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A NATIONAL PROGRAM PLAN FOR WORK
TO ESTABLISH FISSION-PRODUCT BEHAVIOR
IN HTGR SYSTEMS

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1. INTRODUCTION

The purpose of this report is to present (1) an overall discussion of the technical aspects of fission product behavior in HTGR systems and (2) an overall program plan for further development work to characterize fission product behavior in HTGR systems. The program plan encompasses development of analytical methods, laboratory studies of fission product behavior, engineering scale studies, in-reactor studies, and tests to confirm fission product behavior in actual reactor systems. The plan includes consideration of the following:

1. Analysis of key uncertainties in fission product behavior.
2. Integration of further development work with currently scheduled tests in the Peach Bottom HTGR.
3. Investigation of potential methods of confirming fission product transport behavior in HTGR systems.
4. Analysis of methods by which confirmatory fission product data could be obtained from the Peach Bottom HTGR.

Another discussion of fission product behavior in HTGR systems has been prepared by de Nordwall and Bryan.⁽¹⁾

1.1. DESCRIPTION OF HTGR SYSTEMS

HTGR systems employ a uranium-thorium fuel cycle; coated fuel particles; graphite as moderator, fuel cladding, core structure, and reflector; and helium as coolant.

The use of coated uranium-thorium carbide (or oxide) particles in graphite fuel elements results in several advantages: (1) fuel burnups of 50,000 to 100,000 MW-days/ton, (2) excellent inherent neutron economy,

and (3) high heat transfer rates to the coolant, high specific powers, and low fuel inventory. The helium coolant is chemically inert, absorbs essentially no neutrons, and makes no contribution to the reactivity of the system. The core-outlet helium temperature of 1400°F leads to 1000°F modern steam conditions. The particularly good high temperature qualities of graphite, the high heat capacity of the graphite moderator, and the negative neutron temperature coefficient of the graphite core lead to nuclear safety of the HTGR power plant system.

Two operating HTGR systems, the 40-MW(e) Peach Bottom reactor and the 20-MW Dragon reactor, have demonstrated the HTGR concept. The 330-MW(e) Fort St. Vrain HTGR power station for Public Service Company of Colorado is under construction by Gulf General Atomic, and large HTGRs in the 600- to 1100-MW(e) range are under active design at Gulf General Atomic and in England. Related HTGR systems include the UHTREX* at Los Alamos Scientific Laboratory and the AVR in Jülich, West Germany. The major features of each of these HTGR systems are described in Appendix A.

1.2. IN-PILE LOOP EXPERIMENTS

The GAIL and Pluto in-pile loop experiments, described in Appendix B, have contributed substantial information relating to fission product behavior in HTGR systems. Such information has also been supplied by purge-capsule experiments, in particular experiments at Studsvik, Sweden, and Oak Ridge National Laboratory. (In purge-capsule experiments, the steady state release of gaseous fission products is measured during irradiation of fuel specimens.) Also, release information has been provided by unpurged capsule experiments, such as the P-capsule experiments conducted by Gulf General Atomic.

* Operation of the UHTREX reactor has been terminated.

1.3. PURPOSE OF FISSION PRODUCT TRANSPORT STUDIES

The purpose of fission product transport studies is to determine the transport behavior of gaseous and metallic fission products in HTGR systems as a function of time, temperature, and irradiation. This information is important from health, safety, and maintenance standpoints.

Work on fission product transport is of particular importance from the standpoint of safety because in the analysis of accidents, where fission product transport is involved, it is essential to know the distribution of fission products in the primary circuit of an HTGR system under normal operating conditions, since abnormal (accident) situations evolve from normal conditions. For example, the consequences of a depressurization accident are dependent upon the quantity of fission products in the primary circuit (outside of the reactor core) at the time the accident starts.

Incorporation of a secondary containment into the design of the 1100-MW(e) HTGR permits an increase in the tolerance for release of fission products from the primary containment in the event of a depressurization accident. Nevertheless, the amounts of fission products external to the primary circuit in the event of a depressurization accident are limited by an imposed rate of leakage from the secondary containment. Thus, in the analysis of accident situations for the case of an HTGR system with a secondary containment, it is still essential to be able to predict the amounts of fission products in the primary circuit outside of the reactor core.

2. FISSION PRODUCT ELEMENTS

The 35 predominant fission product elements are classified in Table 2.1 according to their fission yields and boiling points. The fission yields of the elements were taken as the sum of the yields of the nuclides having half-lives between 1000 yr and 1 hr or less if precursors have half-lives in the stated range. The fission yields were taken from the DELFIC input.⁽²⁾ The boiling points of the elements were taken from Hultgren⁽³⁾ and Stull and Sinke.⁽⁴⁾

The volatility categories of the elements are listed in the first column of Table 2.1. The elements in the nonvolatile metal category have relatively low vapor pressures and diffusivities and may be considered immobile at HTGR operating temperatures. The following elements in this category form carbides of moderate to great stability:

yttrium	molybdenum	praseodymium	gadolinium
zirconium	lanthanum	neodymium	terbium
niobium	cerium	promethium	

The formation of fission product carbides significantly lowers the vapor pressure of an element. Also, sorption of the elements in graphite at low concentration leads to even lower vapor pressures. It is therefore expected that the nonvolatile metals will remain fixed in the fuel or, at worst, will diffuse out only at extremely low rates.

The fission products in the other volatility categories are either gases or have appreciable vapor pressures at the operating temperatures. The elements in the volatile metals category sorb on graphite, and strontium, barium, samarium, and europium form carbides; but the vapor pressures of the sorbed species or of the carbides are relatively high. Some elements,

Table 2.1
FISSION PRODUCT ELEMENT YIELDS AND BOILING POINTS

Volatility Category of Element	Element	Fission Yield of ^a Element, ΣY (%)	Boiling Point of Element (°K)
Fission gases ↓	Kr	7.8	119
	Xe	13.1	165
	Br	0.54	331
	I	19.4	456
Volatile nonmetals	Se	0	958
	Te	5.74	1261
Volatile metals ↓	As	0.028	885
	Cs	6.51	955
	Rb	0	967
	Cd	0.034	1040
	Sr	22.2	1648
	Sb	1.27	1860
	Eu	0.49	1870
	Sm	0.63	2064
	In	0.009	2343
	Ba	18.6	2398
	Ag	0.043	2436
	Ga	0.00014	2520
Non- volatile Metals ↓	Sn	0.78	2896
	Pm	4.0	3000
	Ge	0.028	3107
	Pd	0.040	3237
	Nd	3.50	3341
	Tb	0.000076	3496
	Gd	0.0010	3539
	Y	28.7	3611
	Ce	18.0	3699
	La	18.9	3730
	Pr	15.6	3785
	Rh	4.27	4000
	Ru	4.25	4392
	Zr	11.7	4682
	Mo	6.1	4883
	Tc	5.4	4900
	Nb	12.0	5017

^aSum of fission yields for nuclides with half-lives between 1 hr and 1000 yr (see text).

such as arsenic, cadmium, germanium, silver, and indium, are unimportant because of their low yields. The following fission product nuclides in the fission gas and volatile categories are of particular interest and importance in considering fission product control in HTGR systems:

1. ^{90}Sr , which is important for safety reasons because (1) it is a long-lived beta-emitter, a bone seeker, and therefore a health hazard, and (2) it has a high yield (5.8%).
2. ^{137}Cs , release of which could result in maintenance difficulties because it has moderately hard gamma activity, a long half-life, and a high yield (6.2%).
3. ^{140}Ba , which could result in maintenance difficulties because it decays to ^{140}La , which emits a very hard gamma. (As will be pointed out in later sections, data exist indicating that ^{140}Ba , because it has a low rate of diffusion in fuel element materials, decays in the fuel element. Therefore direct release of ^{140}Ba into the coolant is not a problem.)
4. ^{131}I , which is important for safety reasons because it concentrates in the thyroid gland and has a sufficiently long half-life, high yield (3.1%), and high activity to be a health hazard. Other iodine isotopes are less important because they have shorter half-lives.
5. ^{90}Kr , which is a precursor of ^{90}Sr .
6. ^{140}Xe , which is a precursor of ^{140}Ba .
7. ^{135}Xe , which is important because of its 2.7×10^5 barn cross section and 6.3% yield.

3. BARRIERS FOR FISSION PRODUCT RETENTION

The primary barrier for the retention of fission products and control of fission product activity within the HTGR primary coolant system is the coating on each individual fuel particle. Two types of coatings must be considered:

1. The BISO coating, consisting of an inner, low density, buffer layer of pyrolytic carbon and an outer, high density, isotropic layer of pyrolytic carbon.
2. The TRISO coating, which has a silicon carbide coating sandwiched between the pyrolytic carbon buffer layer and the outer isotropic pyrolytic carbon layer. In the TRISO-II variation, an isotropic pyrolytic carbon layer is inserted between the buffer and silicon carbide layers.

As will be discussed in later sections, both BISO and TRISO coatings retain fission gases, including iodine, to a satisfactory degree. However, TRISO coatings are superior to BISO coatings in retaining the nuclides of strontium and barium.

Both oxide and carbide kernels can be used with little apparent effect on fission product retention. However, the use of oxide kernels may lead to less fuel contamination in the coating.

The graphite which surrounds the fuel in HTGR systems is an effective secondary barrier for metallic fission products.

4. FISSION PRODUCT CONTROL IN HTGR SYSTEMS UNDER NORMAL CONDITIONS

In considering the transport of fission products in HTGR systems, it is logical to divide the fission products into two groups: (1) fission product gases, including iodine, and (2) fission product metals. These two groups will be considered separately in the following sections.

4.1. CONTROL OF GASEOUS FISSION PRODUCTS IN HTGR SYSTEMS

The mechanism for the transport of krypton and xenon fission product nuclides in an HTGR system is outlined in Fig. 4.1. The basic steps are (1) release from the source, (2) migration through graphite into the coolant, and (3) removal by the purification system or by decay. Some of the krypton and xenon nuclides decay to metallic nuclides, which plateout in the coolant circuit.

The graphite in an HTGR-type fuel element is relatively porous and therefore does not greatly hold up gaseous fission products. Consequently, the fuel particle release rate, the cleanup system removal rate, and the decay rate generally establish the noble gas activity level in the circuit.

The transport behavior of iodine differs from that of the noble gases in that iodine sorbs to some extent in graphite (particularly in the presence of metal atoms) and plates out on steel surfaces (in the temperature range 300° to 500°C) by reaction to form iron iodides. The mechanism for the transport of iodine is given in Fig. 4.2. The basic steps are (1) release from the source, (2) sorption in the matrix and graphite, (3) migration into the coolant, and (4) removal from the coolant by purification, plate-out, and decay.

The sources of release of noble gases and iodine from HTGR fuel elements are (1) fuel contamination in the coatings, (2) fuel contamination

Fig. 4.1. Transport of fission product noble gases
in HTGR systems

Fig. 4.2. Iodine transport in HTGR system

in the fuel rod matrix materials, and (3) particles with defective coatings. Intact coatings of either the TRISO or BISO variety contain fission gases essentially completely, and release from the kernel of intact particles is negligible compared with the release due to fuel contamination and defective particles.

In order to predict the transport behavior of gaseous fission products in an HTGR system, calculation methods and input data are required. The input data include (1) fission product release data, (2) purification rates, (3) decay rates, and (4) plateout data for iodine.

The various aspects of the transport of fission gases and methods for predicting the release of fission gases from HTGR fuel elements are discussed more fully below.

4.1.1. The RAD Code

The primary coolant system activities of noble gases, halogens, other volatile elements, and nonvolatile elements which are assigned a conservative minimum release are calculated using the RAD code. The code accounts for the fission yield, release, decay, and cleanup of each member of the appropriate fission product chain.

The calculational relationships used are outlined below:

Production Rate of Isotope i (A_{PU_i})

$$A_{PU_i} = P \left(Y_i - X_1 Y_{i-1} - X_2 Y_{i-2} \right) + X_1 A_{F(i-1)} + X_2 A_{F(i-2)}$$

where P = power (fissions/sec)

X_1 = branching ratio from parent (i-1)

X_2 = branching ratio from second parent (i-2) if it exists

Y = isotope total fission yield, $Y = (1 - X^{233}) Y^{235} + X^{233} Y^{233}$

Y^{233} = isotope fission yield from U^{233}

Y^{235} = isotope fission yield from U^{235}

X^{233} = fraction of total fissions from U^{233}

Activity Remaining in Fuel (A_{F_i})

$$A_{F_i} = A_{PU_i} (1 - F_i) (1 - e^{-\lambda_i T_C}) ,$$

where

F_i = fission product release fraction from the fuel

= release rate/birth rate

λ_i = decay constant of isotope

T_C = time core is at power P

Gas-Borne Activity (A_{G_i})

Noble gas fission products at equilibrium:

$$A_{G_i} = \left(A_{PU_i} F_i + X_{1_i} A_{P_{i-1}} + X_{2_i} A_{P_{i-2}} \right) \left(\frac{\lambda_i}{\lambda_i + R_N} \right)$$

where

R_N = FP/HI noble gas cleanup rate

FP = purification system flow rate

HI = reactor circuit helium inventory

Nonnoble fission products at equilibrium:

$$A_{G_i} = \left(A_{PU_i} F_i + X_{1_i} A_{G_{i-1}} + X_{2_i} A_{G_{i-2}} \right) \left(\frac{\lambda_i}{\lambda_i + R_C} \right)$$

where

R_C = nonnoble gas fission product cleanup rate = (FP + PP)/HI

PP = equivalent flow rate experiencing plateout cleanup

Equilibrium Coolant Plateout Activity (A_{P_i}):

$$A_{P_i} = \left(A_{PU_i} F_i + X_{1i} A_{P_{i-1}} + X_{2i} A_{P_{i-2}} \right)$$

For calculation of the noble gas activity it is conservatively assumed that all released nonnoble parent activity is available within the primary coolant circuit for decay to the noble gas daughter.

A detailed discussion of the RAD code and its application to HTGR systems is given in Refs. 5 and 6.

4.1.2 Fission Gas Release from Fuel Particles*

The primary barrier to the release of fission gases into the HTGR coolant system is the coating on each fuel particle. The fuel rod matrix material and the graphite in a fuel element are relatively porous and therefore delay significantly only the very short-lived gaseous fission products. The transport time required for gaseous nuclides to traverse the graphite web of the fuel element is considered to be less than 1 min.

Pyrolytic carbon coatings, as well as silicon carbide coatings, have been shown to be excellent barriers for the release of gaseous fission products and iodine from fuel particles in experiments conducted at Gulf General Atomic,⁽⁷⁾ at Oak Ridge National Laboratory,⁽⁸⁻¹¹⁾ under the Dragon project,⁽¹²⁾ and in the GAIL IV irradiation in the GETR.⁽¹³⁾

The end-of-life R/B (release rate/birth rate) data for the GAIL IV loop experiment are plotted against the half-life of the fission products in Fig. 4.3. The GAIL IV fuel element was irradiated in the GETR at full power for 9734 hr while generating a total of 670.6 MW-hr of energy. The fuel element contained pyrolytic carbon coated fuel particles packed into holes. The degree of particle failure at end-of-life was 0.8%.

The release data given in Fig. 4.3 show a $(t_{1/2})^{1/2}$ (square root of half-life) dependence, indicating that the release is controlled by a diffusion mechanism. The fission gas release data measured in most loop

* Reference added in press: R.M. Carroll and O. Sisman, "Evaluating Fuel Behavior During Irradiation by Fission Gas Release," USAEC Report ORNL-4601, September 1970.

Fig. 4.3. R/B values vs half-life for GAIL IV irradiation experiment

experiments and in the Peach Bottom reactor generally show this half-life dependence. Figure 4.3 also illustrates the general observations that (1) xenon release rates are lower than those of krypton, and (2) iodine release rates are like those of xenon.

Measurements have shown that the excellent retention of iodine and noble fission gases by pyrolytic carbon coated fuel particles is unaffected by irradiation up to 24% FIMA.⁽¹⁴⁾

Studies have demonstrated that (1) at temperatures below approximately 1000°C , the release from coated particles is essentially temperature-independent and is governed by recoil processes, and (2) at temperatures above approximately 1000°C , the release becomes temperature-dependent, and is due primarily to diffusion-dependent release of the gaseous nuclides born in or recoiled into the outer pyrolytic carbon coating.⁽¹⁵⁾

The release of fission gases from coated particles at reactor temperatures is primarily from particles with failed coatings and from fuel (uranium and/or thorium) contamination in and on the surface of the outer coating.

A study⁽¹⁶⁾ has shown that there is a noble gas release characteristic of failed fuel particles. Therefore, the reactor designer can establish the design noble gas release levels by determining the amount of fuel failure to be expected and using a release curve obtained from an appropriate experiment having approximately fully failed fuel. The R/B value for the release of Kr-85m from failed coated carbide fuel particles at 1300°C was shown to be around 10^{-2} .

Studies of fuel contamination and its relationship to fission gas release have been performed. One investigation revealed that levels of fuel contamination on the surface and in the outer pyrolytic carbon layers of TRISO-coated particles can be directly related to the fractional release of gaseous fission-product nuclides.^(17,18) Another study showed that release increases in direct proportion to the level of contamination.⁽¹¹⁾

It has been observed that contamination levels of coated particles with oxide kernels are lower by one or two orders of magnitude than those of particles

with carbide kernels coated under similar conditions.^(11, 19, 20) Results indicate that the degree of migration of uranium into particle coatings during application of coatings above 1800°C is significantly higher for carbide kernels than for oxide kernels.

Two types of release, trapping and knockout (in addition to recoil and diffusion), were suggested by ORNL investigators⁽²¹⁾ on the basis of experimental data obtained using uncoated uranium oxide (UO₂) particles.

Other ORNL investigators⁽²²⁾ used mathematical models for interpretation of fission-gas release from pyrolytic carbon coated particles. Their interpretation of the data was that (1) the release of noble gases below 1600°C was due to solid-state diffusion from coating contamination, (2) the release above 1600°C was due to solid-state diffusion of nuclides through the coating, and (3) an overall increase in release rate with time occurred due to migration of U²³⁵ through the coating.

Investigators at ORNL reported that fission gas fractional release from uncoated UO₂ particles is affected by fission density (fission/cm³-sec).⁽²³⁾ It was postulated that traps are created by the fission process and that the number of traps is a function of fission density and temperature.

A study of fission product release from uncoated monogranular uranium carbide (UC₂) particles yielded diffusion coefficients for krypton, xenon, and iodine in uranium carbide. In the paper covering this work,⁽²⁴⁾ a calculational method is described for deriving steady-state fractional release data from short-term (non-steady-state) data for a given fission product nuclide.

Studies have shown that an increase of about one order of magnitude in fission-gas release occurs when coated fuel particles are simply physically mixed with matrix material. Thus, fission gas release increases when fuel particles are fabricated into rods. It is theorized that the increased release occurs because recoil atoms, resulting from fissions of uranium contamination in or on the coating, are stopped in the highly releasing, porous matrix rather than in the more dense, retentive pyrolytic carbon coating of an adjacent particle.⁽²⁵⁾

Investigators at Gulf General Atomic measured the steady-state release of fission gases from an HTGR-type fuel rod sample in a long-term release experiment carried out in the TRIGA Mark III reactor, utilizing a King (graphite tube furnace) and a loop-type system.⁽²⁶⁾ The fuel rod was 2 in. long by 1/2 in. in diameter, contained BISO-coated particles (rather than TRISO-coated particles as indicated in the report), and was made using pitch binder and natural-flake graphite filler material. Steady-state release (R/B) values determined at 30°, 1470°, and 1590°C were very low, indicating satisfactory performance of the fuel rod.

The temperature dependence of fission gas release from a simulated fuel rod has been studied.⁽²⁶⁾ The simulated fuel rod consisted of a mixture of BISO-coated fuel particles mixed with coke matrix material. This study showed that the temperature dependence of the release from the simulated fuel rod is similar to that for coated particles alone.⁽¹⁵⁾ At temperatures below about 800°C, release from the simulated fuel rod was essentially independent of temperature, showed little dependence on isotope, and thus was apparently controlled by recoil. At intermediate temperatures (800° to 1200°C), release was quite temperature dependent; and above 1200°C, release was less temperature dependent. It is speculated that diffusion in the matrix material may be controlling in the intermediate temperature region, whereas diffusion from the particles becomes important at higher temperatures.

The migration of uranium and thorium into pyrolytic carbon coatings on fuel particles and into the fuel rod matrix can lead to an increase in the fission gas release.^(16,26)

Studies at Gulf General Atomic indicate that release of noble gases is high during oxidation-hydrolysis of the carbide kernels of previously failed fuel particles. On the other hand, the release of iodine appears to be low during oxidation-hydrolysis of exposed carbide fuel kernels.

It has been found that iodine and tellurium nuclides are released from pyrolytic carbon coated fuel particles at about the same rate as xenon

isotopes of comparable half-life.⁽²⁷⁾ In the Peach Bottom reactor, I-135 accounted for about one-half of the total Xe-135 release, while I-133 release accounted for about one-third of the Xe-133 (see Section 4.4.2).

In work at ORNL, the release of iodine from coated UO_2 and UC_2 particles with cracked coatings was measured as a function of time and temperature in postirradiation annealing experiments.^(23,28) This work is summarized in Ref. 29. In 8-hr anneals at 1300°C , average iodine releases were 11% for cracked UO_2 particles and 0.9% for cracked UC_2 particles, indicating greater retention by the latter. Mass balances were determined. Activation energies were reported to be 47 kcal/mole at temperatures from 1000° to 1200°C and 92 kcal/mole at temperatures from 1300° to 1500°C .*

In similar studies at GGA,⁽³⁰⁾ the release of iodine from BISO-coated UC_2 particles with cracked coatings was determined. In a 6-hr anneal at 1300°C , the iodine release from cracked particles with 24% FIMA burnup was only 4%.

Needed Work. Work is needed on (1) the half-life dependence of fission gas release from fuel rods, (2) the effect of irradiation rate, time, and burnup on gaseous release from fuel rods, (3) simple quality control methods (such as alpha-counting) for indirect measurement of gaseous release, and (4) the relationship of gaseous release and uranium and thorium contamination in fuel rods. Further studies are needed of the effect on fission gas release of impurity reactions with exposed kernels in particles with cracked coatings. In addition, a better understanding is needed of the mechanisms of fission gas release from fuel particles and fuel rods.

4.1.3 Calculation of Release of Fission Gases from Coated Particles

The transport of fission products in a stagnant medium is generally described by a diffusion equation of the form

$$\frac{\partial C}{\partial t} = \nabla \cdot (D \nabla C) - \lambda C + B, \quad (1)$$

* These activation energies are not consistent with the curves in a plot (see Fig. 5.13, Ref. 29) of iodine release vs reciprocal temperature. Perhaps these activation energies were meant to apply to diffusion coefficients rather than releases. Release is proportional to the square root of the diffusion coefficient. See Ref. 24.

where C is the concentration, t is the time, D is the diffusion coefficient, λ is the decay constant, and B is the birth rate per unit volume. In coated fuel particles with spherical symmetry and a constant diffusion coefficient, this equation takes the form

$$\frac{\partial C}{\partial t} = \frac{D}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial C}{\partial r} \right] - \lambda C + B. \quad (2)$$

This equation can be used to treat the formation, diffusion, and decay of a fission product nuclide in either the kernel or coating, or both. Generally, a buffer layer of low-density pyrolytic carbon exists between kernel and coating, but this can be neglected.

For the case where the birth rate is constant throughout a particle (e.g., a bare uranium carbide particle), the solution to Eq. (2), assuming time-independent properties and zero initial and outside concentrations, is readily obtained. The release of fission products is usually expressed in terms of the R/B value, which is the instantaneous release rate divided by the birth rate. This R/B value as a function of time is given by

$$\begin{aligned} \frac{R}{B}(t) &= - \frac{D 4\pi r^2 \frac{\partial C}{\partial r}}{\frac{4}{3} \pi r^3 B} \Big|_{r=a} \\ &= 3 \left(\frac{D}{\lambda a^2} \right)^{\frac{1}{2}} \left[\coth \left(\frac{\lambda a^2}{D} \right)^{\frac{1}{2}} - \left(\frac{D}{\lambda a^2} \right)^{\frac{1}{2}} \right] - 6e^{-\lambda t} \sum_{n=1}^{\infty} \frac{\exp(-n^2 \pi^2 D t / a^2)}{n^2 \pi^2 + \lambda a^2 / D}, \end{aligned} \quad (3)$$

where a is the outside radius of the particle. This expression starts at zero at time zero and reduces to the first term as t approaches infinity. That is,

$$\frac{R}{B}(\infty) = 3 \left(\frac{D}{\lambda a^2} \right)^{\frac{1}{2}} \left[\coth \left(\frac{\lambda a^2}{D} \right)^{\frac{1}{2}} - \left(\frac{D}{\lambda a^2} \right)^{\frac{1}{2}} \right]. \quad (4)$$

This equation represents the steady-state behavior. The solid line on Fig. 4.4 is a plot of Eq. (4) and illustrates that the release from uncoated (bare) particles should be proportional to the square root of the fission product half-life for R/B values less than 0.1.

If there is a pyrolytic carbon coating on the particle, Eq. (2) must be solved with a step change in the birth rate at the kernel/coating interface--down to a value indicative of coating contamination. The dashed lines in Fig. 4.4 illustrate the behavior that would be expected for different values of the birth rate ratio B_2/B_1 , where B_2 and B_1 are the birth rates in the coating and the kernel, respectively. It should be noted that the behavior is quite different if there is no contamination in the outer coating; i. e., $B_2/B_1 = 0$. However, it should also be noted that the position of the curve for $B_2/B_1 = 0$ depends on the ratio of the kernel and coating diffusion coefficients, the sorptive abilities, and the radii of the two regions.

The release of gaseous (as well as metallic) fission products from coated fuel particles has been treated by several investigators:

Norman and Roos^(31,32) developed the SLIDER code, which is a numerical evaluation of the diffusion of nuclides in a multicoated spherical particle. This code handles both transient and steady-state release.

Dunlap and Gulden⁽³³⁾ developed a two-zone diffusion model to describe diffusion-controlled release. Both the steady-state release and the time to reach steady state are considered.

Walther⁽³⁴⁾ formulated a method for calculating the steady-state fractional release of fission products from coated particles. His method treats release from intact and broken particles as well as from contamination.

Prados and Scott⁽³⁵⁾ developed mathematical relations for estimating the release fractions of gaseous fission products from coated fuel particles and fuel elements.

In general, the formulations consider diffusion coefficients of the element in the fuel and coating, coating and kernel dimensions, distribution coefficients at the fuel-coating interface, contamination fraction, and decay. The formulations are more applicable to the release of metals than to the release of gases from coated fuel particles. This is because contamination

Fig. 4.4. Steady-state R/B values vs half-life and coating contamination

and recoil release is the major source of fission gas release. Thus, release of the gases by diffusion through the coating is relatively unimportant. (See Section 4.2.2 for a discussion of the application of these formulations to metal release.)

Needed Work. There appears to be a continuing need for work in this area consistent in level with experimental studies.

