)	Max. Gas Outlet Temp. From Hottest Region (°F)
Rod withdrawal from full power	1200	800	1480	1275	805	1485
Rod withdrawal from source power	465	340	485	475	310	475

Fig. 2.6--Maximum fuel temperature in hottest element during rod withdrawal transients

Fig. 2.7--Gas outlet temperature in hottest channel during rod withdrawal transient

Task III
FUEL DEVELOPMENT

FUEL ELEMENT DESIGN

The objective of this subtask is to develop the design of the fuel elements to be used in the plant by means of mechanical and flow testing, analysis of temperatures under normal and accident conditions, analyses of stresses, including effects of temperature, irradiation, and creep, and analysis of fission-product concentration in the reactor system.

Mechanical Design

Core Arrangement Design. Work continued this quarter to establish the over-all core restraint concept, side reflector design, and the keyed orifice plenum elements.

The 247 individual columns of fuel and reflector elements are arranged on a triangular array with a pitch spacing of about 14.2 in. As reported previously, the fuel columns are grouped into 37 refueling regions containing seven columns each, except for six outer corner regions which contain five columns each.

Each seven-column region of fuel elements rests on a single large hexagonal graphite core support block. The core support blocks are arranged on a uniform triangular pitch of about 37.6 in. and are keyed together to provide a uniform pitch spacing at the core support plane. The fuel and reflector columns are connected to the core support blocks by graphite dowels to provide lateral restraint and column alignment at the bottom of the core.

Alignment and lateral end restraint of all columns at the top of the core are provided by the top reflector elements, which are keyed together. Keys extend the full length of the element and are arranged perpendicular to the six flat surfaces. The boundary blocks are keyed to the core barrel structure so that at the top and bottom planes of the core every column is effectively fixed relative to the core barrel and support structure. A schematic elevation view of the core showing the top and bottom key arrangement is shown in Fig. 3.1.

The use of the top and bottom keyed arrangement provides a stable means for establishing the pitch spacing of the columns during both reactor operation and shutdown conditions. This arrangement accommodates differences in radial and axial thermal expansion between the core barrel and the core graphite without loss of lateral restraint at each end of the individual columns. The top reflector key elements will allow unrestrained axial movement of individual columns to account for differences in thermal expansion or irradiation-induced contraction of the graphite. The elements within the

Fig. 3.1--Core restraint arrangement

center column of the region are displaced axially downward about 7.5 in. relative to the elements in the surrounding six columns, thereby removing the possibility for a flat plane of shear to exist at element interfaces across the core.

Orifice Assembly. A variable-orifice flow control assembly is located at the inlet to each refueling region to provide for the adjustment of the coolant flow through the region. Changes in fission power generation as a result of fuel burnup and fuel age differences can be compensated for by orificing the coolant flow rate to each region. The flow is controlled by actuating a parted cylindrical orifice valve about 17.5 in. in diameter located over a circular opening at the center of the inlet plenum of each region. The plenum is formed by the surrounding array of six keyed elements. The center column in the region is displaced axially downward about 7.5 in. to expose sufficient flow area to each column. The orifice valve and flow paths are shown in Fig. 3.2.

The orifice valve is mounted on the control rod guide tubes, which engage with the topmost element in the center column of each region. The control rods pass through the guide tubes. The upper ends of the guide tubes are positioned within the control rod drive assembly and provide a stable structure for lateral positioning and support of the orifice valve. A vertical shaft driven from the orifice drive mechanism actuates the orifice valve.

Reflector Elements. The top and bottom reflector elements have the same basic configuration as the fuel elements but do not contain fuel beds. The reflector elements have the same pattern of coolant and control rod channels as the fuel elements within the same columns, except that the bottom reflector elements within each fuel column provide a transition of the coolant channels from the distributed array within the fuel elements to a single large hole that mates with the coolant channels within the core support blocks. Alignment of the fuel and reflector columns with the core support blocks is maintained by dowels on the bottom reflector elements, which fit into the core support blocks. The topmost element of each column within the core is a metal box structure that serves as a plenum for inlet coolant but also contains reflector graphite. Vertical metal keys and keyways are provided on the side faces of each box structure to provide lateral positioning relative to the core barrel structure.

As shown in Fig. 3.3, the first two rings of side reflector columns are of the typical hexagonal-block shape. The remaining side reflector consists of large graphite blocks between the outermost columns of hexagonal-block elements and the core barrel.

The hexagonal side reflector elements surrounding the core have the same envelope dimensions as the fuel element but are solid graphite and contain only a central hole for removal and insertion.

Handling Hole Lifting Ledge Impact Strength Test. The test program to determine whether the lifting ledge within the fuel element handling hole will withstand potential shock loads experienced during grappling by the

Fig. 3.2--Orifice assembly

NOTES

1 FUEL ZONE BOUNDARIES

2 FUEL REGION BOUNDARIES

3 CONTROL ROD COLUMN

SHADED REFLECTOR ELEMENTS
ARE NORMALLY REPLACED WITH
ADJACENT FUEL REGION

Fig. 3.3--Core arrangement

fuel handling machine grapple head has continued this quarter. (A six-year-old block, if momentarily restrained by adjacent elements, could fall from a height of 13/16 in. as a result of operating clearances, overtravel allowances on the fuel handling machine, and the calculated graphite contraction.) During the quarter the first lifting ledge impact strength test was performed using TS-688 graphite with an eccentric ledge grapple tool concept. Drops were made from 1/4, 1/2, and 3/4 in. with the total combined weight of the jig and graphite sample being 146.2 lb. In the 1/4 in. drop a slight (≈ 0.16 in.) crushing of lifting ledge graphite was evident. In both the 1/2 and 3/4 in. drops small pieces of graphite were broken from the inside corner surface of the graphite ledge. However, the integrity of the lifting surface was maintained.

Further lifting ledge load tests are planned using an alternative grapple tool design. A prototype is being fabricated for load testing.

In addition, a direct tensile test is planned to determine the maximum tensile force that can be applied before the graphite fuel element fails. This value can then be used to determine the maximum force that could be applied in an attempt to free a hypothetically stuck fuel element.

Locating-Dowel Shear Strength Tests. Seven tests were performed during the quarter to determine the failure strength and fracture mode of the locating dowel or side wall of the fuel element when subjected to gradually applied horizontal shear loads. The results of the seven tests, which were performed with TS-688 graphite specimens, are shown in Table 3.1.

The first three tests were performed with socket specimens that had the fuel holes drilled completely through the specimen with the exception of those directly behind the socket. Also, the socket specimen was machined with a 90° corner at the bottom of the socket to determine whether special shapes such as a chamfer or radius would significantly increase the strength as determined in subsequent tests. In all the seven tests, failure occurred in the side wall between either the socket or the locating pin and the nearest face of the fuel element, as shown in Fig. 3.4. No failures occurred in the dowel pin. During all tests the parallel graphite faces were maintained about 0.01 in. apart to eliminate friction.

Table 3.1
RESULTS OF SHEAR STRENGTH TESTS

Test No.	Shear Force at Failure(lb)	Side Wall Failure Between Dowel and Fuel Element Face	Side Wall Failure Between Socket and Fuel Element Face
1	1311	Yes	No
2	1081	No	Yes
3	956	No	Yes
4	1036	No	Yes
5	1185	No	Yes
6	1223	Yes	No
7	1242	Yes	No

Fig. 3.4--Fuel element dowel shear test arrangement

The fuel holes in the socket specimens used in tests 4 through 7 were not drilled completely through the face of the specimen to simulate the reference reference fuel element design, and a 3/8-in. radius was incorporated at the bottom of the socket to reduce local stress concentrations. The dowel specimens were identical to those used for the first three tests.

A second phase of dowel shear strength tests to evaluate the impact strength of the fuel element dowels was initiated this quarter. The graphite specimens used for the first three shear tests were salvaged for use in this test. In addition, with minor modifications, the same shear test fixture was usable for these tests.

The back side of the four graphite specimens used for the first three shear tests were machined to make two new dowel pin specimens and two elongated slots for testing the weakest side of the socket specimen. A 3/8-in. radius was used at the bottom of the new socket specimens to limit the stress concentrations at the inside corners.

Four tests were performed this quarter with the results shown in Table 3.2. In all four tests it again was the side wall between either the socket or the locating pin and the nearest face of the fuel element that failed. No failure occurred in the locating dowel pin itself, with the exception that some of the graphite threads were broken off at the thread roots in certain areas (engagement areas between the dowel pin threads and the corresponding threads in the fuel element).

Table 3.2
RESULTS OF IMPACT TESTS

Test No.	Impact Weight on Dowel or Socket (lb)	Drop Height Producing Failure (in.)	Prior to Failure, Impacts were made onto the Dowel or Socket from these Successive Heights (in.)	Side Wall Failure occurred between Dowel or Socket and Fuel Element Face
1	97.8	0.821	0.250, 0.393, 0.536, 0.678	Dowel
2	97.9	0.678	0.250, 0.393, 0.536	Socket
3	97.9	0.821	None	Socket
4	97.5	0.928	None	Dowel

The primary purpose of these tests was to demonstrate experimentally that the dowel pin arrangement will withstand significant seismic or other unexpected impact shear loads experienced within the core.

Lateral Column Deflection Test. Deflection characteristics of a multi-element column subjected to varying lateral load distributions caused by fluid or seismic forces are being experimentally determined using a one-quarter scale column of 16 simulated fuel elements, with no lateral support except at top and bottom. The information will be used to develop improved analytical models for describing the equivalent stiffness and deflection characteristics of the multielement articulated column for core stability analysis.

Four tests were run during the quarter. In two tests, lateral loads of 5% of each element weight were applied to each of the 16 elements. A 25-lb weight was applied to the top of the column in one of these tests to approximate the pressure drop through the PSC core. The two tests were then repeated with 10% lateral loadings. The results are given in Table 3.3. Testing will be continued during the next quarter, with lateral loadings of 20%, 30%, and 50%. The development of the analytical model is in process.

Table 3.3
RESULTS OF LATERAL DEFLECTION TESTS

Lateral Loading (%)	ΔP Weight (lb)	Maximum Deflection (in.)	Location of Max. Deflection ^a Element
5	None	0.010	2 and 5
5	25	0.009	2, 3 and 4
10	None	0.032	5 and 6
10	25	0.017	3

^a Element number from top of column.

Multicolumn Flow and Vibration Test. Work has continued during the quarter to establish the test program details and the preliminary design of the multicolumn air flow test rig. Preliminary design of the test rig is nearly complete, and fabrication of the rig and auxiliary equipment will be initiated next quarter.

Fuel Element Thermal Stress Analysis. Preliminary radial and axial thermal operating stresses at the location of maximum temperature gradient between the fuel and coolant hole were calculated. The maximum temperature gradient occurs at the bottom of the upper axial fuel zone in the maximum power region at the end of the first year. The radial thermal stresses were calculated with the SAFE-PLANE code using irradiated TS-688 graphite material properties. The maximum stresses in the radial or R- θ plane occur in the theta direction. A maximum theta tensile stress of 274 psi occurs next to the coolant hole, and a maximum theta compressive stress of 267 psi occurs next to the fuel hole. The maximum radial stresses are 89 psi tensile and 47 psi compressive. The maximum axial stresses, approximated by one-dimensional calculations, are 295 psi tensile next to the coolant hole and 250 psi compressive next to the fuel hole.

Thermal Design

The present reference design carbon-to-thorium ratio (C/Th) is 225 for the equilibrium core fuel loading. Core flow rates, temperatures, and pressures that have been reported previously all refer to the C/Th = 200 core loading, which has somewhat lower region peaking factors and corresponding fuel temperatures than the current reference design utilizing the lighter fuel loading.

During the past quarter, new thermal and hydraulic analyses were performed from which the following information has been obtained.

Core Coolant Flow Distribution. The heat energy produced within the active core and reflectors is removed by a downward flow of helium coolant. The total core coolant flow rate is 3.410×10^6 lb/hr. A portion of this flow bypasses the active core region through the clearances between the side reflector columns. The side reflector coolant flow, which is dependent on the core pressure gradient, is calculated to be about 5% of the total reactor flow rate under normal full-power operating conditions. It removes an estimated 1% of the total core thermal power generated within the side reflector elements.

To cool the control rods, about 4% of the reactor flow enters the control rod guide tubes through individual orifices that are set to allow an equal gas flow into each tube. This flow, in passing through the control rod channels within the center column of each region, removes heat from the fuel elements as well. It merges with the gas flow from the coolant channels within the elements upon entering the lower plenum.

The power distribution among the 37 regions of the active core requires a controlled coolant flow rate through each region to satisfy the objective of uniform gas exit temperature. This is obtained by appropriate orificing.

Adjustment of the orifice valve position controls the flow rate entering the region. The required flow rate ranges from 160,000 lb/hr through the maximum-power region ($P/\bar{P} = 1.8$) to about 50,000 lb/hr through a low-power region ($P/\bar{P} = 0.5$) in the core. The maximum flow rate, since it depends on the fraction of total core power produced within the region, is needed for only a brief period toward the end of the annual refueling cycle when the region age peaking factor is greatest. The annual average of the maximum power region flow rate is 140,000 lb/hr for equilibrium core conditions.

The maximum flow rate of 160,000 lb/hr of helium through a fully open orifice causes a pressure loss of about 1.2 psi. For a low-power region, where the orifice valve opening is reduced to allow only 50,000 lb/hr to pass, the resulting orifice pressure drop is about 8 psi.

The maximum calculated plenum-to-plenum core pressure drop is 8.3 psi. Included are the loss across the inlet orifice, the pressure drop in the channels resulting from friction and momentum changes, and local pressure losses occurring at the coolant channel interfaces between adjacent fuel elements within each column. The latter losses account for any slight

coolant channel offset at element interfaces due to machining tolerances and small distortion differences during irradiation.

Within an orificed region some flow bypasses the internal coolant channels through the clearance between adjacent fuel element columns. The width of the local gaps includes the combination of design assembly clearances, relative thermal expansion and irradiation-induced contraction of the fuel element graphite. The calculated bypass flow through the gaps between fuel columns ranges from 2-1/2% to 4% of the region flow rate, the smaller number applying to new fuel elements. This flow removes heat from the side surfaces of the fuel elements and reduces the local fuel temperatures. However, no credit is taken for heat removal by the flow between columns in the peak fuel temperature calculation to account for possible local closure of the gaps.

The general effects of crossflow on the coolant distribution have been discussed in detail in the previous quarterly. A computer program for analysis of multiple parallel flow channels with multiple interconnects is 90% completed.

Thermal Performance. Because of coolant orificing, the bulk of the active fuel spans a relatively narrow temperature range. Figure 3.5 shows the percentage of total core fuel volume that operates above a given temperature at rated design power. Only about 2% of the fuel operates above 1800°F. Moreover, a specific fuel element is generally at the highest temperature for less than one year of the six-year fuel life.

Although the mixed-mean temperature at which the coolant leaves a region is 1456°F, the gas temperature within individual coolant channels varies across some elements because of local edge peaking or power depression, depending upon the age of immediately adjacent fuel columns. The coolant exit temperature within the maximum power region varies from a minimum of 1340° to a maximum of 1670°F. The latter value is for channels located near the side reflector where the local heat generation rate is about 2.3 times the core average, as compared to a region average $P/\bar{P} = 1.8$, at the time in life of maximum power generation.

The maximum fuel temperature of 2300°F occurs within the maximum power region near the axial midpoint of the active core, where the axial peak-to-average power ratio is about 1.4. The maximum graphite temperature is 1860°F and occurs near the active core exit of the maximum power region. Figure 3.6 shows a typical radial temperature distribution through the active fuel and graphite heat-transfer path to a coolant channel surface. Thermal performance and coolant flow conditions for full-power operating conditions are summarized in Table 3.4.

Hot-spot Evaluation. The analysis of the fuel temperatures is subject to uncertainties in the many variables affecting the core performance. The deviations from the nominal fuel temperatures that result from these uncertainties have been calculated and are summarized in Table 3.5. The design and operational variations are discussed in the text that follows.

Fig. 3.5--Fuel volume temperature distribution

Fig. 3.6--Typical radial temperature distribution in a fuel element

Table 3.4

THERMAL PERFORMANCE AND COOLANT FLOW CONDITIONS AT FULL POWER

Fuel Temperatures

Maximum temperature, °F.....	2,300
Volume median temperature, °F.....	1,500
Minimum temperature, °F.....	900

Moderator Temperatures (Active Core)

Maximum temperature, °F.....	1,860
Volume median temperature, °F.....	1,350
Minimum temperature, °F.....	900

Surface Heat Flux at Coolant Channel Surface

Maximum flux, Btu/hr-ft ²	120,000
Average flux, Btu/hr-ft ²	45,000
Minimum flux, Btu/hr-ft ²	14,000

Coolant Flow Characteristics

Maximum Reynolds number.....	70,000
Average Reynolds number.....	25,000
Minimum Reynolds number.....	12,000
Max. surface heat-trans. coeff., Btu/hr-ft ² -°F.....	400
Avg. surface heat-trans. coeff., Btu/hr-ft ² -°F.....	230
Min. surface heat-trans. coeff., Btu/hr-ft ² -°F.....	120
Core inlet pressure, psia.....	700
Total core pressure drop, psi.....	8.3

Table 3.5

DEVIATIONS FROM NOMINAL FUEL TEMPERATURES

<u>Variable</u>	<u>Fuel Temp. (°F)</u>
1. Dimensional tolerances.....	17
2. Material properties:	
Fuel conductivity (-10%).....	66
Graphite conductivity (14 Btu/hr-ft°F).....	21
3. Heat-transfer correlation uncertainties.....	0
4. Fuel loading tolerances (+6%).....	65
5. Radial power peak (+10%).....	154
6. Axial power peak (+10%).....	74
7. Exit coolant temp measurement error.....	20
Theoretical total increase	417
Theoretical hot-spot temp. (if applied at point of max. fuel temp.)	2717

1. Fuel Element Dimensional Variances. The nominal coolant channel diameter is 0.625 in. A conservative minimum diameter to account for machining tolerances of 0.620 in. was evaluated. Dimensional tolerances on web thickness between fuel beds and coolant channels will have a very small effect on fuel temperature.
2. Uncertainty in Thermal Conductivity. An irradiated graphite thermal conductivity value of 14 Btu/hr-ft-°F representing a 12% reduction from the design value was selected as a lower limit for hot-spot calculations.

The fuel bed material utilizes a temperature-dependent function for the thermal conductivity. This function was reduced by 10% for the hot-spot calculation to represent the lower limit of measured values.

3. Uncertainty in Heat-transfer correlation. The conventional Dittus-Boelter correlation for fully developed turbulent flow conditions was used for computing the surface heat-transfer coefficients. The validity of the correlation is well established, and in the PSC reactor core, the applicable range of the correlation falls within the limits experienced during normal operation. Any local disturbances in the flow caused by slight coolant channel offset at adjacent element interfaces will increase the coefficient relative to the calculated value for the remainder of the channel length within the element. It is concluded that any uncertainties in the predicted film coefficient, if evaluated, would yield higher values and would therefore reduce the calculated fuel temperature levels.
4. Fuel Loading Tolerances. At the location of maximum core power, an increase of 6% in power corresponding to the maximum local fuel load-tolerance was evaluated.
5. Uncertainty in Radial Power Peaking Factor. For hot-spot analysis, a maximum channel power density over the entire core length of 10% higher than predicted was used to conservatively account for uncertainty in the edge peaking factor values used for design.
6. Uncertainty in the Axial Power Peaking Factor. The power peak at the axial fuel zone boundary was increased 10% above the design value for conditions within the maximum power region to conservatively account for uncertainties in the local neutron flux peaking.
7. Exit Coolant Temperature Measurement Tolerance. Coolant flow through the orifice assembly is controlled by sensing exit coolant temperature from each region. A measurement error tolerance of $\pm 20^\circ\text{F}$ is allowed for sensing inaccuracy.