4.1.4 Primary Coolant Circuit Plateout of Iodine

The iodine released from the fuel region is not expected to be held within the active core fuel element graphite to a significant degree. The coolant gas-borne fraction of the total released equilibrium activity of iodine is determined by the combined effects of decay, removal by the helium purification system flow, and plateout (or exchange) on surfaces within the primary coolant circuit flow path.

Extensive work has been performed at Oak Ridge National Laboratory, (23,28,36-40) at Gulf General Atomic, (30,41-43) and at Harwell⁽⁴⁴⁾ to study the deposition of iodine on steel surfaces. Also, iodine plateout studies were conducted in an out-of-pile loop at Battelle Memorial Institute.^(45,46) This work yielded information on the sorption-desorption characteristics of iodine on steel and other surfaces at reactor temperatures. An ORNL investigator recently prepared a review of the adsorption of iodine on metal and its behavior in loops.⁽⁴⁷⁾

The studies at Gulf General Atomic were conducted using (1) an isopiestic technique in which a metal specimen contained in an evacuated L-shaped tube was equilibrated with tagged iodine vapor, and (2) a dynamic technique in which tagged iodine was deposited from a helium stream onto steel tubing. The information derived from these studies can be summarized as follows:

1. The plateout (or reaction) of iodine at 300° to 500°C on steel surfaces is rapid and quantitative. Iodine chemisorbs on metal surfaces with subsequent formation of iron iodides. The desorption and transport of iodine are due primarily to volatilization of iron iodide or by decomposition of the iodide.

2. The plateout is much less rapid and less quantitative at temperatures below 200°C, which indicates that the reaction to form iodide is much less rapid at the lower temperature. (In other words, it appears that at the lower temperatures, the adsorption process is fast but the chemical process is slow.)
3. The exchange of iodine on an iodine-saturated iron surface is rapid, indicating that gas-borne radioactive iodine nuclides will exchange with plated-out stable iodine nuclides.
4. The plateout of iodine on a carbon-dust-covered steel surface occurs by chemisorption on the metal. Relatively little iodine is sorbed on the dust.

Studies at Gulf General Atomic yielded an adsorption isotherm for iodine on 1% Cr-1/2% Mo alloy at 750°F. ⁽⁴³⁾

These results indicate that steel surfaces in an HTGR system would effectively remove iodine from the helium coolant, resulting in a low level of gas-borne iodine activity.

In a recent paper describing operating experience with the Dragon reactor, ⁽⁴⁸⁾ it was stated that iodine is effectively gettered by the primary circuit components and never exists in measurable quantities in the coolant.

Calculational methods have been developed to handle the plateout of fission products. ^(23,49) The PAD code (Plateout Activity Distribution), under development at Gulf General Atomic, solves equations of the form

$$\frac{\partial N_j}{\partial t} = S_{N_j} + \lambda_i N_i - N_j \left(\lambda_j + \frac{4k}{d} \right) - v \frac{\partial N_j}{\partial x} + \frac{4kK \sum M_j^p}{d(1 - \sum M_j/Ms_j)} \left(\frac{M_j}{\sum M_j} \right)$$

$$\frac{\partial M_j}{\partial t} = S_{M_j} + \lambda_i M_i + kN_j - \lambda_j M_j - \frac{kK \sum M_j^p}{(1 - \sum M_j/Ms_j)} \left(\frac{M_j}{\sum M_j} \right), \quad (5)$$

where

S_{N_j}, S_{M_j} = source terms for coolant concentration and surface concentration of fission product j,

N_j, N_i = atom concentration of fission product j and its precursor i in the coolant stream,

M_j, M_i = atom surface concentration of fission product j and its precursor i on the plateout surface,

λ_j, λ_i = decay constant of fission product j and its precursor i,

x, t = axial distance and time coordinates,

k = the mass transport coefficient,

d = the channel diameter,

V = the coolant flow velocity, and

$$\frac{k \sum M_j^p}{(1 - \sum M_j / M_{s_j})} \left(\frac{M_j}{\sum M_j} \right)$$

is an expression for the desorption of fission product j from the surface into the coolant. The PAD code can handle an arbitrary number of species interacting through the summation term.

Figure 4.5 is a plot showing a typical calculated iodine distribution around a reactor circuit. It can be seen that the iodine concentration is greatly reduced by the reactor circuit components. For this case the loop removal rate is considerably greater than the purification system removal rate and, consequently, the total amount of loop plateout is adequately approximated by assuming instantaneous complete plateout in the circuit.

Needed Work. See Section 5.6.

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Fig. 4.5. Calculated iodine distribution in a reactor circuit

4.1.5. Sorption of Iodine in Graphite

It is known that the sorption of iodine by graphite is significant at temperatures around 500°C^(41, 50) and that the degree of sorption is enhanced by the presence of metallic elements in the graphite. Iodine is an electronegative element and reacts chemically with the metallic elements, which are electropositive. The degree of sorption of iodine nuclides in fuel element graphite may thus be appreciable when the graphite contains end-of-life loadings of metallic fission products. Therefore, studies of the sorption of iodine in graphite containing appropriate loadings of metallic elements are needed.

4.2. CONTROL OF METALLIC FISSION PRODUCTS IN HTGR SYSTEMS

The mechanism of the transport of metallic fission product nuclides in an HTGR system is outlined in Fig. 4.6. The basic steps are (1) release from the source, (2) sorption and diffusion in the matrix materials, (3) sorption and diffusion in the graphite, (4) evaporation through the boundary layer, and (5) transport by helium to graphite and metal plateout surfaces.

The sources of release of metallic fission products from HTGR fuel elements containing TRISO-coated particles are (1) fuel contamination in the coatings, (2) fuel contamination in the fuel rod matrix materials, and (3) particles with defective coatings. Release from BISO-coated particles is primarily controlled by the diffusion rate in the kernel for most metallic fission product elements. However, the release of cesium may also be significantly controlled by the BISO coating.

In order to understand and predict the release of metallic fission product nuclides into the helium coolant of an HTGR system, basic data are required in the following areas:

1. Release of metallic fission products from fuel particles.

Fig. 4.6. Transport of metallic fission products in HTGR systems. (Decay also occurs during the delay at each step.)

2. Sorption and diffusion of metallic fission products in matrix material. (Diffusion in the matrix material is conservatively considered to be relatively fast and noncontrolling.)
3. Distribution of metallic fission products between matrix and graphite materials.
4. Diffusion of metallic fission products in graphite.
5. Vapor pressure of metallic fission products sorbed on graphite.
6. Plateout behavior of metallic fission products.

In addition, calculational methods are required for using the basic data to predict the release of the metal nuclides into the coolant.

4.2.1. Calculation of Release of Metallic Fission Products from HTGR Fuel Elements

Three methods exist for use in calculating the release of metallic fission products from HTGR fuel elements:

1. The FREVAP-8 and FREVAP-9 codes, developed under the Fort St. Vrain reactor program,
2. The FIPER I code, developed under the HTGR Base Program. (The FIPER II code is under development under the HTGR Base Program.)
3. The FIPDIG code, developed in England under the Dragon project.

In addition, the SLIDER code, developed under the HTGR Base Program is used as a source code for calculating release from coated fuel particles.

Important features of the codes are compared in Table 4.1. The codes are described more fully in the following sections.

TABLE 4.1

COMPARISON OF FISSION PRODUCT RELEASE CODES

	FIPER I	FIPDIG	FREVAP-8	FREVAP-9
Geometry	One-dimensional plane, cylindrical, and spherical. Also quasi-two dimensional	One-dimensional plane, cylindrical, and spherical	Quasi-two-dimensional (x, z); coupling via the coolant channel, axial diffusion neglected	
Temperature distribution	Diffusion constants can be made temperature, space, or concentration dependent	Diffusion constants can be made temperature dependent or space dependent	Linear temperature distribution	
Interaction of several isotopes considered	Yes, via the Freundlich isotherms	No	Yes, via the Freundlich isotherms	
Time-step automatically adjusted	No	No	Yes, based on the HAMDE approach	
Time integration method	Exact numerical treatment; stable, implicit technique	Exact numerical treatment; stable, implicit technique	Steady state solution for each time-step has been assumed.	Crude approximation for time-dependence (data fitting!)
			Solution of a system of ordinary differential equations by the Milne-Hamming method ⁽⁵¹⁾	
Treatment of space dependence	Three-point difference equations	Three-point difference equations	Analytical solution for a one-region problem	

TABLE 4.1 (cont)

	FIPER I	FIPDIG	FREVAP-8	FREVAP-9
Number of regions allowed	Multiregion	Multiregion	One-region problem	
Source in fuel region	Arbitrary	Arbitrary	Treated by release rate at fuel-graphite interface	
Soret effect considered (temperature gradient driving force)	Yes, in a general form	No	Yes, in a specialized form	
Continuity	$J_i^- = J_i^+$ $C_i^- = \phi_i C_i^+$	$(D\nabla C_i)^- = (D\nabla C_i)^+$ and $(D\nabla C_i)^- = -H_i(C_i^- - C_i^+)$ or $C_i^- = C_i^+$	-----	
Boundary conditions	$\frac{\partial C_i}{\partial x} + \psi_i C_i = \chi_i$	$(D\nabla C_i)^- = -H_i(C_i^- - C_i^+)$	Fuel-graphite Interface: $C_i = C_i^{(0)}$ Graphite-coolant interface: $\frac{\partial C_i}{\partial x} + \psi_i C_i = \chi_i$	Fuel-graphite $\frac{\partial C_i}{\partial x} = \text{constant}$ Graphite-coolant interface: $\frac{\partial C_i}{\partial x} = 0$ with correction
Iteration of concentration dependent quantities for each time-step possible	Yes	No	No	No

4.2.1.1. FREVAP Code

The FREVAP code has been developed for use in estimating the release of longer-lived metallic fission products from HTGR fuel elements. The most recent version used in reactor analyses is FREVAP-8.⁽⁵²⁾ The code takes into account the combined effects of the retention of metallic fission products by fuel particles, the decay of these fission products during diffusion, and their rather strong sorption by the bonding material and graphite of the fuel elements. Release calculations are made on the basis that the loss of fission product nuclides such as strontium, cesium, and barium is determined by their evaporation from the graphite surfaces and their transpiration induced by the flowing helium coolant. The computation has been devised so that changes of fission rate (fuel element power), fuel temperature, and graphite temperature may be incorporated into the calculation. Temperature is quite important in determining release because, in general, release from fuel particles, diffusion in graphite, and loss by evaporation vary exponentially with the reciprocal of the absolute temperature.

For calculating the effect of decay during diffusion, FREVAP-8 normally assumes that steady state diffusion of fission products through the fuel element graphite is attained. However, the code was modified to treat transient behavior in an approximate manner. Failure to treat transient release adequately leads to a conservative (relatively high) estimate of release, since the actual transient release can only approach steady state release as a limit. The treatment of diffusion includes the effect of temperature gradient on diffusion (for the case where the heat of transport for diffusion of a given fission product element is known or can be estimated).

The FREVAP-8 formulation has an additional index, h (or h'), for chemical element species, which is necessary for the treatment of the effect of the competition of various chemical species (elements) for sorption

sites in graphite or amorphous carbon. In FREVAP, the equation which describes the quantity of initial species in the fuel particle as a function of time is

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

where N is the concentration of metal in the particle, t is the time, B is the production rate, λ is the decay constant, and R is the release constant. Values of R are derived from fission product release data and are described by the equation

$$\ln R = \ln R_o - \frac{E_a}{1.987} \left(\frac{10^3}{T} \right) ,$$

where E_a is the activation energy (kcal/mole), R_o is a constant, and T is in $^{\circ}\text{K}$.

The FREVAP code is not as accurate as the FIPER code and requires much more computer time. FREVAP does not treat transient behavior adequately. It uses the release constant concept (see above) in calculating the release from particles; however, the SLIDER code, which uses a diffusion concept, can be incorporated into the FREVAP code and used in place of the release constant source term.

FREVAP-9,⁽⁵³⁾ a modification of FREVAP-8, takes into account the effect of a relatively fast process of diffusion (see Section 4.2.7) as well as the nonsteady state of a slower diffusional process.

Needed Work. No additional development work on the FREVAP code is anticipated.

4.2.1.2. FIPER Code

The FIPER code⁽⁵⁴⁾ is an exact formulation for predicting the fraction of metallic fission products released into the coolant gas. The diffusion equation in space and time dimensions is solved numerically, taking into account as driving forces both the gradient of the chemical potential and that of the temperature field.

There are two forms of the FIPER code: FIPER I, which treats diffusion in one space and one time dimensions, and FIPER II, which treats diffusion in two space and one time dimensions. FIPER I can also be used in a quasi-two-dimensional mode by coupling segments via the coolant concentration.

The FIPER code is a mathematically precise description of the release of metallic fission products from an HTGR fuel element. It treats complex aspects of the problem, such as transient behavior. It utilizes either a release constant concept (see Section 4.2.1.1) or a diffusion concept for calculating the release from particles. The two-dimensional code FIPER II, now under development, will describe axial transport as well as radial transport.

FIPER I has several advantages over FREVAP-8 (see Section 4.2.1.1). FREVAP-8 tends to overestimate release values primarily because it does not treat transient behavior adequately. Furthermore, the computer time is less for FIPER.

The FIPER I code has undergone some technical modifications to produce a fast, flexible, quasi-two dimensional code (FIPER X) for reactor calculations.⁽⁵⁵⁾ It has been rewritten in a modular form so that the broadest possible range of one-dimensional (or quasi-two dimensional) diffusional problems can be solved. The program has been separated into parts that pertain to the actual solution of the partial differential equations, the geometry, the mesh size, the time step, etc., and parts that contain the physical and chemical parameters describing the adsorption and diffusion of fission products. To achieve flexibility for different release calculations, the program is separated into two distinct parts, the MASTER code and the AUXILIARY code. The MASTER code contains the routines that are problem independent. The AUXILIARY code is composed of the routines that are strongly dependent on the physical problem statement.

The FIPER II code, which solves the diffusion equation in two space dimensions, is being developed for several reasons:

1. Fission product transport analyses should take the complex structure and composition of reactor graphite into account. Graphite is a heterogeneous material, and the metal atoms wander along diffusion paths of this material.
2. The actual reactor geometry is a three-dimensional geometry. By comparing release results for a one- and two-dimensional pseudo-geometry, something can be learned about the "geometrical" error of a one-dimensional calculation.
3. Multiregion nonlinear diffusion problems in two-space dimensions have not been studied before with high-speed digital computers.

The work on the MASTER part of this code, which solves the finite difference equations describing the diffusion in two-space dimensions, has been completed. Some results for the time development of the concentration profile in an r-z cylinder symmetry are presented in Ref. 56. At the present time, the AUXILIARY code is being developed, which allows for flexible boundary conditions and for a general form of the input in form of tables for diffusion and absorption coefficients. A two-dimensional Fermi-Dirac distribution is used to describe the change of physical parameters, such as the diffusion coefficient, at a two-dimensional corner. Next, a change of the distribution coefficient ϕ at such a corner must be formulated. After the AUXILIARY code to FIPER II is completed, the code will be used for a detailed comparison between measured and calculated concentration profiles and release currents.

In addition to its use in analyses of fission product transport in reactor systems, the FIPER I code has recently been applied in (1) the analysis of concentration profiles in the Peach Bottom D06-01 fuel element graphite, ^(57,58) and (2) the analysis of a system characterized by paths of fast and slow diffusion. ⁽⁵⁶⁾

Needed Work. The FIPER code work is important and should be continued. Areas of work needed include:

1. For FIPER X, optimization of computer time with required accuracy.
2. For FIPER II, (1) development of routines for handling input data, (2) comparison of analytical and numerical results, and (3) programming for reactor application.

4.2.1.3. FIPDIG Code

The FIPDIG code, ^(59, 60) developed in England for the Dragon project, is described in reports by Preinreich. As stated in his reports, the code can calculate concentration profiles of decaying fission products in slab, cylinder, or spherical geometry for any number and arrangement of fuel zones and fuel-free zones, allowing for boundary conditions with continuous concentrations as well as discontinuous concentrations (evaporation boundaries). The diffusion coefficient is constant in any given zone. The code also calculates steady state diffusion profiles. For both time dependent and steady state solutions, the fractional release, the R/B value, and the releases in μCi are calculated.

Like the FREVAP and FIPER codes, the FIPDIG code takes into account the temperature and concentration dependence of the diffusion step by employing finite difference techniques and copes with the inner boundary condition by using a fuel release factor to simulate the input current. At the coolant boundary the methods are different. FREVAP and FIPER are designed to use graphite adsorption isotherms and a diffusion step in the gas of the boundary layer. FIPDIG has an evaporation coefficient which is an empirical function of temperature derived from in-pile measurements of release. FIPDIG does not offer any apparent advantage over the other methods.

4.2.2. Calculation of Release of Metallic Fission Products from Coated Fuel Particles

Formulations, described in Section 4.1.3, can be used to treat the release of metallic fission products from coated fuel particles. In addition, the FIPER code (see Section 4.2.1.2) can be used for this purpose. The SLIDER concept has been profitably applied to laboratory fission product release data, resulting in diffusion coefficient data for strontium and barium in pyrolytic carbon coating and carbide kernel materials.⁽⁶¹⁾

The formulations (the SLIDER code in particular) are valuable for use in treating the release of metals (such as strontium and barium) from BISO-coated particles. On the other hand, the formulations are of little use in treating release from TRISO-coated particles. The reason is that in the case of TRISO-coated particles, the silicon carbide coating is a very effective barrier for strontium and barium, and release by diffusion through the coating is unimportant compared with other sources of release (such as contamination and broken particles).⁽⁶²⁾ Also, little input information is available on the diffusion and solubility behavior of strontium, barium, and cesium in silicon carbide (see Section 4.2.4).

Needed Work. The SLIDER code or the FIPER code should be further applied to laboratory fission product release data for the purpose of measuring diffusion coefficient data for strontium, cesium, and barium in coating and kernel materials (see Section 4.2.3.2). The procedure in this work is (1) to anneal irradiated BISO-coated particles, (2) to measure the fractional release of metallic fission products as a function of time, and (3) to apply the SLIDER or FIPER formulations to the results. (This procedure does not work in the case of TRISO-coated fuel particles because, as mentioned above, the degree of release by diffusion of metals through the TRISO coating is too small to be measured.) Earlier kernel diffusion coefficient data measured by this procedure⁽⁶¹⁾ are questionable because the anneals were not carried to the point where the release was clearly due to diffusion from the kernel and not due to release of recoils in the coating.

4.2.3. Release of Metallic Fission Products from Coated Fuel Particles

The release of metallic fission products from coated particles has been studied extensively. In early studies at BMI,⁽⁶³⁾ the release of fission product metals for pyrolytic carbon coated particles was measured. This work indicated pyrolytic carbon to be more effective in retaining fission gases than in retaining fission product metals. Apparent diffusion coefficients for metals in pyrolytic carbon were reported.

The release of metallic fission products was studied in 6-hr post-irradiation anneals at ORNL.⁽²³⁾ No effects of pyrolytic carbon coating characteristics were apparent. No variation in fission product release as a result of burnup (up to 10% U) was observed. Addition of a silicon carbide coating resulted in a marked improvement in the retention of metallic fission products. Silicon carbide and pyrolytic carbon coatings appeared to be equally effective in retaining cesium in the short-term experiments.

British investigators pointed out the value of a silicon barrier layer for retaining fission product metals.⁽⁶⁴⁾

In an ORNL investigation, sweep capsule components were examined for metallic fission product release.⁽⁶⁵⁾ It was concluded that (1) barium and strontium are not retained by a pyrolytic carbon coating, but are retained if a silicon carbide layer is added, (2) cesium traveled furthest from the fuel, (3) a gas gap did not help in stopping the more volatile metallic fission products, and (4) no metallic fission products got beyond the capsule and most were trapped in the graphite holder.

Annealing studies of coated fuel particles irradiated to high burnups of U-235 were performed at ORNL to determine their ability to contain metallic fission products.⁽⁶⁶⁾ Annealing times were in the range of 6 to 32 hr. The release of metallic fission products from samples with silicon carbide layers was much lower than that from other samples. The release of cesium

was relatively low for both pyrolytic carbon and silicon carbide coated particles.

The release of metallic fission products from BISO- and TRISO-coated fuel particles during irradiation in capsule experiments was determined at GGA by gamma spectrometric and radiochemical analyses of irradiated particles and capsule components. The in-pile release results for the various capsule experiments may be summarized as follows:

1. Fractional release values of 0 to 0.08 for Cs-137, 0.22 to 0.26 for Ba-140, and 0.40 to 0.50 for Sr-89 were observed for BISO-coated carbide particles irradiated to around 7% FIMA burnup at around 1300°C in the P12 capsule, which ran at full power for 83 days. (67)
2. Fractional release values of 0.02 to 0.21 for Cs-137, 0.02 to 0.19 for Ce-144, and 0.12 to 0.42 for Sr-89 were observed for BISO-coated carbide particles irradiated for 56 days to 8% FIMA burnup at around 1200°C in the P13E capsule. (68)
3. Fractional release data for Ba-140, Ce-141, Ce-144, Cs-137, Sr-89, Sr-90, and Zr-95 were determined for TRISO- and BISO-coated carbide particles irradiated for 69 full power days in the P13F capsule. Burnup for the TRISO-coated particles (Lot 3332-145E) was around 14% FIMA, and burnup for the BISO-coated particles (Lot 3222-129E) was around 3% FIMA. The data indicate that (a) cesium was retained to an equal degree by the TRISO- and BISO-coated particles at 1300°C (the cesium release data are questionable; see below), and (b) barium and strontium were retained to a much greater degree by the TRISO-coated particles. (69, 70)
4. The releases of Cs-137, Ce-144, Sr-89, and Eu-155 from TRISO-coated carbide particles during the P13H capsule irradiation were determined. Burnups as high as 23% FIMA were obtained, and the time at full power was 111 days. In general, particles irradiated at around 925°C showed good retention of fission product metals, whereas those irradiated at around 1250°C showed some coating failure. (71)

5. Releases of Cs-137, Ce-144, and Sr-90 were measured for (diluent-type)* BISO-coated carbide particles irradiated to about 50% FIMA burnup in the P13K capsule, which ran for 93 full power days. At 900°C, observed fractional releases were 0.05 to 0.20 for Cs-137, 0.02 to 0.05 for Ce-144, and 0.74 to 0.94 for Sr-90. At 1300°C, observed fractional releases were 0.28 to 0.79 for Cs-137, 0.34 to 0.70 for Ce-144, and 0.89 to 0.99 for Sr-90. (72,73)
6. The fractional release of Cs-137 from intact BISO-coated carbide particles during irradiation in the P17, P18, and P19 capsules was determined. Irradiation times were 140 days for P17, 231 days for P18, and 196 days for P19. Burnups as high as 26% FIMA were achieved. The data show that particles irradiated at ~900°C retained essentially all of the Cs-137 and those irradiated at ~1200°C released up to 0.66 fraction of the Cs-137. (74)
7. Cesium release values for TRISO-coated carbide particles irradiated in the P22 capsule were all less than 0.05 (the uncertainty of each determination), indicating the effectiveness of the TRISO coating in retaining cesium. Capsule P22 was a full-exposure test of TRISO-coated particles. It received an estimated fast fluence of 7.8×10^{24} n/cm² with an estimated burnup of 23% FIMA, and the time at full power was 252 days. (75)

The in-pile release data for the P13F and P13H capsules were determined by measuring the metallic fission products collected in the graphite thimbles and crucibles used to hold the particles. It was assumed that all the metallic fission products which escaped the particles were trapped in the thimbles and crucibles. This assumption may not have been valid for cesium. In the other measurements, the in-pile release data were determined by gamma spectrometric analyses of irradiated particles.

In a study at Seibersdorf of cesium release from irradiated uranium oxide kernels coated with TRISO-type coatings, low cesium release was observed for intact particles and high cesium release was observed for

* Fuel particles containing diluents other than thorium in the kernels.

broken particles.⁽⁷⁶⁾ This work indicated the effectiveness of the TRISO-type coating in retaining cesium.

Seibersdorf investigators studied the distribution of metallic fission products in fuel particle coatings.⁽⁷⁷⁾ It was reported that in particles with a plain pyrolytic carbon coating, breakthrough of cesium occurred after 99 irradiation days over a temperature range from 1260° to 1290°C. Measurements on particles with TRISO-type coatings showed that silicon carbide is effective as a barrier for metallic fission products.

A British investigator compiled data on the release of fission product metals.⁽⁷⁸⁾ The data were derived from the Dragon reactor and irradiation loops.

Based on data given in Ref. 31, the diffusion coefficient for strontium in pyrolytic carbon is $\sim 10^{-10}$ cm²/sec at 1000°C and that for cesium in pyrolytic carbon is $\sim 10^{-13}$ cm²/sec at 1000°C. Accordingly, one would expect pyrolytic carbon to be more effective in retaining cesium than strontium. Using these diffusion coefficient values, one can estimate delay times of ~ 500 days at 1000°C for cesium in a 50- μ m pyrolytic carbon coating and ~ 0.5 day at 1000°C for strontium in the coating. The relationship used to calculate these delay times was $x^2 = 6Dt$, where x is the coating thickness, D is the diffusion coefficient, and t is the delay time.

The above studies have yielded data that are useful in predicting the release of metallic fission products from coated fuel particles in HTGR systems and, in addition, have led to the following conclusions:

1. The release of metallic fission products from coated fuel particles is a diffusion controlled, time and temperature dependent phenomenon.
2. TRISO (silicon carbide type) coatings are much more effective than BISO (pyrolytic carbon type) coatings in retaining metallic fission products at normal reactor times and temperatures.
3. Pyrolytic carbon is more effective in retaining cesium than strontium.

4. The refractory fission product metals such as zirconium are found outside coated particles only as a result of fuel contamination.

It was thought earlier that silicon carbide and pyrolytic carbon coatings were equally effective in retaining cesium. However, this belief was based primarily on the results of relatively short-time release measurements on pyrolytic carbon coated particles. These measurements did not allow time for cesium breakthrough to occur.

Additional information on TRISO- and BISO-coated particles is discussed in the following sections.

4.2.3.1. Release of Metals from TRISO Particles

The release of fission product metals from TRISO-coated particles is due to:

1. Uranium or thorium contamination in or on the surface of the outer pyrolytic carbon coating.
2. Particles with defective silicon carbide layers. (A defective silicon carbide layer is one that is either incomplete, cracked, porous, or completely absent.)
3. Release from the kernel and diffusion through the coating.

Under reactor temperature conditions, release by diffusion through the TRISO coating is so low that release due to contamination and defective coatings predominates. Thus, in reactor analyses using the FREVAP and FIPER codes, the calculated direct release from intact TRISO coated particles is small compared with the release from the failed fraction.

For TRISO particles with failed coatings, releases of the metallic nuclides such as Sr-90 and Ba-140 are normally calculated using release constants derived from experimental measurements of the release of these metallic nuclides from BISO (pyrolytic-carbon) coated particles, which (as described in the following section) is essentially the same as the release from the carbide substrate.