The maximum fuel temperature is calculated to be 2300°F. If the very conservative assumption is made that all of the hot-spot factors could physically combine at the point where the maximum fuel temperature occurs, the extremely unlikely but safely acceptable maximum fuel temperature would be 2717°F.

Offset Loss Coefficient Test Program. A portion of the presently accepted core pressure drop results from assumed losses that are due to coolant channel misalignment at the interface of adjacent fuel elements within a column. Based on analytical predictions, a loss coefficient of 0.2 has been applied at each interface. There are two sources of uncertainty in the selection of a loss coefficient. First, it is difficult to predict the losses even if the amount of channel offset is accurately known, and second, the distribution of coolant channel offset throughout the core depends on machining tolerances, differential thermal expansion, and irradiation-induced contraction of the graphite blocks.

To improve the accuracy of the core pressure drop analysis, an experiment has been designed whereby the loss coefficient is determined for accurately measured misalignments ranging from 1% of the channel diameter up to complete misalignment. The test unit contains four sets of matching tubes having inside diameters ranging from 0.5 to 1.0 in. Airflow rates through the tubes are to be measured with a high-precision rotameter. Pressure taps along the length of the tubes allow the loss at the offset to be determined. Results of the test are expected within the next month.

Crossflow-Coefficient Test Program. To predict the flow distribution within the core with crossflow occurring at the element interfaces within the columns, it is necessary to know how crossflow relates to the pressure difference between fuel channels within an element and the surrounding gap. It is planned to obtain this loss coefficient experimentally.

If the static pressure of the flow in the surrounding gap is less than the static pressure of the flow within a fuel element, the amount of crossflow leaving the coolant channels depends on the crossflow resistance and the driving pressure gradient. Presumably, most of the crossflow comes from coolant channels near the element periphery. Resistance to crossflow arises from (1) bending losses, when some of the gas flowing down a coolant channel makes a 90° turn at the element interface, (2) friction in the small interface gap, and (3) bending losses, when crossflow enters the gap around the element and makes a 90° turn. For elements with a smooth, flat interface surface, the loss under item (2) probably determines the over-all resistance. If the element surfaces are distorted and a substantial clearance exists over most of the interface, the bending losses, items (1) and (3), may be the important ones.

Crossflow is not necessarily from each coolant channel to the gap surrounding the element or vice versa, but may well be among several coolant channels individually. It is believed that the important parameters determining crossflow resistance are (1) the flow Reynolds number in the channels and (2) the interface gap height.

Because of the apparent complexity of the problem, it is planned to measure the net amount of flow, ΔW , entering or leaving the fuel element column at an interface between two elements under various conditions of pressure gradient, ΔP , between inside and outside of the element, flow rate through the channels, and interface gap height, s . These quantities may then be related as follows:

$$\Delta P = \frac{K(Re,s)}{2\rho g} \frac{\Delta W^2}{A},$$

from which $K(Re,s)$ may be determined once a suitable area, A , has been defined.

The proposed test setup consists of two full-scale fuel element sectors, arranged along the same axis. Instead of using an entire hexagonal element, it is planned to use only a 1/6 symmetrical section of the element. At the interface, two sides of the resulting equilateral triangle are sealed off while the third is exposed to a gap that runs along one side of the element and is formed by mounting a plate parallel to the side at some small distance simulating the expected clearance in the reactor. The graphite test elements are machined to the actual fuel element specifications, so the condition of the interface at the beginning of fuel life may be duplicated accurately. Distortions of the interface are simulated by introducing small spacers between the blocks at the interface or by tilting the blocks and, if necessary, remachining the end surfaces.

The relative flows entering and leaving each of the 21 coolant channels in the test section will be observed using pitot tubes. The system allows an over-all flow mass balance to be made and provides valuable information regarding crossflow among coolant channels individually, which is important for hot-spot calculations.

Multichannel Flow Analysis Code. Development of the PICA code for the analysis of flow distribution and pressure drop through multiple parallel flow channels with interconnections continued during the quarter. Late in the quarter, several simplified test problems were successfully completed using this code, but when a large problem was attempted for final debugging, instability occurred. The convergence criteria and iteration procedures are currently being reviewed.

Fission-product Transport

Primary Coolant Fission-product Activity. The fission-product activity and inventory of the primary coolant system were revised during the quarter to account for the increased PCRV helium inventory (5125 lb) and system pressure (700 psia) and to incorporate current estimates of isotope plateout distribution on particulate matter, graphite surfaces, and metallic surfaces.

The gas-borne and plateout primary coolant system fission product activities were calculated with the RAD-2 and FREVAP codes. FREVAP was used to determine the direct fuel element release of Sr^{89} , Sr^{90} , Cs^{137} , Ba^{140} , Sm^{151} , and Sm^{153} . The contribution to the activity of these isotopes from precursor decay was calculated by RAD-2 as were the gas-borne and plateout activities for the other species. The RAD-2 calculations are based on a single-valued over-all release fraction from the fuel element for each isotope assuming a noble gas release behavior. While this method of calculation by RAD-2 is expected to be conservative for non-noble species, improved

calculational methods are being studied to consider the adsorption characteristics of the fuel element graphite.

Release data for the RAD-2 calculations were evaluated from the GAIL experiments. In these experiments fission product activity levels were measured during the term of the experiment for the isotopes $\text{Kr}^{85\text{m}}$, Kr^{88} , Kr^{87} , Kr^{89} , Xe^{133} , Xe^{135} , and Xe^{138} . The GAIL IV release data are typical of retentive coated particles and are used as representative of the expected retention characteristics of coated particles for the gas-borne and plateout fission-product activity calculations. The GAIL III-B experiment contained particles with an early type of nonretentive coating and experienced a high rate of coating failure. The GAIL III-B release data were used for calculating design gas-borne and plateout activities resulting from particles with failed coatings.

The model used for the FREVAP calculations described the core as 37 regions divided into five radial zones. The core center region was represented as Zone 1; Zone 2, the first ring of the core, contained six regions; and Zone 3, the second ring of the core, contained 12 regions. Zones 4 and 5 comprised the outer ring of the core: Zone 4 representative of the 12, seven-column regions, Zone 5 representative of the six, five-column regions. For each zone, 16 axial segments were defined corresponding to the two bottom reflector elements, 12 fueled elements and two top reflector elements of a column. With this basic model, the temperature, power, and graphite mass could be specified for each reflector and fuel element. The primary coolant flow and power distribution for each region was specified as a function of region age.

The FREVAP code was used to calculate the direct release from the core for the isotopes Sr^{89} , Sr^{90} , Cs^{137} , Ba^{140} , Sm^{151} , and Sm^{153} considering the coated particle retention of these isotopes and the fuel element graphite adsorption and subsequent evaporation from the graphite surfaces. Gas-borne and plateout primary coolant system activities were evaluated using the FREVAP direct release and RAD-2 precursor decay for the total activity and subjecting the total activity to the appropriate removal mechanisms and decay. Calculations were done for an equilibrium core with the transverse power distribution and age peaking for a carbon to thorium ratio of 200. The two-fuel-zone modified cosine axial power profile with core coolant flow orificing was used.

Removal of Primary-coolant-entrained Particulate Matter. A possible mechanism for the removal of particulate matter from the primary coolant is by filtration from the helium permeating the core graphite. The driving forces for the permeation of helium through the graphite are the axial and radial pressure distributions throughout the core. Although the filtering efficiency of TS-688 graphite is a significant factor to be defined, the objective of this study was to determine the limits of the helium permeation flow.

Helium coolant may permeate the core graphite because of the axial pressure differential across the core (Δp about 8.6 psi) and because of the local pressure differential between fuel element coolant channels of a column and

the coolant bypassing the elements through the column gaps. From this study the axial permeation flow was found to be insignificant because of the 21-ft core height over which the axial pressure differential would be applied as a driving force. However, the radial permeation flow occurring through the nominal 3/16-in. web thickness between the fuel element outer face and coolant can be significant.

Radial helium permeation of core graphite will vary throughout the core as a function of the local pressure in the column gap adjacent to a fuel element and the local coolant pressure within the outer coolant channels of each element. Since core regions will be orificed by age to produce a uniform coolant outlet temperature, the coolant channel local pressure will vary by the region age. The gap local pressure axially will be dependent on the core inlet-to-outlet pressure differential. Gap local pressure for this study was assumed to be uniform across the core, but an upper and lower limit was estimated as a function of gap crossflow. As a lower limit, it was assumed that part of the available gap axial pressure differential could be spent as a driving force for crossflow in the gaps for helium bypass around flow orifices of the older core regions. It was assumed that the crossflow would occur at the top of a column within the length of the first two inlet reflector elements. This effect results in a lower gap local pressure in the fueled part of the core and establishes the lower limit of the pressure driving force for helium permeation flow.

The results of this study showed that the radial flow of helium permeating the fuel element graphite would have an upper limit of 3% of the total core coolant and a lower limit of 0.7% when considering gap crossflow and orifice bypass. This amount of flow would be significant as a naturally occurring filtering mechanism for the removal of particulate-borne fission-product activity in the primary coolant.

MATERIALS

The objectives of this subtask are:

1. To determine the characteristics of beds of loaded coated particles by means of fabrication technique investigations, bonded thermal property tests, packing and impact tests, and measurements of packing density and loading uniformity.
2. To select the type of particle bed to be used in the reactor based on the above work and work carried out in other parts of the program, taking into account the expected characteristics and relative ease of reprocessing, and to determine the suitability of the selected system by means of further tests of the type described above.
3. To determine the characteristics of various candidate graphites for use in the reactor fuel elements by means of measurements of the physical, thermal, and mechanical properties of these graphites.
4. To determine the characteristics of various control materials by means of measurements of physical, mechanical, chemical, and thermal properties.

5. To determine the irradiation stability of candidate graphites for use in the reactor fuel elements by means of irradiation tests conducted in the Engineering Test Reactor at the temperatures and to the integrated fast flux expected in the reactor, followed by post-irradiation examination. Approximately five capsules are planned for this part of the program.
6. To establish the effect of fast flux on fuel particle coating structural integrity as a function of coating structure and exposure, by means of irradiation tests in the Engineering Test Reactor on unfueled samples of particle coatings, postirradiation measurements of coating properties such as dimensional changes, flexural strength, and crystallite spacing, and subsequent reirradiation to design integrated fast flux and remeasurement. These irradiations will be carried out in the graphite capsules used in item (6) above.
7. To determine the irradiation stability of coated fuel particles for use in the reactor by means of a series of (1) short-term tests on coated fuel particles to select a coated fuel particle(s) having over its design life a high degree of structural integrity suitable for use in a nonvented fuel element, followed by (2) longer-term irradiation tests conducted at the temperature and to design burnup and to a significant fraction of the integrated fast flux expected in the plant. The "significant" integrated fast dosage will be determined from the results obtained in item (6) above. Capsule tests will be followed by postirradiation examination of the particles. Approximately seven short-term and three longer-term capsules are planned for this part of the program.

Particle Fuel Systems

Fuel development studies in support of the PSC fuel materials program are intended to provide economical candidate fuel systems that will meet the performance requirements of the Public Service reactor. The reference fuel element includes retentive bonded beds of coated particles in an unpurged element. The bonded beds should retain mechanical integrity during fuel life, be compatible with the graphite element during fuel life, and be amenable to fuel reprocessing at the end of life.

Bonded Particle Systems. Bonded particle fuel systems are employed to avoid or minimize particle spillage should a fuel element crack. Several binders and bonding processes are being investigated in an attempt to provide an adequate bond between coated particles while incorporating a high loading of sorbent material (e.g., activated charcoal or carbon black) for the retention of metallic fission products.

Several activated charcoals were characterized to aid in the selection of a suitable sorbent. These data (see Table 3.6) indicate that types MI-1 and XZ-1 provide high loadings. However, MI-1 is a coarse granular charcoal that has to be ground to a fine powder for incorporation into the fuel column, and both MI-1 and XZ-1 have a high level of impurities. The maximum loading

Table 3.6
DESCRIPTION OF CHARCOAL PROPERTIES

Charcoal	Particle Size	Level of Significant Impurities (ppm)	Nitrogen BET Surface Area (m ² /g)	Maximum Loading for 64% Coated Particle Packing Fraction (lb/element)	Sorbent BET Surface Area per Element (m ² /element)
MI-1	Large granules	Al-80; B-20; Ba-30; Ca-100; Fe-150; Ni-40; Si->1000; Ti-40; Zr-10	879,913 ^a	3.3	1.35x10 ⁶
XZ-1	Fine powder ^b	Al-100; B-4; Ca-100; Fe-100; Si-600; Ti-20	1165 ^c	3.7	1.95x10 ⁶
135	Fine powder ^b	Al-10; Fe-20; Si-100	980 ^c	---	---
AC-4	Large granules	Not tested	947 ^c	---	---
PCB	Large granules	Not tested	438,481,406 ^a	---	---
N.F.	Fine powder ^b	Not tested	456,438 ^b	2.3	0.47x10 ⁶

^a 1600°C outgas.

^b Small enough to pack into interstices of close-packed reference particles.

^c Routine 500°C outgas.

for type 135 has not yet been established; however, preliminary tests have indicated that it will fall between that of MI-1 and XZ-1. Therefore type 135 is presently the prime candidate sorbent.

The effects of processing on the sorptive properties of some charcoals were studied. The results of nitrogen BET and Cs adsorption measurements are shown in Table 3.7. An outgas at 1600°C causes a 20% to 30% reduction in sorptive properties. The incorporation of furfuryl alcohol binder in the admix process (hand blended) caused a significant reduction in BET surface area (mainly as a result of the presence of low surface area binder in the sample), but pressure impregnation of furfuryl alcohol and pitch resulted in an extremely high reduction in BET surface area. The charcoal surfaces were apparently still available to Cs, since impregnation caused only a moderate reduction (15% to 40%) in Cs adsorption. Therefore nitrogen BET cannot be relied upon to predict the sorption of fission products in bonded charcoal. Some of the Cs adsorption data for AC-4 are given in Table 3.8. Six specimens have been prepared for further Cs and Sr adsorption measurements.

Table 3.7
EFFECT OF PROCESSING ON SORPTIVE PROPERTIES OF SOME CHARCOALS

Process	Charcoal	Reduction in Sorptive Properties (%)	
		BET Surface Area	Cs Adsorption at 900° and 1000°C
1600°C outgas	N.F.	25	---
	AC-4	32	30
	135	19.4	---
	XZ-1	18.5	---
Hand blending with furfuryl alcohol, polymerizing, carbonizing at 900°C	N.F.	68	---
Impregnation of furfuryl alcohol at 150 psig, polymerizing, carbonizing at 900°C	N.F.	98.5	---
	AC-4	97	20
	AC-4	99	15-40
Impregnation of pitch at 150 psig, carbonizing at 900°C	AC-4	99	15-40

Table 3.8
Cs ADSORPTION IN AC-4 ACTIVATED CHARCOAL

Charcoal Temp. (°C)	Cs Press. (atm)	Adsorbed Cs (mg Cs/g Charc)
900	10^{-4}	250
900	10^{-7}	118
1000	10^{-4}	218
1000	10^{-7}	82

Several fabrication techniques were investigated for preparing bonded particle columns containing charcoal. In the first method, admixtures of furfuryl alcohol, charcoal, and reference particles were loaded into molds for the preparation of fuel sticks that were loaded directly into graphite tubes after carbonization. Qualitative observations of the carbonized fuel columns revealed that prepolymerization of the furfuryl alcohol resulted in a binder of good mechanical integrity.

In the second method, the admixture was tamped into the fuel hole and carbonized. This resulted in weak layers at the boundary where a new charge was placed over a tamped surface. Substitution of vibration for tamping produced low packing fractions at 1 g. Further tests at higher g-levels were more successful.

In the third method, charcoal-coated particle blends were loaded into a split mold for bonding. The two halves of the mold were held 0.002-in. apart by a shim, and the assembly was soaked in a furfuryl alcohol bath. The furfuryl alcohol was drawn into the fuel column through the 0.002-in. gap by capillary attraction. Since this process does not involve pressure impregnation, the matrix was expected to retain a large portion of its BET surface area. After 18 hr, the fuel column was completely impregnated. The polymerized sample broke during removal from the mold. Another sample was prepared using prepolymerized furfuryl alcohol in an attempt to increase its strength. Impregnation of the sample was incomplete because of the high viscosity of the resin; however, regions of good impregnation were very strong and hard. In a further attempt, a sample was exposed to the prepolymerized furfuryl alcohol for a considerably longer period (7 days); however, impregnation was still incomplete.

A fourth fabrication method involved spray overcoating the coated particles with a pitch-charcoal mixture and then warm pressing the particles into pellets. Several batches of particles were coated in a rotating hexagon shaped drum using a spray gun. The following methods were used:

1. The cascading particles were sprayed with a slurry of pitch dissolved in trichloroethylene.
2. Item (1) was performed, but powdered charcoal was added to the cascading mix.

3. The fuel particles were cascaded together with weighed amounts of powdered pitch and charcoal. The mix was simultaneously sprayed with trichloroethylene. (In methods 1 through 3, dry ice was tumbled with the particles to prevent sticking and agglomeration.)
4. The fourth and most successful method involved spraying the heated cascading PyC coated particles with a slurry of pitch and charcoal in a 1:1 solution of trichloroethylene and acetone.

Since the pitch-charcoal mixture dries upon contact, it was found that the overcoat could be applied at the rate of approximately 1 to 1-1/2 wt-%/min of steady spraying if care was taken to avoid agglomeration. It was also found that the PyC-coated particles could be completely separated from the pitch-carbon coating for reuse by submerging them in trichloroethylene for 5 min in an ultrasonic cleaner.

Attempts to warm-press overcoated particles at 200°C have been unsuccessful because of particle breakage and subsequent hydrolysis of the ThC₂ fuel particles. Vibration compaction at 8 g and 200°C in a 5-in.-long tube produced a column having good mechanical integrity and a 57% packing fraction. However, vibration compaction was unsuccessful in an 11-in. tube (47.7% packing fraction).

A new, promising bonding technique was developed in which a blend of charcoal powder and furfuryl alcohol is injected into the top of a column of loose coated particles using a grease gun. The fuel body is then processed routinely. Impregnation of a 15-in.-long fuel column was found to be complete using a hand operated gun. The charcoal distribution was apparently good (0.18 g/cm³ at the bottom and 0.175 g/cm³ at the top). The over-all charcoal content of 0.19 g/cm³ corresponds to an element loading of about 3-3/4 lb. A loading device with control of injection pressure is being constructed. The grease-gun technique is attractive because of its simplicity and economy.