An attempt was made to correlate uranium contamination with the release of strontium from TRISO-coated particles.⁽¹⁵⁾ A correlation was not found, apparently because the strontium release came from several different sources.

The release of Cs-137 during postirradiation anneals of TRISO-coated UO_2 particles was studied at ORNL.^(29,79) The particles were annealed in steps of 100°C (10 min at each step), starting at 1300°C and ending at 2200°C . Eight of the ten particles tested showed high release of Cs-137, while the two smallest particles released no detectable Cs-137. Results indicated failure of silicon carbide layers underneath the outer pyrolytic carbon layers, which remained intact as indicated by Kr-85 release.

Data obtained by the standard technique of diffusion annealing indicate that the diffusion coefficients for strontium and cesium in silicon carbide are only one or two orders of magnitude lower than corresponding coefficients for these elements in pyrolytic carbon.⁽³¹⁾ Therefore, the low degree of transport can be accounted for only by a relatively low solubility of these metals in the silicon carbide. (See Section 4.2.4.)

Needed Work. The rate-determining step in the release of metallic fission products from the kernels of intact TRISO-coated particles is the diffusion through the silicon carbide barrier layer. The calculation of the rate at which the metallic species are released requires knowledge of the permeability coefficients or a separate determination of the diffusion coefficient and the solubility. These data should be obtained as a function of temperature and burnup or fast flux. Continued utilization of laboratory annealing experiments is needed to evaluate the release behavior of lightly and highly irradiated TRISO-coated particles, loose and bonded. (The level of metallic release is a good measure of the integrity of TRISO coatings.) Continued studies of the relationship between metallic release behavior and the level of fuel contamination are needed. Further in-pile release experiments are needed to evaluate the performance of TRISO-coated particles under long-term irradiation conditions (see Section 4.8.2.)

4.2.3.2. Release of Metals from BISO Particles

The release of barium and strontium from BISO-coated fuel particles is controlled by the diffusion of these metals out of the fuel kernels. (62,80) This conclusion is supported by the results of postactivation fission product annealing experiments, which yielded diffusion coefficients for barium and strontium in both the fuel kernel and in the pyrolytic carbon. (61) Generally, it is found that the diffusion coefficients for both barium and strontium in pyrolytic carbon are about $10^{-9} \text{ cm}^2/\text{sec}$ at 1400°C . By contrast, the diffusion coefficients of these species in the kernel are in the range 10^{-13} to $10^{-16} \text{ cm}^2/\text{sec}$, depending upon the properties of the kernels of individual samples. In any event, the minimum of four orders of magnitude difference between the diffusion coefficients in the kernel and in the coating is sufficient to establish that the release rate is controlled by diffusion out of the kernel. (61)

Few data are available on the effect of burnup or fast flux on the rates of diffusion in the kernels. The diffusion coefficients in the coating do not appear to change by more than a factor of two, if at all. (81)

In FREVAP and FIPER calculations (see Sections 4.2.1.1 and 4.2.1.2), the relationship normally used in calculating the quantity of a metallic species in BISO-coated particles as a function of time is

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

where N is the concentration of metal in the particle, t is the time, B is the production rate, λ is the decay constant, and R is the release constant. (82) Values of R are derived from fission product release data and are described by the equation

$$\ln R = \ln R_o - \frac{E_a}{1.987} \frac{10^3}{T} \quad ,$$

where R is in units of fraction/day, E_a is the activation energy (kcal/mole), R_o is a constant, and T is in $^\circ\text{K}$. Present values of $\ln R_o$ and E_a for strontium

are 12.94 and 45 kcal/mole. The release constant data are obtained from (1) in-pile release (or retention) measurements on fuel particle samples and (2) postirradiation annealing experiments on highly irradiated particles. These measurements are described in Ref. 82.

A two-release-constant model presently under investigation by Gulf General Atomic⁽¹⁶⁾ is defined by the coupled equations

$$\begin{aligned}\frac{dn_1}{dt} &= B_1 - R_1 n_1 - \lambda n_1, \\ \frac{dn_2}{dt} &= B_2 + R_1 n_1 - R_2 n_2 - \lambda n_2,\end{aligned}\tag{6}$$

where n_1 and n_2 are the total inventories in regions 1 and 2 of the fuel particle, B_1 and B_2 are the respective birth rates, R_1 and R_2 are the respective release constants, and λ is the decay constant. (The single-release-constant model normally used in reactor calculations is, of course, defined by the first equation.) It is believed that the two-release-constant model would provide a more accurate approximation to the actual situation and would therefore be useful for reactor calculations of the release from fuel particles.

The release behavior from BISO-coated particles can also be predicted with the SLIDER or FIPER code (see Sections 4.2.1.2 and 4.2.2) by treating the release from the fuel particles as a multizone diffusion problem.

Recent work at ORNL (see Refs. 23, 66, and ORNL-4200 of Ref. 40), has shown that the release of cerium and yttrium is significantly lower from pyrolytic-carbon-coated particles with uranium oxide kernels than from similar particles with uranium carbide kernels. The release of other metals, such as strontium, cerium, and barium, is not significantly different for the two types of coated particles.

German workers investigated the distribution of fission product metals in the coating of irradiated particles and in the graphite which surrounded the fuel particles during irradiation.⁽⁸³⁾

Needed Work. The release of metallic fission products from BISO-coated particles over long time periods (months) needs to be studied. This work would lead to improved release models. The effect of burnup or fast flux on the release of strontium and barium can be evaluated by carrying out postactivation annealing experiments on lightly and highly irradiated particles. In-pile release studies, such as those being conducted in P-capsules and in the Peach Bottom test elements (see Section 4.8.2), should be continued. Further studies are needed on the relative merits of coated uranium oxide kernels and coated uranium carbide kernels.

4.2.4. Diffusion of Metallic Fission Products in Coating Materials

Diffusion coefficient data for important fission product metals in isotropic pyrolytic carbon and in silicon carbide have been measured at Gulf General Atomic.

Method. Graphite wafers were coated with thin layers of pyrolytic carbon (or silicon carbide) and were annealed in a bed of graphite powder containing tagged metals. Oxide or chloride metal sources were used. After the anneal, the coatings were ground off with strips of emery cloth (or diamond-coated cloth in the silicon carbide coating studies). The strips were then counted, and concentration profiles were derived from the counting data. Diffusion coefficient data were obtained by analysis of the counting data. (For details, see Ref. 84.)

Chemical diffusion measurements and self-diffusion measurements were carried out using constant chemical potential sources. In the self-diffusion experiment, pyrolytic carbon wafers were preloaded uniformly with the metal and diffusion anneals were carried out with the preloaded samples in contact with a source of tagged metal at tracer concentration.

Diffusion coefficient data for strontium and barium in pyrolytic carbon have also been obtained by application of the SLIDER code to laboratory release data (see Section 4.2.3.2).

Diffusion Coefficient Data for Pyrolytic Carbon. Diffusion coefficient data were measured for cesium, strontium, barium, and europium in pyrolytic carbon in the temperature range 1000° to 1700°C . These data are reviewed in Ref. 31.

As stated in the review, it is difficult to correlate the diffusion behavior of the different elements in pyrolytic carbon. Self-diffusion coefficients and chemical diffusion coefficients seem to vary little for europium and strontium but significantly for barium. The concentration dependence of the diffusion coefficients is pronounced for barium where there is a large spread in the concentration (approximately three orders of magnitude). A change of one order of magnitude in concentration does not affect the diffusion coefficient for strontium. The few data points obtained for europium and cesium do not allow a comparison at the different concentrations levels.

However, correlation seems to exist between the diffusion coefficients for the different metals and their atomic radii.⁽³¹⁾ It has been found that the diffusion coefficients for an element show a linear relationship when plotted against the reciprocal atomic radius of the element. This allows the prediction of diffusion coefficients for different metals in isotropic pyrolytic carbon.

A comparison of the diffusion coefficients for strontium and barium at 1400°C with values calculated from release measurements⁽³¹⁾ shows good agreement in the barium data but a difference of one order of magnitude in the strontium data. Diffusion coefficients observed for cesium are orders of magnitude lower than those for strontium.

All experiments were carried out using either chloride or oxide sources, and some of the experiments need to be repeated using metal or carbide sources. In a reactor one would expect to have the metals or metal carbides as the diffusion source.

Diffusion Coefficient Data for Silicon Carbide. Diffusion coefficient data for strontium and cesium in silicon carbide have been determined.⁽³¹⁾

The measurements for strontium were carried out using a constant chemical potential source of 1×10^{-3} g Sr/g C. From the temperature dependence of the diffusion coefficients, a diffusion equation was calculated:

$$D_{\text{Sr}} = 0.25 \exp \left(- \frac{68,700}{RT} \right) \text{ cm}^2/\text{sec} .$$

A comparison of the migration of strontium in pyrolytic carbon and in SiC shows that the diffusion coefficients for strontium in SiC are only about two orders of magnitude lower than those for strontium in pyrolytic carbon. Therefore, the effectiveness of SiC as a diffusion barrier for strontium is probably the result of the relatively low solubility of strontium in SiC rather than the result of a very low diffusion coefficient for strontium in SiC.

Experiments on the diffusion of cesium in SiC show a very wide scatter in the data.

Needed Work. Additional work on the diffusion of metals in coating materials is needed in order to improve the accuracy of SLIDER or other calculational methods. Existing diffusion data for metals (in particular strontium and cesium) in pyrolytic carbon should be verified by diffusion experiments using a carbide source, to supplement information derived using oxide or chloride sources in previous studies. Further studies of the diffusion of strontium and cesium in silicon carbide are needed to substantiate existing data and obtain new data. The relative solubility of strontium and cesium in silicon carbide also should be measured to provide input data for calculational methods.

4.2.5. Sorption of Metallic Fission Products on Activated Carbon and Binder Materials

The pseudoisopiestic method (see Section 4.2.8) has been used to obtain cesium sorption measurements on several carbon matrix materials (fuel particle binder). Isotherms measured at 800° , 900° , and 1000°C show that the materials tested were virtually identical in their cesium sorption

characteristics.⁽⁷¹⁾ At 900°C and a cesium pressure of 10^{-5} atm, cesium loadings on the binder material were around 100 mg Cs/g C.

Strontium sorption measurements on several other carbon matrix materials have been performed using the mass spectrometer.⁽⁷⁵⁾ These materials showed more variability in their sorptive capacities. There was an apparent correlation with the sulfur content of the material (some samples contained almost 1% sulfur).

In other laboratory studies, the effect of fast neutron flux on the cesium sorption capacity of charcoal (activated carbon) at conditions of 1000°C and 10^{-5} atm cesium has been measured using the pseudoisopiestic method. Typical data were obtained with a sample of MI-1 charcoal irradiated at 700°C to an exposure of 3.8×10^{21} nvt ($E > 0.18$ MeV).⁽⁸⁵⁾ The cesium loadings of the sample were 85 mg Cs/g C before irradiation and 58 mg Cs/g C after irradiation, indicating a decrease in sorptive capacity due to the irradiation. On the basis of the irradiation effects study, a factor of two decrease in sorptive capacity of binder is sometimes assumed in reactor calculations over a 6-yr period.

It is concluded from these studies of the sorption characteristics of binder materials (1) that the materials have a high sorptive capacity for fission product metals and (2) that irradiation reduces the sorptive capacity of the material.

Needed Work. Additional work is needed, in particular to study the sorption behavior of strontium on charcoal and binder materials. More information is needed on the effect of irradiation on the sorptive capacity for metals of these materials.

4.2.6. Distribution of Metallic Fission Products Between Graphite and Fuel Rod Matrix

An important input parameter for FREVAP and FIPER calculations is the parameter ϕ , the sorption ratio, defined as the ratio of the metal concentration between the graphite and fuel rod matrix:

$$\phi = \frac{\text{conc. of metal in fuel matrix}}{\text{conc. of metal in graphite}}$$

Values of ϕ are obtained from data on the sorption behavior of metals on carbon matrix materials and graphite. *

The value of the sorption ratio for matrix graphite obtained from cesium sorption data⁽⁷¹⁾ is about 200. This ratio showed no appreciable change for measurements at 10^{-5} and 10^{-8} atm cesium pressures and at 900° and 1000°C .

Values of the sorption ratio for strontium can be derived from sorption data given in Ref. 75. For pure carbon materials, the sorption ratio is indicated to be approximately constant and equal to 200. For the less pure materials, the sorption isotherms are not parallel to those for graphite and consequently the sorption ratio is a function of temperature and strontium loading.

In addition, sorption ratios of the order of 100 were derived from the results of strontium permeation experiments.⁽⁸⁶⁾ In these experiments, strontium (tagged with Sr-85) diffused from matrix source material into graphite, and the sorption ratios were calculated from strontium concentrations in the matrix material and the graphite at the source boundary.

The ratio tends to decrease with irradiation owing to a decrease in the sorptive capacity of the matrix material and an increase in the sorptive capacity of the graphite. (See Sections 4.2.5 and 4.2.11.)

Needed Work. There is a need for studies in this area to permit refinement of the strontium sorption ratio data.

4.2.7. Diffusion of Metallic Fission Products in Graphite

Early investigators of the diffusion of metals in graphite generally treated graphite as a homogeneous material and applied Fick's law to their data without regard to concentration and structure dependence. This procedure has led to considerable scatter in reported data (see, for example, Ref. 87.)

*The sorption behavior of metals on matrix materials and graphite is discussed in Sections 4.2.5 and 4.2.8.

Graphite is a material which consists of crystallites bound together with a more or less graphitized binder carbon into a rather porous structure. In 1965, Faircloth, Pummery, and Rolls⁽⁸⁸⁾ observed diffusional transport of cerium, barium, and strontium in several graphites. Their conclusion was that "the movement of these metals through well-formed polycrystalline graphite and pyrographite occurs by activated surface diffusion, combined possibly with vapour phase migration through microstructural defects." Thus, the diffusion of metals in graphite involves complex phenomena, and understanding these phenomena will be possible only through a concerted scientific effort.

In the case of HTGR systems, the diffusion behavior of the following two metals in graphite is of particular concern:

1. Strontium, because of 51-d Sr-89 and 28-y Sr-90, whose release into the coolant could create safety problems
2. Cesium, because of 30-y Cs-137, whose release into the coolant could lead to maintenance problems.

The diffusion behavior of barium is also of some concern because of 12.8-d Ba-140 (and its daughter 40-h La-140), whose release into the coolant could lead to maintenance problems. However, results (for example, see Ref. 89) indicate that because of its short half-life and relatively low diffusivity, Ba-140 decays in the graphite and never reaches the coolant. Other metals are generally not of concern (except perhaps for europium):

(1) because they have low yields or short half-lives, or (2) because they are essentially immobile in graphite at normal reactor temperatures (see Section 2). Flowers has considered the various diffusion (and evaporation) processes in the transport of cesium, strontium, and barium as well as other fission products.⁽⁹⁰⁾

Gulf General Atomic Studies of Diffusion of Metals in Graphite.

A study of the diffusion of metallic fission products in graphite is being conducted at Gulf General Atomic. The purpose of this work is (1) to develop a better understanding of the diffusion behavior of metals in

graphite, and (2) to obtain accurate diffusion coefficient data for use in predicting the release of metallic fission products in HTGR systems. Since diffusion coefficients are probably dependent on graphite structure, information gained from this work is of value in selecting graphite for reactor use.

The phenomena observed in the experimental studies at Gulf General Atomic are summarized in the following:

1. Plane source studies have exhibited "tails" in plots of activity versus the square of the penetration distance. More than one diffusion coefficient or a concentration dependent diffusion coefficient is required to explain these data.
2. Both microprobe and autoradiographic investigations have demonstrated the localization of the diffusing metal.
3. Permeation experiments have demonstrated transport of material in a much shorter time than is credible with a single diffusion coefficient derived from the steep portion of the profiles close to the source.
4. Test results and theoretical considerations have indicated that rates of metal diffusion in graphite are concentration dependent. However, it is clear, from results of the microprobe and autoradiographic studies, that concentration dependence alone will not adequately explain the complexity of the diffusion process.

These observations are described more fully below.

The methods used at Gulf General Atomic for studying the diffusion of metals in graphite include:

1. A plane source method. A source of tagged metal (such as graphite impregnated with Sr^{85} -tagged strontium) is sandwiched between two graphite specimens and annealed at desired temperatures. After the anneal, concentration profiles are determined by analyzing sections of the graphite. Diffusion coefficient data can be derived from the profiles (where the profiles have the ideal shape). (For details, see Ref. 91.)
2. A permeation method. This method involves (1) annealing a source material (such as charcoal impregnated with tagged strontium) in a graphite crucible, (2) measuring the evaporation rate of strontium from the crucible, and (3) determining strontium profiles across the crucible wall. Once the steady state evaporation rate (i. e. , the flux) and the concentration profiles

are known, a permeation coefficient* can be calculated.
(For details, see Ref. 72.)

3. A microprobe method. An electron microprobe is used to measure the distribution of a diffusing metal on the surface of a section cut from a graphite diffusion specimen.
4. An autoradiographic technique. An autoradiograph is used to determine the distribution of metal on the surface of a section cut from a graphite diffusion specimen.

Review of Gulf General Atomic Studies of the Diffusion of Strontium in Graphite. In studies at Gulf General Atomic, evidence for the existence of a relatively fast component of diffusion was obtained in plane source experiments. A typical concentration profile for strontium in graphite obtained using this technique is given in Fig. 4.7. The data show an initial sharp decrease in concentration with penetration depth and a long tail extending into the interior of the graphite specimen. This long tail is indicative of a fast component of diffusion. On a plot such as Fig. 4.7, a simple diffusion mechanism would be represented by a straight line. A more detailed discussion of the fast component of diffusion and a description of attempts to mathematically analyze multicomponent diffusion are given in Ref. 91.

Because of the difficulty of interpreting concentration profile data obtained in the plane source experiments and the need to determine the importance of the fast component of diffusion in governing the release of strontium from HTGR fuel elements, an experimental program utilizing the permeation method (see the above description) was undertaken to measure the permeation coefficients for strontium in graphite. This

* In earlier reports the coefficient measured in permeation experiments was designated "an effective diffusion coefficient" (see, for example, Ref. 72). However, the designation "permeation coefficient" more clearly distinguishes this coefficient from the fundamental (Fick's law) diffusion coefficient, which applies in both steady-state and transient situations. The permeation coefficient is measured under steady-state conditions and applies, without qualification, only to a steady-state situation.

Fig. 4.7. Dependence of the logarithm of the strontium concentration upon the square of the penetration distance. Circles are experimental data, and dashed lines represent independent, simultaneous diffusion problems.

experimental program is proving useful; results are described in Refs. 57, 72,* 73, and 92.

It is quite significant that results of the permeation experiments show (1) the permeation coefficients for strontium in H-327 graphite at low concentrations are very low (in the range 10^{-11} to 10^{-10} cm²/sec), and (2) delay times are only a few hours. If one assumes the permeation coefficient to be the diffusion coefficient for strontium in H-327 graphite, these two observations would appear to be inconsistent on the basis of the following equation, which is used to estimate delay times for diffusion of a species through a slab:

$$x^2 = 6 D t_d ,$$

where x is the slab thickness, D is the diffusion coefficient, and t_d is the delay time.

A low permeation coefficient (or steady-state diffusion coefficient) and short delay time are consistent for a system where the strontium diffuses predominantly in high-diffusivity paths that occupy a small fraction of the cross-sectional area of the graphite. The applicable relationship is

$$P = A_e D_p ,$$

where P is the permeation coefficient, A_e is the effective fractional area occupied by the high-diffusivity path, and D_p is the diffusion coefficient for strontium in the path.

Accordingly, it appears that under the conditions of the permeation measurements, strontium diffuses through graphite along paths of high diffusivity and that this mode of diffusion is predominant at all times over diffusion in the bulk graphite.

* The value of the permeation coefficient (effective diffusion coefficient) (1×10^{-21} cm²/sec) given on page 42 of Ref. 72 is incorrect. The correct value is 1×10^{-10} cm²/sec.

A high-pressure test system was set up to study the diffusion of strontium in graphite under high pressure.⁽⁷³⁾ The results exhibit no apparent effect of pressure on concentration profiles, indicating no apparent pressure dependence. However, the experimental profiles do not provide conclusive evaluation of transport rates.

Another important investigation is the study of the concentration dependence of the diffusion of strontium through graphite. Permeation experiments have already demonstrated a two-order-of-magnitude change in the permeation coefficient with concentration.⁽⁸⁶⁾

The autoradiographic studies are providing important results on the diffusion of metals in graphite. One interpretation of these results is that the graphite diffusion matrix can be looked upon as a series of various-size reservoirs and pipes constituting the flow network. In the autoradiograph studies, strontium has been observed to be localized. It is obvious that taking a cross section of the matrix to get the average content of the diffusant will not provide a physical description of the flow through the network. Correlation of the principal features of the graphite matrix with the localized metal might provide insight into the diffusion process.

A system characterized by paths of fast and slow diffusion was treated by application of the FIPER code.⁽⁵⁶⁾ Two-region cylindrical geometry was assumed. It was found that for a suitable choice of R_1/R_2 and D_1/D_2 (where R_1/R_2 is the ratio of radii for the inner and outer regions and D_1/D_2 is the ratio of diffusion coefficients for the two regions), the release current is initially determined by the inner region ($D_1 \gg D_2$) until finally, by virtue of its larger cross section ($R_1 \ll R_2$), the outer region dominates the release current.

Diffusion Coefficient Data. Diffusion coefficient data, as presently accepted for use in FREVAP and FIPER calculations of the release of the important fission product elements strontium, barium, and cesium, are described by equations of the form

$$\ln D = \ln D^0 - \left(\frac{Q}{R}\right) \frac{10^3}{T} ,$$

where D is the diffusion coefficient in cm^2/sec , and Q is the activation energy in kcal/mole. The constants in this equation which apply to the three metallic elements are given in Table 4.2.

Table 4.2
DIFFUSION COEFFICIENT DATA FOR
STRONTIUM, BARIUM, AND CESIUM
IN FUEL ELEMENT GRAPHITE

Element	$\ln D^0$ (cm^2/sec)	Q (kcal/mole)
Sr	5.7	60
Ba	7.4	70
Cs	-1.0	40

The reference diffusion coefficient data for strontium, given in Table 4.2, were selected several years ago for use in reactor calculations. These data are based on (1) a diffusion coefficient of $2 \times 10^{-8} \text{ cm}^2/\text{sec}$ at 1000°C , selected as the best estimate on the basis of data existing at the time, and (2) an activation energy of 60 kcal/mole observed by Bryant et al. ⁽⁹³⁾ The barium data are derived from work performed at Gulf General Atomic and reported in Refs. 6 and 94 and an activation energy of 70 kcal/mole observed by Bryant et al. ⁽⁹³⁾ The cesium data were taken from the work of Bryant et al. ⁽⁹³⁾

Presently available diffusion coefficient data for strontium (and also for barium) were reviewed, and the results are given in Ref. 87. A spread of several orders of magnitude in diffusion coefficients as well as considerable differences in activation energies were found. The wide spread in the data might be the result of the dependence of diffusion coefficients on concentration and graphite type. The existence of two (or probably more) components of diffusion, a slow diffusion in the bulk graphite and a relatively fast diffusion in pores or channels, also contributes to the problem.

de Nordwall and co-workers^(89, 96) derived diffusion coefficient data from concentration profiles for strontium (Sr-89) in the graphite spine and the fuel tube of Pluto loop fuel elements. Diffusion coefficients obtained at 1000°C were around $5 \times 10^{-9} \text{ cm}^2/\text{sec}$, considerably lower than the reference value of $2 \times 10^{-8} \text{ cm}^2/\text{sec}$ (which one can derive from the data given in Table 4.2). This suggests that the reference data are conservative when used for release calculations in that their use leads to transport rates which are higher than those in the real case. However, only a single diffusion path and a single diffusion coefficient were considered in the analyses of the loop data.

In barium diffusion work conducted at Gulf General Atomic,⁽⁹⁵⁾ diffusion coefficients for barium in SX4 graphite were measured at 1500°C as a function of the barium-source concentration. Apparent diffusion coefficients were not found to be particularly sensitive to changing source concentration (5 to 700 μg barium/g graphite) or orientation of the graphite.

Diffusion Coefficient Data for Other Metals. The usual procedure in reactor calculations is to take samarium diffusion data to be the same as barium data, europium data the same as strontium data, rubidium data the same as cesium data, and cerium data as 1/10 the barium data. This procedure leads to conservatively high values for the diffusion coefficients of samarium, europium, rubidium, and cerium; there is no further immediate need for refinement of diffusion data for these elements.

Needed Work on the Diffusion of Metals in Graphite. Continuation of studies using the permeation method is important. Diffusion studies using tracers at constant elemental chemical potentials are needed. Studies at several chemical potentials should provide a good measure of the concentration dependence.

There is some reason to believe that diffusion coefficients may be dependent on the ambient gas pressure (helium). If a pore dimension is large compared with the mean free path, a pressure dependence for

volatile metals can be expected. Further experiments are needed to test this phenomenon.

The program at Gulf General Atomic is in accordance with the belief that the complexity of the diffusion phenomena of metals in graphite should be recognized and evaluated in a scientific manner. Through the types of studies outlined, the diffusion process can be better understood; and through this understanding, the precision of estimates of the release can be improved. The most conclusive confirmatory test of metallic fission product transport is measurement in an operating reactor. In-pile diffusion studies, as in the Peach Bottom test elements (see Section 4.8.1), are important and should be continued.

4.2.8. Vapor Pressure of Metallic Fission Products Sorbed on Graphite

Important input data for FREVAP and FIPER calculations are equilibrium vapor pressures of fission product metals sorbed on graphite. A considerable amount of vapor pressure data has been obtained. Essentially all of these data were measured at Gulf General Atomic.

The existing data were obtained by two different methods:

1. A mass spectrometric-Knudsen cell technique in which a Knudsen cell is mounted in a mass spectrometer. Knudsen cells used in this work are small metal containers (usually of tantalum, molybdenum, or tungsten) having a small orifice through which the vapor effuses. Powdered graphite impregnated with fission product metal is contained in a cell. The mass spectrometer determines the amount of vapor effusing from the cell as a function of time and temperature. (For details on this method, see Refs. 69 and 97). A variation of this technique has been the use of a radioactive tracer in following effusion from the Knudsen cell. (98)
2. A pseudoisopiestic method in which a graphite specimen contained in a closed evacuated tube is equilibrated with metal vapor. The graphite specimen is held at the temperature of interest. The vapor pressure is fixed by heating a metal source held in one end of the tube. (For more details, see Refs. 69 and 99.)