Bonded particle samples were prepared by impregnation, and precasting and direct loading of admixtures for irradiation testing in Capsule P14B.

Loose and furfuryl-alcohol-bonded charcoal (MI-1, XZ-1, and 135) samples have been prepared for Capsule P15. In addition, one bonded particle sample was prepared for Capsule P15 using the furfuryl alcohol admix technique.

Packing and Vibration Studies. Packing tests are being performed to evaluate the uniformity of charcoal - coated-particle - binder blends, and to determine attainable charcoal and coated particle loadings using vibration or other compaction methods.

Several wet blends using reference coated fertile particles and varying amounts of charcoal (with water as the mixing agent) were prepared to determine the effect of charcoal content on the packing fraction of the particles. These tests were conducted using MI-1 and N.F. charcoals. The results (see Fig. 3.7) substantiate previous data on MI-1 charcoal and show

Fig. 3.7--Packing fraction of reference fertile, coated particles as a function of charcoal content

that significantly greater amounts of MI-1 can be added to the blend without altering the packing fraction appreciably. Data presented in the previous quarterly progress report indicated that larger quantities of N.F. charcoal could be added; however, the tests were made utilizing a suspension-filtration technique in which a very fine (suspended) size fraction of the charcoal was used.

A test was conducted to evaluate the uniformity of a blend containing MI-1 charcoal and reference fertile particles. The blend was prepared using water-blending technique described above. The 15-in.-long by 1/2-in.-diameter sample was split into 15 one-inch segments, and the segments were analyzed for charcoal and coated particle content by weight. The plot in Fig. 3.8 shows the cumulative distribution of charcoal content by weight in the segments. The average charcoal content was 6.4 wt-%; the standard deviation was 0.1 wt-%.

A similar test was conducted to determine the uniformity of a furfuryl alcohol - charcoal - coated particle blend. The admixture was loaded into a split graphite tube and compacted to a 64% coated particle packing fraction. The tube was opened and the column sectioned into 15 one-inch long segments. The furfuryl alcohol was dissolved and the charcoal content determined. The average charcoal content was 6.8 wt-%; the standard deviation was 0.5 wt-%.

A series of tests was conducted to determine the intensity and duration of vibration required to compact admixtures of furfuryl alcohol, charcoal, and coated particles and to determine the extent of damage to the coated particles. The results (see Table 3.9) indicate that the coated particles (reference fertile) can withstand at least 16 g (peak-to-peak) for 2-1/2 hr if sufficient charcoal is included.

Table 3.9

DATA ON VIBRATION COMPACTION OF FURFURYL ALCOHOL-CHARCOAL-COATED PARTICLE BLENDS TO A 60% PACKING FRACTION

Charcoal		Furfuryl Alcohol (g/cm ³)	Vibration		Coated Particle Breakage ^a (ppm)
Type	Content (g/cm ³)		g (peak-to-peak)	Time (hr)	
XZ-1	0.154	0.415	8.3	2	25
XZ-1	0.154	0.343	9	2-1/2	137
135	0.154	0.343	10	2-1/2	27
135	0.195	0.343	16	2-1/2	<14

^a As determined by a leach in 8N HNO₃ at 90°C for 2 hr.

Thermal Conductivity Tests. Tests are being performed to determine the effect of bonding on the thermal conductivity of coated particle beds.

Fig. 3.8--Distribution of charcoal content in 1-in. segments of a 15-in. coated particle-charcoal blend

Preliminary thermal conductivity measurements have been performed on a specimen prepared by the furfuryl alcohol admixture technique. In making the measurements, a comparative technique similar to that used for loose coated particles (see GA-6950) was used. The specimen contained equal parts by volume of reference fissile and fertile particles packed to 60 vol-%, and contained 0.158 g of type 135 charcoal per cm³ of specimen.

Preliminary results indicate that the apparent thermal conductivity was about 2.25 Btu/hr-ft-°F with a range of only ± 0.1 between 950° and 1400°C (median temperature). This is about twice the conductivity obtained for loose coated particles (as shown in Fig. 3.9) at the lower temperatures, but unlike loose particles, the conductivity of the bonded sample was fairly insensitive to temperature.

GRAPHITE DEVELOPMENT

The objective of this program is to evaluate various conventional needle-coke graphites for use in fuel elements. These graphites are obtained from various vendors and are evaluated with respect to strength, purity, thermal properties, and cost.

Graphite Evaluation. Conventional needle-coke graphites are currently being evaluated for use in fuel elements and as reflector graphites. The materials of current interest are TS-688, H-327, H-328, and 9567.

Union Carbide completed their portion of a cooperative tensile testing program on Grade TS-688. Their results compare favorably with those obtained previously by General Atomic and Battelle Northwest, confirming the relatively low tensile strength of the bar chosen for this experiment. There appears to be relatively good agreement among the three laboratories with respect to testing procedures.

Thermal expansivity measurements for the candidate graphites are given in Table 3.10.

Table 3.10
THERMAL EXPANSIVITY OF CANDIDATE GRAPHITES

Materials	Thermal Expansion Coefficient, $10^{-6} - ^\circ\text{C}^{-1}$ (R.T. to 500°C)	
	Parallel	Perpendicular
TS-688 ^a	1.40	2.92
(NC-8)	1.21	2.62
H-327	0.94	3.47
	1.00	3.20
9567	1.17	3.24
	1.05	3.66
	0.96	3.56
H-328	5.69	5.61
	5.69	5.40

^a Measured on a sample from Battelle - Pacific-Northwest Laboratory. Material was obtained originally from ORNL and has been irradiated extensively by BNW.

Fig. 3.9--Thermal conductivity of a mixture of coated fuel particles and charcoal

A minimum production lot of nine logs of 18-in.-diameter by 72-in.-long Grade 9567 graphite is currently being evaluated. An effort is being made to obtain a few logs of Grade H-327 in 18-in. diameters. It is very desirable to have full-size production logs of the higher strength materials for further testing to determine whether the necessary properties are retained during the scaleup operation.

Irradiation Testing

A series of irradiation experiments is being conducted to test reference BISO-coated particles under PSC design irradiation conditions. These experiments are intended to guide the selection of fuel materials which will meet the performance requirements of the PSC reactor. BISO coated carbide fuel particles have been selected as the reference design based on their good stability under high-burnup and high-temperature irradiation environments. Current tests are directed toward evaluating the optimum materials parameters for each layer of the BISO coating, with emphasis on evaluating the stability of these particles under high fast-flux exposures.

Graphite irradiations are directed toward obtaining extensive data on a conventional needle-coke graphite over the complete temperature range and up to the lifetime dose of the reactor. Physical property and dimensional changes under irradiation will be measured.

Capsule P14. Capsule P14 was intended to be a three to six cycle ETR test to evaluate the isotropic layer of the BISO coating and to test bonded particle beds. However, it was discharged after completing only one cycle because of leaks in several of the cells and inability to reach design temperatures. The particles experienced a fast-flux exposure of 9.5×10^{20} nvt ($E > 0.18$ Mev) at average fuel temperatures of 950°C (range 515° to 1125°C) for four of the cells and 715°C (range 385° to 860°C) for the fifth. Fuel burnup analyses are still being evaluated; however, it appears the burnups were about 5% FIMA for the fissile particles and 2% FIMA for the fertile.

Results of the postirradiation microscopic examination of the particles, together with detailed descriptions of the samples, are given in Tables 3.11 and 3.12. No metallographic examination was conducted because of the low temperatures, burnups, and fast flux exposures.

All coated particles were found to survive this low-exposure test. Microscopic examination of radial cross sections of the two bonded beds (pitch and resin binders) indicated no apparent change after irradiation. The following conclusions were drawn from this test:

1. BISO coated fissile particles ($T/D = 0.5$) with coating densities of 1.94 to 2.13 g/cm³ and anisotropy factors of 1.0 to 1.9 survived 5% burnup at 950°C.
2. BISO coated fertile particles with coating densities of 2.0 to 2.1 g/cm³ and T/D ratios of 0.30 to 0.38 survived 2% burnup at 950°C.

Table 3.11
DESCRIPTION AND POSTIRRADIATION EXAMINATION OF COATED PARTICLES TESTED IN CAPSULE P14
(1-cycle ETR Irradiation)

Lot Number	Coating ^a					Particle			T/D Ratio	Test Type	Irradiation Conditions ^b				Microscopic Examination		Remarks	
	Type	Thickness (μ)			Density (g/cm ³)	Anisotropy Factor	Type	Size (μ)			Th:U Ratio	Burnup ^c (% FIMA)	Temp. (°C)			Total Particles Examined		Broken Coatings (%)
		B	I	Total									Avg.	Min.	Max.			
3222-85E	BISO(2100)	27	72	99	1.94	1.0	(Th,U)C ₂	150-250	3:1	0.50	Loose	5	960	565	1055	1100	0	BISO density series
3143-135E	BISO(2000)	31	73	104	2.03	1.1	(Th,U)C ₂	150-250	3:1	0.52	Loose	5	950 ^d	550 ^d	1050 ^d	1500	0	
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	950	525	1125	1100	0	
3222-75E	BISO(2200)	32	74	106	2.13	1.2	(Th,U)C ₂	150-250	3:1	0.53	Loose	5	950 ^d	550 ^d	1050 ^d	1300	0	
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	(d)	(d)	(d)	(d)	0 ^d	BISO preferred orientation series
3222-77E	BISO(2200)	27	74	101	2.11	1.3	(Th,U)C ₂	150-250	3:1	0.51	Loose	5	950 ^d	550 ^d	1050 ^d	1100	0	
3222-145E	BISO(2200)	28	69	97	2.11	1.3 ^e	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	995	575	1045	1200	0	
3222-91E	BISO(2200)	28	73	101	2.10	1.9	(Th,U)C ₂	150-250	3:1	0.51	Loose	5	950 ^d	550 ^d	1050 ^d	1300	0	
3222-71E	BISO(2200) ^f	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	(d)	(d)	(d)	(d)	0 ^d	BISO crystallite size series
3143-133E	BISO(2000) ^f	29	68	97	2.12	1.1	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	945	515	1050	1700	0	
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	(d)	(d)	(d)	(d)	0 ^d	Loose versus bonded particles
3033-151D(P2) ^g	BISO(2200)	32	72	104	2.14	(h)	UC ₂ ⁱ	150-250	---	0.52	Bonded-pitch	0	950 ^d	550 ^d	1050 ^d	(j)	(j)	
3222-71E(U) ^g	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Bonded-resin	5	905	580	1000	(j)	(j)	
3143-135E	BISO(2000)	31	73	104	2.03	1.1	(Th,U)C ₂	150-250	3:1	0.52	Loose	5	715	415	845	1800	0	BISO density and preferred orientation series at lower irradiation temperatures
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	Loose	5	685	385	815	1200	0	
3222-77E	BISO(2200)	27	74	101	2.11	1.3	(Th,U)C ₂	150-250	3:1	0.51	Loose	5	750	415	860	1600	0	
3222-129E	BISO(2200)	31	73	104	2.10	1.2	(Th,U)C ₂	300-420	12:1	0.30	Loose	2	965	540	1065	500	0	BISO fertile particle T/D series
3222-127E	BISO(2200)	34	97	131	2.09	1.2	(Th,U)C ₂	300-420	12:1	0.37	Loose	2	925	535	1050	700	0	
3222-127E	BISO(2200)	34	97	131	2.09	1.2	(Th,U)C ₂	300-420	12:1	0.37	Loose	2	(k)	(k)	(k)	(k)	0 ^k	BISO fertile particle density series
3222-105E	BISO(2200)	34	98	132	2.04	1.1	(Th,U)C ₂	300-420	12:1	0.38	Loose	2	950 ^d	550 ^d	1050 ^d	1000	0	

^a BISO denotes two-layer buffer-isotropic PyC coating; the number in parentheses is deposition temp. of isotropic layer in °C; B = buffer, I = isotropic.

^b Fast-flux exposure was 8×10^{20} nvt (estimated).

^c Estimated; analyses in progress.

^d Same sample as 3222-71E in density series.

^e The anisotropy factor was expected to be about 1.6.

^f The two samples differ in crystallite size.

^g Designation for bonded sample.

^h Not determined.

ⁱ Depleted U.

^j No change in appearance of bonded bed was observed after irradiation.

^k Same sample as 3222-127E in fertile T/D series.

Table 3.12

DEPOSITION CONDITIONS AND STRUCTURAL PARAMETERS OF PyC COATINGS TESTED IN IRRADIATION CAPSULE P14

Coated Particle Lot No.	Coating Type	Metallographic Classification	Deposition Temp. (°C)	Deposition Rate (μ/min)	Deposition Gas	Density (g/cm ³)	Bacon Anisotropy Factor	Crystallite Size (L _c)
3222-85E	BISO	Porous	1100	9.0	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2100	1.6	CH ₄	1.94	1.0	123
3143-135E	BISO	Porous	1100	9.2	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2000	1.1	CH ₄	2.03	1.1	154
3222-71E	BISO	Porous	1100	9.7	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	1.2	CH ₄	2.11	1.2	174
3222-75E	BISO	Porous	1100	9.7	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	0.8	CH ₄	2.13	1.2	184
3222-145E	BISO	Porous	1100	9.3	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	0.5	CH ₄	2.11	1.3	180
3222-91E	BISO	Porous	1100	9.3	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	0.2	CH ₄	2.10	1.9	174
3143-133E	BISO	Porous	1100	9.7	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2000	0.3	CH ₄	2.12	1.1	184
3222-129E	BISO	Porous	1100	39	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	1.4	CH ₄	2.10	1.2	184
3222-127E	BISO	Porous	1100	42	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	1.1	CH ₄	2.09	1.2	174
3222-105E	BISO	Porous	1100	42	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	2.0	CH ₄	2.04	1.1	162
3033-151D	BISO	Porous	1100	11	C ₂ H ₂	(a)	(a)	(a)
		Isotropic	2200	0.7	CH ₄	2.14	(a)	165

(a) Not determined.

3. Bonded coated particle beds were apparently unaffected by this low irradiation exposure.

Examination of the gas tubing and thermocouples revealed extensive fracture of both gas tubing and thermocouple shrouds. The tubes fractured in a brittle fashion. The area of tube breakage was blackened. Metallographic examination of the worst region indicated that the copper braze material had penetrated the walls of several of the thermocouples examined. This was a direct result of the multiple brazing required to achieve a gas-tight seal between the lead tube and capsule secondary containment. It is felt that the binding together of many of the thermocouples and gas tubes by the copper which ran down the tubes from the end cap resulted in an overheating problem as a result of the high gamma heating level and the restriction of free movement of gas tubes and thermocouples due to thermal expansion. Design changes are being incorporated in the P15 and subsequent capsules to ensure that this situation is not repeated.

Capsule P14B. Capsule P14B is similar to capsule P14 with essentially identical coated particle samples. The objectives of this experiment are to evaluate the isotropic layer of the BISO coating and to test bonded particle beds. A description of the coated particles being tested is given in Table 3.13.

The capsule, which is in its first of three to six scheduled ETR irradiation cycles, is operating extremely well. All cells are very close to design temperature with cells two through five (design 1300°C) at 1200° to 1305°C (most samples between 1290° and 1305°C), and cell 1 (design 900°C) at 865° to 905°C. Design burnups are 13% and 4% FIMA (for fissile and fertile particles, respectively) in four cycles. The fast-flux exposures will be 3 to 6 x 10²¹ nvt.

Capsule P15. Capsule P15 is a three to six cycle ETR test to evaluate the buffer layer of reference BISO coatings. A description of the samples to be tested is given in Table 3.14. The experiment has been delayed one cycle because thermocouples were not delivered by the vendor in time to begin irradiation in early June (cycle 81).

The fast-flux exposure will be 3 to 6 x 10²¹ nvt at a design temperature of 1300°C. Design burnups are 20% and 6% FIMA (in four cycles).

HTGR Reference Fuel Tests. HTGR reference coated particles (BISO coated fissile and fertile particles in a packed bed) were irradiated in ORNL Loop 1-15 at a maximum temperature of 1400°C. A low-temperature (600°C) appendage capsule containing a small quantity of both loose- and pitch-bonded coated particles is also included. The capsule was retracted early in the quarter. The final burnup was estimated to be 12.1% FIMA in the fissile particles and 2% FIMA in the fertile particles (upper capsule). The burnup in the fissile particles in the lower capsule was estimated to be 9.4%. The release ratios (R/B) just prior to termination of the test are shown in Table 3.15.

Table 3.13
DESCRIPTION OF COATED PARTICLES TO BE TESTED IN CAPSULE P14B

Lot Number	Coating ^a						Particle			T/D Ratio	Design Irradiation Conditions ^b		Test Type	Remarks
	Type	B	I	Total	Density (g/cm ³)	Anisotropy Factor	Type	Size (μ)	Th:U Ratio		Temp. (°C)	Burnup ^c (% FIMA)		
3222-85E	BISO(2100)	27	72	99	1.94	1.0	(Th,U)C ₂	150-250	3:1	0.50	1300	13	Loose	BISO density series
3143-135E	BISO(2000)	31	73	104	2.03	1.1	(Th,U)C ₂	150-250	3:1	0.52	1300	13	Loose	
3222-71F	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	
3222-75E	BISO(2200)	32	74	106	2.13	1.2	(Th,U)C ₂	150-250	3:1	0.53	1300	13	Loose	
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	BISO preferred orientation series
3222-145E	BISO(2200)	28	69	97	2.11	1.3	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	
3342-5E	BISO(2200)	28	69	97	2.12	1.6	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	
3222-91E	BISO(2200)	28	73	101	2.10	1.9	(Th,U)C ₂	150-250	3:1	0.51	1300	13	Loose	
3222-71E	BISO(2200) ^d	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	BISO crystallite size series
3143-133E	BISO(2000) ^d	29	68	97	2.12	1.1	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Loose	Loose versus bonded particles
3222-71E(1) ^e	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	---	0.52	1300	13	Bonded resin precast	
3222-81E(3) ^e	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	1300	13	Bonded resin admixed	
3143-135E	BISO(2000)	31	73	104	2.03	1.1	(Th,U)C ₂	150-250	3:1	0.52	900	13	Loose	BISO density and preferred orientation series at lower irradiation temperatures
3222-71E	BISO(2200)	29	68	97	2.11	1.2	(Th,U)C ₂	150-250	3:1	0.49	900	13	Loose	
3222-145E	BISO(2200)	28	69	97	2.11	1.3	(Th,U)C ₂	150-250	3:1	0.49	900	13	Loose	
3222-129E	BISO(2200)	31	73	104	2.10	1.2	(Th,U)C ₂	300-420	12:1	0.30	1300	4	Loose	BISO fertile particle T/D series
3222-127E	BISO(2200)	34	97	131	2.09	1.2	(Th,U)C ₂	300-420	12:1	0.37	1300	4	Loose	
2260-26	BISO(2200)	32	72	104	(f)	(f)	ThC ₂	300-500	---	0.30	1300	0	Bonded	Diluent fertile particles for bonded beds

^a BISO denotes two-layer buffer-isotropic PyC coating; the number in parentheses denotes the deposition temperature of the isotropic layer in °C. B = buffer, I = isotropic.

^b Design fast-flux exposure is 4×10^{21} for four cycles in ETR.