The data obtained in these experiments are in the form of vapor pressure versus graphite loading (concentration) at various temperatures. In general, the data exhibit Freundlich-type adsorption behavior. Freundlich behavior is characterized by the equilibrium partial pressure of the sorbate being proportional to a power $u > 1$ of the concentration of the sorbate in the sorbent and is described by the equation

$$P_M = e^v C_M^u, \quad (7)$$

where

$$v = a + b \left(\frac{10^3}{T} \right) \quad \text{and} \quad u = c + d \left(\frac{10^3}{T} \right),$$

or

$$\ln P_M = \left[a + b \left(\frac{10^3}{T} \right) \right] + \left[c + d \left(\frac{10^3}{T} \right) \right] \ln C_M, \quad (8)$$

where P_M is the equilibrium pressure of metal (atm), C_M is the concentration of metal ($\mu\text{moles}/\text{m}^2$ internal (BET) area of the graphite), T is absolute temperature ($^\circ\text{K}$), and $a, b, c,$ and d are constants. The concentration is usually expressed in units of internal (BET) surface area rather than mass or volume of the graphite because it is believed the phenomenon principally involves sorption on internal surface sites.

Bulk graphite has an internal (BET) surface area of about $1 \text{ m}^2/\text{g}$.

Data for use in FIPER and FREVAP calculations are expressed in the form of Eq. 8.

Freundlich sorption behavior is characterized by an increase in the heat of adsorption with decreasing concentration of the sorbed species. However, the magnitude of the heat of adsorption must reach a limit at some concentration. Accordingly, one would expect a transition at low concentrations from Freundlich sorption behavior to Langmuir behavior. The latter is characterized by a

constant heat of adsorption. Transition from Freundlich to Langmuir behavior has been observed in the case of the cesium-graphite system (see below). The transition occurs at a cesium concentration of about 0.03 mg Cs/g C.

The evaporation of fission products from a solid such as graphite has been considered in terms of the laws of ideal solution by a British investigator.⁽¹⁰⁰⁾ It was shown that it is convenient to express the results in terms of an "effective net drive" and an "effective exit resistance."

Adsorption isotherm data exist for strontium, barium, cesium, and rubidium. The data, as presently accepted for use in reactor calculations, are given in Table 4.3. The data, which are described in the form of the constants a, b, c , and d as defined in the above equation, are discussed in the following sections. The data apply to bulk graphite.

Vapor pressure studies, similar to those at Gulf General Atomic, are being performed by R. H. Flowers and co-workers at Harwell in England. The effect of irradiation on the sorption capacity of graphite for fission product metals has been studied (see Section 4.2.11).

Sorption of Cesium on Graphite. The vapor pressure of cesium sorbed on graphite has been studied extensively using the pseudoisopiestic method.^(31,95,99,101) The cesium sorption characteristics of several different types of graphite were studied, and the data are compared in Refs. 31 and 99.

In Fig. 4.8, the data for cesium on TS-688 (EGCR) graphite are plotted. At cesium concentrations less than ~ 0.03 mg Cs/g C, the isotherms show a Langmuir behavior, in which the concentration is proportional to the cesium pressure. At this low concentration, the Clausius-Clapeyron heat of adsorption derived from the data approaches a constant level of 120 kcal/mole, an unusually high value.

Table 4.3
VALUES OF CONSTANTS IN VAPOR PRESSURE-CONCENTRATION
EQUATION

Metal	Method of Measurement	a	b	c	d
Strontium	M. S. ^a	8.65	-36.0	-0.57	4.59
Barium	M. S.	6.48	-33.7	-1.304	7.55
Cesium	Isopiestic ^b	28.4	-56.0	-19.0	31.7
Rubidium	Isopiestic	14.1	-34.1	-9.0	16.8

^a—Mass spectrometric-Knudsen cell technique.

^b—Pseudoisopiestic technique.

Fig. 4.8. Sorption of cesium on TS-688 graphite

The cesium data given in Table 4.3 were measured using H-327 graphite.⁽³¹⁾ The pseudoisopiestic method was used (see above for description of the method). Earlier data, given in the Fort St. Vrain PSAR,⁽⁶⁾ were measured using TS-688 graphite. The vapor pressure data for the two types of graphite are similar. (Note: The crushed graphite used in the cesium vapor pressure measurements had an internal (BET) surface area about 25% higher than bulk graphite, and this was taken into account in deriving values for the constants a, b, c, and d from the data).

Studies of the adsorption of cesium on graphite have been performed at North Carolina State University by Zumwalt and Haire; the results have not yet been reported.

Sorption of Strontium on Graphite. Strontium adsorption isotherms have been measured using the mass spectrometric-Knudsen cell method, described in Ref. 97. Molybdenum Knudsen cells have been determined to be satisfactorily nonreactive with the strontium and have been used in these measurements. The strontium was initially added to the graphite powder sample (37 to 74 μm) as aqueous $\text{Sr}(\text{NO}_3)_2$. It was reduced by pretreating at high temperature in a sealed tantalum container before being placed in the mass spectrometer.

The rate of vaporization of strontium from the cell was kept as small as possible, consistent with sensitivity requirements, in order to minimize kinetic limitations. Experiments indicate the existence of equilibrium conditions; however, this has not been conclusively demonstrated.

Data obtained for H-327 graphite have conformed well to Freundlich behavior and have been adequately reproducible.^(56,102)

The derived isotherms are reproduced in Fig. 4.9. The actual measurements covered the range from 1000° to 1800° K and 5 to 0.003 mg Sr/g C, and extrapolation of the isotherms to reactor conditions is required. This procedure leads to some spread in the predicted vapor pressures. The Freundlich parameters given in Table 4.3 were derived from these data. The transition to Langmuir behavior has not yet been observed for strontium on graphite. (Note: The powdered graphite used in the strontium vapor pressure measurements had an internal (BET) surface area of about 4 m²/g, and this was taken into account in deriving values for the constants a, b, c, and d from the data.) Earlier data, given in the Fort St. Vrain PSAR,⁽⁶⁾ were measured for TS-688 graphite. The data for the two types of graphite are similar.

At one time the input strontium vapor pressure equation used in FREVAP calculations was altered conservatively to yield strontium vapor pressures higher by a factor of 30 until more precise strontium vapor pressures could be obtained (see Ref. 62). The factor of 30 was incorporated into the equation by changing the value of the constant a. The new data have justified dropping the factor of 30.

Attempts were also made to obtain vapor pressure data using the relatively straightforward, simple pseudoisopiestic method, which has proved very successful in studies of the cesium-graphite system. However, these attempts were unsuccessful because a suitable containment tube could not be found.⁽⁸⁶⁾

Sorption of Barium on Graphite. The Knudsen-cell mass spectrometric method has been used to obtain vapor pressure measurements for barium sorbed on graphite.⁽⁶⁷⁾ Two different graphites,

Fig. 4.9. Strontium partial pressure over 37- to 74- μm grain H-327 graphite

TS-688 and HLM-85, were studied, and the two sets of data are in reasonable agreement.

In view of the fact that 12.8-day Ba^{140} , the important nuclide of barium, decays to a considerable extent in the fuel element (see Section 4.2.7), the existing vaporization data for the barium-graphite system are considered adequate.

Sorption of Rubidium on Graphite. Rubidium sorption isotherms obtained using two different types of graphite have been measured using the pseudoisopiestic method.⁽⁹⁷⁾ The resulting data are believed to be adequate for use in reactor calculations.

Sorption of Other Metals on Graphite. As described in previous sections, vaporization data exist for cesium, strontium, barium, and rubidium sorbed on graphite.

With the possible exception of samarium and europium, vaporization data for elements at normal reactor conditions are generally not critical for one of the following reasons: (1) the element is essentially immobile (e. g., zirconium), (2) the nuclides of the element have low fission yields, (3) the nuclides have relatively short half-lives and decay in transit, or (4) the nuclides have relatively long half-lives or are stable.

Samarium and europium are similar to strontium (e. g., their respective boiling points are 1790° , 1600° , and 1375°C), and in reactor calculations the transport behavior of samarium and europium usually is taken to be the same as that of strontium. This assumption leads to conservatively high calculated releases for samarium and europium. Nuclides of the two elements (1) have relatively low fission yields, (2) are not important in safety considerations, and (3) are subordinate in importance to cesium in maintenance considerations.

Needed Work on the Vapor Pressure of Metals Sorbed on Graphite. Studies of the vapor pressure of metals sorbed on graphite should be continued. Further studies are needed to prove

that equilibrium conditions exist in Knudsen cell measurements (i. e., that kinetic effects, such as diffusion of strontium out of the graphite particles, are not controlling). The vaporization of strontium at relatively low concentrations should be measured in an effort to identify the transition from Freundlich to Langmuir behavior, which is expected to occur because of the limitation on sorption energies. Measurements of the sorption of cesium on graphite, utilizing the pseudoisopiestic method, should be continued. (Studies of the metal-graphite systems using the pseudoisopiestic method are being carried out by L. R. Zumwalt and co-workers at North Carolina State University.) The correlation between the sorptive capacity of finely divided and bulk graphite requires better definition. This would provide more insight into the nature of the sorption sites.

4.2.9. Interaction Effects of Metals Sorbed on Graphite

The effect of cerium on the cesium-graphite system has been measured using the pseudoisopiestic method.⁽¹⁰³⁾ Reductions in equilibrium cesium loadings of 42% at 1000°C and 32% at 900°C were observed for graphite which had been preloaded to 0.90 monolayer of cerium. Reductions of 32% at 1000°C and 24% at 900°C were noted for a specimen containing 0.78 monolayer of cerium. No change in the slopes of the isotherms was detected (within the uncertainty of the data) down to a cesium loading of about 0.1 monolayer.

The pseudoisopiestic method was also used to study the co-sorption of cesium and rubidium on graphite.⁽⁹⁷⁾ Preferential sorption of cesium was indicated by the fact that the cesium loading was essentially unchanged from that observed in the cesium-graphite

system alone. However, the presence of cesium reduced the rubidium loading by a factor of about 2.5.

The vaporization behavior of strontium and barium co-sorbed on graphite was studied using the Knudsen cell-mass spectrometric technique.⁽⁶⁸⁾ Barium, which was the more energetically sorbed element, exhibited a behavior similar to that observed in studies of the barium-graphite system. On the other hand, strontium strongly sensed the presence of barium. The barium heat of vaporization data are, within experimental error, quite close to corresponding barium data obtained without strontium present. By contrast, the heats of vaporization obtained for strontium in the presence of barium differ considerably from those obtained without barium.

The conclusion drawn from these experimental studies of the interaction effects of metals on graphite indicates that cerium will displace cesium, cesium will displace rubidium, and barium will displace strontium from sorption sites in graphite. These results also indicate that, in general, the effect of other metals on the transport in graphite of important fission product metals is not great.

Studies of cosorbed cesium and rubidium on graphite have been performed at North Carolina State University by Zumwalt and Haire; the results have not yet been reported.

In addition to the experimental studies of the interaction effects of metallic fission products sorbed in graphite, a theoretical relationship was derived for treating the sorption of mixed fission products in graphite.⁽¹⁰⁴⁾ This relationship was applied to the case of barium and strontium co-sorbed on graphite, and barium and strontium pressure-loading data were calculated.⁽¹⁰³⁾ The calculated data are in good agreement with experimental results for barium, the highly loaded element. In addition, there is some agreement in character

between the experimental and calculated results for strontium. An approximation of this theoretical relationship is used in FREVAP and FIPER.

Needed Work on Interaction Effects. More complete, systematic measurements are needed to quantitatively define the interaction effects of co-adsorbed fission products upon the vaporization isotherms. Additional experimental data on the diffusion effects of mutually sorbed metals on graphite are needed in order to refine the release values of fission product metals (in particular strontium) into the coolant of an HTGR system. Also, existing theoretical relationships for treating interaction effects should be improved.

4.2.10. Vaporization of Strontium from Graphite in the Presence of Carbon Monoxide

Some early measurements⁽⁸⁵⁾ suggested that carbon monoxide would significantly decrease the partial pressure of strontium adsorbed on graphite below the partial pressure observed in vacuum measurements. Recently, mass spectrometric measurements using a special gas-feed Knudsen cell were carried out to further investigate this question.^(58, 73)

Under some circumstances a very strong depression in strontium vapor pressure was achieved. However, under the experimental conditions which more closely approached reactor operating conditions, only a minor reduction (about 10%) was observed. Extrapolation of the data indicates that no significant dependence of strontium partial pressure on carbon monoxide pressure would be expected under reactor operating conditions (about 1 ppm CO partial pressure).

Needed Work. No further work in this area appears warranted.

4.2.11. Effect of Irradiation on Sorption Characteristics of Graphite

It is known that neutron irradiation increases the sorptive capacity of graphite for metallic fission products. Apparently, new sorption sites are created by the irradiation. (See Refs. 62, 85, and 101.) This effect is accounted for in some FREVAP and FIPER calculations by using a factor of six increase in sorption for the graphite (at the fuel matrix-graphite interface) proportionally stepped each year over a 6-yr period.

The pseudoisopiestic method was used to perform cesium sorption measurements on graphite specimens irradiated to fast neutron exposures from 2.0 to 6.0×10^{21} nvt at temperatures ranging from 610° to 1130°C. (101) The data show a continuing increase in the sorption capacity of the graphite with increasing exposure. The maximum increase in cesium loading (over the loading of unirradiated material) was a factor of 12.2, which was observed for a specimen irradiated at 1000°C to 6.0×10^{21} nvt. The amount of cesium that can be sorbed is a function of both the exposure and irradiation temperature, and above 1000°C the sorption sites may anneal out almost as fast as they are produced.

Similar measurements also using the pseudoisopiestic method were conducted to examine the effect of irradiation on the strontium sorption characteristics of graphite. (31) The largest increase observed in the strontium loading was a factor of 6.7, as compared with the factor of 12.2 for cesium in a duplicate specimen.

Needed Work. Experiments should be performed to obtain additional data on the effect of irradiation on the capacity of graphite to sorb strontium. The sorption tests should be conducted

at 800° to 1000°C using graphite specimens irradiated at various temperatures to fast-neutron exposures up to around 8×10^{21} nvt. Either the mass spectrometric -Knudsen cell method or a simplified isopiestic method could be used. (See Section 4.2.8 for a description of these methods.)

4.2.12. Plateout of Metallic Fission Products

Plateout (or deposition) profiles for metallic fission products such as cesium and strontium on steel surfaces in the primary circuit of HTGR type systems have been determined in loop and reactor experiments. (89,105-109) The profiles generally show (1) an initial steep increase (starting on steel surfaces at the downstream end of the fueled region), (2) a peak, and (3) a gradual decrease. The initial increase results from a decrease in temperature and consequent increase in sticking efficiency of the fission product metal on the steel surfaces. The gradual decrease results from depletion of the gas stream. When the plateout results from decay of a relatively long-lived noble gas precursor (e.g., decay of Xe^{140} to produce Ba^{140} , which plates out), the profiles are relatively flat, indicating relatively uniform plateout of the metallic fission product throughout the circuit.

Studies at ORNL and GGA have yielded information on the plateout of cesium and strontium on stainless and mild steel and on carbon dust for use in predicting the extent of plateout on the surfaces of reactor components. (Details on these studies are presented below. Also see Section 5.6.)

Methods utilized at GGA for investigating cesium plateout on steel surfaces at temperatures in the range 343° to 735°C are (1) an isopiestic technique in which a metal specimen contained in an evacuated L-shaped tube was equilibrated with tagged cesium vapor⁽¹¹⁰⁾ and (2) a dynamic technique in which tagged cesium was deposited from a helium stream onto steel tubing.⁽¹¹⁰⁾

Methods utilized at GGA for studying strontium plateout on steel surfaces at temperatures around 1000°C include (1) a transpiration method in which a metal surface was equilibrated with tagged strontium vapor in a helium stream and (2) a Knudsen cell method in which a metal specimen and strontium-impregnated graphite powder, both of which were contained in a small metal cell, were equilibrated under vacuum conditions. (97)

At ORNL, (1) the plateout of fission product metals was studied on samples of graphite, nickel, and carbon steel exposed in the sweep gas downstream from a fuel sample in GRR-ORR Loop No. 2, (45) and (2) the plateout and desorption of fission product metals on steel surfaces was studied by use of an out-of-pile loop. (111) Also, plateout studies were conducted in the out-of-pile loop at Battelle Memorial Institute. (45, 46)

In work performed under the Dragon project in England, plateout profiles were determined at various points in the coolant circuits of the Pluto loop and the Dragon reactor. (106, 108)

At GGA, the isopiestic method was used to obtain cesium plateout data over the temperature range from 343° to 735°C (650° to 1350°F) with cesium pressures of $\sim 10^{-9}$ to 10^{-6} atm. All measurements were made in vacuo. Cesium plateout levels ranged from 0.5 to 90 monolayers,* and a strong dependence on the degree of vacuum baking at elevated temperatures (800° to 1000°C) was noted. It is evident that the thickness of the surface oxide film on the steel must be the controlling factor in determining the degree of cesium plateout. (112)

Cesium plateout data were obtained using the dynamic method on 304 stainless steel and mild steel over the temperature range from 210° to 735°C with cesium pressures of 10^{-8} to 3×10^{-6} atm. In general, the level of cesium plateout was reasonably consistent:

* One monolayer of cesium is equivalent to about 0.93 mg Cs/m² of surface.

~250 monolayers were deposited at 735°C, with deposition increasing uniformly to ~1000 monolayers at 590°C. Very high plateout levels (up to 40,000 monolayers) were observed on mild steel at the lower temperatures, and it is believed that none of these represent steady state sorption levels, since the activity was still increasing at the conclusion of the runs (up to 2400 hr in duration.)^(6,110)

Using these plateout data and data for the cesium-graphite system, one can estimate the relative distribution of cesium on graphite (such as dust) and on stainless steel. At 1350°F (735°C) and a cesium pressure of 10^{-7} atm, the cesium loading on stainless steel is estimated to be $0.8 \mu\text{g Cs/cm}^2$ surface with a corresponding graphite loading of $0.053 \mu\text{g Cs/cm}^2$, a ratio of 15 to 1. This ratio would not be expected to change much at lower cesium pressures.

The data indicate that on a surface area basis, cesium is preferentially sorbed on steel, rather than graphite, surfaces. However, graphite has a relatively high surface area per unit weight (ranging from $\sim 1 \text{ m}^2/\text{g C}$ for bulk graphite to $\sim 8 \text{ m}^2/\text{g C}$ for graphite dust with a particle diameter of $\sim 25 \mu$). Therefore, the fractional plateout on graphite dust outside the reactor core may be relatively high.

A transpiration-type experiment for measuring the plateout of strontium from a helium stream on 304 stainless steel and TS-688 graphite dust indicated movement of strontium only 3 in. downstream from the source with an average strontium vapor pressure of 4×10^{-8} atm (determined from the loss of strontium from the source). The 1/2-in.-o.d stainless steel containment tube, held at 1050°C throughout the run, was cut into 1/2-in. lengths to determine the distribution of strontium along the tube. A sharp strontium gradient was detected, with an equivalent of 700 monolayers of strontium being deposited on the steel tubing

adjacent to the source, which decreased rapidly to 2.7 monolayers about 3 in. downstream. ^{(97)*}

A Knudsen cell test using Sr^{85} -tagged strontium and operated at 1000°C confirmed that steel surfaces have a very high affinity for strontium and act as a very effective sink for strontium nuclides. ⁽⁹⁷⁾

Studies utilizing the ORNL out-of-pile loop have yielded information on both the plateout and desorption of metallic fission products. ⁽¹¹¹⁾ In one experiment a study was made of the effect of introducing low concentrations of steam into a turbulent helium stream flowing in a system with previously deposited fission products. Steam injections produced negligible effect when the partial pressure was 0.07 atm. However, when the steam partial pressure was 0.35 atm, there was a decrease in activity by a factor of 2 or 3.

Calculational methods have been developed to handle the plateout of fission products. For details see Section 4.1.4.

A simple strontium deposition loop is being assembled at Gulf General Atomic in order (1) to study the nature, distribution, and adherence of strontium deposits formed under reactor conditions and (2) to check calculation methods such as the PAD code described in Section 4.1.4.

The above plateout studies have shown that stainless steel and mild steel surfaces have a high affinity for cesium and strontium and act as a very effective sink for the fission product nuclides of these two elements. The studies have also shown that steel surfaces have a higher affinity for strontium than for cesium. The surface oxide film appears to be an important factor in determining plateout levels.

Needed Work. See Section 5.6 for description of needed work on fission product plateout.

* One monolayer of strontium is equivalent to about 0.91 mg Sr/m^2 of surface.

4.3. INTEGRAL (LOOP AND REACTOR) SYSTEMS

As indicated in previous sections, the transport and plateout behavior of fission products under reactor conditions is highly complex. Factors affecting fission product behavior which must be considered and which cannot readily be evaluated in the laboratory include (1) the effect of irradiation, (2) the effect of helium pressure under high-velocity flow conditions, (3) the effect of temperature gradients, and (4) the effect of other fission product elements on the behavior of a given fission product element. Integral systems, such as loop and reactor systems (see Appendixes A and B), in which these influences are operating simultaneously can provide important information on fission product transport and plateout behavior. These integral systems provide, for example, observed fission product release data that can be compared with data calculated by the FREVAP or FIPER codes. Also, observed concentration profiles of fission products in the structural graphite of the loop and reactor systems can be compared with calculated profiles. Such a comparison provides information on the diffusional behavior of fission products in graphite. The major problem with such comparisons is uncertainty in the input information. For example, local temperatures in loop and reactor systems often vary considerably during the test, and this variation affects fission product transport rates.

HTGR-type loop and reactor systems in operation or planned include (1) the Dragon reactor, (2) the Peach Bottom reactor, (3) the Studsvik loop, (4) Pluto loop B (a planned loop), and (5) the (out-of-pile) fission product deposition loop at ORNL. Details on these important in-pile systems are given in Appendixes A and B and in Section 4.4. All of these systems can be considered as integral systems and sources of fission product transport data for comparison with available models.

ORNL Fission Product Deposition Loop. The fission product deposition loop at ORNL has been reactivated to permit a study of the

diffusion of cesium and strontium in graphite and evaporation of the metals into a turbulently flowing helium stream. (23)

The graphite evaporator used in these experiments is in the form of a hollow cylinder with a 1-in. inside diameter and a 6-in. length. One end of the graphite cylinder is sealed and the other is connected to a source of tagged cesium vapor. (Cesium will be used in the initial experiments.) The evaporator is induction-heated. The vapor pressure of cesium in the graphite is fixed by the temperature of the source region. Cesium vapor flows to the capsule by gaseous diffusion. Cesium evaporating from the outside of the graphite cylinder is swept away by the helium stream and the cesium condenses downstream. Measured data include cesium flux through the graphite and concentration gradients in the graphite.

Four runs have been completed and the experimental results are given in Refs. 79 and 113. It was found that in each run the observed rates of transport of cesium through the graphite tube into the outer helium stream were several orders of magnitude higher than one would predict on the basis of known diffusivity and vaporization data. The unexpectedly high rate of transport of cesium was attributed to transverse flow of helium into and out of the graphite evaporator tube. Since graphite is porous, some degree of transverse flow would be expected.

An analysis based on transverse flow considerations was used to predict the rate of cesium transport in each of the four experiments. (114) The predicted values are in reasonable agreement with the experimental data, substantiating that transverse flow of helium through the graphite was responsible for the high rate of cesium transport.

Calculations at GGA show that the high rate of transport observed in the fission product deposition loop experiment is peculiar to that system. In the reactor situation, the collision frequency between migrating cesium atoms

and fuel rod matrix and graphite surfaces in the fuel element will be sufficiently high that equilibrium will essentially exist between the gas and solid phases. Accordingly, cesium vapor pressures will be fixed by the concentration of cesium in the matrix and graphite materials. The cesium concentration will be low; and thus the cesium vapor pressures will be low ($< 10^{-10}$ atm). Therefore, one would expect transport by condensed state diffusion in the graphite to predominate. On the other hand, at the relatively high vapor pressure of cesium (around 10^{-3} atm) in the loop experiment, it is not surprising that transport of cesium by transverse flow (i. e., by vapor phase transport) is predominant over transport by condensed state diffusion.

From another viewpoint, if the vapor transport of cesium is proportional to its vapor pressure (as one would expect it to be) and thus to C^u , reflecting Freundlich behavior (see Section 4.2.8, p. 59), then the fractional vapor transport of cesium per unit time (i. e., the steady-state fractional release by vapor transport) is proportional to C^u/C and, therefore, decreases rapidly with concentration since u is a power well in excess of unity at reactor temperatures.

The fission product deposition loop is important for use in studying the transport of fission product metals under simulated reactor conditions, and this work should be continued. A solid source, such as a simulated fuel rod impregnated with cesium, may be advantageous in place of the present vapor source. In support of the fission product deposition loop work, laboratory experiments to study the effect of helium flow on cesium diffusion in graphite would appear to be worthwhile.

Needed Work. Continued utilization of the Peach Bottom and Dragon reactors and the loop systems as sources of information on fission product transport is needed and is of vital importance.

4.4. PEACH BOTTOM TESTS

Peach Bottom reactor tests continue to be an important source of observed data on fission product behavior. The data are derived from the following tests:

1. Gaseous activity determinations. These tests yield information on the release of krypton and xenon into the purge system and main loop. The data are of particular use in assessing performance of the fuel.
2. Iodine release measurements. These measurements furnish information on release of iodine into the purge system and main loop.
3. Analyses of particulate matter. This work yields information on the character of particulate matter in the helium coolant and on the role of the material in transporting fission products.
4. Diffusion probe experiments. These experiments provide information on the release of condensable fission products from the Peach Bottom core. (These experiments also yield information on particulate matter.)
5. Determinations of the distribution of fission products in Peach Bottom fuel elements. This work yields information on the transport behavior of fission products in HTGR-type fuel elements.

The combined information is useful for (1) assessing the performance of the Peach Bottom reactor, (2) testing methods for calculating the transport of fission products in HTGR systems, and (3) attaining

a better understanding of fission product behavior in HTGR systems. Details on the five types of Peach Bottom tests outlined above and results obtained during Core 1 operation are given in the following sections. Core 1 operation ended on October 3, 1969, following 452 EFPD (effective full power days).

4.4.1. Peach Bottom Gaseous Activity Determinations (P. B. Test JM-1)

Purpose. These tests involve measurements of the radioactive gaseous fission product activity in the main coolant and fuel element purge systems. The purpose is (1) to determine fractional release of krypton and xenon nuclides from the fuel compacts, (2) to measure the effectiveness of the fission product trapping system in removing xenon and krypton nuclides from the fuel element purge stream, (3) to determine gaseous activities in the main coolant helium, and (4) to check the integrity of the fuel elements.

Procedure. Gas samples are taken and measurements are made by gamma-spectra assay of the concentrations of various xenon and krypton isotopes in the main loop helium and in the fuel element purge stream. During the periods while these measurements are being made, the reactor power, temperature, and flow and the fuel element purge flow are held constant. From these measurements the effectiveness of the fission product trapping system and the fission product retentivity of the fuel are calculated. (See Ref. 115.)

Continued surveillance of the level of activity in the main loop helium is maintained using the installed Cary ionization chamber and connected vibrating-reed electrometer.