^c During four cycles in ETR.

^d These two samples differ in crystallite size.

^e Designation for bonded sample

^f Not determined

Table 3.14
DESCRIPTION OF COATED PARTICLES TO BE TESTED IN CAPSULE P15
(Three-to-Six Cycle ETR Irradiation^a)

Lot Number	Coating							Particle				Design _b Burnup _b (% FIMA)	Remarks
	Type	Buffer		Isotropic		Anisotropy Factor	Total Thickness (μ)	Type	Size (μ)	Th:U Ratio	T/D Ratio		
		Thickness (μ)	Density (g/cm ³)	Thickness (μ)	Density (g/cm ³)								
2814-149E	BISO(2200)	25	1.20	80	2.07	1.08	105	(Th,U)C ₂	150-250	1.6:1	0.53	20	True buffer series
2824-11E	BISO(2200)	45	0.96	53	2.05	1.10	98	(Th,U)C ₂	150-250	1.6:1	0.49	20	
2824-9E	BISO(2200)	57	0.67	41	2.06	1.14	98	(Th,U)C ₂	150-250	1.6:1	0.49	20	
2824-27E	BISO(2200)	58	1.25	45	2.13	1.25	103	(Th,U)C ₂	150-250	1.6:1	0.52	20	
2814-151E	BISO(2200)	80	1.11	24	2.04	1.03	104	(Th,U)C ₂	150-250	1.6:1	0.52	20	
2824-29E	BISO(2200)	38	1.00	33	2.05	1.06	71	(Th,U)C ₂	150-250	1.6:1	0.36	20	
28 4-17E	BISO(2200)	29	1.14	74	2.07	1.16	103	(Th,U)C ₂	150-250	1.6:1	0.52	20	High-tempera- ture buffer series
2824-19E	BISO(2200)	46	0.93	51	2.10	1.20	97	(Th,U)C ₂	150-250	1.6:1	0.49	20	
2824-21E	BISO(2200)	63	0.75	39	2.10	1.12	102	(Th,U)C ₂	150-250	1.6:1	0.51	20	
2824-25E	BISO(2200)	34	1.40	65	2.10	1.20	99	(Th,U)C ₂	150-250	1.6:1	0.50	20	Low density isotropic
3552-57E	BISO(2200)	29	1.32	68	2.05	1.13	97	(Th,U)C ₂	300-420	7.8:1	0.49	6	True buffer series on fertile particles
3552-49E	BISO(2200)	44	1.00	60	2.05	1.14	104	(Th,U)C ₂	300-420	7.8:1	0.52	6	
3552-59E	BISO(2200)	60	0.72	35	2.05	1.17	95	(Th,U)C ₂	300-420	7.8:1	0.48	6	
3552-51E	BISO(2200)	50	0.79	48	2.05	1.09	98	(Th,U)C ₂	300-420	7.8:1	0.49	6	
2814-149E	BISO(2200)	25	1.20	80	2.07	1.08	105	(Th,U)C ₂	150-250	1.6:1	0.53	20	Bonded bed (resin admix- ture)
2260-26	BISO(2200)	32	(c)	72	(c)	(c)	104	ThC ₂	300=500	---	0.28	0	

^a Design irradiation temperature for all samples is 1399°C; design fast flux exposure is 3 to 6 x 10²¹ nvt.

^b In four cycles.

^c Not determined.

Table 3.15
FINAL FISSION-GAS RELEASE RATES
IN ORNL LOOP 1-15

<u>Isotope</u>	<u>R/B</u> <u>(x 10⁻⁶)</u>
Kr ⁸⁵	190
Kr ⁸⁷	120
Kr ⁸⁸	250
Xe ¹³³	190
Xe ¹³⁵	51

Postirradiation examination began April 18, 1966. There was evidence of corrosion of the coated particles at the lower (hot) end of the upper capsule and of the graphite sleeve. Only one broken particle was found. The particles were divided into 17 groups to find the uniformity of the fissile-fertile blend. The over-all blend ratio was 36 wt-% fertile - 64 wt-% fissile. One sample contained 66.4 wt-% fertile, another contained 59.5 wt-% fertile, and the balance of the samples fell in the range 29.2 to 51.1 wt-% fertile. The distribution was such that the higher fissile concentrations were at the lower (hot) end of the capsule. It is not known whether this distribution occurred at the time of loading, or during removal and disassembly when the particles were unrestrained.

Capsule G11. A series of capsule experiments has been planned to irradiate conventional needle-coke graphites in the temperature range 550° to 1100°C to a maximum dose of about 9×10^{21} nvt (fast). Capsule G11 contained samples of TS-688, H-327, H-328, and 9567 graphites.

The capsule developed several leaks shortly after startup. These leaks, however, did not pose a threat to the samples, and the experiment was continued for two cycles. It was irradiated in the temperature range 560° to 990°C to a maximum fast-flux exposure of about 1.5×10^{21} nvt. The irradiation has been completed, and the capsule taken to the hot cell where it is awaiting postirradiation examination.

Capsule G12. Capsule G12 is being designed to irradiate candidate graphites. This capsule will contain samples cut from 18-in.-diameter Grade 9567, 20-in. by 20-in. cross section TS-688, 12.75-in.-diameter H-327, and 18-in.-diameter H-328. Samples of pyrocarbon coating material will also be included. The capsule will be designed to operate in the range 700° to 1100°C to a fast-neutron exposure of about 3.5×10^{21} nvt ($E > 0.18$ Mev). The experiment is scheduled to begin irradiation in September 1966 (ETR cycle 84).

CHEMISTRY

The objective of this subtask is to determine:

1. The release behavior of solid fission products that might be important plateout sources and gaseous fission products from the fuel particles being considered for use in the plant as a function of time, temperature, and irradiation by means of analysis and postirradiation annealing and in-TRIGA furnace tests on irradiated fuel particles to determine the effects of such variables as particles diameter coating thickness, and coating structure. The particles used in these tests will be irradiated under other subtests of this program or will be obtained from other sources.
2. The sorption and diffusion behavior of solid fission products in graphites being considered for use in the plant as a function of temperature and concentration by means of analysis and out-of-reactor chemical experiments on graphite samples doped with "fissium."

Fission-product Release

Postirradiation Annealing Experiments. Studies have continued on the release of the metallic fission-product nuclides cesium, strontium, cerium, and barium from two lots of irradiated coated particles. One lot of particles had 98- μ BISO(2200°C) coatings, a 26- μ buffer and a 72- μ isotropic having a density of 2.12 g/cm³. The other lot of particles had 105- μ BISO(2100°C) coatings, a 26- μ buffer and a 79- μ isotropic having a density of 1.99 g/cm³. The kernels used in preparing both batches of particles were 177 to 250 μ (Th,U)C₂, with a Th:U ratio of 4:1. The irradiated particles are being evaluated to determine the effect of irradiation on the release of metallic fission products. These particles are of particular interest because (1) they have coatings of relatively high density and (2) they show, as indicated by earlier release data (see GA-7086), a Cs¹³⁷ release that is lower by a factor of about 10 than BISO-coated particles irradiated in previous irradiation tests.

In postirradiation annealing experiments, samples consisting of five to ten irradiated particles are first gamma-counted to determine the amounts of Cs¹³⁷, Ce¹⁴⁴, Ba¹⁴⁰, and Zr⁹⁵ in the particles. The amount of Zr⁹⁵ produced is used to calculate the number of fissions. The samples, contained in graphite containers, are then annealed in a King furnace at temperatures in the range 1000° to 2000°C for approximately 100 hr. The metallic fission products released are collected in the graphite container and sleeves or on a cold finger. Fission gases are collected in a cold trap. The quantities of metallic fission products released are evaluated by gamma spectrometric and radiochemical analyses of the collection samples taken at various time intervals during the annealing test. To check the release data, the particles used in the annealing experiments are counted afterwards to determine the amounts of metallic fission products remaining in the particles. The calculated release values are based on the amounts of the nuclides present in the particles at the start of the experiment.

Results of the annealing tests at 2000°C are given in Table 3.16. It can be seen that the fractional release values for Ba¹⁴⁰, Sr⁸⁹, and Ce¹⁴⁴ were essentially 1.0 at 60 hr. About 50% of the cesium was released at

100 hr, and fractional release values for Zr^{95} and Kr^{85} were about 0.03 and 10^{-3} , respectively, at 100 hr.

Release constant values for Cs^{137} , Ce^{144} , and Ba^{140} are given in Table 3.17. These values are important because they are used in calculations to predict the release of metallic fission products from HTGR fuel elements. The values given in Table 3.17 were derived from the fractional release data presented in previous reports and from the data given in Table 3.16. The values are based on initial release rates assuming that the release behavior follows the exponential relationship, $F_{\text{ret}} = e^{-Rt}$, where F_{ret} is the fraction retained, R is the release constant, and t is the annealing time. The initial release behavior of particles in an annealing experiment is of primary interest, because this should best represent the release behavior of the particles at the end of the irradiation test.

Other release constants derived from the release data at 2000°C are 0.39 for Sr^{89} , obtained using the BISO(2200°C) particles, and 0.13 for Sr^{89} , 3.4×10^{-4} for Zr^{95} , and 2.5×10^{-5} for Kr^{85} , obtained using the BISO(2100°C) particles.

The release constant values in Table 3.17 are about a factor of ten lower than values found for BISO-coated particles used in previous irradiation tests. This indicates that the retention characteristics of the fuel have been improved.

Figures 3.10 and 3.11 are Arrhenius plots of the release constant data given in Table 3.17. From the slopes of curves drawn through the data for the BISO(2200°C) particles (see Fig. 3.10), neglecting the release constant values obtained at 1750°C which for no apparent reason are too low, one obtains activation energies of 82 kcal/mole for Cs^{137} , 82 kcal/mole for Ce^{144} , and 68 kcal/mole for Ba^{140} ; and for the BISO(2100°C) particles (Fig. 3.11), one obtains 69 kcal/mole for Cs^{137} , 63 kcal/mole for Ce^{144} , and 61 kcal/mole for Ba^{140} . These activation energies are similar to values obtained previously for BISO-coated particles and are typical of values obtained for diffusion-controlled processes.

Activation energies for the BISO(2200°C) particles of 58 kcal/mole for Cs^{137} , 73 kcal/mole for Ce^{144} , and 55 kcal/mole for Ba^{140} were reported in the previous quarterly report (GA-7086). These values are now considered too low because they were based on curves drawn through the 1750°C points which, as mentioned above, are too low.

Retention of Metallic Fission Products during In-pile Irradiation. In measurement of the retention of metallic fission products during in-pile irradiation, quantities of Zr^{95} , Cs^{137} , Ce^{144} , Ba^{140} , Sr^{89} , and Sr^{90} contained in particles (after the irradiation test) are determined using gamma spectrometry and radiochemical analyses. The lithium-drifted germanium crystal detector is particularly useful in this work. The amount of Zr^{95} is used to calculate the number of fissions that occurred and the amount of the various nuclides produced. (Zirconium is essentially completely retained during irradiation testing.)

Table 3.16
FRACTIONAL RELEASE OF Ba^{140} AND Sr^{89} AT 2000°C
FROM BISO-COATED PARTICLES IRRADIATED TO A
BURNUP OF 4% FIMA

Particle Coating	Annealing Time (hr)	Fractional Release	
		Ba^{140}	Sr^{89}
BISO(2200°C)	1.0	0.17	0.39
	3.8	0.54	0.65
	8.1	0.76	0.79
	18.0	0.90	0.89
	41.5	0.95	0.98
	62.5	0.98	0.99
	65.0	0.99	1.00
	82.0	1.00	1.00
	103.7	1.00	1.00
BISO(2100°C)	2.0	0.13	0.26
	8.0	0.67	0.60
	14.6	0.84	0.82
	30.1	0.95	0.96
	50.9	0.98	0.99
	59.3	0.99	1.00
	72.8	0.99	1.00
	93.0	1.00	1.00

Table 3.17
RELEASE CONSTANT DATA FOR PARTICLES IRRADIATED TO
A BURNUP OF 4% FIMA

Particle Coating	Temp. (°C)	Release Constant, Fraction/hour		
		Cs^{137}	Cs^{144}	Ba^{140}
BISO(2200°C)	1300	7.5×10^{-6}	1.3×10^{-5}	1.2×10^{-4}
	1400	2.2×10^{-5}	1.6×10^{-4}	3.5×10^{-3}
	1750	3.3×10^{-4}	3.5×10^{-3}	5.0×10^{-3}
	2000	2.8×10^{-2}	1.0×10^{-1}	1.4×10^{-1}
BISO(2100°C)	1300	2.0×10^{-6}	1.6×10^{-4}	2.0×10^{-4}
	1750	2.1×10^{-4}	8.5×10^{-3}	1.4×10^{-2}
	2000	2.2×10^{-3}	7.1×10^{-2}	8.5×10^{-2}

Fig. 3.10--Arrhenius plot of release constant data for BISO (2200 $^{\circ}\text{C}$)-coated particles irradiated to a burnup of 4% FIMA

Fig. 3.11--Arrhenius plot of release constant data for BISO (2100°C)-coated particles irradiated to a burnup of 4% FIMA

During the quarter, measurements of the fractions of Cs^{137} , Ba^{140} , and Sr^{89} retained by particle samples during the irradiation test were completed. Description of the particles and the fractional retention data are given in Table 3.19. The Cs^{137} and Ba^{140} data are based on gamma spectrometry (using the lithium-drifted germanium crystal detector) and the Sr^{89} data are based on radiochemical analyses. The retention values were calculated from the experimental data using a computer code. The code takes into account shutdown times during the irradiation test.

The fractional retention values for the various particles are the same within the uncertainty of the data. It appears, therefore, that differences in particle properties had no significant effect on the release of cesium, barium, and strontium. As indicated in Table 3.18, the main differences in the kernel and coating properties were in the Th:U ratio and thickness of the isotropic layer. The isotropic layer thickness varied from 48 to 101 μ .

Noble Gas Release Studies. The steady-state release of short-lived fission gases (Kr and Xe nuclides) from two samples of BISO pyrolytic carbon-coated (Th,U) C_2 fuel particles irradiated to approximately 8.5% FIMA burnup was studied over the temperature range from ambient (approximately room temperature) to 1700°C. These experiments were a part of continuing work to determine the effect of burnup on the integrity of the pyrolytic carbon coatings. One sample of particles consisted of 177- to 250- μ (Th,U) C_2 kernels (Th:U = 4:1) with 98- μ BISO (2200°C) coatings, a 26- μ buffer, and a 72- μ isotropic having a density of 2.12 g/cm³. The other sample consisted of 177- to 250- μ (Th,U) C_2 kernels (Th:U = 4:1) with 105- μ BISO (2100°C) coatings, a 26- μ buffer, and a 79- μ isotropic having a density of 1.99 g/cm³.

In steady-state fission gas release work, samples consisting of 5 to 10 particles (or more if the particles have low activity) contained in a graphite crucible are placed in a King furnace mounted in a TRIGA reactor. Release measurements are made by sweeping released fission gases with helium into a small charcoal trap held at -198°C. At the end of the run, the contents of the trap are counted. The results are given in Table 3.19.

An examination of the results shows (1) that the release values increased only slightly with temperature, (2) that half-life had no apparent effect on release, and (3) that the release values were relatively high. This behavior means that recoil release predominated, indicating a relatively high uranium contamination in the coatings.

For diffusion-controlled release, the release values would be more temperature dependent and also would be proportional to the square root of the half life of the fission-gas nuclides. Fuel particles with cracked coatings would tend to show a diffusional type of behavior, because diffusion of the gases from the kernel would be rate limiting.

Steady-state fission-gas release results were performed previously using a sample of the BISO(2200°C) particles irradiated to about 4% FIMA burnup, and the results were reported in the previous quarterly report. The earlier release values are very similar to those in Table 3.19, indicating that irradiation from 4% to 8% FIMA burnup caused no effect on release behavior.

Table 3.18

DESCRIPTION AND FRACTIONAL RETENTION VALUES OF COATED PARTICLES AFTER IRRADIATION

Sample No.	Kernel			Coating					T/D ^d Ratio	Irradiation Conditions		Fractional Retention		
	Type	Th:U Ratio	Size(μ)	Type ^a	Thickness ^b (μ)			Density ^c (g/cm ³)		Burnup (% FIMA)	Temp. (°C)	Cs ¹³⁷	Ba ¹⁴⁰	Sr ⁸⁹
					B	I	T							
1	(Th,U)C ₂	4:1	177-250	BISO (2000)	22	48	70	1.95	0.33	7.4	1200-1440	0.92	0.74	0.58
2	(Th,U)C ₂	4:1	177-250	BISO (2000)	29	68	97	1.91	0.46	6.8	1200-1440	0.95	0.74	0.60
3	(Th,U)C ₂	4:1	177-250	BISO (2000)	40	101	141	1.88	0.66	6.9	1190-1430	0.96	0.76	0.60
4	(Th,U)C ₂	4:1	177-250	BISO (2000)	26	56	82	1.94	0.38	6.7	1200-1440	0.98	0.76	0.58
5	(Th,U)C ₂	17:1	300-350	BISO (2000)	28	76	104	1.91	0.32	2.5	1190-1430	0.98	(e)	(e)
6	(Th,U)C ₂	17:1	350-420	BISO (2000)	45	98	143	1.94	0.37	2.6	1190-1430	1.00	0.78	0.50

^a BISO denotes buffer-isotropic coating; number in parentheses gives the deposition temperature of the outer isotropic layer.^b B denotes buffer layer, I denotes isotropic layer, and T denotes total thickness of coating.^c Density of isotropic layer.^d Ratio of coating thickness to fuel particle diameter.^e Not determined.

Table 3.19

FISSION-GAS RELEASE VALUES FOR BISO-COATED (Th,U)C₂ FUEL PARTICLES IRRADIATED TO 8.5% FIMA BURNUP

Particle Coating	Temp. (°C)	Steady-state Fractional Release					
		Kr ^{85m} (4.4h)	Kr ⁸⁸ (2.8h)	Kr ⁸⁷ (1.3h)	Kr ⁸⁹ (0.053h)	Xe ¹³⁸ (0.28h)	Xe ¹³⁹ (0.011h)
BISO(2200°C)	RT	1.0x10 ⁻⁴	8.0x10 ⁻⁵	9.2x10 ⁻⁵	7.0x10 ⁻⁵	7.2x10 ⁻⁵	1.8x10 ⁻⁵
	1100	9.5x10 ⁻⁵	6.4x10 ⁻⁵	7.0x10 ⁻⁵	7.4x10 ⁻⁵	8.2x10 ⁻⁵	4.4x10 ⁻⁵
	1300	3.1x10 ⁻⁵	1.6x10 ⁻⁵	8.0x10 ⁻⁶	1.1x10 ⁻⁴	9.6x10 ⁻⁵	4.2x10 ⁻⁵
	1700	1.8x10 ⁻⁴	1.7x10 ⁻⁴	1.7x10 ⁻⁴	1.7x10 ⁻⁴	1.5x10 ⁻⁴	7.6x10 ⁻⁵
BISO(2100°C)	RT	---	---	---	4.3x10 ⁻⁵	5.2x10 ⁻⁵	7.0x10 ⁻⁶
	1100	3.2x10 ⁻⁵	7.7x10 ⁻⁶	4.5x10 ⁻⁶	2.5x10 ⁻⁵	2.5x10 ⁻⁵	5.7x10 ⁻⁶
	1400	3.3x10 ⁻⁵	2.0x10 ⁻⁵	1.1x10 ⁻⁵	8.7x10 ⁻⁵	9.2x10 ⁻⁵	4.0x10 ⁻⁵
	1700	1.6x10 ⁻⁴	1.6x10 ⁻⁴	1.3x10 ⁻⁴	1.3x10 ⁻⁴	1.3x10 ⁻⁴	7.3x10 ⁻⁵

Sorption and Diffusion of Fission Products

Vaporization of Metallic Fission Products from Graphite. Measurements are continuing on the vapor pressures of the fission-product metals cesium, strontium, and barium sorbed on graphite. The main purpose of this work is to obtain basic data for use in calculations, which are described later in this task, to predict the release of these metals from PSC fuel elements.