Status of Knowledge on Release into Purge System. Analyses were conducted during Core 1 operation to determine fuel body release fraction (R/B) values for Kr^{85m} , Kr^{88} , Kr^{87} , Kr^{89} , Xe^{133} , Xe^{135} , and Xe^{138} . The data for Kr^{85m} are typical, and a plot of the data for this

nuclide is given in Fig. 4.10. (Plots of data for the other nuclides are given in Ref. 116.) These data show that the R/B values increased by a factor of about 10 during the period from startup to about 3×10^5 MW(t)-hr of accumulated reactor thermal energy. From that point, the R/B values remained relatively constant for a period and then decreased somewhat. The decrease is particularly noticeable for the isotopes with the shortest half-lives, 17-min Xe^{138} and 3.2-min Kr^{89} . The decrease at the very end of each plot may be attributed to the measurements having been taken at reduced power levels (see Table 5.1 in Ref.116). However, general decreases in R/B values may be explained by the expansion of the fuel compacts, which gradually closed the gap between the compacts and the fuel element sleeves. This resulted in improved heat transfer and lower fuel temperatures, and hence lower release rates.

As described in Ref.117, the Core 1 release behavior, except for the decrease, correlates with the release behavior observed during the GAIL III-B experiment, where a full-diameter section of a Peach Bottom fuel element was irradiated for 2 yr in the Gulf General Atomic in-pile loop. (27)

The Peach Bottom R/B values show an approximate square root of half-life dependence, and the release of the xenon nuclides is lower than that of the krypton isotopes. This behavior is the same as was observed in the GAIL III-B experiment (see Ref. 16). In Fig. 4.11 end-of-life R/B data for Core 1 are plotted against the half-life of the fission products.

On the basis of the R/B data, it was estimated that 55% to 70% of the fuel particles in the Peach Bottom core 1 had cracked coatings. (118)

Status of Knowledge on Main Loop Gaseous Activity. The main loop gaseous activity, as measured by the Cary electrometer, remained

Fig. 4.10. R/B values for Kr-85m

Fig. 4.11. R/B values vs half-life for Peach Bottom

essentially constant during the initial period of operation (June 1967 to January 1968). The total activity for all gaseous species in the main loop helium was generally less than 0.4 curie. Considering that the expected activity at the beginning of life was 108 curies and the design activity was 4225 curies, this value is satisfactorily low.

It was observed that even though the purge stream activity increased by about a factor of 10, the main loop activity only doubled during the initial period of operation. ⁽¹¹⁷⁾ Based on the measured values given in the table below, the release fraction through the fuel element sleeves, as determined in October 1967, was about 6×10^{-5} , which corresponds to an effective sleeve permeability of $7 \times 10^{-7} \text{ cm}^2/\text{sec}$:

<u>Stream</u>	<u>Xe¹³⁵ Concentration ($\mu\text{Ci}/\text{cm}^3$)</u>
Fuel element purge.	9.0×10^{-2}
Main coolant loop	5.1×10^{-6}

This release fraction is about two orders of magnitude lower than the GAIL III-B results and about three times lower than the design xenon release fraction of 2×10^{-4} . ⁽¹¹⁹⁾ The reasons for this excellent performance are very complicated; possible explanations are: (1) unlike the Peach Bottom elements, the GAIL III-B elements did not have a purge stream inlet porous plug to develop a significant pressure drop along the entire sleeve; (2) the pore size distribution may be more favorable in the Peach Bottom sleeves than in the GAIL III-B sleeve; and (3) the Peach Bottom fuel element temperatures are generally lower than the GAIL III-B fuel element temperature and the effective permeability is temperature-dependent.

Starting in January 1968, the Cary electrometer showed periodic step increases in main loop activity with time, and this

behavior was related to fuel element failures. These increases brought the indicated main loop activity level to 36 curies at the time of the scheduled 300-day shutdown on October 23, 1968.

Following the 452-EFPD shutdown, 78 improperly purged elements were identified using the failed fuel element locator (FFEL). The maximum indicated loop activity at full power was 270 curies. The resulting average value of 3.5 curies per improperly purged element at full power is consistent with the values at 300 and 168 EFPD. The use of this value to interpret the number of fuel element failures seems to have yielded a reasonably accurate number. (116)

In June 1968, an experiment was conducted to determine whether very-short-lived nuclides were major contributors to the main loop activity. Flow through the Cary electrometer was interrupted by closing the downstream valve. The electrometer was then read every minute for 30 min. No large quantities of short-lived activities with half-lives measured in seconds were detected. The plot appeared to have two components after it had reached equilibrium. Rubidium-88 and Kr-89 could account for the first component and Kr-87 and Kr-88 for the longer-lived component of the decay curve. When the downstream valve was opened, the readings returned to preexperimental levels. (120)

Needed Work. This work is important and should be continued. The measurements are of first priority because the data obtained are of great importance in assessing the performance of the fuel elements and of the helium purification equipment.

4.4.2. Peach Bottom Iodine Release Measurements (P. B. Test JM-8)

Purpose. The purpose of the iodine release measurements is to determine the quantities of radioactive nuclides of iodine (1) in the purge stream system (internal traps and delay bed A-301) and (2) in the main loop, and then to use these data to obtain R/B values.

Procedure. Iodine release measurements are performed when the reactor is shut down after operating at a steady power for a minimum of 1 week with constant purge flow rate. The reactor temperature is dropped to a minimum as quickly as possible in order to minimize any further release from the fuel. Gas samples are taken at intervals of 2 hr over a period of 24 hr from the purge stream and main loop after the krypton activity decays to insignificant levels (less than 1/100 of its original value). The gas samples are analyzed using gamma spectrometry to determine activities of the 5.27-d Xe¹³³ and 9.2-h Xe¹³⁵; and from these data, the quantities of iodine nuclides are calculated. (121)

Status. Several iodine release measurements were performed during Core 1 operation. Quantities (in curies) of iodine nuclides in the purge system and main loop measured at the time of the October 23, 1968, shutdown are given in Table 4.4. The quantities of iodine nuclides in the main loop, even though relatively low, were, as a result of cracked fuel elements, higher than normal. Release fraction values calculated on the basis of release into the purge stream at the time of the October 23, 1968, shutdown were 6.0×10^{-4} for I-135 and 1.9×10^{-3} for I-133. (122)

Table 4.4
QUANTITIES OF IODINE NUCLIDES IN PURGE SYSTEM AND MAIN
LOOP AT TIME OF OCTOBER 23, 1968, SHUTDOWN

Nuclide	Quantities (curies)	
	In Purge System	In Main Loop
6.7-h I ¹³⁵	3,300	29
21-h I ¹³³	12,000	110
8.05-d I ¹³¹	21,000	180

Three iodine release analyses were conducted in July and August 1967. (117) The R/B values for release into the purge system

were 2.3×10^{-3} , 3.4×10^{-4} , and 3.8×10^{-4} for I^{135} and 1.3×10^{-3} , 1.0×10^{-3} , and 1.5×10^{-3} for I^{133} . Comparison of these values with the October 1968 values cited above indicates only a slight increase in R/B values over the time period involved.

Results of the measurements performed in July and August 1967 revealed that 6.7-h I^{135} accounts for about one-half of the total release of Xe^{135} . (The effective Xe^{135} half-life in the core is about 4.5 hr.) This indicates that the releases of both isotopes are very nearly the same. For 21-h I^{133} , the R/B value is about one-third that of 5.27-d Xe^{133} , the somewhat greater release for Xe^{133} being due to its comparatively long half-life. In essence, these data indicate that the release of iodine and xenon is similar for any given half-life.

Needed Work. The iodine release measurements are important and should be performed at intervals of 200 to 300 full-power days. Future measurements are of particular importance for determining the effect on iodine release of the buildup of fission product metal concentrations in the fuel element. (The metals, which are electro-positive, could interact with the iodine atoms, which are electronegative, and thus reduce the degree of iodine release.)

4.4.3. Analysis of Particulate Matter Collected in Peach Bottom Main Loop (P. B. Test JM-14)

Samples of particulate matter are periodically removed from the main-loop dust collectors at Peach Bottom and examined at Gulf General Atomic.

Purpose. The purpose of this work is to determine (1) the amount of particulate matter, (2) the physical and chemical character of the particulate matter, and (3) the role of the particulate matter in transporting fission products and fuel material.

Procedure. The Peach Bottom primary loop contains two bypass centrifugal dust collectors. About 1% of the primary coolant

gas is continually passed through these collectors. The particulate matter collected is removed periodically (during scheduled plant shutdowns), weighed, and analyzed for fission products, carbon, and occasionally iron and trace elements. The samples are collected by Philadelphia Electric and sent to Gulf General Atomic for analysis. Details on the method employed are available in Ref. 123.

When access to other primary system components is available, samples of particulate matter are taken from these components. This was done, for example, during steam generator modification operations in 1967 and early 1968.

Status. Following the attainment of full-power operation in June 1967, samples of particulate matter were taken from the Peach Bottom dust collectors in July 1967, August 1967, January 1968 (after 168 full-power days), October 1968 (after 300 full-power days), and October 1969 (after 452 full-power days). In addition, several samples were taken prior to the attainment of full power. The amount of material in each sample has not exceeded 120 g. Additional samples were removed from fuel element surfaces and from membrane filters that were part of a diffusion-probe experiment.

Samples of particulate matter removed from the dust separators following the 452-EFPD (October 1969) shutdown are to be analyzed at GGA. Samples of particulate matter from the 300- and 452-EFPD diffusion-probe experiments have been transmitted to Oak Ridge National Laboratory, where they are being analyzed. The ORNL analyses will complement those performed at Gulf General Atomic.

Throughout Core 1 operation, the particulate matter consisted primarily of construction debris (weld spatter, grinding chips, shreds of stainless steel insulation, fibrous wool-like material, and fine black dust). Some of the smaller dust particles appeared to be agglomerates

of still smaller particles. The carbon content generally ranged from 10% to 30%; the balance was largely iron (see Ref.118).

An estimated 60 g of dust were removed from the dust separators between July 1967 and January 1968. Following the October 1968 shutdown, 116 g were removed. The concentration of dust in the primary coolant has been very low (approximately 10 $\mu\text{g/lb}$ helium) compared with the concentration (0.1 to 1 $\mu\text{g/lb}$ helium) found in the Gulf General Atomic In-Pile Loop (GAIL). The GAIL system, however, was equipped with a full-flow glass-fiber bed filter and was much less complex.

Tables 4.5 and 4.6 compare the weights, carbon contents, and specific activities of particulate matter collected to October 1968. As expected, the specific activities of activation products such as Cr^{51} , Mn^{54} , Fe^{59} , Co^{60} , and $\text{Ag}^{110\text{m}}$ did not increase markedly with time, the largest increase being noted for Co^{60} . Also, as expected, fission product concentrations in the dust did increase with time.

The release of very small amounts of fuel compact material to the main loop, presumably from fuel elements with cracked sleeves, was indicated by (1) the presence of Zr^{95} , Ru^{106} , and Pa^{233} , (2) increased concentrations of other fission products, (3) $\text{Sr}^{89}/\text{Sr}^{90}$ ratios, and (4) the presence of a small number of recognizable fuel particle coating fragments.

Following 300 EFPD of operation, with about 1.4% (eleven) of the elements having cracked sleeves, only 4×10^{-7} fraction of the fuel compact material was estimated to have been released to the primary coolant. This very small amount of fuel compact material released from the failed fuel elements accounted for almost all the fission product activity associated with the particulate matter.

Thin, generally adherent, black deposits ("sooty" in appearance) were noted on core components. These deposits appeared to consist primarily of carbon and iron. The carbon may have been deposited

Table 4. 5

PARTICULATE MATTER REMOVED FROM
MAIN LOOP DUST COLLECTORS

	July 1967	August 1967	January 1968		October 1968	
Loop	1	2	1	2	1	2
Weight, g	21	11	6	22	60	56
Carbon content, wt-% ^a	10	13	28	25	<10	(b)

^a-Balance construction debris.

^b-Not measured.

Table 4. 6

ANALYSIS OF PARTICULATE MATTER
FROM LOOP NO. 1 DUST COLLECTOR

Nuclide	Concentration (mCi/g)		
	July 1967	January 1968	October 1968
Co ⁶⁰	9.5×10^{-4}	6.0×10^{-3}	2.5×10^{-2}
Sr ⁸⁹	1.4×10^{-4}	0.15	3.7
Sr ⁹⁰	6×10^{-6}	2.9×10^{-3}	0.16
Zr ⁹⁵	(a)	0.35	2.2
Cs ¹³⁷	1×10^{-6}	1.1×10^{-2}	9.5×10^{-2}
Ce ¹⁴⁴	$<3 \times 10^{-5}$	0.16	4.2
Pa ²³³	(a)	≤ 0.43	23

^a-Not detected.

from the gas phase, as a result of the cracking of methane for example, or as fine dust.

Reactor operations were not affected by the thin deposits and particulate matter present in the Peach Bottom primary coolant system.

During the January 1968 shutdown, wipes were taken of metal surfaces at various locations in the No. 1 steam generator. Analysis of these wipes, which was performed at Gulf General Atomic, (118,120) showed that plateout activities were very low. Typical values of the plateout activity (in units of $\mu\text{Ci}/\text{cm}^2$) were 3×10^{-4} for Zr^{95} , 6×10^{-4} for Pa^{233} , 5×10^{-6} for Sr^{90} , and 2×10^{-3} for Cs^{137} . The Zr^{95} and Pa^{233} activities are indicative of fuel contamination in the main loop circuit. The activity value for Sr^{90} corresponds to a total wipeable inventory of 0.1 mCi Sr^{90} . This is reasonably consistent with the total Sr^{90} activity (on main loop surfaces as of January 1968) estimated from diffusion probe measurements to be 1 mCi. (118) There was no evidence to suggest that the strontium isotopes were being released to the main loop other than as their krypton precursors.

Needed Work. Work on the analysis of particulate matter is important and should be continued. Samples of particulate matter should be collected and analyzed at intervals of 6 months to 1 year. Emphasis should be placed on interpretation of the analysis data.

4.4.4. Peach Bottom Diffusion Probe Experiments (P. B. Test JM-15)

Purpose. The Peach Bottom diffusion probe experiments provide information on the behavior of condensable fission products in the primary circuit. Fission products and induced activities in the loop can be identified, plateout concentrations and partial pressures

determined, and some information regarding chemical form obtained. Partial pressure values are a measure of release from the Peach Bottom core and consequently are of particular value for use in testing methods (e. g., FIPER) for calculating fission product release from HTGR-type fuel elements.

Procedure. The diffusion probe method is based on the work of Browning and Ackley⁽¹²⁴⁾ and Gormley and Kennedy.⁽¹²⁵⁾ Roos of Gulf General Atomic confirmed the original theoretical work.⁽¹²⁶⁾ Recent theoretical work concerning diffusion tubes has been performed by Ozisik.^(127, 128) Also, Ozisik and Venerus have considered the problem in some detail.⁽¹²⁹⁻¹³³⁾

The diffusion probe system and procedures are described in detail in Refs. 134 and 135. Briefly, the diffusion probe system consists of (1) a plateout sleeve, (2) three diffusion tubes contained in an outer tube (the probe body), (3) a membrane filter, (4) an activated charcoal trap, and (5) a flow control unit. Each diffusion tube is a small-diameter tube through which a portion of the coolant stream passes in laminar flow. The majority of the condensable fission products diffuse to and react with the inside surface of the tube. After being in the reactor for a sufficient time, the probe, membrane filter, and charcoal trap are removed from the reactor loop and shipped to Gulf General Atomic for examination and analysis. A new probe is installed when the exposed probe is removed. The Peach Bottom diffusion probe is located in a thermowell near the hot valve in Loop 1 and samples the hot coolant gas.

In the laboratory, the diffusion tubes are removed from the probe and sectioned into 1-cm lengths. The sections are gamma-counted, and selected sections are subjected to radiochemical analysis for the determination of nuclides, such as Sr^{89} and Sr^{90} , which cannot be determined by gamma-counting. The resulting data are in the form of activity profiles for each nuclide detected. The shape of an activity

profile is indicative of the form (atoms, molecules, or particulates) in which condensable fission product nuclides are transported in the helium coolant. In the ideal case, diffusion coefficient values can be determined from the profile data. (136,137)

Partial pressures are calculated from data on the total deposition of particular nuclides in the diffusion tube, total helium flow through the tube, and decay constants. Release rates can then be derived from the partial pressure data. Release fractions are calculated by taking into account production rates and release rates. Plateout data are determined by analysis of surfaces of the plateout sleeve and probe body. Concentrations in the helium of nuclides associated with particulate matter are determined by analyzing deposits on the membrane filter.

Diffusion probe experiments were conducted in the Gulf General Atomic in-pile loop during irradiation of the GAIL IV fuel element. As reported in Refs. 137, 138, 139, and 140 useful and important information was derived from the experiments. For example, it was shown that cesium and iodine nuclides were transported primarily in the form of atoms, rather than in the form of molecules or aerosols. (140)

Status. Three diffusion probe sets, each containing an Incoloy 800 plateout sleeve, stainless steel probe body, three diffusion tubes, a 0.45- μ m membrane filter, and an activated charcoal trap, were exposed during Core 1 operation. The first probe was removed following the January 1968 shutdown (168 EFPD), the second following the October 1968 shutdown (300 EFPD), and the third following the October 1969 shutdown (452 EFPD). The emphasis was on the study of condensable fission products present in the helium coolant in atomic or molecular form. All probes were exposed in the same position between the core outlet and steam generator inlet.

From diffusion tube data, partial pressures for the nuclides detected and release fractions for fission products were calculated. Partial pressures were in the range of 10^{-16} to 10^{-14} atm. Fractions of fission products released from the core to the main loop were in the range of 10^{-7} to 10^{-4} . Values for specific nuclides are given in Tables 4.7 and 4.8.

Calculations of fission-product concentrations on diffusion probe No. 3 body specimens were recently completed. The nuclides detected included Mn^{54} , Fe^{59} , Co^{60} , Zr^{95} - Nb^{95} , Mo^{99} - Tc^{99} , Ru^{103} , I^{131} , Te^{132} - I^{132} , Cs^{134} , Cs^{137} , Ba^{140} - La^{140} , Ce^{141} , Ce^{144} , and Pa^{233} . Surface activity concentrations were on the order of 10^{-3} to 10^{-1} $\mu Ci/cm^2$ on exposed surfaces. Concentrations on the stainless steel mandrel upon which the Incoloy plateout sleeve was fitted averaged 22% (range = 4% to 50%) of concentrations on outside exposed surfaces of the plateout sleeve.

Concentrations of selected nuclides on surfaces of the three Incoloy 800 plateout sleeves exposed during Core 1 operation are given in Table 4.9. Between 168 and 300 EFPD, concentrations on outside plateout sleeve surfaces remained the same for Cs^{137} ; doubled for I^{131} , Cs^{134} , and Ba^{140} ; tripled for Zr^{95} ; and increased by factors of 7 and 11, respectively, for Ce^{141} and Pa^{233} . The largest increases were exhibited by nuclides not normally expected in the primary circuit, but likely to be associated with fuel material released from failed elements. Between 300 and 452 EFPD, surface concentrations doubled for Ba^{140} , Ce^{141} , and Pa^{233} , tripled for Zr^{95} and Cs^{137} , quadrupled for Cs^{134} , and increased by factors of 5 and 7, respectively, for Ce^{144} and I^{131} . The relatively large increase in iodine concentration may have been the result of a slight reduction in plateout sleeve temperature during reduced power operation just before the 452-EFPD shutdown.

Table 4.7
CONCENTRATIONS OF NUCLIDES IN
PRIMARY COOLANT EXPRESSED AS
PARTIAL PRESSURES
(From Diffusion Tube Data)

Nuclide	Average p (atm)	
	0-168 EFPD	168-300 EFPD
Cr ⁵¹	2×10^{-16}	2×10^{-16}
Co ⁶⁰	5×10^{-16}	6×10^{-16}
Sr ⁸⁹	$< 3 \times 10^{-17}$	4×10^{-16}
Sr ⁹⁰	$< 5 \times 10^{-17}$	5×10^{-16}
Zr ⁹⁵	5×10^{-17}	2×10^{-16}
I ¹³¹	9×10^{-17}	5×10^{-16}
Cs ¹³⁷	1×10^{-14}	4×10^{-14}
Ba ¹⁴⁰	$\sim 4 \times 10^{-16}$	2×10^{-16}
Ce ¹⁴¹	$\sim 2 \times 10^{-17}$	1×10^{-16}
Ce ¹⁴⁴	$\sim 9 \times 10^{-17}$	5×10^{-16}

Table 4.8
FRACTION OF FISSION PRODUCTS RELEASED
FROM CORE TO MAIN LOOP
(From Diffusion Tube Data)

	0-168 EFPD	168-300 EFPD
Sr ⁸⁹	$<1 \times 10^{-7}$	1×10^{-6}
Sr ⁹⁰	$<1 \times 10^{-7}$	1×10^{-6}
Zr ⁹⁵	9×10^{-8}	4×10^{-7a}
Ru ¹⁰³	1×10^{-7}	4×10^{-7a}
I ¹³¹	4×10^{-7}	2×10^{-6}
Cs ¹³⁷	2×10^{-5}	7×10^{-5}
Ba ¹⁴⁰	$\sim 7 \times 10^{-7}$	4×10^{-7}
Ce ¹⁴¹	$\sim 3 \times 10^{-8}$	3×10^{-7}
Ce ¹⁴⁴	$\sim 2 \times 10^{-7}$	9×10^{-7}

^aCorresponds to ~3 g fuel compact dust containing
0.6 g Th²³² and <0.1 g U²³⁵.

Table 4.9
FISSION-PRODUCT CONCENTRATIONS ON SURFACES OF
INCOLOY PLATEOUT SPECIMENS
($\mu\text{Ci}/\text{cm}^2$)

	Probe 1, 168 EFPD Shutdown, 1 Failed Element	Probe 2, 300 EFPD Shutdown, 11 Failed Elements	Probe 3, 452 EFPD Shutdown, 78 Failed Elements
Zr ⁹⁵	2.8×10^{-3}	8.2×10^{-3}	2.6×10^{-2}
I ¹³¹	4.8×10^{-4}	7.0×10^{-4}	4.8×10^{-3}
Cs ¹³⁴	6.9×10^{-3}	1.3×10^{-2}	4.9×10^{-2}
Cs ¹³⁷	2.3×10^{-2}	2.4×10^{-2}	6.5×10^{-2}
Ba ¹⁴⁰	5.7×10^{-3}	1.0×10^{-1}	1.9×10^{-1}
Ce ¹⁴¹	2.4×10^{-3}	1.7×10^{-2}	4.1×10^{-2}
Ce ¹⁴⁴	ND ^a	1.1×10^{-2}	5.0×10^{-2}
Pa ²³³	3.1×10^{-3}	3.4×10^{-2}	8.0×10^{-2}

NOTE: Specimens positioned at top of diffusion probe inserted into Loop No. 1 hot valve located between reactor vessel outlet and steam generator inlet. Probes were removed and replaced at each of the scheduled shutdowns, and thus individually experienced 168, 132, and 152 EFPD of exposure, respectively.

^aNot detected.

Fission product concentrations on the side of the Incoloy plateout sleeve facing the oncoming hot coolant gas were about twice as high as those on the opposite side (see Ref. 141)

A study⁽¹⁴¹⁾ indicated that the release of Sr^{90} at 168 EFPD, either by way of its precursor Kr^{90} or as strontium vapor, was probably much lower than indicated by the diffusion tube deposits. The greatest contribution was probably from the fission products in fuel particle dust or other fissile coolant stream contaminants.

The fractional release of Zr^{95} and Ru^{103} determined from diffusion tube data indicates that during the operational period from 168-300 EFPD, approximately 4×10^{-7} fraction of the fuel was released to the primary coolant, presumably from failed fuel elements.⁽¹⁴²⁾ Earlier a fraction of 1×10^{-7} was calculated for the same period from plateout and dust data.⁽¹²²⁾ The fraction 4×10^{-7} corresponds to about 3 g of fuel-carbon dust containing 0.6 g of Th^{232} and less than 0.1 g U^{235} . A strontium release fraction of 1×10^{-6} was calculated from diffusion-tube data. The fact that the strontium release value is higher than the fuel release value suggests that strontium is also released as either precursor or vapor, or both.⁽¹⁴²⁾

The analysis of isotope activity ratios (e.g., $\text{Sr}^{89}/\text{Sr}^{90}$) after 300 days indicated further that the major contributor to the metallic fission product activity in the main loop of the reactor was fuel released from cracked fuel elements rather than diffusional release or decay of gaseous precursors.⁽¹⁴²⁾ However, as has been discussed above, the absolute value of the fuel release was small.

Needed Work. The diffusion probe experiments are very important and should be continued. As mentioned above, these experiments yield valuable information on (1) the form in which condensable fission products are transported in the helium coolant, (2) partial pressure and release fractional data, and (3) the presence of fuel in the circuit. This

work becomes increasingly important as fission product activity builds up in the reactor.

4.4.5. Distribution of Fission Products in Peach Bottom Fuel Elements (P. B. Test JM-19)

Work is under way to determine the distribution of fission products in the graphite of fuel elements removed from the Peach Bottom reactor.

Purpose. The purpose of this work is (1) to obtain information for use in testing calculational methods and (2) to gain a better understanding of fission product transport behavior in HTGR-type fuel elements.

Procedure. The procedure, ^(142,143) which is similar to that used in the chemical examination of the GAIL IV fuel element, ⁽¹⁴⁴⁾ is to core the graphite at appropriate locations. Each coring is then sampled at intervals along its length. These samples are analyzed using gamma-counting and radiochemical techniques. The resulting data are reported in the form of concentration profiles in the graphite. Corings are taken in the graphite sleeves, spines, and reflectors of the Peach Bottom fuel elements.

Status. Investigations were performed to determine the distribution of fission products in the graphite of fuel elements E05-05 (an intact fuel element removed after 168 EFPD) and D06-01 (a fractured fuel element removed after 300 EFPD).

Concentrations of fission products in the E05-05 graphite were too low to provide definitive data. However, low fission product concentrations were demonstrated, coring and sectioning techniques were tested, and small amounts (in the ppb range) of impurities in the high-purity graphite were detected. These impurities were scandium, possibly titanium, iron, manganese, cobalt, nickel, possibly copper, molybdenum, niobium, terbium, and possibly europium. Further, it was learned that Sr^{89} and Sr^{90} are apparently present in the outer half

of the graphite sleeve only because of their noble gas precursors. Consistent with this contention were the shape of the profiles, the absence of higher concentrations at the surface of the upper reflector as noticed for cesium and to be expected for strontium, and the fact that the highest strontium concentrations were in the coring opposite the lower reflector compact, where krypton concentrations in the purge stream were highest. Concentrations of Sr^{89} and Sr^{90} were the order of 10^{-8} to 10^{-7} mg/g carbon as compared to 10^{-7} to 10^{-3} mg/g found in GAIL IV graphite.⁽¹⁴⁴⁾ The data indicate that the sleeve and purge stream had been very effective in limiting the migration of fission products into the primary system (see Ref.122).

A fairly complete study of the distribution of fission products in fuel element D06-01 was performed. The element was one of 11 failed elements removed following the 300-EFPD shutdown on October 23, 1968. Radial concentration profiles for Sr^{89} , Sr^{90} , Cs^{134} , Cs^{137} , and other nuclides were determined in the sleeve and spine of D06-01 at several axial locations.

Representative concentration profiles for Sr^{89} , Sr^{90} , Cs^{134} , Cs^{137} , and other metallic fission products in the sleeve and spine graphite of fuel element D06-01 are given in Refs.116 and 142 .

To determine concentration profiles, the graphite sections cut from the sleeve and spine were rotated on a lathe, and small portions were consecutively removed. This method represents an improvement over traditional coring methods. With this method, as little as 0.002 in. per sample could be cut off. Each sample was collected in a vial and gamma-counted with a high-resolution lithium-drifted germanium detector and a Sigma-2 4096 channel computer-analyzer. Concentrations of fission products and activation products in the graphite were calculated from the gamma spectra with the Sigma-2 computer using the GULF code.

The profile data show that the concentration ratio ($\text{Sr}^{90}/\text{Sr}^{89}$) decreases with distance from the source (fuel). This behavior suggests that Sr^{90} and Sr^{89} were transported predominantly as the precursors Kr^{90} and Kr^{89} . The fact that the Kr^{89} has a significantly longer half-life than Kr^{90} (3.2 min compared with 33 sec) would explain the decrease in the ratio $\text{Sr}^{90}/\text{Sr}^{89}$ with distance from the source. The Kr^{90} could be expected to decay to a greater extent during transit in the graphite. If Sr^{90} and Sr^{89} were transported in the graphite in the form of metallic nuclides, the ratio $\text{Sr}^{90}/\text{Sr}^{89}$ would be expected to increase with distance from the source owing to greater decay of Sr^{89} (relative to Sr^{90}) during transit in the graphite. (The half-life of Sr^{89} is 50.6 days; that of Sr^{90} is 28.8 yr.) Assuming a diffusion coefficient of $0.1 \text{ cm}^2 \text{ sec}^{-1}$ and a distance of 0.85 in., the transport time required for gaseous nuclides to traverse the spine is estimated to be 47 sec. This is comparable to the 33-sec half-life of Kr^{90} . (See Refs. 116 and 145.)

Aside from activation products such as Sc^{46} and Co^{60} , which show uniform concentration profiles in the graphite, only Cs and Sr were found to penetrate to the center of the spine. The more refractory metals (protactinium, zirconium, and ruthenium) showed a high concentration only at the fuel-graphite boundary, with a very rapid decrease in concentration with penetration depth. The penetration depth for protactinium, zirconium, and ruthenium was generally of the order of 0.005 to 0.010 in. and showed a maximum of 0.030 in. in one case.

The cesium profiles exhibit a concentration decrease of about one to three orders of magnitude over the diffusion distance, with the concentration of Cs^{137} being higher by a factor of about 20 than the Cs^{134} concentration.*

The profile data show that the concentration ratio ($\text{Cs}^{137}/\text{Cs}^{134}$) in the graphite changes only slightly with distance from the source fuel.

* Cs^{134} is not a fission product; it is produced by activation of Cs^{133} .

It was first thought that this behavior indicated transport by the rare gas precursors 5.27-d Xe^{133} and 4.2-m Xe^{137} . However, it is now concluded that the cesium isotopes were transported into the fuel element graphite primarily as metallic cesium. This conclusion is based primarily on the inability of the fission gas transport model to represent the observed cesium isotope profiles as it does the strontium isotope profiles. Also, cesium diffusion calculations were made which showed similarity in concentration profiles for Cs^{134} and Cs^{137} .⁽⁵⁶⁾ Before this calculation was made, it was not known which of the following two effects would predominate: (1) steepening of the Cs^{134} profile with respect to the Cs^{137} profile caused by decay of Cs^{134} being much faster than decay of Cs^{137} , and (2) flattening of the Cs^{134} profile with respect to the Cs^{137} profile caused by the higher neutron dose experienced by the older Cs^{133} . It appears from this calculation that the latter effect exceeded the former for the given conditions.

The FIPER code (see Section 4.2.1.2) was applied in the analysis of cesium concentration profiles measured in the D06-01 fuel element.⁽⁵⁷⁾ It was shown that under certain circumstances the diffusion profile for an unstable species will have a shape identical to that of a stable species.

Needed Work. Work on the distribution of fission products in Peach Bottom fuel element graphite is important and should be continued. Concentration profiles for the sleeve and spine graphite will furnish information on the diffusion behavior of metallic fission products in graphite. Also, the profiles can be compared with calculated profiles as a test of calculational methods and input data.

Determination of concentration profiles in the graphite of the upper and lower reflectors of the elements is of some interest. In the case of Sr^{90} , analysis of representative outlet reflectors would

indicate the amount of Sr^{90} released from the fueled region of the elements. Analysis of the lower or inlet reflectors would determine the amount of Sr^{90} completing the circuit in one form or another. With data from both sets of reflectors, the fraction of Sr^{90} deposited in the steam generators and other parts of the primary circuit could be estimated.

4.5. TESTING OF CALCULATIONAL METHODS

An important area of work is the testing of the calculational methods, FIPER and FREVAP. The most practical way to test these methods is to compare observed and calculated releases from experimental fuel elements. The sources of observed data are (1) Gulf General Atomic in-pile loops GAIL III-B and -IV, performed in the GETR at Vallecitos, (2) Pluto loops 3, 8, 15, 16, performed at Harwell for the Dragon project, (3) the Dragon reactor, and (4) the Peach Bottom reactor. Information derived from these sources includes (1) release of metals to graphite, (2) concentration profiles in graphite, and (3) release of metals to coolant.

FREVAP-8 was tested by comparing observed and calculated releases from experimental fuel elements irradiated in Pluto loops 8 and 15^(146, 147) and in GAIL-IV.⁽¹⁴⁴⁾ Table 4.10 describes the in-pile loop experiments, and Table 4.11 gives the results of the comparison between experiment and calculation. Analyses of the coolant gas showed that the rates of release of Kr^{89} , Kr^{90} , and Xe^{140} were insufficient to account for the Sr^{89} , Sr^{90} , and Ba^{140} found in the Pluto coolant circuits.

Owing to neglect of transient diffusion, the FREVAP-8 calculation for the GAIL element yielded strontium releases which were 100 times greater than those observed. For the short-lived Ba^{140} , the situation is reversed because the calculation predicts that effectively all the Ba^{140} will decay during its passage through the thick fuel tube,

Table 4.10

DESCRIPTION OF IN-PILE LOOP EXPERIMENTS

Experiment	Fuel Coating	Irradiation Time (days)	Peak Fuel Temp (°C)	Peak Surface Temp (°C)	Fuel-Free Zone (Graphite Matrix) Thickness L (mm)	Coolant Flow Rate (g sec ⁻¹)
Pluto 8	PyC/SiC/PyC	173	1400	1350	1.25	18
Pluto 15	PyC/PyC	162	1370	1320	1.25	18
GAIL IV	PyC/PyC	400	1450	1100	≤ 6.35	35

Note: L^2/D for a Pluto element is $>1/23$ that for GAIL IV; hence, a Pluto element is that much closer to steady state. Other things being equal, a Pluto element represents the behavior of an element with a 6-mm fuel-free zone after more than 10 yr of irradiation.

Table 4.11

COMPARISON OF OBSERVED AND FREVAP-CALCULATED
STRONTIUM AND BARIUM ACTIVITY RELEASE FOR
PLUTO 8, PLUTO 15, AND GAIL IV IRRADIATIONS
(IN CURIES)

Nuclide	Experimental Data	FREVAP-Based Data
Pluto 8		
Sr ⁹⁰ - FFZ ^a	2.7×10^{-2}	8.4×10^{-3}
Sr ⁹⁰ - to coolant	7.3×10^{-3}	6.9×10^{-3}
Sr ⁸⁹ - to FFZ	1.2	3.4×10^{-1}
Sr ⁸⁹ - to coolant	3.2×10^{-1}	2.7×10^{-1}
Ba ¹⁴⁰ - to FFZ	5.2×10^{-2}	3.4×10^{-2}
Ba ¹⁴⁰ - to coolant	2.2×10^{-3}	1.8×10^{-5}
Pluto 15		
Sr ⁹⁰ - to FFZ	Not determined	7.0×10^{-1}
Sr ⁹⁰ - to coolant	Not determined	6.5×10^{-1}
Sr ⁸⁹ - to FFZ	4.4×10^1	3.6×10^1
Sr ⁸⁹ - to coolant	3.5×10^1	3.3×10^1
Ba ¹⁴⁰ - to FFZ	4.8	9.2
Ba ¹⁴⁰ - to coolant	5×10^{-2}	5.0
GAIL IV		
Sr ⁹⁰ - to FFZ	4.7	6.4×10^{1b}
Sr ⁹⁰ - to coolant	2.8×10^{-2}	5.2
Ba ¹⁴⁰ - to coolant	$(3 \text{ to } 12) \times 10^{-5}$	5.8×10^{-11}

^aFuel-free zone (to FFZ signifies the total quantity entering the graphite matrix).

^bBased on a conservative (high) estimate of "release constants." A calculated release of 2.4 curies is obtained using release data from in-pile experiments.

as is in fact found experimentally, whereas in reality there is a small release of Ba^{140} as Xe^{140} . Confirmation that Xe^{140} release can account for observed Ba^{140} has been obtained from GAIL and other experiments using similarly thick fuel tubes.

The measured coolant releases given in Table 4.11 are probably too high because of the presence of strontium contamination from previous loop experiments. It is known that prior to the start of the GAIL IV experiment, the loop was contaminated as a result of operation of the GAIL III-B experiments.

Needed Work. This important effort yields information on the reliability of the calculational methods, input data, and assumptions made in deriving the mathematical relationships and should be continued.

4.6. USE OF GETTERS FOR CONTROLLING STRONTIUM RELEASE

The behavior of the strontium nuclides Sr^{89} and Sr^{90} is of particular interest in HTGR systems because the calculated release of strontium during depressurization accidents tends to be the limiting factor for the design of reactor plant containment for HTGRs. Strontium becomes available in the primary coolant system via four mechanisms:

1. Release from failed fuel particles
2. Diffusion through intact fuel particle coatings. (This source is relatively unimportant in the case of TRISO-coated particles.)
3. Release as a daughter product from gaseous precursors
4. Contamination

It is important from a safety standpoint that strontium be contained within the fuel particles, within the matrix material, or within the graphite structure of the HTGR fuel elements.

One approach is to contain strontium within the fuel elements through the use of chemically active getters such as MgF_2 and ThF_4 .⁽¹⁴⁸⁾

Strontium released from the coated fuel particles would react with the getters, forming strontium compounds. As a compound, strontium would not be as mobile as metallic strontium and would be effectively retained within the fuel body. Another approach is to reduce the strontium activity in the coated particles by increasing the oxygen activity of oxide fuels. Some recent experiments on various carbon materials⁽⁷⁵⁾ have indicated that sulfur is an effective agent in decreasing the strontium vapor pressure over these materials.

Needed Work. A literature search should be performed for the purpose of identifying effective getter materials. Calculations should be made to determine the quantities of getter materials required to immobilize strontium, and the effects of these additives on the reactor physics and fuel cycle economics should be assessed. Experimental studies should be performed to determine the influence of getter materials such as MgF_2 , ThF_4 , metal oxides, and sulfur on the transport behavior of strontium in fuel element materials.

Studies should be undertaken to determine the compatibility of promising getter material candidates with core and coolant loop components to ensure that no detrimental side effects are introduced in the system.

Diffusion studies in the Peach Bottom fuel test elements are expected to provide information on the effect of getter materials (sulfur and magnesium fluoride) on the migration of metals. (See Section 4.8.1)

4.7. PLATEOUT AND DIFFUSION PROBES

Plateout and diffusion probe experiments provide important information on the behavior of metallic fission products in the primary circuit of HTGR systems. Fission products and induced activities in the helium coolant can be identified and partial pressures and plateout concentrations determined.

Plateout and diffusion probe experiments operating or planned include (1) the Dragon sampling tube (so-called Dracule), (2) the Peach Bottom diffusion probe, and (3) the Fort St. Vrain

plateout probes. The Peach Bottom diffusion probe is discussed in Section 4.4.4, and the other two probe experiments are discussed below.

4.7.1. Dragon Sampling Tube

Sampling tube (plateout probe) experiments are being conducted in the Dragon reactor. The sampling tube, known as the Dracule, is inserted into the main hot coolant of the reactor at a point about midway between the core and a heat exchanger. The sampling tube can be removed while the reactor is operating. (See Ref. 149.)

The Dracule yields (1) the concentrations of fission product nuclides in the coolant, (2) their molecular size (by application of diffusion theory to plateout profiles in the tube),* and (3) their plateout behavior. The tube could be used to prepare specimens for blow-off (re-entrainment studies). Other studies under consideration for Dracule include (1) the effect of flow rate on plateout, and (2) the effect of water ingress on the stability of plated-out fission products.

4.7.2. Fort St. Vrain Plateout Probes

It is planned to install two removable and replaceable sample probes within the primary helium circuit to monitor the concentration of vapor- and aerosol-borne metallic fission products within the helium coolant at two points; (1) downstream of the reactor core exit reflector and upstream of the steam generator tube surfaces, and (2) downstream of the steam generator tube surfaces. Sample probes will be installed to monitor the flow in each of the two loops within the primary circuit. (See Refs. 7 and 26.)

Each probe assembly, as presently designed, will have diffusion tubes, a plateout sleeve, and a charcoal trap. Fiberfrax insulation and a graphite thermal bond are arranged to keep the diffusion tube temperatures close to the return coolant gas temperature.

*See Section 4.4.4.

Surveillance of the plateout inventory within the primary circuit will be achieved by periodically removing the probes and performing gamma spectrometric and radiochemical analyses on the probe contents. On the basis of the probe calibrated flow and measured nuclide inventory, the average concentration of nuclides in the coolant gas can be determined.

Needed Work. It is important that the probe experiments be continued as sources of information on fission product transport in HTGR systems.

4.8. PEACH BOTTOM FUEL TEST ELEMENTS

Fuel test elements are being irradiated in the Peach Bottom reactor for the purpose of (1) evaluating the performance of advanced HTGR-type fuels over a wide range of operating conditions and lifetimes, and (2) performing controlled irradiation experiments. (57, 58, 150) Included are experiments on (1) diffusion rates of metallic fission products in an HTGR environment and (2) fission product release of various types of fuel particles. These are discussed in the following two sections. Some of the test elements contain only one type of fuel and are provided with individual purge streams permitting measurement of fission gas release during irradiation.

Needed Work. The fuel test elements are proving to be very important sources of information on the performance of advanced fuels under reactor conditions, and this work should be continued.

4.8.1. Diffusion Studies in Fuel Test Elements (57, 58)

Irradiation of diffusion samples in the Peach Bottom fuel test elements will provide important information on the diffusion of strontium, cesium, barium, and samarium in graphite under reactor conditions. The study will also provide information on the effect of getter materials (sulfur and magnesium fluoride) on the migration of the metals.

The diffusion samples consist of graphite crucibles containing tagged metal sources consisting of metal carbides distributed uniformly on coke material. The graphite crucibles are 1.25 in. long by 1.00 in. in outside diameter with a 0.2-in. wall thickness. Each crucible has a graphite cylinder in the center. The crucibles were coated with polystyrene to avoid hydrolysis of the metal carbides during shipping. The crucibles will be irradiated for periods of 1, 2, and 3 yr at temperatures between 800° and 1150°C.

After irradiation, each graphite crucible and inner graphite cylinder will be sectioned, and metal concentrations in the graphite will be established. The source material will be analyzed to determine the total metal loss from the source. From the total metal losses and the concentration gradients, migration rates and diffusion coefficients will be calculated. Having the graphite cylinder in the center assures concentration profiles for slow migration where no measurement can be made of the fractional release.

Needed Work. Test element diffusion studies will furnish valuable data measured under reactor conditions and should be continued.

4.8.2. Fission Product Release Studies in Fuel Test Elements ^(57, 58)

Samples have been prepared for irradiation in the Peach Bottom fuel test elements to study the release of fission product metals from coated fuel particles. The objective of this work is to relate fission product release behavior to particle characteristics, burnup, time, and temperature.

The samples consist of a mixture of coke material and coated fuel particles contained in a graphite crucible. The coke material and the graphite crucible are sorbent materials for the released metal fission products. After irradiation in the Peach Bottom reactor, the particles, coke material, and crucibles will be separated and analyzed to determine release levels for important metallic fission products such

as Cs^{137} , Sr^{90} , and Ba^{140} . The total number of samples is 86. Samples will be examined at 1-, 2-, and 3-yr intervals.

Each sample contains approximately 500 selected fuel particles coated with either TRISO or BISO coatings. Fifteen of the samples were encapsulated in containers made of Nb-1% Zr alloy. Encapsulation of the samples in a metal (barrier-type) container was necessary in order to determine release values below about 5%. This is because of cross-contamination from other samples and from the reactor fuel. The Nb-1% Zr alloy was chosen as the container material because of its nuclear properties and low carbide formation rate. Calculations indicate crucibles made of this alloy with wall thicknesses of ~ 0.25 in. will survive the time-temperature conditions in the reactor core.

Needed Work. Fission product release studies in the fuel test elements will furnish valuable data measured under reactor conditions and should be continued.

4.9. ORNL FISSION PRODUCT SURVEILLANCE PROGRAM

A program for joint surveillance of fission products in HTGR systems has been proposed by H. J. de Nordwall (ORNL).⁽¹⁵¹⁾ The objective of this program is to monitor fission product behavior in HTGR reactor systems.

Preparations are being made to implement this program by performing experiments in the Peach Bottom reactor with the following objectives:

1. Confirm or perhaps refine the predictions made in HTGR safety analysis reports concerning fission product release and test the theories upon which they are based in an unimpeachably realistic environment.
2. Make an estimate of the role of dust in fission product transport.
3. Deduce the sources of dust.

4. Determine where in the circuit fission products accumulate so that one may assess maintenance hazards for future plants and estimate the hazards that could be associated with perturbations such as temperature changes, coolant velocity changes, and the ingress of steam, which may lead to fission product redistribution.

It is expected that this work will yield needed information in the following areas:

1. Plateout profiles for condensable fission products in the primary circuit.
2. Concentrations of fission products in the helium coolant leaving the core.
3. Concentrations of fission products in the helium coolant at a point downstream of a steam generator.
4. Concentrations of short-lived krypton and xenon precursors in the primary circuit.

The work will add significantly to the amount of information already obtained from studies of fission product transport in the Peach Bottom reactor (see Section 4.4).

Instrumentation in addition to that existing at Peach Bottom (see Section 4.4) would be required for these measurements. For example, achieving item 3 would require the installation of a diffusion probe at a point downstream of a steam generator.

5. CONTROL OF FISSION PRODUCT ELEMENTS IN HTGR SYSTEMS UNDER ABNORMAL CONDITIONS

Information on fission product behavior in HTGR systems is important in the analysis of (1) the depressurization accident and (2) the steam in-leakage accident. Large HTGRs have been provided with additional engineering safeguards to preclude consideration of the loss-of-forced-circulation accident.

The depressurization accident involves an assumed failure of a prestressed concrete reactor vessel (PCRV) penetration closure. Its consequences are limited by the depressurization rate and the releasable fraction of the inventory of fission products within the primary system. Engineered safeguards are incorporated in the plant to effectively reduce the depressurization rate. The fission product inventory within the primary coolant circuit is dependent upon (1) the integrity of the coated fuel particles, which were conservatively designed relative to the type and thicknesses of the coating; (2) the sorption and transport characteristics of the core structural materials; (3) the form of the chemical species (i.e., oxide, carbide, gaseous, or metallic); (4) the plateout characteristics of the fission products; and (5) the cleanup rate in the helium purification system. The releasable fraction of the fission product inventory in the coolant circuit is, in turn, dependent on the quantity and location of the graphite dust on which a portion of the fission products are adsorbed and the amount of this dust discharged during the depressurization event. These latter characteristics are dependent upon the fuel and primary coolant system component temperatures and surface conditions.

In a high temperature gas-cooled reactor producing power by means of high pressure steam, the possibility of steam leakage into the primary coolant exists. Steam ingress may range from slow leaks up to a sudden steam tube failure.

In the case of a slow leak, a large fraction of the water vapor will react with the hot graphite core to form CO and H₂ which, along with

unreacted H_2O , are subsequently removed by the helium purification system. In this case the primary concern is not how much graphite is reacted, but rather how the oxidation attack is distributed over the complete graphite core and how well the fuel particles are protected.

In the case of a large leak, the reactor is scrammed; the fuel and graphite temperatures equalize; and the core cooldown is achieved rather quickly. Much less than 100% of the water will be reacted with the core. The amount of reaction that does occur determines the buildup of pressure in the coolant circuit (two molecules of gas are formed for every one consumed) in competition with the pressure-reducing effects of helium cooldown and any helium transferred to storage.

More information on fission product behavior under steam in-leakage conditions would permit more accurate prediction of the consequence of the accident.

The safety work suggested below is responsive to the high priority which is attached to analysis of these accident situations. It would be very desirable, in addition, to conduct a limited amount of research in three other areas involving fission product behavior: fission product retention in concrete, methyl iodide adsorption, and tritium transport.

5.1. BLOWDOWN EXPERIMENTS

A limiting safety consideration in the design of large HTGR power plants is the off-site bone dose resulting from a postulated rapid depressurization of the reactor pressure vessel. The consequences of this accident are dependent upon the quantity and form of the fission products discharged from the PCRV. The fission product discharge is in turn dependent on the quantity of radioactive materials external to the fuel elements, the location of the fission products within the vessel, the degree of adherence of the fission products on various reactor internal surfaces, the quantity of fission products that may be sorbed on dust or particulates, and the form of the fission products during the depressurization accident (i.e., vapor phase, chemical compounds, aerosols, or large particulates).

In the safety analyses for current HTGRs, conservative assumptions must be made regarding the quantity of fission products potentially discharged during a hypothetical rapid depressurization accident. Blowdown experiments would resolve many of the problems encountered in the present safety analyses by providing experimental data to demonstrate the degree of conservatism now being applied.

Laboratory experiments have been carried out at Gulf General Atomic to determine the fraction of adhering particulate activity which could be removed from metal surfaces during such an accident. (41, 85) Measurements were made of the fractions of dynamically deposited activity which could be removed by an increased helium flow velocity. This test was performed on a section of primary circuit coolant piping from the Gulf General Atomic in-pile loop (GAIL IV, see Appendix B). The pipe sections had been in the GAIL during nearly 10,000 hr of full-power operation. Five test runs were made, and the removal fractions of Cs^{137} , Ce^{144} , and Sr^{90} were measured. The fluid shear stress at the coolant boundary for some tests was increased to a factor of 7.5 times that calculated for the conditions in the GAIL loop during the time the activity was laid down. The maximum measured fraction of removed activity was 46% (this was for Ce^{144} in one run). The maximum fractions of Cs^{137} and Sr^{90} removed were 3% and 28%, respectively.

Potential sources of additional information on blowdown include (1) the Dragon sampling tube, (2) Pluto Loop B, and (3) the Gulf General Atomic strontium deposition loop. The Dragon sampling tube (so-called Dracule) is inserted into the main hot coolant of the Dragon reactor and can be removed while the reactor is operating. (See Section 4.7.) The Dracule could be used to prepare specimens for blowdown studies. Pluto Loop B (see Appendix B) is a planned British loop, and possible use of the loop for simulation of depressurization accidents is being considered. The Gulf General Atomic strontium deposition loop is a small loop which is now under construction (see Section 4.2.12). This loop could be used to prepare specimens for blowdown studies.

Needed Work. Blowdown experiments are needed to obtain test information relating to the discharge of radioactive materials during reactor vessel depressurization accidents to confirm the assumptions made in the safety analyses of such accidents.

The factors which need to be studied include:

1. The plateout characteristics of the fission products in the various portions of the coolant loop as functions of time, temperature, chemical form, reactor component surface conditions, and particulate size.
2. The quantity of fission products discharged from the coolant loop as functions of initial location and concentration, temperature, initial flow rate, and depressurization rate.
3. Correlation of the experimental test results with both the analytical methods used in the safety analyses for large-scale reactor plants and the experience with Peach Bottom, Dragon, and other operating gas-cooled reactors.

5.2. MATHEMATICAL MODELS FOR CALCULATING FISSION PRODUCT RELEASE, TRANSPORT, AND PLATEOUT FROM HTGR CORES

The behavior of metallic fission products in the HTGR core and primary coolant circuit has an overriding effect on the large HTGR plant design. In particular, the safety analysis of the rapid depressurization accident is dependent on the release, sorption, and plateout characteristics of the metallic fission products. These problems are currently managed by applying conservative factors in all analyses and design calculations performed using the mathematical models FREVAP-8 and FIPER developed at Gulf General Atomic. If the behavior of the fission products could be calculated with greater precision, many of the design conditions pertaining to HTGR safety features could be altered. Further work is needed to refine the calculational methods and to extend their capabilities to include higher temperature.

Needed Work. Improved methods are needed for calculating under accident conditions (1) the release behavior of fission products from an HTGR core and (2) the form and location of the fission products in the

primary coolant system. This information will be particularly useful in determining the limits for fission product discharge from the reactor during depressurization accidents.

5.3. STUDIES OF THE STEAM-GRAPHITE REACTION

In the case of a steam in-leakage accident, water will enter the primary circuit and react with the hot graphite core to form CO and H₂. The steam-graphite reaction has been studied to a considerable extent at laboratories in England and the United States. Existing data on the steam-graphite reaction were reviewed recently by Burnette, et al.,⁽¹⁵²⁾ and earlier by Walker, et al.,⁽¹⁵³⁾ and Clark, et al.⁽¹⁵⁴⁾

Studies of the steam-graphite reaction have yielded the following information:

1. It appears that at the high graphite temperatures involved in HTGR systems (800° to 1000°C), the thermal oxidation reactions predominate over the radiolytic process. At lower graphite temperatures (below 800°C), the radiolytic reactions could become significant.
2. The rate of the steam-graphite reaction is controlled by (1) temperature, (2) steam concentration, (3) concentration of hydrogen and other suppressive gases, (4) catalysts present, (5) diffusion parameters, and (6) burnoff. A considerable body of experimental data is available on the effects of these variables.
3. In-pore diffusion appears to be severely restricted at the high helium pressures in an HTGR; therefore, graphite corrosion should occur almost entirely at the coolant surfaces.
4. Up to about 1% burnoff, the reaction rate increases by a factor of about 10. The reaction rate is then relatively constant up to 50% to 60% burnoff. The rate decreases at higher burnoffs.
5. Barium is found to be more catalytic than strontium, and cesium is not catalytic at all.