Vapor pressure measurements have been made on barium sorbed in TS-688 graphite using the Knudsen-cell mass spectrometric technique. A sample of TS-688 graphite powder, prefired in vacuo to 2000°C and loaded to 3 mg Ba/g C, was used. The graphite had a BET (nitrogen adsorption) surface area of 5.87 m²/g C. Experimental procedures were essentially the same as those described in the previous report for an investigation of the strontium-TS-688 graphite system.

The results are presented in Fig. 3.12, which is a plot of log concentration versus log pressure, and Fig. 3.13, which is a plot of log concentration versus heat of vaporization. The data on both plots are linear, within experimental error, indicating conformance to the Freundlich isotherm.

The data are described by the Freundlich equation

$$\log P/1.95 = (-1.304 + 7553/T) \log C/86.4,$$

where P is the barium pressure in atmospheres, T is the temperature in degrees Kelvin, and C is the barium concentration in $\mu\text{moles Ba/m}^2$ graphite.

In the form of the Freundlich equation used in FREVAP calculations (see GA-6830, p.81), the data are described by

$$\ln P_{\text{Ba}} = [a + b(\frac{10^3}{T})] + [c + d(\frac{10^3}{T})] \ln C_{\text{Ba}},$$

where P, T, and C are the same as for the previous equation and $a = 6.48$, $b = -33.69$, $c = -1.304$, and $d = 7.553$.

A comparison of the Freundlich isotherms representing the behavior of barium sorbed on TS-688 graphite and HLM-85 graphite is shown in Fig. 3.14 (data for the barium-HLM-85 graphite system were given in GA-6830). These two sets of data are in reasonable agreement, particularly in the experimental region 10^{-5} to 10^{-10} atm and 1500° to 2000°K.

One can calculate the monolayer concentration on three bases: (1) on the basis of pressure, assuming that when the pressure of barium carbide is obtained the sample is loaded to monolayer concentration; (2) on the basis of heat of vaporization, assuming that when the heat of vaporization is equivalent to that of barium carbide the sample is loaded to monolayer concentration; and (3) on the basis of the atomic size of barium. Values for both the barium-TS-688 graphite and the barium-HLM-85 graphite systems, calculated by these three methods, are given in Table 3.20. Data for BaC₂

Fig. 3.12--Variation of vapor pressure with concentration for barium sorbed on TS-688 graphite at 1900°K

Fig. 3.13--Variation of heat of vaporization with concentration for barium sorbed on TS-688 graphite

Fig. 3.14--Freundlich isotherms for sorption of barium on graphites

obtained by Flowers* were used. The geometrical calculation was based on a diameter of 4.35 Å for the barium atom.

Table 3.20
MONOLAYER CONCENTRATION OF Ba ON TWO GRAPHITES

Graphite	Monolayer Concentration ($\mu\text{moles Ba/m}^2$)		
	Press.	Heat of Vapor.	Geom.
TS-688	3.9	2.1	10.1
HLM-85	3.1	8.1	10.1

Cesium Sorption on Activated Carbon. Measurements are in progress to study the sorption of cesium on activated carbon (charcoal). The purpose of this work is to investigate the value of adding activated carbon, which has a relatively high surface area per unit weight, to the packed bed of fuel particles in the PSC fuel element to provide a sorbent for metallic fission products.

An isopiestic method is used. A sample of graphite, having known BET surface area, is placed in the horizontal leg of an L-shaped Pyrex glass and stainless steel tube. The system is evacuated, and cesium metal tagged with Cs^{137} to a known specific activity is distilled from an auxiliary glass tube into the Pyrex leg of the tube. The entire system is thoroughly out-gassed and sealed off at a pressure of about 10^{-5} mm Hg. During a run, the cesium pressure is fixed by maintaining the cesium metal in the Pyrex leg at a constant temperature. The amount of cesium on the graphite specimen is monitored with a calibrated gamma scintillation counter until an equilibrium condition has been established at the desired specimen temperature and cesium pressure.

Work on the addition of activated carbon is coincident with the development of methods of bonding the particles into the holes of elements to prevent particle spillage in the event of fuel element cracking. The bonding techniques under investigation, including pitch and furfuryl alcohol impregnation, have caused appreciable reduction of the BET (nitrogen adsorption) surface area of the carbon. This reduction may be expected to reduce the sorptive characteristics of the carbon, since chemisorption has been demonstrated to be a surface phenomenon. Accordingly, experimental work is being started to obtain cesium sorption data on the impregnated activated carbon for correlation with the BET surface area data.

Cesium and BET (nitrogen) sorption measurements were made on three samples of Type AC-4 (6 to 10 mesh) activated carbon that had been treated

*Flowers, R. H. and E. G. Rauk, "Studies of the Equilibrium Metal Vapor Pressures Over the Alkaline-earth Carbides," AERE-R 5011, August 1965.

differently. Cesium sorption isotherms were obtained at 900° and 1000°C over a cesium pressure range of 2×10^{-6} to 1×10^{-4} atm. Comparison of the sorption data with cesium sorption isotherms for untreated carbon are shown in Table 3.21.

Table 3.21
CESIUM SORPTION MEASUREMENTS USING ACTIVATED CARBON

Sample	Treatment	BET Area (m ² /g C)	Cesium Loading at 900°C (P _{Cs} = 1×10^{-5} atm) ^a	
			(mg Cs/g C)	(mg Cs/m ² C)
1	Untreatment	947	195	0.206
2	Heated to 1600°C	650	130	0.200
3	Impregnated with pitch and heated to 1600°C	0.56	104	186
4	Impregnated with furfuryl alcohol and heated to 1600°C	0.56	99	178

^a These data were obtained from the 900°C sorption isotherms, all of which were parallel.

Examination of these data show a reduction in the BET surface area of 31% by heating the charcoal to 1600°C. However, the cesium sorption capacity is also reduced by the same amount, so that the cesium loading remains unchanged when expressed on a surface-area basis.

The pitch and furfuryl alcohol impregnations apparently rendered most of the pores in the carbon inaccessible to nitrogen (the gas used in the BET method), whereas cesium sorption remains relatively unhindered by pore closure. The cesium loading was reduced by about 20% (compared to the heat-treated carbon), whereas the BET area was reduced by a factor of 1150. Thus, it is evident that the BET method will not evaluate the effect of the various impregnating and bonding techniques on the capacity of carbon to adsorb metals. Accordingly, cesium and strontium sorption data will be used for evaluation in future studies. Samples will initially be examined for their cesium sorption characteristics, and strontium sorption measurements will be made on the most promising specimens.

Sorption of Strontium on Steel and on Graphite Dust. Measurements on the relative sorption of strontium on TS-688 graphite dust and Type 304 stainless steel are continuing. This work will be useful in predicting the distribution of fission-product strontium in the PSC reactor.

A fluidized-bed apparatus is currently being used in the investigation. This technique loosely simulates expected reactor conditions, i.e., graphite dust loaded with metallic fission products and in dynamic contact with steel surfaces in a helium atmosphere. TS-688 graphite dust (74 to 105 μ), loaded with Sr^{85} -tagged strontium to yield a strontium vapor pressure of 10^{-7} to 10^{-8} atm at about 750°C, is fluidized in helium (flow rate 3.5 l/min in a Type 304 stainless steel tube. Two zirconium getters are placed in the loop to ensure a low oxygen concentration. The second getter also serves as a preheater for the gas stream before entering the fluidized zone. A pressure regulator allows additional helium to bleed into the system as needed to maintain a positive pressure in the loop of about 10 cm Hg. This is necessary to compensate for the helium loss through the diaphragm of the recirculation pump.

Twelve thermocouples are spot-welded to the 18-in.-long reaction tube to monitor continuously the temperature of the steel tube. The tube is lined with Type 304 stainless steel foil (0.005-in. thick) that will be removed at the conclusion of the run and cut into sections. The distribution of strontium will then be determined by gamma counting the dust and each steel section.

The expected distribution of the dust particles in the helium stream was visually observed before startup by using a glass tube having the same dimensions as the vertical steel section. There was a heavy concentration of dust particles in the lower 3 to 4 in., but the distribution seemed relatively uniform throughout the remainder of the tube.

After two weeks operation at 750°C, an analysis of the loop gas showed about 3000 ppm H_2 and 0.03 ppm CO. The hydrogen (formed from 25 ppm H_2O in the helium supply and residual moisture in the system at startup) should ensure a clean steel surface for optimum plateout results. The hydrogen concentration was reduced to about 100 ppm by bleeding out the loop gas at 50 cm^3/min and replacing it with clean helium. This run will continue for six weeks with this reduced hydrogen concentration in the loop.

Determination of Oxide Film Thickness on Stainless Steel. An attempt was made to utilize a vacuum-fusion technique for measuring the thickness of the oxide film on stainless steel specimens. The purpose was to obtain information for use in correlating information on the sorption (plateout) behavior of metallic fission products, such as cesium and strontium, on steel surfaces.

Electropositive elements, such as cesium and strontium, should tend to react with the oxide film on metal surfaces as long as oxygen is present in the film and as long as the vapor pressure of the metal is above the equilibrium pressure of the metal over the resultant oxide. Thus, one might expect that the capacity of a steel surface to sorb metal elements from the gas phase will be directly related to the oxide film thickness.

The vacuum-fusion technique was used to determine the total amount of oxygen and nitrogen in 0.002-in.-thick type 304 stainless steel foil specimens as a function of the temperature and duration of a vacuum-baking period.

The steel foil specimens were melted in a graphite crucible in the presence of powdered graphite and the total gas evolved (nitrogen plus oxygen, the latter in the form of CO) was measured from the increase in system pressure. The CO was converted to CO₂ and trapped out; the nitrogen was determined by the difference in pressure. The nitrogen fraction included the inert gases, which were assumed to be negligible.

Seven specimens were analyzed in this manner, each having a total surface area of 100 cm². Before the analyses, the foils were degreased and rolled up to make a compact specimen, and five of the specimens were vacuum-baked at various temperatures. The results of these measurements are shown in Table 3.22.

The data in Table 3.22 show that appreciable nitrogen, as well as oxygen, was evolved when the specimens were fused. The nitrogen must have come from the interior of the specimens. Accordingly, a portion of the oxygen must have come from the interior, and this precludes use of the oxygen data for estimation of the thickness of the oxide film. It therefore appears that vacuum fusion is not a satisfactory method for use in determining oxide film thickness.

Table 3.22

RESULTS OF VACUUM-FUSION ANALYSES OF STAINLESS STEEL SPECIMENS

Specimen	Temp. (°C)	Time (hr)	($\mu\text{g O}_2/\text{cm}^2$)	($\mu\text{g N}_2/\text{cm}^2$)	(N ₂ /O ₂)
1	(a)	---	2.32	14.3	6.13
2	(a)	---	2.14	12.9	6.03
3	400	18	2.24	12.4	5.53
4	600	12	2.46	13.6	5.53
5	800	8	2.01	12.8	6.37
6	1000	4	1.64	8.0	4.88
7	1000	22	1.92	9.1	4.74

^a As received.

Monolayer Equivalents of Metallic Fission Products. In studies of the sorption of metallic fission-product elements on graphite and steel surfaces, it is often useful to express the results in terms of monolayers. In the past, varied values for monolayer equivalents have been used, calculated from radii obtained from various sources without designation of the packing arrangement of the atoms. This has led to some confusion.

To avoid this problem, a set of standard monolayer equivalents, shown in Table 3.23, has been adopted. The monolayer equivalents were calculated from atomic diameters* that appear to be reasonable in view of the varying

*Barrett, C. S., Structure of Metals, McGraw-Hill Book Co., 1952.

data reported in the literature. A hexagonal close-packing arrangement was assumed, disregarding any spatial discrepancies that might result from carbide formation, i.e., SrC_2 or BaC_2 .

Table 3.23
MONOLAYER EQUIVALENTS FOR METALLIC ELEMENTS

Element	Molecular Weight	(μmole) mg	Atomic Diameter ($\text{\AA}$)	Area ($\text{\AA}^2/\text{atom}$)	Monolayer Equivalent	
					(atoms/ cm^2) $\times 10^{14}$	(mg/ m^2)
Ce	140.1	7.13	3.40	9.1	10.0	2.33
Sr	87.6	11.4	4.31	14.6	6.25	0.91
Ba	137.4	7.28	4.35	14.8	6.15	1.40
Rb	85.5	11.7	4.88	18.7	4.83	0.69
Cs	132.9	7.53	5.25	21.6	4.20	0.93

Diffusion of Metallic Fission Products in Graphite. The diffusion of barium in TS-688 graphite is being studied to obtain data for use in calculations to predict the release of 12.8-d Ba^{140} from PSC fuel elements. Information on the rate of diffusion of Ba^{140} is particularly important, since this nuclide will decay appreciably during passage through the element graphite.

Graphite diffusion work includes (1) the measurement at 900° to 1200°C of the diffusion coefficient for the isotope Ba^{140} in TS-688 graphite loaded with Ba^{133} -tagged barium at various concentration levels and (2) Soret effect (thermal diffusion) experiments to determine the heat of transport, Q_{Ba}^* , for barium in TS-688 graphite.

Studies to determine the tracer diffusion coefficient for 12.8-d Ba^{140} in TS-688 graphite, preloaded with Ba^{133} -tagged barium have continued. Three diffusion experiments were completed during the quarter.

An experiment was completed after showing a Ba^{140} diffusion coefficient of $2.9 \times 10^{-9} \text{ cm}^2/\text{sec}$ after 340 hr at 1000°C . The Ba^{133} diffusion coefficient was calculated to be $3.6 \times 10^{-9} \text{ cm}^2/\text{sec}$ in the same graphite sleeve after 746 hr at 1000°C .

The second experiment was run a total of 270 hr at 1200°C . The sleeve was preloaded with Ba^{133} -tagged barium to about 0.2 mg Ba/g C. The compact containing about 0.9 mg Ba/g C was then loaded with 1 mc Ba^{140} (carrier free). Three corings were taken and the diffusion coefficient was calculated to be $5.3 \times 10^{-9} \text{ cm}^2/\text{sec}$.

The third tracer diffusion experiment was run to observe the diffusion of carrier-free Ba^{140} in TS-688 graphite. A compact loaded with 1 mc of Ba^{140} was heated to 1225°C for 216 hr and monitored at intervals. Approximately 3×10^{-3} percent of the Ba^{140} appeared on the compact saturation sleeve at some time after 134 hr. The sleeves were changed and the temperature was reduced to 1000°C for 261 hr. No Ba^{140} appeared on the sleeve. The temperature was then raised to 1200°C for 72 hr, and again, no Ba^{140} could be detected on the sleeve. This may have been partly due to the small amount of Ba^{140} activity remaining.

A Soret (thermal-diffusion) experiment of barium in TS-688 graphite has been initiated so that the distribution of fission-product barium in a graphite body (such as a PSC fuel element) having a significant temperature gradient may be estimated. This essentially involves determining the heat of transport (Q_{Ba}^*) of barium when it undergoes transport in graphite in a thermal gradient. The data obtained from this type of experiment will provide necessary mathematical input for computer analysis of fission-product transport in fuel elements.

A TS-688 graphite cylinder 1.52 cm in diameter and 8.00 cm long was impregnated with Ba^{133} -tagged barium nitrate to yield a concentration greater than 0.2 mg Ba/g C. The nitrate was decomposed to the oxide by heating the graphite slowly to 900°C for 4 hr, resulting in a loss of about 5% of the initial barium. (Two previous attempts failed to convert the nitrate when heated to only 600°C .) The graphite cylinder was encapsulated in a molybdenum can by arc welding under high vacuum. The clearance between the molybdenum can and the specimen was low (approximately 0.001 in.) to reduce vapor transport of the barium at high temperatures. A longitudinal groove 1.27 mm by 1.27 mm was cut in the specimen to allow evacuation.

The specimen in the can was annealed at 1300°C for 100 hr to ensure uniform distribution of the barium throughout the graphite. The encapsulated specimen was then placed in a small graphite tube furnace providing an almost linear temperature gradient of 315°C (972° to 1287°C) over the length of the 8-cm specimen, and the run began.

Figure 3.15 shows concentration profiles determined from radiation scans of the specimen at the start of the run and after 159.8 hr. The gamma scan was made using a collimating slit of 1/16 in. The data in Fig. 3.15 are plotted on a log C versus $1/T$ plot because the slope of this type of plot is convenient for use in determining the heat of transport. It is apparent that a considerable migration of the barium to the cold end of the specimen is occurring.

This run will continue until equilibrium has been established. The specimen will then be cored both axially and radially in order to establish the exact concentration gradient. The radial concentration gradients will be examined carefully to determine the extent of barium transport in the vapor phase. Higher (or lower) concentrations near the surface of the graphite cylinder, in particular around the slit in the specimen, would suggest vapor transport to be important.

Fig. 3.15--Soret (thermal diffusion) effect study of barium in TS-688 graphite

Mathematical Treatment of Metallic Fission-product Release from HTGR Fuel Elements. The FRELIM code has been developed for making an upper-limit estimate of the release of the less volatile metallic fission products from an HTGR core. The code is particularly useful, for estimating the release of nuclides of the elements Y, Zr, Nb, Mo, Tc, Ru, Rh, Pd, In, Sn, Sb, La, Ce, Pr, and Nd.

The method of calculation is based on the assumption that in the limiting case the longer-lived isotopes* of a given fission-product element have been able to diffuse to the coolant-graphite interfaces of the fuel elements in a time short compared to the time of interest. Furthermore, it is assumed that the isotopes at the interface have the chemical potential corresponding to the carbide (or other form of the given element that is in equilibrium with carbon) at the temperature of the interface.

The FRELIM code is formulated for a single fuel element radial zone in an HTGR core, which includes the corresponding top and bottom reflector blocks. The sum of the releases calculated for the several zones comprising the reactor core then give the total release from the core. The zones are divided in k segments (approximately isothermal zones) and the calculations are made over a period divided into j time steps, as indicated in mathematical formulation given below, Eq. (1).

Production of a fission-product isotope, i, of a particular chemical elemental species h is given by

$$\frac{dM_{hi}(t)}{dt} = B_{hij} - \lambda_{hi}M_{hi}(t), \quad \left(\frac{\text{moles}}{\text{day}}\right) \quad (1)$$

where $M_{hi}(t)$ = the total quantity of a given fission-product isotope hi at time t (moles),

B_{hij} = production rate of the isotope hi, the subscript j indicating the time interval to which B_{hij} applies (moles/day), and

λ_{hi} = radioactive decay constant (1/day).

The quantity of a given fission-product element h at time t is given by

$$M_h = \sum_i M_{hi}, \quad (\text{moles})$$

*The short-lived isotopes (< 1 day in half-life) may be expected to decay before they reach the boundary.

where the sum is over the several isotopic species of the fission-product element.

The rate of escape (E_h) of a given fission-product element h from segment k of a given fuel element radial zone (or region) in time interval j is given by

$$E_{hkj} = J_{hkj} (A/l) l_k, \quad (\text{moles/day}) \quad (2)$$

where J_{hkj} = diffusion current of h through boundary layer in segment k during time interval j (moles/cm² day),

(A/l) = area to length ratio of the coolant channel, in the given fuel element (cm), and

l_k = length of coolant channel in segment k (cm).

Now the diffusion current through the boundary layer in segment k is given by

$$J_{hkj} = H_{hkj} \left(\frac{P_{bh} - P_{ch}}{R_g T_{ckj}} \right), \quad (\text{moles/cm}^2 \text{ day}) \quad (3)$$

where H_{hkj} = the mass transfer coefficient for h at k in time interval j (cm/day),

P_{ch} = partial pressure of fission-product element h in coolant, i.e., in turbulent region (atm),

P_{bh} = equilibrium pressure of fission product as the carbide at the interface temperature, i.e., partial pressure at wall (atm),

T_{ckj} = coolant temperature, segment k , time interval j (°K), and

R_g = gas constant = 82.06 (cm³-atm/°K-mole).

Let us consider calculation of the zeta factor,

$$\zeta_h = \frac{P_{ch}}{P_{bh}},$$

of the several fuel segments, which is an essential part of the FRELIM computation.

Since at steady-state the flow of a given fission-product element (the sum of the isotopes) across the boundary layer into segment k equals loss by convection (neglecting decay of radioisotopes) from segment k , we have for any value of index k , except for $k = 1$,

$$\begin{aligned}
 E_{hkj} &= J_{hkj} (A/1) l_k = H_{hkj} (A/1) l_k \frac{P_{bhk}}{R_g T_{ckj}} (1 - \zeta_{nkj}) \\
 &= \left(\frac{\dot{n}_{He}}{P_{He}} \right) [\zeta_{hkj} P_{bhk} - \zeta_{h(k-1)j} P_{bh(k-1)j}] ; \quad (4)
 \end{aligned}$$

then

$$\zeta_{hkj} = \frac{\left(\frac{\dot{n}_{He}}{P_{He}} \right) \left(\frac{P_{bh(k-1)j}}{P_{bhk}} \right) \zeta_{h(k-1)j} + H_{hkj} \frac{(A/1) l_k}{R_g T_{ckj}}}{\left[\left(\frac{\dot{n}_{He}}{P_{He}} \right) + H_{hkj} \frac{(A/1) l_k}{R_g T_{ckj}} \right]} ,$$

or

$$\zeta_{hkj} = \frac{\left(\frac{P_{bh(k-1)j}}{P_{bhk}} \right) \zeta_{h(k-1)j} + \frac{H_{hkj} (A/1) l_k}{(\dot{n}_{He}/P_{He}) R_g T_{ckj}}}{\left(1 + \frac{H_{hkj} (A/1) l_k}{(\dot{n}_{He}/P_{He}) R_g T_{ckj}} \right)} . \quad (5)$$

In the case where $k = 1$ and $k = k'$ is the last segment (at coolant exit),

$$\begin{aligned}
 E_{hlj} &= H_{hlj} (A/1) l_1 \frac{P_{bhlj}}{R_g T_{clj}} (1 - \zeta_{hlj}) \\
 &= (\dot{n}_{He}/P_{He}) (\zeta_{hlj} P_{bhlj} - \alpha_{hj} \zeta_{hk'j} P_{bhk'j}) \quad (6)
 \end{aligned}$$

$$\zeta_{hlj} \left[(\dot{n}_{He}/P_{He}) P_{bhlj} + \frac{H_{hlj} (A/1) l_1}{R_g T_{clj}} P_{b1j} \right] = (\dot{n}_{He}/P_{He}) \alpha_{hj} \zeta_{hk'j} P_{bhk'j} + \frac{H_{hlj} (A/1) l_1 P_{bhlj}}{R_g T_{clj}}$$

$$\zeta_{hlj} = \frac{\frac{P_{bhk'j}}{P_{bhlj}} \alpha_{hj} \zeta_{hk'j} + \frac{H_{hlj} (A/1) l_1}{(\dot{n}_{He}/P_{He}) R_g T_{clj}}}{1 + \frac{H_{hlj} (A/1) l_1}{(\dot{n}_{He}/P_{He}) R_g T_{clj}}} , \quad (7)$$

where α_{hj} = fraction of h that does not plate out upon passing through the coolant circuit (i.e., fraction returned) in time interval j,

$\dot{n}_{He}$ = flow rate of helium in the coolant channels of the given fuel element radial zone (moles/day),

P_{He} = pressure of helium coolant (atm),

P_{bhkj} = vapor pressure of h at graphite boundary (segment k, time interval j),

$$\log_{10} P_{bhkj} = A_h - B_h \left(\frac{10^3}{T_{gkj}} \right), \text{ and} \quad (8)$$

T_{gkj} = graphite temperature at coolant interface, in segment k, time interval j ($^{\circ}\text{K}$).

The constants A_h and B_h are chosen to give the vapor pressure of chemical element h (in atmospheres) over its carbide (or other condensed form, such as the metal species, which is at equilibrium in the presence of carbon at temperature T_{gkj}) in accordance with the best available data.

The following lists values of A_h and B_h which are considered the best presently available.

Elemental Species (h)	A_h	B_h	Elemental Species (h)	A_h	B_h
Sr	3.74	11.9	In	5.47	12.3
Y	7.08	30.8	Sn	5.32	15.5
Zr	6.43	40.2	Sb	5.38	12.3
Nb	7.14	39.2	Ba	3.13	12.2
Mo	7.34	34.7	La	6.67	28.3
Tc	7.34	34.7	Ce	8.81	33.0
Ru	8.48	35.8	Pr	5.68	25.7
Rh	7.78	30.8	Nd	5.68	24.6
Pd	6.57	21.5	Pm	7.34	34.7
Ag	6.10	14.2	Sm	5.51	18.1

Values of the mass-transfer coefficient are calculated from the formula

$$H_{hkj} = \frac{2.5 \times 10^3 R_m T_{gkj}}{(d_o^2 - d_i^2) MP Sc^{0.67} Re^{0.20}}, \quad (\text{cm/day}) \quad (9)$$

and where in turn the computer calculates

Schmidt No.,

$$Sc = \frac{\mu_{kj}}{\rho_{kj} D}$$

Reynolds No.,

$$Re = \frac{4m}{\pi(d_o + d_i)\mu_{kj}}$$

gas density,

$$\rho_{kj} = \frac{MP}{R_g T_{ckj}} \quad (g/cm^3)$$

diffusion coefficient,

$$D_{kj} = \frac{D_o}{P} \left(\frac{T_{ckj}}{273} \right)^{1.75} \quad (cm^2/sec)$$

and viscosity,

$$\mu_{kj} = 4.17 \times 10^{-6} (T_{ckj})^{0.678} \quad (g/cm \text{ sec})$$

from the following input data:

d_o = outside diameter of coolant annulus or circular cross-section diameter (cm),

d_i = inside diameter (= 0 if coolant passage a circular hole),

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

M = molecular weight of coolant (g/g-mole),

P = coolant gas pressure (atm),

D_o = diffusion coefficient of gas species at 1 atm and $0^\circ C$ (cm^2/sec),

m = mass flow rate of coolant (g/sec), and

T_{ckj} = coolant gas temperature in zone k and at time interval j ($^\circ K$).

The zeta factors ζ_{hkj} are calculated by iteration using Eqs. (5), (7), and (8) with the following input quantities being given: R_g , $\dot{n}_{He}$, P_{He} , (A/l) , l_k , A_h , B_h , α_{hj} , H_{hkj} , T_{ckj} , T_{gkj} . Initial values of $\zeta_{hkj} = 1$ may be used.

The rate of plateout (deposition)* in the coolant circuit of fission product isotope hi , $D_{hi}(t)$ is given by

*This includes removal to the helium coolant purification system.

$$D_{hij}(t) = \left(\frac{\dot{n}_{He}}{P_{He}} \right) (1 - \alpha_{hj}) \zeta_{hk'j} P_{bhk'j} \frac{M_{hi}(t)}{\sum_i M_{hi}(t)} \quad (\text{mole/day}), \quad (10)$$

The integrated deposition in the coolant circuit (plus removal to the coolant purification system) at time t , $ID_{hi}(t)$, is given by

$$ID_{hi}(t) = \int_0^t D_{hi}(t') e^{-\lambda_{hi}(t-t')} dt' \quad (\text{moles}). \quad (11)$$

The quantities $M_{hi}(t)$ are obtained by solving Eq. (1) using the input quantities B_{hij} and λ_{hi} .

The expression for $M_{hi}(t)$ for time $t_j, < t < t_{j+1}$ is given by

$$\begin{aligned} M_{hi}(t) = & \frac{B_{hi1}}{\lambda_{hi}} \left[1 - e^{-\lambda_{hi} t_1} \right] \prod_{j=2}^{j=j'} e^{-\lambda_{hi}(t_j - t_{j-1})} \left[e^{-\lambda_{hi}(t - t_{j'})} \right] \\ & + \dots \\ & + \frac{B_{hij'}}{\lambda_{hi}} \left[1 - e^{-\lambda_{hi}(t - t_{j'})} \right]. \end{aligned} \quad (12)$$

Note that if $\lambda_{hi} = 0$, the exponentials = 1 and $\frac{B_{hik}}{\lambda_{hi}} (1 - e^{-\lambda_{hi} t}) = B_{hik} t$.

The total deposited activity (A_{hi}) at time t is given by

$$A_{hi} = \frac{[ID_{hi}(t)] \lambda_{hi} (6.01 \times 10^{23})}{3.7 \times 10^{10} (8.64 \times 10^4)} \quad (\text{curies}). \quad (13)$$

The fractional release of isotope hi at time t , FR_{hi} , is given by

$$FR_{hi} = \frac{ID_{hi}(t)}{M_{hi}(t)}. \quad (14)$$

HELIUM PURIFICATION SYSTEM

The objectives of this subtask are:

1. To determine operating conditions of the plant's low-temperature adsorption train as a function of bed size and temperature, co-adsorbate interference, regeneration techniques, aerosol removal, and adsorption bed materials, by means of laboratory-type experiments.
2. To predict the extent of carbon transport and deposition in the plant's primary coolant system by means of analysis and out-of-reactor laboratory scale testing under the influence of variables such as temperature, materials, coolant gaseous impurities, and "fissium" concentration.
3. To determine a method of cleaning and protecting the steel components of the plant coolant system prior to startup, by reviewing available methods and testing selected candidates to verify their adequacy.
4. To develop processes for decontamination of the plant components to facilitate maintenance by means of analysis and testing of representative material on samples which have been exposed in a simulated plant environment.
5. To establish the suitability, reliability, and maintainability of the helium purification process for the plant for steady-state and transient operation by means of conceptual design and analysis utilizing data from other parts of the program and/or other sources.
6. To develop criteria for the final design of components making up the helium purification system.

SORBENT BEDS

Low-temperature Adsorption Studies

An investigation of the adsorption of tritium and hydrogen on activated carbon and molecular sieve material at -196°C has continued. The investigation consists of equilibrium isotherm studies to determine the capacity of sorbent beds and dynamic adsorption studies to determine adsorption rates.

In this investigation, mixtures of hydrogen and/or tritium in helium are circulated through an adsorbent bed utilizing the high-pressure helium loop apparatus. In the equilibrium isotherm studies a fixed amount of adsorbate (hydrogen and/or tritium) is added to the apparatus and circulation is continued until equilibrium is reached. In the dynamic adsorption studies the adsorbate is added continuously to the circulating helium upstream from the adsorbent bed, and the time required for breakthrough is measured. Dynamic adsorption studies yield information on H_d ,* the transfer-unit height,

*For more information on H_d see Burnette, R. D., W. W. Graham, and D. C. Morse, The Removal of Radioactive Krypton and Xenon from a Flowing Helium Stream by Fixed Bed Adsorption, General Atomic report GA-2395, October 11, 1961.

which is a measure of the rate of adsorption or the height of bed required for effective delay of adsorbates.

Equilibrium isotherms for tritium and hydrogen at -196°C on activated carbon† and 5A molecular sieves were reported in the previous quarterly report. During the present quarter, tritium and hydrogen isotherms on 4A molecular sieve material have been determined. The data points for the molecular sieve material were found to correspond quite closely with the activated carbon equilibria.

Experiments on the dynamic adsorption of hydrogen and tritium on activated carbon† are essentially complete. Experimentally determined breakthrough fronts at 25°C and -196°C for both species indicate that adsorption is rapid (i.e., equilibrium is attained rapidly).

Dynamic tests using hydrogen at high partial pressures as the adsorbate show that breakthrough profiles are steep and breakthrough times are controlled by the respective saturation values as determined by the equilibrium adsorption isotherm. Figure 3.16 illustrates this effect. As would be expected, the rate of saturation of the adsorbent, and therefore the breakthrough times, are dependent on the initial inlet concentration of the hydrogen.

An experiment was conducted using a mixture of hydrogen and tritium as the adsorbate, with the concentration of tritium in the ppb range. The results are included in Fig. 3.16. It can be seen that both species eluted from the activated carbon bed in essentially the same time.

Dynamic breakthrough tests using tritium alone as the adsorbate in the ppb range) have been carried out. Figure 3.17 shows the effluent concentration profile of a low-temperature, high-pressure run.

The height of the mass-transfer unit, H_d , was measured at widely divergent conditions of pressure, temperature, and flow rate. In sorbent bed studies, flow rate is normally expressed in terms of "particle Reynolds number," defined as $D_p G / \mu$, where D_p is the particle diameter, G is the mass velocity ($\text{lb}/\text{ft}^2\text{-hr}$) of gas, and μ is the viscosity of the carrier gas. Figure 3.18 is a plot of H_d versus particle Reynolds number for both the present experiments using tritium and previous experiments using krypton reported in GA-2395. The fact that H_d increases with particle Reynolds number (flow rate) indicates that the rate of tritium adsorption is controlled by mass transfer of adsorbate to the adsorbent surface. The work shows that the calculational methods applied to krypton and xenon previously described in GA-2395 should be satisfactory for predicting tritium breakthrough profiles.

High-temperature Adsorption Studies

Studies of the dynamic behavior of fission products and carbon aerosols in a high-temperature activated carbon bed have continued. The primary purpose of this work is to determine the degree of transport of carbon

†Type BC-107, 6-10 mesh.

Fig. 3.16--Hydrogen and tritium dynamic breakthrough profiles; tritium concentration plotted in arbitrary units (actual tritium concentration in ppb range)

Fig. 3.17--Tritium breakthrough profile

Fig. 3.18--Height of the mass transfer unit versus particle Reynolds number determined using activated carbon, BQ-107, 6-10 mesh, as absorbent

dust (aerosols) through a carbon bed. The apparatus, designated HITPOT (high-temperature plateout trap), and procedure are described in the previous quarterly report. The trap consists of a 7/8-in.-ID steel tube, about 10 cm long, filled with 6-10 mesh activated carbon. The apparatus is designed to simulate conditions of temperature, purge gas velocity, and helium pressure expected in the PSC high-temperature plateout trap.

During this reporting period, the first experiment using the HITPOT apparatus was completed and the second experiment was started.

In the first experiment, the generation of aerosols in situ through abrasion as a result of gas flow or vibration was studied. In this test a small amount (0.446 g) of Ba^{133} -tagged activated carbon (6-10 mesh) was placed in the column about 3 cm from the inlet end to act as an aerosol source. For details, see the previous quarterly report. The test trap was scanned after various running times--prior to the start of gas flow, and after 9, 45, 46 (following strong vibration), and 56 days accumulated running times (see Fig. 3.19). The test trap was vibrated violently on the 46th day of running by giving the trap 300 sharp raps with a hammer. Very little change from the initial activity distribution was noted in the activity profile after the various running times. The greatest change was noted after the vibration test. The vibration on the 46th day caused the activated carbon to settle, as was noted by a 2-cm shift in the source activity peak. An increased amount of Ba^{133} activity (as well as carbon dust) was detected on the millipore filter following the vibration test (see Table 3.24).

Table 3.24

ACTIVITY DETECTED ON MILLIPORE FILTER AT OUTLET OF EXPERIMENTAL TRAP

Time Elapsed (days)	Collection Time (days)	Ba^{133} -tagged Carbon (μ g)	Loading (%)
9	9	0.13	$\approx 3 \times 10^{-5}$
45	36	0.03	$\approx 7 \times 10^{-6}$
46	1 ^a	15.1	$\approx 3.5 \times 10^{-3}$

^a After vibration.

In general, it was concluded that gas flow alone is not sufficient to cause attrition of the carbon granules. The combination of gas flow and vibration, however, will cause some dusting of the carbon and, therefore, some transport of fission products out of the trap via aerosols or dust particles.

In the second test, a sample of Sr^{85} -tagged dust (carbon black) was injected into the HITPOT upstream from the carbon bed while gas flow and full pressure were maintained. After five days the apparatus was shut down and gamma-scanned, as in the previous test. It was found that approximately

Fig. 3.19--Distribution of Ba¹³³-tagged carbon dust in carbon bed after various running times (in situ aerosol experiment)

40 mg of carbon black was distributed exponentially throughout the bed (see Fig. 3.20) and 0.5 mg, or 1.2%, had penetrated the activated carbon column and collected in the millipore filter. A second scan, after an additional 28 days of operation, showed that the distribution of dust throughout the bed was virtually unchanged and that the amount of additional dust which penetrated the bed was only 6.2×10^{-3} mg, or $1.5 \times 10^{-2}\%$ of the total in the packed carbon bed.

The test trap was returned to the HITPOT apparatus for an additional month of running and will be reexamined soon.

Helium Purification System Components

Since the design criteria of the purification system units have been established, research and development effort, in general, is now needed not to provide design information, but to determine the performance of the units. Presently under study is the testing of a full-size low-temperature adsorber. The required test loop would consist of equipment comparable to that to be used in the plant. This equipment could be used for the test program prior to shipment to the plant site for installation. Such a program, although primarily for testing the low-temperature adsorber, would also prove the adequacy of the other units and provide other useful information such as the freezeout capacity of the low-temperature gas-to-gas exchanger and heat leakage in cryogenic units.

Carbon Transport and Deposition

Studies are under way to gain further information on the effect of metal surfaces and gas composition on the rate and degree of carbon deposition resulting from reactions involving CO and H₂.

The experimental approach for the study of carbon deposition on metals of technical interest is twofold: (1) long-term experiments in which three alloys are exposed to two different H₂-CO-He mixtures and (2) short-term experiments using an automatic recording microbalance in a thermogravimetric analysis (TGA) technique. The metals under study are Incoloy 800, low alloy (2-1/4% Cr - 1% Mo) steel, and Type 316 stainless steel. The experimental conditions and results of each of these approaches are discussed below.