Studies^(155, 156) have shown that loose, irradiated pyrolytic-carbon-coated fuel particles fail rapidly when exposed to steam at high temperatures (~1300° to 1400°C) for sufficient time, but are quite resistant to long-term steam attack at lower temperatures (900° to 1000°C). However, at

temperatures above 1000°C fuel particles contained in graphite are protected from oxidation due to the sacrificial nature of the container. At 1000°C and below, the protective effect of surrounding graphite is not apparent. This is because at these temperatures the steam-carbon reaction rate is so low as to allow diffusion of oxidant throughout the porous material virtually undepleted. These results, which need to be verified by in-pile studies, indicate that sacrificial graphite is quite protective at temperatures where protection is needed. At lower temperatures, protection is not needed because particle coating reaction rates are low. Thus, in the event of a steam in-leakage accident, it is expected that fuel particle coatings will not fail and fission products will be safely retained inside the coatings.

Experiments were performed to determine the behavior of strontium sorbed on graphite during steam attack.⁽¹⁰³⁾ Steam at high gas velocities (about 200 ft/sec) was passed over 1.5-g specimens with strontium loadings of 0.1 to 0.3 mg Sr/g C. Tests at 840° and 940°C for carbon burnoffs to 25% have shown that strontium is neither volatilized nor lost as an aerosol during steam attack.

Computer codes have been developed at Gulf General Atomic,⁽⁶⁾ Oak Ridge National Laboratory,⁽¹⁵⁷⁾ and Battelle Northwest Laboratory⁽¹⁵⁸⁾ for analyzing core corrosion in HTGR systems.

The design change of incorporating a secondary containment into the large HTGR has led to renewed interest in steam-graphite reaction rate data. The rate data are needed for analyzing the consequences of the postulated compound accident involving simultaneous steam leakage and failure of the PCRV. The events in this accident are (1) rupture of a steam tube, (2) reaction of the steam with hot graphite in the core of the reactor (before the core cools) to form hydrogen and carbon monoxide, and (3) escape of the resulting H_2 -CO- H_2O -He mixture into the secondary containment. If air exists in the secondary containment, the potential exists for accumulation of a flammable mixture. Accordingly, the

steam-graphite reaction rate data have a bearing on (1) the need for inerting the secondary containment, (2) the amount of steam in-leakage that can be tolerated, and (3) operational procedures to be followed in the event of a steam in-leakage accident.

Steam-graphite reaction rate data are also important for use in fixing the tolerable rate of steam in-leakage during normal reactor operation. The problem in this area is that carbon deposition will tend to occur on steam generator metal surfaces, provided the metal surfaces are in a catalytic (reduced, iron oxide-free) state; and the ratio of H_2 to H_2O in the coolant controls the formation of iron oxide on the metal surfaces. (For details on carbon deposition phenomena, see Ref. 152.)

Important work on the steam-graphite reaction is being performed at ORNL, BNWL, and GGA, as outlined below.

BNWL (Under AEC Nuclear Graphite Program)

1. Resumption of studies of the radiation-induced steam-graphite reaction.
2. Continuation of computer code (GOP) work with revision of code to account for reactions (such as the carbon deposition reaction) occurring in the HTGR circuit.

ORNL (under AEC Safety Program)

1. Continuation of in-pile studies of the reaction of steam with fuel rod samples contained in graphite crucibles.
2. Continuation of out-of-pile basic kinetics study of steam-graphite reaction.

GGA (under AEC Base Program)

1. Effect of steam oxidation on the strength of H-327 graphite.
2. Effect of neutron irradiation on the reactivity of H-327 graphite toward steam oxidation.

Needed Work. In-pile and out-of-pile studies of the steam oxidation reaction should be continued to gain further information on the effect of irradiation, temperature, catalysts, and other factors on the relative steam oxidation rate of particle coating, binders, and graphite. In-pile studies are of particular importance for verifying the results of out-of-pile studies. Development of analytical methods to describe the oxidation behavior should be continued. The effect of helium pressure on the rate of the steam-graphite reaction should be determined.

5.4. FISSION PRODUCT RETENTION IN CONCRETE

Fission products leaking through a cracked PCR V liner would be filtered and plated out in the PCR V concrete. No data are currently available on the filtering or sorbing characteristics of concrete for fission products.

Needed Work. Tests are needed to determine the effectiveness of concrete in filtering fission products, such as iodine, resulting from cracks in the PCR V liner. Experimental data regarding the filtering action in concrete would be useful in determining the effects of primary coolant leakage through defects in the PCR V liner, both during normal plant operations and under potential accident conditions.

5.5. FORMATION OF METHYL IODIDE

Methyl iodide (CH_3I) is a penetrating form of iodine which is not readily removed at ambient temperature by activated-carbon sorber systems.

Methyl iodide is a colorless, pungent liquid which melts at -66°C and boils at 45°C . It is the most reactive of the methyl halides and is used extensively as a methylating agent in organic syntheses. Methyl iodide can be made in low yield by reacting iodine with methane.

Several investigations at various facilities both in England and the United States have yielded information on the formation and behavior of methyl iodide. A good source of information is the proceedings of a conference on fission product release and transport under reaction conditions held at ORNL in 1965.⁽¹⁵⁹⁾ Adams, et al.,⁽¹⁶⁰⁾ have summarized much of the available information on methyl iodide formation in reactor accidents.

Under normal conditions, the CH_3I level in the main coolant of an HTGR system is expected to be negligible because (1) impurity concentrations of H_2 and CH_4 , either of which could lead to CH_3I formation, are very low, i.e., <10 ppm, (2) CH_3I decomposes thermally and radiolytically, and (3) CH_3I would be removed by the purification system.

Needed Work. The fraction of methyl iodide in the radioiodine released during depressurization for HTGRs should be determined. HTGR environments differ considerably from those of water reactors. AEC research on sorption media for methyl iodide currently under way at other sites should be expanded to include HTGR accident conditions.

5.6. FISSION PRODUCT PLATEOUT

During normal reactor operations, metallic fission products and iodine released from the core of an HTGR are plated out on various surfaces in the primary coolant loop (see Sections 4.1.4 and 4.2.12). In the design of advanced HTGRs, concentrations, chemical forms, and locations of the metallic fission products in the primary loops are needed to determine reactor maintenance procedures, shielding requirements, and the potential fission product release during reactor blowdown accidents.

Needed Work. Tests are needed to supply data on the sorption-desorption characteristics of metallic fission products and iodine as a function of temperature, deposition surface material (i.e., stainless

steel or mild steel), surface condition, and the form of metallic fission products (i. e., as metals, oxides, and other chemical species). Calculational models, such as the PAD code, are needed for predicting plate-out distributions in HTGR systems.

5.7. TRITIUM

The principal sources of tritium in HTGR systems are ternary fission and neutron activation of (1) He^3 present in the helium coolant as an impurity, (2) Li^6 and Li^7 present as impurities in the core and reflector materials, and (3) B^{10} in the control rods and reflector materials. Other tritium-producing reactions are relatively unimportant.

A preliminary GGA study of tritium in the Peach Bottom reactor has shown that comparatively little tritium arises from either Li^6 or He^3 activation. The major source appears to be ternary fission. Little tritium has been found in the Peach Bottom waste water, indicating that release to the main coolant is relatively low, only about one curie per month during normal operation. In addition, only small quantities of tritium have been found in the graphite of an irradiated fuel element. These results suggest that appreciable quantities of tritium are retained within the fuel; however, little information is available on the retention of tritium in HTGR-type fuel. In a study at ORNL of Dragon fuel, it was found that tritium is retained by TRISO-coated particles. ⁽¹⁶¹⁾

Needed Work. Additional studies are needed to establish the distribution of tritium in the Peach Bottom reactor. In particular, studies are needed of the retention of tritium by fuel particles, both intact and broken and both unirradiated and irradiated. (P-capsules are a good source of irradiated particles.)

6. PLAN OF ACTION

This section contains a list of the subtasks and plans of action required for development work on fission product behavior in HTGR systems.

Subtask 4.1.1. The RAD Code (see Section 4.1.1 for justification)

There is no apparent need at this time for development work on the RAD code.

Subtask 4.1.2. Fission Gas Release Studies (see Section 4.1.2 for justification)

Studies of the release of fission gases (including iodine) from coated fuel particles need to be continued. The studies at GGA include (1) short-term (hours) fission gas release measurements in the TRIGA Mark I reactor, utilizing a King (graphite-tube) furnace and (2) long-term (weeks or months) fission gas release measurements in the TRIGA Mark III reactor, utilizing a King furnace and loop system. (Use of the King furnace permits release measurements at temperatures up to 1800°C.) Measurement of fission gas release from fuel samples during irradiation in P-capsule tests is planned. The release behavior of TRISO- and BISO-coated particles, bonded and unbonded, is to be studied. Measurement of the release of fission gases from Peach Bottom Core 2 and test elements is expected to yield valuable data. All of this effort will be augmented by fission gas release studies to be performed at ORNL and under the European HTGR programs.

This subtask includes studies of:

(a) The effect of time, temperature, irradiation, and thermal cycling on the fission gas release behavior and integrity of fuel rods and fuel particles.

(b) The relationship of gaseous release and fuel contamination (1) on the surface of coated particles, (2) in the outer coating of coated particles, and (3) in the fuel rod matrix.

(c) Simple quality control methods (such as use of alpha counting) as an indirect indication of the level of fission gas release.

(d) The measurement of fission gas release from Peach Bottom test elements (see Section 4.8).

(e) The effect of rod fabrication on fission gas release.

(f) The relative fission gas release behavior of coated carbides and coated oxides.

(g) The effect of coolant impurities and burnup on the release behavior of fuel particles with broken coatings.

(h) The variation of release processes with half-life, temperature, neutron flux, and helium pressure. (Better understanding of the recoil release process is a particular need.)

Information pertaining to all these areas exists but more is needed to refine the data and understand the phenomena involved.

Subtask 4.1.3. Calculation of Release of Fission Gases from Coated Fuel Particles (see Section 4.1.3 for justification)

Calculational methods will be applied to the release of fission gases from coated fuel particles as required.

Subtask 4.1.4. Plateout of Iodine (see Section 4.1.4 for justification and Subtask 5.6 for plan of action)

Subtask 4.1.5. Sorption of Iodine in Graphite (see Section 4.1.5 for justification)

This work consists of studies of the sorption of iodine in graphite with appropriate loadings of metallic elements.

Subtask 4.2.1. Methods for Calculating Release of Metallic Fission Products from HTGR Fuel Elements (see Section 4.2.1 for justification)

(a) Work on FIPER X* development consists of the optimization of computer time with required accuracy.

(b) Work on FIPER II includes (1) development of routines for handling input data, (2) comparison of analytical and numerical results, and (3) programing for reactor application.

* FIPER X is the engineering version of the FIPER I code.

Subtask 4.2.2. Calculation of Release of Metallic Fission Products from Coated Fuel Particles (see Section 4.2.2 for justification)

This work consists of application of the SLIDER code or FIPER code, which treat the release of fission products from coated fuel particles, to laboratory release data obtained by annealing irradiated BISO-coated fuel particles and measuring the fractional release of metallic fission products as a function of time. In these experiments, the anneal is continued until the fractional release is high enough so that release by diffusion from the kernel, rather than release due to recoils in the coatings, is dominant. This work is expected to yield information leading to increased accuracy in predicting the release of fission product metals from BISO-coated particles.

Subtask 4.2.3. Release of Metallic Fission Products from Coated Fuel Particles (see Section 4.2.4 for justification)

This work consists of studies of the release of metallic fission products from coated fuel particles and fuel rods. Methods include (1) post-activation experiments in which lightly irradiated (in-TRIGA) particles are annealed and the release is measured as a function of time,* (2) postirradiation experiments in which highly irradiated particles are annealed and release is measured as a function of time, and (3) in-pile release experiments in which the fractional releases of metallic fission products during long-term irradiation are measured by postirradiation examination of the irradiated fuel particles and graphite crucibles used to contain the coated particles in capsule irradiation tests.

Expected areas of study include:

(a) Postactivation and postirradiation experiments to evaluate the effect of time, temperature, and irradiation on the metallic release behavior of coated particles and fuel rods. (The level of strontium and barium release in these experiments is a good measure of the integrity of the coatings of TRISO-coated particles.)

* Postactivation experiments can also be conducted on previously irradiated particles (e.g., P-capsule particles after sufficient aging).

(b) Experiments to determine the in-pile metallic release behavior of particle and rod samples (including Peach Bottom test element samples, see Section 4.8.2).

(c) Refinement of release constant models using data obtained in items (a) and (b).

(d) Experiments to determine the relationship between the metallic release behavior of TRISO-coated particles and the level of fuel contamination in outer pyrolytic carbon coatings and fuel rod matrix material.

(e) Comparison of the metallic release behavior of coated carbides and oxides.

Considerable information exists concerning the release of fission product metals from HTGR fuel, but more information is needed to refine the data and understand the phenomena involved.

Subtask 4.2.4. Diffusion of Metallic Fission Products in Coating Materials
(see Section 4.2.4 for justification)

The diffusion of metallic fission products (in particular strontium and cesium) in coating materials needs to be studied further to obtain data required for accurate calculation of the release of these metals from coated fuel particles. Work on this subtask includes:

(a) Diffusion experiments in which coated wafers are annealed in contact with a metal source in the temperature range 800° to 1500°C .

(b) Measurement of the solubility of fission product metals in coating materials.

(c) Analyses of fission product metal release data.

Subtask 4.2.5. Sorption of Metallic Fission Products in Fuel Rod Matrix Material (see Section 4.2.5 for justification)

This work consists of vapor equilibration experiments to measure:

(a) The sorptive capacity for strontium and cesium of reference fuel rod matrix material.

(b) The effect of irradiation on the sorptive capacity of this material.

Subtask 4.2.6. Distribution of Metallic Fission Products Between Matrix and Graphite Materials (see Section 4.2.6 for justification)

(a) Experiments are to be performed to determine the distribution coefficient for strontium between graphite and matrix material with the purpose of reducing the uncertainty of existing data. The technique will involve annealing graphite in contact with matrix material impregnated with tagged strontium. The experimental temperature range will be 700° to 1000°C.

(b) Diffusion experiments being conducted in the Peach Bottom reactor (see Subtask 4.8.1) are expected to yield distribution coefficient data under reactor conditions. The procedure involves measurement of the distribution of strontium at the interface between the source material (which simulates the fuel rod matrix material) and graphite in the diffusion specimen.

Subtask 4.2.7. Diffusion of Metallic Fission Products in Graphite (see Section 4.2.7 for justification and description of experimental methods)

For this subtask, the experimental effort consists of laboratory studies of the diffusion of fission product metals in graphite. Experiments are under way and planned using (1) the permeation method, (2) the plane-source method (including self-diffusion studies), (3) the autoradiographic technique, and (4) the microprobe technique. Fission product metals are to be studied in the following order of priority: strontium, cesium, barium.

The work in this subtask includes:

(a) A study of the phenomenology of the diffusion of metals (in particular strontium) in graphite using the plane-source, permeation, autoradiograph, and microprobe methods in the temperature range 800° to 1200°C. (This work will provide a better understanding of the diffusion process, particularly the modes of transport.)

(b) A study of the concentration dependence of the diffusion of metals (in particular strontium) in graphite using permeation and plane-source techniques at 900° to 1100°C. This work includes self-diffusion experiments

in which a graphite specimen is loaded uniformly with metal (e.g., strontium) and then tagged metal at tracer level is diffused into the specimen.

(c) A study of the temperature dependence of the diffusion of metals (in particular strontium) in graphite using plane-source and permeation methods in the temperature range 800° to 1200°C .

(d) Experiments to correlate graphite type (or characteristics) and diffusion behavior. Information derived from this work will aid in selecting types of graphite for reactor use.

(e) A study of the effect of other metallic elements (such as barium) on the diffusion behavior of a given fission product metal (such as strontium) to be determined by (1) studying the simultaneous diffusion behavior of one or more metals and (2) measuring the diffusion rate of a given metal in graphite preloaded with another metal.

(f) Experiments in the Peach Bottom fuel test elements to study the diffusion of metals in graphite under reactor conditions (see Section 4.8.1).

(g) Further studies of the effect of a thermal gradient on the diffusion behavior of metals in graphite.

Considerable information exists concerning the diffusion of metals in graphite, but more information is needed to refine the data and understand the phenomena involved.

Subtask 4.2.8. Vapor Pressure of Metallic Fission Products Sorbed on Graphite (see Section 4.2.8 for justification)

Studies of the vapor pressure of metals sorbed on graphite are to be continued, the main goal being determination of interaction effects (see Subtask 4.2.9). The Knudsen cell-mass spectrometric method will be used for most of this work, but a further attempt to adapt the pseudoisopiestic method to strontium vapor pressure studies may prove advisable. The temperature range of the studies will be 1000° to 1700°C . (Strontium vapor pressures at lower temperatures (below 1000°C) are generally too low to be measured using the mass spectrometric technique. However,

use of the pseudoisopiestic method would allow measurements at lower temperatures.) A further effort to prove that vapor pressure data are measured under equilibrium conditions in the mass spectrometer (i.e., that kinetic effects, such as diffusion of a metal out of the graphite particles, are not controlling) may be worthwhile. This effort may require the use of a double Knudsen cell, with graphite powder being contained in one Knudsen cell and strontium metal contained in a connecting cell.

The work in this subtask includes:

(a) Measurement of vapor pressures of fission product metals in support of interaction effect studies (see Subtask 4.2.9).

(b) Measurement of the vaporization of strontium at relatively low concentrations in an effort to identify the transition from Freundlich to Langmuir behavior, which is expected to occur because of the limitation on sorption energies.

(c) Further studies of the effect of graphite characteristics on the vapor pressure of strontium.

Subtask 4.2.9. Interaction Effect of Metals Sorbed on Graphite (see Section 4.2.9 for justification)

The effect of other metals on the vapor pressure of a given metal (strontium in particular) sorbed on graphite is to be further studied using both experimental and theoretical approaches. This work includes:

(a) Use of the Knudsen cell-mass spectrometric method to measure experimentally the vapor pressure of a given metal over graphite loaded with two or more metals.

(b) Refinement of existing theoretical relationships for treating the sorption of mixed fission product metals on graphite. This work will permit accurate estimate of interaction effects.

Subtask 4.2.10. Vaporization of Strontium from Graphite in the Presence of Carbon Monoxide Gas (see Section 4.2.10 for justification)

No further work in this area appears warranted.

Subtask 4.2.11. Effect of Irradiation on Sorption Characteristics of Graphite (see Section 4.2.11 for justification)

Experiments to determine the effect of irradiation on the capacity of graphite to sorb strontium are planned. Either the pseudoisopiestic or the mass spectrometric method will be utilized. The sorption tests will be conducted at 800° to 1000°C using graphite specimens irradiated at various temperatures to fast neutron exposures up to around 8×10^{21} nvt.

Subtask 4.2.12. Plateout of Metallic Fission Products (see Sections 4.2.12 and 5.6 for justification and Subtask 5.6 for plan of action)

Subtask 4.3. Integral (Loop and Reactor) Systems (see Section 4.3 for justification)

It is expected that a study of cesium transport utilizing the modified fission-product-deposition loop at ORNL will continue. The objective of this study is to obtain information on cesium transport for use in testing the FIPER calculational method. (The evaporator portion of the apparatus needs to be modified to eliminate transverse flow effects; see Section 4.3.)

Subtask 4.4.1. Peach Bottom Gaseous Activity Determinations (see Section 4.4.1 for justification and procedure)

Work is to be continued on (1) measurement of krypton and xenon fractional release into the purge system and (2) surveillance of the level of activity in the main loop helium. It is expected that measurement of fractional release into the purge system will be performed at least once a month, and more often if anomalous changes occur in the main loop activity. It is further expected that special tests will be performed, such as measurement of the effect of core temperature on release of fission gases into the purge stream. Existing apparatus will be utilized in these measurements.

The data will be used mainly in assessing the performance of the fuel elements and of the helium purification equipment. It is expected that this work will be augmented by work under the ORNL Fission Product Surveillance Program; see Section 4.9.

Subtask 4.4.2. Peach Bottom Iodine Release Measurements (see Section 4.4.2 for justification)

It is expected that iodine release will be measured periodically. The measurements should be performed at intervals of approximately 300 full-power days (at scheduled shutdowns), or more often if unusual situations develop that involve fission product behavior. Of particular interest will be determination of the effect on iodine release of the buildup of fission product metal concentrations in the fuel element. (The metals, which are electro-positive, should interact with the iodine, which is electronegative, and thus reduce the degree of iodine release.)

Subtask 4.4.3. Analysis of Particulate Matter Collected In Peach Bottom Main Loop (see Section 4.4.3 for justification)

Work on the laboratory analysis of particulate matter collected in the Peach Bottom main loop collectors is to be continued. It is expected that samples of particulate matter will be collected at intervals of approximately 200 full-power days (at scheduled shutdowns) and examined at GGA and/or ORNL to determine (1) type of dust (carbonaceous or metallic), (2) particle size, (3) concentrations of fission products, (4) concentrations of induced activities, and (5) concentrations of fuel material. It is expected that this work will be augmented by work under the ORNL Fission Product Surveillance Program; see Section 4.9.

Subtask 4.4.4. Peach Bottom Diffusion Probe Experiments (see Section 4.4.4 for justification)

Work is to be continued on diffusion probe experiments to gain information on (1) the form in which condensable fission products are transported in the helium coolant, (2) partial pressure and release fractions,

(3) the presence of fuel in the circuit, and (4) particulate matter transport. Diffusion probes will be exposed in the Peach Bottom reactor for periods of approximately 150 full-power days. The exposed probes will be examined at GGA and/or ORNL. The data obtained will be used in (1) assessing the performance of the Peach Bottom fuel, (2) determining the behavior of condensable fission products, and (3) testing models and calculational methods. It is expected that this work will be augmented by work under the ORNL Fission Product Surveillance Program; see Section 4.9.

Subtask 4.4.5. Distribution of Fission Products in Peach Bottom Fuel Elements (see Section 4.4.5 for justification)

Work is to be continued (1) to determine the distribution of fission products in the graphite of fuel elements removed from the Peach Bottom reactor, and (2) to derive from these data information on the transport of fission products in HTGR systems. Corings will be taken in the graphite at appropriate locations in the sleeve, spine, and reflector regions of selected fuel elements removed from the reactor. These corings will be analyzed, and data will be obtained in the form of concentration profiles for important nuclides in the graphite. The profiles will then be compared with calculated profiles as a test of calculational methods and laboratory data. Concentration data obtained from analysis of corings from the outlet and inlet reflector graphite may be used to estimate the fraction of a given condensable nuclide completing the circuit.

Subtask 4.5. Testing of Calculational Methods (see Section 4.5 for justification)

It is expected that calculation methods, such as FREVAP and FIPER, will be tested by comparing the calculated and observed behavior of fission product elements. This will be done as observed data become available. Sources of observed data include (1) the Peach Bottom reactor, (2) the Dragon reactor, and (3) loop experiments. Areas where calculated and observed data can be compared include: (1) release of metals to graphite, (2) concentration profiles in graphite, (3) release of metals to coolant, and

(4) release of fission gases to coolant. Results of these comparisons will be used to assess the reliability of the calculational methods and input data.

Subtask 4.6. Use of Getters for Controlling Strontium Release (see Section 4.6 for justification)

The work in this subtask includes:

(a) A literature search to identify effective getter materials.

(b) Calculations to determine the quantities of getter materials required to immobilize strontium. (The effects of these additives on reactor physics and fuel cycle economics will be assessed.)

(c) Experimental studies to determine the influence of getter materials, such as MgF_2 , ThF_4 , and metal oxides, on the transport behavior of strontium in fuel element materials.

(d) Studies of the compatibility of promising getter material candidates with core and primary loop components to assure that no detrimental side effects are introduced into the system

Subtask 4.7. Plateout and Diffusion Probes (see Section 4.7 for justification)

Work is to be continued on the design of equipment to measure the plateout inventory in the Fort St. Vrain reactor. (Planned work on the Peach Bottom diffusion probe is covered in Subtask 4.4.4.)

Subtask 4.8. Peach Bottom Fuel Test Elements (see Section 4.8 for justification)

Test element fission product release experiments and diffusion experiments are covered under Subtasks 4.1.2, 4.2.3, and 4.2.7, respectively.

Subtask 4.9. ORNL Fission Product Surveillance Program (see Section 4.9 for justification)

It is expected that experiments will be performed in the Peach Bottom reactor to measure (1) plateout profiles for condensable fission products in the primary circuit, (2) concentrations of fission products in the helium coolant leaving the core, (3) concentrations of fission products

in the helium coolant at a point downstream of a steam generator, and (4) concentrations of short-lived krypton and xenon precursors in the primary circuit. This work is covered under Subtasks 4.4.1, 4.4.3, 4.4.4, and 5.6.

Subtask 5.1. Blowdown Experiments (see Section 5.1 for justification)

It is expected that blowdown experiments will be conducted using plateout specimens obtained from, for example, the GGA strontium deposition loop (see Subtask 5.6).

Subtask 5.2. Mathematical Models for Calculating Fission Product Release, Transport, and Plateout from HTGR Cores (see Section 5.2 for justification)

Mathematical models are to be improved and extended as needed for analysis of fission product behavior under accident conditions.

Subtask 5.3. Studies of the Steam-Graphite Reaction (see Section 5.3 for justification)

The work in this subtask includes:

(a) Out-of-pile studies to determine the effect of various factors on the relative steam oxidation rate of fuel particle coatings, binder, and graphite.

(b) In-pile studies to verify the results of out-of-pile studies. (Data on fission product release and transport during steam attack will be emphasized.)

(c) Experiments on the irradiation-induced steam-graphite reaction using a Co⁶⁰ source.

(d) A study of the effect of helium pressure on steam-graphite reaction rates.

(e) Development of models and analytical methods to describe the steam oxidation behavior of HTGR cores.

Subtask 5.4. Fission Product Retention in Concrete (see Section 5.4 for justification)

This work consists of the determination of the effectiveness of concrete surfaces for filtering and sorbing fission products. Concrete

specimens containing large and small cracks will be used. Delay time data will be measured.

Subtask 5.5. Formation of Methyl Iodide (see Section 5.5 for justification)

The work in this subtask includes:

(a) Performance of required experiments to study formation of methyl iodide under reactor conditions.

(b) Analysis of data obtained from these experiments.

Subtask 5.6. Fission Product Plateout (see Sections 4.1.4, 4.2.12, and 5.6 for justification)

The work in this subtask includes:

(a) Performance of laboratory experiments, such as the GGA strontium deposition loop, to gain needed information on the plateout characteristics of strontium, iodine, and cesium as a function of temperature and type of metallic surface. Of particular interest will be determination of the distribution of metallic fission products and iodine between carbon dust and metallic surfaces.

(b) Determination of plateout profiles in the primary circuit of the Peach Bottom reactor. (This effort is a part of the ORNL Fission Product Surveillance Program; see Section 4.9.)

(c) Development of adequate mathematical models and correlation between the mathematical models, literature data, and the experimental data obtained in laboratory, reactor, and loop experiments.

Subtask 5.7. Tritium (see Section 5.7 for justification)

Work in this subtask includes:

(a) Studies of the distribution and transport of tritium in the Peach Bottom reactor (or other HTGR or loop systems).

(b) Studies of the retention of tritium in irradiated coated fuel particles.

7. BUDGET AND SCHEDULING

The following table gives the areas of required work under each subtask and the estimated cost of each subtask during the five-year period FY-71 through FY-75. The table also indicates the laboratory where the work could best be performed. (ORNL signifies Oak Ridge National Laboratory; BNWL, Battelle Northwest Laboratory; PE, Philadelphia Electric; and GGA, Gulf General Atomic.)

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.1.1. <u>RAD Code Development</u> (No apparent need for further development work on the RAD code.)							
4.1.2. <u>Fission Gas Release Studies</u>		140	200	200	220	220	980
(a) Study effect of time, temperature, irradiation, and thermal cycling on fission gas release. (ORNL, GGA)							
(b) Study relationship of gaseous release and fuel contamination. (GGA)							
(c) Develop simple quality control methods. (GGA)							
(d) Measure fission gas release for test elements. (GGA)							
(e) Study effect of rod fabrication on fission gas release. (GGA)							
(f) Study relative fission gas release behavior of coated oxides and carbides. (GGA)							
(g) Study effect of coolant impurities and burnup on gaseous release. (GGA)							
(h) Study variation of release processes with half-life, temperature, neutron flux, and helium pressure. (GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/ FY	1971	1972	1973	1974	1975	Total
4.1.3. <u>Calculation of Release of Fission Gases from Coated Fuel Particles</u>		5	10	10	10	10	45
Apply methods for analyzing re-release of fission gases from coated particles as required. (GGA)							
4.1.4. <u>Plateout of Iodine</u>		-	-	-	-	-	
(See Subtask 5.6.)							
4.1.5. <u>Sorption of Iodine in Graphite</u>		-	-	-	20	-	20
Study sorption of iodine in graphite. (GGA)							
4.2.1. <u>Methods for Calculating Release of Metallic Fission Products from HTGR Fuel Elements</u>		20	30	30	20	20	120
(a) Continue FIPER I development. (GGA)							
(b) Continue FIPER II development. (GGA)							
4.2.2. <u>Calculation of Release of Metallic Fission Products from Coated Fuel Particles</u>		5	10	10	10	10	45
Apply SLIDER (or FIPER) code to laboratory release data for BISO-coated particles. (GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.2.3. <u>Release of Metallic Fission Products from Coated Fuel Particles</u>		150	170	160	150	150	780
(a) Evaluate metal release in post-activation and postirradiation experiments. (ORNL, GGA)							
(b) Determine in-pile metal release. (GGA)							
(c) Refine release constant models. (GGA)		→					
(d) Determine relationship between release behavior and fuel contamination. (GGA)		→					
(e) Compare metal release of coated carbide and oxides.		→					
4.2.4. <u>Diffusion of Metallic Fission Products in Coating Materials</u>		-	30	20	20	-	70
(a) Measure diffusion coefficients for metals in coating materials. (GGA)		→					
(b) Measure solubility of metals in coating materials. (GGA)		→					
(c) Analyze release data to determine diffusion coefficients. (GGA)		→					
4.2.5. <u>Sorption of Metallic Fission Products in Fuel Rod Matrix Material</u>		-	20	40	40	40	140
(a) Measure sorptive capacity for Sr and Cs of reference matrix material. (GGA)		→					
(b) Determine effect of irradiation on sorptive capacity of reference matrix material. (GGA)		→					

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/ FY	1971	1972	1973	1974	1975	Total
4.2.6. <u>Distribution of Metallic Fission Products Between Matrix and Graphite Materials</u>		-	20	30	20	-	70
(a) Study distribution of Sr between matrix and graphite material using annealing technique. (GGA)							
(b) Perform in-pile experiments to study distribution of Sr between matrix and graphite material. (GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.2.7. <u>Diffusion of Metallic Fission Products in Graphite.</u> (In the following studies, the diffusion of strontium will be emphasized.)		60	140	160	160	150	670
(a) Study phenomenology of diffusion (e. g., multiprocess diffusion) of metals in graphite using plane source, permeation, autoradiograph, and microprobe techniques in temperature range 800° to 1200°C. (GGA)							
(b) Study concentration dependence of diffusion of metals in graphite using plane source and permeation techniques at 900° to 1100°C. (GGA)							
(c) Study temperature dependence of diffusion of metals in graphite using plane source and permeation techniques in the temperature range 800° to 1200°C. (GGA)							
(d) Study effect of graphite characteristics on diffusion of metals in graphite. (GGA)							
(e) Study effect of other metals (such as barium) on diffusion rate of a given metal (such as Sr). (GGA)							
(f) Conduct in-pile experiments (in Peach Bottom HTGR) to study diffusion of metals under reactor conditions. (GGA)							
(g) Study effect of thermal gradient on diffusion of metals in graphite. (GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.2.8. <u>Vapor Pressure of Metallic Fission Products Sorbed on Graphite</u>		40	40	30	20	20	150
(a) Measure vapor pressures of fission product metals in support of interaction effect studies.(GGA)							
(b) Measure vapor pressure of Sr at low concentrations. (GGA)							
(c) Determine effect of graphite characteristics on Sr vapor pressure. (GGA)							
4.2.9. <u>Interaction Effect of Metals Sorbed on Graphite</u>		-	60	60	80	60	260
(a) Study interaction effects of metals sorbed on graphite. (GGA)							
(b) Refine theoretical relationships for treating sorption of mixed metals on graphite. (GGA)							
4.2.10. <u>Vaporization of Strontium from Graphite in the Presence of Carbon Monoxide Gas</u>		-	-	-	-	-	
(No further work in planned on this subtask.)							
4.2.11. <u>Effect of Irradiation on Sorption Characteristics of Graphite</u>		-	-	20	20	20	60
Determine effect of irradiation on capacity of graphite to sorb Sr. (GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.2.12. <u>Plateout of Metallic Fission Products</u>							
(See Subtask 5.6)							
4.3. <u>Integral (Loop and Reactor) Systems</u>		150	160	170	150	100	730
Study Cs and Sr transport utilizing modified fission product deposition loop at ORNL, including related laboratory work. (ORNL)							
4.4.1. <u>Peach Bottom Gaseous Activity Determination</u>		25	40	40	20	-	125
Measure gaseous activity in purge system and main loop at Peach Bottom. (PE, ORNL, GGA)							
4.4.2. <u>Peach Bottom Iodine Release Measurements</u>		10	10	10	5	-	35
Measure iodine release into purge system and main loop at Peach Bottom. (PE, GGA)							
4.4.3. <u>Analysis of Particulate Matter Collected in Peach Bottom Main Loop</u>		25	60	60	80	50	275
Analyze particulate matter collected in Peach Bottom main loop collectors. (ORNL, GGA)							
4.4.4. <u>Peach Bottom Diffusion Probe Experiments</u>		60	120	100	90	40	410
Conduct diffusion probe experiments at Peach Bottom. (PE, ORNL, GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4. 4. 5. <u>Distribution of Fission Products in Peach Bottom Fuel Elements</u>		30	40	40	120	80	310
Study distribution of fission products in Peach Bottom fuel elements. (GGA)							
4. 5. <u>Testing of Calculational Methods</u>		10	10	10	10	20	60
Test calculational methods using data from integral experiments. (GGA)							
4. 6. <u>Strontium Gettering</u>		10	40	60	60	60	230
(a) Perform literature search. (GGA)							
(b) Determine quantities needed and effect on reactor performance. (GGA)							
(c) Perform experiments to determine influence of getters on strontium transport. (GGA)							
(d) Perform compatibility studies. (GGA)							
4. 7. <u>Plateout and Diffusion Probes</u> (See Subtask 4. 4. 4)		30	30	-	-	-	60
Complete design of equipment for monitoring fission product transport in Fort St. Vrain reactor. (GGA)							
4. 8. <u>Peach Bottom Fuel Test Elements</u>							
(Test element fission product release and diffusion studies are covered under Subtasks 4.1.2, 4.2.3, and 4.2.7, respectively.)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
4.9. <u>ORNL Fission Product Surveillance Program</u>							
(This work is covered under Subtasks 4.4.1, 4.4.3, 4.4.4, and 5.6.)							
5.1. <u>Blowdown Experiments</u>		-	40	60	50	50	200
Perform blowdown experiments using plateout specimens. (GGA)							
5.2. <u>Mathematical Models for Calculating Fission Product Release, Transport, and Plateout from HTGR Cores</u>		20	30	20	10	10	90
Modify codes as required. (GGA)							
5.3. <u>Studies of the Steam-Graphite Reaction</u>		375	380	300	240	200	1495
(a) Study (out-of-pile) the steam-graphite reaction. (ORNL, BNWL, GGA)							
(b) Study (in-pile) the steam-graphite reaction. (ORNL)							
(c) Study the irradiation-induced steam-graphite reaction. (BNWL)							
(d) Study the effect of helium pressure on the rate of the steam-graphite reaction. (GGA)							
(e) Develop analytical methods. (ORNL, BNWL, GGA)							

Budget Projection by Fiscal Year (in thousands of dollars)							
No. Subtask	/FY	1971	1972	1973	1974	1975	Total
5. 4. <u>Fission Product Retention in Concrete</u>		20	20	-	-	-	40
Measure the filtering effectiveness of concrete. (GGA)	→						
5. 5. <u>Formation of Methyl Iodide</u>		-	-	50	50	30	130
(a) Perform experiments on methyl iodine. (ORNL, GGA)	→						
(b) Analyze data obtained from these experiments. (GGA)	→						
5. 6. <u>Fission Product Plateout</u>		110	180	180	180	150	800
(a) Study plateout characteristics of fission products as a function of temperature and type of surface. (ORNL, GGA)	→						
(b) Determine plateout profiles in Peach Bottom reactor. (PE, ORNL, GGA)	→						
(c) Develop mathematical models and plateout data. (ORNL, GGA)	→						
5. 7. <u>Tritium</u>		10	40	40	30	30	150
(a) Study tritium distribution and transport in reactor and loop systems. (PE, GGA)	→						
(b) Study retention of tritium in irradiated fuel particles. (GGA)	→						
TOTAL		1305	1930	1910	1885	1520	8550

8. FUNDING

Present and proposed funding in support of the National Program for Development Work on Fission Product Behavior is as follows:

	<u>Funding (in thousands of dollars)^a</u>		
	<u>FY-71</u>	<u>FY-72</u>	<u>FY-73</u>
AEC Base Program			
ORNL	\$ 280	\$ 400	\$ 350
GGA	200	400	350
AEC Nuclear Graphite Program			
BNWL	75	140	150
AEC Safety Program			
ORNL	415	530	600
GGA	0	100	100
Industry Reactor Programs			
PE	25	30	30
GGA	<u>310</u>	<u>330</u>	<u>330</u>
TOTAL	\$ 1305	\$ 1930	\$ 1910

^aThe funding levels for ORNL, BNWL, and PE are estimates.

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APPENDIX A

DESCRIPTION OF HTGR SYSTEMS

The purpose of this appendix is to describe briefly the major features of HTGR systems, in particular features related to fission product behavior. The reactor systems described are:

1. The Peach Bottom nuclear power station
2. The Dragon experimental reactor
3. The Fort St. Vrain nuclear power station
4. The UHTREX
5. The AVR

Reviews of operation experiences for these HTGR systems were presented by Trauger^{(1)*} and Rennie.⁽²⁾

A. 1. THE PEACH BOTTOM NUCLEAR POWER STATION⁽³⁾

The Peach Bottom HTGR power station was built by Philadelphia Electric Company and 52 other U. S. utility companies of the HTRDA group in conjunction with Gulf General Atomic under the USAEC nuclear power demonstration program, with substantial research and development funding by the AEC. Bechtel Corporation was architect-engineer-contractor. The Peach Bottom power station, which has a power of 40 MW(e), was the first nuclear power station in the world to produce commercial electric power at modern steam conditions of 1000^oF temperature and 1450 psi pressure.

The core of the Peach Bottom HTGR is composed of 804 individual fuel elements vertically oriented in a close-packed array. The core array is cylindrical, with an effective diameter of 9 ft and an active height of 7.5 ft. Helium coolant gas, at an operating pressure of 350 psia, passes upward through the tricuspid-shaped passages formed by the elements.

*References are listed at the end of each appendix.

The Peach Bottom fuel element, shown in Fig. A. 1., has an overall length of 12 ft and an outside diameter of 3.5 in. and consists of an upper reflector section, a fuel-bearing middle section, a bottom reflector section, and an internal fission product trap containing activated carbon. A low permeability sleeve extending from the upper reflector to the bottom reflector contains the fuel compacts and the internal fission product trap. The fuel compacts consist of fuel particles in a graphite matrix. The fuel particles are between 200 and 600 μ in diameter and consist of uranium-thorium carbide particles coated with pyrolytic carbon.⁽⁴⁾

Helium purge gas enters each fuel element through a porous plug in the upper-reflector piece and then flows downward and around the fuel compacts, sweeping volatile fission products out of the grooved space between the fuel compacts and the graphite sleeve. The purge gas then flows through the internal trap, where some of the fission products are adsorbed by the activated carbon. Volatile fission products leaving each internal trap enter a purge line leading to the external fission product trap.

The primary coolant system consists of the reactor, two steam generators, and two helium compressors arranged to form two parallel loops for circulating the helium coolant.

Commercial operation of the Peach Bottom power station began in June 1967. In October 1969, the reactor was shut down after 450 EFPD (effective full-power days). The reactor attained full-power operation on July 14, 1970 with its second core.

A. 2. THE DRAGON EXPERIMENTAL REACTOR⁽⁵⁾

The Dragon HTGR, located at Winfrith, England, is a 20-MW(t), helium-cooled experimental reactor. The purpose of the reactor was to demonstrate the viability of the HTGR concept. The reactor does not produce power.

The Dragon core consists of 37 fuel elements which form an approximate cylinder 3-1/2 ft in diameter and 5-1/4 ft in length. Individual fuel elements contain seven fuel rods, each of which is composed of a graphite

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Fig. A. 1. Peach Bottom fuel element

sleeve, annular fuel cartridges, and a graphite spine. Each fuel cartridge consists of coated fuel particles compacted into a graphite matrix.

Every fuel element is purged, and the purge stream passes through delay beds to permit decay of krypton and xenon activity. The helium coolant, at a pressure of 20 atm, passes upward through the core. After leaving the core, the hot gas passes through the six heat exchangers. The inlet helium temperature is 662°F (350°C) and the outlet temperature is 1382°F (750°C).

A. 3. THE FORT ST. VRAIN NUCLEAR POWER STATION^(6, 7)

The 330-MW(e) Fort St. Vrain nuclear generating station is being built by Gulf General Atomic for Public Service Company of Colorado as a part of the USAEC Power Reactor Demonstration Program. The plant, which will be located approximately 35 miles north of Denver, Colorado, is scheduled to begin commercial operation in the 1971-1972 period.

Like the Peach Bottom plant, the Fort St. Vrain HTGR will utilize high temperature helium coolant to produce steam (superheated and reheated) at a temperature of 1000°F to match modern thermal station conditions. The plant will have a net overall efficiency of approximately 40%.

Major features of the primary coolant systems of the Fort St. Vrain station are the use of a prestressed concrete reactor vessel (PCRv), once-through modular steam generators with integral superheaters and reheaters, steam-driven axial flow helium circulators, and hexagonal graphite fuel elements incorporating improved, carbon-coated fuel particles.

The reactor arrangement is shown in Fig. A.2. The coolant flow from the reactor core bifurcates into two identical coolant loops, each consisting of a six-module steam generator, a steam generator outlet plenum, and two helium circulators. The circulators of both loops discharge into a common plenum below the core support floor. All of the flow passes upward around the core support floor and the barrel to the core inlet plenum above the reactor core.

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Fig. A.2. Primary coolant system arrangement and flow paths

The helium coolant, at a pressure of about 700 psia, flows downward through the reactor core, where it is heated to about 1430°F. The helium is then directed to the steam generators beneath the reactor core to produce superheated and reheated steam. After passing through the steam generators, the coolant is returned to the reactor at about 760°F by steam-turbine-driven circulators. Each circulator unit is mounted vertically within a penetration through the bottom of the reactor vessel.

The reactor core is composed of approximately 1500 hexagonal graphite fuel elements. These elements are stacked in 247 vertical columns to form an active core approximately 19-1/2 ft in diameter and 15-1/2 ft high. The individual fuel elements, shown in Fig. A.3., are made of graphite and are hexagonal in cross section, measuring approximately 14 in. across the flats by 31 in. high. Vertical coolant holes within each element are aligned with coolant holes in elements above and below. Coated fuel bound in a fuel rod is contained in an array of small-diameter holes, which are parallel to the coolant holes. An engagement hole at the center of each fuel element is provided for handling purposes. Three graphite dowels for aligning the elements within a column are located on the top faces of the elements. The fuel element graphite is conventional nuclear grade graphite.

In the equilibrium fuel cycle, the average fuel element contains about 0.5 kg of fissionable uranium and 10 kg of thorium and generates about 600 kW of thermal power. The specific power is thus about 1200 kW/kg.

The fuel particles are spherical kernels of carbides of thorium and uranium coated with layers of pyrolytic carbon and silicon carbide. This method of coating provides the particles with excellent retention characteristics for all fission products. Coated particles of this type are called

Fig. A. 3. Fort St. Vrain fuel element
(all dimensions are in inches)

TRISO particles. Interim design conditions and specifications for these coated particles are given in Fig. A.4.

A.4. THE UHTREX⁽⁸⁾

The Ultra High Temperature Reactor Experiment (UHTREX), located at the Los Alamos Scientific Laboratory, was built to gain information for the development of more economic nuclear fuel. The UHTREX utilized unclad fuel elements in a helium-cooled, graphite-moderated reactor operated at 3 MW and a maximum coolant temperature of 2400°F.

Fuel elements were unclad, hollow, cylindrical compacts, 1 in. in outside diameter and 5.5 in. long, and consisted of coated fuel particles held in a graphite matrix. Four fuel elements rested end-to-end in each of 315 fuel channels to make a total of 1248 elements in a complete core loading.

The entire core, together with portions of the reflector and insulation, could be rotated about a cylindrical axis to accomplish fuel loading and unloading. A fuel loader, mounted on the side of the pressure vessel, moved fuel elements into and out of the core, even while the reactor was operating. As a new fuel element was inserted, a spent element was discharged into a slot and was conveyed to a fuel handling cell.

Helium coolant, at pressures up to 550 psia, entered at the bottom of the reactor, flowed up through the plenum around the central plug, passed through the fuel channels, collected in an annular plenum around the outside of the movable core, and left the reactor through a graphite pipe at the side.

In June 1969, the UHTREX reactor attained its full design operating condition; it was operated at 2400°F coolant outlet temperature and a power of 3 MW. Reactor operations were terminated in early 1970, and all fuel elements were unloaded.

FISSILE
 (Th,U)C₂
 4.25
 6 YEARS
 1260°C
 20% FIMA
 8.0 X 10²¹ N/CM²

FORM
 Th:U RATIO
 LIFETIME
 TEMPERATURE (MAX)
 BURNUP (MAX)
 FAST FLUX EXPOSURE (MAX)

FERTILE
 ThC₂
 (ALL Th)
 6 YEARS
 1260°C
 7% FIMA
 8.0 X 10²¹ N/CM²

A (SMALL)

B (LARGE)

100-175μ

175-275μ

50μ

50μ

20μ

20μ

20μ

20μ

30μ

40μ

120μ

130μ

PARTICLE SIZE

BUFFER COATING

ISOTROPIC PyC COATING

SiC COATING

ISOTROPIC PyC COATING

TOTAL COATING

A (SMALL)

B (LARGE)

300-450μ

450-600μ

50μ

50μ

20μ

20μ

20μ

20μ

40μ

50μ

130μ

140μ

Fig. A.4. Interim design conditions and specifications for Fort St. Vrain TRISO-coated particle fuel (particles are drawn to scale)

A. 5. THE AVR⁽⁹⁾

The Arbeitsgemeinschaft Versuchsreaktor (AVR), located in Jülich, West Germany, is a 15-MW(e), helium-cooled power reactor. This reactor is unique in that it is a "pebble bed" reactor.

The AVR core consists of a moving bed of graphite balls, each 2.36 in. in diameter. Each ball contains a core of coated uranium carbide particles in a graphite matrix.

The fuel moderator balls are introduced at the top of the reactor and removed from the bottom, even while the reactor is in operation. The core is 10 ft in diameter and 12 ft high.

The AVR went critical on August 26, 1966, and power operation commenced in September 1967.⁽⁹⁾ Since that time, the plant has operated intermittently. Difficulties have been encountered with the gas purification plant and the fuel handling facility.

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APPENDIX B

DESCRIPTION OF IN-PILE LOOP AND PURGED SYSTEMS

B.1. GAIL

The Gulf General Atomic in-pile loop (GAIL) at the General Electric Test Reactor (GETR) was designed to obtain information of a confirmatory nature under simulated reactor conditions. In the in-pile loop test, a fuel element could be irradiated in a thermal neutron flux slightly higher than 10^{13} nv in a stream of purified helium at 350 psia. Provisions were made for purging the fuel element and passing the purge stream through internal and external fission product traps. The GAIL is described in Ref. 1.

The major items of equipment in the primary loop were the facility tube containing the test fuel element, circulators for pumping the helium coolant through the loop, a cooler for dumping the heat produced in the element, a 50-kW electric heater for providing fine control of the inlet helium temperature, and associated piping, valves, and instrumentation.

B.1.1. GAIL IV Experiment

The GAIL IV fuel element (described in detail in Ref. 2) consisted of a 12-hole, telephone dial, cross-sectional configuration with loose Triplex pyrolytic-carbon-coated $(Th,U)C_2$ fuel particles packed into the holes. The fuel holes were located 30° apart. The fuel element assembly (fabricated from Speer 780-S graphite) was about 40-7/8 in. in overall length and 3-1/2 in. in diameter. The active length of fuel particles in the element was about 22-1/2 in. The end plugs in fuel hole No. 1 contained thin cobalt wafers for use in axial length measurements of the irradiated graphite body.

A 16-in. -long Speer 780-S graphite spine was located in the central base of the fuel element. Above the graphite spine, a 7-3/4-in. -long BeO spine extended upward to the level of the top of the active fuel bed. A 10-in. -long by 3/4-in. -diameter acoustic resonance cavity centered 9 in. above the bottom of the active fuel was incorporated in the graphite spine.

Helium at a pressure of 350 psia flowed downward between an outer baffle and the inside wall of the pressure vessel. At the bottom of the outer baffle, the flow was turned upward and passed over the surface of the fuel element. A purification system removed fission products and impurities from the helium.

Provision was made for purging the fuel element. In the purged mode of operation, helium was drawn radially inward through the graphite body into the annular space between the spines and the surface of the central hole in the fuel element. Purge helium and fission products were drawn into an internal fission product trap which contained activated charcoal.

The postirradiation examination of the GAIL IV fuel element is described in Refs. 2 and 3. Conclusions drawn from the examination were: (1) the packed bed fuel element design operated very satisfactorily; (2) the Triplex-coated fuel particles were in good condition; (3) good in-pile fission gas retention was achieved despite a few (~0.8%) broken fuel particles; and (4) all metal and graphite components were in good condition.

B. 1.2. THE GAIL III-B

The GAIL III-B fuel element was a graphite assembly with an overall length of about 43-3/8 in. and an outside diameter of 3-1/2 in. The 22-1/2-in. -long active fuel region was composed of 15 annular fuel compacts 1-1/2 in. long, 2-3/4 in. in outside diameter, and 1/2 in. thick radially. The fuel compacts were made by hot-pressing a graphite matrix containing approximately 30 vol-% of pyrolytic-carbon-coated (Th,U)C₂ particles of active fuel. The nearly spherical uncoated carbide particles

ranged from 150 to 354 μ in diameter and contained about 25% fully enriched uranium, 61% thorium, and 15% carbon. The average thickness of the pyrolytic carbon coating was 55 μ . The total amount of U^{235} contained in the 15 compacts was 195 g, and the average Th/U ratio was about 2.5. A graphite spine extended the full length of the active fuel through the central hole in the annular fuel compacts. The GAIL III-B experiment is described in Ref. 4.

B. 2.1. Pluto Loop A

The Pluto loop A system was basically a single channel of the Dragon reactor and simulated the majority of components and processes involved in the reactor. The loop was located in the heavy-water reflector adjacent to the core of the Pluto reactor at Harwell, England. The fuel elements were full size except for a 40% reduction in length. The purpose of the loop experiments was to investigate problems concerning the coolant, corrosion, mass transfer, and fission product emission and control.

A total of 16 experiments were conducted in Pluto Loop A, and the loop is no longer operative. Extensive fission product transport data were obtained from experiments 3, 8, 15, and 16 (see Refs. 5-8). These data are of particular use for correlation with calculated fission product transport data.

B. 2.2. Pluto Loop B

It is intended that a reactor loop, to be known as Pluto Loop B, will be constructed at Harwell within the next two years.⁽⁹⁾ The purpose of the loop is to check FIPDIG predictions (see Section 4.2.1.3) and to study plateout characteristics of HTGR systems. By means of this loop, it will be possible to obtain fission product release data, plateout factors, distribution profiles, and blow-off factors.

It is planned that the loop will comprise a 10-in. fuel element with very small axial gradient, a cross-flow boiler of reactor design, and a helium circulation system. Attempts will be made to simulate heat fluxes in both fuel element and boiler surfaces. The fuel element

will be made to operate like the hottest element in a reactor by heating the helium electrically to simulate heating by upstream fuel elements in a reactor. Experiments are planned in which the fuel element will contain a known fraction of broken particles.

B. 3. STUDSVIK LOOP

A fuel irradiation test loop exists in the R2 reactor at Studsvik, Sweden. This loop is a part of the Dragon program. The fission gas release behavior of fuel samples can be measured in this loop.

B. 4. 6F2 SWEEP CAPSULE EXPERIMENT

The GA2-284-6F2 experiment was designed as a high temperature helium-purged irradiation capsule to determine the fission gas release characteristics of three different types of HTGR fuel bodies.⁽¹⁰⁾ This was an early test of HTGR-type fuel and was performed in 1963. The fuel bodies contained coated (Th, U)C₂ fuel particles and were of three types: (1) fuel particles coated with alternate layers of silicon carbide and pyrolytic carbon dispersed in a hot-pressed graphite matrix, (2) fuel particles with a monolithic laminar pyrolytic carbon coating dispersed in an extruded graphite matrix, and (3) fuel particles with a monolithic laminar pyrolytic carbon coating dispersed in a hot-pressed graphite matrix (Peach Bottom prototype).

These fuel bodies were irradiated in the core of the GETR at temperatures of around 1300°C for a total of 2640 hr. Purge gas samples were collected at periodic intervals and analyzed for noble gas fission products. The isotopic distributions in these samples indicated that the observed fission product release may have been primarily from fuel particles which had undergone some type of coating failure. The fission product release rates increased throughout the irradiation, with the multilayered silicon carbide-pyrolytic carbon coated fuel particles showing the greatest retention of fission products. A postirradiation examination of the fuel particles indicated that a high percentage of the particle coatings had failed during the irradiation

period. However, the fission product release data remained below the fractional release normally obtained on uncoated (Th, U)C₂ particles.

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