Long-term Tests. In the long-term experiments, ten metal specimens placed in series are annealed isothermally in a 1/2-in.-diameter quartz reaction tube (a different alloy in each tube). Each specimen is a 1-in.-wide by 12-in.-long strip of 0.005-in.-thick foil, loosely coiled. This configuration yields a high catalytic surface area (1548 cm²) which facilitates measurement of the gaseous reaction products CO₂ and H₂O. The two gas mixtures used in these experiments are 1000 ppm and 250 ppm each of CO and H₂ in helium at one atmosphere. Flow rates are around 100 cm³/min. The CO and H₂ levels are obtained by premixing. Oxidizing impurities (e.g., H₂O) are removed from the gas stream by passage over graphite chips at 1200°C and finally through a train containing ascarite and magnesium perchlorate for final reduction of CO₂ and H₂O to less than 1 ppm.

Fig. 3.20--Distribution of Sr⁸⁵-tagged carbon dust in carbon bed after various running times

The results of these experiments after 1400 hr of exposure at 480°C are shown in Table 3.25.

Table 3.25
RESULTS OF 1400-HR EXPOSURE OF STEEL SPECIMENS TO MIXTURES
OF H₂ AND CO IN HELIUM AT 480°C

Material	Gas Mixture	Carbon Deposition Rate ^a (μg C/min)	Observations
Incoloy 800	1000 ppm CO 1000 ppm H ₂	0.25	Specimens uniformly dark grey; some isolated patches of carbon deposition; grey color may be due to oxide formation.
2-1/4% Cr - 1% Mo		0.38	First specimen covered with carbon deposit; remaining samples shiny.
316 SS		0.0015	Leading edge of first sample covered with carbon; other samples slightly discolored.
Incoloy 800	770-240 ppm CO ^b 770-240 ppm H ₂	0.10	Specimens uniformly grey in color.
2-1/4% Cr - 1% Mo		0.028	No carbon deposit; all specimens shiny.
316 SS		<0.0002	No carbon deposit; all specimens shiny.

^a Carbon deposition rate by CO₂ measurement; total deposition rate has been found to be approximately double this rate (i.e., H₂O production ≈ CO₂ production).

^b Composition of gas mixture varied as follows: 0-380 hr, 770 ppm; 380-980 hr, 400 ppm; 980-1400 hr, 240 ppm.

Figures 3.21 and 3.22 show by CO₂ measurement how the carbon deposition rates have changed during the 1400-hr run.

The observation that carbon deposition is occurring only on the leading specimens, in the case of 2-1/4% Cr - 1% Mo steel and Type 316 SS at

Fig. 3.21--Long-term carbon deposition experiment (conditions: temp, 480°C; gas composition, 1000 ppm H₂ and 1000 ppm CO; gas flow, 100 cm³/min at 1 atm; specimen surface, 1548 cm²)

Fig. 3.22--Long-term carbon deposition experiment (conditions: temp, 480°C; gas composition, ≈ 200 to 800 ppm H_2 and CO; gas flow, 100 cm³/min at 1 atm; specimen surface, 1548 cm²)

the higher CO and H₂ levels (see Table 3.25), indicates that reaction products CO₂ and H₂O generated by reaction of CO and H₂ with the first specimen are inhibiting the reaction in the downstream specimens. Everett and Kinsey* have shown that for the conditions of this experiment, 480°C and 1000 ppm CO and H₂, the critical H₂/H₂O ratio below which carbon deposition cannot occur is 20. The H₂/H₂O value for this experiment (for the 2-1/4% Cr, 1% Mo specimens) is approximately 100. It appears that there is not only an absolute thermodynamic limitation, as calculated by Everett and Kinsey, but also a kinetic limitation that prevails at even relatively high H₂/H₂O ratios. This phenomenon is not new. Carbon deposition invariably occurs to a greater extent on the leading edge of even quite small specimens, where low gas flow rates are used.

The results of the Incoloy 800 experiments are inconclusive. No leading edge effect is noted, and the cause of the grey coloration is not understood.

Short-term Tests. The experimental apparatus for the short-term tests is a Cahn automatic recording microbalance from which test specimens (nominally 0.5 g) are suspended in a quartz reaction tube. The apparatus permits both weight changes and gas concentration changes to be easily and sensitively monitored.

More latitude in experimental parameters is employed in the short-term tests. Variations in temperature, gas composition, sample composition, and H₂/H₂O ratio have elucidated much of the reaction kinetics.

A low alloy steel specimen (2-1/4% Cr - 1% Mo) was exposed to 1% CO and 1% H₂ in helium for 140 hr. A steady-state reaction rate was observed for the last 100 hr. The H₂ and CO concentrations were changed 5 times, and steady-state reaction rates were observed for a period of 100 hr or more for each gas mixture. Table 3.26 shows the reaction rates observed for the different gas compositions. The reactions involved are:

Figure 3.23 shows log-log plots of $(P_{\text{CO}})^2$ as a function of reaction rate for reaction (1), and $P_{\text{H}_2} P_{\text{CO}}$ as a function of reaction rate for reaction (2). Both plots are linear and show slopes corresponding to a reaction order of 0.8. Therefore, the rate for reaction (1) is

$$K_1 (P_{\text{CO}})^{1.6},$$

*Everett, M. R., and D. V. Kinsey, Some Aspects of Carbon Transport In High Temperature Gas Cooled Reactors, Dragon D. P. report 365, August 1965.

Fig. 3.23--Effect of reactant pressure on carbon deposition rate (conditions: temp, 500°C; gas flow, 50 cm³/min at 1 atm; specimen was 2-1/4% Cr-1% Mo alloy, 0.487 g, 16 cm² surface area)

and the rate for reaction (2) is

$$K_2(P_{\text{CO}} \times P_{\text{H}_2})^{0.8}.$$

The reaction rate constants, K_1 and K_2 , are of course variable, because they depend largely on surface pretreatment, amount of carbon deposited, roughness factor, and alloy composition. It is therefore difficult to arrive at absolute reaction rates that can be extrapolated to the reactor system.

Table 3.26
CARBON DEPOSITION RATES ON LOW-ALLOY STEEL^a

Gas Mixture		Carbon Deposition Rate ($\mu\text{g C/cm}^2\text{-hr}$)	
CO (atm)	H ₂ (atm)	By Total Weight Gain Measurement	By CO ₂ Measurement
10 ⁻²	10 ⁻²	7.0	35
10 ⁻³	10 ⁻²	6.0	0.64
10 ⁻³	10 ⁻³	1.25	0.60
2.68x10 ⁻⁴	2.64x10 ⁻⁴	0.17	0.10
6.95x10 ⁻⁴	8.06x10 ⁻²	20	0.55

^a Specimen was 0.487 g 2-1/4% Cr - 1% Mo alloy of 16 cm² geometric surface area. Temperature was 500°C and the gas flow rate was 50 cm³/min.

At the conclusion of the experiment cited above, controlled amounts of water vapor were injected into the reaction vessel to determine the effect of varying H₂/H₂O ratio on the carbon deposition rate.

The results of this test are shown in Fig. 3.24. Each experimental point is numbered, showing the test sequence. It is seen that for the specimen with the heavy carbon deposit, almost 2000 ppm H₂O vapor (H₂/H₂O = 5) is needed to completely poison the reaction. (This result checks Everett and Kinsey's thermodynamic prediction.) The inhibiting effect of water vapor on the reaction rate is also evident where H₂/H₂O > 5. The possibility of a kinetic limitation was discussed above in connection with the long-term tests.

Perhaps more important is the effect of water vapor on the catalytic activity of a new specimen. As shown in Fig. 3.24, about 480 ppm H₂O (H₂/H₂O = 21) is sufficient to prohibit incipient carbon deposition. However, as soon as some reaction is allowed (by eliminating water vapor in the inlet gas), more water vapor is required to quench the reaction.

A plot of reaction rate as a function of temperature for three gas mixtures is shown in Fig. 3.25. It can be seen that reaction rates maximize

Fig. 3.24--Effect of water concentration on carbon deposition rates (conditions: temp, 500°C; gas composition, 1% CO - 1% H₂ in He; gas flow, 50 cm³/min at 1 atm; specimen, ≈0.5 g 2-1/4% Cr - 1% Mo alloy)

Fig. 3.25--Effect of temperature on carbon deposition rate (conditions: gas flow, $50 \text{ cm}^3/\text{min}$ at 1 atm; specimen, 2-1/4% Cr - 1% Mo alloy, 0.487g, 16 cm^2)

in the temperature range 400° to 500°C. The relatively narrow temperature range for maximum reaction rates has been previously demonstrated by Everett and Kinsey.

Studies using Incoloy 800 have just begun. Some preliminary carbon deposition rates using 1% H₂ - 1% CO in helium at one atmosphere are shown in Table 3.27.

Table 3.27
CARBON DEPOSITION ON INCOLOY 800

Temp. (°C)	Time at Temp. (hr)	Avg Carbon Deposition Rate ($\mu\text{g}/\text{cm}^2\text{-hr}$)
500	1	5.7
500	1	2.9
500	87	1.7
530	15	0.29
560	3	1.43
560	22	0
530	6	0.71
500	16	0
460	20	0
434	63	0.07
495	20	0
630	6	1.43
630	46	0.29
700	2	3.6
700	4	2.1
700	17	1.3

Table 3.27 shows that carbon deposition on Incoloy 800 begins immediately (with no incubation period), but soon after initiation the rate decreases. Although there is some scatter in the data, this phenomenon (i.e., the decrease in reaction rate with time) is observed to occur each time the temperature is raised to a higher level. This is somewhat similar to diffusion barrier behavior, in which the buildup of a protective layer (perhaps oxide or carbide) on the surface inhibits further reaction. Raising the temperature enables the reacting species to partially overcome the diffusion barrier, but further reaction only increases the barrier thickness and eventually inhibits the reaction. This explanation is of course conjectural, and further experiments will be performed to confirm whether the observed phenomenon is indeed peculiar to Incoloy 800.

Alloys of high chromium content are known to resist carbon deposition, presumably because chromium oxide is not reduced by H₂ or CO. It follows that a continuous chromium oxide film on the metal surface should restrict carbon deposition reactions until the oxide film is broken. A protective Cr₂O₃ film can possibly be prepared by preferential oxidation of the surface,

that is, by preventing the concurrent oxidation of iron. This could be accomplished by using very low pressure of oxidant or by using the proper $H_2 - H_2O$ mixture in an inert gas atmosphere.

Accordingly, selective oxidation of Incoloy 800 was attempted. A specimen was exposed to 0.1% H_2O - 1.0% H_2 - 1.0% CO in He at 500°C for 65 hr. After this treatment, the water vapor source was removed, and carbon deposition from the 1% CO- H_2 mixtures was resumed. Very little deposition was seen after 30 hr exposure. The average reaction rate was 0.28 μg C/cm²-hr. These results, although not conclusive, are encouraging and indicate that selective preoxidation of Incoloy 800 may be feasible.

PART II

Task I

FUEL TRANSFER MACHINE

The objective of this task is to establish the suitability and maintainability of the fuel transfer machine for use in the plant by means of testing (probably in air) of a full-scale prototype of the fuel transfer machine with a segment of a dummy core, and by carrying out tests in helium at the temperature and chemical impurity level expected in the plant of critical-service bearings used in the machine and other critical-service components as necessary.

DESIGN

Design studies mentioned in the previous quarterly report resulted in a preliminary design of a fuel transfer machine with internal storage for 63 elements. The basic features of the transfer machinery included a single, vertical, tubular section containing power sources for azimuth rotation of the lower section, arm extension, and grapple operation. The tubular section was raised and lowered by simple means, such as chains or cables operated by drives located at the top of the machine.

The reach of the machine was 30 in. radially, thereby providing capability for reaching one stack of reflector elements beyond the fueled core. Vertically the machine was capable of servicing all top and bottom reflector elements, in addition to the fuel elements. Grappling was performed by a collet type of grapple utilizing the maximum ledge area available in the fuel block.

A refueling nozzle liner will be placed in a nozzle for the transfer machine to work within during refueling. The liner will be placed in the nozzle by the auxiliary cask after the control rod drive has been removed.

A carousel type of storage magazine with a loading system for the core blocks was utilized. A redundant cooling system formed an integral part of the machine.

The fuel transfer machine assembly will be transported by the reactor building crane.

Work continued on this basis until it became apparent that the carousel-type storage was not optimum for the 63-element configuration. A study layout drawing was then produced of a fuel transfer machine with a capacity of 63 single-length elements but suitable for handling single-length elements, double-length elements, and keyed reflector elements.

This design eliminates the carousel, the stacker mechanism, and the pickup pad and substitutes a static rack with water cooling. The prime objective of

this study was to produce a design that could easily be maintained when contaminated. The mechanism is reduced to a minimum and is contained within the transfer machine, which can be lowered into the hot service facility by the machine hoist mechanism.

FUEL TRANSFER MACHINE TESTS

The second 1000-hr test was completed on a bearing with DF-700 as the lubricant. Results obtained were similar to those obtained on the prior run. The bearings from both tests were disassembled, photomacrographed, cleaned, and reassembled for continued testing. In addition to showing only limited signs of wear in comparison with the observations made of bearings coated with other lubricants, it was interesting that during these last two tests there was no observable pickup on the bearing balls.

Testing has been continued with these bearings to obtain an end-of-life point in terms of total revolutions. In the interest of reducing the total testing hours, the shaft speed for the continued tests has been changed from 50 rpm to 1000 rpm. During reworking of the motor mountings to accommodate the higher test speed defective parts were discovered in the torque readout system.

The motor mount was modified, and the torque readout system was repaired; the front bearing is now being tested at the new speed.

Task II

SERIES-STEAM-TURBINE-DRIVEN CIRCULATOR

The objective of this task is to establish the suitability of the main helium circulators and steam turbine drives for use in the plant by means of analysis, component tests, and prototype testing (or if prototype is less than full scale, full-scale production unit testing) under conditions of temperature and speed expected in the plant and up to approximately 50% of design horsepower. Compressor blade and rotor vibration measurements will be made during these tests. The duration of the prototype or full-scale production unit testing will be approximately 200 hr.

The aerodynamic design of the circulator for the PSC plant has been completed. The design reflects changes that have been made in the previously published reference design. There are four circulators in the PSC power plant. Each circulator consists of a single-stage rotor followed by a single-stage stator with an accelerating section preceding the rotor and axial diffuser downstream of the stator. Table II.1 gives the pertinent design information for the compressor, and Fig. II.1 shows the performance of the compressor over its operating range in the reactor. The ordinate is pressure drop (ΔP) divided by inlet pressure, P_1 . The abscissa is normalized mass flow, $\dot{M}\sqrt{\theta}/\delta$. The reactor operating line is shown with the 100%, 75%, 50%, and 25% load points noted as A, B, C, and D. The circulator aerodynamic speed, $N/\sqrt{\theta}$, is the parameter. The reactor operating lines for two out of four circulators operating, for the cases of two in the same loop and one in each loop, are also shown, as is the case for a single circulator operating.

The circulator performance requirements to satisfy emergency operating conditions are shown in Fig. II.2. The reactor operating line changes from the normal operating condition because of the changes in the conditions within the closed circuit.

Figure II.3 shows the torque speed requirements together with the torque speed characteristics of the emergency water turbine drive, the design of which is summarized for the two different heads across the water turbine. The higher head, 2040 psi, relates to the turbine design condition with the boiler feed pump as the source of water supply while the lower one, 250 psi, relates to the condition where the condensate pumps act as the water supply.

Since the selection of the compressor design was based on a value for the aerodynamic loading parameter that can be exceeded, i.e., the diffusion factor; two other compressor designs have been made for higher diffusion factors. These blading designs are such that the same disk can be used for increases of the aerodynamic loading parameter by 10% and 20%. The same disk can be used as these two designs are for the same number of blades at the same stagger angles as the reference design. It is anticipated that

Table II.1
SUMMARY OF HELIUM COMPRESSOR DESIGN

Helium Conditions

Mass flow rate, lb/sec	242
Inlet pressure, psi (static)	686
Outlet pressure, psi (static)	700
Inlet temperature, °F	742

Aerodynamic and Mechanical Conditions

Rotational speed, rpm	9550
Inner diameter, in.	18.0
Outer diameter, in.	26.6
Rotor centrifugal stress, psi	19,300
Rotor bending stress, psi	3760
Stator bending stress, psi	4500
Number of rotor blades	31
Number of stator blades	33
Over-all compressor efficiency, %	78.9
Maximum diffusion factor	0.505
Maximum rotor camber	1.48

Diffusion and Inlet Condition

Inlet efficiency, %	99
Diffusor efficiency, %	80
Diffusor area ratio	3.5:1

Fig. II.1--Circulator performance over operating range in the reactor

Fig. II.2--Circulator performance in emergency operating condition

Fig. II.3--Torque speed requirements and characteristics of the emergency water turbine drive

Table II.2
SUMMARY OF STEAM TURBINE DESIGN

Steam Conditions

Mass flow rate, lb/sec	148
Inlet pressure, psia	910
Inlet enthalpy, Btu/lb	1362.6
Enthalpy drop, Btu/lb	30

Aerodynamic and Mechanical Conditions

Rotational speed, rpm	9550
Inner diameter, in.	15.0
Outer diameter, in.	17.1
Rotor centrifugal stress, psi	3662
Rotor bending stress, psi	6130
Nozzle bending stress, psi	9040
Number of rotor blades	91
Number of nozzle blades	67
Over-all turbine efficiency, %	81.9
Hub reaction, %	5

Diffusion Conditions

Area ratio	3.0:1
Efficiency, %	70

the performance of these designs will be evaluated in a development program and will provide information for the design of future plants with higher pumping power. Figures II.4 and II.5 show the part-load performance of these compressor designs.

Another means by which the pressure rise can be increased is by an increase in axial gas velocity. The effect of both these variables is shown in Fig. II.6 with pressure rise as a parameter. It should be borne in mind that an increase in axial velocity results in an increase in mass flow. As a matter of fact, for flow adjustment of the compressor for a particular reactor plant, it is anticipated that the blade height will be varied by either increasing or decreasing the flow at constant pressure-rise ratio. The machine under development will be for a flow of 14% in excess of the reactor core full load requirement.

Figure II.7 shows the performance for the development compressor. A summary of the steam-turbine-drive design is shown in Table II.2 for the reference design case. If compressor modification is required in future plants, the turbine design will be adjusted by a change in blading. Turbine performance for the reference blading is shown in Fig. II.8. The part-load performance of the steam turbine for the 25%, 50%, 75%, and 100% are summarized in Table II.3. The Pelton wheel design summary is given in Table II.4; its torque-speed characteristic was previously shown in Fig. II.3. The turbine and compressor disk stresses are shown in Figs. II.9 and II.10.

The final design of the circulator is continuing. Material procurement for long-lead items for the inertia rig test has begun, and various parts are being released for manufacture. The final design and construction of the facility for the prototype type test has begun.

Table II.3
PART-LOAD PERFORMANCE FOR STEAM TURBINE

Load (%)	Steam Flow Available to Turbine (lb/sec)	Steam Flow Through Turbine	Pressure Available to Turbine	Pressure at Turbine Inlet	Pressure at Turbine Outlet	Circulator Speed (RPM)
100	155	137.5	866.4	860.0	644.6	9400
75	117.5	89.6	647.7	605.0	488.0	7210
50	78.6	49.4	430.2	357.0	328.0	5020
25	38.5	18.6	209.5	174.5	161.0	2900

Fig. II.4--Part-load performance of the helium circulator with a maximum diffusion factor of 0.55

Fig. II.5--Part-load performance of the helium circulator with a maximum diffusion factor of 0.60

Fig. II-6--Maximum rotor diffusion factor and compressor horsepower as a function of axial velocity

Fig. II.7--Part-load performance of the development circulator: $P_1 = 661$ psia with a pressure rise of 14 psi; design flow rate is 269 lb/sec of He

Fig. 11.8--Torque-speed performance of the steam turbine for various pressure ratios

Table II.4
 PELTON WHEEL DESIGN SUMMARY

Basic Configuration

No. Jets.....	2
Shaft.....	Vertical
Pitch diam. of jet.....	5.25 in.
Jet diameter.....	0.4 in.
No. of buckets.....	20
Bucket-to-jet diam. proportions.....	Normal

	<u>Design Point</u>	<u>Typical Pelton Turbine</u>
Speed, rpm.....	10,800	--
Power, hp.....	400	--
Efficiency, %.....	80	80-90
Head:		
Ft.....	4700	1000-5000
Psi.....	2040	--
Total flow, gpm.....	420	--
Bucket velocity, ft/sec.....	247	--
Jet velocity, ft/sec.....	538	--
Specific speed per jet.....	3.92	2-9
Wheel ratio.....	13.1	27-6
Ratio of wheel speed to theo. jet speed.....	0.45	0.45 ^a

^a For a specific speed of 4.

Fig. II-9---Turbine and compressor disk stresses

Fig. II.10--Turbine and compressor disk stresses

Task III
CONTROL ROD DRIVE

The objective of this task is to determine the suitability, maintainability, and reliability of the control rod drives for the plant under the expected temperature and helium chemical impurity conditions predicted for the reactor by means of testing, including accelerated life testing, of a full-scale prototype of the control rods and drive.

Following the termination of the Phase II environmental test late last quarter, a performance evaluation was undertaken to determine whether or not any changes in operating characteristics had taken place since the start of the test. No evidence was found indicating deterioration or malfunction of the mechanism other than the limit switches and slack cable switch that had malfunctioned for several months.

A control rod guide tube displacement test was performed with the lower guide tubes misaligned relative to the upper guide tubes by 1.0 in. No change in drive characteristics was observed.

When the drive was removed from the autoclave, it was apparent that the drive had been subjected to an excessive temperature for a limited period during which the drive had not been operated. This condition was evidenced by the lead counterweight having melted, by a deposit of cadmium oxide on the cable drums and adjacent structure, and by charring of some of the electrical leads.

The drive was completely dismantled and examined. All parts so far appear to be in good condition with little or no evidence of wear. All dry-film-lubricated bearings still ran smoothly, including the one drum bearing fitted with 440C balls (the other bearings were all fitted with tungsten carbide balls). All grease-lubricated bearings were dry, and some showed uneven wear.

Task IV

STEAM GENERATOR

The objective of this task is to establish the suitability and maintainability of the design of the steam generators for the conditions of temperature and pressure expected in the plant by means of conceptual design, analyses, and testing of the critical aspects of the design. The tests are expected to include gas flow distribution and pressure drops, helium-side heat transfer, tube vibration, and experimental investigation of material compatibility with coolant impurities. The development of a high-temperature design basis and investigation of experience relative to tube integrity is also planned.

TUBE VIBRATION TEST

Drawings for the modification of the vibration test rig were completed. The parts and equipment necessary for this modification were received and assembly has started. The modification in the test loop will provide a mechanism that is capable of continuously moving the flow screens while the test loop is in operation (see Fig. IV.1). This will make it possible to establish the combinations of plena sizes that cause a resonating vibration condition within the tube bundle. This condition results when the combination of plena sizes and the mass effect of the tube bundle air flow channel create a Helmholtz resonator of the same frequency as the tube or tube bundle natural frequencies.

The design of the second tube bundle has been completed, and preparations are being made for fabrication. This bundle will have tubes oriented in an in-line pattern with respect to the direction of flow. Two strain gauges will be applied to each of the tubes in the bundle.

Testing using the modified rig will be started early in the coming period.

STEAM/WATER FLOW STABILITY

Various loop designs for the proposed steam flow stability test have been analyzed with respect to performance at various operating conditions, and the results are presently being evaluated.

The analog computer stability investigation is being revised to incorporate parallel tube coupling effects in the model. The analog computer is a more versatile tool, in this instance, than the digital computer, because use of the latter would require extensive programming modifications.

Fig. IV.1--Steam generator tube vibration test loop

An analytical study and literature survey of moisture carryover from the evaporator to the superheater has been completed. This effect is of interest with respect to corrosion and solids deposition.

STEAM GENERATOR TUBE PLUGGING MACHINE

The development of welding techniques for Incoloy 800 has been delayed owing to material unavailability. The material is now on hand, however, and fabrication of specimens is well advanced.

Task V

COATED PARTICLE DEVELOPMENT

The objective of this task is to determine the process variables for fabrication of coated fuel particles suitable for use in the Plant by means of investigation of various methods of fabricating fuel particles, investigation of coating process variables, and preirradiation evaluation of coated particles.

BUFFER COATING STUDIES

An intensive study of the inner layer of the reference two-layer BISO-coated particles is being conducted. This inner layer is interposed between the fuel particle and the outer, dense coating layer to accommodate swelling of the fuel particle and to absorb fission recoils. The objective of the study is a more complete understanding of the properties required for the inner layer to enhance the radiation stability of the coated fuel particle. Information gained during this study will aid in the optimization of the reference coating process.

Three different approaches to producing a coating that will satisfy the requirements of the inner layer of the BISO fuel particles are being considered. First, the proven concept of using a soft, low-density, isotropic coating (true buffer coating) is being systematically studied. Second, a low-density, isotropic coating that is not soft is being considered because it should be more economical to produce than the true buffer coating; the deposition and evaluation of this type coating has been reported previously.* Third, the concept of a laminar inner coating that contains the core particle loosely within its central void may be evaluated, though it does not now appear economically attractive.

The systematic studies of the true buffer coating have been completed. All of the coatings were deposited from acetylene at temperatures ranging from 900° to 1200°C and in concentrations ranging from 60% to 100%. The results indicate a buffer density minimum at 950° to 1000°C deposition temperature, with other parameters held constant. In all cases, buffer density decreases with increasing acetylene concentration. Over the range of parameters studied, the minimum buffer density achieved on fissile size particles (150 to 250 μ diameter) was 0.7 to 0.8 g/cm³. Methods of characterizing the mechanical strengths of these coatings are being studied.

HIGH-DENSITY ISOTROPIC COATINGS

A study of high-density, isotropic PyC coatings is being conducted. The results of the G9 irradiations on pyrolytic carbon strips indicate that

*Bokros, T. C., The Structure of Pyrolytic Carbon Deposited in a Fluidized Bed, General Atomic Report GA-5163, July 1964.

this structure should have the best radiation stability of the structures tested to date. The object of this work, therefore, is an understanding of the high-density isotropic coating region and optimization of the coating process necessary to obtain the high-density structure.

Work on the deposition of high-density coatings from gases other than methane at low temperatures and low partial pressures have continued. While characterization of the coatings produced is not yet complete, preliminary results show that coatings with densities up to 2.1 g/cm^3 can be deposited; however, preferred orientation measurements have not yet been made to determine whether these high-density coatings are suitably isotropic.

An investigation of the effect of hydrogen and nitrogen diluents (rather than helium) on coating structure was begun. Both hydrogen and nitrogen perturb the coating structures, compared to the helium-diluent-deposited coatings. As can be predicted from chemical equilibrium considerations, the perturbations of hydrogen diluent are greater than those of nitrogen diluent. Characterization of the structures produced in this study is under way.

COATED PARTICLE PREPARATION

Irradiation testing of promising types of reference coated particles is being carried out in the PSC program described in Part I of this report. Supporting irradiation tests are also being conducted at ORNL, Petten, Dragon, and Dounreay. These irradiation tests are intended to guide the development of BISO coatings with optimum radiation stability and retentivity and with minimum fabrication costs. During the past reporting period, coated particles were prepared for the following experiments.

Capsule P15

The P15 irradiation capsule will be a test of buffer structures under simulated operating conditions. Test specimens include the true buffer structure, the high-temperature true buffer, and the low-density isotropic inner layer. Sample selection and preparation for Capsule P15 have been completed.

Petten Irradiation

Six pyrocarbon structures are to be tested in the Petten (Netherlands) reactor. The six structures were chosen from among those prepared for Capsule P14. The irradiation samples are restrained and unrestrained PyC disks. No coated particles are to be irradiated.

COATING EVALUATION TECHNIQUES

Particle Geometry Studies

Irradiation tests of coated particles have shown that the ratio of coating thickness to core particle diameter (T/D) is one of the most important

materials parameters influencing radiation stability. A study is therefore in progress to more accurately determine the T/D ratios to be used in analyzing and correlating the results of the various irradiation experiments. The new T/D value is believed to be more accurate for correlation purposes because:

1. It is based on a minimum coating thickness for each particle rather than an average.
2. It is based on a radiographic average of the maximum and minimum fuel core diameters rather than the arithmetic average of the range of core particle sieve sizes.
3. It is measured on coated particles actually tested in the irradiation experiment rather than on a random sample taken from the entire particle batch.

In the first application of this method, a radiograph of each coated particle sample that went into an irradiation experiment was examined, and the minimum coating thickness and average core diameter of each of 55 particles were recorded.

The data were used to calculate individual and group average T/D ratios. The results are shown in Table V.1 along with the previous data for each sample. These results show (1) that new core diameters are larger, (2) that minimum coating averages are smaller, and (3) that new T/D's are smaller. A plot of the range of T/D values for each sample versus percent failure after irradiation, shown in Fig. V.1, shows (1) that coating failure increases as T/D decreases, as expected, and (2) that coated particles with porous kernels survived with lower T/D ratios than did those with dense kernels, also as expected.

Correlations of these data with other parameters are being made, and the study is being extended to samples from other irradiation experiments. It is anticipated that these studies will lead to a more accurate determination of T/D ratios required for coating survival.

Coating Contamination Studies

The extent of coating contamination in coated particles for irradiation capsules is being determined by alpha counting techniques. The alpha counts can yield quantitative values of uranium contamination alone, but only estimates for combined thorium and uranium.

Alpha emission measurements were made on some coated particles that were tested in an irradiation capsule to determine the amount of fuel contamination on the surface of the coating and the amount within approximately 15 μ of the surface, if the fuel is uranium. If the fuel contains thorium also, the measurements cannot distinguish between the two and yield an average value. From this average, an estimate of the contamination can be made on the assumptions that thorium and uranium diffuse at about the same rate and that the contamination is picked up in the same Th:U ratio as in the fuel core. Each group of particles was measured for alpha emission and the results are listed in Table V.2.

Table V.1
PREVIOUS AND NEW T/D DATA

Sample No.	Previous Particle Data			New Particle Data by Radiography					
	Average ^a Core Diameter (μ)	Average ^b PyC Thickness (μ)	T/D	Average ^c Core Diameter (μ)	Average ^c Min PyC Thickness (μ)	T/D	Std. Dev. (σ)	Max T/D	Min T/D
1	300	107	0.35	327	99	0.30	0.03	0.38	0.22
2	300	103	0.34	340	98	0.29	0.04	0.38	0.16
3	300	103	0.34	316	95	0.30	0.03	0.38	0.26
4	300	85	0.28	313	73	0.24	0.03	0.29	0.18
5	300	127	0.42	311	116	0.38	0.04	0.50	0.27
6	300	192	0.64	325	170	0.53	0.06	0.67	0.36
7	300	92	0.31	296	91	0.31	0.03	0.38	0.24
8	300	100	0.33	302	100	0.33	0.03	0.38	0.25
9	360	114	0.32	408	98	0.24	0.04	0.36	0.13
10	360	146	0.40	400	123	0.31	0.05	0.43	0.24
11	360	170	0.47	406	144	0.36	0.05	0.52	0.27
12	200	138	0.69	183	125	0.69	0.11	0.93	0.45

^a By screening analysis.

^b Based on 27 samples by radiography.

^c Based on 55 samples.

Fig. V.1--New T/D curves showing calculated mean, maximum, and minimum T/D versus percent failed coatings during irradiation at 1350° to 1700°C to 10% to 13% burnup (FIMA) and a fast-flux exposure of 5×10^{20} nvt

Table V.2
COATED PARTICLE CONTAMINATION

Sample No.	Th:U Ratio	$\alpha \text{ min}^{-1}\text{cm}^{-2}$	$\mu\text{g Th} + \text{U}^{235}$ per cm^2 of Sample
3222-85E	3:1	25	0.71
3143-135E	3:1	30	0.83
3222-71E	3:1	34	0.96
3222-75E	3:1	36	1.02
3222-77E	3:1	41	1.17
3222-145E	3:1	42	1.18
3222-91E	3:1	47	1.32
3143-133E	3:1	33	0.92
3222-129E	12:1	24	0.67
3222-127E	12:1	15	0.43
3222-105E	12:1	20	0.55

Acid leaching may be one method of reducing surface contamination. Alpha emission has been decreased by acid leaching in some cases to less than one-fourth the original count.

Coating Density Measurements

The X-ray technique discussed in previous reports was used to measure the density of buffer coatings. The measured buffer densities were only approximations because the coatings were thin and difficulties were encountered in enlarging the photographic plate.

A plate was sent to Kodak Laboratories for enlargement. The enlargement was successfully prepared, and good densitometer measurements were made from the resulting film. The film contained X-rays of 30 to 50- μ buffer coatings. Approximate density values for these samples had been determined earlier, and the new measured values will be compared to the approximations.

THERMAL STABILITY STUDIES

Long-term thermal stability tests of BISO coated particles at 1400°C are being conducted to evaluate the stability of reference particles in the expected maximum HTGR fuel temperature range. Three samples of BISO-coated (Th,U)C₂ particles with high-density coatings are being tested. These particles have been shown to have good radiation stability in P-capsule experiments. The samples have reached 4600 hr at 1400°C. Previous examination after 2200 hr of heating indicated negligible migration. The samples will be examined next at 5000 hr.

FISSION-PRODUCT RELEASE

Postactivation release experiments were conducted on a series of BISO (buffer-isotropic) pyrolytic carbon coated particles. The results are given in Table V.3.

The results, in general, show that (1) all the particles released strontium and barium to an appreciable extent, (2) the total release increased with time as might be expected for a diffusion-controlled process, and (3) strontium releases were higher than barium releases, indicating a higher diffusivity for strontium, in agreement with the results of post-irradiation experiments.

Table V.4 compares accumulated release fraction values at around 8 hr for this series of particles. The values are listed in order of increasing release. It can be seen that the fractional release values for Sr^{91} differ by as much as a factor of five and those for Ba^{140} by as much as a factor of seven; however, release values, coating densities, and coating thicknesses do not appear to correlate in any obvious manner. Particles from Sample No. 1 show the lowest release values, and particles from Sample No. 8 show the highest release values. The relatively high release values for the latter particles, as well as for particles from Sample No. 7, suggest the possibility of a relatively high degree of uranium contamination in the coatings because of the high coating temperatures involved.

The results of the release studies on pyrolytic carbon coated fuel particles, in general, show that (1) all the particles released strontium and barium to an appreciable extent, (2) the total release increased with time, as might be expected for a diffusion-controlled process, (3) strontium releases were higher than barium releases, indicating a higher diffusivity for strontium, and (4) a high pyrolytic carbon density ($\rho > 2.0 \text{ g/cm}^3$) does not appear to improve the retention of metallic fission products--contrary to what one might intuitively expect in the absence of uranium contamination.

Table V.3

FRACTIONAL RELEASE VALUES FOR BISO-COATED CARBIDE PARTICLES
(Determined by Postactivation Annealing
Experiments Conducted at 1400°C)

Sample No. ^a	Annealing Time (hr)	Fractional Release	
		Ba ¹⁴⁰	Sr ⁹¹
1	1.0	0.0018	0.014
	3.0	0.0077	0.029
	7.0	0.021	0.041
	24	0.043	0.050
2	1.0	0.0022	0.017
	3.0	0.0059	0.036
	7.0	0.022	0.059
	24	0.046	0.083
3	1.0	0.0096	0.040
	3.0	0.025	0.081
	8.5	0.058	0.14
	17.8	0.11	0.21
4	1.0	0.0067	0.038
	3.0	0.014	0.058
	8.5	0.055	0.13
	17.8	0.080	---
5	1.0	0.011	0.046
	3.0	0.024	0.076
	4.8	0.046	0.11
	9.3	0.070	0.16
6	1.0	0.0011	0.017
	3.0	0.010	0.051
	4.8	0.021	0.077
	9.3	0.036	0.11
7	1.0	0.010	0.071
	4.0	0.086	0.12
	7.3	0.14	0.14
	11.3	0.15	0.14
8	1.0	0.0053	0.037
	4.0	0.067	0.13
	7.3	0.15	0.19
	11.3	0.17	0.21

^a For detailed description of particles,
see Table V.4.

Table V.4
COMPARISON OF FRACTIONAL RELEASE VALUES FOR COATED CARBIDE PARTICLES
(Determined by Postactivation Annealing
Experiments Conducted at 1400°C)

Sample No.	Kernel			Coating					T/D ^d Ratio	Annealing Time (hr)	Fractional Release	
	Type	Th:U Ratio	Size (μ)	Type ^a	Thickness ^b (μ)			Density ^c (g/cm ³)			Ba ¹⁴⁰	Sr ⁹¹
					B	I	T					
1	UC ₂	---	150-250	BISO (2000)	32	70	102	2.02	0.51	7.0	0.021	0.041
2	(Th,U)C ₂	3:1	250-350	BISO (2000)	31	71	102	1.97	0.34	7.0	0.022	0.059
3	(Th,U)C ₂	4:1	177-250	BISO (2000)	22	48	70	1.95	0.33	8.5	0.058	0.14
4	(Th,U)C ₂	4:1	177-250	BISO (2000)	26	56	82	1.94	0.38	8.5	0.055	0.13
5	(Th,U)C ₂	4:1	177-250	BISO (2000)	29	68	97	1.91	0.46	9.3	0.07	0.16
6	(Th,U)C ₂	4:1	177-250	BISO (2000)	40	101	141	1.88	0.66	9.3	0.036	0.11
7	(Th,U)C ₂	4:1	177-250	BISO (2200)	26	72	98	2.12	0.46	7.3	0.14	0.14
8	(Th,U)C ₂	4:1	177-250	BISO (2100)	26	79	105	1.99	0.53	7.3	0.15	0.19

^a BISO denotes buffer-isotropic coating; number in parentheses is the deposition temperature of the outer isotropic layer.

^b B denotes buffer layer, I denotes isotropic layer, and T denotes total thickness of coating.

^c Density of isotropic layer.

^d Ratio of coating thickness to fuel particle diameter.